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Positron 2D-ACAR Study of Divacancies in Si: Experiments and Theory

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Abstract. Two-dimensional angular correlation of annihilation (2D-ACAR) for divacancies in Si with definite charge states (V_2^0, V_2^{1-} and V_2^{2-}) are studied theoretically and experimentally. The 2D-ACAR for the various charge states shows almost isotropic and very close to each other. However, using a specimen with aligned divacancies (V_2^{1-}), we have obtained a small but definite anisotropy in the 2D-ACAR. All these features are well reproduced by first principles calculations based on the two-component density-functional theory. Furthermore calculated anisotropies for the different charge states show systematic changes arising from the bonding character of the divacancy electrons. Theoretical 2D-ACAR is also presented for monovacancies. The present calculation and experiment demonstrate that the 2D-ACAR is an effective tool to provide microscopic information about vacancy-type defects.

The positron annihilation technique has recently emerged as a powerful tool for the studies of vacancy-type defects in semiconductors [1]. Especially the two-dimensional angular correlation of annihilation radiation (2D-ACAR) technique gives more detailed microscopic information about defects, compared with positron lifetime or Doppler broadening spectrum [2-10]. In the 2D-ACAR studies of defects, it is very important to carry out the experiments on samples containing well-defined defects. Furthermore it is indispensable to make first principles-calculation on the defect component to get microscopic information on the defects. In this study our target is divacancy in Si, which is one of the fundamental stable defects existing at room temperature in Si and is crucially important in the current Si technology.

The preparation method for the samples containing aligned divacancies with definite charge states has been reported in our previous papers [7-10]. The experimental procedures for 2D-ACAR measurements and for obtaining the spectra for the divacancy components are also described in them.

We employed the two-component density functional (TCDF) theory within the local density approximation (LDA) [5,6,10] in the framework of pseudopotential scheme. Detailed calculational procedures are described in the previous paper [10] and these proceedings [11,12].

Before presenting the results for divacancies, we show the calculated 2D-ACAR $N(p_x,p_y)$ of [100] projection for bulk and neutral (mono)vacancies and their anisotropies in Fig.1, to compare with those for divacancies. The 2D-ACAR for the bulk shows characteristic large anisotropy due to the band structure of Si [7,11,12]. This agrees very well with the experiment as demonstrated in [11,12]. We extract the anisotropy $A(p_x, p_y)$ from the 2D-ACAR as $A(p_x,p_y) = N(p_x,p_y) - C(p_x, p_y)$, where $C(p_x,p_y)$ is a smooth cylindrical average of $N(p_x,p_y)$ around the projection direction (p_z). The anisotropy of Fig. 1(c) exhibits large anisotropy amplitude (valley-to-peak altitude), 17% of the peak height of the 2D-ACAR, and shows the peaks and valleys with the four-fold symmetry. This anisotropy is also in good agreement with the experimental one.

The theoretical 2D-ACAR and its anisotropy for the vacancies are shown in Fig. 1(b) and (d). The 2D-ACAR is practically isotropic and hence exhibits very small anisotropy. The anisotropy shows small peaks and valleys with four-fold symmetry, as expected from the symmetry of vacancies. The four-fold symmetry is also observed in that for the bulk (Fig.1(c)). However, it is noted that the peaks and valleys for the vacancies are broader and shifted to the higher momentum, compared with those for the bulk; for example the four-fold peaks are located at about 8 mrad along [011] and [01-1], while those for the bulk at about 5 mrad. Then it is very interesting to measure 2D-ACAR of the vacancies. Unfortunately, however, the vacancies in Si can not survive at room temperature and exist only as divacancies or vacancy-impurity complexes , because the vacancies can migrate well below room temperature[12]. Then the 2D-ACAR experiments on the vacancies is not easy to be made and have not been done yet.

The 2D-ACAR for divacancies (V_2^{1-}) aligned along [1-11] and [11-1] in the (011) plane are shown in Fig. 2. The

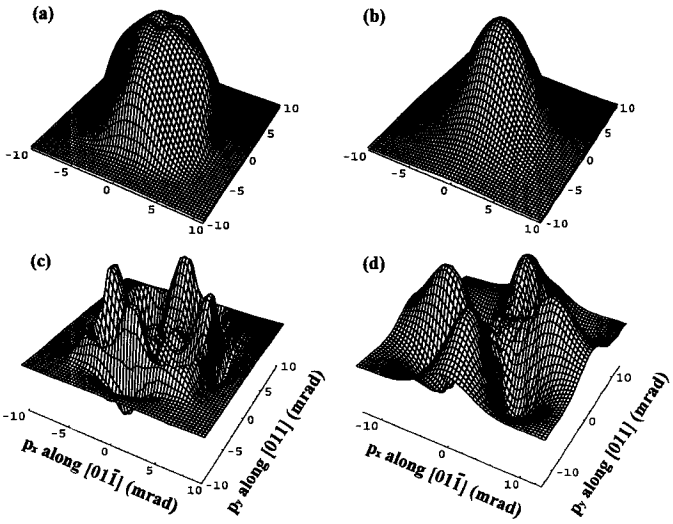

Fig. 1. Theoretical 2D-ACAR of [100] projection for the bulk (a), monovacancies (V_o) (b), and their anisotropies (c), (d). The anisotropy amplitude are 17% for the bulk and 2.0% for the monovacancies, relative to the 2D-ACAR peak height. In this figure the anisotropy is normalized to the same amplitude.

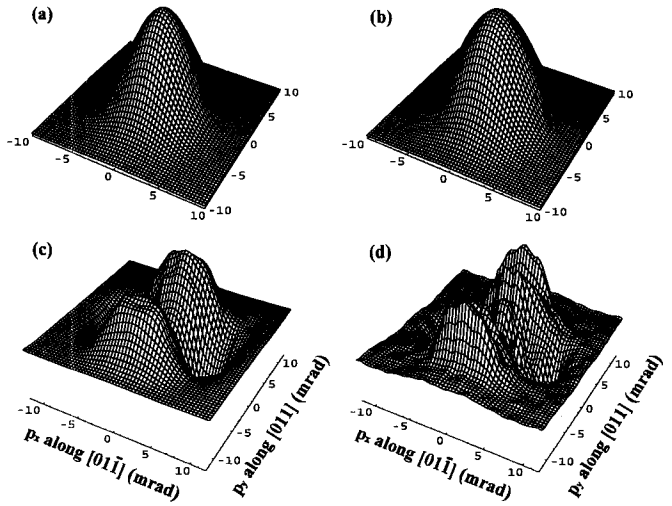

Fig. 2. The 2D-ACAR for aligned divacancy (V_2^{1-}) and their anisotropies: (a) theory , (b) experiment, and their anisotropies ((c) theory and (d) experiment). The anisotropy amplitude is 4.1% of the peak height of 2D-ACAR for the theoretical spectrum, and 1.8% for the experimental one. The anisotropy for V_2^{1-} of Fig. 2(c) is also shown in Fig. 3(b) in a contour map. The anisotropy is normalized to the same amplitude.

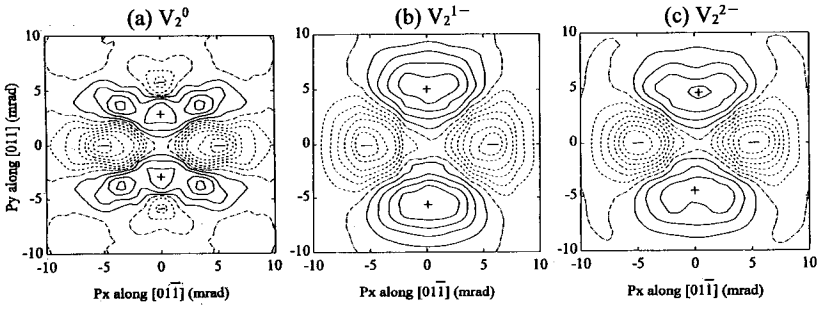

Fig.3. Calculated anisotropies of divacancies with different charge states: (a) V_2^0, (b) V_2^{1-}, and (c) V_2^{2-}. The contour spacings are 1/11 of the anisotropy amplitudes. The solid (dashed) lines indicate positive (negative) values.

2D-ACAR is almost isotropic. However, the 2D-ACAR is slightly broader along [011] than [01-1]. Then small but definite anisotropy is observed in the anisotropies in Fig. 2. Peaks are seen around ± 6 mrad along [011], while valleys around ±5mrad along [01-1] in Fig.2 (c) and (d). The experimental 2D-ACAR is slightly more isotropic than theoretical one. Therefore the experimental anisotropy amplitude is slightly smaller than theoretical one (Fig.2). It should be noted that the experimental anisotropy has two-fold symmetry as expected from the aligned divacancies and that the positions of the peaks and valleys are in good agreement with those by first principles-calculation. The fact that even for the aligned divacancies the 2D-ACAR shows the very small anisotropy is well explained by the electron-positron density overlap distribution around the divacancies [10].

The 2D-ACAR for the aligned divacancies with the other different charge states have been also calculated and found to be almost the same with each other. However, systematic changes are observed for the different charge states, as shown in Fig. 3. The relative anisotropy amplitudes are 2.4% for V_2^0, 4.1% for V_2^{1-}, and 4.8% for V_2^{2-}. The anisotropy for V_2^{1-} is nearly the same as that for V_2^{2-}, but is somewhat different from that for V_2^0. On comparing the contour map shown in Fig.2 for V_2^0 with that for V_2^{1-}, the peaks are shifted to the origin along [011] and are flattened along [01-1], and consequently the valleys has narrowed along [011]. Furthermore small valleys appear around p_y=6 (p_x=0) mrad. These changes are possibly due to the bonding character of electrons around the divacancies in Si. In a simple LCAO theory [14], for V_2^0 the uppermost two electrons occupy the extended-bond orbital, while for V_2^{1-} (V_2^{2-}) the extra electron(s) occupies the next extended-antibonding orbital. Then the different feature in the anisotropy for V_2^0 and V_2^{1-} (V_2^{2-}) reflects the different orbitals occupied by the divacancy electrons. The 2D-ACAR experiments for aligned divacancies of V_2^0 and V_2^{2-} are very necessary.

In summary, 2D-ACAR for divacancies and monovacancy in Si has been studied experimentally and theoretically. The experimental 2D-ACAR and its characteristic anisotropy for aligned divacancies of singly negative charge is well reproduced by the calculation based on the TCDF theory within the LDA. The calculation for the divacancies with the other charge states and for monovacancies suggest that 2D-ACAR spectra is promising for the studies of microscopic structure of defects.

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