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POSITRONIUM EMISSION SPECTROSCOPY

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POSITRONIUM EMISSION SPECTROSCOPY

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Abstract

Measurements of the intensity, velocity, and angular distribution of positronium emitted from solid samples of metals and insulators have been performed using the intense, pulsed positron beam from the 100 MeV electron linac. From these data it is possible to determine properties of both the surface interactions and volume potentials of the materials studied. Examples of these effects will be given using measurements of positronium time of flight performed with the Livermore intense positron beam. The time of flight data have been augmented by positron lifetime and angular correlation measurements performed with the beam. Measurements resulting in workfunctions, deformation potentials and surface interaction effects will be reported for both metals and insulators. In this paper we will review the results of experiments conducted on metal surfaces with the Lawrence Livermore National Laboratory intense positron beam. Results on insulators are covered in a separate report in this volume. Positron energy loss and positronium production differ between metals and insulators due to the effects of the large band gap and low electron density in insulators. A more extensive review of this work may be found in reference 1. In metals positrons reach full thermalization through electron hole pair formation and only bare positrons are stable on time scales long enough for diffusion to take place.

In metals positrons that pass directly through the surface can trap, pass into the vacuum or form positronium by capturing an electron from the filled electron bands. The positronium escapes into the vacuum with an energy 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 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 are used with a positron beam to measure properties of positronium: They are two dimensional angular correlation of annihilation radiation 2D-ACAR, and positronium velocity measured by time of flight methods, PS-TOF.

2D-ACAR measurements provide a direct measurement of the momentum of the annihilating electron-positron pair a plane usually chosen normal to the sample surface. These measurements have provided good evidence that the kinetic rules described above are obeyed and that qualitative agreement is obtained between a free electron model and the data for Cu and Al[2,3]. Later data on separate Al surfaces have shown that Ps angular distributions can be related to band filling at the sample surface[4]. Detailed calculations have been performed on these data[5,6].

More recently at Livermore, the intense, pulsed positron beam[7] has been used to measure the time of flight of positronium in a vacuum over a known distance. The time difference between the beam pulse and the annihilation of triplet positronium is detected in a highly collimated detector. Details of the apparatus can be found in references 1 and 7.

Much of what is known about positronium has been determined using PS-TOF Thermal positronium desorption from surface traps was found using PS-TOF and a bunched radioactive beam [8,9]. Also conservation of energy was demonstrated, the positronium workfunction established and single step electron pickup in positronium formation was postulated from results of measurements with that beam [10].

With PS-TOF measurements using the linac beam we can quickly obtain high statistics spectra. 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 [11]. Thus positive values for positron workfunctions can be determined with this technique[12]. We have seen that the positronium workfunction is insensitive to surface conditions but does depend sensitively on the temperature of the sample [13].

We have measured the positronium energy distribution to test the relationship between the positronium energy and electron energy distributions. With PS-TOF measurements we have observed energetic positronium resulting from electron pickup by nonthermalized positrons scattered out of the metal samples[14]. The effects observable in PS-TOF can be seen in figure 1. The data in figure 1 have been corrected for effects of decay in flight and detector efficiency to reflect the velocity distribution of positronium as it leaves the sample. These corrections are completely described elsewhere[1]. The prompt narrow peak at the left of this spectrum arises from the detection of scattered gamma rays produced from the prompt annihilations.

The spectrum in the top frame was taken with 2000eV positrons incident on a clean, annealed Ni(100) surface at room temperature and the middle frame 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 of surface trapped positronium. The flight time of the fastest positronium is noticeably longer at high temperatures due to the thermally induced shift in the workfunction value. In the bottom frame the contribution from fast positronium formed by backscattered positrons dominates the spectrum obtained with a positron energy of 50eV. application of positronium measurements that we explored 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 have different electron density of states near the surface. The data and a model fit based on a free electron density are shown in figure 2. The electron band structure would lead us to expect larger differences in the spectra than were observed. Near the Fermi 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 [11]. More detailed calculations based on the electron bands that also include the effects of electron screening at the surface and higher order breakup and formation terms are able to give a good description of the Ni data[15].

The present data from PS-TDF and 2D-ACAR suggest that while the existence of surface band gaps and other similar features can be viewed with positronium spectroscopy the values of the inner potentials, i.e. the positron zero point energy and electron Fermi energy can be accurately monitored. However detailed information about the population of electron states appears to be obscured by the effects of multielectron and multistep interactions.

The energy of the most energetic positronium is a linear function of the sample temperature for all of the samples observed to date [13]. The slope of the work function-temperature line is similar for all of the samples ranging from 5 to 9 meV/ 9 K. These data can be approximated by a simple relationship between the volume change with temperature and the positronium work function using the deformation potential[16]. Complete calculations of positronium workfunctions, temperature variations and deformation potential values have been published in reference[17].

At low initial positron energies there is a large flux of non-thermalized positrons back scattering out of the sample [14]. Many of these pickup an electron and form positronium with energies higher than the work function as seen in the bottom frame of figure 1. 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. This extra positronium presents a difficulty for the execution of back-diffusion experiments that are based on total positronium yield[18]. The energy spectra of positronium formed from backscattered positrons has been measured for lead and other metals. The positronium intensity appears to depend on the positronium energy by a power law relationship of approximately E^{-2} .

The variation of the intensity of the high energy positronium with sample and positron incident energy was studied for samples from A1 to Au. Good agreement between positronium intensities and electron or positron backscattering data can be obtained by scaling the low energy fraction of positronium backscattering 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. The positronium produced from backscattered positrons would then be proportional to BdE/E where B is the backscattering coefficient measured for electrons and varies slowly with electron bombarding energy for most elements.

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Figure 1. PS-TOF spectra for Ni.



Figure 2. Positronium emission energy spectra for several metals.