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LETTER TO THE EDITOR

Positron states at point defects in GaAs measured by 2D-ACAR

R Ambigapathy †, C Corbel ‡, P Hautojärvi §, A A Manuel† and K Saarinen§

† Département de Physique de la Matière Condensée, Université de Genève, 24 quai Ernest Ansermet, CH-1211 Genève 4, Switzerland

‡ Centre d'Etudes Nucléaires de Saclay, Institut National des Sciences et Techniques Nucléaires, F-91191 Gif-sur-Yvette Cédex, France

§ Laboratory of Physics, Helsinki University of Technology, SF-02150 Espoo, Finland

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Abstract. The electron–positron momentum distribution in the Ga vacancy (V_{Ga}^{3-}) and Ga antisite (Ga_{As}^{2-}) have been obtained by performing 2D-ACAR measurements on electron-irradiated semi-insulating GaAs. These results are compared to our previous results for the As vacancy. All the vacancy distributions are structureless, though their anisotropies indicate that there is a residual bulk-like component. The magnitude of this component varies from vacancy to vacancy and indicates how much the localized positron wave function extends in the crystal bulk around the vacancy. Our results show that this extension of the wave function in V_{Ga}^{3-} and V_{As}^{-} is similar to and clearly greater than that in V_{As}^{0}. The momentum distribution sampled by the positron in a Rydberg state about Ga_{As}^{2-}, though very similar to that of the bulk, can nevertheless be distinguished from the latter.

It is well known that defects play a significant role in a number of properties of the solid state. In the case of semiconductors, their presence can drastically modify the optical and electrical properties and hence have an influence on the proper functioning of electronic devices. It is thus important to characterize these defects in a complete manner [1]. Positrons have proven to be a useful probe for this task in both metals and semiconductors [2]. So far, the most commonly used techniques for studying defects with positrons have been positron lifetime and Doppler broadening of the annihilation radiation methods. For a review of results obtained for GaAs see [3]. Recently, the two-dimensional angular correlation of positron annihilation radiation (2D-ACAR) set-up has been applied for studying the electronic structure of point defects in semiconductors [4, 5, 6, 7, 8]. These studies have offered some very interesting results and provide a new perspective on the field.

In the case of a compound semiconductor like GaAs a number of different types of defect and associated complex may be present. These defects and their known properties have been discussed in [9]. In a recent study [4] we probed native monovacancies in Si-doped GaAs using 2D-ACAR. Using trapping fractions obtained from the results of positron lifetime experiments [10] the electron–positron momentum distributions for the As vacancy in its neutral (V_{As}^{0}) and negative (V_{As}^{-}) states were extracted from the measurements. The results obtained confirmed the findings of molecular dynamics calculations [11]; these were that (1) the electronic structure of the As vacancy is very much dependent on its ionization state and (2) V_{As}^{-} is characterized by a smaller open volume than V_{As}^{0}. More recently, another *ab initio* calculation was performed for bulk GaAs and V_{As}^{-} [12], in which the vacancy was allowed to relax in the presence of the positron. The results of this calculation have been compared to our experimental 2D-ACAR spectra and there is a good qualitative agreement.

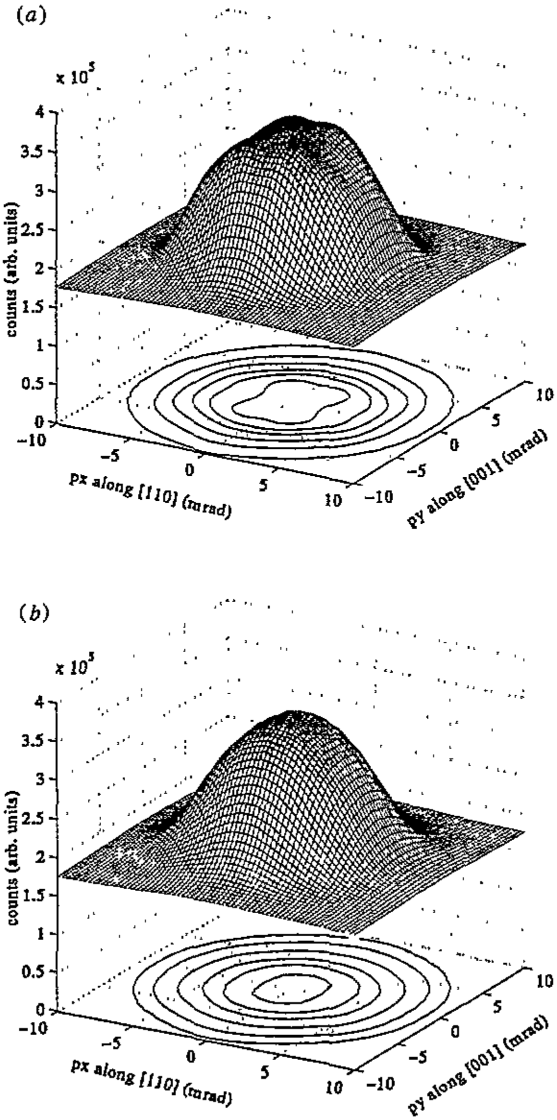

Figure 1. Combined perspective and contour plots of the 2D-ACAR of the Rydberg state around the Ga_{As}^{2-} (top) and V_{Ga}^{3-} (bottom). Each spectrum has been normalized to the positron lifetime *for that state. The vertical scale is the same in all three cases, and all the plots were offset* vertically by the same amount so as to not hide the contour plot below.

In this letter we present the electron–positron momentum density for the electron-irradiation-induced Ga vacancy (V_{Ga}^{3-}) and gallium antisite (Ga_{As}^{2-}). These measurements are discussed and compared to the above-mentioned results for the As vacancy and bulk GaAs.

The experiments were performed using a standard 2D-ACAR spectrometer described in [13]. In such an experiment the two 511 keV gamma rays that are emitted upon the annihilation of the positron in a sample are measured in coincidence using two position-sensitive detectors. The resulting distribution $N(p_x, p_y)$ is a projection of the electron–positron momentum density $\rho^{2\gamma}(p)$. Consequently, $N(p_x, p_y)$ contains information about the electronic structure as seen by the positron. When the positron is trapped in a vacancy and its wave function is localized, $N(p_x, p_y)$ will contain information about the electronic structure of the vacancy. However, in a given sample with defects, only a fraction of the positrons will annihilate at a defect; the rest will annihilate delocalized in the bulk. In consequence all of our measured spectra contain several components and the separation of these components will be discussed below.

The sample studied was an undoped semi-insulating (SI) wafer of GaAs grown using the liquid-encapsulated Czochralski method. Prior to any measurements it was irradiated at 20 K with 1.5 MeV electrons with a fluence of 5×10^{17} cm^{-2} and subsequently aged at room temperature. Lifetime measurements at 300 K only found one type of vacancy with an associated positron lifetime of 260 ps and it was identified as the gallium vacancy [14]. The data also showed that at 90 K this defect is in competition for trapping positrons with a negative ion that acts as a shallow trap which localizes the positron in a Rydberg state. This shallow trap was subsequently inferred to be the Ga antisite Ga_{As}^{2-} [14].

The details of the experimental set-up are identical to those described in our study of the As vacancy [4]. Two spectra were acquired, one at 90 K and the other at 300 K; both of these experiments were done in a magnetic field of 4 T. The sample was aligned to within 1° with the [110] direction along the projection axis. The activity of the ^{22}Na positron source was about 35 mCi. Both the spectra contain approximately 1.5×10^8 coincidence events and the typical acquisition time of a spectrum was about four days. Finally, all the raw data sets were treated in a similar manner to that described in [4].

In the As vacancy study [4] we also showed how it is possible to extract the trapping fractions for a sample that has been well characterized by positron lifetime measurements. In the present case, for the electron-irradiated sample, using results from [14], we find that $N_{e^-irr}^{300}(p_x, p_y)$, the 2D-ACAR spectrum at 300 K, can be decomposed in the following manner:

$$N_{e^-irr}^{300}(p_x, p_y) = 0.67 N_b^{300}(p_x, p_y) + 0.33 N_{V_{Ga}^{3-}}^{300}(p_x, p_y) \quad (1)$$

where $N_b^{300}(p_x, p_y)$ and $N_{V_{Ga}^{3-}}^{300}(p_x, p_y)$ are the 2D-ACAR distributions of the bulk and the gallium vacancy at 300 K. Since $N_b^{300}(p_x, p_y)$ had been previously measured we were then able to extract the distribution for V_{Ga}^{3-} using equation (1). Similarly at 90 K the measured spectrum $N_{e^-irr}^{90}(p_x, p_y)$ can be decomposed as:

$$N_{e^-irr}^{90}(p_x, p_y) = 0.06 N_b^{90}(p_x, p_y) + 0.10 N_{V_{Ga}^{3-}}^{90}(p_x, p_y) + 0.84 N_{RS}^{90}(p_x, p_y) \quad (2)$$

where $N_b^{90}(p_x, p_y)$, $N_{V_{Ga}^{3-}}^{90}(p_x, p_y)$ and $N_{RS}^{90}(p_x, p_y)$ are respectively the 2D-ACAR distributions of the bulk, the gallium vacancy and the Rydberg state around the Ga antisite at 90 K. The uncertainty of the fractions in equation (1) and 2 is estimated to be 0.02. $N_{RS}^{90}(p_x, p_y)$ can be extracted from equation (2) by using our previous 90 K bulk measurement and by making the substitution $N_{V_{Ga}^{3-}}^{90}(p_x, p_y) = N_{V_{Ga}^{3-}}^{300}(p)$. This latter substitution is in fact an approximation and can be justified in the following manner. We had noticed that for the bulk the 2D-ACAR changes very little from 300 K to 90 K. The small difference can be accounted for by thermal expansion of the lattice. In consequence, it can be argued that similarly the 2D-ACAR of V_{Ga}^{3-} must not vary in a significant manner from 300 K to 90 K.

The 2D-ACAR distributions extracted for Ga_{As}^{2-} and V_{Ga}^{3-} are shown in figure 1. The first thing one sees when looking at the figure is that the 2D-ACAR distribution for the Rydberg state (figure 1, top panel) is very similar to that of the bulk. This is not surprising though, as the lifetime of the positron in a Rydberg state around Ga_{As}^{2-} is the same as that of the positron in the bulk at 90 K, namely 230 ps [14, 15]. Nevertheless, small differences are observed (see figure 4 of [5]). They can be characterized for example by the shape parameter S which, in the [110] direction, is equal to 0.479 and 0.485 for the bulk and Ga_{As}^{2-}. The Ga vacancy distribution (figure 1, bottom panel) on the other hand is much more isotropic and has few apparent features. This is in agreement with what we previously found for the As vacancy which is also characterized by an isotropic electron–positron momentum distribution. In our previous study we also discovered that even though the As vacancy distributions seem fairly isotropic and structureless when compared to the bulk, there still remain some anisotropies with structures that are bulk-like. This is an indication that the wave function of the localized positrons extends beyond the vacancy and samples the bulk. In the case of V_{Ga}^{3-}, once again, there are anisotropies in the momentum distribution that have the same symmetry as the bulk.

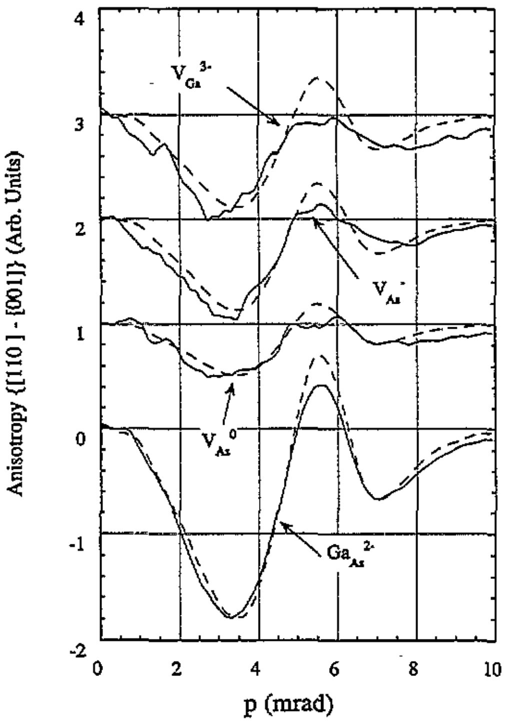

Figure 2. The solid lines show the difference between the 2D-ACAR in the [110] and [001] directions for the various defects: Ga_{As}^{2-}, V_{As}^{0}, V_{As}^{-}, and V_{Ga}^{3-}. For clarity, the zero levels of the three vacancy curves have been shifted. The dashed lines show the same difference for the bulk, scaled to fit the solid line (see the text for more details).

In order to visualize these anisotropies in a simple way, we define them in the following manner: we take a 2D-ACAR line along the [110] direction and subtract from it another 2D-ACAR line with the same momentum along the [001] direction. The results of this operation for various defects are shown in figure 2. In each case we have also plotted the best fit of the bulk 90 K anisotropy to the defect anisotropy. The fitted parameter is a scaling factor, applied to the [110]–[001] difference for the bulk. When looking at the plot for Ga_{As}^{2-} (figure 2) we notice that the fit is very good except around the peak at 5 mrad. This strongly suggests that the positron has been trapped in a shallow trap because, as is the case for the other defects shown in this figure, the amplitude of the peak is smaller than it is for the bulk. This difference is significant despite the slight uncertainty of the trapping fractions (0.02) and the approximation $N^{90}_{V_{Ga}^{3-}}(p_x, p_y) = N^{300}_{V_{Ga}^{3-}}(p_x, p_y)$. This leads to an interesting point that is worth highlighting. Using lifetime measurements we are not able to directly distinguish between the Rydberg state around Ga_{As}^{2-} and the bulk at 90 K. However, we have just shown that we clearly see a difference and this indicates the power of discrimination that the 2D-ACAR set-up possesses. For the vacancies we can also use the difference curve to get an indication on the degree to which the positron extends into the bulk by looking at the scaling factor that gives the best fit of the bulk 90 K anisotropy to the vacancy anisotropy. The values we get for our data are 0.27, 0.48, 0.49 and 0.99 for V_{As}^{0}, V_{As}^{-}, V_{Ga}^{3-} and Ga_{As}^{2-} respectively. This difference can be seen also in figure 2 by looking at the local minima of the curves at around 3 mrad. Thus, the bulk-like character of the previously measured V_{As}^{0} is less than that of both V_{As}^{-} and V_{Ga}^{3-} indicating that the positron is less localized in V_{As}^{-} and V_{Ga}^{3-} than in V_{As}^{0}.

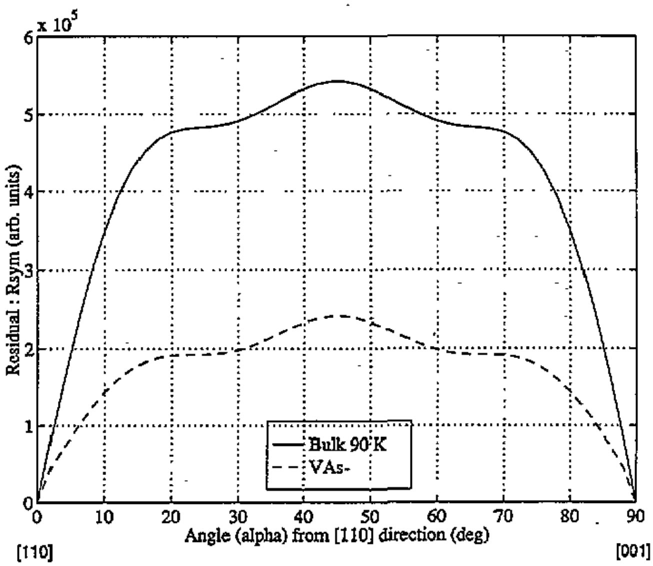

Figure 3. A plot of residuals (see the text for the definition) of the bulk and V_{As}^{-}. Note how the spectra are characterized by curves with the same topology, thus indicating that we see no lowering of symmetry in the vacancy 2D-ACAR.

In their theoretical calculation Gilgien *et al* [12] noticed that in real space V_{As}^{-} undergoes a Jahn–Teller distortion and this results in the lowering of the symmetry compared to that of the bulk. We thought it would be interesting to see if we could observe any similar reduction of symmetry in momentum space. To do so we took a line of symmetry starting along the [110] direction (p_x) and rotated it through 90° to the [001] direction (p_y) in steps of 1°. At each angle we determined the extent of mirror symmetry present by calculating the following parameter R which we term the 'residual':

$$R = \sqrt{\sum_{p}(N(p) - N(p'))^2} \qquad (3)$$

where the index p of the sum only runs through one of the halves of the mirror line and p' is the equivalent point of p on the other side of the symmetry line. The result of this operation is depicted in figure 3. Let us mention that it is unchanged when estimated from the raw (unsymmetrized) data. In this figure we have plotted the residual for the bulk and V_{As}^- as a function of angle of rotation of the symmetry line. One notices in both cases that the only symmetry lines that exist are along the [110] direction (0° rotation) and [001] (90° rotation) and that the topologies of the two curves are quite similar. This similarity in the topology indicates once again that the momentum distribution of V_{As}^- has to a certain extent a bulk-like nature. We, however, see no reduction in the symmetry of the vacancy 2D-ACAR distribution. This is not surprising as even if the vacancies undergo a distortion, they will not all be aligned in a preferred direction. As a consequence in our experiment, with 10^8 counts, vacancies aligned in all the possible directions will be equivalently sampled and the reduction of symmetry will not be apparent in our spectra.

In conclusion, a new set of 2D-ACAR measurements have been performed on a SI sample of GaAs that had been irradiated with 1.5 MeV electrons. Using these measurements and fruitfully combining results from lifetime measurements we have been able to extract the electron–positron momentum distribution of the electron-irradiation-induced Ga antisite and the Ga vacancy in the (110) plane. The results were compared to previously obtained results for bulk GaAs and the As vacancy in its negative and neutral ionization states. We notice that there is a close resemblance between the momentum distribution of the Rydberg state of the positron around Ga_{As}^{2-} and that of the bulk. A closer look at the anisotropies, however, reveals that we can indeed distinguish between them. This clearly indicates that 2D-ACAR measurements have a resolution and sensitivity that is not present in lifetime measurements. The anisotropies also revealed that when trapped in V_{As}^0 the positron is much more confined in the vacancy than it is when it is trapped in V_{As}^- or V_{Ga}^{3-}.

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