Positron Dynamics in Rare-Gas Solids

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Using a beam of slow positrons in ultrahigh vacuum, we estimate the positron mean free path and energy-loss rate and measure the inelastic thresholds due to exciton, electron-hole pair, and positronium formation in solid rare-gas targets. The measurements are used to explain the large emission energies of positrons from the wide-band-gap solids in terms of a hot-positron model. The large diffusion length of the hot positrons in the solid rare gases makes them very efficient moderators for producing slow positrons.

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There have been many studies of positrons in ionic crystals.[1] Measurements have revealed the positronium binding energies, wave function, effective mass, and formation mechanisms. Powdered insulators provided the first sources of positronium in vacuum[2,3] and the first practical efficient moderator for producing slow positrons.[4] Recently there have been studies of the positronium formation mechanism at the surface of ice,[5] of positronium emission from quartz,[6] and of positron reemission by various ionic solids.[7-9] Unfortunately, this large body of work has not led to a precise picture of how a low-energy positron interacts with an insulator. The present study attempts to rectify this situation by careful measurements on the ideal wide-band-gap insulators, the rare-gas solids.

When positrons are implanted into an ionic solid at kiloelectronvolt energies, one observes the reemission of positrons with a spread of kinetic energies comparable to the band-gap energy. In Ref. 9 this reemission was interpreted in terms of a modified version of the positronium breakup mechanism originally proposed by Canter *et al.*[4] In this model the reemitted positrons result from positronium formed in the bulk which diffuses to the surface and breaks up, emitting the positron via an Auger-like process. We now find that positrons are copiously emitted with several electronvolts of kinetic energy from solid Ne, Ar, Kr, and Xe. However, because of the simplicity of these materials we have also been able to examine the emission process in detail by measuring the mean free path and inelastic thresholds for few-electronvolt positrons. The measurements are not consistent with positronium breakup and suggest that the emitted positrons are simply those that have not thermalized and happen to scatter back to the surface. Our new results also show that the rare-gas solids are a new class of very efficient slow-positron moderators.

We propose to explain the observed positron reemission as follows. In any material, energetic positrons lose energy rapidly by making inelastic collisions involving electronic transitions. However, in an insulator there can be no more of these events once a positron has less energy than is needed to make an electron-hole pair, an exciton, or positronium. The positron continues to lose energy by creating phonons, but since the maximum phonon energy ($E_{\max}$) is small, the diffusion length of the hot positron is large. Positrons reaching the surface before their energy falls below the positron work function ($\phi_+ > 0$) may escape.[10] We shall refer to this process as the hot-positron model.

The data for the present experiment were obtained with a magnetically guided slow-positron beam in a vacuum chamber with a base pressure of $\approx 2\times 10^{-10}$ Torr. The rare-gas samples were condensed at a pressure of $\approx 10^{-6}$ Torr onto a Ni(100) substrate cooled by a ≈ 10-K refrigerator and shielded by a ≈ 60-K shroud. Film thicknesses were obtained from the pressure during deposition and the total time of exposure, and calibrated by measurement of the positron yield versus exposure. The slow positrons ($\approx 10^4$ $\sec^{-1}$) were produced by a Ni(100) moderator and an 8-mCi ^{58}Co source.

Figure 1 shows the positron reemission spectra for Ar, Kr, and Xe. Positrons were implanted at 1.8 or 4.8 eV into the solids, and the energy spectra of the reemerging positrons were measured with a retarding grid. The emission spectra were recorded with the sample in a large magnetic field so that the energy scale is the total positron kinetic energy.[11] The spectra for the two incident energies were normalized to the same area. The following observations are in agreement with the hot-positron model. (1) The maximum energy of the emission spectra corresponds closely with the inelastic threshold, whose measurement we discuss below. (2) The emitted positrons tend to have lower energies when the incident positrons are implanted with higher energy. (3) The total positron reemission yields extrapolated to zero implantation energy, y_0, given in Table I, are much greater than can be explained by positronium breakup. For example, the value of 67% for Ar far exceeds the positronium formation probability in the solid, which is only 10%.[12] The structure in Fig. 1 presumably is due to the combined effects of the posi-

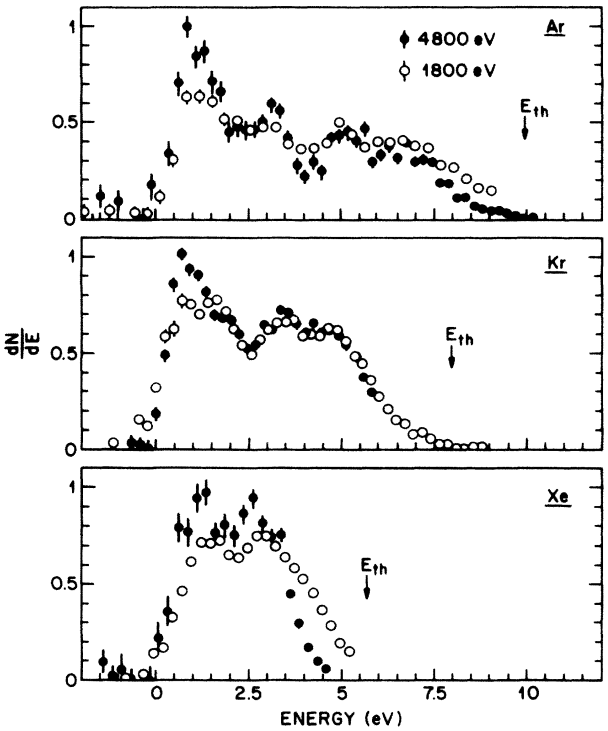

FIG. 1. Spectra of the total energy of positrons reemitted from Ar, Kr, and Xe solid surfaces. The inelastic threshold E_{th} is indicated. The positrons were implanted at 1800 and 4800 eV.

TABLE I. Properties of the rare-gas solids.

	Ar	Kr	Xe
	Electronic properties		
E_{max} (meV)[a]	8.3	6.2	5.4
E_g (eV)[a]	14.16	11.60	9.28
E_a (eV)[b,c]	−0.25	0.3	0.4
E_x (eV)[a]	12.1	10.25	8.36
	Positronic properties		
$E_g + E_a - 6.8$ (eV)	7.1	5.1	2.9
E_{Bragg} (eV)	2.3(1)	2.0(1)	1.4(1)
ϕ_+ (eV)	1.7(1)	1.5(1)	1.6(1)
E_{th} (eV)	9.95(5)	7.9(1)	5.67(5)
E_b (eV)	2.5(1)	2.2(2)	2.1(1)
$-\phi_{Ps}$ (eV)	2.8(1)	2.8(1)	2.8(1)
E_0 (keV)	7.1(5)	7.1(5)	6.1(5)
y_0 (%)	67(3)	63(3)	50(3)
δE (meV)	6(2)	3(1)	3(1)
τ (nsec)[d]	0.43(3)		0.40(3)

[a]*Rare Gas Solids*, Ref. 14.
[b]Perluzzo *et al.*, Ref. 14.
[c]Schwentner *et al.*, Ref. 14.
[d]Liu and Roberts, Ref. 15.

tron, electron, and hole density of states in the solid.

The inelastic thresholds are determined by measurement of the positron reemission probability, R, and the positronium yield as functions of the incident positron energy. The positronium yield is obtained by the peak-to-total method,[13] and the reemitted positrons are detected without energy discrimination by a Channeltron. Figure 2 shows examples of such measurements for solid Ar targets ≈ 20 Å thick [Fig. 2(a)] and ≈ 15000 Å thick [Fig. 2(b)]. In Fig. 2(a), a Bragg peak is observed in R at $E_{Bragg} = 2.3$ eV. If we assume that the Ar grows in the (111) orientation,[14] the position of the Bragg peak implies that the positron work function is $\phi_+ = 1.7$ eV. Between 4 and 10 eV R is only about 0.32 and independent of the incident positron energy. Since no free Ps is being formed at these energies the remaining positrons must be trapped either in the Ar film or at the substrate. When the thickness of the film is increased [Fig. 2(b)], the value of R in the same energy range increases dramatically, indicating that the trapping occurs at the Ni substrate. In Fig. 2(b), the Bragg peak is unobservable because the reemission probability is near unity for energies below 10 eV. The sharp drop in R below 2 eV occurs because $\phi_+ > 0$ and, therefore, a positron which loses its incident energy while inside the solid will become trapped. The width of this dip, ΔE, is related to the mean energy loss per collision, as will be discussed. Between 6 and 10 eV in Fig. 2(b) there is a slow decrease in R that we attribute to formation of positronium in vacuum by electron pickup at the surface. The amount of Ps formed by this process increases with increasing incident energy since the positrons lose energy while inside the solid and may reemerge with an energy below the threshold. The slow decrease in R becomes more pronounced with time, presumably as a result of the surface's becoming contaminated. We identify a sharp drop in R at $E_{th} = 9.95 \pm 0.05$ eV as the elastic threshold. This corresponds to the threshold for positronium formation in the solid as we show below. At a slightly higher energy than E_{th} there is an increase in R at the threshold for formation of an exciton, E_x. The reemission probability rises at this threshold since a positron which creates an exciton will no longer have sufficient energy to form Ps and may be reemitted. A second increase in R is identified with the threshold for formation of an electron-hole pair at E_g. Our estimates $E_x = 12.0 \pm 0.3$ eV and $E_g = 14.2 \pm 0.3$ eV agree with optical measurements.[14] The positron work function does not affect these thresholds because the positron is escaping from the solid after the inelastic event. We have also measured E_x and E_g using positron energy-loss spectroscopy and obtained results in agreement with the above measurements.

Our identification of E_{th} as the threshold for positronium formation in the solid is confirmed by our measurements of the positronium yield in Fig. 2. This exhibits a sharp increase at a positron energy $E = E_{th}$

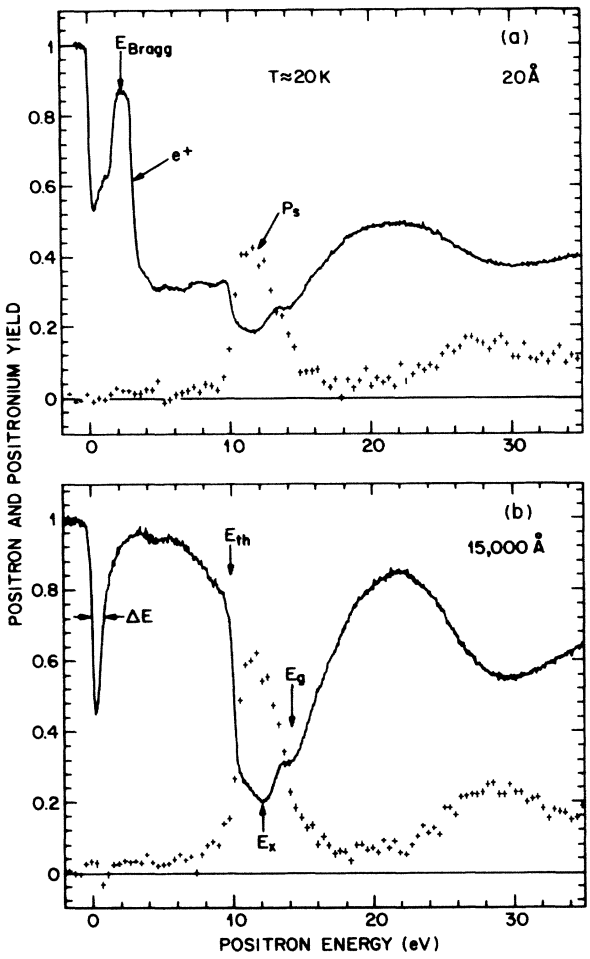

FIG. 2. Positron (e^+) reemission yield (solid line) and positronium (Ps) yield (crosses) for solid Ar films of two different thicknesses. The inelastic threshold, E_{th}, is found to be the threshold for Ps formation. The exciton threshold energy, E_x, and band gap, E_g, are indicated.

and falls again when the incident positron energy is sufficient to excite an exciton. There is also some positronium formation for $E < E_{th}$ that is sensitive to surface contamination and thus can be associated with the production of vacuum positronium at the surface. The undulations at higher energies in a similar measurement on ice have been explained by the Ore model.[5] The positronium binding energy in solid Ar is $E_b = E_g - E_{th} - \phi_+ = 2.5(1)$ eV. The positronium work function is $\phi_{Ps} = E_g + E_a - \frac{1}{2}R_\infty - E_{th} = 2.8(1)$ eV, where E_a is the electron affinity. These measurements have been collected in Table I along with similarly obtained values for Kr and Xe.

Having established the position of the inelastic threshold we would like to know the energy-loss rate for a positron with energy less than E_{th}. We determine the mean energy loss per collision, δE, from the width of the dip in $R(E)$ near $E = 0$ in Fig. 2(b). Since the

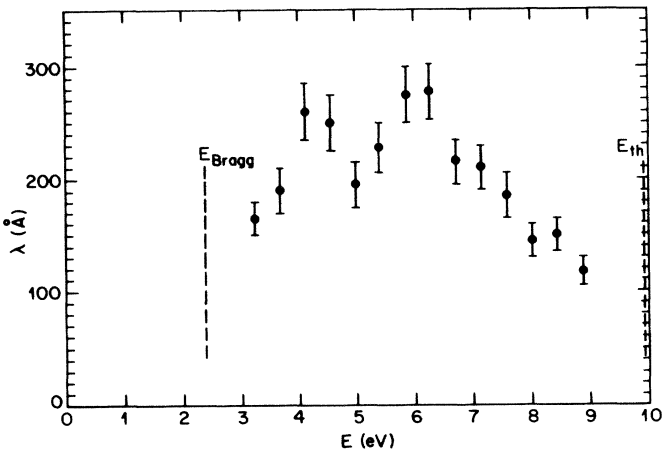

FIG. 3. Positron mean free path vs energy for solid Ar.

positron work function is positive, positrons with sufficiently small incident energies will lose that energy quickly enough to become trapped, thus causing the dip in $R(E)$. A Monte Carlo calculation was performed with the assumptions that (1) the positrons begin a random walk in the solid at a depth exponentially distributed with a mean depth λ, (2) λ is independent of energy, (3) the scattering is isotropic, (4) the mean energy loss per collision is δE, (5) a positron colliding with the surface from within may escape if the perpendicular component of its energy is greater than ϕ_+, and (6) the probability for escape is given by the quantum-mechanical transmission of a plane wave past a step potential of height ϕ_+. The calculation yields a dip in R that can be approximated by $R(E) = 1 - [1 + (E/\Delta E)^p]^{-1}$, with δE and ΔE related by $\delta E/\phi_+ = a(\Delta E/\phi_+)^n$, and with $p = 1.80$, $a = 0.0255$, and $n = 2.68$. We attribute the nonzero value of R at $E = 0$ in Fig. 2(b) to the combined effects of the finite energy spread of the incident beam, 0.2 eV, and reflection of the incident beam at the surface. Using the measured width $\Delta E = 0.1$ eV and $\phi_+ = 1.7$ eV we obtain $\delta E = 6(2)$ meV for Ar. Since the values for δE are not very different from the maximum phonon energies E_{max} (see Table I), a probable conclusion would be that acoustic-phonon generation is the primary energy-loss mechanism for positrons with energies below the inelastic threshold.

We have determined the positron mean free path λ by measuring R versus the thickness, x, of the absorbed gas layer. We calculate λ by fitting R to the form $1 - \alpha \exp(-x/\lambda)$ near $x = 0$ and plot the results for Ar in Fig. 3. Knowledge of λ and δE enables us to estimate the hot-positron diffusion length, $L \approx (E_{th}/3\delta E)^{1/2}\lambda \approx 5000$ Å for Ar. This estimate for L agrees with our measurement of E_0, the positron implantation energy at which only half the positrons return to the surface (see Table I). The characteristic time for a

positron to lose half its energy is roughly $\Delta t = \frac{1}{2}(E_{th}/\delta E)\lambda(m/2E_{th})^{1/2} \approx 10$ psec, much shorter than the positron annihilation lifetime τ.[15]

In conclusion, positron reemission from the rare-gas solids is consistent with the hot-positron model, and cannot be explained by positronium breakup. Presumably, the same model is also applicable to other insulators, and indeed it is in agreement with the polarized-positron data of Ref. 7. Unfortunately, contrary to Ref. 9, it now does not appear that ionic crystals would make very good moderators for muons,[16] although solid Ne could possibly give a useful slow-muon yield. On the other hand, the large yield of reemitted positrons and their long diffusion length suggests that the rare gases will be excellent slow-positron moderators. As an example, we find that Ne condensed onto a small ^{22}Na source yields slow positrons with an efficiency of 0.3%, better than the current best transmission moderator[17] and equal to the best backscattering moderator.[18] Solid Ne would be an easily fabricated low-Z moderator useful for producing spin-polarized slow positrons[19] and for increasing the efficiency of an intense ^{64}Cu slow-positron beam.[20] The high reemission coefficient for positrons with energy below the inelastic threshold could be useful in ultrahigh-efficiency multiple-bounce moderator geometries and in experiments requiring positron bottling and bunching techniques.[21]

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