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Multiple encounters of thermal positrons with surfaces

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We propose that the thermal-positron reflection by surface potentials, including the potential-well effects, is more substantial than predicted by a simple step-potential model. Thus, positrons may encounter the surface potential many times to participate in surface-escape processes. The physical picture presented here is not only more realistic than the step-potential model, it also suggests possible explanations of the related experimental findings.

It is known that when positrons are implanted into solids, they quickly lose their energies and come to equilibrium with the solids. The thermalized positrons may diffuse back to the surface where they can be emitted as positronium (Ps) and/or positrons (when the positron work function is negative); they may also be trapped into a surface image potential.[1] Because of the thermal energy involved, it was suggested that the positrons can be reflected by the surface potential back into the bulk solids,[2] so inhibiting the positrons from participating in the surface-escape processes.

This internal surface reflection was demonstrated by Britton *et al.*[3] from positron- and Ps-emission measurements. The modeling of positron reflection by the surface potential is adopted from Ref. 2, where a surface-potential step accounting for the positron work function (ϕ_+) is employed. The reflection and transmission of thermal positrons by this potential step are deduced as being of the same order of magnitude. In fact, the transmission coefficient used in Ref. 3 is

$$\Gamma = \frac{4[k_B T(k_B T - 2\phi_+)]}{\{k_B T - \phi_+ + [k_B T(k_B T - 2\phi_+)]^{1/2}\}^2}\ , \qquad (1)$$

where $k_B T$ is the thermal-positron energy. Recently, Brandes[4] measured positron reemission yield from a thin Ni film and deduced the internal surface-reflection coefficient as $R = 0.63 \pm 0.04$, consistent in order of magnitude with that predicated by Eq. (1).

However, there are problems related to modeling the surface potential by a simple potential step as in Eq. (1). It is recognized that a potential well exists in between the vacuum and the solid which is responsible for the positron surface states on many metal and semiconductor surfaces.[1] Although the dimension of the potential well is of the order ~ 1 Å (note that the thermal-positron de Broglie wavelength is $\sim(62\,\text{Å})[(300\,\text{K})/T]^{1/2}$) it will significantly affect the positron reflection from the surface potential. The larger mismatch of the wave numbers for a positron inside the solid and inside the potential-well region can increase the quantum-mechanical reflection by an order of magnitude. In fact, for a square potential well of depth V_0 at a zero-work-function surface, the transmission coefficient is[5]

$$\Gamma = (1 + \tfrac{1}{4}\{[k_B T/(k_B T + V_o)]^{1/2} - [(k_B T + V_0)/k_B T]^{1/2}\}^2 \sin^2\{[2m(k_B T + V_0)a^2/\hbar^2]^{1/2}\})^{-1}\ , \qquad (2)$$

where m is the positron effective mass in solids, $\hbar$ is the Planck constant, and a and V_0 are the width and depth of the potential well, respectively. In our case, a is of the order of 1 Å and V_0 is about the Ps vacuum binding energy (6.8 eV). Equation (2) is approximately decided by $4k_B T/V_0$, which is of the order of 10^{-2} in the temperature range of interest. The corresponding transmission coefficient will then be an order of magnitude smaller than that suggested by Eq. (1) and the aforementioned experimental results. For negative-work-function surfaces at high temperatures, the transmission coefficient can be around 10^{-1}; nevertheless, it is much smaller than it would be from Eq. (1). For positive work-function surfaces, the reflection coefficient will be unity in terms of thermal-positron reemissions, even when a potential well is present. Figure 1 shows the temperature-dependent reflection coefficients from Eq. (1) (solid line); the dash-dotted line shows the results when the potential-well effect is considered. The positron work function is taken as -0.1 V, the potential well has a width of 1.5 Å, and a depth of 6.8 eV.

The experimentally deduced reflection coefficients in Refs. 3 and 4 can be interpreted more realistically when stronger reflection effects are taken into account. The key here is the fate of the reflected positrons. It is the implicit assumption in the modeling of Refs. 3 and 4 that the reflected positrons will no longer contribute to positron surface processes. In reality, however, positrons reflected by the surface potential will experience scatterings since they are still present in the solids. The average

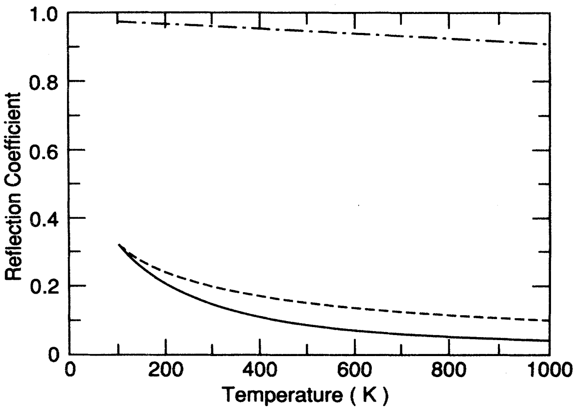

FIG. 1. Reflection coefficients for $\phi_+ = -0.1$ eV when Eq. (1) is used (solid line); when the potential-well effect is considered (dash-dotted line); and when the multiple-encounter model Eq. (3) is used (dashed line).

scattering angle of a thermal positron should be around 120°;[2] thus a sizable fraction of the positrons will approach the surface again before they wander further from the surface and annihilate in the bulk solid. Once at the surface region, positrons can participate in one of the surface escape processes, i.e., emission as positrons, or Ps, and trapping into the surface state.

In Refs. 3 and 4, it is not the outcome of a single encounter of a positron at the surface that is measured. The measured reemitted positron fraction is the sum of the many meetings between the positron and the surface potential. The reemitted fraction will not be sensitive toward the strong reflection that the positrons experience during one attempt. One can model the multiple encounters of a positron with the surface potential in a simple way as follows. Assume that at a single encounter, the positron reflection coefficient is r and the transmission coefficient is $t=1-r$, and for these reflected positrons, a fraction α (≤ 1) on average will come back to the surface and join the surface processes again. After many such repeats, the total surface transmitted fraction is

$$\tau = t + \alpha r t + \alpha^2 r^2 t + \alpha^3 r^3 t + \cdots = \frac{t}{1-\alpha r} . \quad (3)$$

From Eq. (3), if all the reflected positrons are lost in the solid ($\alpha=0$), then the measured transmission coefficient will be the same as that for a single encounter. When α is nonzero, there will then be a finite number of positrons coming back to the surface and participate in the surface processes. In this case, the transmission coefficient obtained from Eq. (3) τ will be larger than that for a single encounter t. The extreme case would be for $\alpha=1$, when positrons will all be transmitted, i.e., a total absorbing boundary. In Fig. 1, we have also shown τ (the dashed line) with $\alpha=0.99$ and t from the dash-dotted line. The temperature dependence of τ obtained here closely resembles that of Eq. (2) (solid line) without any parameter adjustments.

If τ is assumed to equal 0.1–0.9, which is of the same magnitude as deduced from Eq. (1) and Refs. 3 and 4, and if t equals 1%, we can deduce from Eq. (3) that $\alpha \approx 91-99.9\,\%$. In other words, there are about 0.1–9 % of positrons "lost" in the bulk solids after one reflectoin by surface potentials. Since experiments in Refs. 3 and 4 were carried out on relatively clean and defect-free samples, it is natural to assume that the lost positrons are due to annihilation events. The total bulk annihilation fractions of the reflected positrons corresponding to $\tau=0.1-0.9$ are between 0.9 and 0.1, which is actually the measured reflection coefficient in Refs. 3 and 4.

There has actually been some experimental evidence for a large reflection of positrons at the surface. Köymen, Gidley, and Capehart[6] suggested from a simple classical model that their experimental results indicate a reflection coefficient of 95% for positrons reaching the surfaces; thus, positrons may encounter the surface potential many times through which they join the surface processes. Another example which may directly indicate the multiple encounter process is the positron microscope measurements of positron diffusion and emission in solids.[4] By combining a classical diffusion equation with a nontotal absorbing boundary, the positron reemission yield and the spatial profile widths of reemitted positrons have been analyzed.[4] The positron-emission-yield measurement gives a reflection coefficient consistent with Eq. (1) as discussed above, the experimentally measured spatial profile widths, however, are larger than the calculated results from the diffusion equations. Among other possibilities, a larger reflection coefficient (the authors in Ref. 4 suggested $R \geq 0.92$) can explain the discrepancy. We point out that it will be the reflection at a single encounter that is important for introducing widened spatial profile of positron emission, while the net result of multiple reflection is important for the positron emission yield measurements. Thus, it is realistic to explain the findings of Ref. 4 with the multiple-encounter model proposed here. We note also that analyzing the experimental results with the diffusion equation approach may not be appropriate for evaluating quantum-mechanical properties such as reflection, but the qualitative features should at least hold for experiments in Ref. 4.

A further example where surface reflection should be accounted for is the positron-annihilation-induced Auger electrons from solid surfaces. Positron-annihilation-induced Auger electron spectroscopy (PAES) has recently been developed as an unique probe for solid surfaces.[7,8] It is believed that positrons originating the core holes for the subsequent Auger deexcitations are those which are trapped in the surface image potential. The experimental support for this claim may be divided into two parts. On the one hand, temperature-dependent PAES signals are found to be complementary to that of thermal Ps desorption from surfaces, which are both attributed to surface trapped positrons.[7,8] On the other hand, surface adsorption effects on PAES have been measured simultaneously with the electron-collision-induced Auger electron spectroscopy (EAES). The observed differences in PAES and EAES responses were explained by assuming that positrons producing the PAES signal are trapped at the surface potential.[9] These experiments, however, do not exclude the possibility that a small percent of the PAES signal is due to bulk delocalized positrons.

Recently, Soininen, Schwab, and Lynn[7] measured the temperature dependence of PAES signal from a Ge(100) surface with relatively high accuracy. While they observed an essential desorption of the surface positrons through the Ps signal, the PAES signal was, nevertheless, nonzero and was about 5% of the room-temperature intensity at elevated temperatures. The authors introduced other positron surface trapping centers to explain the nonzero PAES signal. We suggest that positron surface reflection can also explain the nonvanishing PAES signal. Positron work function at a Ge(111) surface was calculated to be +1.98 eV; [10] it is possible that the Ge(100) surface also has a positive work function.[11] While the thermal-positron reemission channel is blocked in this case, Ps emission and positron surface trapping are still possible; thus, the effective reflection probability is not unity, though it is much enhanced compared with a negative work-function surface. We note that the trapping branching ratio is of the same order of magnitude as the net reflection τ, and the contribution of the reflected positrons to PAES will be due to those which annihilate while within the Auger electron escape depths (~10 Å). Considering that thermal-positron random-walk diffusion length before they annihilate is about 10^3 Å, we can roughly estimate that 10^{-2} subsurface positron core annihilations may produce Auger electrons when compared with the surface trapped positrons. Here, it is not necessary to introduce the multiple-encounter picture to account for the subsurface PAES contribution, but multiple-encounter will certainly enhance the probability of such a contribution. The above estimate agrees in order of magnitude with the results in Ref. 7. Figure 2 shows two sets of model calculations of temperature-dependent PAES with (i) the surface positron desorption model (the parameters are the same as those for thermal Ps desorption in Ref. 7), and (ii) the desorption model plus a bulk delocalized positron contribution of 6% of the surface trapped fraction at lower temperatures. The low-temperature data are normalized to unity. The figure shows that the difference between the two models compares well with the observed difference of Ps desorption at PAES desorption behaviors on a Ge(100) surface.[7]

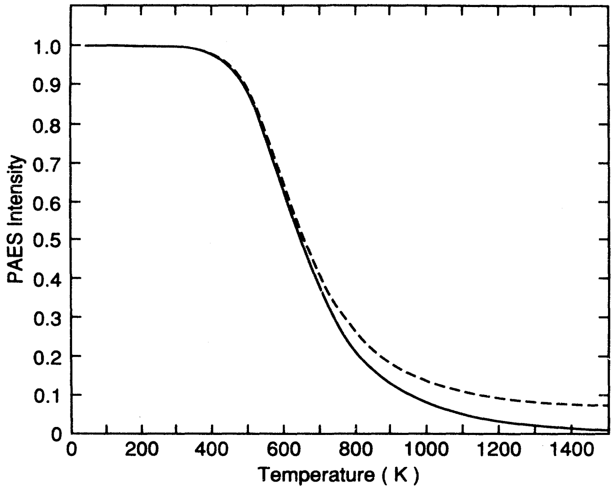

FIG. 2. The calculations for the PAES temperature dependence from a pure desorption model (solid line), and from a desorption model plus a constant bulk delocalized positron contribution (dashed line).

In conclusion, we have suggested that positron reflection from the surface potential is more substantial than that previously suggested. Positrons will thus encounter the surfaces with many attempts to participate in the surface processes. For a clean, defect-free sample, a few percent of the reflected positrons will annihilate in solids after each encounter. The suggested picture is possible to explain the difficulties in previous positron measurements. The experimental data of Refs.3 and 4 can also be interpreted more physically taking into account the fate of the reflected positrons.

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