Brief Reports

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Calculation of momentum distribution of positronium ejected from surfaces

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The two-dimensional momentum distribution of positronium formed and ejected from a surface is calculated and compared with experimental results. It is shown that the higher-order Born contribution is very important in explaining the experimental momentum distributions of positronium ejected from the surface.

In the recent years, it has been recognized that positron beams are a very useful probe for studying solid surfaces.[1] Experimental methods such as low-energy positron diffraction and positron-energy-loss spectroscopy present complementary information to that obtained from electron-beam experiments. The characteristic that most distinguishes positron beams from electron beams is the formation of the electron-positron bound state called positronium, or Ps, which is very sensitive to the surface electronic structure because Ps formation occurs at the solid surface.[2] The kinetic energy of the implanted and reejected positrons thermalized inside the solid is the negative of the positron work function if it is negative. The energy distribution of the formed and ejected positronium is thought to be proportional to the density of states of the surface electrons which combine with the positron to make positronium.[3] Therefore, the angular and energy distributions of the emitted positronium atoms may be used as a surface-sensitive probe, which we call positronium formation spectroscopy (PsFS), to measure the density of states. For this purpose, a knowledge of the elementary process of positronium formation is needed to extract the surface density of states from the measured energy and angular distributions of Ps. Recently, Ps formation experiments have progressed to measure these distributions in detail.[3-6] In a previous paper,[7] we presented a theory of Ps formation using an analogy with the quantum theory of resonant ion neutralization[8] at surfaces, and calculated the energy distribution of Ps to compare with the experiment by Mills and Pfeiffer.[4] The purpose of the present paper is to apply our previous theory to the calculation of two-dimensional Ps momentum distribution, which will be compared with recent experiments by Howell, Meyer, Rosenberg, and Floss[5] and Lynn *et al.*[6]

Because the energy level of the positronium 1S state is located inside the band of most metals, Ps formation can be theoretically described as a kind of resonant charge-exchange process (or chemisorption process) at the surface. Ps formation is distinguished from ion neutralization because of its light mass, which requires a quantum description of the motion of the center of mass of the positronium atom. In a previous paper,[7] we proposed a theory of Ps formation using the golden-rule formula for resonant charge exchange,[8] which is

$$P=(2\pi/\hbar)\sum_f\sum_{\mathbf{k}} N_{\mathbf{k}}|V_{a\mathbf{k}}|^2|u_{\mathbf{p}}|^2\delta(E_i+e_{\mathbf{k}}-E_f-e_a) , \quad (1)$$

where $N_{\mathbf{k}}$ is the Fermi-Dirac function, $V_{a\mathbf{k}}$ the hopping term of the electron between the substrate, $e_{\mathbf{k}}$, and positronium, e_a states, and $u_{\mathbf{p}}$ is the matrix element for the center of mass of the electron and positron system. E_i, the kinetic energy of the emitted positron, is effectively the negative of the positron work function $(-\phi_+)$ and $E_f=\mathbf{p}^2/2M$ is the kinetic energy of the emitted positronium atom whose mass, $M=2m$, is twice the electron mass. The separation of the matrix element into the relative coordinate part and the center-of-mass part is assumed in Eq. (1). By introducing the chemisorption function, $\Delta(e)$, as

$$\Delta(e)=\pi\sum_{\mathbf{k}}|V_{a\mathbf{k}}|^2\delta(e-e_{\mathbf{k}}) , \quad (2)$$

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and by dividing P of Eq. (1) by the initial flux of the center of mass $v=(-2\phi_+/M)^{1/2}$, we have the positronium formation probability as follows:

$$n=(2/\hbar v)\int \frac{d^3\mathbf{p}}{(2\pi)^3}\int de\, N(e)\Delta(e)\,|u_{p_\perp}|^2 \times\delta(E_i+e-E_f-e_a)\ , \quad (3)$$

where $\mathbf{p}$ is the formed positronium momentum and $p_\perp$ is its normal component.

Now let us assume the "wide-band limit" where $\Delta(e)$ is independent of e, which is widely used in theories of ion neutralization at surfaces.[9] In fact, the purpose of PsFS is to determine the shape of $\Delta(e)$ which is proportional to the surface density of states when $V_{a\mathbf{k}}$ in Eq. (2) is independent of $\mathbf{k}$. However, since our present purpose is to give the whole shape of the angular distribution, we simplify the electronic structure.

In the recent experiments by Howell *et al.*[5] and by Lynn *et al.*,[6] the two-dimensional angular correlation of the annihilation γ ray is used to measure the momentum distribution of formed and ejected positronium at surfaces. This quantity can be calculated from Eq. (3) as follows:

$$\frac{d^2n}{dp_x dp_\perp}=(\Delta/\pi\hbar v)(2\pi)^{-2}\sqrt{p_{\max}^2-(p_\perp^2+p_x^2)}\,|u_{p_\perp}|^2\ , \quad (4)$$

where it is assumed that the Fermi energy is much larger than the positronium work function

$$\phi_{\rm Ps}=-p_{\max}^2/2M=e_a+\phi_-+\phi_+\ ,$$

which is valid in present cases. Then, under the assumption of the wide-band limit, the momentum distribution of the ejected positronium is determined by $|u_{p_\perp}|^2$, which is the matrix element concerning the translational motion of the positronium.

The calculation of $|u_{p_\perp}|^2$ in Eq. (3) was done in a previous paper,[7] where it was shown that the first Born approximation is not sufficient but that higher-order Born contributions are important in Ps formation. In the first Born approximation, $|u_{p_\perp}|^2$ is

$$|u_{p_\perp}|^2=\left|\int_{-\infty}^{\infty}e^{ip_iz}u(z)e^{-ip_\perp z}dz\right|^2 =1/[\alpha^2+(p_i-p_\perp)^2]\ , \quad (5a)$$

when a simple exponential attenuation function from the surface is chosen for $u(z)$, $u(z)=\exp(-\alpha z)\theta(z)$, where α^{-1} is the value of the order of attenuation length of electron density from the surface, and $p_i[=(-2M\phi_+)^{1/2}]$ is the initial momentum of the center of mass. The higher-order Born contribution is taken into account when we introduce the optical potential to the positronium state.[7] The normalized Born approximation is derived from the optical potential theory[8] from which $|u_{p_\perp}|^2$ becomes

$$|u_{p_\perp}|^2=|u_{p_\perp}|^2_{\rm Born}\frac{1}{(1+\frac{1}{4}n_{\rm Born})^2} =\frac{1}{\alpha^2+(p_i-p_\perp)^2}\,\frac{1}{|1+(M/2\hbar^2p_\perp)\Delta/2\alpha[\frac{1}{2}+(1/\pi)\tan^{-1}(p_\perp/\alpha)]|^2}\ , \quad (5b)$$

where $n_{\rm Born}$ is the total neutralization probability of Eq. (3), but the initial momentum of the center of mass is $p_\perp$.

By substituting Eq. (5) into Eq. (4), we have the theoretical two-dimensional momentum distribution of the ejected positronium atoms from the surface. Figures 1(a) and 1(b) show the calculated contour map of the distribution using the first Born [Eq. 5(a)] and the normalized Born [Eq. 5(b)] approximations, respectively. Figure 1(c) is the experimental contour map obtained by Lynn *et al.*[6] using the Al(100) surface.

It can be seen that the agreement of our normalized Born approximation with experiment is much better than that of the first Born approximation. This tendency does not depend on the parameters and the functional form of $u(z)$ but indicates the inadequacy of the Born approximation in the low-$p_\perp$ region. In particular, in the low-$p_\perp$ limit, $u_{p_\perp}$ becomes zero using the higher-order Born theory while it remains finite in the first Born approximation.

Evidently, the experimental contour map of Fig. 1(c) supports the higher-order Born theory in the low-$p_\perp$ region.

Now let us mention the theory recently proposed by Walker and Nieminen,[9] where the calculation of the Ps momentum distribution is done in agreement with the experiment. In their theory, the first Born approximation with different assumptions for the matrix element is used. The most significant difference from the present theory is that they subtract the positronium flux toward bulk ($|u_{-p_\perp}|^2$ using our notation) from the flux toward the vacuum. This procedure enables them to fit their calculation to the experiment in the low-$p_\perp$ region, even using the first Born approximation. However, the experimentally measured momentum distribution of Fig. 1(c) is that of the higher-energy (hot) part of Ps formed due to the direct process, which is theoretically described by Eq. (1). The isotropic momentum distribution of the adsorbed (cold) Ps is subtracted to obtain the direct process experimentally,[5,6] which cannot be the direct Ps flux toward bulk.

In conclusion, we calculated the two-dimensional angular distribution of formed and ejected Ps from surfaces using the normalized Born approximation, taking into account the higher-order Born contribution, which agrees well with the experiment. To explain the lower normal-momentum part of the experimental distribution, the higher-order Born effect is very important. The assumptions used for the present calculations, the separation of the matrix element, the choice of the function of $u(z)$, and the wide-band limit, do not seriously affect the present results.

The extension of the present theory to positronium for-

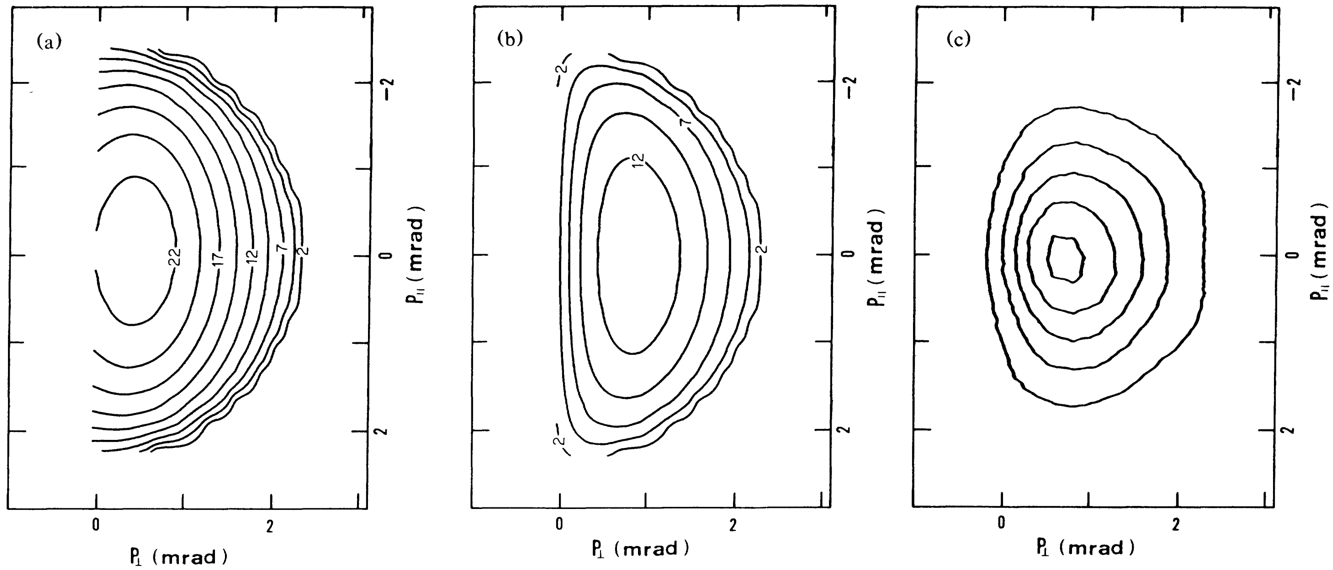

FIG. 1. (a) Contour map of the ejected Ps momentum distribution calculated using Born approximation of Eq. (5a). Parameters are chosen as $\alpha=0.5$ and $\Delta=0.1$ a.u. (b) Same as for (a) but using the normalized Born approximation of Eq. (5b). (c) Experimental contour map obtained by Lynn *et al.* (Ref. 6) from the Al(100) surface.

mation in the scattering of high-energy positrons from surfaces[10] is in progress.

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