Cette thèse étudie la conduction électrique sous champ magnétique de l'interface entre un substrat de SrTiO3 et une mince couche cristalline de LaAlO3. Ce système combine les propriétés remarquables des gaz bidimensionnels et la richesse de la physique des oxydes complexes. Grâce à nos mesures, nous révélons le caractère multi-bande de ce système et mettons à jour une transition, induite par l'effet de champ, entre un régime dominé par des corrections quantiques et un régime semi-classique. Lorsque le peuplement des nouvelles sous-bandes est faible, nous montrons qu'il existe une sous-bande pour laquelle le couplage spin-orbite Rashba est si important que sa densité d'états est modifiée quand l'orientation du champ magnétique est variée. Nous mettons également à jour le comportement complexe des oscillations quantiques, lequel peut être relié au rôle primordial de l'interaction spin-orbite dans ce système. Pour finir, nous révélons une évolution subtile des oscillations quantiques avec l'effet de champ.


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Magnetotransport Experiments at the \LaAlO_3/SrTiO_3 Interface

THÈSE

présentée à la Faculté des Sciences de l'Université de Genève pour obtenir le grade de docteur ès Sciences, mention Physique

par

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de
Cortébert (BE)

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Genève, le 17 avril 2014

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Le Doyen, Jean-Marc TRISCONE

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<tr>
<td>2D</td>
<td>Two-Dimensional</td>
</tr>
<tr>
<td>2DEG</td>
<td>Two-Dimensional Electron Gas</td>
</tr>
<tr>
<td>MR</td>
<td>Magnetoresistance</td>
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<tr>
<td>HE</td>
<td>Hall Effect</td>
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<tr>
<td>MC</td>
<td>Magnetococonductance</td>
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<tr>
<td>DOS</td>
<td>Density Of States</td>
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<tr>
<td>FS</td>
<td>Fermi Surface</td>
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<tr>
<td>LL</td>
<td>Landau Level</td>
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<tr>
<td>PLD</td>
<td>Pulsed Laser Deposition</td>
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<tr>
<td>RHEED</td>
<td>Reflection High-Energy Electron Diffraction</td>
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<tr>
<td>SdH</td>
<td>Shubnikov-de Haas</td>
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<tr>
<td>uc</td>
<td>Unit Cell</td>
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<tr>
<td>( q )</td>
<td>Electric charge</td>
</tr>
<tr>
<td>( e )</td>
<td>Electron charge ( (e &lt; 0) )</td>
</tr>
<tr>
<td>( m_e )</td>
<td>Free electron mass</td>
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<tr>
<td>( c )</td>
<td>Speed of light</td>
</tr>
<tr>
<td>( h )</td>
<td>Reduced Planck constant</td>
</tr>
<tr>
<td>( m^* )</td>
<td>Electron’s effective mass</td>
</tr>
<tr>
<td>( n_{2D} )</td>
<td>Sheet carrier density</td>
</tr>
<tr>
<td>( \mu )</td>
<td>Electron’s mobility</td>
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<tr>
<td>( \omega_c )</td>
<td>Cyclotron frequency</td>
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<tr>
<td>( \sigma_{2D} )</td>
<td>Sheet conductance</td>
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<td>( R_s )</td>
<td>Sheet resistance</td>
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<td>( R_{xy} )</td>
<td>Hall resistance</td>
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<td>( R_H )</td>
<td>Hall coefficient</td>
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<tr>
<td>( \mathbf{A} )</td>
<td>Notation for a vector</td>
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<td>( A )</td>
<td>Norm of ( \mathbf{A} )</td>
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<tr>
<td>( \mathbf{B} )</td>
<td>Notation for a tensor</td>
</tr>
<tr>
<td>( \mathbf{C} )</td>
<td>Notation for an operator</td>
</tr>
<tr>
<td>( \mathbf{k} )</td>
<td>Reciprocal space wave vector</td>
</tr>
<tr>
<td>( \mathbf{E} )</td>
<td>Electric field vector</td>
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<tr>
<td>( \mathbf{B} )</td>
<td>Magnetic field vector</td>
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<tr>
<td>( \sigma )</td>
<td>Tensor of conductance</td>
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<tr>
<td>( \rho )</td>
<td>Tensor of resistance</td>
</tr>
<tr>
<td>( f^0 )</td>
<td>Fermi-Dirac (equilibrium) distribution function</td>
</tr>
<tr>
<td>( f )</td>
<td>General (out-of-equilibrium) distribution function</td>
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**List of symbols**

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
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<tbody>
<tr>
<td>$g_k$</td>
<td>$f_k - f_k^0$</td>
</tr>
<tr>
<td>$\dot{u}$</td>
<td>Time-derivative of the function $u$</td>
</tr>
<tr>
<td>$\ddot{u}$</td>
<td>Second time-derivative of the function $u$</td>
</tr>
<tr>
<td>$h'$</td>
<td>First derivative of the function $h$</td>
</tr>
<tr>
<td>$k_B$</td>
<td>Boltzmann constant</td>
</tr>
<tr>
<td>$\epsilon_F$</td>
<td>Fermi energy</td>
</tr>
<tr>
<td>$\Theta$</td>
<td>Heaviside function</td>
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Cette thèse étudie les propriétés de conduction électrique sous champ magnétique de l’interface entre un substrat de SrTiO$_3$ et une mince couche cristalline de LaAlO$_3$ ($\approx 5$ nm).

Ce système est fascinant car il combine les propriétés remarquables des gaz bidimensionnels, lesquelles ont largement contribué à la popularité des hétérostructures semiconductrices, et la richesse de la physique des oxydes complexes. A titre d’exemple, l’interface LaAlO$_3$/SrTiO$_3$ est supraconductrice, présente un fort couplage spin-orbite de type Rashba et, suivant les conditions de préparation, est ferromagnétique. Plus intéressant encore, la plupart de ces propriétés sont sensibles à l’effet d’un champ électrique, ce qui permet leur modulation sans avoir à recourir à un dopage chimique. Ces considérations sont le sujet du premier chapitre de cette thèse, lequel vise à donner un panorama aussi large que possible des études menées à ce jour sur ce système.

Le chapitre 2 naît du constat qu’il est absolument nécessaire, si l’on veut se forger une interprétation fiable des mesures de (magneto-)transport dans ce système, d’avoir une idée précise des scénarios justifiant la conduction électrique à l’interface LaAlO$_3$/SrTiO$_3$ et de leurs implications en termes de structure de bande. Dans ce but, nous passons tout d’abord en revue les principaux modèles décrivant l’origine de la conduction interfaciale et étudions ensuite en détail la structure de bande issue du scénario dit de “reconstruction électronique”. Pour ce faire nous avons réalisé plusieurs simulations numériques qui nous permettent de faire ressortir plusieurs facteurs importants pour la configuration électronique du système. L’un d’eux est la présence d’états localisés dans la bande interdite du SrTiO$_3$ qui doivent être considérés dans l’électrostatique du problème pour rendre compte correctement du champ de confinement.


Le premier se concentre sur des interfaces dites “standard” (synthétisées à 800 °C).
Résumé en français

car leurs propriétés sont celles de la majeure partie des mesures publiées dans le domaine; ces échantillons ont une densité de porteurs de charge de l’ordre de $10^{13} \text{ cm}^{-2}$ et une mobilité électronique à 1.5 K aux alentours de 1000 cm$^2$ V$^{-1}$ s$^{-1}$. En particulier, nous étudions la magnétoresistance et l’effet Hall de ces échantillons en fonction de la température et de l’effet de champ. L’analyse de ces mesures nous permet de révéler le caractère multi-bande du système LaAlO$_3$/SrTiO$_3$. Nous mettons également à jour une transition, induite par l’effet de champ, entre un régime dans lequel le magnetotransport est dominé par des corrections quantiques (faible magneto-resistance) et un régime dans lequel la présence de sous-bandes de différentes symétries induit une contribution semi-classique importante (grande magneto-resistance). Ces études nous permettent également de fournir les bases nécessaires à l’interprétation de l’effet Hall dans ce système.

Dans le régime où le peuplement des nouvelles sous-bandes est faible, nous montrons qu’il existe une sous-bande pour laquelle le couplage spin-orbite Rashba est si important que sa densité d’états est modifiée lorsque le champ magnétique est appliqué dans le plan de l’interface et son orientation variée par rapport au courant. Expérimentalement, nous démontrons que cette situation provoque une oscillation périodique de la résistance en fonction de l’angle entre le champ magnétique et le courant. Nous remarquons également que le peuplement de cette sous-bande est lié à une augmentation du temps de diffusion élastique, un phénomène qui soulève des questions intéressantes quant à la présence d’états protégés à l’interface LaAlO$_3$/SrTiO$_3$.

Le chapitre 5 étudie une nouvelle classe d’interfaces LaAlO$_3$/SrTiO$_3$ découverte en 2010. Celles-ci sont synthétisées à plus basse température (650 °C) et, de ce fait, présentent un nombre de porteurs mobiles réduit (de l’ordre de $10^{12} \text{ cm}^{-2}$) et une mobilité augmentée (aux alentours de 7000 cm$^2$ V$^{-1}$ s$^{-1}$ à 1.5 K). Pour cette raison, nous les appelons interfaces “haute mobilité”. Etant donné le grand libre parcours moyen des électron dans ces échantillons, ces interfaces ont le grand avantage de permettre l’étude des oscillations quantiques à des champs magnétiques relativement faibles ($\approx 8$ T), lesquelles sont directement reliées à la structure de bande du système.

Expérimentalement, nous étudions ces oscillations dans la magnetorésistance de notre système (effet Shubnikov-de Haas). Leur étude en champs magnétiques télés (jusqu’à 33 T) nous permet d’attester du caractère bidimensionnel des porteurs les plus mobiles de ce système. En outre, en travaillant à très basses températures (entre 50 mK et 800 mK), nous mettons à jour un comportement très complexe des oscillations quantiques. Nous montrons que celui-ci peut être compris dans un modèle multi-bandes mais qu’un modèle muni d’une seule bande “splitée” par l’interaction spin-orbite de type Rashba est au moins tout aussi satisfaisant. Remarquablement, le couplage spin-orbite et la masse effective des porteurs que nous extrayons dans le scénario Rashba sont très similaires aux valeurs obtenues au chapitre 4. Nous interprétons ce résultat comme une nouvelle signature de l’importance d’une sous-bande relativement lourde et fortement “splitée” dans le magnetotransport des interfaces LaAlO$_3$/SrTiO$_3$. Pour finir, nous révélons une évolution subtile des oscillations Shubnikov-de Haas lorsque nous utilisons l’effet de champs pour moduler la densité de porteurs présents aux interfaces haute mobilité. En effet, alors que les fréquences d’oscillations sont globalement réduites lorsque le dopage électrostatique est diminué, nous observons des déviations...
Résumé en français

rapides (ou des sauts) dans la trajectoire des maxima des oscillations en fonction de la tension de grille. A notre avis, ces mesures ne peuvent pas être comprises dans un modèle d’électrons indépendants. Nous proposons donc que ces déviations sont créées par le croisements de niveaux de Landau de chiralité (dans un modèle Rashba) ou de sous-bande (dans un modèle multi-bandes) différentes.
CHAPTER 1

INTRODUCTION

All men dream, but not equally. Those who dream by night in the dusty recesses of their minds, wake in the day to find that it was vanity: but the dreamers of the day are dangerous men, for they may act on their dreams with open eyes, to make them possible.

T. E. Lawrence

1.1 Introduction

Nowadays an easy way to make a solid state physicist smile is to start a discussion with “quantum computer” or “room temperature superconductivity”. Indeed, these two topics are such big challenges for today’s scientists that they may appear to many as dreams. Yet, at the same time, the possibilities these realizations would bring with them drive the intense activity of many fields of research.

Let’s focus on the case of quantum computer. Why is it such a difficult task? The most exact answer to this question is probably: “because a quantum computer is quantum”. We agree that it is a quite disappointing answer, but in fact this statement covers all the complexity of the topic.

A classical computer is based on the classical (or common) logic. The information is stored in series of “bits” that can take either the value 0 or 1. These bits are then processed through gates in order to produce an answer to the query of the user. On the contrary, in a quantum computer the information is stored in “quantum bits” (qbits) that can take the values \(|0\rangle, |1\rangle\) and... \(\alpha|0\rangle + \beta|1\rangle\) (the notation |⟩ stands for a qbit) meaning that the qbit can be simultaneously in the state 0 and 1 (superposition state). This is already very weird and potentially useful as the qbit can thus explore simultaneously several solutions of the gate sequence. Yet, if this were everything about the quantum computer, the latter would probably not have attracted so much attention.

Where quantum computer becomes really quantum is when many qbits are considered. Indeed, similarly to a single qbit that can be put in a superposition state, qbits sequences can be “superposed”. Let’s take the example of a two qbits computer:
1. INTRODUCTION

this means that, in addition to the traditional $|0\rangle|0\rangle$, $|0\rangle|1\rangle$, $|1\rangle|0\rangle$ and $|1\rangle|1\rangle$ qbits sequences, the quantum computer has also to deal with strange combinations like $\alpha|0\rangle|0\rangle + \beta|1\rangle|1\rangle$. These combinations are called “entangled states”. Interestingly, considering the above example of “entangled state”, quantum mechanics tells you than as soon as you measure one of the two qbits composing it to be, for example, $|1\rangle$, the total configuration will instantaneously collapse to $|1\rangle|1\rangle$. This implies that, when qbits are entangled, some sort of link exists between the individual qbits, although it cannot be interpreted as a communication because of its instantaneity. The quantum computer is a device that takes advantage of this property.

Yet, controlled entanglement is a property that is difficult to preserve as the qbit tends to entangle with its environment when it is subjected to physical interactions with it (decoherence problem). In this context, the use of topological states as qbits has been proposed as a solution to this problem [1]. Indeed these states are naturally protected against external perturbations. For example, Majorana zero-energy modes have been pointed out to be “intrinsically immune to decoherence” [2].

In this thesis, we study the electronic properties of the electron gas present at the LaAlO$_3$/SrTiO$_3$ interface, created by depositing a few atomic layers of the insulating perovskite LaAlO$_3$ on top of a TiO$_2$-terminated SrTiO$_3$ substrate, another insulating perovskite. The metallic interface thereby created has been shown to be endowed with a lot of fascinating properties among them two-dimensional superconductivity and large Rashba spin-orbit coupling, both of them being tunable by an electric field. As a consequence, it has been proposed that Majorana fermions could be realized in this system [3]. We will not address directly this topic in this thesis. Yet, by studying across its phase diagram the electronic structure of this system, we intend to provide a better picture of the mechanisms at play in this heterostructure. This is a necessary step in order to possibly engineer Majorana physics at the LaAlO$_3$/SrTiO$_3$ interface. In addition to that, as we will see, the fascinating properties displayed by this oxide system link to a much broader field of research including high-temperature superconductivity and spin-based electronics.

1.2 The LaAlO$_3$/SrTiO$_3$ interface

In 2004, Ohtomo and Hwang discovered that the interface between an insulating LaAlO$_3$ layer and an insulating SrTiO$_3$ substrate is metallic [4]. This observation sparked a burst of research activity aimed at understanding the origin of the carriers and the properties of the system [7–9]; this thesis, participating to this effort, testifies that the system still hides some surprise.

The question on the origin of the conduction is still debated, owing to the complexity of the materials composing this heterostructure. Two main scenarios have been proposed: the first related to the intrinsic properties of the system, the second linked to the possible defects created during the growth.

The intrinsic scenario stems from the analysis of the ionic charge of the crystallographic planes along the [001] growth direction. Looking at fig. 1.1a, we see that the sequence of planes at the interface is SrO$^0$/TiO$_2^0$/LaO$^{+1}$/AlO$_2^{-1}$ (n-type interface). According to this picture, LaAlO$_3$ has a polar surface facing the non-polar material.
1.2 The LaAlO$_3$/SrTiO$_3$ interface

Figure 1.1: The metallic LaAlO$_3$/SrTiO$_3$ interface. (Left) Atomic structure of the interface. White refers to the Oxygen atoms, blue to the Strontium atoms, red to the Lanthanum atoms, cyan to the Titanium atoms and gray to the Aluminium atoms. On the left are the growth direction (001) and the chemical content of the crystallographic planes. The $0$, $+$ and $-$ refer to the formal ionic charge of the planes. (Right) Temperature dependence of the sheet resistance at the LaAlO$_3$/SrTiO$_3$ interface for different oxygen pressures during growth. From [4].

Figure 1.2: The role of oxygen vacancies on the transport properties of the interface (Left) Sheet resistance versus temperature for 6 samples grown in different oxygen environments. No annealing step is performed. From [5]. (Right) Map of the electrical resistance across the LaAlO$_3$/SrTiO$_3$ interface. The scan is performed using an atomic force microscope mounted with a conducting tip and scanning along the thickness of the sample. (Right top) Sample grown at $1 \times 10^{-6}$ mbar and annealed in 300 mbar of oxygen after growth. (Right bottom) Sample grown at $1 \times 10^{-5}$ mbar and not annealed. From [6].
1. INTRODUCTION

Figure 1.3: Critical thickness and room temperature field effect (Left) Sheet conductance at room temperature as a function of the number of LaAlO$_3$ unit cells (uc). A clear metal-insulator transition is observed at 4 uc. (Right) Modulation of the (room-temperature) sheet resistance of a 3 uc sample upon application of a back-gate voltage. Reproducible metal-insulator transitions are obtained. From [11].

SrTiO$_3$: this polar discontinuity has been suggested to be at the origin of the electron gas observed at the interface, hence screening the polarization [10], as will be described in detail in chap. 2. For the second scenario, the role of the stoichiometry of the heterostructure has been discussed since its discovery. In figure 1.1b and fig. 1.2a the dependence of the sheet resistance on the oxygen pressure used during the growth of the LaAlO$_3$ layer is shown. As further studies have revealed, there exist two main regimes for the conduction at the interface: for high oxygen pressure or using oxygen annealings, the electron gas is confined at the interface, as the image of the local resistivity obtained by conducting atomic force microscopy shows in fig. 1.2b. On the contrary, for low growth oxygen pressure, oxygen vacancies are created in the STO substrate, extending deeply in the bulk, as shown in fig. 1.2c. These two regimes can be dubbed “2D” and “3D” and their properties are different. In this thesis we will focus on the 2D state. In this regime, an important behavior of the n-type interface has been reported, first by Thiel et al. [11], then by many groups working on the LaAlO$_3$/SrTiO$_3$ interface. This is shown in fig. 1.3 that displays room-temperature measurements of the conductance vs thickness of the LaAlO$_3$ layer: we see that the interface is insulating for LaAlO$_3$ layers up to 3 uc thick, then it switches into a metallic state, with a conductance that becomes independent of the LaAlO$_3$ thickness. This insulator-to-metal transition can be elegantly explained within the framework of the intrinsic polar discontinuity [12]. Remarkably, a metal-to-insulator transition could also be achieved using the electrostatic field effect, as shown in fig. 1.3.

The occurrences of superconductivity and ferromagnetism at the LaAlO$_3$/SrTiO$_3$ interface are probably the two phenomena that made this system so famous in the oxide community. The first one was discovered in our lab by Reyren et al. [13]. Figure 1.4 shows that at about 200 mK a metal to superconductor transition is indeed observed. This transition temperature is similar to the temperature at which bulk doped SrTiO$_3$ is known to superconduct [14], yet, analysis of the superconducting transition at the LaAlO$_3$/SrTiO$_3$ interface revealed a behavior compatible with 2D superconductivity (transition of the Berezinskii-Kosterlitz-Thouless type). Concerning ferromagnetism, it
1.2 The LaAlO$_3$/SrTiO$_3$ interface

was first measured by the Twente group [5]. It attracted a lot of attention as an interface emergent phenomenon, since it is absent in bulk SrTiO$_3$ and bulk LaAlO$_3$. Figure 1.4 shows the hysteresis loop observed in the low-temperature magnetotransport of the interface.

Unfortunately, for a long time, the coexistence of these two phenomena was not evidenced as the above measurements were performed on samples prepared in different conditions (see caption of fig. 1.4). It is only rather recently that Li et al. [15] and Bert et al. [16] independently measured a ferromagnetic signal in superconducting LaAlO$_3$/SrTiO$_3$ interfaces using respectively torque magnetometry and SQUID measurements. In the latter study, the authors could resolve spatially the magnetic signal. From their analysis, they concluded a nanoscale phase separation at the interface, meaning that the carriers that are giving rise to superconductivity and ferromagnetism are distinct, the latter probably being localized at the interface. Nevertheless, the close proximity of these two orders is of fundamental interest not only for the fields of superconductivity and magnetism but also for the search for the more exotic quantum states described above.

Contrarily to ferromagnetism that is insensitive to field effect [17], superconductivity at the LaAlO$_3$/SrTiO$_3$ interface can be largely tuned using a gate voltage. This was discovered in Geneva by Caviglia et al. [18]. Figure 1.5 shows that the effect is indeed very important. It is materialized, in the associated phase diagram, by the presence of a quantum critical point (QCP) separating a superconducting region from an insulating one. The dome-shaped form of the superconducting region is reminiscent of the phase

Figure 1.4: Superconductivity and magnetism (Left) Sheet resistance as a function of temperature, in the milli kelvin regime, for different strengths of out-of plane magnetic field. A transition to a superconducting state is observed around 200 mK. The interface was grown in $6 \times 10^{-5}$ mbar of oxygen and annealed after growth. From [13]. (Right) Sheet resistance as a function of magnetic field at 300 mK. No superconductivity is observed in this sample. The interface was grown in $1 \times 10^{-3}$ mbar of oxygen without post-annealing. From [5].
1. INTRODUCTION

Figure 1.5: Tunable superconductivity (Left) Sheet resistance as a function of temperature, in the milli kelvin regime, for different gate voltages (field effect experiment). At low temperature a superconductor-insulator transition is realized. (Right) Phase diagram of the LaAlO$_3$/SrTiO$_3$ interface. The transition temperature to the superconducting state ($T_{\text{BKT}}$) can be continuously tuned from 0 to $\approx 300$ mK. At low temperature a quantum critical point (QCP) is evidenced. From [18].

diagram of cuprate superconductors. The presence of a quantum phase transition is another hallmark of the LaAlO$_3$/SrTiO$_3$ system and triggered many studies [17, 19–21]. These results were rapidly reproduced by Bell et al. [22] who showed that the quantum phase transition is accompanied by a sharp increase in the electron mobility of the system.

The thickness of the conducting layer at the LaAlO$_3$/SrTiO$_3$ interface was also studied. Following the work initiated by Basletic et al., Copie et al. found a remarkable temperature dependence of the thickness of the 2DEG as a function of temperature. Figure 1.6 shows the lateral map of the resistance they recorded at low and high temperature. They estimated its width to $\approx 12$ nm at 8 K and to $\approx 4$ nm (a value probably limited by the resolution of the tip) at room temperature [23]. Similar estimations of the electron gas thickness were obtained using infrared ellipsometry [24], photoelectron spectroscopy [25] and anisotropy of the superconducting transport properties [26].

An ingredient that we have not introduced yet, but that plays a key role on the physics of the LaAlO$_3$/SrTiO$_3$ interface, is the presence of a strong Rashba spin-orbit interaction [27, 28]. It is a consequence of the strong confining potential, which, by breaking the inversion symmetry induces a spin-splitting of the electronic states away from the $\Gamma$ point (with the help of the bulk atomic spin-orbit of SrTiO$_3$, see chapter 2). As a large portion of this thesis is dedicated to its study, we will not enter into the detail here. Yet, let us just mention that spin-orbit coupling is at the origin of interesting phenomena at the LaAlO$_3$/SrTiO$_3$ interface such as weak-antilocalization, complex in-plane magnetotransport and most probably a non-trivial pattern of Shubnikov-de Haas
1.2 The LaAlO$_3$/SrTiO$_3$ interface

![Figure 1.6: Thickness of the conducting layer](image)

Room temperature (left) and low temperature (right) map of the resistance across the LaAlO$_3$/SrTiO$_3$ interface (see fig. 1.2). At 300 K, due to the tip size, only an upper bound for the thickness of the conducting layer can be given (4 nm). At 8 K they 2DEG is estimated to be 10 nm thick. From [23].

oscillations.

In addition to all the investigations on the physics of this fascinating system, research groups have also been trying to optimize the electronic mobility at the LaAlO$_3$/SrTiO$_3$ interface. A significant enhancement of the mobility was achieved, in different ways, by our group [29] and by Huijben et al. [30]. We will describe in chapter 3 the way we proceeded. Concerning the Twente group, they capped their LaAlO$_3$/SrTiO$_3$ heterostructures with 1-3 uc of SrCuO$_2$, and by 2 uc of SrTiO$_3$. In both cases, the enhanced electron mobility was accompanied by a net decrease in the sheet carrier density and a lowering(or absence) of superconducting transition temperature. As we shall see in chapter 5, the analysis of the quantum oscillations reveals a 2D character and points to a strong Rashba spin-orbit interaction.

Many other exciting developments of the LaAlO$_3$/SrTiO$_3$ platform are currently pursued having in mind the idea of coupling the very rich physics of this system with mesoscopic phenomena. For example, the possibility to create “on-demand” very thin conducting channels at an (insulating) 3 uc LaAlO$_3$/SrTiO$_3$ interface, by applying a bias voltage between an interfacial metallic contact and the conducting tip of an atomic force microscope [31]. Also, recently, spin-polarized electrons have been successfully injected at the interface by the Paris group [32], a study that paves the way to investigations of the spin transport in this complex system. Following the success of strain-engineering in semi-conductors, the effect of growing LaAlO$_3$ on top of SrTiO$_3$ grown on top of lattice mismatched substrates is currently explored [33]. Above room temperature, field effect transistors made of LaAlO$_3$/SrTiO$_3$ heterostructures (using LaAlO$_3$ as gate dielectric) were operated up to 100 °C [34]. Finally, the integration of the LaAlO$_3$/SrTiO$_3$ system on Si has been proven to be possible [35].
1. INTRODUCTION

In this thesis, we focused on the magnetotransport properties of the LaAlO$_3$/SrTiO$_3$ interface. In chapter 2, we present band structure calculations, as they are important to be able to interpret the magnetotransport data. In chapter 3, we detail the growth procedure and the techniques we use to characterize our samples. Chapter 4 is devoted to the magnetotransport properties of our “standard” samples. Finally in chapter 5 we study the remarkable quantum oscillations that are observed in samples where the electronic mobility has been optimized. By tuning the carrier concentration by gate field, a complex fan diagram is brought into light.
CHAPTER 2

BAND STRUCTURE

Science never solves a problem without creating ten more.

George Bernard Shaw

2.1 Introduction

In this chapter, we investigate the origin of the electron gas and a model for the band structure of the LaAlO$_3$/SrTiO$_3$ interface. As we shall see, solving the quantum confinement problem is a complicated issue because it mixes the dielectric and electronic properties of SrTiO$_3$.

2.2 Origin of the conduction at the LaAlO$_3$/SrTiO$_3$ interface

In order to calculate the electronic band structure of the LaAlO$_3$/SrTiO$_3$ system, an assumption has to be made concerning the origin of the charges at the interface. In the following section, we discuss the different scenarios accounting for the presence of these charge carriers.

2.2.1 The polar discontinuity problem

At the center of many considerations on the origin of the mobile charge at the LaAlO$_3$/SrTiO$_3$ interface there is the polar discontinuity occurring along the [0 0 1] direction.

In the following discussion, we use an ionic picture to describe the charge distribution in the perovskite unit cell of LaAlO$_3$ and SrTiO$_3$. As shown in fig. 2.1, while the two types of planes that SrTiO$_3$ displays in the [0 0 1] direction are neutral (SrO$^0$ and TiO$_2^0$), this is not the case for the planes of LaAlO$_3$ ((LaO)$^{+1}$ and (AlO)$_2^{-1}$). Thus, an internal electric field is expected within the unit cell of LaAlO$_3$. 
2. BAND STRUCTURE

![Diagram of SrTiO3 and LaAlO3 with charge distribution](image)

Figure 2.1: Charge distribution inside the perovskite unit cell of SrTiO$_3$ and LaAlO$_3$. Using an ionic model the charge of the planes oriented along the [0 0 1] direction can be determined. In this direction SrTiO$_3$ and LaAlO$_3$ present respectively non-polar and polar atomic planes. Q is the charge per atomic plane per unit cell.

As a matter of fact, this observation is far from being anodyne for anyone interested in growing ideal (sharp) interfaces with LaAlO$_3$. Indeed, already back in the ’70s many studies addressed the question of the surface stability of polar termination since the electrostatic energy of the polar surface of a macroscopic crystal is infinite (see for example [36]). It was found that the only way such surfaces can be stabilized is by introducing some structural reconstruction, faceting or by adsorption of additional charges, effects that neutralize the surface polarization.

In fig. 2.2, we show that the very same problem is likely to appear in the LaAlO$_3$/SrTiO$_3$ heterostructure. Indeed, when growing just a few unit cells (uc) of LaAlO$_3$ on top of a (0 0 1)-oriented SrTiO$_3$ substrate a difference of several volts between the top surface and the interfacial layer is attained. From the electrostatic point of view, one can assimilate the entire heterostructure as a stacking of parallel plate capacitors, which are not charged in SrTiO$_3$ but loaded with +1/−1 electron charge per uc in LaAlO$_3$. Integrating the electric field along the out-of-plane direction indeed leads to a potential $V$ which increases with the film thickness.

Thus, we expect the perfect arrangement of fig. 2.2 to become energetically unfavorable as the number of layers of LaAlO$_3$ is increased. The system can solve the polar discontinuity problem either by different structural rearrangements or by an electronic reconstruction. In this case, due to the presence of Ti, a transition metal which can change its valence from +4 to +3, electrons can be injected at the interface.

2.2.2 The (intrinsic) electronic reconstruction scenario

Since its proposition by Nakagawa and coworkers [10], the electronic reconstruction scenario has received increasing attention due to its ability to explain several important experimental observations.
2.2 Origin of the conduction at the LaAlO$_3$/SrTiO$_3$ interface

Figure 2.2: Simple electrostatic analysis of the LaAlO$_3$/SrTiO$_3$ interface. The LaAlO$_3$/SrTiO$_3$ system can be assimilated to a stacking of parallel plate capacitors. Integrating the resulting electric field along the out-of-plane direction leads to a diverging potential as a function of film thickness.

Figure 2.3: Electronic reconstruction scenario. Left: simplified band structure of the LaAlO$_3$/SrTiO$_3$ heterostructure. The presence of an electric field in the film shifts the LaAlO$_3$ valence band (VB) to higher energy, eventually reaching the energy of the conduction band (CB) of SrTiO$_3$. Thanks to the accessible +3 valence of the Ti atoms, a Zener breakdown occurs. Right: the transfer of half a charge per uc at the LaAlO$_3$/SrTiO$_3$ interface from the top layer of LaAlO$_3$ cancels the potential increase observed in the non-reconstructed interfaces.
2. BAND STRUCTURE

In Figure 2.3, we sketch the idea for a heterostructure composed of $n$ unit-cells of LaAlO$_3$ grown on TiO$_2$-terminated SrTiO$_3$. Following Cancellieri et al. [37], we present the band structure of the interface (left part of fig. 2.3). We see that, due to the strong electric field inside the film, the valence band of LaAlO$_3$ is tilted. As the number of layers $n$ increases, the valence band of the surface layer of LaAlO$_3$ reaches the energy level of the conduction band of SrTiO$_3$: at this point a Zener breakdown occurs (critical thickness). As a consequence, electrons are transferred from the top surface of the heterostructure to the interfacial Ti (i.e. some interfacial Ti ions are changing their valence from +4 to +3), and holes are left on the top surface. This electron transfer creates a dipole that counterbalances the increase of the potential inside the LaAlO$_3$ layer.

Above the critical thickness, the transfer of electrons occurs progressively, as only the amount of charge needed to keep the valence band of LaAlO$_3$ at the conduction band level of SrTiO$_3$ is transferred. In this model, only for thick LaAlO$_3$ films is the internal potential completely canceled: the transfer is then of half an electron per uc from the LaAlO$_3$ surface to the interface (right part of fig. 2.3).

Interestingly, at least four predictions can be made from the electronic reconstruction scenario:

1. In the case of LaAlO$_3$ grown on SrO terminated SrTiO$_3$ holes need to be transferred at the interface to counterbalance the potential inside LaAlO$_3$. Since Ti cannot have a valence higher than +4, these holes are supposed to be welcomed by the O atoms. Yet, the ionization energy of the oxygen atoms is twice the one of the Ti atoms. Other reconstruction mechanisms than charge transfer may thus prevail for this type of heterostructure.

2. By modelling the stacking of LaAlO$_3$ planes as parallel plate capacitors in series and taking the dielectric constant of LaAlO$_3$ to be $\epsilon_r = 25$ one can calculate that the LaAlO$_3$ valence band is shifted by $\approx 1$ V per uc. Thus one expects Zener breakdown for films thicker than 3 uc only (the SrTiO$_3$ band gap is 3.2 eV).

3. The electronic density transferred to the interface is $3.3 \times 10^{14}$ cm$^{-2}$ when the electric field in the film is completely compensated.

4. If the number of LaAlO$_3$ uc is lower than 4, an electric field should be present in the layer.

As mentioned above, these predictions have been confirmed experimentally. For example, in their seminal paper [4] Ohtomo and Hwang showed that the interface created by depositing LaAlO$_3$ on SrO terminated SrTiO$_3$ is insulating. Likewise, Thiel et al. demonstrated that only films with thickness larger than 3 uc lead to a conducting interface [11]. These observations were then reproduced by many other research laboratories around the world, including ours.

Concerning the carrier density, analysis of the Hall effect measurements yields values typically in the range of $2 - 5 \times 10^{13}$ cm$^{-2}$ at low temperature. This value is notably lower than the prediction made by the electronic reconstruction scenario, and triggered many studies aiming to explain this difference. We can already mention
2.2 Origin of the conduction at the LaAlO$_3$/SrTiO$_3$ interface

![Figure 2.4](image)

Figure 2.4: LaAlO$_3$ a- and c-axis parameters plotted as a function of the film thickness. The solid lines are a guide to the eye. The dashed horizontal line indicates the pseudocubic LaAlO$_3$ bulk lattice constant. The inset shows in detail the expansion of the c axis for low LAO thicknesses.

Two points relevant to this discussion. As we shall see in the next chapters, the estimation of the carrier density from the Hall data is not straightforward in the case of a multi-band system and transport experiments only count mobile charges. Secondly, other experimental probes (photoemission spectroscopy [25, 38] and local magnetic susceptibility [16]) indicate the presence of localized interfacial charges: their estimate of the total carrier density is in agreement with the electronic reconstruction scenario.

The presence of an internal field in the LaAlO$_3$ layers has also been questioned. Using core-level photoemission spectroscopy Segal et al. [39] found no energy shift of the atomic levels of La and Al as a function of film thickness (2-9 uc) for LaAlO$_3$ grown on TiO$_2$ or SrO terminated SrTiO$_3$. On their side Takizawa et al. [38] found an electric field roughly 10 times smaller than the one predicted by the polar catastrophe scenario, but having the same orientation for LaAlO$_3$ grown on the two substrate terminations. Carrying out tunneling experiments across the LaAlO$_3$ barrier Singh-Bhalla et al. [40] found a very low built-in electric field (80 meV Å$^{-1}$). In this last experiment, however, the presence of a metallic electrode on top of LaAlO$_3$ probably modifies the boundary conditions of the system compared to the pure LaAlO$_3$/SrTiO$_3$ interface. Finally, using surface x-ray diffraction, Pauli et al. [41] showed that a buckling of the atomic planes exists in 2 unit cells thick LaAlO$_3$ films and is reduced as the number of layers is increased.

On our side, we investigated electrostriction of the LaAlO$_3$ film [37] as a probe of the electric field. Indeed, the presence of a strong electric field is expected to stretch the LaAlO$_3$ film, elongating its c-axis. Figure 2.4 shows that, indeed, for ultra-thin films (<6uc, see Inset), the c-axis of the LaAlO$_3$ layers is significantly larger than the value one would infer from a purely elastic deformation due to the epitaxial strain. Above 5 uc, the electron transfer reduces the electric field and the c-axis shrinks to the elastic
2. BAND STRUCTURE

limit (3.74 Å) given by the in-plane strain of the substrate. Above 20 uc the c-axis and the a-axis come back to their bulk value, a strong indication of strain relaxation.

Combining these data with ab initio calculations, we could show that electrostriction reproduces the c-axis expansion of our films before the electronic reconstruction. We mention also another explanation that can describe the data: the presence of 2 uc of LaAlO$_3$ intermixed at 50% with SrTiO$_3$ at the interface leads to a c-axis behavior close to the one observed. Intermixing of the interfacial layers has been observed using surface X-ray diffraction [41, 42] and TEM [10]. According to the electrostriction scenario, however, the observation of a c-axis value for $n > 5$ equal to the elastic limit suggests that the electric field inside the LaAlO$_3$ layer has been strongly reduced by the interface charges.

2.2.3 The (extrinsic) intermixing scenario

In fact, atomic reconstruction at the LaAlO$_3$/SrTiO$_3$ interface has been proposed to be responsible for the observed metallicity in the system. In this scenario, due to the polar discontinuity problem, the interfacial atoms of the LaAlO$_3$ and SrTiO$_3$ planes mix, resulting in a rougher interface as observed by TEM and X-ray diffraction. Metallicity in the system is then conceivable based on the fact that bulk La-doped SrTiO$_3$ is conducting.

In a study to probe the different scenarios, we artificially altered the composition of all the LaAlO$_3$ layers by mixing them with SrTiO$_3$ [43] (Sr$\leftrightarrow$La, Ti$\leftrightarrow$Al). We obtained two main results. First, in agreement with the fact that in this case the formal charge of the layers is lower than $+1/-1$ and in agreement with the electronic reconstruction scenario, the onset of conductance was shifted to higher values of film thicknesses. We note that such a precise dependence with film thickness and composition is hardly reconcilable with the intermixing scenario. Secondly, the intermixed films were insulating for all the concentration ratios, showing that this type of intermixing is probably not at the origin of the observed conduction in standard LaAlO$_3$/SrTiO$_3$ interfaces.

Other mechanisms have been proposed to explain the origin of the conductance at the interface between these two wide band gap insulators. Often these propositions, however, overlook the polar discontinuity problem: additional charges at the interface and at the surface (or a charge transfer) are (is) needed to solve the polar catastrophe problem.

2.2.4 The (extrinsic) oxygen vacancies scenario

SrTiO$_3$ is a material that undergoes a metal/insulator transition at a remarkably low carrier density $\approx 1 \times 10^{16}$ cm$^{-3}$ [44]. This implies that a slight deviation from stoichiometry that would create an oxygen vacancy every 150 unit cells, and hence release two electrons in the conduction band of SrTiO$_3$, is enough to turn the crystal conducting. Since in general the growth of the LaAlO$_3$ layers is performed at high temperature and in a rather reducing oxygen pressure, a contribution from oxygen vacancies (O-vacancies) to the total carrier density cannot be excluded.

As explained in chapter 3, an efficient way to minimize the importance of this extrinsic doping is after the growth to anneal the interfaces at high temperature (530°C)
in a high oxygen pressure (0.2 bar) for at least 1 hour. In a previous study [45], we showed that using this annealing procedure it is possible to obtain samples with very similar transport properties despite being grown in oxygen pressures ranging from $1 \times 10^{-6}$ mbar to $1 \times 10^{-2}$ mbar. As samples grown at $1 \times 10^{-6}$ mbar without annealing have a large density of O-vacancies and display 3D conductivity, this result indicates that oxygen annealing is efficient in refilling the O-vacancies.

### 2.2.5 Oxygen vacancies in LaAlO$_3$

As explained above, in the case of LaAlO$_3$ on TiO$_2$ terminated SrTiO$_3$, the electronic reconstruction scenario proposed by Nakagawa and coworkers requires the ionization of some of the oxygen atoms belonging to the uppermost AlO$_2$ layer. The occurrence of this effect has to be energetically compared to other mechanisms. One such other process is the creation of O-vacancies on the top surface of LaAlO$_3$. Indeed, according to recent studies [37, 46–49], the formation energy of an O-vacancy on the top surface of the LaAlO$_3$ film is strongly reduced in the LaAlO$_3$/SrTiO$_3$ system, due to the electric field in the LaAlO$_3$ layer. In this scenario, the electrons liberated by the O vacancies are transferred to the interface, solving the polar discontinuity problem (the O vacancies create in-gap states located just below the LaAlO$_3$ conduction band). A critical thickness also exist in this framework due to the balance between the energy needed to create an O vacancy and the energy gained by transferring electrons at the interface. In addition to that, Bristowe et al. proposed that, for ultra-thin films, the presence of the vacancies only a few unit cells above the SrTiO$_3$ surface may localize the transferred carriers. Hence, according to these authors, a real insulator to metal transition occurs as the LaAlO$_3$ thickness is increased.

### 2.3 Band structure calculations

In this section we describe a model for the conduction band of SrTiO$_3$ when electrons are injected at its surface according to the electronic reconstruction scenario.

#### 2.3.1 Bulk SrTiO$_3$ without spin-orbit interaction

##### 2.3.1.1 Elements of crystal field theory

SrTiO$_3$ has a very rich physics which has been studied since the ’60s: it is a band insulator with a large gap ($E_{\text{gap}} = 3.2$ eV) separating a conduction band, dominated by the $3d$ orbitals of the Ti atoms ($d_{xy}, d_{yz}, d_{xz}, d_{3z^2-r^2}$ and $d_{x^2-y^2}$), from a valence band, dominated by the $2p$ orbitals of the O atoms ($p_x, p_y, p_z$) [50, 51]. As a consequence, some of the properties of the conduction band of SrTiO$_3$ can be discussed looking at the simpler model of isolated Ti atoms in the crystal field. In this perspective, an interesting tool is group theory, because, by labeling the atomic orbitals according to their symmetry properties and taking into account the symmetry of the crystal field, this theory can make predictions on the degree of orbital degeneracy of the system. Indeed a central result of this theory is that orbitals belonging to the same irreducible representation (hence having the same label) are degenerate. The amplitudes and
2. BAND STRUCTURE

![Diagram showing band structure](image)

Figure 2.5: Energy of the Ti $3d$ orbitals in the presence of crystal field. Energy levels of the Ti $3d$ orbitals in a spherical, cubic, tetragonal and non inversion-symmetric tetragonal environment. The splittings were determined from experimental and theoretical studies [50–55]. The green labels refer to the irreducible representation to which the orbital(s) belong(s).

Signs of the splittings, however, have to be determined from numerical calculations or experimental data, as they depend on the specific crystal distortion under study.

Figure 2.5 shows an analysis on the $3d$ energy levels of Ti in different environments, using group theory as well as experimental and numerical data [50–55]. We omit the effect of atomic spin-orbit coupling in this analysis as it will be treated in section 2.3.3. In the following description, we also omit the spin state of each energy level. We see that in the absence of any crystal field (spherical symmetry) the $3d$ orbitals of Ti are degenerate. Indeed, they belong to the same irreducible representation of the continuous group of rotations labeled by the angular momentum $L=2$. Going from the spherical symmetry to the cubic symmetry ($O_h$ point group) this quintuplet of states breaks into a triplet (labeled $T_{2g}$) and a doublet (labeled $E_g$). It is the oxygen octahedron surrounding the Ti atom in the perovskite structure that is at the origin of this splitting: the triplet, composed of orbitals with lobes oriented at $45^\circ$ with respect to the O atoms ($d_{xy}$, $d_{yz}$ and $d_{xz}$ orbitals) lies approximately $2$ eV lower in energy compared to the doublet ($d_{3z^2-r^2}$ and $d_{x^2-y^2}$ orbitals) with lobes pointing towards the oxygen atoms. As a consequence, the bottom of the conduction band of SrTiO$_3$ is composed by the $3d - T_{2g}$ states of Ti.

Below 105 K, SrTiO$_3$ undergoes a structural transition from cubic to tetragonal ($D_{4h}$ point group) with a rotation of the oxygen octahedra accompanied by a small extension of the c-axis (at 4 K $c/a \approx 1.001$) [54, 56, 57]. From the point of view of group theory, this implies that both the $E_g$ and the $T_{2g}$ states break into two new groups of states. This transition slightly handicaps the planar orbitals, shifting the $d_{xy}$ state $\Delta_T \approx 3$ meV higher in energy relative to the $d_{yz}$ and $d_{xz}$ orbitals (and similarly for the $E_g$ doublet) [54].

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2.3 Band structure calculations

Figure 2.6: Dominant contributions to Ti-$d_{xy}$ to Ti-$d_{xy}$ and Ti-$d_{xz}$ to Ti-$d_{xz}$ hopping in the xy-plane of SrTiO$_3$. Left: hopping between two Ti-$xy$ orbitals through an O-$p_y$ (\(\hat{y}\)-direction) or O-$p_x$ orbital (\(\hat{x}\)-direction). Right: hopping in the \(\hat{x}\)-direction between two Ti-$xz$ orbitals through an O-$p_z$ orbital. In the nearest neighbor approximation, the hopping between Ti-$xz$ orbitals vanishes along the \(\hat{y}\)-direction. By symmetry, similar conclusion can be obtained for hopping between Ti-$yz$ orbitals. This picture was strongly inspired by [58].

2.3.1.2 Tight binding model of bulk SrTiO$_3$

Moving from this crystal field picture to a description of the conduction band bottom of SrTiO$_3$ can be efficiently achieved using a tight binding model (see for example [58–60]). Figure 2.6 shows the relevant hopping processes in the xy-plane, in the nearest neighbor approximation. For the model, we consider only the \(T_{2g}\) states of Ti as these orbitals lie much lower in energy than the \(E_g\) states. We see that \(d_{xy}\) to \(d_{xy}\) hopping proceeds through an O-$p_y$ along the \(\hat{x}\)-direction and through an O-$p_x$ along the \(\hat{y}\) direction (\(t\)). For the hopping between \(d_{xz}(d_{yz})\) orbitals, the O-$p_z$ orbital is involved in the \(\hat{x}(\hat{y})\)-direction while in the \(\hat{y}(\hat{x})\)-direction symmetry considerations in the nearest neighbor approximation predict a vanishing hopping. Higher order approximations restore a hopping in these directions albeit much smaller (\(t'\)). These considerations lead to the following tight binding Hamiltonian (in the cubic phase):

\[
\hat{H}_{0}^{TB} = \begin{pmatrix}
-2t' & 0 & 0 \\
0 & -2t & 0 \\
0 & 0 & -2t'
\end{pmatrix} \cos(k_z a) + \begin{pmatrix}
-2t & 0 & 0 \\
0 & -2t' & 0 \\
0 & 0 & -2t
\end{pmatrix} \cos(k_y a) \\
+ \begin{pmatrix}
-2t & 0 & 0 \\
0 & -2t & 0 \\
0 & 0 & -2t'
\end{pmatrix} \cos(k_x a)
\]

\[
\psi_k = \begin{pmatrix}
\psi_{yz}^k \\
\psi_{xz}^k \\
\psi_{xy}^k
\end{pmatrix}
\]

with \(t = \frac{t_{pd}^2}{\Delta_{pd}}\) describing the hopping through the oxygen states (\(\Delta_{pd} \approx 3\) eV is the energy gap between the O-$p$ and the Ti-$d$ states and \(t_{pd}\) is the virtual hopping between the titanium and oxygen states) and \(a\) the SrTiO$_3$ lattice parameter. The eigenvector \(\psi_k\) is composed by three Bloch functions with the same symmetry (at the \(\Gamma\) point) as the atomic orbitals they are built from (\(yz, xz\) and \(xy\)). Hereafter we call the associated energy bands, respectively, the \(yz, xz\)- and \(xy\)-band.
2. BAND STRUCTURE

Figure 2.7: Conduction band bottom of tetragonal bulk SrTiO$_3$ in the absence of spin-orbit coupling. Dispersion of the yz-,xz- and xy bands along the $\Gamma - X$ and $\Gamma - Y$ directions in our tight binding model. We used $m^*_l = 0.7m_e$, $m^*_h = 7.5m_e$ and $\Delta_T = 3$ meV. The energy scale has been shifted for clarity.

We have seen that the effect of the 105 K tetragonal distortion of SrTiO$_3$ is principally to lift the degeneracy between the $T_{2g}$ orbitals, shifting the $d_{xy}$ state $\Delta_T$ higher in energy. This can be included in the present tight binding model by adding the following contribution to the Hamiltonian of the system :

$$\hat{H}_{\Gamma}^{TB} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \Delta_T \end{pmatrix}$$ (2.2)

Close to the Brillouin zone center, theoretical and experimental estimations of the parameters relevant to the description of the conduction band bottom of SrTiO$_3$ have been proposed [53, 57, 60–62]. In the absence of spin-orbit, they agree on the fact that each band has two effective masses : one light ($m^*_l = 0.4 - 1.2m_e$) linked to the hopping parameter $t$ and one heavy ($m^*_h = 6 - 14m_e$) related to hopping parameter $t'$ ($t = \frac{\hbar^2}{2a^2m^*_l}$, $t' = \frac{\hbar^2}{2a^2m^*_h}$). For example, the yz-band is light in the $\hat{y}$ and $\hat{z}$ direction and heavy along the $\hat{x}$ direction. Concerning the tetragonal distortion $\Delta_T$, recent estimations suggest a splitting of around 3 meV [54]. Figure 2.7, shows the bottom of the conduction band of SrTiO$_3$ along the $\Gamma - X$ and $\Gamma - Y$ directions, derived from the above model.

2.3.2 The LaAlO$_3$/SrTiO$_3$ interface without spin-orbit interaction

2.3.2.1 Elements of crystal field theory

Coming back briefly to the crystal field picture presented in fig. 2.5, we observe that reducing further the symmetry of the system by creating an interface (i.e. moving to the point group $D_{4h}$ to $C_{4v}$) doesn’t lead to additional lifting of degeneracies. Yet, because of the confining potential thereby created, we are going to see that the sign and the
2.3 Band structure calculations

\[ \varepsilon_r(E) \]

\[ V_{\text{bottom}} = 0 \]

\[ D_{\text{top}} = |e| n_{2D} \hat{z} \]

Figure 2.8: Geometry and boundary conditions used in the Poisson-Schrödinger model. Progressive modelization of the system. Left, the LaAlO$_3$/SrTiO$_3$ heterostructure. Middle, a material with an electric field dependent dielectric constant $\varepsilon_r(E)$ and boundary conditions $D_{\text{top}} = |e| n_{2D} \hat{z}$ and $V_{\text{bottom}} = 0$. Right, resolution of the Poisson and the Schrödinger equation only along the out-of-plane direction (in-plane the system is supposed homogeneous and in-plane parabolic band dispersion is assumed).

Amplitude of the splittings between the in-plane and out-of-plane orbitals within the $T_{2g}$ and $E_g$ groups are completely modified. Experimentally, an average splitting of 50 meV between the $d_{xy}$ and the $d_{xz}/d_{yz}$ states and 100 meV for the $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ states was estimated by Salluzzo et al. using X-ray absorption spectroscopy [55].

2.3.2.2 The Poisson-Schrödinger model

Modelization of the heterostructure Starting from the above considerations for the band structure of SrTiO$_3$, we try to model a quantum confined electron gas at the LaAlO$_3$/SrTiO$_3$ interface. We follow the theoretical approach proposed by Stengel [59]. A sketch of the geometry of the model is given in fig. 2.8.

First, we consider that the LaAlO$_3$ layer fixes the value of the displacement field at the interface: $D_{\text{top}} = |e| n_{2D} \hat{z}$, with $n_{2D}$ the 2D density of carriers injected at the interface. The energy of the conduction band at the bottom side of SrTiO$_3$ is set to zero. The thickness of the substrate varies between 110 and 410 nm depending on the charge extension in each particular simulation. These values are much larger than the extension of the electron gas that is estimated from experiments to be 10 nm at low temperature.

We make use of the effective mass and envelope function approximations. This means that the wave functions attributed to carriers belonging to the sub-band $n$ generated by the quantum confinement of the bulk band $\alpha$ can be factorized as follows:

\[ \psi_{n,k}^{\alpha}(r) = \frac{e^{i k_{||} r_{||}}}{2\pi} \xi_n^{\alpha}(z) \]

(2.3)

with $k_{||} = (k_x, k_y)$, $r_{||} = (x, y)$ and $\xi_n^{\alpha}(z)$ an envelope function that modulates the amplitude of the plane wave along the $z$ direction. The energy of this state is simply:

\[ E_n^{\alpha}(k_{||}) = \frac{\hbar^2 k_x^2}{2 m_{x,\alpha}^*} + \frac{\hbar^2 k_y^2}{2 m_{y,\alpha}^*} + \varepsilon_n^{\alpha} \]

(2.4)

with $m_{i,\alpha}^*$ the bulk band masses and $\varepsilon_n^{\alpha}$ the solution of the following 1D-Schrödinger
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\[ -\frac{\hbar^2}{2m^*_\alpha z^2} \frac{\partial^2 \xi^\alpha_n(z)}{\partial z^2} + eV(z)\xi^\alpha_n(z) + E^\alpha_{0} \xi^\alpha_n(z) = \epsilon^\alpha_n \xi^\alpha_n(z) \]  

(2.5)

with \( V(z) \) the confining potential and \( E^\alpha_{0} \) the energy of the bottom of the band \( \alpha \) in the bulk. Stengel [59] showed that, due to the strong interfacial electric field, the band-widths of the bulk bands are modified (by up to 30\%). In our case, we could probably include this effect in our calculation by allowing an electric field dependence of \( m^*_x, m^*_y, \) and \( m^*_z. \) Unfortunately, a reliable estimation of the variation of these parameters implies \textit{ab initio} calculations that are out of our skills.

Under these assumptions the profile of the 3D charge density is:

\[ \rho_{3D}(z) = e \sum_{n,\alpha} \Theta(E_F - \epsilon^\alpha_n) \sqrt{\frac{m^*_x m^*_y}{\pi\hbar^2}} (E_F - \epsilon^\alpha_n) |\xi^\alpha_n(z)|^2 \]  

(2.6)

with \( \Theta \) the heaviside function (our calculation is for \( T = 0 \) K). \( E_F \) is chosen to obtain a given sheet carrier density \( n_{2D} \):

\[ n_{2D} = \sum_{n,\alpha} \Theta(E_F - \epsilon^\alpha_n) \sqrt{\frac{m^*_x m^*_y}{\pi\hbar^2}} (E_F - \epsilon^\alpha_n) \]  

(2.7)

The 3D carrier density is linked to the confining potential \( V(z) \) via Poisson’s equation:

\[ -\frac{\partial}{\partial z} \left( \epsilon_0 \epsilon_r(E) \frac{\partial V(z)}{\partial z} \right) = \rho_{3D}(z) \]  

(2.8)

with \( \epsilon_r(E) \) the electric field dependent relative dielectric constant.

Thus, eq. (2.5) and eq. (2.8) have to be solved self-consistently.

**Self-consistency** Figure 2.9 shows schematically how these two equations can be solved in a consistent manner. We arbitrarily choose to start with a triangular potential well, representing a constant electric field inside SrTiO\(_3\) (of typically 5 MV m\(^{-1}\)). Then the sequence of energy levels in the potential well is calculated for each bulk band \( \alpha \). With this information \( \rho_{3D} \) is computed and used to find a new potential well that fits the charge distribution. At this stage, the new potential well and the initial one are compared via:

\[ \delta_V = \frac{1}{L} \int_0^L \left( \frac{V_i(z) - V_i^{in}(z)}{V_i(z)} \right)^2 dz \]  

(2.9)

with \( L \) the thickness of the simulated SrTiO\(_3\). At the first iteration, \( V_i^{in}(z) = V_0(z) \), but, for \( i > 1 \), it is a linear combination of \( V_{i-1}(z) \) and \( V_{i-1}^{in}(z) \). If the computed difference is larger than a critical difference \( \delta_V \) (typically set to \( \delta_V \approx 10^{-5} \)) then the new potential is re-injected in the Schrödinger equation. The process is repeated until \( \delta_V \) becomes smaller than \( \delta_V \).

To ensure convergence of the algorithm, the \textit{over-relaxation} method was used (see for example [63]). This method uses the following combination of the results of the \((i - 1)\)th iteration as input for the new round:

\[ V_i^{in}(z) = \zeta V_{i-1}(z) + (1 - \zeta)V_{i-1}^{in}(z) \]  

(2.10)
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Figure 2.9: Scheme for the self-consistent solving of the Poisson-Schrödinger system of equations. At step 0 a triangular potential well is chosen as an input for the Schrödinger equation. Its resolution allows the 3D charge distribution ($\rho_{3D}$) to be determined. $\rho_{3D}$ is then used to calculate a new potential via Poisson’s equation. The comparison between the initial and the new potential determines if the charge density has to be recomputed or not.

In our studies we set $\zeta = \min(0.4, 1000\delta_V)$ or simply $\zeta = 0.4$ for calculations for which reaching convergence was easy.

This approach is very useful since Schrödinger’s equation tends to delocalize the electrons while Poisson’s equation tends to accumulate them at the interface. As a consequence, injecting directly the result of one equation into the other would end in a “bouncing” sequence of solutions.

Dielectric constant Before moving to the results of the simulation, let us briefly comment on the dielectric constant of SrTiO$_3$ as eq. (2.8) states that it is an important quantity of the calculation.

SrTiO$_3$ is a quantum paraelectric: quantum fluctuations avoid the establishment of a ferroelectric order at low temperature. The dielectric properties manifest this proximity to a ferroelectric state: the dielectric constant is strongly temperature and electric field dependent. In figure 2.10, we show the temperature dependence of the dielectric constant of one of our SrTiO$_3$ single crystals. Impressively, at low temperature, the dielectric constant reaches $\epsilon_r = 21600$. Yet this value depends on the electric field applied to the material. We measured the voltage dependence of $\epsilon_r$ using capacitance measurements at 4 K. Due to relatively large thickness of our samples (0.5 mm) the maximum electric field we could apply was $E_{\text{max}} = 200 \text{ V/0.5 mm} = 400 \text{ kV m}^{-1}$. Our measurements are shown in fig. 2.11 together with the dependence proposed by Ueno et al. and Stengel [59, 64]. At low field, our dielectric constant agrees very well with the dependence chosen by Ueno et al.. At very high field, however, the extrapolation of our data seems to match Stengel’s estimation.

We fitted (gray dashed line in fig. 2.11) our low field measurements together with
Figure 2.10: Temperature evolution of the dielectric constant of SrTiO$_3$. The dielectric constant of our SrTiO$_3$ single crystals was determined using differential capacitance measurements. In section 3.5 we give a description of this technique.

Figure 2.11: Field dependence of the dielectric constant at 4 K. Dielectric constant measured for a SrTiO$_3$ single crystal (blue dots), and according to different references. The function used in our Poisson-Schrödinger simulation of the LaAlO$_3$/SrTiO$_3$ interface is shown as the dashed line.
the high field behavior calculated by Stengel. The following mathematical form was obtained and used for our low-temperature Poisson-Schrödinger simulations:

\[ \epsilon_r(E) = 1 + \frac{B}{[1 + (E/E_0)^2]^{1/3}} \]  

(2.11)

with \( B = 25462 \) and \( E_0 = 82213 \) V m\(^{-1}\). While this model predicts a dielectric constant always larger than 1 (it becomes equal to 1 only for infinite electric field), Ueno’s model yields \( \epsilon_r = 1 \) already at \( \approx 2 \) GV m\(^{-1}\) and then becomes even smaller than 1.

**Confinement for a perfect interface**  In the electronic reconstruction scenario all the charges transferred to the interface are mobile. But, according to first principle calculations [37, 65], the total charge of 0.5 electron/uc is transferred to the interface only for infinite LaAlO\(_3\) thickness. Thus, in our first calculations, we considered a sheet carrier density \( n_{2D} \) of 0.25 electron/uc. We compared the case of an electric field independent dielectric constant (\( \epsilon_r^{\text{cst}} = 350 \)) with the results obtained using the dielectric constant given in eq. (2.11). The first model is a simplification of the room temperature situation for which a dependence of the dielectric constant on the electric field probably exists as well; yet we were not able to observe it within our accessible electric fields (\( E_{\text{max}} = 400 \) kV m\(^{-1}\)). The second situation is a more realistic approximation of the low temperature behavior of SrTiO\(_3\).

Figures 2.12 and 2.13 show the result of our computations. In both cases, we observe that \( xy \) states having planar symmetry lie at the lowest energy and that a complex sequence of occupied sub-bands, which include also bands with \( yz/xz \) symmetry, is present. The two simulations differ substantially by their charge distribution along the vertical direction. Indeed, due to the lower polarizability of SrTiO\(_3\) for the case \( \epsilon_r^{\text{cst}} = 350 \), the first model predicts a more confined charge density. These observations are in line with published work [23, 59, 65–67]. Note that this argument applies for \( z > 5 \) nm where the confining potential is sufficiently shallow for the electric field dependent \( \epsilon_r(E) \) to be bigger than 350 (at \( z = 5 \) nm, \( E \approx 4 \) MV m\(^{-1}\)). A more realistic modelization of the room temperature dielectric constant would probably change this last result, by confining even more the charge distribution of fig. 2.12 at the interface.

As a consequence, while for the \( \epsilon_r^{\text{cst}} = 350 \) case, the 3D charge density reaches the critical density for the metal-insulator transition in bulk SrTiO\(_3\) (\( n_{3D}^{\text{min}} \approx 1 \times 10^{-5} \) nm\(^{-3}\)) at \( z \approx 40 \) nm, this value is shifted to 180 nm in the case of large and field dependent \( \epsilon_r(E) \). For consistency, we extended the thickness of our SrTiO\(_3\) to 410 nm for the second simulation.

Another striking feature that comes out of our calculations is the very small splitting between the sub-bands as their band bottom approaches the Fermi energy. This feature is common to other studies but has surprisingly received little attention in the past. The reason behind this “stacking” of sub-bands at the Fermi level can be explained by noting that the confining potential displayed in the middle panel of fig. 2.12 and 2.13 is filled to the very top, where it becomes almost flat. As a consequence, the occupied sub-bands with the highest energy are much less confined, hence the very low splitting
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Figure 2.12: Results of Poisson-Schrödinger simulation assuming a transferred in-plane carrier density of 0.25 e/uc and $\varepsilon_r^{\text{ext}} = 350$. Left panel: band structure of the interface with $1.6 \times 10^{14}$ cm$^{-2}$ carriers. Green: sub-bands with $xy$ symmetry. Red: sub-bands with $yz$ symmetry. Blue: sub-bands with $xz$ symmetry. Middle panel: confining potential (black) and 3D electron density (light blue). Right panel: sub-band bottom energy as a function of the average electric field felt by the carriers of a particular sub-band (see eq. (2.12)). The size of the simulated SrTiO$_3$ is 110 nm and $E_F = 0$.

Figure 2.13: Results of Poisson-Schrödinger simulation assuming a transferred in-plane carrier density of 0.25 e/uc and $\varepsilon_r(E)$ given by eq. (2.11). Left panel: band structure of the interface with $1.6 \times 10^{14}$ cm$^{-2}$ carriers. Green: sub-bands with $xy$ symmetry. Red: sub-bands with $yz$ symmetry. Blue: sub-bands with $xz$ symmetry. Middle panel: confining potential (black) and 3D electron density (light blue). Right panel: sub-band bottom energy as a function of the average electric field felt by the carriers of a particular sub-band (see eq. (2.12)). In this case, the size of the simulated SrTiO$_3$ had to be extended to 410 nm.
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between them. In turn, these states are responsible for the queue in the 3D carrier density \( n_{3D} \) that extends deep into the substrate at “low temperature”.

In order to quantify this effect, we estimated the average electric field “felt” by each sub-band independently via:

\[
E_{\text{field}}^{\text{av}} = \int_0^L |\xi_\alpha^a(z)|^2 \left( -\frac{\partial V}{\partial z} \right) \, dz
\]

The result is shown on the right panel of fig. 2.12 and 2.13. We see that \( E_{\text{field}}^{\text{av}} \) is smaller for sub-bands with lower binding energy, eventually reaching zero for sub-bands lying very close to the Fermi level. This implies that the very weakly filled sub-bands of our calculation are not well confined at the interface and hence may display 3D behavior.

From the experimental point of view, however, evidences of the confinement of weakly filled sub-bands at the LaAlO\(_3\)/SrTiO\(_3\) interface were brought into light. For example:

- 2D quantum oscillations of the resistance were evidenced by Caviglia et al. [29]. The analysis of the frequencies observed in that work showed that the area occupied by the electron responsible for the oscillations is only 0.1\% of the Brillouin zone.

- As we shall see in the next chapters, both in-plane magnetoresistance oscillations and Shubnikov-de Haas oscillations measurements seem to reveal the importance of a very weakly filled but strongly Rashba spin-orbit split sub-band [68]. Confinement is a prerequisite for Rashba spin-orbit interaction to emerge.

- The 2D character of superconductivity was demonstrated by Reyren et al. [26] and analyses of the superconducting state in parallel and perpendicular fields yield a conducting thickness of \( \approx 10 \text{ nm} \).

Coming back to the simulation of fig. 2.13, we emphasized that \( n_{3D} \) is reduced below the metal insulator transition of bulk SrTiO\(_3\) for \( z > d = 180 \text{ nm} \). This suggests that the threshold 2D carrier density for conductance in the LaAlO\(_3\)/SrTiO\(_3\) system should be roughly \( d \times n_{3D}^{\text{min}} \approx 2 \times 10^{11} \text{ cm}^{-2} \). This is a rather low value if we compare with the typical 2D carrier density at which the metal-insulator transition is experimentally observed at the LaAlO\(_3\)/SrTiO\(_3\) interface. Liao et al., for example, found that the metal to insulator transition takes place at \( n_{2D}^{\text{min,LAO/STO}} \approx 8 \times 10^{12} \text{ cm}^{-2} \) [69].

Interestingly, if now we perform the reverse estimation, assuming a thickness of \( \approx 10 \text{ nm} \) for the electron gas, the estimation of Liao et al. translates to a critical 3D carrier density equal to \( n_{3D}^{\text{min,LAO/STO}} \approx 0.01 \text{ nm}^{-3} \). Nicely, this value is comparable with the magnitude of the charge density in fig. 2.13 around \( 10 - 20 \text{ nm} \).

To rationalize these arguments, we propose that, in the region of the interface, the metal-insulator transition takes place at a higher 3D carrier density, probably due to a higher degree of disorder compared to bulk SrTiO\(_3\). Finally, we note that integrating the area below \( n_{3D}^{\text{min,LAO/STO}} \) in fig. 2.13 yields an estimated localized carrier density of \( \approx 3 \times 10^{13} \text{ cm}^{-2} \), a quantity that represents a substantial portion of the electrons coming from the polar catastrophe.
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Figure 2.14: Band bending and carriers distribution in presence of in-gap states. Left: diagrammatic view of the conduction band of the LaAlO$_3$/SrTiO$_3$ interface in the presence of in-gap states (green dots). In the bulk of SrTiO$_3$ the Fermi energy (in red) lies below the in-gap states (in our model they are supposed to be empty). Approaching the interface, however, the band bending induced by the electronic charge transfer brings these levels below the Fermi level (only localized charges region). At the interface, the band bending is strong enough to also bring the conduction band below the Fermi level and a 2DEG is created. Right: in this model the 3D charge density is the sum of the contribution of the 2DEG (in blue) and of the localized charges (in green).
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This motivated us to consider a model of the interface in which a portion of the charge transferred to the interface ends up in localized states. Indeed, there is no reason why, as electrons accumulate at the interface some of them do not get trapped in defect states. In a band structure picture, it means that in addition to the states of the conduction band, in-gap states are also accessible to the transferred electrons. Figure 2.14 shows that in this case a depletion region is created. We note that in-gap states at the SrTiO$_3$ surface were experimentally observed [46, 70–72], but, to our knowledge, this is the first time that their impact on the band structure of the LaAlO$_3$/SrTiO$_3$ system is considered.

Adding localized states To include localized charges at the interface, we followed Stern et al. [73] and added to the free charge density ($\rho_{\text{free}}^{3D}$) a constant background charge of thickness $d$ (see fig. 2.14). In our picture, this background charge is caused by the filling of in-gap states by the polar catastrophe.

$$\rho_{\text{tot}}^{3D}(z) = \rho_{\text{free}}^{3D}(z) + \rho_{\text{loc}}^{3D}$$

(2.13)

We kept the same total carrier density

$$n_{\text{tot}}^{2D} = n_{\text{free}}^{2D} + n_{\text{loc}}^{2D} = 0.25/\text{uc}$$

and we set $n_{\text{free}}^{2D} = 0.1/\text{uc}$. For consistency $\frac{n_{\text{loc}}}{d} < n_{\text{min,LAO/STO}}^{3D}$, thus we set $d$ to 100 nm. We get:

$$\rho_{\text{loc}}^{3D} = \frac{0.15e}{a^2} \frac{\Theta(z)\Theta(d-z)}{d}$$

(2.14)

Due to our simplistic model $n_{\text{loc}}^{3D} \approx 0.01 \text{nm}^{-3}$ even tens of nanometers from the interface. Further refinement of our calculation could take into account a “$z$” dependent $n_{\text{min,LAO/STO}}^{3D}$. This would result in a non-rectangular distribution for the localized electrons and in turn to a larger admissible spread of the localized charges.

Figure 2.15 shows the result of our calculation using $\epsilon_r(E)$ as dielectric constant and $\rho_{\text{tot}}^{3D}(z)$ as charge density for the Poisson equation. Clearly, in this situation a strong confining electric field is “felt” by all the sub-bands and, as a consequence, the 2DEG is well confined at the interface. The electronic structure remains complex with several $xy$ sub-bands occupied as well as $xz/yz$ sub-bands.

We would like to stress that the band structure discussed here can still differ substantially from the one present at the LaAlO$_3$/SrTiO$_3$ interface. As we have seen, the electronic configuration depends on many parameters such as the special choice of the dielectric constant, the effective masses, the total number of carriers, and the localized charge density profile. In addition to that, contrarily to an ab initio calculation, our Poisson-Schrödinger simulation is far from taking into account all the subtleties of the LaAlO$_3$/SrTiO$_3$ system. Yet, this model has the advantage to be very flexible. Good examples are, for example, the ability to perform simulations of the band structure given different dielectric constants, sizes of the system or the presence of in-gap states.

2.3.3 Bulk SrTiO$_3$ with spin-orbit interaction

To simplify the calculation, the role of spin-orbit (SO) coupling in the system has been omitted in all the above analysis. Recently, many studies have reached the
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Figure 2.15: Results of Poisson-Schrödinger simulation assuming a total transferred in-plane carrier density of 0.25 e/uc, 0.15 e/uc of it being localized. \( \epsilon_r(E) \) is given by eq. (2.11). Left panel: band structure of the interface with a total of \( 1.6 \times 10^{14} \) cm\(^{-2} \) carriers, \( 9.8 \times 10^{13} \) cm\(^{-2} \) of them being localized. Green: sub-bands with \( xy \) symmetry. Red: sub-bands with \( yz \) symmetry. Blue: sub-bands with \( xz \) symmetry. Middle panel: confining potential (black) and 3D electron density (light blue). Right panel: sub-band bottom energy as a function of the average electric field felt by the carriers of a particular sub-band (see eq. (2.12)).

Conclusion that it is an essential ingredient to describe the physics at the \( \text{LaAlO}_3/\text{SrTiO}_3 \) interface [27, 28, 60, 68, 74, 75].

Spin-orbit coupling is an additional contribution to the Hamiltonian of the system that describes the interaction of the particle spin with the gradient of the electrostatic potential. It can be deduced from the non-relativistic limit of the Dirac equation and takes the general form:

\[
\hat{H}_{SO} = -\frac{\hbar}{4m_0^2c^2} \vec{\sigma} \cdot \vec{p} \times (\nabla U)
\]  

(2.15)

where \( c \) is the speed of light, \( \vec{\sigma} \) is the vector of Pauli matrices, \( \vec{p} \) is the linear momentum operator and \( \nabla U \) is the gradient of the electrostatic potential energy.

In the simple case of the hydrogen atom \( U = -\frac{e^2}{4\pi\varepsilon_0 r} \), \( \hat{H}_{so} \) takes the well known form:

\[
\hat{H}_{SO} = \frac{e^2}{8\pi m_0^2c^2\varepsilon_0 r^3} \vec{L} \cdot \vec{S}
\]

(2.16)

with \( \vec{L} = \vec{r} \times \vec{p} \) the orbital angular momentum operator.

The case of an electron in a crystal lattice is more complex, since the potential energy in eq. (2.15) is the potential energy of the lattice. The gradient of this potential, however, is expected to be relevant only close to the (heavy) atoms composing the crystal, where the atomic potential is a good approximation to the lattice potential. In addition to that, in this region, the tight binding approach states that the atomic orbitals are a good approximation to the Bloch state. Thus, the matrix elements of \( \hat{H}_{so} \) with the crystal wave functions can be replaced by the simpler atomic matrix elements of \( \hat{H}_{so} \).
In the case of our tight binding model used to describe bulk SrTiO$_3$, this translates mathematically to:

\[
\langle \psi_i^k, s | \hat{H}_{\text{SO}} | \psi_j^k, s' \rangle \approx \langle d_i, s | \hat{H}_{\text{SO}} | d_j, s' \rangle
\]  

(2.17)

with $i, j = yz, xz, xy$ and $s, s'$ the spin states. Computing these matrix elements gives:

\[
\hat{H}_{\text{SO}}^{\text{TB}} = \frac{i\Delta_{\text{SO}}}{3} \begin{pmatrix}
0_{2\times2} & \sigma_z & -\sigma_y \\
-\sigma_z & 0_{2\times2} & \sigma_x \\
\sigma_y & -\sigma_x & 0_{2\times2}
\end{pmatrix}
\]  

(2.18)

with $\sigma_x, \sigma_y, \sigma_z$ the Pauli matrices and $\Delta_{\text{SO}}$ the energy splitting induced by the SO interaction in the bulk. $0_{2\times2}$ is the $2 \times 2$ null matrix.

Using theoretical calculations, Zhong et al. [60] set the value of $\Delta_{\text{SO}}$ in SrTiO$_3$ to 29 meV. According to Bistritzer et al. [61], however, the value of $\Delta_{\text{SO}}$ is not so clearly defined as “the experiments that do exist appear to partially contradict one another”. Experimentally, $\Delta_{\text{SO}}$ is difficult to determine as it mixes with the tetragonal distortion term. As underlined at the beginning of this section, our point of view on the band structure of bulk SrTiO$_3$ and LaAlO$_3$/SrTiO$_3$ is only qualitative. Thus we arbitrarily choose to follow Zhong et al.

Fig. 2.16 (left) shows the band structure obtained by Zhong et al. using ab initio modeling of cubic SrTiO$_3$ in the absence (dashed lines) and in the presence (red lines) of SO interaction. Clearly the degenerate $xy, yz$ and $xz$ bands presented in fig. 2.7 split into two groups when atomic SO coupling is taken into account. More precisely, as SO mixes the electronic states with different spatial symmetry, the initial $T_{2g}$ states (called $\Gamma^+_{5}$ in the notations used by Zhong) are no longer eigenstates of the system at the $\Gamma$ point; thus new states are formed. The first ones, labeled by the irreducible representation $\Gamma^+_{8}$ are four times degenerate at the $\Gamma$ point and lie at lower energy (remember that now spin is taken into account). The second ones, labeled by the irreducible representation $\Gamma^+_{7}$, are only twice degenerated and lie at higher energy.

Fig. 2.16 (right) shows the band structure obtained by diagonalizing $\hat{H}_{\text{SO}}^{\text{TB}}$ (note that in [60], an improved version of $\hat{H}_{\text{SO}}^{\text{TB}}$ can be found, that is more correct at large $k$-values). A very good matching between density functional theory (DFT) and the tight binding calculation is observed with an SO interaction induced splitting around $\approx 30$ meV. Moreover, the tight binding model easily gives access to the ($\Gamma$
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Figure 2.16: Comparison between ab initio calculations and the tight binding approach in bulk SrTiO$_3$. Left: DFT results for the band structure of bulk SrTiO$_3$ without (dashed grey lines) and with (red lines) atomic SO interaction. Right: same analysis using a tight binding approach. The figure was borrowed from [60]. $\Gamma^7$ and $\Gamma^8$ correspond to the irreducible representations of the $O_h$-double group to which the $\Gamma$-point states of the spin-orbit split bands belong.

Additional complications of the bulk band structure arise when the tetragonal distortion is taken into account (see for example [52]). For the sake of simplicity, we do not go into the description of the band structure of tetragonal bulk SrTiO$_3$ in the presence of SO interaction. Recent papers by Janotti et al. and El-Mellouhi et al. [53, 54] deal with this point.

2.3.4 The LaAlO$_3$/SrTiO$_3$ interface with spin-orbit interaction

In the previous sub-sections we have seen that moving from the bulk to the interface of SrTiO$_3$ has dramatic effects on the band structure, even in the absence of SO interaction. In particular, the continuous energy dispersion along the out-of-plane direction of bulk SrTiO$_3$ is replaced at the interface by a set of sub-bands dispersing only along the planar directions ($k_z$ being quantized). Also, the sequence in energy of the bands with

\[
|\Gamma^+_8\rangle : \frac{1}{\sqrt{6}} (i\psi^{yz,\uparrow}_k + \psi^{xz,\uparrow}_k + 2i\psi^{xy,\downarrow}_k) \\
\frac{1}{\sqrt{2}} (-i\psi^{yz,\downarrow}_k - \psi^{xz,\downarrow}_k) \\
\frac{1}{\sqrt{2}} (i\psi^{yz,\uparrow}_k - \psi^{xz,\uparrow}_k) \\
\frac{1}{\sqrt{6}} (-i\psi^{yz,\downarrow}_k + \psi^{xz,\downarrow}_k + 2i\psi^{xy,\uparrow}_k)
\]

\[
|\Gamma^+_7\rangle : \frac{1}{\sqrt{3}} (-i\psi^{yz,\downarrow}_k + \psi^{xz,\uparrow}_k - i\psi^{xy,\uparrow}_k) \\
\frac{1}{\sqrt{3}} (-i\psi^{yz,\uparrow}_k - \psi^{xz,\uparrow}_k + i\psi^{xy,\downarrow}_k)
\]
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different symmetries is reversed at the interface, the \( xy \) sub-bands lying on average at lower energy than the \( xz/yz \) sub-bands.

Now, in order to consider the effect of SO, we follow Zhong et al. [60] and Kim et al. [75] and reduce the band structure of the interface to a three sub-band model, that can be regarded as a “close-up” on the band structure discussed in subsection 2.3.2. It consists of two degenerate 2D \( xz/yz \) sub-bands separated from one sub-band with \( xy \) symmetry by an energy \( \Delta E \). Thus, close to the \( \Gamma \) point we get the following model Hamiltonian:

\[
\hat{H}_{0,2D}^{TB} = \begin{pmatrix}
\frac{k^2}{2}(\frac{k_x^2}{m_x} + \frac{k_y^2}{m_y}) & 0 & 0 \\
0 & \frac{k^2}{2}(\frac{k_x^2}{m_x} + \frac{k_y^2}{m_y}) & 0 \\
0 & 0 & \frac{k^2}{2}(\frac{k_x^2}{m_x} + \frac{k_y^2}{m_y}) - \Delta E
\end{pmatrix}, \quad \psi^{2D}_{k_\parallel} = \begin{pmatrix}
\psi^{2D, yz}_{k_\parallel} \\
\psi^{2D, xz}_{k_\parallel} \\
\psi^{2D, xy}_{k_\parallel}
\end{pmatrix}
\]

(2.20)

We choose \( \Delta E = 50 \) meV in reference to the X-ray absorption spectroscopy study by Salluzzo et al. [55].

In addition to represent the minimal band scheme necessary to explain some of the features of the (magneto-)transport measurements at the LaAlO\(_3\)/SrTiO\(_3\) interface (as we shall see), this special choice of \( \hat{H}_{0,2D}^{TB} \) has the advantage to allow the same matrix representation as in eq. (2.18) to be used for the atomic SO at the interface (for details see [60]).

At this stage, the total Hamiltonian of the interface, \( \hat{H}_{0,2D}^{TB} + \hat{H}_{SO}^{TB} \), is symmetric under the inversion symmetry operation \( (r \rightarrow -r) \), which is not true in the case of the heterostructure itself. This inconsistency can be solved by taking into account the effect of the strong confining electric field present at the interface on the atomic orbitals of the Ti and O atoms [60, 74].

Figure 2.17 shows that the confining electric field has two main effects. The first one (top) is to deform the lobes of the atomic orbitals (orbital polarization), the electrons being attracted towards the interface. The second one (bottom) is to move the Ti and O ions away from their equilibrium position (lattice polarization). Due to their ionic charge, the Ti atoms are pushed farther from the interface while the O atoms are attracted, resulting in a small buckling of the TiO\(_2\) plane. We note that this second effect has been observed in the LaAlO\(_3\)/SrTiO\(_3\) system using surface X-ray diffraction by Pauli et al. [41]. According to Khalsa et al. [74], it is this phenomenon rather than the orbital polarization that leads to the strong Rashba interaction observed at the LaAlO\(_3\)/SrTiO\(_3\) interface (see below).

An important consequence of the orbital and lattice polarization is the appearance of new channels in the bonding network linking the \( T_{2g} \) orbitals. Indeed, due to these structural perturbations the transfer integral between two neighboring Ti-\( d_{yz} \) and O-\( p_y \) orbitals (for example) is no longer canceled by symmetry (see fig. 2.17). As a consequence, in the presence of a confining electric field, the hopping between a \( d_{yz} \) and a \( d_{xy} \) orbital is allowed (in contrast with the bulk case where this process is forbidden). Interestingly, from fig. 2.17 one can easily obtain that the hopping from a \( d_{xy} \) to a \( d_{yz} \) orbital equals \( t_R = \frac{u^2 + \gamma \Delta \nu_d}{\Delta_{pd}} \) while the hopping from a \( d_{yz} \) to a \( d_{xy} \) orbital equals \( -t_R \) \( (E \) is the modulus of the electric field and \( n \) is the z-direction cosine of the Ti-O bond).
2. BAND STRUCTURE

Figure 2.17: Orbital and lattice polarization due to the strong confining electric field present at the LaAlO$_3$/SrTiO$_3$ interface. Row of atoms composed by Ti-O-Ti-O-Ti. Top: Due to the distortion of the atomic orbitals in the presence of a strong electric field, hopping between (for example) a Ti-$d_{yz}$ and a O-$p_y$ orbital is possible. Bottom: A similar effect is obtained via the polarization of the SrTiO$_3$ lattice. This picture was inspired by [74].

These additional hopping paths lead to the following inversion asymmetric contribution to the total Hamiltonian of the system (at small wave vector $k_{\parallel}$):

$$\hat{H}_{TB}^R = i \Delta_R \begin{pmatrix} 0 & 0 & -k_x a \\ 0 & 0 & -k_y a \\ k_x a & k_y a & 0 \end{pmatrix}$$

(2.21)

with $\Delta_R = 2t_R$.

In fig. 2.19 and 2.20, we show respectively the band structure and the Fermi surfaces resulting from the diagonalisation of the model Hamiltonian $\hat{H}_{TB}^0 + \hat{H}_{TB}^{SO} + \hat{H}_{TB}^R$. We used the same parameters as Kim et al. [75] (mapped on the results of Zhong et al. [60]): $\Delta_{SO} = 27$ meV, $\Delta_R = 20$ meV, $m_l^* = 0.41m_e$ and $m_h^* = 6.8m_e$. Thus, in this analysis, the effective masses are different from the ones used in our Poisson-Schrödinger model. We made this special choice, as the above values of $\Delta_{SO}$, $\Delta_R$, $m_l^*$ and $m_h^*$ were determined altogether by fitting a DFT calculation.

To analyze the calculated band structure and Fermi surfaces, we used a color code (RGB=%Red, %Green, %Blue) that represents the relative weight of each $T_{2g}$ state in the eigenstate of energy $E$:

$$\text{color} = \text{RGB} (P^{yz}, P^{zy}, P^{xz})$$

(2.22) with:

$$P^i = \sqrt{|\psi_{k_{\parallel}}^{2D,i,\uparrow}|^2 + |\psi_{k_{\parallel}}^{2D,i,\downarrow}|^2}$$

(2.23)
2.3 Band structure calculations

Figure 2.18: Band structure and spin orientation in the linear and cubic Rashba model. Spin orientations are in red. (Left) Linearly Rashba spin-split parabolic band structure. (Right) Cubicly Rashba spin-split parabolic band structure.

Looking at fig. 2.19, we observe that some of the branches keep their orbital character. This is for instance the case for the xy-band (green color) except for the crossing points where $\hat{H}_{TB}^{SO} + \hat{H}_{TB}^{R}$ lifts the degeneracy and mixes the $T_{2g}$ states.

The main effect of the breaking of inversion symmetry is to spin-split every sub-band for $k \neq 0$ (i.e. for systems with time and inversion symmetry $E(k, \uparrow) = E(k, \downarrow)$). In our case, as the inversion asymmetry is created artificially, this splitting is called “structure inversion asymmetry splitting” or SIA splitting (to be distinguished from the “bulk inversion asymmetry splitting” or BIA splitting). The simplest model Hamiltonian to describe SIA splitting is due to E. I. Rashba [76] and takes the following form:

$$\hat{H}_{TB}^{R} = \lambda \hat{\sigma} \cdot (k \times E)$$ (2.25)

with $\lambda$ a material specific Rashba coefficient and $E$ the confining electric field. In the case of a single band isotropic 2DEG lying in the xy-plane, it is easy to show that the band structure in the presence of linear-Rashba coupling is:

$$E_{\pm}(k) = \frac{\hbar^2 k^2}{2m^*} \pm \lambda E_z k$$ (2.26)

where $\pm$ refer to the two spin-split bands. The associated eigenstates are:

$$\psi^{2D}_{k_{\parallel}, \pm} = e^{ik_{\parallel} r_{\parallel}} \frac{(\pm ie^{-i\theta}, 1)}{\sqrt{2}}$$ (2.27)

with $k_{\parallel} = (k \cos(\theta), k \sin(\theta))$. They correspond to states with the spin always oriented perpendicular to the wave vector $k$ (see the left part of fig. 2.18).
2. BAND STRUCTURE

As we shall see, in more complex situations, it can happen that \( k \)-linear SIA splitting is forbidden. In these cases, \( k \)-cubic Rashba splitting occurs. It is in general described by the following model Hamiltonian:

\[
\hat{H}_{\text{R3}}^{\text{eff}} = \lambda_3 E_z i (k_-^3 \sigma_+ - k_+^3 \sigma_-)
\]  

(2.28)

with \( k_\pm = k_x \pm ik_y \), \( \sigma_\pm = \frac{1}{2} (\sigma_x \pm i\sigma_y) \) and \( \lambda_3 \) a material specific cubic-Rashba coefficient.

Taking, as for the case of a linear-Rashba splitting, the example of a 2DEG we obtain:

\[
E_{\pm}(k) = \frac{\hbar^2 k^2}{2m^*} \pm \lambda_3 E_z |k|^{3/2}
\]  

(2.29)

and:

\[
\psi_{2D}^{k||,\pm} = e^{ik_\parallel r_\parallel} \left( \pm i e^{-i3\theta}, 1 \right) \sqrt{2}
\]  

(2.30)

It is interesting to note that, moving from the linear to the cubic case, not only the band structure splits differently, but the spin orientations are also changed. In figure 2.18(right), we see that in the presence of cubic Rashba SO, going around the Fermi surface, the spin rotates three times instead of one.

In the tight binding simulation, SIA splitting is particularly large close to where the \( d_{yz} \) and \( d_{xy} \) sub-bands would cross in the absence of SO (see [60]). Moreover, around these crossings, its energy and orientation dependence is very complex. The situation becomes simpler at the sub-band edges. In fig. 2.19 we observe that, indeed, the sub-band dispersions close to the \( \Gamma \) point can be fitted using either a linear (for the lowest and highest sub-bands) or a cubic spin-splitting (for the middle band).

The spin orientation of a particular state belonging to a particular sub-band can be approached by calculating:

\[
S = \langle \psi_{2D}^{k||} | \Sigma | \psi_{2D}^{k||} \rangle
\]  

(2.31)

with

\[
\Sigma_i = \mathbb{I}_{\text{orb}} \otimes \hat{\sigma}_i
\]  

(2.32)

and \( \mathbb{I}_{\text{orb}} \) the identity operator acting on the orbital part of \( \psi_{2D}^{k||} \) (in our case it is the 3x3 identity matrix). \( S \) is nothing but the average orientation of the spin in state \( \psi_{2D}^{k||} \). In the particular situation where the spin of the electron has a well defined orientation (as in the above Rashba model Hamiltonians) \(|S| = 1\). Otherwise, \(|S| < 1\), illustrating the fact that the spin is no longer a good quantum number.

In figure 2.20, we show three different Fermi surfaces obtained from the band structure plotted in fig. 2.19. The three corresponding Fermi energies were fixed slightly higher than the three sub-band bottoms. On top of that, we plotted the vectors \( S \) (black arrows) and a reference vector of unit size (red arrow on each graph). Far from the avoided band crossings one can observe that spin is still a good quantum number. Similarly, the linear Rashba model describes correctly the bottom of the lowest sub-band. In general, however, the spin configuration obtained from the tight binding model of the interface is very complex. For example neither the linear, nor the cubic Rashba model is able to describe the spin orientations at the bottom of the two highest
2.3 Band structure calculations

sub bands. Indeed, at these locations $|S|$ is very small indicating that $\psi_{k_\parallel}^{3D}$ doesn’t have a preferential spin direction.

From the discussion above, one could be concerned by the ability to interpret the (magneto-)transport experiments in such a complicated system. In general, the dynamics of electrons, \textit{a fortiori} under magnetic field, is a complex question. As we will see, it turns out that relatively simple models, like the linear Rashba model, are able to describe satisfactorily many experiments. Moreover the parameters extracted from these analyses often converge to a common interpretation.
Figure 2.19: Band structure in the tight binding model. Top left: Color code representing the weight of each $T_{2g}$ state in $\psi_{k\parallel}^{2D}$. Bottom left: Band structure obtained by diagonalizing $\tilde{H}_{TB}^{0} + \tilde{H}_{SO}^{TB} + \tilde{H}_{R}^{TB}$. Right: Detail of the spin-split band structure at the bottom of the three sub-bands. Black lines are the fits to the linear or cubic Rashba models.
Figure 2.20: Fermi surfaces in the tight binding model. Fermi surfaces of the band structure presented in fig. 2.19 taken at three different Fermi energies ($E_f = 12$, -8.4 and -51.8 meV). Color code is the same as fig. 2.19. Graphs in the same rectangle are different "close-ups" of the same Fermi surface. Black arrows indicate the spin direction and amplitude ($S$). The red arrow on each graph shows the size of a spin unit vector.
3.1 Introduction

Growing a crystal on top of another crystal is not an easy task, even in the case of thin films. It depends on many parameters such as for example the surface state of the substrate, the kinetic energy of the deposited atoms, their oxidation state (in the case of oxides), the lattice mismatch between the two materials etc... At the same time, on the crystal grower side, there are just a limited number of parameters that can be tuned. These are in general the growth temperature of the substrate, gas(es) pressure(s), the deposition rate, the relative orientation and position between the substrate and the target in the deposition chamber and the chemical composition of the plasma (in the case of physical vapor deposition). This implies that, for many materials, a coherent growth of the thin film is only achieved in a very narrow window of parameters that needs to be determined. It is also essential to be able to check the crystal quality of the heterostructure before performing other measurements.

In this chapter, we explain how high quality LaAlO$_3$/SrTiO$_3$ interfaces can be produced by pulsed laser deposition (PLD). We also show how the deposition parameters influence the (electronic) properties of the interface. Finally, we present the field effect devices used throughout this thesis to tune the electronic properties of the 2DEG.

3.2 PLD thin film growth

As mentioned in the introduction, in our lab, we grow the LaAlO$_3$ layer by pulsed laser deposition. We use this technique as it was demonstrated to be an efficient way to
3. SAMPLE GROWTH AND CHARACTERIZATION

Figure 3.1: Pulsed laser deposition system (Left) Picture of our PLD chamber. The red dashed line symbolizes the UV laser pulse coming from the back of the picture. (Middle) Close up through the encircled viewport at the precise moment when a laser pulse hits the target. The very bright spot is the plasma ejected from the target, called the plume. (Right) Scheme describing the elements of the middle picture. Only the RHEED gun and the phosphor screen were added.

produce high quality heterostructures. More recently, other deposition techniques have also been used [77].

In PLD, an intense ($\approx 50$ mJ) laser pulse of 25 ns is used to ablate a target made from the material one wishes to deposit on the substrate. This process leads to the formation of a plasma “plume” (see fig. 3.1) that propagates perpendicularly to the target surface due to Coulomb repulsion. If a background gas is present in the chamber, the plume interacts with it during its path to the substrate. This is a crucial step for the deposition of many materials with two important consequences:

- The plasma can react chemically with the background gas (for instance, oxidizing the atomic/ionic species).
- The background gas reduces the kinetic energy of the plume, avoiding etching of the surface of the substrate by the plasma.

Once landed, the ad-atoms diffuse on the surface of the substrate until they reach a local minimum energy position. The substrate temperature and the binding forces between the ad-atoms and the surface modify the diffusion length, resulting in different growth mode (see below). Figure 3.1 shows a picture of our PLD system together with a scheme demonstrating its working principle.

In practice the surface of the substrate presents a step and terrace structure (see fig. 3.3): it consists of atomically flat terraces followed by abrupt, one unit cell high, steps. This implies that the optical surface makes a small angle (the “miscut” angle) with the crystallographic planes (typically our LaAlO$_3$/SrTiO$_3$ interfaces are grown starting with miscut angles $< 0.1^\circ$). Given this important surface property three layer growth modes can be observed depending on the temperature:
3.2 PLD thin film growth

![Image of growth modes](image.png)

**Figure 3.2: Growth modes** (From left to right) Step flow, layer by layer and 3D growth mode. The associated typical RHEED profiles are also shown. Note that the time axis is not preserved between the graphs as for step flow growth each reduction of the intensity corresponds to one laser pulse, while for layer by layer growth a finite number of pulses are needed to create an oscillation.

- **Step flow growth**: if the diffusion length of the species deposited on the surface is comparable to the terrace width, the ad-atoms will diffuse until they reach a terrace edge, where they will attach. The deposition effectively leads to a forward expansion of each step.

- **Layer by layer growth**: if the diffusion length of the species deposited on the surface is smaller, then many of the ad-atoms will not reach the terrace edges. Rather, they will collide and stick together on the terrace acting as a nucleation center for species landing nearby. Thus, one unit cell high “islands” are going to grow directly on the terraces. When adjacent “islands” merge, one layer of the film is deposited.

- **3D growth**: if the diffusion length of the species deposited on the surface is very small (as for example at room temperature), new atoms landing on the surface will simply pile up on top of each other, destroying the initial steps and terraces structure of the surface.

Experimentally, the growth mode can be determined *in-situ* by using Reflection High-Energy Electron Diffraction (RHEED). This technique makes use of an accelerated electron-beam (21 kV) to probe the surface state of the sample. The electrons are sent in grazing incidence (< 2°) so that they are reflected only by the very top layers of the growing material. Then, a diffraction image forms on a fluorescent screen (see fig. 3.1).

The heart of this method relies on the fact that constructive interferences of the reflected electrons will only happen on a crystalline surface. Looking at the intensity of the interference spots allows one to monitor the surface order during the deposition.
3. SAMPLE GROWTH AND CHARACTERIZATION

(The image is the reciprocal of the atomic structure present at the surface). The different evolution of the RHEED intensity for the three growth modes are shown in fig. 3.2. Thus, in the case of layer by layer growth, one expects substantial and periodic variations in the intensity of the diffracted beam, the so-called RHEED oscillations. On the contrary, in the case of step flow growth, one expects a sharp decrease of the RHEED signal just after each laser pulse directly followed by a recovery of the intensity (on the time scale of the diffusion time). Finally, extinction of the diffracted beam happens in the case of 3D growth.

In Appendix A we detail step by step the procedure we follow to prepare LaAlO$_3$/SrTiO$_3$ interfaces by PLD. Note that we work exclusively with SrTiO$_3$ substrates presenting a (001)-oriented TiO$_2$-terminated surface, as growing LaAlO$_3$ on a SrO terminated surface leads to insulating interfaces [78] (see chapter 2).

3.3 Quality check

Before investigating in detail the transport properties, it is very desirable to be able to check the quality of the deposited layers. An important piece of information is readily accessible thanks to the RHEED oscillations. In fig. 3.3 we show that, in our growth conditions, layer by layer growth of the LaAlO$_3$ is obtained.

Typically, we use three additional characterization techniques: atomic force microscopy (AFM), X-ray diffraction and electronic transport.

3.3.1 AFM

Atomic force microscopy (AFM) is probably one of the experimental probes with the highest ratio between conceptual simplicity and power of investigation. It makes use of a very soft cantilever at the end of which a small tip is located to image, for example, the topography of a surface on the nanometer scale (its resolution is typically 1–10 nm in the surface plane and 0.1 nm out-of-plane). These two elements are themselves technological achievements since the best cantilevers nowadays are able to sense forces down to 1 pN and the tips end can be less than 2 nm in radius.

A very short and simple minded description of the AFM working principle is the following. To measure topography, the cantilever is displaced on the surface of the sample using piezo-electric crystals. Due to the forces between the surface and the tip, the cantilever bends. This deflection is amplified, via an optical detection technique: the reflection of a laser beam on the top surface of the cantilever is recorded by an array of photodiodes. This measurement is then converted to a height profile. Other measurement modes, are also possible in non-contact: in this case the change of the resonant frequency of the cantilever due to the surface-tip forces is detected.

In fig 3.3, we show the surface topography of a 10 unit cell thick LaAlO$_3$/SrTiO$_3$ sample grown in our lab. Clearly the deposition process reproduces the substrate step and terrace structure, with abrupt 1 unit cell steps.
Figure 3.3: Characterization of our LaAlO$_3$/SrTiO$_3$ samples. (Top left) RHEED oscillations of a 9uc sample. Layer by layer growth is observed. (Top right) X-ray analysis of the same sample. $\theta - 2\theta$ scan around the (001) Bragg peak of SrTiO$_3$ reveals finite size oscillations. (Bottom left) Temperature dependence of the electrical resistance for the same sample. A metallic behavior is observed. (Bottom right) AFM surface topography of a 10uc sample. Abrupt unit cell steps followed by flat terraces are visible.
3. SAMPLE GROWTH AND CHARACTERIZATION

3.3.2 X-ray diffraction

We use high resolution x-ray diffraction (HRXRD) to probe the crystalline perfection of our films. This technique is based on the elastic scattering of x-ray photons on the valence electrons of the atoms composing a crystal. In our setup, we use the K α\textsubscript{1} emission line of Cu (λ = 1.54056 Å). This sub-nanometric wavelength allows information on the crystal structure to be obtained (one unit cell of a perovskite structure is typically 3.5 Å).

The intensity of the reflected light in a diffraction experiment on a periodical lattice is maximal at angles that satisfy the Laue equation:

\begin{align}
    a \cdot \Delta k &= 2\pi h \\
    b \cdot \Delta k &= 2\pi k \\
    c \cdot \Delta k &= 2\pi l
\end{align}

(3.1)

with \(a\), \(b\) and \(c\) the primitive vectors of the lattice, \(\Delta k\): the difference in wave vector between the incident and reflected light and \((h, k, l)\) the reciprocal lattice indices that specify from which set of crystallographic planes the light is scattered.

Defining \(\theta\) as the angle between the incident beam and a particular set of crystallographic planes and \(d\) the distance between them, eq. (3.1) simplifies to Bragg’s relation:

\[2d \sin \theta = n\lambda\]

(3.2)

with \(n\) an integer.

Thus, by varying the incident \(\theta\) and the reflected \(2\theta\) angles with respect to the sample and recording the intensity of the reflected beam one can obtain \(d\). This type of scan is called a \(\theta - 2\theta\) scan.

In fig. 3.3 we show a \(\theta - 2\theta\) scan, of a 9 unit cells LaAlO\textsubscript{3}/SrTiO\textsubscript{3} sample, around the (0 0 1) reflexion of SrTiO\textsubscript{3}. Consistently with Bragg’s law, a sharp peak is observed at \(2\theta \approx 22.75^\circ\) (c\textsubscript{cubic} \text{STO} = 3.905 Å). We also see a very broad peak: we attribute it to the film. Due to the large tensile strain applied by the substrate on the LaAlO\textsubscript{3} film (c\textsubscript{pseudo-cubic} \text{LAO} = 3.791 Å), its position is displaced to higher angles, implying that the c-axis of the LaAlO\textsubscript{3} film is reduced to c\textsubscript{pseudo-cubic} \text{LAO-film} = 3.745 Å. In addition to that, finite size oscillations are visible. Fitting their positions yields an estimation of the film thickness of 9 uc, in accordance with the number of RHEED oscillations. Reciprocal space maps, not shown here (see for example [79]), reveal that our films are fully strained.

It is interesting to note that, far from the film Bragg peak, a simple formula holds for the determination of the film thickness via the finite size oscillations induced by the (0 0 1) planes. Indeed:

\[
    N = \frac{\sin \theta_B}{\sin \theta_{m+1} - \sin \theta_m}
\]

(3.3)

with \(N\) the film thickness in unit cells, \(\theta_B\) the angle of the film Bragg peak and \(\theta_m\) the angle of the \(m\)-th finite size oscillation.
3.3 Quality check

Figure 3.4: Sample design (Left) Sketch of our Hall bar configuration for transport measurements at the LaAlO$_3$/SrTiO$_3$ interface. Grey areas are insulating regions. The back-gate electrode is visible by transparency (on the back of the SrTiO$_3$ substrate). The color gradient refers to the value of the local electrical potential, obtained via the simulation of the electrical transport through the Hall bar in the presence of an out-of-plane magnetic field (using COMSOL Multiphysics and the 2DEG approximation). As can be observed, in this case the isopotential lines are kinked due to the development of a transverse electric field (Hall effect). (Right) Picture of the real device.

3.3.3 Electrical resistance

We use standard photolithographic techniques to pattern a conducting Hall bar for transport measurements (see Appendix A). A sketch and a picture of our final device are given in fig. 3.4. The length of the channel (i.e. the distance between the $V_+$ and $V_-$ contacts) is $l = 2000 \mu$m while its width is $w = 500 \mu$m. The current is injected from the $I_+$ contact and extracted from the $I_-$ contact which is in general grounded. Ohm’s law is used to estimate the electrical resistance:

$$\rho \mathbf{J} = \mathbf{E}$$  \hspace{1cm} (3.4)

with $\mathbf{J}$ the density of current, $\mathbf{E}$ the electric field and $\rho$ the tensor of resistance. Choosing the direction of the current to be along the “$x$” direction we get:

$$\rho_{xx} = \frac{E_x}{J_x}$$  \hspace{1cm} (3.5)

By definition, $J_x = I/(w \times t)$ in three dimensions ($t$ is the thickness of the conducting layer) and $J_x = I/w$ in two dimensions. The electric field is $E_x = (V_+ - V_-)/l = \Delta V/l$. Thus:

$$\rho_{xx}^{3D} = \frac{\Delta V \ w \ t}{I \ l} \text{ and } \rho_{xx}^{2D} = \frac{\Delta V \ w}{I \ l}$$  \hspace{1cm} (3.6)

$\rho_{xx}^{3D}$ is called the resistivity and $\rho_{xx}^{2D}$ is the sheet resistance (often denoted $R_s$). Since we are studying a 2DEG at the LaAlO$_3$/SrTiO$_3$ interface we work with the sheet resistance:
the room temperature (296 K) value is typically $\rho^{2D}_{xx} = 10^{-40}$ kΩ. Fig. 3.3 shows a typical example of the temperature dependence of the electrical resistance at the LaAlO$_3$/SrTiO$_3$ interface. Reducing the temperature to liquid helium temperatures a metallic behavior is observed. At 4 K the sheet resistance lies around 400 Ω.

In the presence of an out-of-plane magnetic field ($B$), a potential difference develops transversely to the propagation of the electrical current. This is the Hall effect. Again, this can be measured easily in a Hall bar configuration. Referring to the sketch in fig. 3.4, we find $E_y = (V_+ - V_H)/w$. Thus:

$$\rho^{3D}_{yx} = \frac{V_+ - V_H}{I} \quad \text{and} \quad \rho^{2D}_{yx} = \frac{V_+ - V_H}{I}$$

(3.7)

Defining the Hall constant $R^{3D,2D}_H = \rho^{3D,2D}_{yx}/B$, the carrier density in 3D and 2D can be estimated via (these expressions are only valid in the simplest theory, see chapter 4):

$$n_{3D} = (R^{3D}_H q)^{-1} \quad \text{and} \quad n_{2D} = (R^{2D}_H q)^{-1}$$

(3.8)

with $q$ the charge of the carriers. Thus, the inverse of the Hall constant contains information on the charge density. Note that $\rho^{2D}_{yx}$ is often written $R_{xy}$.

Making use of the Drude formula for the conductivity (or the conductance)

$$\frac{1}{\rho^{3D,2D}_{xx}} = n_{3D,2D} q \mu$$

(3.9)

the electron mobility ($\mu$ also called the Hall mobility) can be estimated:

$$\mu = \frac{R^{3D,2D}_H}{\rho^{3D,2D}_{xx}}$$

(3.10)

As we shall see, the inverse Hall constant and Hall mobility vary as a function of temperature for the “standard” LaAlO$_3$/SrTiO$_3$ interface. Typically, if the measurements are made up to a magnetic field $B = 4$ T, at room temperature $(R^{3D}_H q)^{-1} = 7 \times 10^{13}$ cm$^{-2}$ and $\mu = 10$ cm$^2$ V$^{-1}$ s$^{-1}$, while at low temperature $(R^{2D}_H q)^{-1} = 2 \times 10^{13}$ cm$^{-2}$ and $\mu = 1000$ cm$^2$ V$^{-1}$ s$^{-1}$. The high mobility observed at low temperature reveals the high crystalline quality of the interface.

### 3.4 The role of the deposition parameters

In what follows, we give a description of the dependence of the transport properties of LaAlO$_3$/SrTiO$_3$ interfaces as a function of the thickness of the LaAlO$_3$ layers, of the oxygen pressure during growth and of the growth temperature. If not indicated, the parameters for the growth of the LaAlO$_3$ film are the same as in Appendix A.

#### 3.4.1 Film thickness

In fig. 3.5, we show the evolution of the sheet resistance at 5 K as a function of the number of unit cells of LaAlO$_3$ deposited along the [001] direction (similarly to [80]). A clear insulator to metal transition is observable as the thickness of the film exceeds 3
3.4 The role of the deposition parameters

3.4.1 Optimal thickness window

Figure 3.5: Thickness dependence of the transport properties at the LaAlO$_3$/SrTiO$_3$ interface. For three different growth temperatures, sheet conductance at low temperature (5 K) as a function of the thickness of the LaAlO$_3$ film. For all the samples, a jump in the conductance is observed at 4uc, followed by a gradual decrease of the conductance above 15-20 unit cells. Such a transition is also observable at room temperature. As we have seen in chapter 2, this feature of the LaAlO$_3$/SrTiO$_3$ system is one of the crucial observations [11] in favor of the polar catastrophe model proposed by Nakagawa et al. [10] to explain the conduction in this system. The decrease of conductance above ≈ 20 uc is concomitant with the relaxation of the LaAlO$_3$ c-axis to its bulk value (see [37] or fig. 2.4). Moreover, AFM topography images of LaAlO$_3$ films thicker than 20 uc reveal a crack network (see for example [79]). Thus, high quality LaAlO$_3$/SrTiO$_3$ interfaces can only be obtained in a restricted window of film thicknesses (see fig. 3.5). Interestingly the maximum thickness depends on the growth temperature, being higher at lower growth temperature.

3.4.2 Oxygen pressure during growth

In our standard growth procedure (see Appendix A), the LaAlO$_3$ deposition is always followed by an in situ high temperature annealing in 0.2 bar of oxygen for 1 h. The reason for this treatment is that SrTiO$_3$ is very prone to lose oxygen if heated in vacuum and that oxygen deficient SrTiO$_3$ is a conductor. In fig. 3.6 we show the transport properties of three samples grown in very different oxygen pressures ($10^{-2}$, $10^{-4}$ and $10^{-6}$mbar) without annealing in oxygen. Clearly, the sample grown at $10^{-6}$ mbar has very different properties than the other two. For example, its low temperature sheet resistance is 5 to 6 orders of magnitude lower than that of samples grown in higher oxygen pressure. Similarly its inverse Hall coefficient is the largest, by about three
orders of magnitude.

These experimental observations, together with the conductance profile measured by Basletic et al. [6] (see fig. 1.2) indicate that interfaces grown in reducing conditions exhibit bulk conduction due to oxygen vacancies. An estimate of the 3D carrier density can be obtained via $(R_2D H)^{-1}$ : taking for the thickness of the sample $t = 0.5$ mm we get $n_{3D} = 6 \times 10^{17}$ cm$^{-3}$. For such a carrier density, Spinelli et al. [44], on doped bulk SrTiO$_3$, find a mobility around $10^4$ cm$^2$ V$^{-1}$ s$^{-1}$ in agreement with the data in fig. 3.6c. The observation of 3D Shubnikov-de Haas oscillations by Herranz et al. [81] for samples grown in similar conditions corroborate this picture.

In fig. 3.7, we show the transport properties of interfaces grown in the same conditions as the samples presented in fig. 3.6 but annealed right after growth. This additional step makes the transport properties of heterostructures grown in very different oxygen pressures collapse on the same curve : these are the electronic properties of the “standard” samples we have investigated in this thesis.

### 3.4.3 Growth temperature

As we have seen in fig. 3.5, the growth temperature has a role on the transport properties of the LaAlO$_3$/SrTiO$_3$ system. We further investigated its effect by studying the magnetotransport ($B$ applied perpendicularly to the interface) of interfaces grown in “standard” conditions but varying the substrate temperature $T_g$. As magnetotransport measurements have devoted chapters in this thesis, we will go rather briefly into the
3.4 The role of the deposition parameters

Figure 3.7: Transport properties of annealed (0.2 bar of oxygen pressure) samples with a 5 uc LaAlO₃ film and grown at different oxygen pressures (a) Sheet resistance, (b) inverse Hall constant and (c) Hall mobility as a function of temperature and (d) magnetoresistance at 1.5 K.

description of these experiments.

Figure 3.8 (top) shows the room temperature (296 K) sheet resistance plotted as a function of the growth temperature. We note that upon decreasing \( T_g \) to 650 °C the sheet resistance increases by roughly an order of magnitude. This evolution is mainly related to a reduction of the inverse Hall constant (right axis), the increase in the Hall mobility being less than a factor of two. In fact, carrier mobility at room temperature is determined by phonon scattering and is independent of the carrier density [82]. The middle panel of fig. 3.8 shows a large increase in the electron mobility at low temperatures. As \( T_g \) is reduced from 900 to 650 °C, \( \mu \) rises from 600 to 8000 cm² V⁻¹ s⁻¹.

In order to compare the behavior of these interfaces with the properties of doped-STO single crystals, we assume a uniform thickness of 10 nm for the electron gas for all the samples [6, 26]. As displayed in the bottom panel of fig. 3.8, the evolution of the electron mobility as a function of the carrier density for LaAlO₃/SrTiO₃ interfaces tracks the behavior of bulk SrTiO₃. This suggests that the dominant mechanism controlling the electron mobility is similar in the two systems.

The observed magnetotransport at the interface is however different from the one of doped-SrTiO₃. In fig 3.9, we plot (left) the Hall resistance measured at 1.5 K as a function of the magnetic field and (right) its field derivative as a function of the magnetic field and temperature for heterostructures grown at three different temperatures.

Samples grown at \( T_g = 650 \) °C exhibit a linear Hall effect essentially temperature independent. For growth temperatures of 800 and 900 °C, the Hall resistance displays a
3. SAMPLE GROWTH AND CHARACTERIZATION

![Graphs showing sample growth and characterization data]

**Figure 3.8:** Top panel: Evolution of the sheet resistance measured at 296 K (left scale) and inverse Hall constant measured at 260 K (right scale) as a function of the growth temperature $T_g$. The lines are guides to the eye. Middle panel: Hall mobility as a function of temperature for samples grown at 900 °C (brown triangles), 800 °C (gold dots) and 650 °C (blue squares). The dashed line shows a fit assuming $\mu \propto T^\alpha$ with $\alpha$ being -2.8. This temperature dependence is also observed in bulk doped-STO. Bottom panel: Comparison of LaAlO$_3$/SrTiO$_3$ interfaces and doped-SrTiO$_3$ (green dots) showing the dependence of the low temperature mobility on the carrier density (measured at 260 K) assuming a uniform gas thickness of 10 nm. The same color code is used for LaAlO$_3$/SrTiO$_3$ interfaces. Data for bulk SrTiO$_3$ are taken from Ref. [44].
3.4 The role of the deposition parameters

Figure 3.9: $R_{xy}$ vs. magnetic field $B$ measured at 1.5 K for three growth temperatures. (right) First derivative of $R_{xy}$ as a function of magnetic field and temperature. For each growth temperature, the value of the derivative has been normalized to its value at 160 K to highlight the presence of non-linearities.
more complex behavior. In the 100-300 K temperature range, $R_{xy}$ is linear in magnetic field. Below $\approx 100$ K, the Hall response becomes a non-linear function of the magnetic field and its low field derivative increases at low temperature.

Figure 3.10 compares the magnetoresistance of a high mobility sample (5050 cm$^2$ V$^{-1}$ s$^{-1}$) with the behavior of a sample grown at 800 °C. Using field effect (see next section), the inverse Hall coefficient of the latter was tuned to match that of the high mobility sample ($\left(R_{H}^{2D}q\right)^{-1} = 8 \times 10^{12}$ cm$^{-2}$). As a result, the mobility of the 800 °C sample was reduced to 34 cm$^2$ V$^{-1}$ s$^{-1}$. Figure 3.10 shows that while the high mobility sample displays a negative magnetoconductance (positive magnetoresistance), the change in conductance of the 800 °C sample is positive and on the scale of $e^2/\pi h$. This behavior has been shown to be the signature of weak-localization [27]. We give a theoretical description of this phenomenon in chapter 4.

This experiment reveals that carrier mobility, which determines the conduction regime, is a complex function of the carrier density, field effect and disorder. The origin of the high mobility in “low” $T_g$ samples, however, has not yet been determined. A possible source for the observed differences is an influence of the growth temperature on the interface perfection [83, 84]. The enhanced quality of the RHEED oscillations and X-ray diffraction of LaAlO$_3$/SrTiO$_3$ interfaces grown at 650 °C seem to corroborate this idea [29].

3.5 Field effect devices

As mentioned earlier SrTiO$_3$ has exceptional dielectric properties at low temperature. As a consequence, our 0.5 mm thick substrate can be used as dielectric to build efficient field effect devices. This greatly simplifies the realization of field effect experiments at the LaAlO$_3$/SrTiO$_3$ interface as no processing of the LaAlO$_3$ surface is necessary (needed in the more conventional top-gate geometry). The metallic (back-)gate electrode (typically gold) can be directly deposited on the unpolished side of the SrTiO$_3$ substrate (see Appendix A).

A standard field effect device realized in our lab is shown/sketched in fig. 3.4. By applying a bias voltage between the Hall bar and the gate contact ($V_g$), charges accumulate at the metallic electrodes, just like in a capacitor. As a result the doping of these electrodes changes. While this doping difference does not change the properties of the metallic back-gate contact, the impact on the low density 2DEG at the LaAlO$_3$/SrTiO$_3$ interface is important.

As an illustration, we show, in fig. 3.11, a simulation of the electric field distribution in our field effect devices. The induced charge density in the channel ($\delta n_{2D}$) is given by the color gradient. For a bias voltage of 200 V, we find respectively at room temperature and 1.5 K, $\delta n_{2D}^{RT} \approx 7.3 \times 10^{13}$ cm$^{-2}$ ($\epsilon_r = 350$) and $\delta n_{2D}^{1.5K} \approx 1.5 \times 10^{13}$ cm$^{-2}$ ($\epsilon_r$ given by eq. (2.11)). An estimate of the doping induced by field effect can also be attempted using an infinite parallel plate capacitor model. It gives $\delta n_{2D}^{RT} \approx 7.7 \times 10^{11}$ cm$^{-2}$ assuming $\epsilon_r = 350$ and $\delta n_{2D}^{1.5K} \approx 4.9 \times 10^{13}$ cm$^{-2}$ assuming $\epsilon_r = 22000$. The rather big difference at low temperature between our simulation and the parallel plate capacitor approximation is due to the large electric field dependence of the dielectric constant of SrTiO$_3$ (see fig. 2.11 chapter 2).
Figure 3.10: Comparison between the magnetotransport of two interfaces exhibiting the same inverse Hall constant but grown at different temperatures. Top: Magnetoresistance at 1.5 K of a high mobility sample ($\mu = 5050 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) grown at 650 °C. Shubnikov-de-Haas oscillations appear above 4 T. Bottom: Magnetoconductance normalized by the quantum of conductance ($e^2/\pi h$) of a sample grown at 800 °C. For this device, the electric field effect was used to reduce the carrier density. A low value for the mobility ($34 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) is measured and analyses of magnetotransport reveal a weak localization regime.
3. SAMPLE GROWTH AND CHARACTERIZATION

Figure 3.11: Simulation of the electric field in our field effect devices COMSOL. Multiphysics simulation of the electric field dispersion inside our SrTiO$_3$ substrate upon application of 200 V of back-gate bias at low temperature ($\epsilon_r(E)$ is given by eq. (2.11)). The color code refers to the tuning of the local carrier density.

We performed low temperature (1.5 K) differential capacitance measurements to check for the real evolution of the total carrier density in our heterostructure upon application of gate voltage. The differential capacitance is defined as:

$$C(V) = \frac{dQ}{dV} \tag{3.11}$$

with $Q = qSn_{2D}$ being the charge accumulated on the top or bottom electrode, one of them being in our case the 2DEG. We take $S$ to be the area of the back-gate electrode. From this quantity, we can recover the variation in carrier density induced by a given gate voltage variation:

$$\delta n_{2D} = \frac{1}{Sq} \int_{V_0}^{V_0+\Delta V} C(V) \, dV \tag{3.12}$$

In fig. 3.12, we show the result of this measurement. We see that varying the voltage from $-200$ V to $200$ V, leads to a change in carrier density of $\approx 4 \times 10^{13}$ cm$^{-2}$. This modulation is of the same order as the carrier density inferred from Hall effect measurements, explaining the large tuning of the electronic properties achieved at the LaAlO$_3$/SrTiO$_3$ interface. We also note that, measuring the charging current on a SrTiO$_3$ single crystal equipped with top and bottom gold electrodes, we could confirm the modulation of the carrier density displayed in fig. 3.12 (for technical details see for example [79]).

In our lab, we call the state of the system prior to the application of any field effect the “virgin” or “as-grown” state. This is motivated by the fact that a large hysteresis, in
3.5 Field effect devices

the transport properties, develops during the first sweep in gate voltage. Figure 3.13 shows an example of the variation of the sheet resistance upon gate voltage sweeps. Clearly, the value of the sheet resistance in the “virgin” state is quite different from the values recorded at 0 V after field effect.

As a consequence, all the measurements under field effect presented in this thesis are preceded by a “forming process” which consists of ramping the gate voltage to the maximum positive value before starting the study at different gate voltages.

Another important point concerning field effect experiments at the LaAlO\textsubscript{3}/SrTiO\textsubscript{3} interface, is the following. For the sample shown in fig. 3.13, the sheet resistance can be tuned continuously from about 200\,Ω to 5000\,Ω. This is a quite different range than the one spanned by the sample we published in [27]. In this paper, the sheet resistance could be tuned from \( \approx 500\,Ω \) to \( \approx 23\,kΩ \). This can be interpreted as the fact that the exact same virgin state is not always realized at the LaAlO\textsubscript{3}/SrTiO\textsubscript{3} interface. Indeed, due to the low carrier density in this system, to its two-dimensional nature and to the proximity of the air-LaAlO\textsubscript{3} interface, this heterostructure is very sensitive to external perturbations. It is thus reasonable to think that, for example, slight changes in the interfacial crystal quality or photodoping processes can alter the low temperature behavior of the heterostructure.

As a consequence, to compare the transport properties of different samples, we use the zero-magnetic-field sheet conductance \( (σ_{\text{2D}}^0) \). Figure 3.14 shows the magnetoresistance \( (B \text{ applied out-of-plane}) \) of two different samples; the sample studied in [27] and

---

**Figure 3.12:** Tuning of the carrier density at the LaAlO\textsubscript{3}/SrTiO\textsubscript{3} interface using field effect. Experimental determination of the tuning of the carrier density in a back-gate geometry using differential capacitance measurements.
Figure 3.13: Gate voltage tuning of the sheet resistance Dependence of the resistance on the applied back-gate voltage. A large hysteresis is visible during the first sweep in voltage (forming process).
Figure 3.14: Comparison between the magnetotransport of two samples displaying the same sheet conductance. Variation of the resistance due to the application of an out-of-plane magnetic field. Two samples are presented: the sample studied in [27] and that of fig. 3.13. Field effect was used to bring their zero-magnetic field sheet conductance to $\approx 2 \text{ mS}$.

that of fig. 3.13. For both of them, field effect was used to bring the sheet conductance in the absence of magnetic field to $\approx 2 \text{ mS}$ ($R_s \approx 500 \Omega$). A similar magnetic field dependence of the resistance is observed, despite some deviations in the field scale, confirming the choice of $\sigma_{2D}^0$ as a good parameter to compare different samples.
3. SAMPLE GROWTH AND CHARACTERIZATION
4.1 Introduction

The primary effect of a magnetic field on moving charged particles is to bend their trajectories. In a solid, this phenomenon affects the longitudinal and transverse electrical resistance and can be described using a semi-classical treatment of the electronic system. Subtle effects, linked to the quantum nature of electrons, need however a full quantum mechanical approach.

In this chapter, we give a succinct theoretical description of the semi-classical approach and discuss examples of quantum corrections that can impact the magnetotransport. We then analyze the magnetotransport of LaAlO$_3$/SrTiO$_3$ interfaces grown at 800°C for different magnetic field orientations.

4.2 Semi-classical treatment

4.2.1 General formalism

In the semi-classical approach, the conducting system is assumed to be composed of independent particles that are governed by the Pauli exclusion principle (hence described by the Fermi-Dirac statistics). From a quantum mechanical point of view, a charge carrier corresponds to a wave packet traveling with a well defined group velocity:

$$ v_k = \frac{1}{\hbar} \frac{\partial \epsilon_k}{\partial k} \quad (4.1) $$
where $\epsilon_k$ is the energy of the wave packet with average wave vector $k$. For an electron gas $\epsilon_k$ simply takes the well known form:

$$\epsilon_k = \frac{\hbar^2 k^2}{2m_e}$$

(4.2)

When the interactions with the crystal are taken into account the electron mass is often replaced by an effective mass $m^*$. In more general situations, however, the energy spectrum $\epsilon_k$ can be non-parabolic as well as anisotropic. In these cases, a single effective mass cannot be obtained and $m^*$ is to be treated as a tensor.

In this framework, the electrical current density ($J$) is readily written as:

$$J = \int qv_k f_k \frac{d\mathbf{k}}{2^{d-1} \pi^d}$$

(4.3)

with $f_k$ the distribution function of the carriers, and $\frac{1}{2\pi^{d-1}}$ the spin-degenerated density of states in $k$-space in dimension $d$ ($d > 0$). In the examples below, we will restrict ourselves to the two-dimensional case ($d = 2$). In the absence of external fields and temperature gradients, $f_k$ can be identified with the equilibrium Fermi-Dirac distribution $f_0^k$. In this case, no net current is obtained.

In a transport experiment, the application of an electric and magnetic field brings the system to a steady state for which:

$$\dot{f}_k = 0$$

(4.4)

Notwithstanding its very simple form, this equation is difficult, if not impossible to solve. This is because in reality $\dot{f}_k$ is a complicated function depending on $f_k$ itself and also on its partial derivatives:

$$\dot{f}_k = \dot{f}_k|_{\text{fields}} + \dot{f}_k|_{\text{scatterings}} + \dot{f}_k|_{\text{diffusion}}$$

(4.5)

with:

$$\dot{f}_k|_{\text{fields}} = -\frac{q}{\hbar} (E + v_k \times B) \cdot \frac{\partial f_k}{\partial k}$$

$$\dot{f}_k|_{\text{scatterings}} = -\frac{f_k - f_0^k}{\tau(k)}$$

$$\dot{f}_k|_{\text{diffusion}} = -v_k \frac{\partial f_k}{\partial r}$$

(4.6)

The first term is nothing but the effect of the Lorentz force on the distribution function. The second one expresses the tendency of the system to return to the equilibrium state: after switching off the external fields and temperature gradients this term brings the system back to the equilibrium distribution, $f_0^k$, on a time scale $\tau(k)$. $\tau(k)$ is to be interpreted as a $k$ dependent scattering time, introduced in the theory “by hand” to simplify the problem. Finally, the last term describes the evolution of the particle distribution due, typically, to a thermal gradient: this contribution, relevant for thermoelectric effects, will be omitted in the following discussion.
4.2 Semi-classical treatment

The first approximation is done by setting $f_k = f_k^0 + g_k$ with $g_k$ being linear in $E$. This step leads to the linearization of the problem and is valid if we consider only small deviations from equilibrium. Consistently, we retain only terms that are first order in $E$ in eqs. 4.6 and we write:

$$\dot{f}_k|_{E\text{-field}} \approx -\frac{q}{\hbar} E \cdot \frac{\partial f_k^0}{\partial k} = -\frac{q}{\hbar} \partial \epsilon_k E \cdot v_k \quad (4.7)$$

The magnetic field term has to be treated with more care:

$$\dot{f}_k|_{B\text{-field}} \approx -\frac{q}{\hbar} (v_k \times B) \cdot \frac{\partial f_k^0}{\partial k} = -\frac{q}{\hbar} \partial \epsilon_k (v_k \times B) \cdot v_k = 0 \quad (4.8)$$

Thus, as for the scattering term, the effect of the magnetic field is written in terms of the deviation from equilibrium of the distribution function:

$$\dot{f}_k|_{B\text{-field}} = -\frac{q}{\hbar} (v_k \times B) \cdot \frac{\partial g_k}{\partial k} \quad (4.9)$$

According to eq. (4.4):

$$-q \frac{\partial f_k^0}{\partial \epsilon_k} E \cdot v_k = \frac{g_k}{\tau (k)} + q (v_k \times B) \cdot \frac{\partial g_k}{\partial k} = \left( \frac{1}{\tau (k)} + q (v_k \times B) \cdot M^{-1} \frac{\partial}{\partial v_k} \right) g(v_k) \quad (4.10)$$

where we have used the definition of the inverse mass tensor:

$$M^{-1} = \frac{1}{\hbar} \frac{\partial v_k}{\partial k} \quad (4.11)$$

If we define:

$$\tilde{\Omega} = q \tau (k) (v_k \times B) \cdot M^{-1} \frac{\partial}{\partial v_k} \quad (4.12)$$

we obtain a rather compact form for the out-of-equilibrium part of the distribution function:

$$g(v_k) = \left( 1 + \tilde{\Omega} \right)^{-1} \left( -q \tau (k) \frac{\partial f_k^0}{\partial \epsilon_k} E \cdot v_k \right) \quad (4.13)$$

Since Ohm’s law defines the tensor of conductance ($\sigma$) as:

$$J = \sigma E \quad (4.14)$$

By using 4.3 one easily finds:

$$\sigma_{ij} = \int q v_i \left( 1 + \tilde{\Omega} \right)^{-1} \left( -q \tau (k) \frac{\partial f_k^0}{\partial \epsilon} v_j \right) \frac{dk}{2^{d-1} \pi^{d}} \quad (4.15)$$

where we have omitted the $k$ index for convenience.

Due to the presence of the derivative of the Fermi distribution in the integral of eq. (4.15), if $k_B T \ll \epsilon_F$, the conductance of the system will be determined only by the states close to the Fermi level (this condition is often satisfied in metals or at liquid helium temperatures). An important consequence is that electronic transport measurements are probing only the Fermi surface properties.
4. MAGNETOTRANSPORT

4.2.2 The Drude model with an effective mass

In practice, as a first model for understanding the transport experiments, the Drude model is often used. The interactions of the charge carriers with the crystal are introduced via a finite, energy independent and isotropic elastic scattering time $\tau$ and an effective mass ($m^*$) different from the free electron mass ($m_e$). These assumptions allow eq. (4.15) to be solved analytically.

For a magnetic field set perpendicular to the conduction plane (along "z"), in two dimensions and at zero temperature we get:

$$
\sigma_{xx} = \sigma_{yy} = \frac{1}{1 + \left(\frac{q}{m^*}\right)^2 \frac{B^2}{n} \frac{2}{\tau}} = \frac{1}{1 + \mu^2 B^2 \sigma_0^{2D}}
$$

$$
\sigma_{xy} = -\sigma_{yx} = \frac{\mu B \frac{n}{q^2 \tau}}{1 + \left(\frac{q}{m^*}\right)^2 \frac{B^2}{n} \frac{2}{\tau}} = \frac{\mu B}{1 + \mu^2 B^2 \sigma_0^{2D}}
$$

(4.16)

where we have used the relations $\mu = q\tau/m^*$, $\sigma_0^{2D} = nq\mu$ and defined the components of the tensor of conductance as:

$$
J = \begin{pmatrix}
\sigma_{xx} & \sigma_{xy} \\
\sigma_{yx} & \sigma_{yy}
\end{pmatrix} E
$$

(4.17)

In transport measurements the direction and intensity of the current are fixed and we measure a difference of potential on the path of this current. Thus, experimentally, $J$ is known quite accurately while $E$ is determined from the voltage probes.

As a consequence, the quantity estimated by our measurements is the tensor of resistance ($\rho$) rather than the tensor of conductance. The former is simply the inverse of the latter since:

$$
\rho J = E
$$

(4.18)

and takes the following form under the assumptions of this paragraph:

$$
\rho = \begin{pmatrix}
\frac{1}{\sigma_0^{2D}} & -\frac{B}{q \nu_{2D}} \\
\frac{B}{q \nu_{2D}} & \frac{1}{\sigma_0^{2D}}
\end{pmatrix}
$$

(4.19)

We find two interesting results. Firstly, the carrier density ($n_{2D}$) and the carrier charge ($q$) can be easily determined by looking at the evolution of the transverse resistance $\rho_{xy}$ in field; this is the very famous Hall effect (HE). Secondly, in this model, no magnetoresistance (MR) is expected since $\rho_{xx} = \frac{1}{\sigma_0^{2D}}$. This situation can be understood by observing that the transverse electric field $E_y$ generated by the HE leads to an opposite force to the one produced by the Lorentz term. Thus in this model the only net force acting on the charge carriers comes from $E_x$, illustrating why no MR is predicted.

4.2.3 Multi band conduction

In many cases, one cannot assign a single scattering time and/or single effective mass to all the carriers. For instance, in low-dimensional systems, several (sub-)bands can
be occupied simultaneously and their contribution to the transport has to be taken into account carefully.

In the most crude approximation, these different carriers are considered to be independent. Thus one can simply write the total tensor of conductance as:

$$\sigma = \sum_i \sigma_i$$  \hspace{1cm} (4.20)

where $i$ labels the bands contributing to transport.

Making the same approximations for each type of carriers as in the previous paragraph we get:

$$\sigma = \sum_i \frac{\sigma_{1D,i}^0}{1 + \mu_i^2 B^2} \begin{pmatrix} 1 & \mu_i B \\ -\mu_i B & 1 \end{pmatrix}$$  \hspace{1cm} (4.21)

This tensor is obviously invertible. However the expressions for $\rho_{xx}$ and $\rho_{xy}$ rapidly become complicated with increasing number of bands. Let us consider the case of two types of carrier only. We find:

$$\rho_{xx} = \frac{(\sigma_{1D,1}^0 + \sigma_{1D,2}^0) + (\sigma_{1D,1}^0 \mu_2^2 + \sigma_{1D,2}^0 \mu_1^2) B^2}{(\sigma_{1D,1}^0 + \sigma_{1D,2}^0)^2 + (\sigma_{1D,1}^0 \mu_2 + \sigma_{1D,2}^0 \mu_1)^2 B^2}$$

$$\rho_{xy} = -\frac{(\sigma_{1D,1}^0 \mu_1 + \sigma_{1D,2}^0 \mu_2) + (\sigma_{1D,1}^0 \mu_2 + \sigma_{1D,2}^0 \mu_1) \mu_1 \mu_2 B^2}{(\sigma_{1D,1}^0 + \sigma_{1D,2}^0)^2 + (\sigma_{1D,1}^0 \mu_2 + \sigma_{1D,2}^0 \mu_1)^2 B^2}$$  \hspace{1cm} (4.22)

We observe that in these conditions, neither the resistance nor the slope of $\rho_{xy}$ is constant when an external magnetic field is swept. Indeed, as Ziman writes [85]:

*When there are two types of carrier, the Hall field must be a compromise between the two [Hall fields that would be created by each type of carrier independently], and therefore cannot be exactly right to keep either carrier from being deflected by the magnetic field and lost to the current.*

An interesting limit of eq. (4.22) happens when $B \to \infty$:

$$\rho_{xx} (B \to \infty) = \frac{\sigma_{1D,1}^0 \mu_2^2 + \sigma_{1D,2}^0 \mu_1^2}{(\sigma_{1D,1}^0 \mu_2 + \sigma_{1D,2}^0 \mu_1)^2}$$

$$\rho_{xy} (B \to \infty) = -\frac{B}{q(n_{1D,1} + n_{1D,2})}$$  \hspace{1cm} (4.23)

The resistance becomes constant and the HE is a direct measure of the total number of carriers in the system. One can understand this result considering that at very high magnetic fields, the difference between velocities of electrons belonging to distinct bands becomes irrelevant.
Boltzmann equation in the low field approximation

In the last two sub-sections the approximations on the mass and scattering time allowed us to obtain formulae for the conductance which often describe the experimental results well. However, there are situations where these approximations are no longer valid and the variations of the scattering time and effective mass on the Fermi surface become relevant. When these complications are taken into account, it brings the modeling of the magnetoresistive phenomena to a new level of complexity.

To illustrate this, let us make no assumption on the Fermi surface geometry or the elastic scattering time. In this case an analytic treatment of eq. (4.15) is possible if we restrict ourselves to low enough magnetic fields (method of Jones and Zener). In this case the operator

\[(1 + \tilde{\Omega})^{-1}\]

can be written as:

\[
(1 + \tilde{\Omega})^{-1} \approx 1 - \tilde{\Omega} + \tilde{\Omega}^2
\]

(4.24)

This leads to the following development of the tensor of conductance:

\[
\sigma_{ij} = \sigma_{ij}^{0} + \sigma_{ij}^{I} + \sigma_{ij}^{II}
\]

(4.25)

with the labels 0, I, II referring to the order in \(B\). In the case of cubic symmetry, at zero temperature and in two dimensions, it can be shown that the only non-zero components are:

\[
\sigma_{xx}^{0} = \sigma_{yy}^{0} = \frac{q^2}{4\pi^2\hbar} \int |v_F| \tau(k) \, ds
\]

\[
\sigma_{xy}^{I} = -\sigma_{yx}^{I} = \frac{q^3 B_z}{4\pi^2\hbar} \int |v_F| \tau(k)^2 \left( \frac{M_{yy}^{-1} - \text{sgn}(v_{F,x}v_{F,y})M_{xy}^{-1}}{M_{yy}^{-1} - \text{sgn}(v_{F,x}v_{F,y})M_{xy}^{-1}} \right) \, ds
\]

\[
\sigma_{xx}^{II} = \sigma_{yy}^{II} = \frac{q^4 \epsilon_{\nu\rho} \epsilon_{\sigma\mu} B_z^2}{2\pi^2\hbar} \int \frac{v_{F,x}v_{F,\rho}}{|v_F|} \tau(k)^3 \left( \frac{M_{\sigma,x}^{-1}M_{\mu,\rho}^{-1} + \frac{v_{F,\mu}}{\hbar} \frac{\partial M_{\sigma,\rho}}{\partial k_x}}{M_{\sigma,x}^{-1}M_{\mu,\rho}^{-1}} \right) \, ds
\]

(4.26)

with \(v_F\) the Fermi velocity and \(\epsilon_{\nu\rho}\) the permutation tensor. The integration is over the Fermi surface (for a 2D system, it is a path integral). We get an expression which directly relates the band structure and the scattering time to the tensor of conductance.

One case of interest is the rather simple situation where the band structure consists of a single isotropic parabolic band but with a scattering time that depends on \(k\). Using eq. (4.26) we find:

\[
\rho_{xx}^{II} - \rho_{xx}^{0} \rho_{xx}^{0} = \frac{e^2 (\tau(k))^2}{m^2} B_z^2 \left( \frac{(\tau(k))^3}{(\tau(k))^3} - \frac{\langle \tau(k) \rangle^2}{(\tau(k))^4} \right)
\]

(4.27)

\[
\rho_{xy}^{I} = -\frac{B_z}{q n_{2D}} \frac{\langle \tau(k) \rangle^2}{(\tau(k))^2}
\]

with:

\[
\langle x \rangle = \frac{\int x \, ds}{\int ds}
\]

(4.28)
4.3 Quantum corrections

In this situation a MR is expected and the HE at low field is renormalized by averages of the scattering time over the Fermi surface. This simple example shows that the link between the magnetotransport and the intrinsic parameters of the system, for example the number of carriers, is not straightforward at all. Analysis of the transport properties under magnetic field should thus go hand in hand with other measurement techniques and theoretical simulations of the system.

4.2.5 Kohler’s rule

Before moving to the magnetotransport of the LaAlO$_3$/SrTiO$_3$ system, let us briefly look at the case of an arbitrary band structure consisting of a single type of carrier with a constant scattering time over the Fermi surface. This is in fact an inverse situation of the one considered in eq. (4.27), because now $\tau$ is a constant but $M^{-1}$ and $v_F$ depend arbitrarily on $k$.

In this case, simply due to the complexity of the integrals to compute, we expect a non-zero MR and a HE not straightforwardly linked to the total number of carriers in the system.

This situation has been investigated by Kohler who postulated that [86]:

$$
\frac{\rho_{xx}(B) - \rho_{xx}(0)}{\rho_{xx}(0)} = h \left( \frac{B}{\rho_{xx}(0)} \right)
$$

(4.29)

with $h$ a function depending on the material and on the geometrical configuration. To any order in $B$, the Kohler’s rule predicts a MR that is a function of the zero-magnetic-field resistance and $B$ only.

A qualitative understanding of this rule can be obtained by noting that in eq. (4.15) $B$ only appears multiplied by $\tau$. Accordingly, the variations in conductance induced by the application of an external magnetic field can be seen as depending on a new variable, $B' = B\tau$. However $\tau$ is not always directly accessible experimentally, hence the choice of $\rho_{xx}(0) \propto 1/\tau$ to rescale the magnetic field scale. This last step relies of course on the fact that neither the band filling nor the band structure change significantly during the measurement.

This scaling has obtained a lot of success in the context of metals and alloys where even when the band structure is complex the hypotheses behind Kohler’s formula are in general well satisfied. The most common test is to record the MR of the system in a region of temperatures where the scattering processes are not expected to change in nature. Then by simply plotting the data as a function of $B/\rho_{xx}(0)$, one could infer whether the system is composed of a single type of carriers with a well defined scattering time and thus get insight into the fundamental details of the electronic system.

4.3 Quantum corrections

Quantum mechanics enters into the semi-classical approach via the description of the particle statistics, of the particle energy and of the scattering processes. However, other quantum phenomena may occur. For instance, an electron experiencing a series
of scattering events can manifest coherent phenomena due to its quantum nature. These effects manifest spectacularly at low temperature and are more relevant for low dimensional systems.

In the following section, we will discuss the principles of phase coherent transport, without providing a complete treatment that is out of the scope of this thesis.

4.3.1 Weak-localization

In Feynman’s path integral approach to quantum mechanics [87], the probability to detect a particle at point \( r_f \) at time \( t_f \) knowing it was at point \( r_i \) at time \( t_i \) is:

\[
P = |K(r_i, r_f; t_i, t_f)|^2
\]

\[
K(r_i, r_f; t_i, t_f) = \sum_{r(t)} \exp \left( i \frac{\hbar}{\hbar} \int_{t_i}^{t_f} \mathcal{L}(\dot{r}(t), r(t)) \, dt \right)
\]

(4.30)

with \( \mathcal{L} \) the Lagrangian of the system and \( r(t) \) a path going from \( A = (r_i, t_i) \) to \( B = (r_f, t_f) \). \( K \) is called the propagator because it describes the propagation of a particle from \( A \) to \( B \), and the sum is over all the possible paths \( r(t) \). Since the sum over \( r(t) \) comes before the absolute value in the expression for \( P \), interference effects are accounted for. This is very different from the classical approach in which the probability of each path would be summed to obtain \( P \).

Let us consider a diffusive system, with elastic collisions only (i.e. no loss of phase coherence induced by the scattering events). In this context, one class of paths of particular interest is the class of closed time reversed paths:

\[
r_2(t) = r_1(t_f - t)
\]

(4.31)

with \( r_1, r_2 \) a pair of time reversed paths, an example of them being displayed in fig. 4.1.

Defining

\[
K_n(r_1, r_1; t_i, t_f) = \exp \left( i \frac{\hbar}{\hbar} \int_{t_i}^{t_f} \mathcal{L}(\dot{r}_n(t), r_n(t)) \, dt \right)
\]

(4.32)

it is easy to show that the contribution of \( K_2 \) to \( K \) equals the contribution of \( K_1 \). As a consequence, when it comes to the calculation of the probability, pairs of closed time reversed paths will interfere constructively, effectively leading to an overall increase of the probability to find the particle at position \( r_i \) at time \( t_f \). Indeed, if we compute the probability of occurrence of a special pair of paths we get:

\[
P = |K_1 + K_2|^2 = |2K_1|^2 = 4|K_1|^2
\]

(4.33)

whereas in the classical limit:

\[
P = |K_1|^2 + |K_2|^2 = 2|K_1|^2
\]

(4.34)

Due to the enhancement of the localization of the electrons, we expect an increase of the electrical resistance. This phenomenon is called weak-localization (WL). In the

66
4.3 Quantum corrections

Figure 4.1: An example of a pair of time reversed paths. This figure has been adapted from [88]. At the big dots, the electron experiences an elastic scattering event.

absence of any phase decoherence, arbitrarily sized closed paths like the ones depicted in fig. 4.1 contribute to this effect. However, inelastic processes, like electron-phonon and electron-electron interactions, limit the lengthscale on which coherent scattering can take place and hence the number of loops contributing to WL. A more complete calculation, taking into account a finite inelastic ($\tau_i$) scattering time in 2D, leads to the following correction to the conductance [88]:

$$\delta \sigma_{xx}^{WL} = -\frac{e^2}{\pi \hbar} \ln \left( \frac{\tau_i}{\tau_{el}} \right)$$  \hspace{1cm} (4.35)

with $\tau_{el}$ the elastic scattering time.

Considering the general temperature dependence of the inelastic scattering time: $\tau_i \propto T^{-p}$ we obtain:

$$\delta \sigma_{xx}^{WL} = \frac{e^2}{\pi \hbar} p \ln(T) + \text{cst.}$$  \hspace{1cm} (4.36)

The existence of time-reversed paths is a consequence of preserved time-reversal symmetry. Accordingly, we expect an external magnetic field to suppress weak-localization as $B$ is known to break time-reversal symmetry. To illustrate this, we introduce a new Lagrangian that includes the effect of the magnetic field on the orbital part of the electron’s wave function:

$$\mathcal{L}' = \mathcal{L} + q v \cdot A$$  \hspace{1cm} (4.37)

with $A = \text{curl}(B)$. Considering $B$ to be perpendicular to the interface plane and using
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Stokes’ theorem to integrate the vector potential term we get:

\[
\frac{i}{\hbar} \int_{t_i}^{t_f} \mathbf{L}'(\mathbf{r}_n(t), \mathbf{r}_n(t)) \, dt = \frac{i}{\hbar} \int_{t_i}^{t_f} \mathbf{L}(\mathbf{r}_n(t), \mathbf{r}_n(t)) \, dt + \frac{i}{\hbar} q \int_{t_i}^{t_f} \mathbf{v} \cdot \mathbf{A} \, dt
\]

\[
= \frac{i}{\hbar} \int_{t_i}^{t_f} \mathbf{L}(\mathbf{r}_n(t), \mathbf{r}_n(t)) \, dt \pm \frac{i}{\hbar} q B S
\]

(4.38)

where \( S \) is the surface delimited by the path \( \mathbf{r}_n(t) \) and \( \pm \) refer, respectively, to the case of a path running counterclockwise or clockwise. Thus, in this situation, the propagators \( K'_1 \) and \( K'_2 \) are not equal. They read:

\[
K'_1 = K_1 e^{-i q B S / \hbar}, \quad K'_2 = K_1 e^{i q B S / \hbar}
\]

(4.39)

leading to the probability:

\[
|K'_1 + K'_2|^2 = 2 |K_1|^2 + 2 |K_1|^2 \cos \left( \frac{2q B S}{\hbar} \right)
\]

(4.40)

Because of the \( \cos \) term in eq. (4.40), that can be positive or negative depending on \( B \) and the area of the path, for \( B \neq 0 \) T the sum over all the paths \( \mathbf{r}(t) \) will reduce the correction to conductance induced by WL. For a macroscopic sample it was shown using diagrammatic methods that the magnetoconductance takes the following form \((\tau_i \gg \tau_{el})\) [88] :

\[
\Delta \sigma^{WL}_{xx}(B) = \delta \sigma^{WL}_{xx}(B) - \delta \sigma^{WL}_{xx}(0) = \frac{e^2}{\pi \hbar} \left[ \psi \left( \frac{1}{2} + \frac{B_i}{B} \right) - \psi \left( \frac{1}{2} + \frac{B_{el}}{B} \right) + \ln \left( \frac{B_{el}}{B_i} \right) \right]
\]

(4.41)

with \( \psi \) the digamma function, \( B_i = \hbar / 4 e D \tau_i, \ B_{el} = \hbar / 4 e D \tau_{el} \) and \( D \) the diffusion constant. A set of curves computed using eq. (4.41) is shown in fig. 4.2. First, we see that increasing \( \tau_i \) leads to a sharpening of the minimum around \( B = 0 \) T. This is a consequence of the increase of the WL phenomenon induced by the increase in the number of (coherent) paths for which eq. (4.40) has to be evaluated.

Secondly, if the magnetic field is strong enough to induce a complete rotation of the phase of the electron during the shortest time-reversed path (of area \( \sim l_{el}^2 \approx (v_F \tau_{el})^2 \)), we expect the WL phenomena to be almost canceled. This happens at the field \( B^* \) (shown on fig. 4.2 as the border between the green and white areas):

\[
\frac{2q B^* l_{el}^2}{\hbar} = 2\pi
\]

\[
B^* = \frac{h \pi}{q l_{el}^2}
\]

(4.42)

4.3.2 Weak anti-localization

In the previous discussion, we neglected the spin of the electron, assuming its state to be conserved along the propagation. But, for instance, in the presence of spin-orbit
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Figure 4.2: Magnetoconductances (MC) in the WL regime. The depth of the minimum is controlled by \( \tau_i \) while the width of the dip is controlled by \( 1/\tau_{el} \).

coupling, the diffusive motion of the electron will imply rotations of its spin. In this context it is useful to define a spin-resolved propagator:

\[
K^so(r_i, r_f; t_i, t_f; s_i, s_f) = \sum_{r(t)} \exp \left( -\frac{i}{\hbar} \int_{t_i}^{t_f} L(\dot{r}(t), r(t)) \, dt \right) \langle s_f | \hat{R}(r(t)) | s_i \rangle
\]

with \( |s_i\rangle \) and \( |s_f\rangle \) the initial and final spin-state, and \( \hat{R} \) the operator representing the spin-rotation undergone while traveling along \( r(t) \).

Considering again the case of a pair of closed time-reversed paths in the absence of external magnetic field, we get the two following spin-resolved propagators:

\[
K^so_1 = K \langle s_i | \hat{R} | s_i \rangle
\]

\[
K^so_2 = K_2 \langle s_i | \hat{R}^{-1} | s_i \rangle = K_1 \langle s_i | \hat{R}^\dagger | s_i \rangle
\]

Thus the probability of finding the electron at position \( r_i \) at time \( t_f \) having experienced the spin-rotation \( \hat{R} \) reads:

\[
P_{\hat{R}} = \sum_{|s_i\rangle} |K^so_1 + K^so_2|^2 = |K_1|^2 \left( 2 + 2Re \left( \langle s_i | \hat{R}^2 | s_i \rangle \right) \right)
\]

At this point, two interesting limits can be considered. The first one corresponds to the case with no spin-orbit scattering (i.e. no rotation of the spin). It implies taking \( \hat{R} \) to be the identity operator. In this case it is easy to show that \( \langle s_i | \hat{R}^2 | s_i \rangle = 1 \) so that we recover the weak-localization correction \( (P = 4|K_1|^2) \). The second one corresponds to the case of a very strong spin-orbit interaction so that the spin of the electron is completely randomized as it travels along \( r_{1,2}(t) \). In this case we need to average over
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![Diagram showing examples of magnetoconductance curves in three regimes of WL/WAL.]

**Figure 4.3:** Examples of magnetoconductance curves in three regimes of WL/WAL. Black curve: $\tau_{el} < \tau_i < \tau_{so}$. Red curve: $\tau_{el} < \tau_{so} < \tau_i$. Blue curve: $\tau_{so} \leq \tau_{el} < \tau_i$.

all the possible rotations. We get :

$$
\mathcal{P} = \frac{1}{4\pi^3} \int_0^{2\pi} d\alpha \int_0^\pi \int_0^{2\pi} \mathcal{P}_R d\beta d\gamma = |K_1|^2 \left( 2 + 2 \left( -\frac{1}{2} \right) \right) = |K_1|^2 \tag{4.46}
$$

with $\alpha$, $\beta$, $\gamma$ the Euler angles. Strikingly, averaging over all the possible spin rotations reduces the classical probability to half of its classical value. This phenomenon is called weak-antilocalization (WAL).

The presence of an external magnetic field suppresses this positive contribution to the conductance, hence decreasing the conductance value. However, a more careful analysis shows that to predict the shape of the MC the relative values of $\tau_{el}$, $\tau_i$, and $\tau_{so}$ have to be considered (with $\tau_{so}$ the spin-orbit scattering time). Three regimes can be identified and are illustrated in fig. 4.3.

The first one, $\tau_{el} < \tau_i < \tau_{so}$, leads to a completely positive MC. Indeed, this is the case of weak localization for which the value $\tau_i < \tau_{so}$ does not allow the randomization of the spin direction, brought by the spin-orbit interaction, to be effective before phase decoherence.

In the second regime where $\tau_{el} < \tau_{so} < \tau_i$, two types of time reversed path come up: those of duration between $\tau_{so}$ and $\tau_i$ and those of duration between $\tau_{el}$ and $\tau_{so}$. The first type are expected to lead to WAL while the second type are expected to lead to WL. Since an external magnetic field sets an upper value to the size of the interfering time-reversed path, a negative MC appears at low field followed by a positive MC at higher fields (i.e. at low field the effect of the magnetic field is to cancel the contribution of the paths of duration ranging between $\tau_{so}$ and $\tau_i$).

Finally, if $\tau_{so} \approx \tau_{el} < \tau_i$ the spin is randomized on all the time-reversed paths. Thus a completely negative MC is observed.
4.3 Quantum corrections

In addition to its effect on the orbital part of the electron’s wave function, an external magnetic field also tries to anti-align the electron’s spin via the Zeeman interaction. Thus, as the spin will tend to get a preferential direction, we expect weak-antilocalization to be diminished by the Zeeman contribution. This situation has been investigated using diagrammatic techniques by Maekawa and Fukuyama [89]. The perturbative nature of this approach reduces its range of validity to not too strong spin-orbit interaction and to low magnetic fields ($\tau_{el} \ll \tau_{so}$, $\tau_B = \frac{l_B^2}{B}$ and $l_B = \sqrt{\frac{\hbar}{eB}}$), however, as we shall see later, it will be very useful in describing the magnetotransport of the LAO/STO interface. The Maekawa-Fukuyama formula for the MC in the weak-(anti)localized regime reads:

\[ \Delta \sigma_{\text{ex}}^{W(A)L}(B) = \frac{e^2}{\pi \hbar} \left[ \Psi \left( \frac{B}{B_i + B_{so}} \right) + \frac{1}{2\sqrt{1 - \gamma^2}} \Psi \left( \frac{B}{B_i + B_{so}(1 + \sqrt{1 - \gamma^2})} \right) \right. \]

\[ - \left. \frac{1}{2\sqrt{1 - \gamma^2}} \Psi \left( \frac{B}{B_i + B_{so}(1 - \sqrt{1 - \gamma^2})} \right) \right] \tag{4.47} \]

with $\Psi(x) = \ln(x) + \psi\left(\frac{1}{2} + \frac{1}{x}\right)$, $B_{so} = \hbar/4eDB_{so}$, $\gamma = g^*\mu_BB/(4eDB_{so})$ and $g^*$ the effective g factor.

4.3.3 Relationship between $\tau_{el}$ and $\tau_{so}$

Is there a relation between the elastic and the spin-orbit scattering time? Naively one can imagine an electron undergoing series of scattering events, some of them effectively leading to a rotation of its spin, for example due to the presence of heavy impurities acting as strong spin-orbit scattering centers. So, the SO scattering time is a multiple of the elastic scattering time; this occurs for atomic spin-orbit interaction. This relation takes the name of Elliot-Yafet relation:

\[ \tau_{so} \propto \tau_{el} \tag{4.48} \]

However, when inversion symmetry is broken, another class of mechanisms leading to spin relaxation emerges, the so called D'yakonov Perel mechanisms, for which, contrarily to the Elliot-Yafet case, the spin rotations do not occur at the scattering events but in-between the collisions. This is possible, as the breaking of inversion symmetry, and the associated electric field, induce an effective $k$-dependent magnetic field acting on the spin of the electron. Thus, in-between two collisions the spin precesses at the Larmor frequency by an angle $\delta \phi = \omega_{\text{Larmor}}\tau_{el}$. The sequence of rotations after each scattering event on a time $t \gg \tau_{el}$, is efficiently described by a random walk of the spin on the Bloch sphere. From the central limit theorem we get that the variance of $\delta \phi$ goes as:

\[ \langle \delta \phi^2 \rangle = \omega_{\text{Larmor}}^2\tau_{el} t \tag{4.49} \]
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Figure 4.4: Detail of the magnetic field orientations used in this thesis. $\phi$ is the angle between the current ($J$) and the in-plane magnetic field $B_{\text{in-plane}}$.

It is natural to define the spin-orbit relaxation time as the time needed to lose the information on initial spin orientation ($\langle \delta \phi^2 \rangle \approx 1$). Thus we get:

$$\tau_{\text{SO}} \approx \frac{1}{\omega^2_{\text{Larmor}} \tau_{\text{el}}} \propto \frac{1}{\tau_{\text{el}}} \quad (4.50)$$

This is the D’yakonov Perel relation, and it applies for example in systems with Rashba and/or Dresselhaus spin-orbit interactions.

4.4 The magnetotransport of the LaAlO$_3$/SrTiO$_3$ interface

4.4.1 Out-of-plane magnetic field

We now turn to the magnetotransport of the LaAlO$_3$/SrTiO$_3$ interface. We will first consider the case of a magnetic field applied perpendicular to the conducting plane (see geometry in fig. 4.4).

In chapter 3, we showed preliminary analyses of the LaAlO$_3$/SrTiO$_3$ magneto-transport data using a simple approach. Here, we refine these treatments taking into account the theoretical developments presented in the previous sections and using field effect to discriminate between different scenarios.

4.4.1.1 Temperature dependence

In fig. 4.5a and b, we show respectively the relative MR and the transverse resistance $R_{xy}$ as a function of temperature for a LaAlO$_3$/SrTiO$_3$ interface grown at 800 °C. Clearly as the temperature is decreased, non-zero MR and non-linear HE strongly develop. This is more easily seen in fig. 4.5c and d where we plot as a function of temperature the amplitude of the relative MR at 7 T and the relative difference between the slope of $R_{xy}$ calculated at high field and at low field. Within our experimental resolution, finite MR and non-linear HE are present at temperatures as high as 150 K.
4.4 The magnetotransport of the LaAlO$_3$/SrTiO$_3$ interface

Figure 4.5: Temperature dependence of the out-of-plane magnetotransport of an ungated LaAlO$_3$/SrTiO$_3$ interface. a and b, respectively, relative magnetoresistance (MR) and Hall effect (HE) for temperature ranging between 1.5 and 260 K. c, amplitude of the relative magnetoresistance at 7 T. d, relative difference between the slope of $R_{xy}$ at 7 T and 0 T (in absolute value). The color code indicates the temperature in agreement with data in graph c or d.
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In fig. 4.6 we plot the inverse of the Hall coefficient ($R_H = R_{xy}/B$) calculated at different magnetic field strengths, and the ratio between the Hall coefficient and the zero-field sheet resistance. In a model consisting of a single parabolic band with isotropic scattering time, these two quantities correspond respectively to the carrier density and to the mobility.

Above 150–200 K, the evolution of these transport properties follows a simple rule, the Hall coefficient being roughly constant as a function of magnetic field and temperature, and the inferred mobility following a $T^\alpha$ evolution with $\alpha \sim -2.3$. Probably, this is due to the averaging of the electronic properties around the Fermi surface, caused by the broadening of the Fermi distribution and the dominant phonon scattering. Thus at high temperature, the total number of mobile carriers is likely to be directly readable on fig. 4.6 and would be $\sim 6.8 \times 10^{13}$ cm$^{-2}$. At lower temperature, the parameters extracted in fig. 4.6 are very much dependent on both temperature and magnetic field.

The central question is now to understand the origin of the low temperature deviation from the behavior observed at the highest temperatures. Interestingly enough, it could be linked to the cubic to tetragonal transition of SrTiO$_3$ (observed at 105 K in the bulk). Indeed it was shown experimentally that the surface of SrTiO$_3$ has a tendency to undergo this transition at temperatures as high as 150 K [90–93]. As we have seen in chapter 2, this transition lifts the orbital degeneracy.

Concerning the large magnetoresistance, similar observations of anomalously large and quasi-linear behavior have been reported for doped silver chalcogenides [94]. Parish and Littlewood have proposed an interesting scenario to explain such anomalous behavior: the magnetoresistance could originate from a disordered and inhomogeneous semiconductor that should be described as a network of resistances [95]. This model is not very sensitive to microscopic processes but only to the macroscopic conductivity of each node. In our situation, the structural transition of SrTiO$_3$ generates tetragonal domains oriented in different directions across the sample (for a recent description see [96]). These domains could generate the network of resistances required by the Parish and Littlewood model and hence determine the large and quasi-linear magnetoresistance.
This interpretation requires further investigation, in particular in relation with the field effect experiments where an electric field acts on the distribution of the tetragonal domains and in relation to recent measurements on nanodevices where conductance is likely to be restricted to a monodomainal area.

In a less disordered semiconductor, it is well known [97–101] that, at low temperature, a positive transverse magnetoresistance often develops. This has been attributed to an energy dependent scattering time. For example, if one makes the assumption of a mean free path, \( l \), which is constant in energy then the scattering time evolves with energy \( \epsilon \) as:

\[
\tau(\epsilon) = \frac{l}{v_F} \propto \frac{1}{\sqrt{\epsilon}}
\] (4.51)

The above relation pertains to an acoustic deformation potential scattering: for other scattering mechanisms, the mean free path is not constant and the power law linking \( \tau \) and \( \epsilon \) is modified. In the following, we use \( \tau \propto \epsilon^{-1/2} \) as our conclusions are not changed by making a different reasonable choice.

It can be shown [102, 103] that, in the isotropic case and approximating the Fermi distribution by the Boltzmann one, eq. (4.51) leads to the following magnetoresistance (at low magnetic field):

\[
\frac{R_s(B) - R_s(0)}{R_s(0)} \approx \alpha \mu^2 B^2
\] (4.52)

with \( \mu \) the mobility of the charge carriers and \( \alpha \) a factor which depends on the dimensionality, the scattering mechanism and on the relative orientation of \( B \) and \( J \). For example, in the 3D case with \( B \) normal to \( J \), \( \alpha \approx 0.38 \) while in 2D, with \( B \) normal to the conducting plane, \( \alpha = 0.93 \) (with \( \tau \propto \epsilon^{-1/2} \)).

In fig. 4.6, we have seen that the Hall mobility at the LaAlO\(_3\)/SrTiO\(_3\) interface seems to increase markedly as the temperature is reduced. Hence, it is very tempting to attribute the large increase in the amplitude of the magnetoresistance observed in fig. 4.5 to the energy dependence of the scattering time. Yet, a more careful analysis of our experimental situation shows that, contrarily to the semiconductor case, it is not correct to use the Boltzmann distribution to approximate the Fermi-Dirac statistics in our case. In fact, using the Fermi-Dirac distribution to compute the low field magnetoresistance of a 2D isotropic electron gas in the constant mean free path approximation leads to:

\[
\frac{R_s(B) - R_s(0)}{R_s(0)} \approx h(\eta)^2 \mu^2 B^2
\] (4.53)

with \( h \) a function of \( \eta = \frac{\epsilon_F}{k_B T} \) only, that can be numerically computed and roughly fitted to \( \eta^{-1.55} \). This is the reason why in metals (which have \( \epsilon_F \gtrsim 1 \text{ eV} \)) with isotropic Fermi surfaces no magnetoresistance is expected to come from the energy dependence of \( \tau \) [104, 105]. This can be easily understood by noting that, at low temperature, the Fermi Dirac distribution selects the states with same energy, hence the energy dependence of \( \tau \) does not play a role.

As shown in chapter 2, the Fermi energy at the LaAlO\(_3\)/SrTiO\(_3\) interface is typically on the order of 50–100 meV. This implies that \( h(\eta) \approx 0.0001 \) at 1.5 K and \( h(\eta) \approx 0.4 \) at room temperature. Thus, in this scenario, even with the large temperature dependence of the Hall mobility shown above, we would not expect a magnetoresistance larger
Figure 4.7: Kohler plot of the out-of-plane magnetotransport of an ungated LaAlO₃/SrTiO₃ interface. Relative magnetoresistance (MR) as a function of the magnetic field divided by the zero-field sheet resistance. The panel on the right is a low-field close-up. The same color code as in fig. 4.5 is used.

than $2 \times 10^{-5}$ at 7 T at any point in the temperature range we explored. Moreover, since $h(\eta)$ strongly decreases with $T$, the amplitude of the magnetoresistance would be expected to be reduced at low temperature, in contradiction with the evolution shown in fig. 4.5.

As mentioned earlier, a tool of choice in the analysis of the magnetoresistance data is Kohler’s rule. We show a test of this scaling in fig. 4.7, using the same data as in fig. 4.5; evidently, our data do not scale according to Kohler’s rule. Similar failure of scaling has already been observed, for example in the normal state of cuprate superconductors or in quasi-two-dimensional organic metals [106, 107] and implies that the band structure of the system is not composed of a unique band with a unique scattering time.

Among the invoked scenarios to explain the failure of this scaling are:

- A scattering time that varies around the Fermi surface and whose temperature dependence is different for different regions of the Fermi surface.

- Temperature dependent band structure parameters, leading to a dependence of the Fermi surface geometry on temperature.

- Multiple band population, with the associated mobilities not sharing the same temperature dependence.

The first proposition probably implies, in our case, a very large modulation of the scattering time around the Fermi surface. Indeed, considering eq. (4.27), an isotropic Fermi surface and (arbitrarily) choosing a scattering time evolving around the Fermi surface as $\tau_0 (1 - p \cos \phi)$, we find that the anisotropy parameter $p$ has to reach values of at least 0.5 in order to reproduce the large magnetoresistance we observe at low temperature. The same result holds irrespective of the orientation of the anisotropy. In chapter 2, we have seen that, in SrTiO₃, the conduction band states are built from the
very anisotropic $d$–orbitals of Ti. It would be interesting to investigate theoretically the result of such an anisotropy on the scattering time and if, combined with the anisotropy in the effective mass it can give rise to the large magnetoresistance observed in this section.

Given the huge temperature dependence of the dielectric constant of bulk SrTiO$_3$ between 1.5 and 105 K, and the shift in the transition temperature observed at the surface of this material, the second suggestion needs some consideration. Indeed, since the potential well in which the mobile charges evolve is strongly influenced by the dielectric constant of SrTiO$_3$ via Poisson’s equation, the band structure (see chapter 2) could also change dramatically. However, as we have seen, the confining electric field damps the dielectric constant of SrTiO$_3$ at low temperature. Thus advanced theoretical modelings are necessary to quantify the real impact of the temperature dependence of the polarizability of SrTiO$_3$ on the band structure of the interface. From our data, it is clear that the change in band structure has to be dramatic to explain by itself the pronounced features observed here.

Due to the multi orbital character of the conduction in SrTiO$_3$ and to the relatively high effective masses of the charge carriers along the [0 0 1] direction, we could observe in the preceding chapters that the band structure of the interface is probably composed of several sub-bands. Depending on their relative filling and associated mobilities, it could well be that they give rise to the features observed here. This third scenario has the advantage to be partially testable, using for the MR and the HE the analytical results obtained for multi band conduction with isotropic scattering times (eq. (4.22)). In what follows we limit ourselves to a two channel model as it requires only four parameters (a similar approach is presented in [108, 109]).

Figure 4.8 presents the results of this analysis for temperatures ranging from 1.5 to 100 K. The fits using eq. (4.22) (black lines, graphs a and b), show an overall agreement with the data even though below 30 K significant deviations between theory and experiment are observed in $R_s(B)$. We would like to stress here that the theoretical curves were optimized to fit both $R_s(B)$ and $R_{xy}(B)$ since in this model the transport parameters ($n_{2D}, \mu$) of the bands determine completely the tensor of resistance.

Evidently, the quality of the low temperature fits could be enhanced repeating the same procedure with an additional carrier type because it would imply 6 instead of 4 free parameters. However, we do not think it is a good direction to take since in establishing eq. (4.22) other assumptions were made, like the parabolic dispersion, which, considering recent band structure calculations, that include the effect of spin-orbit coupling [60,67,74,75], should be released.

Figures 4.8c and d show the temperature evolution of the transport parameters resulting from the two band fits. We obtain two types of carrier, both n-type, with very different characteristics, the first one having a low density with a mobility above 2000 cm$^2$V$^{-1}$s$^{-1}$ and the second being 4 times denser with a mobility around 500 cm$^2$V$^{-1}$s$^{-1}$. We observe that $n_{2D}$ for the low density species is increased by $\sim$50% when the temperature is varied from 100 to 1.5 K while in the same range of temperature the carrier density of the denser species is decreased by $\sim$70% leading to a global reduction of the total electronic population as the temperature is decreased. Carrier freezing could be invoked to explain the decrease in population of the second
Figure 4.8: Fitting of the out-of-plane magnetotransport of an ungated LaAlO$_3$/SrTiO$_3$ interface using an isotropic two band model. a and b, respectively, relative magnetoresistance (MR) and Hall effect (HE) for temperature ranging between 1.5 and 66 K. Curves are shifted for clarity. Colored lines correspond to the experimental data while black lines are the fits. c and d, temperature evolution of the four fitting parameters ($n_{2D,1}$, $n_{2D,2}$, $\mu_1$, $\mu_2$) of the theoretical curves presented in a and b. Triangles and rhombuses refer to the two carrier species. The color code indicates the temperature in agreement with data in graph c or d.
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group of carriers.

Analysing the fitting parameters in fig. 4.8c and d an important point has to be stressed. Compared to the low temperature measurements, above 30 K, the evolution of $R_s(B)$ and $R_{xy}(B)$ is simpler (in the range of our accessible magnetic fields): the magnetoresistance is quadratic and the Hall effect is almost linear. This implies that only 3 fitting parameters are needed to describe satisfactorily the shape of $R_s(B)$ and $R_{xy}(B)$ (i.e. $R_s(B) = a + bB^2$ and $R_{xy}(B) = cB$). Hence, in this range of temperature, our fit is underdetermined and the extracted parameters are less reliable.

Yet, our analysis shows that the multi band scenario is a serious candidate for the origin of the large low temperature magnetoresistance and non-linear Hall effect observed at the LaAlO$_3$/SrTiO$_3$ interface. Indeed, it reproduces correctly the amplitude of the phenomena and the typical field at which non-parabolic and non-linear contribution respectively appear in the magnetoresistance and Hall effect data. However, being it that the fits are not perfect in the low $T$ regime, one should keep in mind that more subtle effects coming from the population of additional sub-bands, the non-diagonal form of $M_e^{-1}$ and the evolution of the scattering time around the Fermi surface can possibly alter the reliability of the computed charge densities and mobilities.

Within the multi band model we also expect the high magnetic field estimation of $R_H$ to lead to a reliable estimation of the total carrier density (c.f. eq. (4.23)). Figures 4.5 and 4.6 show that, while at 7 T the field dependence of $R_{xy}$ is not completely linear, the number of carriers extracted at 1.5 K for the highest fields is far from the room temperature estimation. On the other hand, we note that, in line with a multi band scenario, going to high fields the temperature dependence of $1/(R_{He})$ is damped. One possibility is the presence of carrier freezing. In these conditions, at low temperature, the total number of carriers would be really different from that at high temperature and should be evaluated at the highest magnetic field.

4.4.1.2 Field effect experiments

We now move to the analysis of the low temperature dependence of the magnetotransport under field effect applied in a back gate geometry (see section 3.5).

In fig. 4.9 we present the same analysis of the MR and HE as in fig. 4.5, using this time the field effect to tune the electronic properties of the interface. Sheet conductance in the absence of magnetic field ($\sigma_{02D}$) is used in order to define the state of the system because it is better suited than gate voltage for comparing data from different samples (see chapter 3). As an illustration, we plot in fig. 4.9c and d the data extracted at low temperature for the sample studied in the previous paragraphs (purple crosses). Using the sheet conductance criterium, we can observe that the two samples fall on the same curve, even with having been grown 4 years and 400 samples apart.

Strikingly, the behavior observed in fig. 4.9 is very similar to that observed in fig. 4.5. Indeed, above a certain threshold sheet conductance (~1-2 mS), finite positive MR and non-linear HE appear. Since field effect tunes the chemical potential of the system, it is very tempting to consider a change in (sub-)band population leading to a multi band conduction.

In an attempt to corroborate this, we plot in fig. 4.10, the doping evolution of the inverse of $R_H$ and of the ratio between $R_H$ and the zero field sheet resistance. At low
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Figure 4.9: Gate voltage dependence of the low temperature out-of-plane magneto-transport of a LaAlO$_3$/SrTiO$_3$ interface. a and b, respectively, relative magnetoresistance (MR) and Hall effect (HE) recorded at various gate voltages. c and d, respectively, amplitude of the relative MR at 7 T and relative difference between the slope of $R_{xy}$ at 7 T and 0 T (in absolute value). The purple crosses indicate the values extracted from the ungated sample presented in the preceding paragraphs. Sheet conductance at zero magnetic field ($\sigma_{2D}^0$) is used as an indicator of the state of the system or, equivalently, of its filling level and defines the color code in agreement with data in graph c or d.
magnetic field, the inverse of the Hall coefficient follows a rather peculiar evolution, which, if interpreted as an estimation of the carrier density, would imply a reduction in the electronic density. Moving to higher fields, $1/(R_{He})$ recovers a positive evolution but, surprisingly, displays a plateau around 1-2 mS.

As a second check for the evolution of the total carrier density we added in fig. 4.10 the estimation of the variation of the carrier density obtained using differential capacitance measurements at low temperature (green dashed line, offset for a better comparison). Clearly, these measurements point to a monotonic increase of the carrier density as the gate voltage is increased, with a slope (in log-log scale) very consistent with the evolution of the inverse Hall coefficient below $\sim 0.5$-1 mS. Above this value, however, this capacitance data does not show any saturation of the carrier density.

To clarify this situation, we present in fig. 4.11 and 4.12 the very same analysis on data acquired up to a magnetic field of 14 T, on another interface grown in the same conditions. Since the measurements were performed in a dilution refrigerator the temperature is now much lower and superconductivity is observed at low $B$. To check for consistency, we added to fig. 4.11c and d the evaluation of the transport properties at 7 T. Clearly they are very similar to the data obtained so far.

The effect of moving to higher $B$ is twofold. First, the onset of non-linear HE and finite positive MR is shifted to lower conductance. We note, however, that this effect is partly due to the temperature dependence of the zero field sheet conductance. Secondly, the width of the plateau in the evolution of $1/(R_{He})$ as a function of sheet conductance is reduced and the measurements performed at the highest conductances and magnetic field are aligned with the extrapolation of the data below 0.6 mS. This analysis suggests that, at high doping, the Hall coefficient at 14 T is a good estimation of the total number of mobile carriers.

To get an estimate of the magnetic field needed to attain a correct estimation of
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Figure 4.11: Gate voltage dependence of the low temperature out-of-plane magnetotransport of a LaAlO$_3$/SrTiO$_3$ interface up to 14 T. a and b, respectively, relative magnetoresistance (MR) and Hall effect (HE) recorded at various gate voltages. c and d, respectively, amplitude of the relative MR at 7 and 14 T and relative difference between the slope of $R_{xy}$ at 7 or 14 T and 0 T (in absolute value). Sheet conductance at zero magnetic field is used as an indicator of the state of the system or, equivalently, of its filling level and defines the color code in agreement with data in graph c or d. Due to the presence of superconductivity, the relative amplitudes are calculated using an extrapolation of the data below $\approx 1$ T.
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Figure 4.12: Gate voltage and magnetic field dependence of the electronic properties up to 14 T. Gate voltage (or sheet conductance) and magnetic field evolution of the inverse of the Hall coefficient and of the ratio between the Hall coefficient and the sheet resistance at zero magnetic field.

For comparison, we show in fig. 4.14, a close-up on the low conductance regime of fig. 4.9a, the MC curves analyzed in [27] and, for completeness, the MR of 2DEGs in InAlAs/InGaAs/InAlAs quantum wells, where Rashba spin-orbit coupling was successfully tuned by changing the degree of structure inversion asymmetry [111]. Clearly,

In the first place, the effect of the magnetic field is almost always to increase the resistance. The few special cases of negative MR are probably due to quite a different mechanism [than the one described by the semi-classical approach], – [...].

In the case of LaAlO$_3$/SrTiO$_3$ interfaces, the origin of the negative MR was convincingly ascribed to the effect of an external magnetic field on a 2D weak (anti)-localized state by Caviglia et al. [27]. The authors could show that the $B$ dependence of the magnetotransport is quantitatively captured for a broad range of gate voltages using a weak-localization (WL) expression for non interacting fermions [89]. Very interestingly, they could also link the magnetic field and gate voltage dependence of the resistance with a steep rise of a linear Rashba-type spin-orbit interaction. In sheet conductance “language”, their fitting was successful below $\sim 1$ mS.
Figure 4.13: Saturation of the Hall resistance slope across the phase diagram. Magnetic field derivative of the Hall coefficient extracted at 13.5 T, as a function of the sheet conductance.

Figure 4.14: Low doping out-of-plane magnetotransport of a gated LaAlO$_3$/SrTiO$_3$ interface. a Close-up on the low conductance regime of fig. 4.9. b Magnetoconductance (MC) curves analyzed in [27, 110]. c Magnetoresistance (MR) of 2DEGs in InAlAs/InGaAs/InAlAs quantum wells where the structure inversion asymmetry was tuned via impurity doping [111].

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the measurements performed on LaAlO$_3$/SrTiO$_3$ interfaces mimic those performed on InAlAs/InGaAs/InAlAs quantum wells.

Accordingly, we repeat the same analysis as in [27] using the data of fig. 4.9 and the Maekawa-Fukuyama expression for the MC (see 4.3.2). The accuracy of the fits allows us to estimate the values of the inelastic field, $B_i$, and of the spin-orbit field $B_{so}$ as a function of the gate voltage $V_g$ (or equivalently $\sigma_{0}^{2D}$). Figure 4.15 displays the evolution of these two quantities and of the superconducting $T_c$ versus $\sigma_{0}^{2D}$.

As mentioned above, previous studies [27, 28] have shown that a linear Rashba model can describe important properties of the magnetotransport at the LaAlO$_3$/SrTiO$_3$ interface. This implies that we can determine the effective Drude mass $m^*$ using the D’yakonov-Perel’ relation between the spin-orbit ($\tau_{so}$) and elastic ($\tau_{el}$) scattering times:

$$\frac{2\pi}{\tau_{so}} = \Omega_{so} \frac{2}{\tau_{el}}$$  \hspace{1cm} (4.54)

where $\Delta_{so} \equiv h\Omega_{so} = 2\lambda E_z k_F$ is the Rashba energy ($\lambda$ is the material specific Rashba spin-orbit coefficient, $E_z$ is the interfacial electric field along $z$, the direction normal to the interface, $k_F$ is the Fermi momentum) [112].

Indeed, in the diffusive regime, the spin-orbit field is defined as $B_{so} = \frac{\Phi_0}{4\pi D\tau_{so}}$, where $\Phi_0 = \frac{\hbar}{2}$ and $D = \frac{1}{2} v_F^2 \tau_{el}$ is the Drude (bare) diffusion constant (i.e. without the WL corrections). Using the D’yakonov-Perel’ relation, it follows that $D\tau_{so} = \frac{1}{2} v_F^2 \frac{2\pi}{\tau_{el}}$.

Taking into account the fact that $m^* v_F = \hbar k_F$ and that $\Omega_{so} = \frac{2\lambda E_z k_F}{\hbar}$ we obtain

$$\frac{\Phi_0}{B_{so}} = \frac{h^4}{4\pi D\tau_{so}} = \frac{1}{(4\pi)^2} \frac{1}{(\lambda E_z m^*)^2}$$  \hspace{1cm} (4.55)

Hence

$$m^* = \frac{\hbar^2}{4\pi \lambda E_z \sqrt{\Phi_0}}$$  \hspace{1cm} (4.56)

For the estimation of $\tau_{el}$ (or $B_{el}$ the elastic magnetic field) the value of $\sigma_{0}^{2D}$ in absence of WL is needed [68].

$$\sigma_{0}^{2D} = \sigma_{0}^{2D} - \frac{e^2}{\pi \hbar} \ln[(1 + \frac{B_{so}}{B_i})(1 + \frac{2B_{so}}{B_i})] - \ln[\frac{B_{el}}{B_i}]$$  \hspace{1cm} (4.57)

We get:

$$\tau_{el} = \frac{\sigma_{0}^{2D} m^*}{n_{2D} e^2}$$

$$D = \frac{\pi \hbar^2 \sigma_{0}^{2D}}{e^2 m^*}$$

$$B_{el} = \frac{\Phi_0}{4\pi D\tau_{el}}$$  \hspace{1cm} (4.58)

When eq. (4.56) holds, i.e. in the range of $V_g$ such that spin-orbit terms contribute to diffusive processes, we consider that $\lambda$ has little $V_g$ dependence. By contrast, changing the gate voltage modifies the sheet carrier density $n_{2D}$ and causes a variation of $E_z$. We model the change in $E_z$ through a simple electrostatic model of the confined
Figure 4.15: (a) Evolution of the inelastic ($B_i$) and spin-orbit ($B_{so}$) fields as a function of $\sigma_{2D}^0$ for the sample of this work and a reference sample (data from ref. [27]) in the diffusive regime. The top voltage scale indicates the corresponding applied voltages for the reference sample. The superconducting phase (2D SC, blue region) and the transition temperature $T_c$ (triangles) are also shown. (b) Evolution of the elastic ($B_{el}$) and spin-orbit fields with $\sigma_{2D}^0$ highlighting the crossing at $\sigma_{2D}^0 \sim 0.6$ mS. Data points extend slightly beyond the diffusive regime. (c) Variation of the transport effective mass $m^*$ with $\sigma_{2D}^0$. Here $m^*_{max} \sim 2.2m_e$. WL, WAL, AM shaded regions denote the weak localization, weak antilocalization and anomalous metal regimes respectively.
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electron gas, taking into account the electric field dependence of the SrTiO$_3$ dielectric constant [64] and taking $n_{2D}$ as a linear function of $V_g$. Using ARPES measurements on cleaved SrTiO$_3$ surface, the authors in [113] found a confining electric field of 83 MV/m, in the triangular potential well approximation. This value is similar to the one we calculated in chapter 2, in the presence of in-gap states and for the sub bands closest to the Fermi level. We assign this value to $E_z$ when $\sigma_{2D}^0 \sim 1.1$ mS, as we surmise that the field $E_z$ on the SrTiO$_3$ side of the interface is caused by all the charge carriers whether they are mobile or not.

Using these prescriptions, we present the variations of $B_{el}$, $B_{so}$ and $m^*$ with $\sigma_{2D}^0$ (or $V_g$) in fig. 4.15b and c. To highlight the trends, we include data points slightly beyond the diffusive regime (see below). We note that $m^*$ is $\sim 3.5$ times lighter in the underdoped regime than in the optimally doped regime, which is consistent with the picture of interface charge transport evolving from being $d_{xy}$-dominated to being $d_{xz}/d_{yz}$-dominated, upon increasing $V_g$ beyond $\sigma_{2D}^0 \sim 0.5$ mS [12, 23, 27–29, 108, 114]. Thus, $\sigma_{2D}^0 \sim 0.3$ mS signals a transition from a single band to multi band conduction at the LaAlO$_3$/SrTiO$_3$ interface.

In addition to that, as reported in [27, 28], $\tau_{so}$ decreases sharply across a threshold sheet conductance, to the extent that $\tau_{so} \sim \tau_{el}$ for $\sigma_{2D}^0 \sim 0.6$ mS (fig. 4.15b). When this occurs spin-orbit processes cease to be diffusive. The spin-orbit time is no longer given by the D’yakonov-Perel’ expression and the Rashba term becomes a bona fide new energy scale in the problem, on equal footing with the kinetic part. Band structure needs to be recalculated in the presence of the (Fermi surface reconstructing) Rashba Hamiltonian. The concomitant sharp rise in the elastic scattering time is thus suggestive of a scenario of spin-orbit protected transport (against disorder) in the 2D conducting sheet. We also note the saturation of $\tau_{el}$ for $\sigma_{2D}^0 < 0.1 – 0.2$ mS which suggests that in the insulating region of the phase diagram the decrease in carrier density doesn’t affect the scattering rate.

4.4.2 In-plane magnetic field

When the magnetic field is applied in the plane of the 2DEG, the semi-classical theory of transport, presented at the beginning of this chapter, predicts no MR. This is due to the fact that no orbital motion is possible in the out-of-plane direction since the value of $k_z$ is fixed for each sub-band. Yet, quantum corrections linked to the interaction between the magnetic field and the spin of the electron are expected to restore the MR when the magnetic field is applied in-plane. This can be due, for example, to the Zeeman effect or to SO coupling.

Alternatively, when a small dispersion along the “z” direction exists, two in-plane orientations of the magnetic field have to be considered; in-plane parallel to the current flow and in-plane perpendicular to the current flow. In the first case, the Lorentz force is zero while in the second case it is finite and oriented along the out-of-plane direction. As a consequence, a larger resistance is expected when the magnetic field is perpendicular to the current flow.

In our interfaces, for conductances below $\approx 2$ mS, we have observed a weak MR when the magnetic field is applied in the plane of conduction [110]. In order to understand its origin we have investigated its dependence with respect to the direction
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Figure 4.16: Dependence of the elastic scattering time (left scale) and of the effective mass (right scale) on the sheet conductance $\sigma_{2D}^0$.

of the current.

Figure 4.17 shows the variation of the conductance at the LaAlO$_3$/SrTiO$_3$ interface applying an in-plane magnetic field $B$ and varying the angle $\phi$ between the current and $B$ (see fig. 4.4). Four values of the magnetic field are presented when $\sigma_{2D}^0$ is 1 mS (a) and $\sigma_{2D}^0$ is 2 mS (b). Maxima of the conductance are seen for both dopings when the external magnetic field is applied perpendicular to the current, pointing to a non-orbital origin of the oscillations. In addition to that, the maximum of the oscillation $\sigma_{2D}(B, \phi = \pi/2)$ evolves in a non monotonic way for the first doping which is not the case for the higher conductance state. Interestingly enough, these features are not observed in symmetric heterostructures made from Nb-doped SrTiO$_3$ thin films [115].

Oscillations of the MC in the parallel field geometry were also reported by Ben Shalom et al. [116]. For the range of sheet conductances and the field intensity that they considered (typically higher than ours) they found a positive MC for $\phi = 0$, and suggested that a magnetic order forms at the interface. For the doping range considered here, we may understand the behavior of the parallel MC as the field is rotated within the 2DEG plane if we consider the anisotropy brought by the $d_{xz}$ and $d_{yz}$ orbitals to the Fermi surface reconstructed by the SO interaction of the LaAlO$_3$/SrTiO$_3$ interface.

As a first approximation, we model the $xz$ and $yz$ sub-bands as 1D-like since they have very different in-plane masses ($m_+^* = 0.4 - 1.2m_e$ and $m_h^* = 6 - 14m_e$). We focus on the former type, which gives a higher contribution to transport since we set the current flow along $\hat{x}$. Interestingly, in the presence of the linear Rashba term identified in the previous section, the quasi 1D spin-split bands exhibit an energy gap at the $\Gamma$ point when $B$ is along $\hat{x}$ ($\phi = 0$) and a Zeeman-like offset when $B$ is along $\hat{y}$ ($\phi = \pi/2$) (see fig. 4.18). The impact of this effect on transport depends on the position of $E_F$ relative to $E_\Gamma$, the energy of the electronic states at the $\Gamma$ point when $B = 0$, since the
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Figure 4.17: (a) and (b) Experimental and theoretical plots of the conductance $\sigma_{2D}(B, \phi)$ versus $\phi$, the angle between the in-plane magnetic field $B$ and the current for various values of $B$. (a) corresponds to $\sigma_{2D}^0 = 1$ mS and (b) to $\sigma_{2D}^0 = 2$ mS. (c) and (d) are the experimental and theoretical dependences of the conductance on the in-plane magnetic field for $\phi=\pi/2$. 
density of states (DOS) at the Fermi energy, \( g(E_F) \), enters the expression of \( \sigma_{2D} \). The conductivity will thus show a dip for \( \phi = 0 \), provided \( E_F \sim E_\Gamma \). If \( E_F \) is not close to \( E_\Gamma \), \( g(E_F) \) is almost unchanged as compared to its \( B = 0 \) value.

Figure 4.18: Schematic view of the band structure according to our model. a) We show a \( xz \) sub-band split by a spin-orbit coupling of \( 5 \times 10^{-12} \) eV m. The effective mass used in the calculation is \( m_{\text{eff}}^{xz} = 0.64 m_e \). \( E_\Gamma \) defines the crossing point of the spin-split bands. b) Applying a magnetic field parallel to the current direction ([100] direction, \( \phi = 0 \)) opens a gap at the gamma point: when the Fermi level is at \( E_\Gamma \), the change in carrier density leads to a decrease of conductance. c) When the magnetic field is in plane and perpendicular to the current ([010] direction, \( \phi = \pi/2 \)), a Zeeman effect is observed in the band structure.

Beyond the qualitative 1D model, we have modeled the evolution of the conduction band in the applied \( B \). We use a tight binding model featuring kinetic and Rashba terms and we take into account the finite value of \( m_{\text{eff}}^{xz} \) and the anisotropy of \( \lambda \) in the \( xy \) plane for the \( xz, yz \) sub-bands. The mobility, \( \lambda \) and the gyromagnetic factor \( g \) all depend on \( V_g \), but we consider that they do not change appreciably with \( \phi \) nor with the magnitude of \( B \) in our experiments. For a given \( V_g \), the variation of \( \delta n \) – i.e. the change in the
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Figure 4.19: Experimental and model determined plots of $\Delta \sigma_{2D}$ versus $B$ for $\sigma_{2D}^0 = 1 \text{ mS}$ and $\sigma_{2D}^0 = 2 \text{ mS}$.

$x z/y z$ carrier concentrations – with $\phi$ and $B$ depends on the values of the spin-orbit and Zeeman energies. The conductance $\sigma_{2D}(B, \phi)$ is then proportional to $\delta n$. Experimental data and plots obtained from the model are shown in fig. 4.17 (c) and (d). We note that while $\sigma_{2D}(B, \phi)$ increases monotonically with $B$ at fixed $\phi$ for $\sigma_{2D}^0 = 2 \text{ mS}$, this is not the case for $\sigma_{2D}^0 = 1 \text{ mS}$, a feature which is correctly captured by our model. Figure 4.19 displays the evolution of $\Delta \sigma_{2D} = \sigma_{2D}(B, \phi = \pi/2) - \sigma_{2D}(B, \phi = 0)$ versus $B$ according to our model and the experimental results. We find good agreement using $\Delta_{SO} = 7(2.5) \text{ meV}$ for $\sigma_{2D}^0 = 2(1) \text{ mS}$ respectively. These values fall within the range of previous experimental estimates [27, 28].

Figure 4.20 shows the evolution of the experimental oscillation amplitude $\Delta \sigma_{2D}$ as a function of $\sigma_{2D}^0$ for different magnetic fields. As can be seen, $\Delta \sigma_{2D}$ tends to zero for a sheet conductance in the $0.1 - 0.3 \text{ mS}$ range. These sheet conductance values – which lie in the diffusive regime – are close to the QCP, suggesting a potentially important role played by the $d_{xz}, d_{yz}$ orbitals in establishing superconductivity.

Our findings underscore the evolution that takes place in the LaAlO$_3$/SrTiO$_3$ heterostructure, as one tunes the gate voltage in the range where superconductivity is observed at low temperature. For low $V_g$, conduction is diffusive and is dominated by the $d_{xy}$ orbitals, as the impact of disorder is expected to be more severe for the
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![Graph showing the evolution of the amplitude of the experimental oscillations as a function of $\sigma_{2D}^0$.]

**Figure 4.20:** Evolution of the amplitude of the experimental oscillations $\Delta \sigma_{2D}$ ($\Delta \sigma_{2D} = \sigma_{2D}(B, \phi = \frac{\pi}{2}) - \sigma_{2D}(B, \phi = 0)$) as a function of $\sigma_{2D}^0$. "Plus" symbols pertain to measurements performed on an additional sample at 7 T.

1D-like $d_{xz}, d_{yz}$ states. For larger values of $V_g$, the nature of transport changes and the out-of-plane $d_{xz}, d_{yz}$ orbitals start contributing to the zero field conductivity, as evidenced by the evolution of $m^*$, $B_{el}$, $B_{so}$ and by the oscillations of the in-plane magnetoconductance. For these orbitals, the effect of the strong spin-orbit interaction has to be taken into account at the band structure level for the calculation of their contribution to the transport.
5.1 Introduction

In this final chapter, we focus on the magnetotransport of samples grown at 650 °C. As mentioned in chapter 3, these interfaces display a much larger electronic mobility than samples grown at 800 °C. Hence, for them, the phenomenon of quantization of the electron orbits in a magnetic field is important.

We will first give a theoretical description of this effect and detail the consequences on some physical properties. Then, we will look at the magnetotransport and its doping dependence at these high-mobility LaAlO$_3$/SrTiO$_3$ interfaces. Thanks to the quality of the low temperature measurements, we could perform a detailed analysis aimed at understanding the band structure of the interface and at revealing the role of the SO interaction on the physics of the 2DEG.

5.2 The semi-classical approach

Contrarily to the theoretical development of section 4.2, we focus here on the trajectory of the semi-classical particles rather than on their distribution function. In the following two sections, we will not consider scattering of the particles as they move along their trajectories. We start with a 3-dimensional model (3D). In the presence of an external magnetic field $B$ (no electric field) the semiclassical equations are:

$$\begin{align*}
v_k &= \dot{\mathbf{r}} = \frac{1}{\hbar} \frac{\partial \epsilon_k}{\partial \mathbf{k}} \\
\dot{\mathbf{p}} &= \hbar \dot{\mathbf{k}} = q(\dot{\mathbf{r}} \times \mathbf{B})
\end{align*}$$

(5.1)
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The first equation is simply eq. (4.1) and the second one is Newton's second law describing the action of the Lorentz force on a charged particle of velocity $v_k$ ($\dot{p}$ is the time derivative of the linear momentum).

One can get two important pieces of information from these two equations. The first one, is that the projection of $k$ along the direction of the magnetic field is constant in time:

$$\dot{k}_\parallel = \hat{k} \cdot \frac{B}{B} = \frac{q}{\hbar} (\dot{r} \times B) \cdot \frac{B}{B} = 0 \quad (5.2)$$

The second, is that the energy of the particle is constant too. Indeed from the scalar product of $\dot{k}$ with $\dot{r}$:

$$0 = \dot{r} \cdot \dot{k} = \frac{1}{\hbar} \frac{\partial \epsilon_k}{\partial k} \cdot \dot{k} = \frac{1}{\hbar} \frac{d\epsilon_k}{dt} \quad (5.3)$$

As a consequence, the orbit of the particle in $k$-space is restricted to the intersection of a plane perpendicular to $B$ and a constant energy surface. As transport is mainly probing electrons at the Fermi level (see chapter 4), resistance will measure electrons performing orbits on the Fermi surface (FS). Figure 5.1a) shows an arbitrary FS and some of the possible particle trajectories in $k$-space for three different field orientations ($B$ along $\hat{x}$, $\hat{y}$ or $\hat{z}$). Taking the vector product of $\dot{k}$ with the direction of the magnetic field yields:

$$B \times \hbar \dot{k} = \frac{q}{B} B \times (\dot{r} \times B) = q B \dot{r}_\perp \quad (5.4)$$

where we have used the fact that the perpendicular (to $B$) component of $\dot{r}$ reads, $\dot{r}_\perp = \frac{B \times (\dot{r} \times B)}{B^2}$. Since $\dot{k}$ has no parallel (to $B$) component we get:

$$\dot{r}_\perp = \frac{\hbar}{qB} \left( B \times \frac{\dot{k}_\perp}{\hbar} \right) \quad (5.5)$$

This implies that the electron motion in real-space projected on the plane perpendicular to $B$ mirrors its trajectory on the FS. Concerning the real-space trajectory along the magnetic field axis contrarily to $\dot{k}_\parallel$, $\dot{r}_\parallel$ does not need to be zero or even constant, in this semi-classical treatment. $r_\parallel(t)$ can have a non trivial evolution given by:

$$r_\parallel(t) = r_\parallel(0) + \frac{1}{\hbar} \int_0^t \frac{\partial \epsilon_k}{\partial k_\parallel} \, dt' \quad (5.6)$$

5.3 The Bohr-Sommerfeld quantization rule

The Bohr-Sommerfeld quantization rule is a way to select, from the continuum of classically allowed states, those allowed by quantum mechanics (at first order in $\hbar$). It states that the line integral of the generalized momentum along any real space closed path is quantized:

$$\oint \pi \cdot dl = 2\pi \hbar (n + \gamma) \quad (5.7)$$

with $\gamma = \frac{1}{2}$ for free electrons, the generalized momentum being defined via the Lagrangian:

$$\pi = \frac{\partial L}{\partial \dot{r}} \quad (5.8)$$
5.3 The Bohr-Sommerfeld quantization rule

Figure 5.1: Semi-classical orbits on the Fermi surface and the Bohr-Sommerfeld quantization rule. a) Examples of semi-classically allowed orbits (red, blue) on the Fermi surface (green) for three different magnetic field orientations ($B$ along $\hat{x}$, $\hat{y}$ or $\hat{z}$). These orbits are always in a plane perpendicular to $B$. Orbits in blue are extremal trajectories while orbits in red are examples of non-extremal trajectories. b) c) and d) Examples of situations in which a minimum (b)) or a maximum (c) and (d)) of the conductance is expected. The brown cylinders physically represent the Onsager relation (eq. (5.9)), derived from the Bohr-Sommerfeld quantization rule.
5. QUANTUM OSCILLATIONS

Onsager [118], used this quantization rule to select, from the semi-classical orbits on the FS, those physically realized. The Onsager relation states that:

\[ A = (n + \gamma)\Delta A \quad \text{with} \quad \Delta A = \frac{2\pi|q|B}{\hbar} = \frac{2\pi}{l_B} \quad (5.9) \]

with \( A \) the surface enclosed by an orbit in \( k \)-space, \( n \) a non-negative integer called the Landau level index and \( \gamma \) a constant equal to \( \frac{1}{2} \) for free electrons. We derive it for the special case of a 3DEG in Appendix B. Note that this rule is, in general, only valid for \( n \gg 1 \). In the simple geometry of fig. 5.1 this condition is represented by concentric cylinders aligned in the direction of the field and with a cross sectional area given by eq. (5.9). Thus, an electron on the FS can only be running along an orbit located at the intersection of the “n-th” tube and the FS; in this case the electron is said to belong to the “n-th” Landau level (LL).

Alternatively one may be interested to know at which fields \( (B^*) \) a given orbit is authorized. From eq. (5.9) we get:

\[ \frac{1}{B^*} = (n + \gamma)\frac{2\pi|q|}{\hbar A} \quad (5.10) \]

Thus the states along an orbit of area \( A \) will be allowed at equidistant values of \( 1/B \).

The period of this repetition is simply:

\[ \frac{1}{B^*_{n+1}} - \frac{1}{B^*_n} = \frac{2\pi|q|}{\hbar A} \quad (5.11) \]

and hence the associated frequency is proportional to the area \( A \):

\[ F = \frac{\hbar}{2\pi|q|A} \quad (5.12) \]

The 2D case Before going on with the 3D case let us consider the case of a two dimensional FS as it is simpler to treat. Figure 5.2 shows the example of an isotropic 2D FS. Clearly, in this simple geometry, only one type of orbit on the FS is possible (blue line).

Setting the field, for example, perpendicular to the conducting plane, one sees that there are values of \( B \) such that no \( n \) satisfies the condition 5.10 (see for example the situation depicted in fig. 5.2 where the electron trajectory lies in-between two neighboring cylinders). At these fields, in this simple treatment, there is no orbit on the FS that is allowed by quantum mechanics. Since from eq. (5.11) one expects this situation to be repeated periodically as a function of \( 1/B \), many physical properties will oscillate as a function of \( 1/B \), at the frequency \( F \). These oscillations are called the quantum oscillations. This is the case, for example, of the electrical resistance/conductance and leads to the so-called Shubnikov-de Haas oscillations that were first observed in 1930 [119]. Another example are the oscillations in the magnetization that were first observed by de Haas and van Alphen [120].

From what precedes it is clear that the quantum oscillations are very useful for investigating the FS properties, as their frequency is directly related to the FS area.
5.3 The Bohr-Sommerfeld quantization rule

Figure 5.2: Onsager relation for a 2D Fermi surface. (Left) 2D Fermi surface (in green but hidden by the only possible orbit in blue). The brown cylinders materialize the Onsager relation (eq. (5.9)), derived from the Bohr-Sommerfeld quantization rule. (Right) In 2D, when the magnetic field orientation is tilted from the perpendicular orientation, only its out-of-plane component influences the trajectory of the electron.

(see eq. (5.12)). Note that this argument also holds for more complex systems than the one presented in fig. 5.2. Indeed, for example, for systems with several Fermi sheets, quantum oscillations with different frequencies will, in the first approximation, simply sum up. Hence, in the free electron approximation, the total number of carriers estimated via the quantum oscillations is:

\[ n_{\text{SDH}}^{2\text{D}} = \frac{|q|}{2\pi\hbar} \sum_{i} F_{i} \nu_{s}^{i} \nu_{v}^{i} \]  \hspace{1cm} (5.13)

where the sum is on the Fermi surface sheets with different areas in \( k \)-space and \( \nu_{s}^{i}/\nu_{v}^{i} \) are the spin/valley degeneracies of the \( i \)th Fermi surface sheet. To establish this formula we have used the fact that the non-spin-degenerate density of states in \( k \)-space is equal to \( 1/(2\pi)^2 \).

A hallmark of a 2D system is the dependence of the quantum oscillations on the magnetic field orientation. Indeed, since in 2D the velocity of the particle along the out-of-plane direction is zero, only the component of \( B \) along the out-of-plane direction has an effect on the electron trajectory. Hence, in 2D eq. 5.10 becomes:

\[ \frac{1}{B^* \cos \theta} = (n + \gamma) \frac{2\pi|q|}{\hbar A} \]  \hspace{1cm} (5.14)

This means that only the projection of the magnetic field along the normal to the conducting plane determines the position of the quantum oscillations. In particular, if \( B \) is in the conducting plane, no quantum oscillations are expected, since there is no finite \( B^* \) that can fulfill eq. (5.14).

The 3D case  Contrarily to the 2D case, in 3D, when \( n \gg 1 \), for every magnitude and orientation of \( B^* \), there are many LLs at the Fermi surface (see fig. 5.1b c) and d)). Thus one naively expects no quantum oscillations in any field orientation. One can also reach this conclusion, considering all the possible semi-classical orbits in a
plane perpendicular to the magnetic field in fig. 5.1. Their number is quasi infinite for a macroscopic sample. Since, via eq. (5.12), most of them are associated to a different frequency, they will interfere destructively, hence the absence of quantum oscillations.

A more rigorous derivation of this result can be found for example in [121], yet, with a subtle difference. Indeed, the above argument is true only for non-extremal orbits. An extremal (or stationary) orbit is such that:

\[
\frac{\partial A}{\partial k_\parallel} = 0
\]  

(5.15)

In fig. 5.1 the extremal orbits are colored in blue. Clearly there are many fewer of these. They sum to two when \( B \) is along the \( \hat{z} \) direction, and only to one when \( B \) is in the x-y plane. In this sense, the 3D case is similar to the 2D case where only a limited number of orbits on the FS are allowed. Thus quantum oscillations are also expected in the 3D case, yet with a number of frequencies depending not only on the number of Fermi sheets but also on the corrugation of the FS along the magnetic field direction.

Concerning the above analysis, an important point has to be stressed. The quantization of the electron orbits is achieved only if there is no scattering event before the electron completes an orbit. For an electron gas this condition can be expressed as (see Appendix B):

\[
|\omega_c| \tau \geq 1
\]  

(5.16)

with \( \omega_c = eB/m^* \).

In term of the electron mobility (for an electron gas \( \mu = e\tau/m^* \)):

\[
|\mu|B \geq 1
\]  

(5.17)

As a consequence, the mobility of the electron gas defines at which magnetic field the quantum oscillations are observable. For the samples of the last chapter, only above \( \sigma_{2D}^0 = 10 \text{ mS} \) the estimated mobility reaches 2000–3000 cm\(^2\) V\(^{-1}\) s\(^{-1}\). This implies that, for our “standard” samples, in the most doped states, quantum oscillations should start above 3–5 T. Returning to figure 4.11 of chapter 4, we indeed observe weak Shubnikov-de Haas oscillations at high fields for the most conducting states (see also [108]). Yet, in what follows, we will focus on the quantum oscillations of samples grown at “low” temperature as they have higher mobilities (see chapter 3) and present bigger oscillations.

### 5.4 Quantum-mechanical approach

In what follows we study the problem of Bloch electrons in an external magnetic field, using a quantum-mechanical treatment and the effective mass approximation. Not only does this strategy allow us to get information on the real nature of the states in our system under a quantizing field but it also permits us to include the effect of Zeeman and Rashba interactions in rather an easy way. We restrict our treatment to the 2D case as we are interested to apply our results to the electron gas present at the LaAlO\(_3\)/SrTiO\(_3\) interface.
5.4 Quantum-mechanical approach

5.4.1 2DEG without Zeeman/Rashba

5.4.1.1 Eigenvalues and eigenstates

In a perpendicular magnetic field \( B = B \hat{z} \), the Hamiltonian of an electron of charge \( e \) \((e < 0)\) and effective mass \( m^* \) confined in the \((x,y)\) plane is:

\[
\hat{H}_L = \frac{\hat{\pi}^2}{2m^*}, \quad \hat{\pi} = \hat{p} - e\hat{A}(r) = \hat{p} - m^*\omega_c \hat{A}(r)/B \tag{5.18}
\]

this Hamiltonian can be derived from the 2D-quantum version of the Lagrangian presented in Appendix B. \( \hat{\pi} \) is the kinetic momentum operator.

The Landau gauge is either \( \hat{A} = -B\hat{y}\hat{x} \) or \( \hat{A} = B\hat{x}\hat{y} \). We take the second choice, which means that:

\[
\hat{\pi}_x = \hat{p}_x, \quad \hat{\pi}_y = \hat{p}_y - m^*\omega_c \hat{x}, \quad \hat{H}_L = \frac{\hat{p}_x^2}{2m^*} + \frac{(\hat{p}_y - m^*\omega_c \hat{x})^2}{2m^*} \tag{5.19}
\]

making the canonical change of variables:

\[
\hat{X} = \hat{x} - \frac{\hat{p}_y}{m^*\omega_c}, \quad \left[ \hat{X}, \hat{p}_x \right] = \left[ \hat{x}, \hat{p}_x \right] = i\hbar \tag{5.20}
\]

The Landau Hamiltonian becomes a one-dimensional harmonic oscillator:

\[
\hat{H}_L = \frac{\hat{p}_X^2}{2m^*} + \frac{1}{2} m^* \omega_c^2 \hat{X}^2 \tag{5.21}
\]

Recasting it in terms of the creation and annihilation operators:

\[
\hat{a}^\dagger_L = \sqrt{\frac{m^*|\omega_c|}{2\hbar}} \hat{X} - \frac{i}{\sqrt{2m^*\hbar|\omega_c|}} \hat{p}_x \\
\hat{a}_L = \sqrt{\frac{m^*|\omega_c|}{2\hbar}} \hat{X} + \frac{i}{\sqrt{2m^*\hbar|\omega_c|}} \hat{p}_x \tag{5.22}
\]

the Landau Hamiltonian becomes simply

\[
\hat{H}_L = E_c \left( \hat{a}^\dagger_L \hat{a}_L + \frac{1}{2} \right) \tag{5.23}
\]

with the well known eigenvalues:

\[
E_n = E_c \left( n + \frac{1}{2} \right), \quad n = 0, 1, 2, \ldots \tag{5.24}
\]

with \( E_c = \hbar|\omega_c| \). Thus, in the presence of an out-of-plane magnetic field, the density of states of a 2DEG moves from a constant value \( (g(E) = \frac{m^*}{\pi\hbar^2}) \) to a set of peaks (see fig. 5.3). The energy levels \( E_n \) are the Landau levels (LLs) and their energy corresponds to the kinetic energy a semi-classical electron would have if it was restricted to move around the tubes of fig. 5.2.
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Figure 5.3: 2D Density of states in the absence/presence of an external magnetic field. (Left) In the absence of an external out-of-plane magnetic field the 2D density of states is constant and is equal to $m^* / \pi \hbar^2$. (Right) With an external magnetic field applied perpendicular to the 2DEG, one observes the creation of LLs separated by a field dependent cyclotron gap $\hbar |\omega_c|$. $\mu$ is the chemical potential that separates the filled states (in blue) from the empty states (in white). $n$ is the LL index.

Since the spacing between two levels depends linearly on the magnetic field strength, increasing the amplitude of $B$ moves the LLs across the chemical potential ($\mu$). The quantum oscillations observed experimentally originally by Shubnikov, de Haas and van Alphen originate from this modulation of the DOS at the Fermi level.

Note that, while the energy spectrum in eq. (5.24) is not gauge-dependent, the creation and annihilation operators are gauge-dependent and the eigen wave functions also. Since in the Landau gauge the Landau Hamiltonian $\hat{H}_L$ depends on $\hat{\mathbf{p}}_x$, $\hat{\mathbf{p}}_y$, and $\hat{\mathbf{k}}_y$, but not on $y$, it commutes with $\hat{\mathbf{p}}_y$; the wave number $k_y$ is therefore a good quantum number. We can choose eigenvectors of $\hat{H}_L$ which are also eigenvectors of $\hat{\mathbf{p}}_y$; these are:

$$\psi_{n,k_y}(x, y) = \langle x, y | k_y, n \rangle = \frac{1}{\sqrt{2\pi}} e^{ik_yy} \phi_n \left( x - \frac{\hbar k_y}{m^* \omega_c} \right)$$

$$\phi_n(X) = \frac{1}{\sqrt{2^n n!}} \left( \frac{m^* |\omega_c|}{\pi \hbar} \right)^{\frac{1}{4}} H_n \left( \sqrt{\frac{m^* |\omega_c|}{\hbar}} X \right) e^{-\frac{m^* |\omega_c|}{2\hbar} X^2} \quad (5.25)$$

where $H_n$ is the Hermite polynomial of degree $n$. These wave functions are the product of a plane wave along the $\hat{y}$ direction and an eigenstate of the harmonic oscillator along the $\hat{x}$ direction. Yet, the energy spectrum is independent of $k_y$ (eq. (5.24)), meaning that each LL contains a certain number of degenerate states (labeled by $k_y$).

The center of the harmonic oscillator wave function depends on $k_y$. This is a useful consideration in order to determine the degeneracy of each LL. Indeed, a reasonable assumption is that the center of the harmonic oscillator has to lie inside the sample. Defining the sample size as $L$ along the $\hat{x}$ direction and Born-von Karman boundary
5.4 Quantum-mechanical approach

condition along the \( \hat{y} \) direction one obtains that:

\[
- \frac{L}{2} \leq \frac{\hbar k_y}{m^*\omega_c} \leq \frac{L}{2}
\]  

(5.26)

with \( k_y = \eta \frac{2\pi}{L} \) and \( \eta \) an integer. The number of \( k_y \) values per unit surface allowed by these boundary conditions (i.e. the degeneracy of a LL) is thus:

\[
\frac{2\eta_{\text{max}}}{L^2} = \frac{m^*|\omega_c|}{2\pi\hbar}
\]  

(5.27)

Since this argument holds for each LL, their degeneracy does not depend on the index \( n \).

One could be surprised to observe that the wave functions of eq. (5.25) break in-plane rotational symmetry; a symmetry which is preserved in the initial problem. This is due to the choice of the gauge, which has an in-plane orientation. Another possible choice that preserves rotational symmetry is the symmetric gauge, \( \vec{A}_{\text{sym}} = -\frac{B}{2} \hat{y} \hat{x} + \frac{B}{2} \hat{x} \hat{y} \).

As no measurable property should depend on the gauge, the Landau level spectrum should be the same using \( \vec{A} \) or \( \vec{A}_{\text{sym}} \) as vector potential. The eigen wave functions are yet different. Using the symmetric gauge they read:

\[
\psi_{n,l}^{\text{sym}}(x, y) = \langle x, y|n,l \rangle_{\text{sym}} = \frac{1}{\sqrt{n!}} \left[ -i \sqrt{\frac{\hbar}{2m^*|\omega_c|}} \left( 2 \frac{\partial}{\partial z} - \frac{m^*|\omega_c|}{2\hbar} \hat{z} \right) \right]^n N_l e^{-\frac{m^*|\omega_c|\|ix\|^2}{4\hbar}}
\]  

(5.28)

with \( n \) the Landau level index and \( l \) a quantum number that replaces \( k_y \). \( l \) are the eigenvalues of an angular momentum based on the canonical momentum rather than on the kinetic momentum. Hence it is not a quantity that is easy to physically interpret. In fact, the average value of the “kinetic” angular momentum in the state \( |n,l\rangle_{\text{sym}} \) is equal to \(-2(n + 1/2)\) which does not depend on \( l \).

One example of \( |\psi_{n,k_y}\rangle^2 \) and of \( |\psi_{n,l}\rangle^2 \) is shown in figure 5.4. One could be tempted to think that the \( |\psi_{n,l}\rangle^2 \) are better representing the semi-classical orbits we studied in the last section. Yet, the sets \( \psi_{n,k_y} \) and \( \psi_{n,l}^{\text{sym}} \) are just two different bases of the degenerate vectorial space of the \( n \)th LL.

5.4.1.2 Density of states and conductance

Using eq. (5.24) and (5.27) the density of states of a 2DEG in a perpendicular magnetic field can be calculated. However, scattering processes modify the energetic line shape of the LLs introducing a broadening of the levels as shown in fig. 5.3. If we choose a Gaussian line shape with variance \( \Gamma \), the density of states is:

\[
D(E) = \sum_{n,s} m^*|\omega_c| \frac{1}{2\pi\hbar} \sqrt{2\pi\Gamma} \exp \left[ -\frac{1}{2} \left( \frac{E - E_n}{\Gamma} \right)^2 \right]
\]  

(5.29)
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Figure 5.4: Dependence of the eigenstates of eq. (5.18) on the gauge choice. Two examples of the eigenwave functions obtained using either the Landau (left) or the symmetric (right) gauge.

with $s$ the spin-orientation.

Other line shapes can be used to describe the LL broadening. For example, Lorentzian and semi-elliptic broadenings can be considered. A constant background is sometimes added to the total DOS. In many cases, according to ref. [122], a Gaussian line shape with a variance proportional to $\sqrt{B}$, or a Lorentzian broadening are considered.

Using the first Born approximation to the self-energy and the above DOS a rather simple relation between the conductance and the density of states can be found [123, 124]:

$$\sigma = \frac{e^2}{2\pi\hbar} \int_{-\infty}^{\infty} dE \left( -\frac{\partial f_0(E - \mu)}{\partial E} \right) \times \sum_{n,s} \left( n + \frac{1}{2} \right) \exp \left[ -\left( \frac{E - E_n}{\Gamma} \right)^2 \right]$$

(5.30)

As in eq. (4.15), due to the presence of the derivative of the Fermi distribution, if $k_B T \ll \epsilon_F$, the conductance of the system is determined by the states at the Fermi level.

Contrarily to what we have been implicitly assuming in our semi-classical treatment, the chemical potential $\mu$ (or the Fermi energy) depends on the magnetic field strength. It is the carrier concentration that does not evolve with $B$. In order to calculate $\mu(B)$, we solve the equation numerically giving the carrier concentration at a given temperature:

$$n_{2D} = \int_{-\infty}^{\infty} f_0(E - \mu) D(E) \, dE$$

(5.31)

Figure 5.5 shows the evolution of $E_F$ in presence and in absence of LL broadening calculated at zero temperature. Clear oscillations of $E_F$ are visible. Yet, at low $B$ ($n \gg 1$), $E_F$ is almost constant, validating a posteriori the assumptions made in the
5.4 Quantum-mechanical approach

Figure 5.5: Evolution of the Fermi energy with magnetic field. (blue lines) Evolution of the Landau levels given by eq. (5.24) with magnetic field. Related magnetic field dependence of the Fermi energy with \( B \), in the absence (red line) and in the presence (black dashed curve) of disorder. The green line is the position of the Fermi level in the absence of magnetic field.

preceding section. The LL positions (eq. (5.24)) as a function of \( E \) and \( B \) are plotted in the background. Such a plot is named a fan diagram.

Strikingly, in the absence of disorder the Fermi level is always aligned with a LL. Referring to eq. (5.30) this implies a constant conductance of the system and thus no quantum oscillations. In the presence of disorder, the Fermi level (black line) is in general not aligned with a LL and quantum oscillations are expected. Thus, disorder is an essential ingredient to observe Shubnikov-de Haas oscillations.

5.4.1.3 The Lifshitz-Kosevich formula

We have seen that under the approximations of the last paragraphs the DOS of a 2DEG in a magnetic field becomes a periodic function of the energy. Hence, it can be expressed in the form of a Fourier series. As we shall see, this is a particularly useful operation when the magnetic field is not too strong and/or the disorder is high enough for the neighboring LLs to overlap greatly. Figure 5.6 shows such a situation. In this case, the DOS can be efficiently described using the fundamental term of the Fourier series:

\[
D(E) \approx C_1 + C_2 \cos \left( \frac{2\pi E}{\hbar|\omega_c|} - \pi \right) \tag{5.32}
\]

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\[ T = \hbar |\omega_c| \]
\[ \varphi = \pi \]
\[ C_1 \]
\[ C_2 \]

Figure 5.6: Modeling of the 2D density of states in a perpendicular magnetic field in the LK description of the quantum oscillations. Density of states (blue) in the high LL limit where individual peaks strongly overlap. Magnetic field is set to an arbitrary value. (green dashed line) Approximation of the density of states using only the fundamental term of a Fourier series with period in energy \( T \) and phase \( \varphi \). \( C_1 \) and \( C_2 \) are defined in eq. (5.32).

This observation is at the basis of the Lifshitz-Kosevich (LK) formula [125, 126] that describes the quantum oscillations at low magnetic field:

\[ \Delta \sigma_{xx}(B) = \sigma_{xx}(B) - \sigma_{xx,0}(B) = 4\sigma_{xx,0}(B)e^{-\alpha_{LK}T_D} \frac{\alpha_{LK}T}{\sinh(\alpha_{LK}T)} \cos\left(\frac{2\pi F}{B} - \pi\right) \]  

(5.33)

with \( \alpha_{LK} = \frac{2\pi^2 k_B}{\hbar \omega_c} \), \( k_B \) the Boltzmann constant, \( T_D \) the Dingle temperature and \( \sigma_{xx,0} \) the non-oscillating part of the magnetoconductance. The first factor is the Dingle term and is related to the disorder in the system (i.e. to the LL broadening): it induces a damping of the oscillations that decreases as the magnetic field is increased. Its precise form depends on the line shape of the LLs. Here it corresponds to Lorentzian LLs but in the Gaussian case it is replaced by \( \exp\left(-\frac{\pi^2 T^2}{\hbar^2 \omega_c^2}\right) \). Hence, a plot of the amplitude of the quantum oscillations as a function of \( 1/B \) can provide information on the nature of the disorder in the system. Such a plot is called a Dingle plot.

The second term is linked to the damping of the oscillations with temperature. Its origin can be traced to the integral over the energy of eq. (5.30), where the presence of the derivative of the Fermi distribution mixes the contribution of different LLs if the temperature is high enough (i.e. if \( k_BT \approx \hbar \omega_c \)). This temperature dependence of the quantum oscillations is determined by the effective mass \( m^* \), and thus allows \( m^* \) to be estimated. A plot of the amplitude of the oscillations at a given magnetic field as a function of the temperature is called an effective mass plot.

The last term is the oscillating part of \( \Delta \sigma_{xx}(B) \). As expected, the oscillations are periodic on a \( 1/B \) scale. Since, the range of validity of the LK formula is similar to the one of the semi-classical treatment, the Onsager relation (eq. (5.12)) is in general used to link \( F \) with the Fermi surface area. The phase factor, here set to \( -\pi \), carries important information. It can be determined by, for example, the dimensionality of
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the Fermi surface, the presence of inter-band scattering, the presence of spin(-orbit) interaction(s) and the character of the carriers (electrons, holes, dirac fermions..). For a 2DEG, for \( n \gg 1 \), one can combine eq. (5.10) and eq. (5.12) to get an equation for the maxima of the quantum oscillations:

\[
\frac{1}{B^*} = (n + \frac{1}{2}) \frac{1}{F}
\]

meaning that:

\[
2\pi \frac{F}{B^*} - \pi = 2\pi n
\]

The phase factor is indeed \(-\pi\) in this case. In the 3D case, it is modified by \( \pm \pi/4 \), the \( \pm \) sign corresponding to the minimum/maximum cross sections of the FS [127, 128].

In the presence of a more complex DOS, the description of the quantum oscillations is obviously not achieved keeping only the fundamental term of the Fourier series (as in eq. (5.32)). In this case, more terms of the type of eq. (5.33) are needed. We distinguish here three cases:

The first one is when the spacing between the LLs is large enough in order that the sum of their line shapes cannot be approximated by a single \( \cos \) function. In this case, higher orders of the Fourier series have to be taken into account. This effect leads to the appearance of harmonics of the fundamental frequency \( (F_p = pF, p = 2, 3, \ldots) [129] \). Since the \( \alpha_{LK} \) for these harmonics increases with \( p \), the latter are only observed in low disorder samples and at low temperature.

In the second case, the complexity of the DOS arises because of the presence of different types of carriers, each one giving rise to an independent set of LLs. Assuming that the conductances are simply summing up and that each individual set of LLs is described by an expansion of the type given in eq. (5.32), each type of carrier will give rise to its own frequency \( F_i \). The \( F_i \)s as well as the particular thermal/disorder driven dampings are, in this scenario, unrelated.

Finally, as we are going to see just following, the DOS of particles under magnetic field can be, in certain circumstances, an aperiodic function of \( E \). In this case, a Fourier expansion of the DOS is not strictly possible and hence the Lifshitz-Kosevich formula should not be used.

5.4.2 2DEG with Zeeman/Rashba

The goal of this section is to calculate the LL spectrum in the presence of Zeeman interaction and then also in the presence of linear Rashba spin-orbit coupling.

5.4.2.1 Including Zeeman

The Zeeman term only acts in the spin sector. Therefore combining the Landau and Zeeman energies we have (in the presence of a perpendicular magnetic field and in the Landau gauge):

\[
\hat{H}_{LZ} = E_c \left( a^\dagger_L a_L + \frac{1}{2} \right) \mathbb{1} + E_Z \sigma_z
\]
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with \( E_Z = \frac{g^* \mu_B B}{2} \), \( g^* \) an effective \( g \)-factor and \( \mu_B = \frac{|e| h}{2m_e} \) is the Bohr magneton. Thus :

\[
E_{ns} = E_c \left( n + \frac{1}{2} \right) + sE_Z, \quad s = \pm 1
\]  

(5.37)

with eigenvectors :

\[
|k_y n s\rangle \quad \text{with} \quad \hat{H}_{LZ}|k_y n s\rangle = E_{ns}|k_y n s\rangle
\]  

(5.38)

\( s \) being the spin orientation.

Eq. (5.37) can be rewritten as :

\[
E_{ns} = \hbar |\omega_c| \left[ n + \frac{1}{2} \left( 1 + s g^* m^* \right) \right]
\]  

(5.39)

Since the splitting between two successive spin-up or spin-down levels is the same as in the spinless case, eq. (5.39) gives rise, in the high LL limit, to two sets of quantum oscillations with the same frequency \( F \). Yet the phase of these two sets is not identical. Indeed, a similar calculation to the one above (eq. (5.34) and eq. (5.35)) gives :

\[
2\pi F_B - \pi \left( 1 + \frac{g^* m^*}{2m_e} \right) = 2\pi n
\]  

(5.40)

Assuming the same damping terms for each spin-orientation we get, for the conductance :

\[
\Delta \sigma_{xx}(B) \propto e^{-\alpha_{LK} T D} \frac{\alpha_{LK} T}{\sinh(\alpha_{LK} T)} \left[ \cos \left( \frac{2\pi F}{B} - \pi \left( 1 + \frac{g^* m^*}{2m_e} \right) \right) + \cos \left( \frac{2\pi F}{B} - \pi \left( 1 - \frac{g^* m^*}{2m_e} \right) \right) \right]
\]  

(5.41)

Hence:

\[
\Delta \sigma_{xx}(B) \propto e^{-\alpha_{LK} T D} \frac{\alpha_{LK} T}{\sinh(\alpha_{LK} T)} \cos \left( \frac{\pi g^* m^*}{2m_e} \right) \cos \left( \frac{2\pi F}{B} - \pi \right)
\]  

(5.42)

In this scenario, the phase of the oscillations is either \(-\pi\) or 0. An interesting situation arises when \( g^* m^* \) is an odd integer. In this case, the quantum oscillations vanish due to the destructive interference of the oscillations caused by the two spin-split species. This effect is called the spin-zero effect (see for example [130]), and is the result of the competition between the orbital (Landau) and the Zeeman splitting. In the spin-zero configuration, the Zeeman term shifts the Landau levels by exactly one quarter of the cyclotron gap (respectively up and down in energy for the spin-up and spin-down electrons). At high \( n \), it has the effect to restore the DOS which decreases in-between the Landau levels in the absence of Zeeman interaction. This results in no quantum oscillations. This effect has been used to determine the value of the \( g^* \) factor in several compounds by performing experiments in titled magnetic field configuration.

5.4.2.2 Including Rashba

In addition to the Zeeman term, we now consider the modifications in the LL spectrum induced by the Rashba interaction. Using the linear Rashba Hamiltonian (eq. (2.25))
and the definition of the kinetic momentum in the presence of an out-of-plane magnetic field (eq. (5.19)) we get (with \( \alpha = \lambda E_z \)):

\[
\hat{H}_R = \frac{\alpha}{\hbar} (\sigma_x \hat{p}_y - \sigma_y \hat{p}_x) \\
= \frac{\alpha}{\hbar} [\sigma_x (\hat{p}_y - m^* \omega_c \hat{x}) - \sigma_y \hat{p}_x] \\
= -\frac{\alpha}{\hbar} (\sigma_y \hat{p}_x + m^* \omega_c \sigma_x \hat{X}) \\
= -\frac{\alpha}{\hbar} \begin{pmatrix}
0 & m^* \omega_c \hat{X} - i \hat{p}_x \\
0 & 0
\end{pmatrix}
\]  

(5.43)

For electrons \( \omega_c < 0 \), hence :

\[
m^* \omega_c \hat{X} - i \hat{p}_x = -\sqrt{2m^* \hbar |\omega_c|} \left( \sqrt{\frac{m^* |\omega_c|}{2\hbar}} \hat{X} + \frac{i}{\sqrt{2m^* \hbar |\omega_c|}} \hat{p}_x \right) = -\sqrt{2m^* \hbar |\omega_c|} \hat{a}_L
\]

\[
m^* \omega_c \hat{X} + i \hat{p}_x = -\sqrt{2m^* \hbar |\omega_c|} \hat{a}_L
\]

(5.44)

Therefore :

\[
\hat{H}_R = \frac{\alpha}{\hbar} \sqrt{2m^* \hbar |\omega_c|} \begin{pmatrix}
0 & \hat{a}_L \\
\hat{a}_L^\dagger & 0
\end{pmatrix}
\]  

(5.45)

We find it more convenient to write \( \hat{H}_R \) in term of the matrices

\[
\sigma_\pm = \frac{1}{2} (\sigma_x \pm i \sigma_y), \quad \sigma_+ = \begin{pmatrix}
0 & 1 \\
0 & 0
\end{pmatrix}, \quad \sigma_- = \begin{pmatrix}
0 & 0 \\
1 & 0
\end{pmatrix}
\]

(5.46)

which have the following action on the eigenstates of \( \hat{H}_{LZ} \):

\[
\sigma_+ |k_y n, s \rangle = \delta_{s,-1} |k_y n, +1 \rangle, \quad \sigma_- |k_y n, s \rangle = \delta_{s,+1} |k_y n, -1 \rangle
\]

(5.47)

The Rashba Hamiltonian becomes

\[
\hat{H}_R = \alpha \sqrt{\frac{2m^* |\omega_c|}{\hbar}} \left( \hat{a}_L \sigma_+ + \hat{a}_L^\dagger \sigma_- \right) \equiv E_\alpha (\hat{a}_L \sigma_+ + \hat{a}_L^\dagger \sigma_-)
\]

(5.48)

with \( E_\alpha = \alpha \sqrt{\frac{2|\omega_c|}{\hbar}} \). In summary, we have to diagonalize the Hamiltonian :

\[
\hat{H}_{LZ} = E_c \left( \hat{a}_L^\dagger \hat{a}_L + \frac{1}{2} \right) \mathbb{I} + E_Z \sigma_z + E_\alpha (\hat{a}_L \sigma_+ + \hat{a}_L^\dagger \sigma_-)
\]

(5.49)

Calculating the matrix elements yields (\( E_{ns} \) is the LL spectrum in the presence of the orbital and Zeeman terms only):

\[
\langle k_y n s \mid \hat{H}_{LZ} \mid k'_y n' s' \rangle = \delta_{k_y k'_y} \left( E_{ns} \delta_{nn'} \delta_{ss'} + E_\alpha \sqrt{n + 1} \delta_{n+1,n'} \delta_{s,-1} \delta_{s',+1} + E_\alpha \sqrt{n} \delta_{n-1,n'} \delta_{s,-1} \delta_{s',-1} \right)
\]

(5.50)
5. QUANTUM OSCILLATIONS

Ordering the states as follows (we omit temporarily \( k_y \))

\[
|0, -1\rangle |0, +1\rangle |1, -1\rangle |1, +1\rangle |2, -1\rangle \cdots
\]

we see that the \( \hat{H}_{LZR} \) has a block-diagonal form:

\[
\begin{array}{cccccc}
|0, -1\rangle & |0, +1\rangle & |1, -1\rangle & |1, +1\rangle & |2, -1\rangle & \cdots \\
\langle 0, -1 | & E_{0,-1} & 0 & 0 & 0 & 0 & \ldots \\
\langle 0, +1 | & 0 & E_{0,+1} & E_\alpha & 0 & 0 & \ldots \\
\langle 1, -1 | & 0 & E_\alpha & E_{1,-1} & 0 & 0 & \ldots \\
\langle 1, +1 | & 0 & 0 & 0 & E_{1,+1} & \sqrt{2}E_\alpha & \ldots \\
\langle 2, -1 | & 0 & 0 & 0 & \sqrt{2}E_\alpha & E_{2,-1} & \ldots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\
\end{array}
\]

Each \( 2 \times 2 \) block has the following structure

\[
\hat{H}_{LZR}^N = \begin{pmatrix} E_{N-1,+1} & \sqrt{N}E_\alpha \\ \sqrt{N}E_\alpha & E_{N,-1} \end{pmatrix}, \quad N = 1, 2, 3, \ldots
\]

that can be easily diagonalized to give:

\[
E_{\pm}^N = NE_c \mp \sqrt{\left( \frac{E_c}{2} - E_Z \right)^2 + NE_\alpha^2}
\]

The corresponding normalized eigenvectors can be taken as (we use the notation \( |N; \pm\rangle \) for the “new” eigenvectors):

\[
|N; - \rangle = \cos \frac{\theta}{2} |N - 1, +1\rangle - \sin \frac{\theta}{2} |N, -1\rangle \\
|N; + \rangle = \sin \frac{\theta}{2} |N - 1, +1\rangle + \cos \frac{\theta}{2} |N, -1\rangle
\]

\[
\tan \theta = \frac{\sqrt{N}E_\alpha}{E_c/2 - E_Z}, \quad 0 \leq \theta < \pi \quad \Rightarrow \quad \theta = \tan^{-1} \left( \frac{\sqrt{N}E_\alpha}{E_c/2 - E_Z} \right) + \begin{cases} 0 & E_Z < E_c/2 \\ \pi & E_Z > E_c/2 \end{cases}
\]

The new LLs are mixtures of the \((N - 1)th\) spin-up and the \(Nth\) spin-down levels. Only the \(N = 0\) level (top left part of the above Hamiltonian) stays spin polarized. It reads:

\[
|0\rangle = |0, -1\rangle \quad E_0 = \frac{E_c}{2} - E_Z
\]

We see that, in the presence of Rashba interaction, the LLs are not equally spaced in energy. Thus we expect the Lifshitz-Kosevich formula to be unusable and must rely on numerical computations of the DOS and \( \sigma_{xx} \).
5.4 Quantum-mechanical approach

5.4.2.3 Numerical computations

The first step in a numerical simulation is to identify the minimum number of free variables in the computation. Interestingly, introducing the energy scale $E_{c,1} = E_c(B = 1 \text{ T})$ to normalize the energies, $\tilde{E} = E/E_{c,1}$, eq. (5.53) and eq. (5.55) can be simplified:

$$\tilde{E}_0 = \frac{E_c}{2E_{c,1}} - \frac{E_Z}{E_{c,1}} = \frac{B}{2} \left( 1 - \frac{g^* m^*}{2m_e} \right) = \frac{Ba}{2}$$

(5.56)

with $a = 1 - g^* m^*/(2m_e)$. Similarly,

$$\tilde{E}_N^\pm = N\frac{E_c}{E_{c,1}} \pm \sqrt{\left( \frac{E_c}{2E_{c,1}} - \frac{E_Z}{E_{c,1}} \right)^2 + N \frac{E^2_{c,1}}{E_{c,1}}},$$

$$\tilde{E}_0 = \frac{B}{2} \left( 1 - \frac{g^* m^*}{2m_e} \right) $$

(5.57)

with $d = 8(\alpha m^*)^2/(|e| h^3)$. We observe that, in these units, the LL spectrum is completely determined by the use of three parameters: those are $B$, $a$ and $d$.

In these units, eq. (5.29), eq. (5.30) and eq. (5.31) become:

$$\tilde{\sigma}_{xx} = \frac{e^2}{2\pi\hbar} \int_{-\infty}^\infty d\tilde{E} \left( -\frac{\partial \tilde{f}_0(\tilde{E} - \tilde{\mu})}{\partial \tilde{E}} \right)$$

$$\times \sum_{N,\pm} \left( n + \frac{1}{2} \right) \exp \left[ -\left( \frac{\tilde{E} - \tilde{E}_N^\pm}{\Gamma_\pm} \right)^2 \right].$$

(5.59)

$$n_{2D} = \int_{-\infty}^\infty \tilde{f}_0(\tilde{E} - \tilde{\mu}) \tilde{D}(\tilde{E}) d\tilde{E}$$

(5.60)

with $\tilde{f}_0(\tilde{E} - \tilde{\mu}) = \left( e^{(\tilde{E} - \tilde{\mu})/k_B T} + 1 \right)^{-1}$. The $\tilde{E}_0$ level is implicitly included in the above LL summation and we have included a “±”-dependent broadening of the levels for more generality.

At low enough temperature ($k_B T \ll \mu$) the Fermi Dirac distribution can be approximated by a Heaviside function. Numerically, since we use Gaussian LLs, this implies that the integral determining the Fermi level is just a sum of error functions of $E_{F} - E_N^\pm/\Gamma_\pm$ and that the integral in the expression of the conductance selects only the states at $E_F$. Apart from the gain in computation time, this implies that, at low temperature, $\tilde{\sigma}_{xx}(B)$ is completely determined by $n_{2D}$, $\Gamma_\pm$, $a$ and $d$. Hence, from a fit of the low
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temperature oscillations in the conductance, only the value of $m^* g^*$, $m^* \alpha$, $m^* \Gamma_\pm$ and $m^* E_F$ can be obtained. The value of the effective mass is accessible via $m^* = \frac{\hbar |e| k_B T}{k_B T}$, the temperature evolution of the oscillations is again a crucial piece of information.

To calculate $\sigma_{xx}(B)$ at finite temperature becomes a much more complex problem mainly because of the integral equation determining the chemical potential. One solution is to rely on the expansion of the equilibrium Fermi distribution proposed by Ozaki [131]. Ozaki’s approximation of $f_0$ converges exponentially faster than the more standard Matsubara’s expansion. This special choice becomes particularly important at low magnetic field when the spacing between LLs is small compared to $k_B T$ or when the broadening of the LLs is large. Without going further into the details, let us just mention that, with Ozaki’s approximation to $f_0$, the integrated DOS can be expressed as a sum of the imaginary part of Faddeeva functions (and similarly for $\sigma_{xx}$).

As mentioned above there are multiple choices of LL broadening, the two most widely used being the Gaussian line shape with a variance proportional to $\sqrt{B}$ and the Lorentzian broadening. These two broadenings are in fact relatively similar, as they both give rise, at high LL index, to an increase of the amplitude of the quantum oscillation as the magnetic field is swept going as $\propto e^{-C/B}$, with $C$ a constant different in the two cases (similar to the Lifshitz-Kosevich formula). Yet, numerically, the choice of the Gaussian line shape is much more convenient due its rapid decay away from the LL center. Thus, when calculating the conductance oscillations in the presence of Rashba/Zeeman interaction, we set : $\Gamma_\pm = \gamma \pm \sqrt{B}$. Ideally, this choice should be motivated by a microscopic theory of the scattering processes in our system. The consequence of this arbitrary choice is that the numerical model best fits the position of the oscillations rather than their amplitude.

Finally, in addition to an oscillating part, eq. (5.59) produces a “background” (or “non-oscillating”) evolution of $\sigma_{xx}$ (going roughly as given by the semi-classical equation 4.16). Hence, the oscillating part $\Delta \sigma$ of the conductance is obtained by subtracting a curve computed using a sufficiently high temperature $T_{\text{high}}$, at which the quantum oscillations are completely suppressed :

$$\Delta \sigma(B, T) = \sigma_{xx}(B, T) - \sigma_{xx}(B, T_{\text{high}})$$  (5.61)

5.5 The LaAlO$_3$/SrTiO$_3$ interface

As mentioned in the section 3.4.3 of this thesis, using appropriate growth conditions, the electronic mobility at the LaAlO$_3$/SrTiO$_3$ interface can be as high as $5000 - 8000$ cm$^2$ V$^{-1}$ s$^{-1}$, at low temperatures. In these conditions, $\omega_c \tau = 1$ at approximatively 1.5 – 2T and hence clear Shubnikov-de Haas (SdH) oscillations are expected to appear in the magnetoresistance signal.

Figure 5.7 (left) shows the variation of the longitudinal resistance of a sample grown at 650 °C upon application of an out-of-plane magnetic field. Clearly, as the temperature of measurement is decreased from 10 K to 1.5 K, quantum oscillations develop. A large background magnetoresistance, similar to the one observed in the last chapter for $\sigma_{02D}^0 \approx 10$ mS, is also present. To remove this contribution, we take the derivative of the magnetoresistance data before plotting it on a $1/B$ scale (remember that, according to
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Figure 5.7: Shubnikov-de Haas oscillations at the LaAlO$_3$/SrTiO$_3$ interface. a Variation of the resistance $\Delta R = R(B) - R(0)$ in response to the application of a magnetic field $B$ oriented perpendicular to the LaAlO$_3$/SrTiO$_3$ interface, recorded at different temperatures $T$. b Numerical derivative $\frac{dR}{dB}$ as a function of the inverse of the magnetic field.

the Bohr-Sommerfeld quantization rule, the quantum oscillations are periodic in $1/B$, see eq. (5.11)). We observe that the oscillations are, in first approximation, periodic in $1/B$ but that the behavior of their amplitude is rather complex.

Below, we will see that further reducing the temperature below 1 K, the pattern of the oscillations becomes even more subtle. But before that, we focus on the dependence of the SdH oscillations on the angle $\theta$ between the magnetic field and the normal to the conducting plane.

5.5.1 Angular dependence of the SdH oscillations

Figure 5.8 shows, for another sample grown at 650 $^\circ$C, the evolution of the SdH oscillations at 1.5 K varying the angle $\theta$. Clearly, the oscillations disappear as $B$ is oriented parallel to the interface plane ($\theta = 90^\circ$). Consistent with previous reports on the parallel magneto-transport in the high conductance regime \cite{27,28,116} a negative magnetoresistance is observed in this orientation. The analysis of the resistance maxima and minima shown in fig. 5.8 reveals that their position in magnetic field scales as $(B \cos \theta)^{-1}$ rather than $B^{-1}$, following the prediction of the semi-classical model (eq. (5.14)) for a 2DEG.

In fig. 5.9, we show the very same analysis repeated at very low temperature (90 mK) and very high magnetic fields (up to 33 T). Similarly to the results at 1.5 K, the position of the oscillations globally aligns on a $(B \cos \theta)^{-1}$ scale. Yet, at high fields or high angles deviations from this law are observed.

Recently McCollam et al. \cite{132} reported similar observations. This seems to indi-
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Figure 5.8: Angular dependence of the quantum oscillations at the LaAlO$_3$/SrTiO$_3$ interface. a Sheet resistance $R_s$ as a function of magnetic field $B$ recorded at different orientations (measured by the angle) with respect to the direction normal to the substrate. b Numerical derivative $\frac{dR}{dB}$ as a function of the inverse of the magnetic field recorded at different orientations. c Numerical derivative $\frac{dR}{dB}$ as a function of the inverse of the component of the magnetic field perpendicular to the interface plane. An offset has been introduced in each curve for clarity. The lines are guide to the eye.

Figure 5.9: Angular dependence of the quantum oscillations at the LaAlO$_3$/SrTiO$_3$ interface up to 33 T. a Sheet resistance $R_s$ as a function of magnetic field $B$ recorded at different orientations (measured by the angle) with respect to the direction normal to the substrate. c Numerical derivative $\frac{dR}{dB}$ as a function of the inverse of the component of the magnetic field perpendicular to the interface plane. An offset has been introduced in each curve for clarity. The lines are guide to the eye.
cate that the situation becomes more complex when the Shubnikov-de Haas oscillations are better resolved. There are different interesting scenarios to account for this, including spin-(orbit) related effects, inter-subband scattering or interaction between LLs. As we will see, we obtained similar deviations performing field effect experiments on samples displaying large SdH oscillations. Yet, the overall dependence in $(B \cos \theta)^{-1}$ of the maxima of the quantum oscillations (fig. 5.8c) and the absence of oscillations in parallel orientation are strong indications of the 2D character of the electronic states giving rise to the quantum oscillations at the LaAlO$_3$/SrTiO$_3$ interface.

5.5.2 Shubnikov-de Haas oscillations in the milli-Kelvin regime

Figure 5.10a displays a set of sheet resistance versus magnetic field curves, recorded on a “high mobility” sample, for temperatures ranging from 800 to 50 mK. As can be seen, the magnitude and the complexity of the Shubnikov-de Haas oscillations increases markedly as the temperature is lowered. At 50 mK and in high magnetic field, the amplitude of the SdH oscillations is about 10–15% of the sheet resistance value. On this sample we used field effect to change the carrier density. Figure 5.10c shows that the transverse resistance $R_{xy}$ varies linearly with magnetic field for all dopings. From fig. 5.10d, we see that ramping the gate voltage up to larger positive values (largest $\sigma_{2D}$) leads to an increase of the inverse Hall coefficient. Analysing the Hall signal using a single-band model, we extract a carrier density that increases from 2.5 to $4.8 \times 10^{12}$ cm$^{-2}$ as the sheet conductance at 50 mK increases from 1.6 to 5.27 mS.

Along with this modulation of the electron density, field-effect doping leads to an increase in the electron mobility $\mu$ as shown in fig. 5.10e where one can observe that $\mu$ evolves from 3900 to 6900 cm$^2$ V$^{-1}$ s$^{-1}$. As can be seen in fig. 5.10b, the changes in electron mobility and density strongly modify the structure of the SdH oscillations with a clear change in both the peak positions and the period of the oscillations.

In order to proceed with the analysis of the SdH data presented in fig. 5.10, we subtracted the large non-oscillating background from the sheet conductance, calculating $\Delta \sigma(B)$ as:

$$\Delta \sigma(B) = \frac{R_s(B)}{[R_s(B)]^2 + [R_{xy}(B)]^2} - \sigma_0(B)$$  \hspace{1cm} (5.62)

with $\sigma_0(B)$ representing a polynomial background. The resulting curves can be found in fig. 5.17a.

Contrarily to the last paragraphs, here we choose to work with the sheet conductance. The reason is that, as illustrated by the theoretical developments of the last sections, the DOS is more directly linked to the electrical conductance than to the resistance. In fact, the hypothesis implicitly made when working with $R_s$ rather than $\sigma_{xx}$ is that $R_{xy} \gg R_s$. This leads to (see eq. (4.18)):

$$R_s \approx \sigma_{xx} R_{xy}^2$$  \hspace{1cm} (5.63)

If $R_{xy}$ does not display plateaux (i.e. we are not in the quantum Hall regime), it represents just a factor of proportionality between the conductance and the resistance of the system.
Figure 5.10: Transport properties under magnetic field. 

(a) Temperature evolution of the sheet resistance ($R_s$) versus magnetic field for a doping level corresponding to a sheet conductance of 5.23 mS and a mobility of $\mu \approx 7000$ cm$^2$/Vs at 50 mK. Curves are offset for clarity. The sheet conductance at 50 mK ($\sigma_{2D}$) is used as a reference for the doping level. 

(b) $[R_s(B) - R_s(0)]/R_s(0)$ for different dopings, illustrating the evolution of the SdH oscillations with gate voltage. Curves are offset for clarity. 

(c) Hall resistance versus magnetic field at 50 mK for different dopings. 

(d) Inverse Hall coefficient and the corresponding Hall mobility at 50 mK as a function of the sheet conductance.
5.5 The LaAlO$_3$/SrTiO$_3$ interface

Figure 5.11: Analysis of the SdH oscillations and comparison with the two-band model. a FT of the experimental signals for all the dopings. Dashed grey lines are guides to the eye. b Comparison between $\Delta \sigma$ versus $1/B$ calculated within the two-band model (red) and the experimental data (black) for the doping with the highest conductance (5.27 mS). The exponential factor $e^{5/B}$ is used to magnify the low-field region.

Given the value of $R_{xy}$ in our samples (at 2 T and 5.23 mS $R_{xy} \approx 270 \, \Omega$), eq. (5.63) would not be a reliable approximation for the analysis of the oscillations. In addition to that, as one possible explanation concerning the complex pattern of oscillations observed in fig. 5.10 is the presence of several bands contributing the quantum oscillations, working with $\sigma_{xx}$ allows their contribution to be easily summed (see eq. (4.20)).

We first analyse $\Delta \sigma(B)$ using Fourier Transform (FT) since in the LK formalism (eq. 5.33) the quantum oscillations have well defined frequencies. Figure 5.11a displays the FT of the magnetoconductance for the different dopings. Looking at the highest doping (largest conductance), we observe a well-defined peak at 55.5 T, a second peak around 18.5 T and a third maximum at 5 T. The doping dependence of the FT spectrum shows that, while the position of the 5 T peak remains relatively unchanged, the other two shift to lower frequencies upon decreasing the carrier density, as one would expect. From this qualitative analysis, we may infer the existence of two distinct trajectories on the Fermi surface. Thus, we consider a model with two parabolic bands for which the magnetoconductance can be calculated using the LK formula [125]. We fit the data for the largest conductance introducing an arbitrary phase for each frequency.

5.5.2.1 The two-band model

As can be seen from fig. 5.11b, a good fit to the data can be obtained using the two frequencies 18 T and 55.9 T. Considering the Onsager relation (or more precisely eq. (5.13)) with a spin degeneracy $\nu_s = 2$ and a valley degeneracy $\nu_v = 1$ for the two bands, we find the carrier densities for the two bands to be 0.87 and $2.7 \times 10^{12} \text{ cm}^{-2}$, yielding a total carrier concentration of $\sim 3.6 \times 10^{12} \text{ cm}^{-2}$. This value is of the same order of magnitude as that extracted from Hall effect measurements, albeit slightly lower. We note that similar discrepancies between the sheet carrier densities obtained using Hall effect data and SdH oscillations have already been reported [29,108,133,134]. A possible explanation is that carriers with a lower mobility contribute to the Hall
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response, but not to the oscillations.

As explained above, in the LK formalism, the temperature evolution of the oscillations has the following functional form:

\[
\Delta \sigma_i(B_m, T) \propto \frac{\alpha_{LK,i} T}{\sinh(\alpha_{LK,i} T)}
\]  

(5.64)

with \( \alpha_{LK,i} = \frac{2\pi k_B}{\hbar \omega_{c,i}} \), \( \omega_{c,i} = eB_m/m_i^* \), \( B_m \) the field at which the extremum is observed and \( i \) the band index.

In fig. 5.12 we show the effective mass plot for the high and low frequency (HF and LF) parts of the SdH oscillations shown in fig. 5.11b. We used filtering techniques to extract the HF and LF parts from \( \Delta \sigma \). Hence, we were able to perform the effective mass estimation on 17 and 4 extrema for the HF and LF, respectively. Good agreement between theory and experiment is obtained by choosing an effective mass of \( 2.7m_e \) for the HF and \( 1.25m_e \) for the LF.

The value of the lower mass is in line with previously reported estimates for STO based heterostructures [29, 133, 134]. Concerning the higher mass, in chapter 4, by analysing the magnetotransport in perpendicular and parallel fields, we have uncovered the presence of carriers with such a mass at the LaAlO\(_3\)/SrTiO\(_3\) interface. We note that, according to the LK formula, oscillations associated with a large effective mass are more affected by disorder and/or temperature-induced Landau level smearing, hence more difficult to observe.

With the information extracted from this analysis, the electronic structure of the two-band model can be reconstructed and the splitting at the Fermi level between the heavy and the light bands can be determined (see fig. 5.13):

\[
\Delta E = \left| E_1(\vec{k}_F) - E_2(\vec{k}_F) \right|, \quad E_i(k) = \frac{\hbar^2}{2m_i^*} \left( k^2 - k_{F,i}^2 \right).
\]  

(5.65)

\( k_{F,i} \) is the Fermi momentum in the \( i \)-th sub-band obtained from the area \( A_i = \pi k_{F,i}^2 \) calculated using the Onsager relation, and \( \vec{k}_F = (k_{F,1} + k_{F,2})/2 \). We find \( \Delta E \approx 2.45 \text{ meV} \).

Remarkably, the calculated band splitting \( \Delta E \) is similar to the typical spin-orbit splitting estimated for LAO/STO heterostructures [27, 28]. We also note that, the low magnetic field region of the data presented in fig. 5.10a and b, reveals the presence of an antilocalization cusp (see fig. 5.14). In line with eq. (4.42), due to the higher mobility of this sample, the WAL correction is restricted to much lower values of the magnetic field than for standard LAO/STO interfaces for which \( l_{el} \) is approximatively ten times smaller.

5.5.2.2 Analysis of the SdH oscillations within the Rashba+Zeeman model

Here, we apply the linear Rashba/Zeeman scenario developed in the preceding theoretical paragraphs of this chapter to our data. In this model, the SO interaction splits the LLs of a single band into two families (see eq. (5.55) and (5.53)), giving rise to two types of quantum oscillations. The comparison between the data and the best fit is displayed in fig. 5.15: as can be seen, the model fits the data well. Remarkably, while
Figure 5.12: Extraction of the effective mass from the temperature dependence of the Shubnikov-de Haas oscillations. Using filtering techniques the temperature evolution of 17 and 4 extrema, for the HF and LF respectively, is considered. Each color indicates the temperature evolution of a single extrema. Optimizing the value of $m^*$, the horizontal scale is adapted in order that the measurements lie on a straight line of slope 1 and of 0 ordinate.
Figure 5.13: Band structure obtained from the analysis of the Shubnikov-de Haas oscillations in the two-band model.

Figure 5.14: Variation of the sheet resistance versus magnetic field for different dopings (close up of the low magnetic field regime of fig. 5.10b). A characteristic antilocalization cusp is observed for $B < 0.06$ T.
in this second scenario the energies of the LLs are controlled by only three parameters \((E_F, a, b, d)\), the fit is at least as good as the two-band fit that requires four parameters \((F_1, \phi_1, F_2, \phi_2)\). Panel \(b\) displays the derivatives of the theoretical and experimental curves allowing the positions of the maxima and minima to be compared.

The carrier density extracted from this analysis is \(n_{2D}^{\mathrm{SdH}} = 1.83 \times 10^{12} \text{ cm}^{-2}\), substantially lower than the one found using the Hall effect measurements. As discussed for the two-band model, this is not surprising, as only bands with a mobility larger than \(\approx 2000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\) will give rise to observable SdH oscillations for the fields used here. The magnitude of the obtained Rashba spin-orbit constant \((\alpha = 3.4 \times 10^{-12} \text{ eV m})\) agrees very well with values obtained from weak localization analyses and from modeling of the transport data in parallel fields \([27, 28, 68]\). We note that, given the small value of \(k_F\) in our samples, a \(k\)-cubic Rashba interaction inducing a spin-splitting of \(\approx 2 \text{ meV}\) would require a very large coupling constant \(\alpha_3\), beyond the values recently reported by H. Nakamura et al. \([135]\).

To obtain the effective mass, we selected three peaks from a region of magnetic fields where the amplitude of the oscillations is large. Figure 5.15c shows that the data can be fit perfectly using an effective mass of \(2.19 \pm 0.06 m_e\). Surprisingly, this value indicates that the electronic state of the oscillating carriers is probably not dominated by Ti \(d_{xy}\) orbitals, as one would then expect a lower effective mass (\(\sim m_k\)). Instead, the higher mass obtained in this analysis can be understood by taking into account the contribution of \(d_{xz}/d_{yz}\) orbitals to the electronic states.

This observation corroborates our results on standard LAO/STO interfaces, where a sharp decrease in the elastic scattering rate was correlated to the progressive appearance, at the Fermi level, of heavier carriers \([68]\). The average effective mass of the carriers was observed to evolve from \(\sim 0.7 m_e\) to \(\sim 2.2 m_e\). 2.2\(m_e\) being precisely the value observed here. These results appear to showcase the critical role played by the heavier sub-band in establishing high-mobility at the \(\LaAlO_3/\SrTiO_3\) interface. We note that this feature gives further substance to a scenario of spin-orbit protected transport due to a Fermi surface reconstruction induced by the Rashba interaction \([68]\).

Because the Zeeman energy enters equation 5.53 only as a squared term, for the LLs with \(N > 0\), we find two solutions for the \(g^*\)-factor, namely 5.2 or \(-3.4\), values significantly different from 2, as is also the case for semiconductor heterostructures. We note that, as seen in chapter 2, atomic SO mixes the orbital character of the electronic states at the \(\LaAlO_3/\SrTiO_3\) interface. In this case, the spin is not a good quantum number as assumed for the diagonalization of the Landau Hamiltonian in the presence of Rashba and Zeeman interaction. The impact on the estimation of the \(g^*\)-factor maybe important as recently pointed out by van der Marel et al. in bulk \(\SrTiO_3\) \([52]\).

From the value of the LL broadenings (\(\Gamma^\pm\)) the quantum lifetime (\(\tau_Q\)) and hence the quantum mobility (\(\mu_Q\)) can be computed. We obtain \(\mu_Q \approx 2000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\) for the “+” states and \(\mu_Q \approx 1000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}\) for the “−” states. These values are both lower than the mobility extracted from the Hall measurements, in agreement with literature \([126]\).

We can estimate the Rashba splitting and the Fermi energy of the high-mobility

\[\text{1The quantum scattering time is related to the total scattering rate while the transport lifetime is weighted by a factor } (1 - \cos \theta) \text{ with } \theta \text{ the scattering angle.}\]
Figure 5.15: Modeling of the SdH oscillations within the Rashba/Zeeman scenario. 

a Comparison between $\Delta \sigma$ measured experimentally for $\sigma_{2D} = 5.27$ mS (black) and calculated using our single-band model with Rashba and Zeeman interactions (green). 

b Derivative with respect to $B^{-1}$ of the curves presented in a. The red dots indicate the values of $B^{-1}$ where extrema are observed in the modelled curve. The exponential factor $e^{5/B}$ is used to magnify the low-field region. 

c Field and temperature dependence of the oscillations over the two ranges of applied field used to extract the effective mass. Black dashed lines are the theoretical curves computed using $m^* \sim 2.2 m_e$, while the thick colored lines are the experimental data.
carriers. Interestingly, we find that both are of the same order of magnitude: \( \Delta R = 2.2 \text{ meV} \) and \( E_F = 1.65 \text{ meV} \), a situation very different from the one of semiconductor 2DEGs, where generally the Fermi energy dominates. The corresponding band structure is displayed in fig. 5.16. We note that, at the Fermi level, it is very similar to the band structure displayed in fig. 5.13, hence the difficulty to discriminate between the two models. Yet, in our opinion, the Rashba model better fits with the overall interpretation of the LaAlO\(_3\)/SrTiO\(_3\) magnetotransport data.

5.5.2.3 Gate voltage evolution of the Shubnikov-de Haas oscillations

We now discuss the gate voltage dependence of the SdH oscillations. Figure 5.17a shows the change in conductance as a function of \( 1/B \) for various \( V_g \), after subtraction of the background. A clear evolution of the SdH oscillations with doping is visible and is compatible with the shrinking of the Fermi surface expected from Hall measurements. Surprisingly, we find that the one-band model described above, including Zeeman and Rashba interactions, is only able to reproduce (with slowly varying fitting parameters) the evolution of the experimental data in a very narrow voltage region (typically over a 4–5 V range).

To understand this behaviour, we identify the position in magnetic field of each maximum as a function of the applied gate voltage. We focus on the “+” levels of equation 5.53 (corresponding to the high frequency modulation) since these extrema are better defined. A convenient way to pinpoint the positions of these maxima is to determine the magnetic fields at which \(-\Delta \sigma''(1/B) = -\partial^2 \Delta \sigma / \partial (1/B)^2 \) is maximal (the second derivative amplifies the high frequencies). The maxima determined in this
Figure 5.17: Analysis of the doping dependence of the SdH oscillations. a Doping dependence of $\Delta \sigma$. Curves are shifted for clarity. Black dots indicate the position where maxima occur in $-\Delta \sigma''(1/B)$. b Fan diagram showing $-\Delta \sigma''(1/B)$; yellow corresponds to the maxima and black to negative values. c Fan diagram calculated using the energy levels from equation 5.53. The parameters of the fit presented in fig. 5.15 were used for the highest doping. For lower dopings, a smooth evolution of $E_F$ with $\sigma_{2D}$ was assumed.
way are indicated in fig. 5.17a by black dots. We expect that the trajectories traced out by the black dots as a function of $V_g$ correspond to the evolution of each LL as a function of the chemical potential. Strikingly, we see that these trajectories present sharp deviations or jumps upon varying $V_g$. This observation explains why a fit of the SdH oscillations using the Rashba-Zeeman model does not produce a smooth evolution of the parameters with gate voltage.

Figure 5.17b displays a fan diagram showing $-\Delta\sigma''(1/B)$. This nicely illustrates the fact that the positions of the “+” LLs follow a simple evolution only for limited regions of the diagram. Conversely, we observe that at precise locations the amplitude of the SdH oscillations is strongly suppressed.

This fan diagram is reminiscent of those observed in semiconductor 2DEGs when LLs originating from different sub-bands show crossings [136–140]. In that case, the behaviour of LLs deviates from the independent-level picture, electron-electron interactions lifting the degeneracy at the crossing points. We note that a similar scenario can be at play in our system.

We also investigated the Rashba scenario described above, calculating a fan diagram using the energy levels in equation 5.53. As starting parameters, we took the values from the fit shown in fig. 5.15, and assumed a smooth evolution of the Fermi energy with gate voltage. The result, displayed in fig. 5.17c, confirms indeed that LLs from the two different families (“+” and “−”) exhibit crossings. While the matching between theory and experiment is not perfect here, “anomalies” in fig. 5.17b nevertheless appear to correlate with regions where a crossing between LLs of different sign (±) is predicted from fig. 5.17c. We thus attribute the jumps in the trajectories of the “+” LLs presented in fig. 5.17a, to anti-crossings with “−” levels, that occur at the Fermi energy. Indeed, in this case the independent-level picture breaks down due to Coulomb interactions between electrons belonging to adjacent LLs. Interestingly, these many-body effects can lead to a variety of phenomena ranging from quantum Hall ferromagnetism to charge-ordered states [141, 142].

In conclusion, the Shubnikov-de Haas oscillations at the LAO/STO interface present a remarkably complex behaviour. While our analysis reveals the important role played by the Rashba SO interaction on the electronic band structure, the evolution of the LL spectrum as a function of doping and magnetic field suggests that electron-electron interactions substantially modify the independent-level picture, as a result of level crossings at the Fermi energy.
Summary

The LaAlO$_3$/SrTiO$_3$ interface is a fascinating two-dimensional electronic system. It displays a room-temperature insulator to metal transition that can be induced either by changing the thickness of the LaAlO$_3$ layer or by field effect. At low temperature, a very complex magnetotransport is observed, probably owing to the occupation of multiple sub-bands and to the presence of a strong Rashba spin-orbit coupling induced by the huge electric field confining the charges at the interface. In the milli-kelvin regime 2D superconductivity is present. Using field effect in a back gate geometry, the transition temperature can be tuned revealing a phase diagram endowed with a quantum critical point that separates an insulating region from a superconducting one. Magnetotransport is also strongly modified by the application of an electric field: detailed analyses reveal a large evolution of the characteristic scattering times across the quantum critical point.

In this study, we have measured and analyzed the magnetotransport properties of the LaAlO$_3$/SrTiO$_3$ interface across its phase diagram, with the aim to obtain a better understanding of the band structure of the system. Through detailed modeling, we have investigated the role of some key parameters that should be considered when discussing the band structure of the interface. This is, for example, the case for in-gap states that were shown to modify the confining potential of the electron gas.

The analysis of the magnetotransport data as a function of gate voltage indicates a change in sub-band population and an important role played by spin-orbit interaction in the band structure of the system. These observations provide strong motivation to investigate the role of the different bands with different symmetries and of the Rashba spin-orbit interaction on the superconducting state.

We also studied in detail the low temperature magnetotransport properties of a new class of LaAlO$_3$/SrTiO$_3$ interfaces, grown at a lower temperature and displaying a much larger electron mobility. The analysis of the spectrum of the Shubnikov-de Haas oscillations suggests a large impact of the Rashba interaction on the electronic structure. Interestingly, in the low carrier density regime where we observe the quantum
oscillations the Fermi energy is of the same order of magnitude as the Rashba spin-orbit splitting. Studying in detail the gate voltage tuning of the SdH oscillations, we put into evidence sharp deviations in the Landau level evolution as a function of magnetic field and doping, an observation that suggests physics beyond the independent particle approximation at play in this system.

Perspectives
Of course, there are still many open issues in the LaAlO$_3$/SrTiO$_3$ system. For example, the precise determination of the shape of the Fermi surface, the detailed mechanism leading to interfacial doping, the nature of the superconductivity pairing and the difference(s) between high-mobility and standard samples. Those are critical points if one is interested in building more complex heterostructures, like, for example, multilayer LaAlO$_3$/SrTiO$_3$ heterostructures or junctions with other materials. Hence, new experiments aimed to clarify these points are highly desirable. Yet, the discovery of high mobility interfaces with very low carrier densities is opening new directions of study.

Indeed, in the last part of our work, the mobile carrier density was so low that the Hall effect reached a maximum of 2 kΩ at 8 T. Applying higher magnetic fields could bring us into the quantum Hall regime. This would certainly be an exciting opportunity to further explore the physics of this heterostructure and, given the complex evolution of the Landau levels we observed in chapter 5, would probably bring many surprises.

Recently, we have demonstrated the feasibility of Hall bars with lateral size ≈ 500 nm [143]. As this dimension will be reduced towards the electron mean free path at high-mobility interfaces (≈ 200 nm), we can envisage ballistic transport experiments at an oxide interface. For sure, those will be exciting experiments as they will give access to mesoscopic phenomena.

To make a long story short, the LaAlO$_3$/SrTiO$_3$ system just celebrated its 10th anniversary at the beginning of this year, which, compared to the 60 year old silicon transistor, makes this system look like a promising young adolescent.
A.1 Sample preparation

In this appendix we detail step by step the procedure we follow to prepare high quality LaAlO$_3$/SrTiO$_3$ interfaces and to transform them into field effect devices. Our supplier for SrTiO$_3$ substrate is CrysTec GmbH. We buy in general crystals with dimensions $5 \text{ mm} \times 5 \text{ mm} \times 0.5 \text{ mm}$ ($W \times L \times H$), with a (001) oriented TiO$_2$ terminated surface, one side polished and a miscut angle $<$ $0.1^\circ$.

A.1.1 Patterning

- In the clean room, we check the cleanliness of the polished substrate surface. If some residue is present, we bathe the substrate in acetone and shake it using the ultrasonic bath. The same procedure is then repeated with ethanol as acetone is likely to leave some residue on the surface. To get rid of the ethanol, we blow on the surface of the substrate with nitrogen gas. If it happens that the surface is still dirty a cotton tip wet with acetone can be used to gently brush the surface. Solvents can also be heated.

- When the substrate is clean, we use a spinner to deposit on it a $1.3\mu m$ thick layer of a photosensitive polymer (the photoresist). We use the resin S1813 G2 from Rohm & Haas. By spinning it at $4000\text{ t/min}$ we get the desired layer thickness. Because of the small dimension of the substrate we want to pattern, special care must be taken when fixing it on the spinner, as resin accumulation on the surface edge can easily occur. To avoid that, the position of the substrate on the spinner has to be off-centered. In addition to that, the sample can be set in modeling clay to the surface edge. Finally, the photosensitive polymer should cover a larger area than the sample surface before spinning.

- The resin is dried at $115^\circ C$ for 1 min.
A. GROWTH AND PATTERNING OF LAO/STO INTERFACES: STEP BY STEP PROCEDURE

- The polymer S1813 is a positive resist meaning that if some parts of the layer are exposed to UV light they will become soluble in a developer. We use a 200 W mercury short arc UV lamp (Osram) in combination with a shadow mask to expose selected regions of the photoresist. These regions are subjected to 12 mW cm$^{-2}$ of UV radiation. The exposure lasts $\approx 12$ s. The shadow mask is a simple piece of glass with black patterns (UV-opaque) drawn on it. It has to be put in contact with the surface of the substrate. When preparing LaAlO$_3$/SrTiO$_3$ interfaces we use a shadow mask that protects the Hall bar we want to design. With this technique the smallest structures we can design have lateral sizes of $\approx 2\mu$m. To produce smaller structure electron beam lithography can be attempted.

- After exposure, the sample is immersed in a developer for 45-60s (Microposit 351 from Rohm & Haas) and rapidly after in de-ionized water to stop the development process. The sample is dried by blowing on the surface with nitrogen gas. After this operation, the substrate surface is clean apart from the Hall bar regions where it is covered by the unexposed photoresist. In case of failure of the above process, the resin can be easily removed bathing the substrate in acetone and then ethanol.

- The major drawback of using a polymer is that it cannot be heated to our LaAlO$_3$ deposition temperature (650-900°C). This is the reason why we deposit amorphous SrTiO$_3$ on the sample surface (the channel being protected by photoresist). We use PLD. As the deposition takes place at room temperature the substrate can be stuck with adhesive tape on the nickel plate of the sample holder (see below). During deposition an oxygen pressure of $8 \times 10^{-5}$ mbar is set (with vacuum, the plume would etch the substrate). The laser we use to ablate the SrTiO$_3$ target is a KrF excimer laser (pulsed laser), with a wavelength of 248 nm. The target is a $10\,\text{mm} \times 10\,\text{mm} \times 0.5\,\text{mm}$ SrTiO$_3$ single crystal placed 7 cm from the substrate surface. The laser energy is set to 50 mJ and the repetition rate is 5 Hz. The laser pulse is focused on the surface of the target to an area of $\approx 5\,\text{mm}^2$ (fluence of 1 J cm$^{-2}$). We deposit in general $\approx 30$ nm of amorphous SrTiO$_3$, which corresponds to 3000 pulses of laser (or 10 min of deposition). After growth, the nickel plate is cleaned in acetone to remove the scotch residues.

- To finalize the patterning process we get rid of the photoresist by bathing the substrate in acetone and then ethanol (“lift-off” process). The sample is dried by blowing on the surface with nitrogen gas. At this stage, the only exposed (to air) part of the substrate surface is the Hall bar.

A.1.2 LaAlO$_3$ growth

- As the LaAlO$_3$ growth is realized at rather high temperatures (650-900°C), the substrate has to be fixed on the PLD sample holder in a manner that ensures good adhesion and good thermalization. To this purpose, we stick the (patterned) SrTiO$_3$ substrate on a $10\,\text{mm} \times 10\,\text{mm}$ nickel plate using silver epoxy (P10 from Epoxy Technology), and dry it for at least 1 h at 120 °C in air. This results in
very good adhesion, so a minimum of silver epoxy should be used, the important point being that all the unpolished side of the crystal is covered (we will see below that residues of epoxy can be annoying to create a field effect device in a back-gate configuration). If a temperature higher than 900 °C is desired another fixing scheme has to be adopted, as the silver epoxy will melt.

- To heat the sample, we use an infrared laser of 114 W able to provide a heating current of up to 30 A. The beam is focused on the back-side of the nickel plate. Thus, we insert a (transparent) 10 mm × 10 mm sapphire crystal in-between the PLD sample holder and the nickel plate to avoid heat losses (to the holder). Screws are used to fix together the holder, the sapphire crystal and the nickel plate. Thermal decoupling is achieved by screwing them as lightly as possible.

- The next step is to remove adsorbates from the exposed surface of the SrTiO\textsubscript{3} substrate. To this purpose, we heat the sample to 290 °C in vacuum (1 × 10\textsuperscript{-6} mbar) for 30 min before transferring it in situ to the deposition chamber (we use a resistive heater located in a adjacent vacuum chamber).

- The substrate is further heated (in the deposition chamber) in an oxygen pressure of 8 × 10\textsuperscript{-5} mbar, in 1 h to 800 °C (about 19 A) for “standard samples” or 650 °C (about 13 A) for “high-mobility samples” using the infrared laser. The temperature is monitored using an infrared pyrometer measuring the light emitted by the silver epoxy because of heating (the emissivity of silver epoxy is known to be 0.85).

- RHEED is used to monitor the LaAlO\textsubscript{3} growth. Due to its surface sensitivity it cannot by used on the surfaces covered by the amorphous SrTiO\textsubscript{3}. As the projection of the beam on the surface of the sample is a rectangle of about 2 mm × 70 μm while the channel is only 500 μm wide, it can sometimes be challenging to find a nice diffracting area (fortunately the electron beam and sample position/orientation can be modified in situ). The acceleration voltage, filament current and emission current are set respectively to 22 kV, 2.3 A and 75 μA. Between 650-900 °C the growth proceeds layer by layer, allowing precise determination of the thickness through RHEED oscillations to be made.

- The repetition rate of the pulsed laser is 1 Hz. The target is made of a pressed LaAlO\textsubscript{3} powder. The deposition parameters are otherwise the same as for the deposition of amorphous SrTiO\textsubscript{3}. In these conditions, ≈ 60 laser pulses are necessary to grow one unit cell of LaAlO\textsubscript{3}. We typically grow between 5 and 15 unit cells of LaAlO\textsubscript{3} on top of the SrTiO\textsubscript{3} substrate as for thicker samples relaxation of the film occurs.

- After growth, the deposition chamber is further filled with 0.2 bar of oxygen. This has the effect to reduce the temperature of the sample due to thermal conduction. The intensity of the infrared laser is adjusted to reach a temperature of 530 °C. The sample is left for one hour in this environment (oxygen annealing procedure). Then the laser intensity is brought to 0 A in hour.
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• Finally, the sample is detached from the nickel plate. This is not an easy task, as mechanical stress on the LaAlO$_3$/SrTiO$_3$ heterostructure should be minimized.

A.1.3 Creation of Ohmic contact pads

Generally, the use of aluminum bonding wires to directly contact the 2DEG at the LaAlO$_3$/SrTiO$_3$ interface suffices. Yet, in certain circumstances, it is desirable to create metallic contact pads which lead to a lower contact resistance.

• In the clean room, we check the LaAlO$_3$ surface, cleaning it if necessary. Note that, as the impact the ultrasonic bath has on the integrity of the heterostructure is not clear, the best is to keep the sample as clean as possible after growth.

• The same photolithographic procedure described in sub-section A.1.1 is applied to shape a photo-sensitive layer on top of LaAlO$_3$. In this case a shadow mask with transparent areas only at the electrical contact locations is aligned with the pre-patterned Hall bar.

• We make use of an ion beam etching system combined with an electron beam physical vapor deposition system. The goal here is to remove, in the selected regions, the deposited LaAlO$_3$ layers and $\approx 10$ nm of SrTiO$_3$. Then we want to fill the formed well with Ti and cap it with Au.

• The pressure in the chamber is first reduced to $\approx 5 \times 10^{-7}$ mbar. Then $1.1 \times 10^{-4}$ mbar of argon is introduced.

• We etch the area of the contacts for 13.5 min. Since the sample surface is insulating charging of the surface occurs and a rather intense etching current needs to be used. To prevent excessive heating of both the photoresist and the interface, 2 min pauses are done every 3 min of etching. During the pauses a shutter is brought in front of the sample and the argon gun is turned off.

• The energy of the argon ions is set to 470 eV and the associated current density is 0.85 mA cm$^{-2}$.

• After etching the argon gas is pumped out. When the pressure is around $3 \times 10^{-7}$ mbar, we start evaporating Ti in the chamber with a shutter in front of the sample (pre-deposition). The quantity of evaporated material is measured using a quartz crystal resonator. We pre-deposit 10 nm of Ti. This further reduce the pressure of the chamber to $8 \times 10^{-8}$ mbar due to the high oxidation rate of Ti.

• The shutter is removed and 15 nm of Ti are deposited on the sample surface at a rate of 0.1 nm s$^{-1}$.

• Subsequently, 20 nm of Au are deposited at a rate of 0.1 nm s$^{-1}$.

• We lift-off of the photoresist (see above). To avoid using the ultrasonic bath the heterostructure is left 1 night in (hot) acetone.
A.1 Sample preparation

A.1.4 Back gate deposition

- As mentioned above, the use of silver epoxy to fix the SrTiO$_3$ substrate on the PLD nickel plate leaves residue on the unpolished side the sample. Thus, the first step is to get rid of this residue. First, we use a scalpel to scrape the epoxy accumulated on the sides of the crystal. This is a difficult task as the sample has to stand on its edge. We use a soft tissue as work surface and blow out all the silver particles that detach from the substrate as soon as they fall on the work surface.

- To clean the unpolished side of the sample we turn it upside down and use sand paper (if a lot of silver epoxy is present we peel off the excess with the scalpel). There is a risk that silver and/or sand paper particles slip in-between the crystalline surface and the work surface, so it’s necessary to press the substrate against the tissue during abrasion. We never press directly with tweezers on the unpolished crystal surface, as they will leave some residues on the surface. We blow the detached particles away and ensure always to work on a clean surface, eventually changing the position of the sample on the work surface.

- When the sample is clean, by transparency, we can see the Ti/Au electrodes looking from the backside of the crystal. We lie the sample, with the crystalline surface facing down, on a piece of teflon and fix it. Then we cover all the unpolished side of the substrate apart from a rectangle aligned with the channel. Note that not all the channel must be covered, just the part where resistance measurements are performed. Typically the size of a back-gate electrode is 0.75 mm × 2.5 mm.

- We use electron beam physical vapor deposition to deposit 20 nm of Au on the backside of the substrate (see above).

- After deposition, a drop of silver paint (DuPont 4929N) is used to stick a 0.1 mm copper wire to the electrode. We use coated copper wires (i.e. conducting copper wires coated with an insulating matrix), thus, a portion of the end of the wires has to be tinplated before sticking. At least 1 h of drying is necessary, but as it depends on the quantity of solvent to evaporate we usually let the silver paint dry for one night.
A. GROWTH AND PATTERNING OF LAO/STO INTERFACES: STEP BY STEP PROCEDURE
The Lagrangian of a free classical electron in a magnetic field reads (e<0):

\[ L = \frac{1}{2} m \dot{\mathbf{r}}^2 + e \mathbf{r} \cdot \mathbf{A} \]  

(B.1)

with \( \mathbf{r} \) the generalized coordinates of the particle and \( \mathbf{A} \) the vector potential defined via \( \mathbf{B} = \frac{\partial}{\partial r} \times \mathbf{A} \). We choose \( \mathbf{A} = (0, Bx, 0) \) leading to a magnetic field of magnitude \( B \) oriented along \( \hat{z} \). From eq. (5.8) we get:

\[ \pi = m^* \dot{\mathbf{r}} + e \mathbf{A} \]  

(B.2)

hence:

\[ m \oint \dot{\mathbf{r}} \cdot d\mathbf{l} + e \oint \mathbf{A} \cdot d\mathbf{l} = 2\pi \hbar (n + \frac{1}{2}) \]  

(B.3)

To get \( \dot{\mathbf{r}} \) we solve the associated Lagrange equations:

\[
\begin{align*}
x(t) &= \sqrt{\frac{v_{0,\perp}^2}{\omega_c^2}} \cos (\omega_c t + \phi) \\
y(t) &= -\sqrt{\frac{v_{0,\perp}^2}{\omega_c^2}} \sin (\omega_c t + \phi) \\
z(t) &= v_{0,z} t
\end{align*}
\]  

(B.4)

with \( v_{0,\perp} \) the in-plane velocity of the particle at \( t = 0 \) (here \( v_{0,\perp}^2 = v_{0,x}^2 + v_{0,y}^2 \)), \( v_{0,z} \) the initial out-of-plane velocity, \( \phi \) a phase shift depending on the initial in-plane velocities and \( \omega_c = \frac{eB}{m^*} \) the cyclotron frequency. Thus, for a particle with a given initial velocity in-plane, the frequency and the radius (\( R = \sqrt{\frac{v_{0,\perp}^2}{\omega_c^2}} \)) of the cyclotron motion are fixed by the magnetic field.

Computing \( \dot{\mathbf{r}} \) from eq. (B.4), expressing it as a function of \( \mathbf{r} \) and substituting in eq. (B.3) leads to:

\[ m^* \oint \begin{pmatrix} \omega_c y \\ -\omega_c x \\ v_{0,z} \end{pmatrix} \cdot d\mathbf{l} + q \oint \mathbf{A} \cdot d\mathbf{l} = 2\pi \hbar (n + \frac{1}{2}) \]  

(B.5)
B. DERIVATION OF THE ONSAGER RELATION FOR A 3DEG

Using Stokes’ theorem for both integrals we find:

\[-2m^*\omega_c S + eBS = -eBS = 2\pi\hbar(n + \frac{1}{2})\] \hspace{1cm} (B.6)

with \(S = \pi R^2\) the surface of the closed orbit in-plane. As a consequence, \(BS\) is the flux (\(\Phi\)) of \(B\) through the real-space orbit of the particle. Hence:

\[\Phi = \frac{2\pi\hbar}{|e|}(n + \frac{1}{2}) = \Phi_0(n + \frac{1}{2})\] \hspace{1cm} (B.7)

where we have introduced the magnetic flux quantum \(\Phi_0 = \frac{2\pi\hbar}{|e|}\). Thus, quantum mechanics restricts the orbits of a free particle in a magnetic field to the one through which \((n + 1/2)\) quantum flux pass.

We have seen in chapter 4, that, in a solid, the electrons that contribute to the electrical conduction are at the Fermi level and thus have the Fermi velocity. Moreover, in the electron gas approximation \(v_{F,\perp} = \frac{\hbar}{m^*}k_{F,\perp}\). Using this formula to compute \(R\) and then \(\Phi\) in eq. (B.7) we get:

\[B\pi\frac{\hbar^2 k_{F,\perp}^2}{e^2B^2} = \frac{\Phi_0^2}{4\pi^2} = \frac{\pi k_{F,\perp}^2}{B} = \Phi_0(n + \frac{1}{2})\] \hspace{1cm} (B.8)

which can be recast in terms of the projection of the \(k\)-space area enclosed by the electron orbit \(A = \pi k_{F,\perp}^2\):

\[\frac{1}{B} = \frac{4\pi^2}{A\Phi_0(n + \frac{1}{2})}\] \hspace{1cm} (B.9)
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Education
2009–2014 PhD, University of Geneva (UNIGE), DPMC
Supervisors : Prof. Jean-Marc Triscone/Dr. Stefano Gariglio.

with internship at the National University of Singapore (NUS).
Master thesis : From the correlations of Dirac particles to the EPR2 approach to non-locality.
Supervisors : Valerio Scarani (NUS)/Marc-André Dupertuis (EPFL).


Teaching Experience
2009–2014 Teaching assistant at Physiscope, UNIGE, Participative physics learning, on a variety of topics (electricity, heat, mechanics, waves...).
Work in focus group to design sessions with new subjects.


Publications
A. Fête, S. Gariglio, C. Berthod, D. Li, D. Stornaiuolo, M. Gabay and J.-M. Triscone
Large Rashba induced modulation of the Shubnikov-de Haas oscillations at the LaAlO$_3$-SrTiO$_3$ interface, submitted

A. Fête, S. Gariglio, A. D. Caviglia, J.-M. Triscone, M. Gabay


C. Cancellieri, N. Reyren, S. Gariglio, A.D. Caviglia, A. Fête, J.-M. Triscone

**Presentations at Conferences and Workshops**

**Workshop on Oxide Electronics 20**, *Singapore (Singapore)*, September 2013.
Interacting Landau levels at the LaAlO$_3$/SrTiO$_3$ Interface

**Swiss Physical Society Annual Meeting**, *ETH (Zurich)*, June 2012.
Magnetotransport properties of LaAlO$_3$/SrTiO$_3$ interfaces

**Materials Research Society Spring Meeting**, *San Francisco (USA)*, April 2012.
Magnetotransport properties of LaAlO$_3$/SrTiO$_3$ interfaces

Magnetotransport properties of LaAlO$_3$/SrTiO$_3$ interfaces

**MAMA workshop**, *Ercolano (Italy)*, October 2011.
Magnetotransport properties of LaAlO$_3$/SrTiO$_3$ interfaces

**Swiss Physical Society Annual Meeting**, *EPFL (Lausanne)*, June 2011.
Quantum oscillation in high-mobility LaAlO$_3$/SrTiO$_3$ interfaces

**OXIDes workshop**, *Olia (Italy)*, May 2011.
Quantum oscillation in high-mobility LaAlO$_3$/SrTiO$_3$ interfaces

**Posters**.
7 poster presentations, including at WOE, SCEF and SPS.
Outreach activities


2012  **FameLab** Qualification to the Swiss Final of FameLab a program whose aim is to find the new voices of science and engineering across the world. For more information: [http://www.famelab.ch](http://www.famelab.ch). Participation to the FameLab master class, a training aiming to develop skills associated with working with media.

2011  **RTS decouverte** Participation in the making of a popular science program for the Swiss television (RTS): [http://www.rts.ch/video/#vid=3160004](http://www.rts.ch/video/#vid=3160004)

2009-2014  **Physiscope** Regular organization of special sessions, including lab visits, for the general public: [http://www.physiscope.ch](http://www.physiscope.ch)

Additional skills

- **Languages**  French (mother tongue), English (fluent), Italian (fluent), German (basic).
- **Programming and software**  C/C++, Fortran 95, Mathematica, Matlab, Comsol, Labview.