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Optical spectra of the heavy fermion uniaxial ferromagnet UGe$_2$

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We report a detailed study of UGe$_2$ single crystals using infrared reflectivity and spectroscopic ellipsometry. The optical conductivity suggests the presence of a low-frequency interband transition and a narrow free-carrier response with strong frequency dependence of the scattering rate and effective mass. We observe sharp increase in the low-frequency mass and reduction in scattering rate below the upper ferromagnetic transition $T_C=53$ K indicating the emergence of a heavy fermion state triggered by the ferromagnetic order. The characteristic changes are exhibited most strongly at an energy scale below 12 meV. They recover their unrenormalized value above $T_C$ and for $\omega>40$ meV. In contrast no sign of an anomaly is seen at the lower transition temperature of unknown nature, $T_s\sim 30$ K, observed in transport and thermodynamic experiments.

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The possibility of unconventional superconductivity mediated by ferromagnetic fluctuations has long been a subject of theoretical speculation. 1,2 Interest in this subject has been recently piqued with the discovery of superconductivity coexisting with the ferromagnetic state of UGe$_2$ under pressure. 3 UGe$_2$ is a strongly anisotropic uniaxial ferromagnet with partially filled 5f electron states. Due to correlations and conduction band-5f hybridization, carrier masses are found to be strongly enhanced 4–5 $(10–25)m_0$, although specific-heat coefficients still fall an order of magnitude short of the largest values found in antiferromagnetic uranium-based heavy fermion (HF) compounds. UGe$_2$ exhibits a Curie temperature that strongly decreases with increasing pressure from about 53 K at ambient pressure to full suppression around 16 kbar. Superconductivity exists in a pressure region from 10 to 16 kbar, just below the complete suppression around 16 kbar. Superconductivity exists in a pressure region from 10 to 16 kbar, just below the complete suppression at ambient pressure 4,5 The optical conductivity suggests the presence of a low-frequency interband transition and a narrow free-carrier response with strong frequency dependence of the scattering rate and effective mass. We observe sharp increase in the low-frequency mass and reduction in scattering rate below the upper ferromagnetic transition $T_C=53$ K indicating the emergence of a heavy fermion state triggered by the ferromagnetic order. The characteristic changes are exhibited most strongly at an energy scale below 12 meV. They recover their unrenormalized value above $T_C$ and for $\omega>40$ meV. In contrast no sign of an anomaly is seen at the lower transition temperature of unknown nature, $T_s\sim 30$ K, observed in transport and thermodynamic experiments.

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properties such as, for instance, the complex conductivity \( \sigma_\omega = \sigma_1 + i\sigma_2 \). The sample used for this Brief Report was grown at the CEA, Grenoble by the Czochralski technique. Measurements were taken in quasinormal incidence to the \( ac \) plane using linearly polarized light. We found only a small shift of the spectra between the two crystal directions with any differences smaller than our experimental accuracy (1%). The displayed spectrum is the average of the two directions.

Figure 1 shows the reflectivity spectra \( R(\omega) \) over the full energy range. At high temperature \( R(\omega) \) exhibits a monotonic increase as expected for a metal with \( R(\omega) \rightarrow 1 \) as \( \omega \rightarrow 0 \). As the sample is cooled below \( T_C \) the reflectivity shows a significant increase with the largest effects at low frequency (see Fig. 1 inset). No sign of an anomaly is found at the temperature \( T_x \), which may be not surprising considering its weak signature in transport and thermodynamics at ambient pressure. It is interesting to mention that URu_2Si_2 also has a magnetic transition of unknown nature at low temperatures, which appears very pronounced in the infrared spectrum. The fact that in UGe_2 we see no signature of the second magnetic transition in the optical spectra, suggesting a very different nature of this transition in the two materials.

We observe that the reflectivity has no significant temperature dependence above 0.12 eV, which suggests a negligible temperature dependence at even higher frequencies. We used therefore room-temperature ellipsometry for frequencies greater than 0.74 eV to calculate the complex optical conductivity over the entire spectral range displayed in Fig. 2(a). We assign the peak at approximately 1 eV to an interband transition. At high temperature the low-frequency optical conductivity \( \sigma_1(\omega) \) shows a broad Drude-type behavior as expected for a metal. The low-frequency conductivity is strongly temperature and frequency dependent. To clarify this trend we show an enlargement of the low-frequency data in Fig. 2(b). Several remarkable low-frequency structures form at low temperatures out of the broad Drude peak, including a very narrow zero-frequency mode which represents the intraband response of the heavy quasiparticles. The presence of a narrow zero-frequency mode is born out by two independent pieces of evidence: First, the imaginary part of

\[ \sigma_\omega \text{ [see inset Fig. 2(b)] rises sharply when temperature is lowered below } T_C \text{. This implies a strong increase in metallic screening originating from the zero-frequency mode below the 6 meV threshold of the reflectivity data. The second piece of evidence is that comparing the dc resistivity at 11 and 290 K, } \sigma_1(\omega=0) \text{ increases by a factor of } 48. \] This ratio is very close to the extrapolated conductivity ratio \( \sigma_1(\omega=0,11 \text{ K})/\sigma_1(\omega=0,290 \text{ K})=50 \). This proves that there is really a narrow zero-frequency mode in the optical spectra at low temperatures. Concomitantly, a maximum develops at 13.6 meV and a weak structure at 37 meV. As will be shown below, these features reflect various aspects of the coherent HF and ferromagnetic states.

In order to further analyze the shape of the low-frequency spectra we use the extended Drude model. In this formalism the optical constants are expressed in terms of a frequency-dependent effective mass \( m^*(\omega)/m \) and scattering rate \( 1/\tau(\omega) \) by the following expression:

\[ \sigma_\omega = \frac{e^2 \tau(\omega)}{m^*(\omega)/m} \]

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where $\hbar \omega_p = \sqrt{4 \pi n e^2/m_0} = 3.5$ eV is the total Drude plasma frequency and $\sigma(\omega)$ is the complex optical conductivity. $\hbar \omega_p$, which is determined through $m^*(\omega)/m = 1$ at 290 K and $\omega = 62$ meV, acts as a normalization constant and does not affect the trends as a function of $\omega$ and $T$.

Figure 3 displays the spectra of $1/\tau(\omega)$ and $m^*(\omega)/m$ as a function of frequency obtained from the extended Drude model for different temperatures. At room temperature both the scattering rate and the effective mass are nearly frequency independent. As the sample is cooled and the magnetic order develops the effective mass is strongly enhanced and the scattering rate suppressed. Such behavior suggests the development of heavy quasiparticles at low temperature.

We observe that the strong increase in the effective mass and the rapid decrease in the scattering rate are largest below approximately 12 meV which can be regarded as the characteristic energy of the heavy quasiparticles. Below 6 meV we use the extrapolation toward zero frequency which gives a quasiparticle effective mass of over 25 for $\omega \rightarrow 0$ at the lowest $T$. Qualitatively this value is consistent with the cyclotron masses of $(10-25)m_0$ obtained by de Haas-van Alphen measurements.

In Fig. 4 we present the temperature dependence of the effective mass and the scattering rate at 0 and 8 meV obtained from Fig. 3. Note that at high temperature, in the paramagnetic state, both quantities are temperature independent. However, starting exactly at $T_C$, the scattering rate becomes strongly suppressed and the effective mass enhanced. Such behavior contrasts with the usual situation in HF compounds, where mass renormalizations develop below a coherence temperature $T'$ different than the transition temperature to a magnetic state. Moreover, the conventional view is that the coherent HF state should actually be suppressed at a magnetic transition as the dominance of the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction is expected to quench the Kondo effect. The observed behavior is not completely unprecedented; however, a number of other multi-$f$ electron compounds have been found to undergo additional mass enhancements at the magnetic transition. In the present case however the magnetic transition appears to actually trigger the HF state suggesting an intrinsic $T' < T_C$. This behavior may be related to multiple occupation of the $5f$ levels although we note that a similar effect in 3$d$ electron systems has also been observed at the $T_C$ of ferromagnetic Yb$_2$Mn$_2$Sb$_3$ (Ref. 18) and helimagnetic FeGe.

The relatively large mass enhancement observed in UGe$_2$ suggests the evolution of renormalized itinerant charge carriers out of a Fermi gas coupled to a lattice of $5f$ local orbitals. It is generally accepted that the periodic Anderson model (PAM), which describes the hybridization of a localized level with a conduction band, captures the essential physics of such systems. In Fig. 5 we plot a schematic band dispersion of the PAM together with its optical conductivity at low and high temperature. At high $T$ only a dispersive conduction band crosses the Fermi level ($E_F$), whereas the (partially filled) $5f$ bands at $E_F$ are dispersionless. The intraband transitions give rise to a broad Drude conductivity sketched in Fig. 5(e). The additional scattering suppression which onsets at $T_C$ implies a strong coupling between HF effects and magnetic ones with important implications for superconductivity. Therefore the occurrence of the HF and the ferromagnetic states cannot be explained using only the hybridization model; the effects of ferromagnetism have to be included. As the temperature is lowered, the material enters in a magnetically ordered state, which on one hand suppresses the channel for inelastic spin-flip scattering near $E_F$ and on the other hand converts the $5f$ spin degrees of freedom into a narrow band of heavy charge carriers. This coherent band, partially occupied, and therefore pinned to $E_F$ exhibits avoided crossings with the wide conduction bands due to hybridization. Excitations between the split bands create the possibility for new interband transitions [Fig. 5(b)] and redistribute spectral weight between high and low energies as shown in Fig. 5(d).

Although this general picture should hold in UGe$_2$, additional features in principle are expected as a result of uranium’s nominal $5f^5$ configuration which results in several $5f$ bands being involved in the heavy electron state. Moreover, as has been suggested for CeCo$_{1-x}$Ir$_x$In$_5$, a distribution of hybridization gaps due to a momentum dependence of the $f$-$d$ coupling parameter may result in multiple features in $\sigma$ as seen in Fig. 2(b) at 37 and 13.6 meV.

The observed suppression in the scattering rate is addi-
tional to that expected generically for HF compounds below their coherence temperatures. The usual expectation is that the effective mass \( m^* \) and the quasiparticle lifetime \( \tau \) are renormalized by approximately the same factor.\textsuperscript{22} In contrast, comparing the low-temperature scattering rates and masses with their high-temperature unrenormalized values we find the ratios \( \frac{m^*}{m} \sim 6 \) and \( \frac{\tau}{\tau_0} \sim 50 \), which disagree by a factor of 8. A similar analysis using instead the high-frequency values gives a similar discrepancy. It is also interesting to note that as the temperature increases the energy scale of the threshold in \( 1/\tau(\omega) \) does not appear to close at the transition, but instead the gap “fills in” and a remnant of this suppression persists even up to 290 K. Similar effects have been observed in ferromagnetic nickel.\textsuperscript{26} This observation of gaps which fill in instead of closing is a common occurrence in strongly correlated systems.\textsuperscript{27}

In the optical spectra shown in Fig. 3 we observe a rather strong but incomplete suppression of \( 1/\tau(\omega) \) for frequencies smaller than about 6 meV. The suppression of the scattering rate, which onsets at \( T_C \) is reminiscent of that which occurs at energies below the Stoner gap in fully spin-polarized ferromagnets such as \( \text{CrO}_2 \).\textsuperscript{28} In such cases longitudinal Stoner-type spin-flip scattering is forbidden at energies below a threshold set by the energy difference from the bottom of the minority band to the Fermi level (the Stoner gap). \( \text{UGe}_2 \) is not fully spin polarized but has only a small minority spin population at \( E_F \).\textsuperscript{29,30} Moreover, the behavior of properties such as the pressure-dependent magnetization and mechanisms of the pairing mechanism\textsuperscript{9} have been interpreted as a consequence of narrow peaks in the density of states, which could give an effective gap to spin-flip excitations. Such a density of states is supported by band calculations.\textsuperscript{10} Longitudinal fluctuations, which can possibly mediate exotic superconductivity, have been found by neutron scattering.\textsuperscript{31} We note that conventional magnons are not expected to play a large role in a strongly uniaxial compound such as \( \text{UGe}_2 \) as their energy scales will be much higher. An effective gap to longitudinal spin-flip excitations has also been inferred through a Stoner model fit to the strength of magnetic Bragg peaks at low temperature with a gap that is on the order of the threshold in the optical scattering rate.\textsuperscript{32} It is interesting that our spectra give a strong indication of a coupling of charge to these longitudinal fluctuations that were originally proposed as a possibility to mediate superconductivity in ferromagnetic compounds.

Our observations suggest an interesting interplay between spin polarization and the HF coherent state. We believe that this is at the origin of the rather rich behavior of the optical conductivity resulting in two structures appearing below \( T_C \) (13.6 and 37 meV) and large quasiparticle renormalizations. The optical data indicate that the magnetic order triggers the transition into a state characterized by heavy and weakly scattered charge carriers. Our data are consistent with a strong coupling to the longitudinal magnetic modes which have been suggested to mediate superconductivity in this compound.

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15 Due to the irregular sample geometry the absolute value for the dc resistivity has not been obtained. Therefore the \( \rho_{dc} \) values are not included in the fitting procedure.