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Search for adiabatic positronium emission from a metal surface

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We measure by time of flight the energy distribution of positronium (Ps) emitted normally to a clean Al(111) surface that is bombarded with energetic (1.5–4.5 keV) positron pulses. The data are consistent with the predictions of a free-electron model for nonadiabatic Ps emission with less than 10^{-3} of the Ps emitted normal to the surface in a monoenergetic peak at the maximum allowable energy. The Ps work function at 300 K is (-2.536 ± 0.035) eV in agreement with the (-2.519 ± 0.027) eV obtained from the sum of the electron and positron work functions minus the binding energy of Ps.

I. INTRODUCTION

To a good first approximation positronium (Ps) emission into the vacuum due to the presence of a thermalized positron near the surface inside a simple metal occurs with the sudden removal of one of the electrons from the Fermi sea.[1] Consequently, the momentum spectrum of the emitted Ps has features in common with the angle-resolved electron spectrum due to photoemission. The Ps kinetic energy E is no greater than the Ps negative work function, given by the Ps binding energy in vacuum, $\frac{1}{2}R_\infty$, less the sum of the electron and positron work functions: $-\phi_{\rm Ps}=\frac{1}{2}R_\infty-\phi_--\phi_+$. The Ps spectrum exhibits a broad distribution of energies that has a sharp step at $E=-\phi_{\rm Ps}$ corresponding to the Fermi surface. The measured angular distribution of the Ps emitted from various Al surfaces has been approximately reproduced by the simplest Fermi "golden rule" theory using a surface-projected nearly-free-electron conduction band for Al, a constant Ps-formation matrix element, and conservation of $k_\parallel$ and energy.[2]

In order to have a practical basis for measuring the electronic properties of the surfaces of metals, we need to test the accuracy of our model. In simple metals any corrections to the lowest-order picture would be of intrinsic interest as regards to understanding the many-body interactions of a light impurity. As in photoemission, the Ps emission spectrum will at some level be distorted by inelastic effects.[3] In a shake-up event, the metal is left in an excited state having one or more electron hole pairs in addition to the original hole due to the electron pickup by the positron. In the case of photoemission from a core state one observes an infrared divergent tail extending to the low-energy side of an otherwise monoenergetic electron peak. The opposite effect would be a shake-down event in which the hole percolates toward the Fermi surface by Auger-like transitions that give more energy to the outgoing particle. The most extreme example of shake-down would be adiabatic emission that leaves the metal in its ground state and the emitted particle having the full energy and no component of momentum parallel to the surface. Considering the relative smallness of the Ps negative work function compared to the Fermi energy or the plasma frequency, one might suppose that the Ps would be emitted slowly compared to the characteristic response time of the electron gas and that the conditions for adiabatic passage of the positron from the metal into the vacuum would be satisfied. It is to be noted that adiabatic photoelectron emission from alkali-metal surfaces for low photon energies is a possibility that does not seem to be contradicted by the data,[4] although most of the effects can be explained by the nearly-free-electron model.[5]

We describe here an experiment to search for a monoenergetic component in the Ps spectrum from a clean Al(111) sample. Since the metal would be left in its ground state, the monoenergetic peak would correspond to emission normal to the surface with an angular spread due only to thermal effects. Thus, with an experiment that looks only in a small solid angle about the normal direction we are able to establish a fairly stringent upper limit on the intensity of the adiabatic Ps. In view of how well the Ps would appear to satisfy the requirements for adiabatic emission, the small adiabatic intensity seems surprising. Possibly the neutrality of the Ps makes higher-order effects on its velocity spectrum occur with small probability.

II. EXPERIMENT

Slow positrons[6] were obtained from the electron beam dump of a microtron accelerator[7] operating at 18.5 MeV and a 30-Hz repetition rate. The 16-μsec-long 40 mA electron pulses each produced about 10^5 slow positrons by means of a W shower converter and a $W(110)$ single-crystal slow-positron moderator.[8] The slow positrons were stored in an rf-trapped magnetic bottle and formed into 8-nsec full width half maximum (FWHM) pulses containing positrons with kinetic energies spread over the range 0–3 keV. A delay of 7 μsec after the end of the accelerator pulse was introduced to reduce the background due to prompt neutrons and γ's from the beam dump.

The 12.7-mm-diam 2.5-mm-thick 99.999% pure Al(111) crystal was electropolished in a 1:5 mixture of

perchloric acid and ethanol. Before each data run the sample was cleaned *in situ* while annealing near the melting point by bombarding at a 45° angle of incidence with 5 μA of 500-eV Ar^+ ions for periods of more than 1 h. Auger electron spectra were measured using a double-pass cylindrical-mirror analyzer, 3-keV exciting electrons, and 4-eV peak-to-peak modulation. The initial amplitudes of the O 510-eV and C 272-eV peaks relative to the 68-eV Al peak were typically 0.01 corresponding to 0.005 and 0.02 monolayers, respectively. In our ultrahigh vacuum chamber (1.0×10^{-10} torr gauge reading) the O contamination grew at a rate of 0.02 monolayers per 22 h, so that the O contamination averaged over the course of an 8-h run was about 0.01 monolayer.

Figure 1 shows the geometry of the sample, the baffles that prevent the detection of scattered Ps, the Pb slit defining the fiducial volume, and the plastic scintillator detector. The latter is in the form of an annulus 16 in. square, 8 in. inside diameter, and 0.25 in. thick. The sample is attached to a heater stage on the end of a long travel vacuum manipulator. The sample and the inner wall of the vacuum chamber in its vicinity are biased at -1500 V relative to ground to provide optimum timing for the positron bunches. As a result, the positrons are implanted into the Al crystal with kinetic energies from 1.5 to 4.5 keV, and the Ps spectra should be relatively free of the effects of nonthermal positrons.[9]

To establish precisely the distance from the sample to the slit a 1.9 μCi source of ^{22}Na was attached to the face of the Al sample after the experiment was concluded. The manipulator was extended to the vicinity of the slit and the count rate versus position is shown in Fig. 2. The centroid must be corrected for the 1.0 mm thickness of the source by adding 0.5 mm.

Time-of-flight data was obtained using a time-to-amplitude converter and multichannel analyzer. The start signal was derived from the high-voltage bunching pulse and the stop signal was taken from a constant fraction discriminator fed by the summed anode pulses from the two photomultiplier tubes attached to the annular scintillator. Spectra shown in Figs. 3(a) and 3(b) were taken with the manipulator set at $z=240.0$ mm and

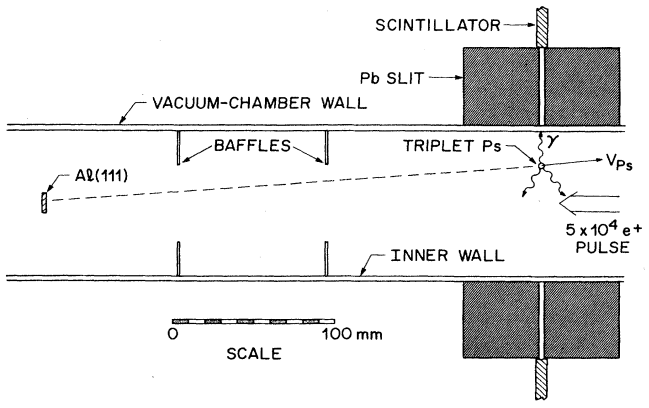

FIG. 1. Apparatus for measuring Ps velocities by time-of-flight using a pulsed positron beam.

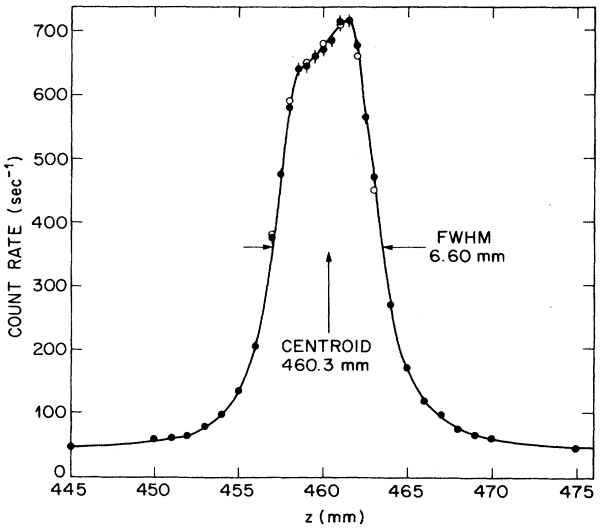

FIG. 2. Spatial resolution function of the detector.

157.0 mm, corresponding to sample-to-slit separations $\Delta z=220.8$ and 303.8 mm, respectively. A measurement of the time distribution of positrons incident on the target, Fig. 3(c), was taken using a 20 mm diam$\times$20 mm long plastic scintillator detector located about 150 mm from the target and viewing the target directly. The amplitude of the prompt peak was reduced by a factor of about 10 by a differential discriminator set to reject large pulse amplitudes. There is consequently a 60 nsec dead time after the prompt peak in Fig. 3(c). Since the time-to-amplitude converter can only process one event per positron bunch, the spectra have been corrected for the likelihood that early events reduce the number of later events. Spectrum (a) contains 2.89×10^5 pulses and employed a time per channel of 0.8142 nsec, as measured with a crystal-controlled calibrator. Spectra (b) and (c) were taken using 1.000 nsec per channel and contain, respectively, 1.35×10^6 and 4.86×10^4 pulses. The probability of detecting a Ps signal is of order 10^{-2} per bunch in Figs. 3(a) and 3(b), and is about 0.5 per bunch in Fig. 3(c). The spectra have all been summed over pairs of channels for display.

The delayed structure in the positron pulse shape of Fig. 3(c) is presumably caused by the release after the bunching pulse of positrons that had been trapped outside of the buncher. We have removed the background due to delayed positrons by fitting spectrum (c) separately to spectrum (a) over the range 88–445 nsec and to (b) over the ranges 155–435 and 705–875 nsec. Subtracting the fitted spectrum (c) from (a) and (b) leaves the delayed peaks due to the Ps time of flight from the sample shown in Figs. 3(d) and 3(e). Arguing that we are only interested in the behavior of the spectrum at the Fermi edge and that the Ps component formed from epithermal positrons is probably very small, we subtract a small background [dashed lines in Figs. 3(d) and 3(e)] with a value chosen simultaneously to remove any epithermal component and to make the spectrum well behaved at low energies. As

in Ref. 1, we obtain the differential spectra of the perpendicular component of the Ps energy, $E_\perp$ shown in Fig. 4, by multiplying the data by $t^2\exp\{t/\tau\}$, where τ is the lifetime of triplet Ps in vacuum, 142 nsec. The spectra have been collected into energy bins of constant 0.04 eV width.

We use Eq. (6) of Ref. 1 as a model:

$$dN/dE_\perp \propto E_\perp^{1/2}\tan^2\alpha\,\Theta(-\phi_{Ps}\cos^2\alpha - E_\perp) + E_\perp^{-1/2}(-\phi_{Ps}-E_\perp)\Theta(-\phi_{Ps}-E_\perp) \times\Theta(E_\perp+\phi_{Ps}\cos^2\alpha) \quad (1)$$

Here α is the collimator half angle and $\Theta(x)$ is the unit step function. From the geometry of Fig. 1 we estimate that the values of α for the two sample-to-slit separations

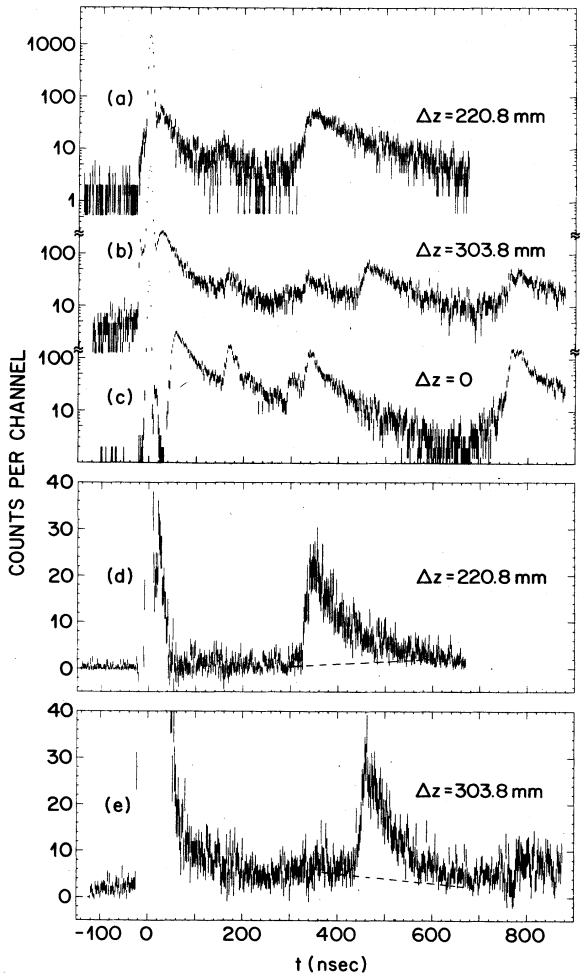

FIG. 3. Time-of-flight spectra for a clean Al(111) sample. (a), (b) Raw data for two sample-to-slit separations Δz. (c) Data obtained viewing the target directly ($\Delta z=0$) corrected for the loss of counting efficiency at later times due to the high probability of getting a count at earlier times. The prompt peak has been suppressed electronically and there is consequently a dead space just after the prompt peak. (d) and (e) Spectra obtained after subtracting a background proportional to curve (c).

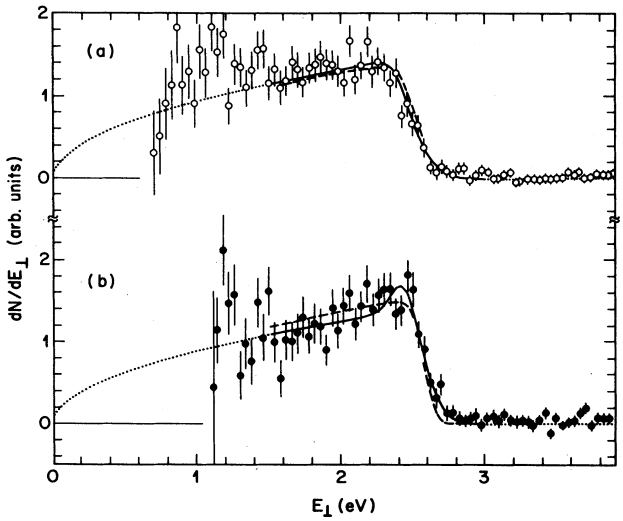

FIG. 4. Spectra of the perpendicular component of the Ps energy, $E_\perp$ from clean Al(111) deduced directly from the time-of-flight spectra of Fig. 3. The dashed curve is a fit using the free electron golden rule model. The solid curve is the fit including an adiabatic Ps peak at $E_\perp=-\phi_{Ps}$. The dotted line shows the extension of the model outside the region fitted, $1.5<E_\perp<3.0$ eV.

are $\alpha=0.230$ rad for $\Delta z=220.8$ mm, and $\alpha=0.146$ rad for $\Delta z=303.8$ mm. The model must be smoothed using the approximately Gaussian energy resolution of our apparatus at the position of the Fermi step in Fig. 4. From the 8-nsec time resolution and the 6.6-mm spatial resolution we find that the FWHM ΔE is 0.196 and 0.143 eV in Figs. 4(a) and 4(b), respectively.

A least-squares fit, shown by the dashed lines in Fig. 4, over the range 1.5 eV $<E_\perp<$ 3.0 eV with separate amplitudes for the two spectra yields $-\phi_{Ps}=(2.572\pm0.004)$ eV with a χ^2 per degree of freedom, $\chi^2/\nu=127.44/73$. The error estimate has been increased by the factor $(\chi^2/\nu)^{1/2}$. If we include a δ-function elastic Ps peak at $E_\perp=-\phi_{Ps}$, the fit, shown by the solid lines, implies that the probability for elastic emission is $(5.0\pm0.5)\times10^{-4}$ of the total Ps, the Ps work function is $-\phi_{Ps}=(2.501\pm0.010)$ eV, and $\chi^2/\nu=83.35/72$. Now the statistical error estimate on $-\phi_{Ps}$ is slightly larger because of correlation with the elastic amplitude. In both cases we have subtracted 0.025 eV from $-\phi_{Ps}$ to account for the thermal motion of the positron in the solid, since the positron work function used in calculating $-\phi_{Ps}$ is also so corrected.[10] At 300 K Ref. 10 reports that the positron work functions for two faces of Al are $\phi_+(100)=(+65\pm30)$ meV, and $\phi_+(111)=(-155\pm30)$ meV. For the same two faces the electron work functions obtained photoelectrically are[11] $\phi_-(100)=(4.41\pm0.03)$ eV and $\phi_-(111)=(4.24\pm0.02)$ eV. Subtracting $\frac{1}{2}R_\infty=6.803$ eV from the sum of the work functions we have $-\phi_{Ps}(100)=(2.548\pm0.042)$ eV and $-\phi_{Ps}(111)=(2.498\pm0.036)$ eV. Since the Ps work function is a bulk property independent of crystal orientation,[1] we take the weighted mean of the two values: $-\phi_{Ps}=(2.519\pm0.027)$ eV.

III. DISCUSSION

The χ^2/ν corresponding to the hypothesis that there is no adiabatic peak represents a four standard deviation departure from a satisfactory fit, whereas inclusion of a small peak gives a good fit. Furthermore, the Ps work function from the separate measurements of ϕ_+ and ϕ_- agrees best with the value deduced from fit that includes the peak. Nevertheless, considering that there might be possible systematic errors, it is best to conclude that we have established an upper limit of 10^{-3} on the amplitude of an adiabatic peak in the spectrum of Ps from Al (111). To include our uncertainty about which model is correct, we take as our value of $-\phi_{Ps}$ the average from the two fits to the data in Fig. 4 with an error estimate that includes both values: $-\phi_{Ps}=(2.536\pm0.035)$ eV. Our new value for $-\phi_{Ps}$ is a little smaller than that of Ref. 1 corrected for the positron temperature $-\phi_{Ps}=(2.59\pm0.04)$ eV and is in agreement with but more precise than that of Howell *et al.*,[12] $-\phi_{Ps}=(2.78\pm0.28)$ eV.

Notwithstanding our cautious conclusion, the evidence in favor of the existence of an adiabatic component is probably sufficient to justify a more sensitive measurement. A decrease of the time, angle, and spatial resolutions by factors of 2 would be needed in order to resolve and make directly visible a hypothetical peak having an amplitude of 5×10^{-4}.

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