Positron Annihilation: Industrial Applications Develop

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Abstract

Positrons are often associated with high-energy physics and accelerators, and hence are thought to occupy the realm of basic science. Less well known is the important role they play in materials science after it was discovered in the second half of the 1960s that the results of positron annihilation experiments in solids are strongly dependent on the history of the sample. Following some 25 years of research and development in the laboratory, positron annihilation is now finding its way into industry. This was illustrated by the first Europhysics Industrial Workshop (EIW) on Industrial Applications of Positron Annihilation (10-12 March 1994), where contributions could roughly be divided into three categories: defects in metals, defects in semiconductor materials and devices, and industrial applications of positron tomography.

Reference


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Two Principal Techniques

There are, grosso modo, two different modes in which positrons are used to provide information on defects in materials. First of all, upon analysis, the lifetime spectrum of positrons in a material may be shown to contain several lifetime components, reflecting the various channels via which positrons decay. For example, positrons may annihilate from the bulk through the formation of positronium (Ps) atoms either in a void or at an interface after being trapped in various types of defects. While the sample contains open-volume defects such as vacancies or voids, the positron lifetime spectrum shows a clearly resolved long-lived component. This reflects the average density of electrons "seen" by the positron since it is trapped in a defect. In general, the lifetime of a positron in a defect depends on the size of the open volume of the defect.

Similarly, the momentum density of a positron-electron pair trapped in a defect gives a relatively large component at low momenta, in agreement with the fact that a vacancy implies the absence of an atom with its bound electrons. This difference can be seen by measuring the Doppler broadening of the 511 keV annihilation line and observing the variations of the fractions $S$ and $W$ of positrons annihilating with low and high momentum (mainly core) electrons, respectively. When compared with the results of measurements in a carefully annealed sample, the relative lack of high electronic momenta in a defect is reflected in a smaller amount of Doppler broadening of the 511 keV line, and hence a higher value of the $S$ parameter and a lower value of the $W$ parameter.

The $S$ and $W$ parameters can be used in depth-profiling measurements in which the energy $E$ of a monochromatic beam of slow positrons is varied between say 100 eV and 50 keV and the $S$ or $W$ parameter is measured as a function of $E$. Since the positron energy determines the depth at which the positrons are implanted in the material, this provides a method of measuring defect type and concentration in a depth-dependent way (see insert).

The sensitivity of the method is high: monovacancy concentrations of $10^{-6}$ per atom can be measured at concentrations greater than $10^{-5}$ per atom, the trapping saturates.

Precision Depth Profiling

When a monoenergetic beam of slow positrons strikes a sample, the positrons penetrate into the material and reach thermal equilibrium with the lattice in a period which is two orders of magnitude shorter than the positron lifetime. The implantation takes place according to a profile which peaks at a depth which depends on the initial energy of the positrons. After thermalization, the positrons diffuse through the material over a distance given by the diffusion length, which depends on the perfection of the material. In perfect crystalline Si, the diffusion length is $\approx 2500 \AA$ while in Si with a high concentration of defects it can be as small as $20 \AA$. In this way it is possible to deposit the positrons approximately at a pre-selected depth which increases with the positron energy.

The figure shows the $S$ parameter (the fraction of positrons annihilating with low momentum electrons) as a function of positron energy for a layer of hydrogenated amorphous silicon grown on a silicon substrate. The high values of the parameter at low positron energies indicate a large concentration of voids in the deposited layer. The void concentration decreases with increasing deposition temperature. As the positron energy increases, a larger fraction of the positrons diffuses into the substrate and the $S$ parameter tends to the value for the crystalline substrate.

Position-annihilation depth-profiling measurements of a 1 µm thick hydrogenated amorphous silicon layer for solar-cell applications grown on a crystalline silicon substrate at different temperatures using a plasma-enhanced chemical vapour deposition method. a, upper) Experimental data and the results of modelling calculations (lines). b, lower) Schematic illustration showing the distribution of voids in a cross-section through the silicon layer.
material affect the electrical properties of the device, as is shown by the different electrical characteristics of Schottky diodes made from material which has been found using positrons to be defect-free or not. Defects in the metallic lines on the semiconducting substrate are also important since they can coalesce into larger clusters and even voids following heating under operating conditions. A series of voids may then lead to rupture of the line, resulting in device failure.

Another region where defects are formed is at the SiO₂/Al(Cu) interface. These defects, which result in contact peeling, cannot be observed by transmission electron microscopy, but are easily detected with the aid of positron depth-profiling. Electromigration probably plays a role, since the results depend on the time that current has been passed through the device.

In compound semiconductor materials, there are new types of defects in addition to the ones encountered in homogeneous semiconductors (they include vacancies on the different sub-lattices and atoms sitting on the wrong sublattice). Several results concerning technologically significant GaAs and GaAlAs materials were reported at the EIW in invited talks by P. Hautojarvi (Helsinki University of Technology) and M.R. Brozel (Manchester Institute of Science and Technology). Positron annihilation is particularly successful in identifying defects such times in good agreement with experimental values [5].

More precise information on the behaviour of electrons in the vicinity of an empty lattice site requires two-dimensional ACAR measurements. This method [6] provides maps of the electron momentum distribution, as seen by the positrons. Initially applied to investigate Fermi surface features in metals and superconductors, it has been used [2] to measure the electron-positron momentum distribution in bulk GaAs and at various kinds of vacancies and antisites.

For the bulk and the singly negative As vacancy, there is a good agreement between the ACAR profiles and distributions calculated using the CP approach (see the right hand-side of the figure). One notes that the shapes of ACAR distributions are quite different for the bulk and the vacancy, outlining the sensitivity of ACAR measurements and the precision of the CP-calculations. The results are very encouraging as they open a new way to investigate and characterize defects in many semiconductors.

Using samples carefully characterized by positron lifetime and Doppler broadening measurements, ACAR maps have been obtained for antisites and the As and Ga vacancies in various negatively and neutrally charged states. The measurements show that each type of structural defect in GaAs has a characteristic ACAR distribution which depends on the charge state of the defect. This information could possibly be used in the calculations to design a tool capable of identifying and quantifying defects in industrial materials. The method looks promising for GaAs and could be extended to other elemental and compound semiconductors. In appreciating the opportunities, several groups worldwide have started to work along these lines; the EIW workshop offered an unique occasion to present some of the new developments.

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as negative vacancies associated with the metastable state of the native mid-gap donor, the so-called EL2 centres, which control compensation mechanisms in semi-insulating GaAs (see figure). The technique is able to show that the open volume of these vacancies is smaller than that of Ga or As vacancies, strongly supporting recent models which propose a [111]-oriented vacancy-interstitial complex for the structures of these centres. Furthermore, As vacancies seem to play an important role as recombination (RC) centres in GaAs since positron annihilation has demonstrated that their concentration is correlated with that of RC centres. The latter are detected by infra-red absorption and have been shown to control the minority carrier lifetime in bulk semi-insulating GaAs.

Proof that the characterisation of defects in semiconductors by positron annihilation is becoming more firmly rooted also stems from interesting studies by a group based in Geneva and Lausanne of the negatively charged As vacancy in doped GaAs. By combining Car/Parrinello-type ab initio electronic structure/molecular dynamics calculations with experimental measurement of the momentum density of electron-positron pairs by observing the two-dimensional angular correlation between the 511 keV annihilation quanta (the so-called two-dimensional ACAR technique), it can be established that the atoms around the vacancy relax in an outward direction when a positron is trapped in the vacancy (see insert). This type of study is important for understanding the behaviour of positrons in defects. A simple but powerful extension of the Doppler broadening method has been shown to allow one to distinguish between various kinds of defects. It is actually a revival of an old idea [Mentl S. & W. Triftshäuser W., Phys. Rev. B 17 (1978) 1845] and consists of simultaneously examining the variations of the S and W parameters. It has been shown for CdTe and that the data points representing samples with different concentrations of a specific type of defect (e.g., monovacancies) fall on a straight line.

Other Materials, Large Scales

Reports at the workshop about depth profiling in diamond-like coatings, magnetic multilayers and layers of paint show that the application of positron annihilation is not limited to metals and semiconductors. Nor are positron methods restricted to the microscopic realm, as is witnessed by work on positron emission tomography at Shell that was reported by G. Jonkers. With the aid of gamma-ray cameras and positron emitting tracers it is possible to study processes as diverse as the flooding of oil-filled reservoir rock with water, the movement of particles in a rotating drum and the oil flow in engines while they are running. The value of the use of positron annihilation is the feasibility of safe in situ imaging of complicated processes without disturbing them.

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POSITRON FACILITIES

Facilities-based Capabilities Increase

W. Triftshäuser from the Institut für Nukleare Festkörperphysik, Universität der Bundeswehr, Munich, described at the Europhysics Industrial Workshop 'Industrial Applications of Positron Annihilation' Europe's move towards larger and more sophisticated facilities for positron annihilation.

Continuous Beams

The application of "conventional" low-energy continuous positron beams has increased significantly these last few years owing to the development of more efficient moderators to give beams with a narrower distribution of energy. Positrons from primary radioactive sources are passed into a moderator acting as a secondary source of low-energy positrons and the low- and high-energy positrons are then separated electrically and/or magnetically to the target. However, the intensity of radiosotope source-based beams are limited by self-absorption. One approach to increase the intensity is based on pair production from Bremsstrahlung in a linac, but heating and cooling of the target foil used to produce the positrons impose serious limitations. Another disadvantage is that most linacs are pulsed so the positron beam is also pulsed. A group at the University of Gent uses a specially designed Penning trap to generate a semi-continuous beam of some 4 x 10^13 positrons/s by smearing out pairs of pulses.

The participants at the first Europhysics Industrial Workshop (EIW) on Industrial Applications of Positron Annihilation (10-12 March 1994) which was held in a secluded hotel near the picturesque village of Oisterwijk in the south of The Netherlands. EIW's aim to bring together scientists from universities and from industry in order to promote the application of new physical methods to industrial problems. The workshop, the 12th in the series of EIW's and chaired by A. van Veen (IRI, Delft) and C. Corbel (Saclay), attracted some 40 scientists from both sectors. The proceedings of the workshop will be published as a supplement to the Journal de Physique.