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Positron spectroscopy of solid N_2

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We have measured the positron and positronium emission yields from a solid N_2 target surface bombarded with slow positrons. We determine the inelastic threshold energy for positrons in N_2, $E_{th}=7.08\pm0.10$ eV, and the threshold for forming positronium, $E_{Ps}=10.0\pm0.5$ eV. In contrast to the rare-gas solids, the low-lying electronic excitations cause E_{th} to lie below E_{Ps}. Unlike electrons, positrons should not excite triplet states at a detectable level, and therefore E_{th} is the energy of the lowest-lying singlet vibrational level less the positron affinity, ϕ_+. We thus determine $\phi_+=1.31\pm0.10$ eV. From the positronium formation threshold we obtain the positronium binding energy in the solid, $E_b=3.8\pm0.5$ eV. Below the inelastic threshold a positron is found to lose an energy 30 ± 5 meV per collision in agreement with a model in which the principal energy-loss mechanism is the excitation of optical phonons.

I. INTRODUCTION

Various properties of positrons and positronium in the rare-gas solids have been deduced from positron diffraction measurements and from the energy dependence of the positron and positronium yields.[1-5] The table of the positronic properties of the rare-gas solids now includes such quantities as the positronium binding energies in the solids E_b, the positron affinities ϕ_+, the inelastic mean free paths, and the positron reemission yield extrapolated to zero positron implantation energy y_0. The positron information is useful for understanding the dynamics of positrons in solids, provides independent checks on the inelastic thresholds measured by other techniques, and is the basis for the most efficient moderator for producing slow positron beams.[3] We have extended our study to a molecular solid in order to test the model developed for the rare-gas solids in a wide-gap insulator that has optical-phonon modes and low-lying electronic excitations.

II. EXPERIMENT

The experiment was performed on a magnetically-guided slow positron beam equipped with an ultrahigh-vacuum chamber and a cryogenic target stage located inside a 50-K radiation shield. The substrate for forming the solid N_2 was a single crystal of Ni(100) that had been cleaned by ion bombardment and annealing, and then left for several days in the vacuum. The low base pressure of 10^{-10} Torr and low-temperature shields ensured that the sample surface would remain uncontaminated for the several hour period required by the measurements. The sample was prepared by cooling the substrate to 20 K and condensing N_2 gas at 3×10^{-6} Torr for 10 min. The solid N_2 layer was thus about 5000 Å thick, and therefore greater than the implantation depth and escape depth of the positrons.

A radioactive source of 8 mCi ^{58}Co and Ni(100) moderator[1] produced a beam of 10^4 slow positrons per second. The total reflection coefficient for positrons, the positron reemission yield, and the positronium formation probability were measured as previously described.[2,5]

III. RESULTS

In Fig. 1(a) we display the positron reemission probability for positron incident energies from 0 to 16 eV. The dip near zero energy is caused by positrons being unable to escape from the solid before they fall below the escape energy. The full width at half maximum of the dip is $\Delta E=(1.28\pm0.07)$ eV. The structure visible from 2 to 7 eV is probably caused by diffraction effects which are ob-

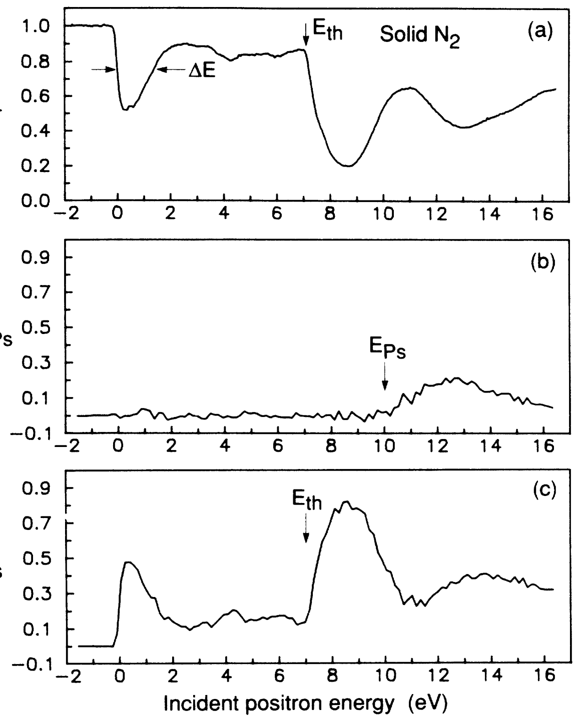

FIG. 1. Energy dependences of the probabilities for (a) positron reemission, Y_+; (b) positronium emission, Y_{Ps}; and (c) positron annihilation in the solid, Y_s. The sample is a solid N_2 surface.

scured by the high reemission probability of the thick target. At higher energies, the reemission probability suddenly drops due to the onset of inelastic channels for the positron. The inelastic threshold energy, $E_{th}=7.08\pm0.10$ eV, was determined from the intersection of two straight-line segments fitted to the data in the vicinity of the threshold. The error estimate is twice that due to statistics to allow for possible systematic effects. The positronium formation fraction versus positron incident energy is shown in Fig. 1(b). There is essentially no positronium formation below a threshold energy $E_{Ps}=10.0\pm0.5$ eV. In Fig. 1(c), we see no obvious excess positron annihilation in the solid at an incident energy near E_{Ps}, and thus there is no evidence for an anomalously short Ps diffusion length near the Ps formation threshold as was found for the rare-gas solids.[5]

The reemitted positron energy spectrum for solid N_2 bombarded with 2-keV positrons (Fig. 2 inset) was obtained with a 4-kG permanent magnet placed behind the sample.[6] After being transported to the analysis electrode, the reemitted positrons have nearly all their momentum directed along the axis of the beam, and thus the energy in the inset to Fig. 2 is the total kinetic energy of the reemitted positrons. The total slow positron yield as a function of positron implantation energy is shown in Fig. 2. The yield extrapolated to low energy is $y_0=(65\pm5)\%$, and the energy at which the yield falls to half its initial value is $E_{1/2}=(3.0\pm0.3)$ keV.

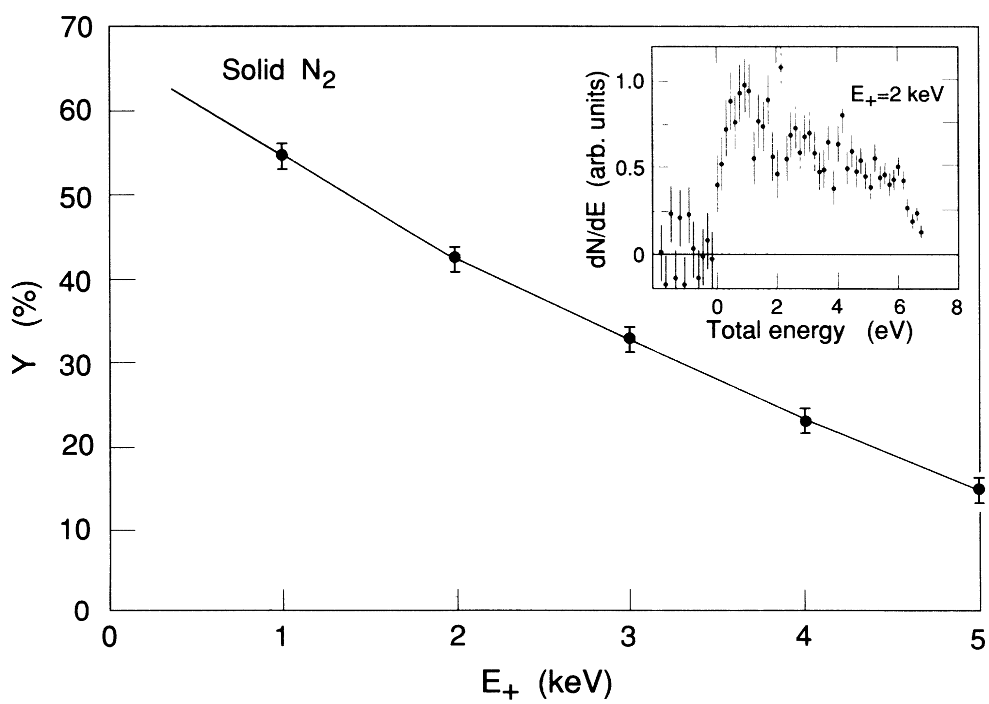

FIG. 2. Positron yield vs positron implantation energy for solid N_2. The inset shows the spectrum of the total positron energy for positrons reemitted from solid N_2 bombarded with 2-keV positrons.

IV. DISCUSSION

We determine the positron affinity ϕ_+ for solid N_2 as follows. Unlike electrons, low-energy positrons will not appreciably excite triplet states in collisions with solid N_2 due to the absence of the exchange force and because of the smallness of the spin-orbit coupling for a slow particle that is repelled from the nuclei. The positron inelastic threshold $E_{\rm th}$ therefore corresponds to the positron implantation energy that is just sufficient to excite the lowest singlet level in the solid, the $\nu=0$ vibrational level[7] of the $a'\,{}^1\Sigma_u^-$ band E_0. Since a positron gains an energy equal to ϕ_+ when it enters the solid, we have

$$E_0=E_{\rm th}+\phi_+ \ . \tag{1}$$

We do not know of any direct measurement of E_0 in the solid, but Marsolais *et al.*[7] using electron-energy-loss spectroscopy find the singlet levels E_ν for $\nu=5,6,\ldots,12$. We extrapolate to find the $\nu=0$ level in solid N_2 by fitting the vibrational energy levels for the Morse potential,[8]

$$E_\nu=E_\alpha+E_\beta[(\nu+\tfrac{1}{2})-x(\nu+\tfrac{1}{2})^2] \ , \tag{2}$$

to the eight measured $a'\,{}^1\Sigma_u^-$ levels given in Table I of Ref. 7. We obtain $E_\alpha=8.296$ eV, $E_\beta=0.188$ eV, $x=0.007\,54$, and $E_0=(8.390\pm0.005)$ eV. In the gas, the optical threshold is[9] 8.398 eV, which corresponds to the lowest-lying singlet state. In the solid, the optical experiments[10] could only determine the $a\,{}^1\Pi_g$ and $w\,{}^1\Delta_u$ thresholds, which turned out to be, respectively, 0.035 and 0.050 eV lower than in the gas. Since the Σ states would be perturbed less by being in the solid than the Π and Δ states, a reasonable estimate for the $a'\,{}^1\Sigma_u^-$ optical threshold would be 8.380 eV, in agreement with the extrapolated $\nu=0$ level in the solid. A value that reflects both estimates with a reasonable error bar would be $E_0=(8.385\pm0.010)$ eV. Substituting into Eq. (1) we find $\phi_+=(1.31\pm0.10)$ eV.

We interpret the dip near zero energy in Fig. 1 using a model from Ref. 2. Positrons begin a random walk in the solid at an exponentially distributed depth. As they slow down, the positrons scatter isotropically, losing a mean energy δE in each collision. The positrons colliding with the surface escape with a probability given by the transmission probability of a plane wave past a step potential of height ϕ_+. A Monte Carlo calculation shows[2] that the width of the dip and the mean energy loss per collision are related by $\delta E/\phi_+=a(\Delta E/\phi_+)^n$, with $a=0.0255$ and $n=2.68$. The mean energy loss per collision is therefore $\delta E=(30\pm5)$ meV, significantly greater than in the rare-gas solids, and suggesting that optical phonons as well as acoustic phonons are involved. We note that optical phonons in solid N_2 have an energy greater than 188 meV corresponding to the vibrational frequency of the electronic ground state.[11]

At a positron incident energy equal to the positronium threshold, the positron energy inside the solid is

$$E_{\rm Ps}+\phi_+=E_g-E_b \ , \tag{3}$$

where E_g is the energy gap in the solid (the minimum energy to raise an electron to the conduction band) and E_b is the binding energy of positronium in the solid. Since the electron affinity of solid N_2 is negative,[7] $\phi_-=-0.8$ eV, the photoemission threshold will be the same as the energy gap. From the experiment of Lau *et al.*[12] we find $E_g=15.1$ eV, and thus conclude that $E_b=(3.8\pm0.5)$ eV. In Ref. 4 we found that the positronium binding energies in the rare-gas solids are closely approximated by $\epsilon^{-2}\frac{1}{2}R_\infty$, where ϵ is the low-frequency dielectric constant, and $\frac{1}{2}R_\infty$ is the Ps binding energy in vacuum. Using the handbook value $\epsilon=1.45$ for liquid N_2, and assuming the same value would be found for solid N_2, we would predict $E_b=3.22$ eV, somewhat less than our measurement.

The reemitted positron energy spectrum in the inset to Fig. 2 cuts off at the inelastic threshold as expected. We will now use the data of Fig. 2 to determine the mean free path λ of positrons diffusing in the solid below the inelastic threshold.[2] The implantation energy at which the slow positron yield falls to half its low energy value, $E_{1/2}=(3.0\pm0.3)$ keV, is related to the hot positron diffusion length L by the expression $L=(A/A')E_0^n$ from Eq. (28) of Schultz and Lynn;[1] we will assume that the stopping profile is a derivative of a Gaussian so that $A'=1.26$. The parameters $A=4.0\pm0.3$ μg/cm^2 and $n=1.62\pm0.05$ are from Ref. 13. We find $L=(1800\pm300)$ Å. From the relation[2] $L\approx(E_{\rm th}/3\delta E)^{1/2}\lambda$, we find $\lambda=(200\pm40)$ Å, a value indistinguishable from the mean free path we determined for solid Ar.[2]

V. CONCLUSION

The main differences between solid N_2 and a rare-gas solid are (1) the inelastic threshold is below the positronium formation threshold due to the existence of low-lying electronic excitations, and (2) a hot positron loses energy more rapidly in the molecular solid because of the emission of optical phonons. On the other hand, the solid rare gases and solid N_2 have positron affinities in the 1–2-eV range, and the positronium binding energies in the solids are close to what one would predict for a positronium atom in a uniform dielectric medium. We conclude that positrons in a wide band-gap solid have a behavior that is largely independent of whether the solid is molecular or not.

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