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RECENT PROGRESS IN ANNIHILATION RELATED STUDIES BY SLOW POSITRONS

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1. INTRODUCTION

The field of slow-positron physics has expanded significantly in the last few years to include particle and atomic physics but has been most extensive in those associated with condensed matter or material science. This can primarily be attributed to the development of more efficient moderators (conversion of fast-to-slow positrons). These moderators have been associated with both laboratory-and facility based beams. In this paper I will focus only on the material-science aspects however. Positrons can and are being used to examine all of the various fields. I feel the contribution in all these areas will be significant. I will primarily discuss those developments that have been developed in the area of interface science; a field that has both scientific and technological importance and has a limited number of nondestructive probes used in studying a buried interface. Interfaces are technologically imporant for applications such as electrical properties (semiconductor devices) and mechanical properties (adhesion). Such applications help to motivate the fundamental research of interface properties and dynamics, which is necessary to develop the basic understanding of new types of interfaces. The role of the interface is also important (i.e. grain boundaries) since it contributes to the strength of a solid. I will only discuss this area owing to the limited length of this paper, however those interested can read recent reviews by Schultz and Lynn[1] for solid-state studies and for atomic physcs reviews by Charlton,[2] and Stein and Kauppila.[3]

Results will be presented on interface studies that have occurred in the last year, including some unpublished results obtained at Brookhaven over the past few months. This field is in the early stages and I expect that the full utilization of this relatively new probe can be anticipated in the next few years. I expect that future studies will be made using laboratory-based beams (10^6 positrons/sec), as well as those based on intense positron beams ($>5\times10^7$ positrons/sec) which require both small and high brightness positron beams, as will be presented by K. Canter in this workshop.

The research on interfaces with variable-energy positron beams can be reduced, at least in the first instance, to measurements of the γ-rays resulting from annihilation of positrons in delocalized, trapped, or Ps states. Occasionally other signals are employed such as direct Ps detection, characteristic x-rays, secondary or Auger electrons. In this review most of the data will be that obtained with high resolution semiconductor detectors [intrinsic Ge or Ge(Li)]. The annihilation line-shape can be deconvoluted to extract the electron momentum distribution, but more often it is simply quantified by a line-shape parameter such as "S", which is the ratio of counts in a central portion of the annihilation photopeak to the total counts in the peak.

In order to utilized variable-energy positrons to study overlayers and their interfaces one must be able to adequately describe the positron implantation profile. The requirement to adequately describe the stopping profile in typical positron beam experiments is more demanding than previous electron studies. It is not surprising that the interaction of an energetic positron is different from electrons with similar velocities. The differences can be associated with the relative differential and total elastic cross-sections, and also with the different energy loss processes for the two particles. These variations are partly associated with the opposite charge, and partly with the fact that there is no Fermi sea of positrons in the sample so that there is no exchange part to the potential.

Results of the positron implantation profile are shown in Fig. 1 for four different incident positron energies. Typically 4,000-10,000 particles are needed to accurately define the profile. From these results we find the backscattered fractions (not shown in the figure) are a sensitive test of the relative weighting of the elastic to inelastic mean-free paths. Moreover, there is still a disagreement between the backscattered fraction determined from the Monte Carlo and those determined experimentally (Baker and Coleman[4] and Nielsen and Lynn.[5] This discrepancy is not fully understood but could be associated with an inaccurate description of correlation of positrons to the electron gas as well as the angle variation associated with inelastic collision. It is worth noting that agreemnt is generally good in determining the electron backscattered fraction.

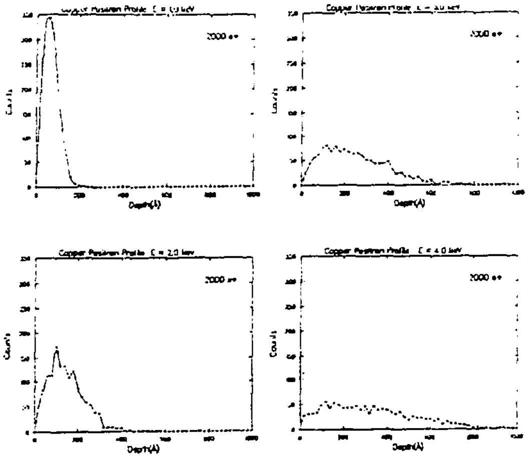


Fig. 1. Monte Carlo simulations of positron stopping profiles for Cu.

The curves shown by Lynn and McKeown[6] in Fig. 1 can be approximately represented by a derivative of a Gaussian as noted by Valkealahti and Nieminen.[7] However, work on parameterizing these profiles in various multi-layer systems is necessary so that experimenters can utilize analytical expressions of the implantation profiles for various composite systems.

Figure 2 shows a highly simplified schematic of positron interface trapping. The first observation that positrons were trapped at interfacial defects was for copper overlayers on a W(110) crystal (Schultz, Lynn, Frieze, and Vehanen[8]). The Cu, which forms an epitaxial overlayer of Cu(111), takes up the strain of the lattice mismatch in the first two atomic layers. From the results shown in Fig. 3, it can be seen that the yield of 2-keV incident positrons reemitted from the Cu(111) overlayer is only ~30% of the anticipated yield. This reduced yield continued until the Cu/W(110) system was annealed to above 1222 K, which is very close to the temperature required to thermally activate the first atomic layer of Cu on W(110) (Bauer et al.[9]) and is well above the temperature required to anneal out any point defects in the bulk Cu itself.

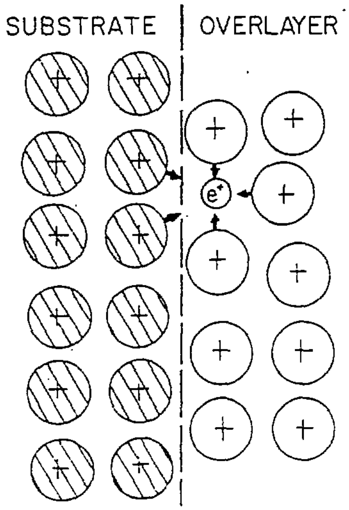


Fig. 2. Schematic representation of positron trapping at an open-volume defect at an interface.

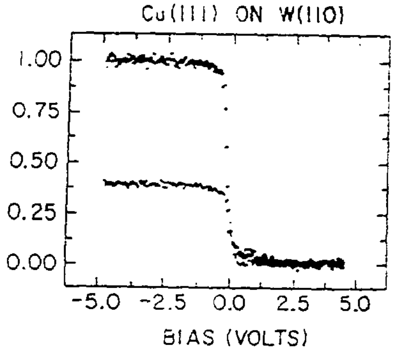
The observed recovery of the interfacial defects at the Cu/W(110) interface was supplemented with measurements of the yield versus energy in the as-deposited state. These results showed approximately a 50% decrease in the reemitted yield for incident positron energies above ~1 keV, which is consistent with the interfacial trapping.

An example of a "trapping" overlayer on a crystalline substrate, in which the positron diffuses, is that of SiO_2 on Si. Iwase, Uedono, and Tanigawa[10] first reported qualitative measurements in this system, showing an increase in the overfall fraction of positrons trapped following γ irradiation. More recently Nielsen, Lynn, Chen, and Welch[11] have made measurements in SiO_2 on Si(110) which fit with a superposition of parameters, including the diffusion in the substrate. Their results, shown in Fig. 4, yield the same value for L_+ as unmodified Si(110) for the portion at energies ≥ 7.5 keV, and the interface is seen very clearly to be near 7 keV. Using Eq. (1) for the mean penetration length, this corresponds to 3200 A, which is in adequate agreement with the known thickness of 3500 A.

$$L_+ = (D_+\tau_{eff})^{1/2} = (A/A')E_0^n , \qquad (1)$$

In another study of this type, Nielsen, Lynn, Leung,[12] found a different behavior when a 520-A oxide was grown on a Si(100) substrate. To reduce the effects of the electric field, the authors measured the sample at 500°C, obtaining the data shown in Fig. 5. At depths greater than the interface, the bulk diffusion length for the data shown was consistent with the field-free value. The increase of the line-shape parameter observed at the SiO_2/Si interface indicates positron trapping in open-volume defects. We have associated this increase with 3γ annihilations, suggesting the o-Ps is being formed in large open-volume spaces (voids) near the interface.

A different example is epitaxially grown semiconductor/semiconductor interfaces. An example described above (Fig. 4) illustrated the sensitivity of the variable-energy positron technique to the SiO_2/Si(110) interface. New studies presently underway are investigating some of the structural and electronic properties of heterostructures that are nominally epitaxial, grown using standard MBE (molecular-beam epitaxy) techniques (e.g., Bean[13]). Materials presently being investigated with variable-energy positrons include GaAs and Si_xGe_y alloys, which have (among others) applications as potential optical sources or detectors that can be matched to existing fiber optics. Other experiments have been performed for Si/Si superlattices, revealing new information about



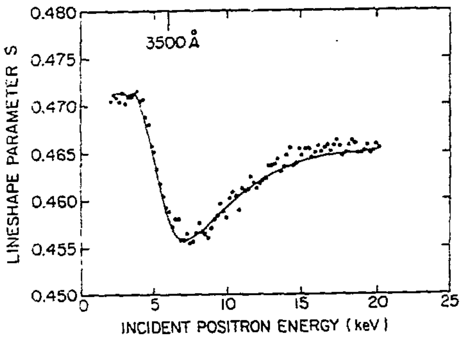


Fig. 3. Doppler-broadening parameter "S" as a function of incident positron energy in SiO_2 on Si(110). The curvature for energy greater than ~7 keV corresponds to positron diffusion in the crystalline Si. The influence of the interface is observable in the limiting value of the line-shape parameter, which the curve approaches (Nielsen, Lynn, Chen, and Welch[11]).

Fig. 4. Positron line-shape parameter S (open circles) and Ps-fraction (crosses) vs incident energy in Si with a thermally grown 52 nm overlayer of SiO_2. The sample had been heated to 970 K and the measurements were made at 800 K.

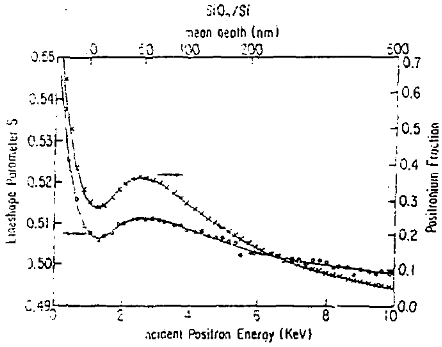


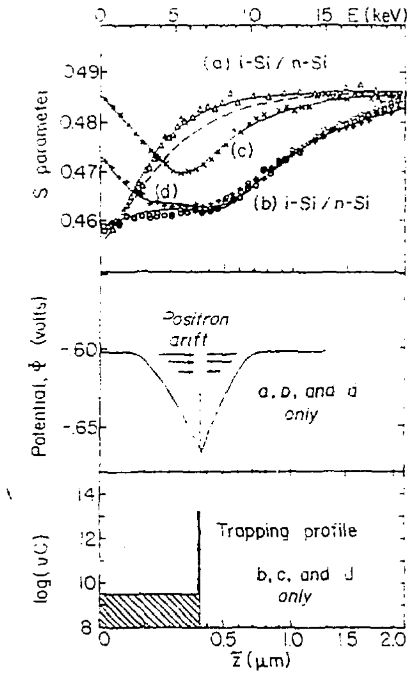
Fig. 5. Positron line-shape parameter S (open circles) and Ps-fraction (crosses) vs incident energy in Si with a thermally grown 52 nm overlayer of SiO_2. The sample had been heated at 970 C and the measurements were made at 300 K.

both electric field effects introduced by electrically active impurities in the epilayers and structural properties associated with the MBE fabrication of the material. Figure 6(a) shows data for a ~3000-A epilayer grown on an N-type Si(100) substrate. The results are only slightly different from those for "bulk" material due to a layer of boron trapped at the interface, which results in a bipolar field of ~2×10^3 V/cm directed towards the interface. The data in Figs. 6(b)-6(d) are for a ~3500-A epilayer on Si(100) that contains oxide-like defects near the interface. The various data sets shown in Fig. 6(b) at 20°C (as grown), Fig. 6(c) at 300°C, and Fig. 6(d) after returning to 20°C. The solid curves through the data are the result of the iterative modeling of the diffusion equation discussed above. Included in the modeling are the effects of positron drift velocity and trapping in defects, observed to be concentrated at the interface and spread (dilutely) throughout the overlayer. The figure also shows the bipolar potential calculated for this sample, and the defect distribution used in the model (Schultz, Tandberg, et al.[14]).

One of the important considerations for positron trapping in semiconductors is the charge state of the defect. For example, the data in Fig. 5 show clear signs of trapping in defects at the interface, which implies that they are either neutral or negatively charged. On the other hand, studies of thick silicon epilayers (4-6 μm) on Si substrates containing varying numbers of dislocations (from ~5×10^3 to $\geq 10^8$ cm^{-2}) show no signs of positron trapping, indicating that these defects are positively charged. It is clear that more detailed studies of the charge states for various defects will be pursued in the future, and in particular studies will investigate whether or not the electric fields associated with charged defects are leading to prethermalized trapping of positrons. Puska et al.[15] have discussed some theoretical aspects of positron states in defects in semiconductors, and Dlubek and Krause[16] and Dannefaer[17] have reviewed some of the bulk solid studies of semiconductors that have been conducted with positrons.

In a more recent study of Pd-Ta on Si(100) G. J. Van der Kolk et al.[18] have used Auger electron Spectroscopy, RBS and positrons to study applicability of Pd_xTa_{1-x} as a diffusion barrier on Si. The positron data shows the defect concentrations in the as-deposited state is very high where the positron diffusion length is one the order of 1 nm. The reaction of the metal overlayer with Si is easily detectable with positrons (see Fig. 7). The changes that occur before 600°C is associated with annealing out point defects and the alloy formation occurs between 600 -700°C.

Although still preliminary, studies of this type are providing the groundwork for using positrons to determine nondestructively depth profiles of defects in the near surface region of a solid. It is already possible to solve the problem without assuming a functional form for the defect profile, but without significant advancements in the numerical procedure the technique is limited by the experimental precision of the data. These concerns, and the correlation of defect and implantation profiles, will eventually establish the limits to which a profile of unknown defects can be uniquely determined experimentally.



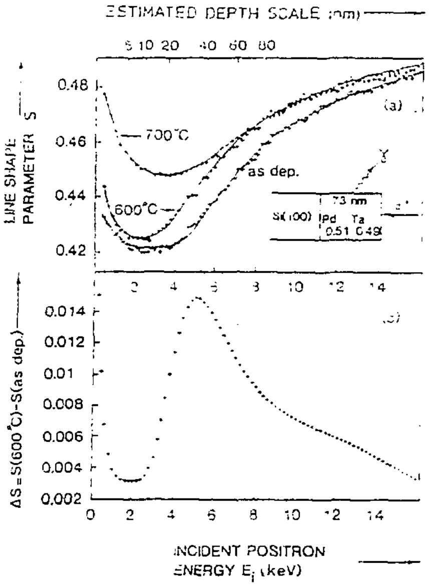


Fig. 6. S-parameters data for MBE-grown intrinsic Si epilayers on n-type Si(100). Data are (a) without defect trapping, and (b)-(d) for a sample containing oxide-type defects. All data are for samples at 20°C, except (c), which is at 300°C. The solid curves are obtained by iteratively solving the diffusion equation, including both defect trapping [profile νC(z)] and positron drift. The dashed curve is for bulk material. From Schultz, Tandberg, et al., 1988.

Fig. 7. (a) Line-shape parameter S of the 511 keV annihilation gamma peak as a function of the energy of a positron beam, incident on a 73 nm thick $Pd_{0.51}Ta_{0.49}$ overlayer on Si(100); as-deposited and after annealing at 600 C and 700 C. Lines are drawn to guide the eye. (b) ΔS=S(600 C) minus S(as-deposited).

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REFERENCES

1. P. J. Schultz and K. G. Lynn, Rev. Mod. Phys. 60, 701 (1988).
2. M. Charlton, Rep. Prog. Phys. 48, 737 (1985).
3. T. S. Stein and W. E. Kauppila, Adv. At. Mol. Phys. 18, 53 (1982).
4. J. K. Baker and P. G. Coleman, J. Phys. C: Solid State Phys. 21, L875 (1988).
5. B. Nielsen and K. G. Lynn, J. Appl. Phys. (June 1989) to be published.
6. K. G. Lynn and M. McKeown, unpublished.
7. S. Valkealahti and R. M. Nieminen, Appl. Phys. A 32, 51 (1984).
8. P. J. Schultz, K. G. Lynn, W. E. Frieze, and A. Vehanen, Phys. Rev. B. 27, 6626 (1983).
9. E. Bauer, H. Poppa, G. Todd, and F. Bonczek, J. Appl. Phys. 45, 5164 (1974).
10. Y. Iwase, A. Uedono, and S. Tanigawa, in ICPA85, p. 977, 1985; see also A. Uedono, S. Tanigawa, and Y. Ohsi, Phys. Letts. A 133, 82 (1988).

11. B. Nielsen, K. G. Lynn, Yen-C. Chen, and D. O. Welch, Appl. Phys. Lett. 51, 1022 (1987).
12. B. Nielsen, K. G. Lynn, T. C. Leung, D. O. Welch, and G. Rubloff, in *Defects in Electronic Materials*, edited by M. Stavola, S. J. Pearton, and G. Davies (Materials Research Society Symposia Proceedings, Boston, MA 1987).
13. J. Bean, Physics Today 39 No. 10, 36 (1986).
14. P. E. Schultz, E. Tandberg, K. G. Lynn, Bent Nielsen, T. E. Jackman, M. W. Denhoff, and G. C. Aers, Phys. Rev. Lett. 1988 (in press).
15. M. J. Puska, O. Jepsen, O. Gunnarsson, and R. M. Nieminen, Phys. Rev. B 34, 2695 (1986).
16. G. Dlubek and R. Krause, Phys. Status Solidi (a) 102, 443 (1987).
17. S. Dannefaer, Phys. Statud Solidi (a) 102, 481 (1987).