PHYSICAL REVIEW B VOLUME 33, NUMBER 5 1 MARCH 1986

Reinterpretation of oxygen on Al samples by variable-energy positrons

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(Received 24 June 1985)

When low-energy positrons are implanted into crystals and diffuse back to the crystal surface, they can form positronium by escaping from the crystal. This positronium formation fraction is a function of temperature for an aluminum sample coated with oxygen. Lynn observed large differences in this positronium fraction in the heating and cooling curves and attributed the hysteretic behavior to the oxygen on the aluminum surface undergoing a phase transition from an amorphous to a crystalline state. This Brief Report offers an alternative explanation for the hysteretic behavior of the positronium formation fraction, namely, the oxygen leaves the surface region either by desorption or by migration into the bulk Al crystal.

Within the last decade, it has become experimentally feasible to form controllable low-energy positron beams. Such probes have subsequently been used to study the properties of solid surfaces.[1] By bombarding a single crystal with variable-energy positrons and determining the relative fraction of reemitted positrons from the crystal and/or the relative positronium fraction, several workers have obtained information about the nature and concentration of defects near the surface region.

In 1980, for example, Lynn[2] performed just such an experiment on clean and oxygen-coated Al(111) crystal. He observed the relative positronium fraction (F_{Ps}) as a function of temperature. The oxygen goes into a spatially disordered state when it is deposited on aluminum at room temperature and the probability of positronium formation is low due to the trapping of positrons in the oxide region. After heating the oxygen coated sample above 500 °C, the relative positronium fraction increased dramatically. Lynn attributed this increase in the positronium fraction to the removal of surface traps because the oxygen on the aluminum surface underwent a structural phase transition from an amorphous to an ordered state.

This Brief Report proposes an alternative explanation for the experimental observations of Lynn.[2] Specifically, this article suggests that the large increase of F_{Ps} at high temperatures is associated with the depletion of the oxygen from the aluminum surface either by thermal desorption or by migration of the oxygen into the aluminum bulk. All the experimental data of Lynn seem to be correct although new experimental information and a different interpretation of his old data is added.

There are essentially two reasons for offering this explanation:

(1) Figure 1 of Ref. 1 displays the relative F_{Ps} as a function of temperature for positrons incident on a clean Al crystal at various energies. Lynn attributes the initial slope to positrons desorbing from surface states. Figure 3 of the same article is a plot of F_{Ps} versus temperature, a similar plot for oxygenated aluminum during heating and subsequent cooling of the sample. As noted earlier, there is a large increase in the positronium fraction when the sample is heated to 500 °C. Upon subsequent cooling of the crystal, the F_{Ps} in Fig. 3 resembles, but does not match, the heating part of Fig. 1 and the hysteresis behavior is noted. The hysteretic cooling curve of Fig. 3 is similar to the heating curve for clean aluminum (Fig. 1) when the comparison is made between curves of similar positron energy. There is a small vertical offset for the two sets of curves but the initial slopes appear to be equal. The difference in the F_{Ps} versus positron energy curves for clean and oxygen coated aluminum is less than the difference in the same set of curves for two different faces [(111) and (110)] of clean aluminum. The small difference is probably due to the presence of a small residue of oxygen on the surface.

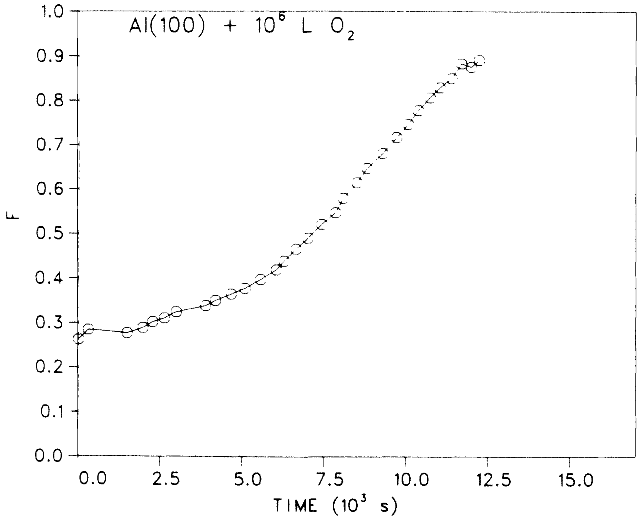

FIG. 1. Shows the measured positronium fraction (F_{Ps}) vs time for an Al(100) at 550 °C exposed to 10^6 langmuirs of oxygen ($1\mathrm{L}=10^{-6}$ torr sec) at room temperature. This behavior shows the removal rate of oxygen from the sample surface. Auger-electron spectroscopy measurements were made on a similar sample, which showed the decrease of the oxygen peak at 505 eV compared to the Al peak at 68 eV during heating. The incident positron beam energy is 500 eV.

(2) Once the oxides associated with mechanical and electrolytic polishing have been removed, the surfaces of two Al(110) and one Al(100) crystals were exposed to air for three days and cleaned in this laboratory by heating the sample to 600 °C for about one hour instead of undergoing the normal sputtering-annealing cycle. A small oxide peak was still observed after this heat treatment. The composition of the surfaces was determined by Auger electron spectroscopy (AES) and noting the relative peak heights of the aluminum at 68 eV to oxygen at 505 eV. The ratio of the oxygen to aluminum peak heights was reduced by an order of magnitude after heating. After we observed such a phenomenon, we discovered that similar observations had been reported by Bedair, Hofmann, and Smith.[3] In fact, Bedair *et al.* could only form crystalline oxides by depositing the oxygen at low exposure ($\approx 15\times10^{-6}$ torr sec) and at a limited elevated temperature range (300—500 °C). In Fig. 3 of Lynn, each data point at high temperatures took minutes during which time the oxygen could have been removed from the aluminum surface.

We present here new data showing time dependence of F_{Ps} for $\mathrm{Al}_x\mathrm{O}_y$ when the sample is maintained at 550 °C and incident positron beam energy of 500 eV. This curve can be interpreted as due to the disappearance of oxygen and removal of surface traps from the surface although an AES study of a similar crystal confirms this interpretation. It is not clear whether the oxygen is thermally desorbed or migrates into the crystal. If the oxygen diffuses into the crystal, such a phenomenon could explain an unidentified shallow positron trap that occurs in positron beam studies[4] at temperatures below 150 K. The resolution of this point awaits more detailed beam studies.

Low-energy positron beams may be used as a probe of surface disorder. To observe the structural order-disorder phase transition for aluminum oxide may require a more complicated heating and cooling cycle than that used by Lynn along with detailed Auger and LEED measurements.

We would like to thank R. Howell for reporting a similar observation, and D. Gidley and W. Frieze for technical discussions. Work supported by Division of Materials Sciences U.S. D.O.E. under Contract No. DE-AC02-76CH00016.

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[4] P. S. Schultz, K. G. Lynn, R. N. West, C. L. Snead, Jr., I. K. MacKenzie, and R. W. Hendricks, Phys. Rev. B **25**, 3637 (1982).