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ELECTRON CAPTURE FROM SOLIDS BY POSITRONS

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Since the atomic system of positronium is intrinsically interesting and the electron pickup process is analogous to electron capture by other charged species, the study of positronium formation in gasses was one of the first areas of positron research. It was recognized later that positronium was formed in the interaction of positrons with solids. Electron capture by positrons from solids results in the formation of positronium in both the ground and excited states. Positronium has been observed in insulators in the solid matrix and in the vacuum emitted from metal and semiconductor surfaces.

The capture of electrons in solids is modified from that in gasses by several factors. The most important is the collective interaction of the electrons which results in a density of electron states in the solid in wide bands. Also the high density of electrons in many solids gives a high frequency of interaction as compared to gasses, and quickly destroys any electron-positron states in the metal matrix. Consequently, most positrons implanted in a metal will rapidly thermalize, and unless they reach the surface will annihilate with an electron in an uncorrelated state. Positronium formation from positrons scattered at a metal surface is analogous to ion neutralization, however, most of the positronium comes from positrons passing through the surface from the bulk. The dominant motivation for studying positronium formation has been the hope that the distribution of the electrons at the surface would be obtained through the annihilation properties of positrons trapped at the surface or through analysis of the energy and angular distributions of the positronium emitted into the vacuum.

There are three mechanisms known to emit positronium into the vacuum from metal surfaces, and they are differentiated by the state of the positron at the surface. A thermalized positron may elastically pass through the surface or trap in a surface state. Positrons may also traverse the surface with epithermal energies. Those positrons that pass directly through the surface can form positronium by capturing an electron from the filled electron bands. The energy of the positronium will then be determined by the electron and positron work functions and the energy of the electron relative to the Fermi level. If the positrons have epithermal energies, then that is added to the positronium energy as well.

Like the ejection of photoelectrons, positronium formation

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positrons passing through the surface conserves energy and total momentum in the direction parallel to the surface. This results in distinctive energy and angular distributions for the positronium emitted into the vacuum. There are two measurements that can be used to measure these properties of positronium. They are two-dimensional angular correlation of annihilation radiation, ACAR, and positronium velocity measured by time-of-flight methods, PS-TOF.

Angular correlation of annihilation radiation measurements, ACAR, provide a direct measurement of the momentum of the annihilating electron-positron pair, and can be used to describe electron densities and defect properties in bulk materials. ACAR measurements for positrons trapped at the surface and for positronium emitted into the vacuum have been performed²⁻³ with position sensitive detectors that allow the angular deviation to be measured in two dimensions. These axes for the momenta in these measurements have been chosen with one parallel to the surface of the sample and one perpendicular to that surface. From the first experiments we learned that the surface trap potential well results in positron-electron angular correlations that are very similar to those measured for trapped positrons in the bulk. The distributions were nearly isotropic. This was in contradiction to the predictions of models of the surface trap based on perfect surfaces that predicted larger angular deviations for the direction perpendicular to the sample surface. At present, the resolution of this contradiction is incomplete, but trapping at surface defects offers a plausible alternative.⁴

Angular correlation measurements were the first demonstration that the positronium formed at a metal surface obeyed the simple kinetic rules described above. If we assume that the pickup process removes an electron from a free electron gas, then we can make a qualitative description of the momentum distribution of the positronium. Good qualitative agreement was obtained for both Cu and Al. Later data on different surfaces of carefully cleaned Al showed deviations from free electron behavior that can be related to the band filling at the surface of the samples.⁵ A complete theoretical description of these measurements is now under development.⁶⁻⁷

More recently, the linac beam at Livermore⁸ has been used to perform measurements of positronium velocity distributions by measuring the time-of-flight of positronium in a vacuum. The positronium time-of-flight was determined by measuring the time difference between the positron beam arrival and radiation from the spontaneous annihilation of long lived, triplet positronium in the active volume of a highly collimated detector placed a known distance from the sample. The TOF detector collimator excludes most annihilation radiation from outside a planar volume 10.5 ± 0.35 cm from the sample collimator. Details of the apparatus can be seen below in Figure 1.

Experiments were performed in an ultrahigh vacuum chamber having 1×10^{-10} torr base pressure on samples that were sputter cleaned, annealed and Auger analyzed. The temperature of a sample could be set during the experiment from room temperature to 900°C. Positronium time-of-flight and lifetime spectra were obtained for incident positron energies from 0 to 3 keV set by the bias of the sample to the transport energy. The running time and total linac electron beam charge were recorded for each run.

Many of the established facts have been determined using positronium time-of-flight techniques. The positronium formed with positrons desorbed from the surface trap was shown to be in a thermal distribution using Ps-TOF and a bunched radioactive beam.⁹⁻¹⁰ The demonstration

Fig.1 Schematic of the experimental chamber and time-of-flight detection system.

that energy was conserved so that there is a work function in positronium production, and the subsequent supposition that positronium is formed by a positron that picks up an electron and leaves behind a single electron hole, also resulted from measurements with that beam.¹¹

With Ps-FOF measurements, using the linac beam, we can obtain high statistics spectra very quickly so that we can take good quality spectra for many sample or beam conditions. Using this tool, we have studied single-crystalline samples of several metals, and have confirmed that positronium emission by a work function process is a general property of metals.¹² This allows the determination of positive values for positron work functions that can not be measured from observation of emitted positrons.¹³ We have seen that the work function value is insensitive to surface conditions, but depends sensitively on the temperature of the sample.¹⁴

We have measured the energy distribution of positronium for energies between zero and the work function to test the relationship between the positronium and electron energy distributions. With Ps-TOF measurements, we have observed energetic positronium resulting from electron pickup by

non-thermalized positrons that have scattered out of the metal samples.¹⁶ Energetic positronium formed by transmission through a very thin carbon foil was reported in reference.¹⁷

The experimental evidence for these effects can be seen in the spectra in Fig. 2. The spectrum in Fig. 2a was taken with 2000 eV positrons incident on a clean, annealed Ni(100) surface at room temperature, and Fig. 2b was taken on the same sample at 900°C. The hot sample produced more positronium with slow flight times, i.e., low energies due to the contribution to the spectrum from thermal desorption, and the flight time of the fastest positronium is noticeably longer due to the thermally-induced shift in the work function value. In fig. 2c, the contribution from fast positronium formed by backscattered positrons dominates the spectrum obtained with a positron energy of 50 eV. The data in Fig. 2 have been corrected to reflect the velocity distribution of positronium as it leaves the sample. First, a time-independent background was subtracted and the result was multiplied by $e(t/142\text{ns})$ for the decay in flight of the positronium. A second background, due to scattered annihilation radiation from triplet positronium, was then subtracted, and the remaining data were multiplied by a factor proportional to t^{-1} to account for the velocity dependent part of the detector efficiency. After these corrections, the spectrum in Fig. 2 corresponds directly to the distribution of positronium as it leaves the sample when binned in equal increments of velocity.⁻¹ The narrow peak at the left of this spectrum arises from the detection of scattered gamma rays produced from the prompt annihilation of positrons and singlet positronium at or near the surface of the sample. The shape of the prompt peak is indicative of the overall timing resolution of the production, transport and timing circuitry, and also serves as a useful zero time marker. In many cases, this is the best representation to use to view the data as the system resolution is nearly constant as a function of time-of-flight. However, when the positronium energy distribution is desired, the time axis data may be converted into bins per unit of energy by multiplying the intensities by t^3 and plotting on an energy scale such as seen in Figs. 3 and 4.

One potential application of positronium measurements is to determine the electron density of states at the metal surface. This will be possible if a single electron hole is left behind in the metal. We have measured positronium energy distributions for several metals that are thought to have different electron density of states near the surface. The data and a model fit based on a free electron density are shown in Fig. 3. The strength and work function values were the only parameters adjusted in the model. This simple model describes the positronium energy distribution near the work function energy well, but fails to describe the spectra at lower positronium energies where more positronium is measured than predicted. The electron band structure would lead us to expect larger differences in the spectra than were observed. Near the surface, copper and aluminum are free electron like while nickel has sharp structure. The structure in the nickel bands would be seen in the data if it were transferred directly to the positronium.¹²

The energy of the most energetic positronium is a linear function of the sample temperature for all of the samples observed to date.¹⁴ The slope of the work function-temperature line is similar for all of the samples ranging from 5 to 9 meV/°K. This is remarkable consistency, and suggests that there is a simple relationship between the the temperature-induced changes in the positronium work function values and the basic structure of the metal.

Fig. 2 Time-of-flight spectra taken on a clean, well annealed Ni sample for initial positron energies of 2000 and 50 eV. In the spectrum at 900°C, the contribution from thermal positronium is seen at long times. At lower initial positron energies, the contribution to positronium formation from backscattered positrons becomes large.

Fig. 3 Energy distributions of positronium emitted from clean metal samples. The solid curves are modeled on a free electron gas density of states.

At low initial positron energies there is a large flux of non-thermalized positrons back scattering out of the sample.¹⁵ Many of these pick up an electron and form positronium with energies higher than the work function. Positronium formed in this manner will account for over half of the total amount at low initial positron energies, and is non-negligible, even at incident energies of three keV. The energy spectra of positronium formed from bombardment of lead is seen for several incident positron energies in Fig. 4. The positronium formed by work function emission is seen as a peak below one eV, and the positronium formed by pickup of non-thermal positronium is seen at higher energies. There is more higher energy positronium at low incident positron energies since there are more positrons backscattered from the sample at energies that efficiently form positronium. The positronium

Fig. 4 Positronium energy distributions taken on a clean, well annealed Pb sample for initial positron energies of 3000 to 25 eV showing the E^{-2} variation in high energy positronium formation.

intensity appears to depend on the positronium energy by a power law relationship near E^{-2} . The exact power of energy differs for different positron beam energies, but an exponent of -2 is a close value for all cases. We could obtain the value of the pickup matrix element if these data were combined with careful calculations and measurements of the epithermal positron emission with matching momentum limits.

The energy and sample dependence of the total intensity of positronium formed by backscattered positrons can be seen in Fig. 5. All samples have less high energy positronium formed at high incident positron energies, but there are sample-to-sample variations. Epithermal positrons produce more positronium from aluminum at low incident energies and gold produces more at high incident energies.

Due to the general similarities in the scattering functions, the number of positrons that backscatter at some incident positron energy will be closely related to the number of electrons that backscatter at the same incident energy. Since there are extensive data available for the total backscattering fraction, B , for electrons we have developed a simple model for scaling the positronium formation due to epithermal, backscattered positrons to the total electron backscattering coefficient. Positronium is only efficiently formed by positrons that have relatively low energies compared to the incident positron energy and the energy spectrum of backscattered positrons or electrons spans the whole range from the incident energy down to thermalization. This suggests that the simplest scaling of positronium production to the backscatter coefficient is obtained by taking the fraction of positrons backscattering with low energy to the total. For simple energy distributions of backscattered positrons, this fraction can be approximated by dE/E for a fixed, small value of dE . This results in an E^{-1} dependence of the positronium intensity with incident positron energy if the backscatter coefficient is slowly varying. The positronium produced from backscattered positrons would then be proportional to BdE/E measured for electrons. Positronium intensities calculated using this model normalized at one point are shown in the inset for Fig. 5.

There is a remarkable correspondence between the results of the model scaled to electron data and our measurements. The good performance of this model indicates that the positron pickup matrix element does not vary much from sample-to-sample, and that the variations in the positronium production are the result of differences in the flux of epithermal positrons. Also the ratio of positrons emitted bare to those forming positronium is nearly constant for all positrons leaving the sample. This suggests that the pickup matrix element is slowly varying with positron energy, and that the energy distribution of the epithermal positrons sets the positronium energy distribution.

The model will be an assistance to those researchers that wish to use positronium fraction measurements to indicate positron diffusion to the sample surface in diffusion or defect profiling studies. Failure to account for the contribution from backscattered positrons can lead to large differences in the parameters derived from the data. Using available electron backscattering coefficients positronium fraction values can now be corrected to remove the contribution from non-thermal positrons at the surface of many samples.

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Fig. 5 Total strength of the high energy positronium distribution for several samples.

REFERENCES

1. D. W. Gidley, R. Mayer, W. E. Frieze, K. G. Lynn, Phys. Rev. Lett. **58**, 595 (1987).
2. K. G. Lynn, A. P. Mills, Jr., R. N. West, S. Berko, K. F. Canter, L. O. Roellig, Phys. Rev. Lett. **54**, 1702 (1985).
3. R. H. Howell, P. Meyer, I. J. Rosenberg, M. J. Fluss, Phys. Rev. Lett. **54**, 1698 (1985).
4. A. R. Koymen, D. W. Gidley, T. W. Capehart, Phys. Rev. B **35**, 1034 (1987).
5. D. M. Chen, S. Berko, K. F. Canter, K. G. Lynn, A. P. Mills, Jr., L. O. Roellig, P. Sferlazzo, M. Weinert, R. N. West, Phys. Rev. Lett. **58**, 921 (1987).
6. A. B. Walker and R. M. Nieminen, J. Phys. F **16**, L295 (1986).
7. A. Ishii and S. Shindo, Phys. Rev. B **35**, 6521 (1987).
8. R. H. Howell, M. J. Fluss, I. J. Rosenberg, P. Meyer, Nucl. Inst. Meth. **B10**, 373 (1985).
9. A. P. Mills, Jr., and L. Pfeiffer, Phys. Rev. B **32**, 53 (1985).
10. A. P. Mills, Jr., L. Pfeiffer, Phys. Rev. Lett. **26**, 1161 (1979).
11. A. P. Mills, Jr., L. Pfeiffer, P. M. Platzman, Phys. Rev. Lett. **51**, 1085 (1983).
12. R. H. Howell, I. J. Rosenberg, M. J. Fluss, R. E. Goldberg and R. B. Laughlin, Phys. Rev. B **35**, 5303 (1987).
13. R. H. Howell, I. J. Rosenberg, M. J. Fluss, P. Meyer, Phys. Rev. B **35**, 4555 (1987).
14. I. J. Rosenberg, R. H. Howell, M. J. Fluss, Phys. Rev. B **35**, 2083 (1987).
15. R. H. Howell, I. J. Rosenberg, M. J. Fluss, Phys. Rev. B **34**, 3069 (1986).
16. A. P. Mills, Jr., and W. S. Crane, Phys. Rev. A **31**, 593 (1985).
17. H. J. Fitting, Phys. Stat. Sol. **a26**, 525 (1974).