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ON THE RESOLVABILITY OF POSITRON DECAY CHANNELS*

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Many data analysis treatments of positron experiments attempt to resolve two or more positron decay or exit channels which may be open simultaneously. Examples of the need to employ such treatments of the experimental results can be found in the resolution of the constituents of a defect ensemble, or in the analysis of the complex spectra which arise from the interaction of slow positrons at or near the surfaces of solids. Experimental one- and two-dimensional angular correlation of annihilation radiation experiments in Al single crystals have shown that two defect species (mono- and divacancies) can be resolved under suitable conditions. Recent experiments at LLNL indicate that there are a variety of complex exit channels open to positrons interacting at surfaces, and ultimately these decay channels must also be suitably resolved from one another.

DIVACANCIES IN ALUMINUM

It is generally, although not universally accepted that the mechanism for enhanced high temperature self-diffusion in aluminum is a consequence of a significant equilibrium divacancy population. Aluminum is an ideal candidate material for a study of the resolving power of positron annihilation techniques for small defect clusters, in this case the separation of monovacancy and divacancy positron annihilation signals. The comparatively large total vacancy concentration at melting $(C_v(T_m)-10^{-3})$ in aluminum results in a wide temperature where >90% of the annihilations take place from positron-vacancy trapped states associated with an increasing divacancy population. Sufficiently high resolution two dimensional angular correlation of annihilation radiation (2D-ACAR) studies on single crystals of Al will eventually reveal the detailed anisotropies that may be associated with trapping in divacancies. However, recent 2D-ACAR experiments [1] performed at Brandeis University were only of sufficient resolution to allow one to follow the narrowing of the ACAR distribution with increasing temperature and to verify that lattice expansion alone could not explain the observed temperature dependence of the observed momentum distribution changes at high temperatures. Thus, one was forced to introduce an additional experimental argument, temperature (T), to compensate for the lack of defect resolving power.

By following the temperature dependence of the positron-electron annihilation signal it is possible to determine various vacancy point defect properties, particularly the vacancy formation enthalpy for monovacancies (H^F_{1v}) [2]. An example of such a study for aluminum is shown in Fig. 1. The temperature dependent "S" shaped curve was obtained from a 1D-ACAR experiment by measuring the peak counting rate. The full 2D-ACAR spectra for the positron Bloch and positron vacancy trapped states are shown as well so as to emphasize the overall spectral changes which occur upon trapping of the positron. As noted by Fluss and co-workers [3] it is possible to determine divacancy properties, which will manifest themselves in the high temperature region of the data shown in Fig. 1, if a priori information exists about the annihilation signal for both monovacancy and divacancy trapped positrons. Since such theoretically derived information was available for positrons in aluminum [3,4] the present application was possible.

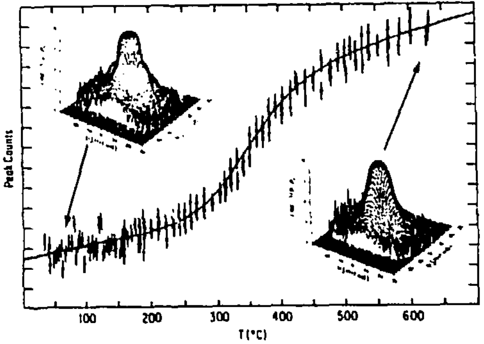

Fig. 1:One and two dimensional angular correlation data obtained from an aluminum single crystal, illustrating the peak count rate and 2D-ACAR experimental techniques. [1,3]

With several experimental and analysis constraints a mono-divacancy model analysis of the data could be obtained which was unique, and which yielded a divacancy binding energy of 0.30 ± 0.13 eV. The constraints used were: 1) the independent determination of H^F_{1v} from lifetime experiments at low temperatures, where the contribution from divacancies to the positron signal could be expected to be minimal, 2) the theoretically predicted peak count rates from 1D-ACAR experiments for positrons in monovacancies and divacancies, along with the predicted temperature dependencies from lattice expansion, and 3) a simultaneous or global analysis of data taken for three different crystal orientations.

The most important lesson learned in this analytical analysis was the critical role played by the last of these constraints. By "viewing" the temperature dependent signal from several crystal orientations differences in response were obtained. These differences apparently imposed a much more stringent set of criteria for the model fit than that which would have been obtained by simply repeating the same experiment three times at the same orientation. Such tomographic constraints are not new to the use of positron ACAR data, but this application to defect studies is! The sensitivity of this analytical method is illustrated in Fig. 2. Here, for the purpose of demonstration, the divacancy binding energy is fixed at either 0.2 or 0.3 eV for three independent analyses of data from three crystal orientations. Since a physically sensible analysis requires defect concentrations to be independent of crystal orientation, it is concluded that the analyses where the divacancy binding energy is 0.3 eV is preferable to the one where it is 0.2 eV. This demonstrates the value of the tomographic constraint which was imposed by simultaneously minimizing the mono- divacancy model to the temperature dependent data obtained from the *three crystal orientations*.

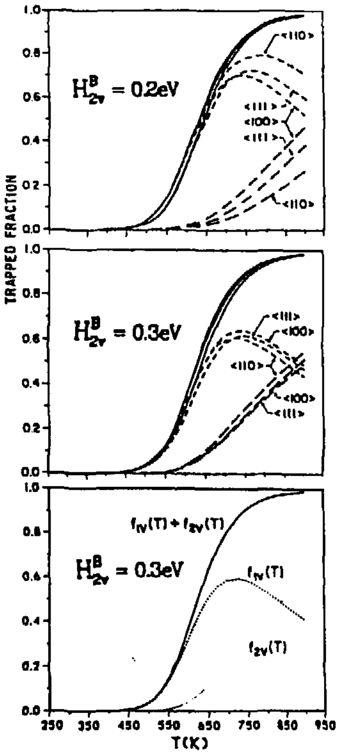

Fig. 2: Comparison of constrained mono-divacancy analyses for three crystal orientations in aluminum. The results are presented in the form of the fraction of the positron population annihilating from monovacancies, f_{1v}, and divacancies, f_{2v}, and the total fraction, f. [3]

Such a general method should be very important in future experiments where similar resolving power may be required. Extension to other defect systems is anticipated, particularly vacancy-solute clusters.

SURFACE ANNIHILATION STUDIES

With the recent development of intense, monoenergetic, variable energy positron beams at LLNL, it is now possible to study electron momentum densities at the surfaces of solids using positron annihilation techniques. Moreover, when the positron is energetically ejected from the surface into the vacuum as *positronium* (Ps) *it* is shifted into the forward hemisphere by an energy approximately equal to the work function energy for the Ps production process. This particular positron exit channel can also be studied using time-of-flight (TOF) techniques. This mode of annihilation may be of particular importance since the information about the surface properties of the sample is carried into a volume of space where it can be easily separated from those annihilations which take place from positrons localized or trapped in the near surface region.

The positron beams are obtained by moderating high energy positrons produced by pair production in a tungsten target irradiated by a pulsed electron beam at the LLNL 100 MeV Electron Linac [5,6]. The high energy positrons are moderated in *tungsten* vanes and a small fraction are emitted from the moderator surface due to the negative work function of the tungsten. These positrons are then electrostatically accelerated and transported along a magnetic guiding field to a UHV ($2x10^{-10}$ torr) chamber. Time of flight (TOF) and 2D-ACAR experiments are used to resolve the annihilation events coming from Ps in flight and positrons bound to the surface.

Fig. 3 shows the 2D-ACAR spectra obtained for positron beams incident on a [121] plane of Cu. Beam energies of 18 keV and 740 eV were used; for 18 keV a spectrum indicative of the Fermi surface of Cu is seen, while for 740 eV a high degree of anisotropy indicative of energetic Ps annihilating in flight is observed. The Ps component was resolved from this, and is shown in the top of Fig. 4. A simple model, which was calculated for a free electron gas, is shown for comparison. The momentum distribution of the Ps is determined by kinematics, the positronelectron interaction, and the electronic density of states. Underlying this spectrum is what is believed to be a spectrum from positrons which are trapped or bound at the surface. The asymmetry observed in this spectrum, which is shown in the bottom of Fig. 5, compared to the high energy spectrum, apparently arises from the localization of the electrons perpendicular to the surface and the delocalization parallel to the surface.

TOF spectra were obtained for the 740 eV incident positron beam. Here, the annihilation of Ps travelling away from the surface is detected by a collimated detector 10 cm from the Cu sample. Momentum resolutions similar to that achievable with ACAR are possible. It is noteworthy that the general features of the TOF spectra obtained to date confirm the 2D-ACAR distributions obtained for the same energetic Ps component.

The future for the use of positron beams seems to be a rich one. TOF and ACAR momentum techniques are complementary, viable spectroscopic tools for such studies, and as has been demonstrated here, different positron annihilation channels can be resolved from one another.

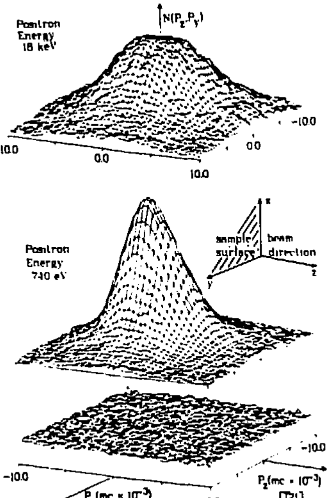

Fig. 3: The 2D-ACAR spectra for positron beams of 18 keV and 740 eV impinging on a single crystal of copper. [6]

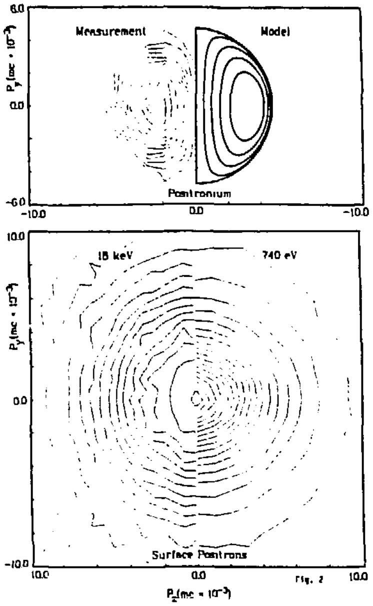

Fig. 4: Top) Contour maps comparing measured and computed momentum distributions for positronium ejected from the copper surface. Bottom) The bulk and surface trapped positron contours for the 18 keV and 740 eV beams. [6]

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