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NEW EXPERIMENTS WITH BRIGHT POSITRON AND POSITRONIUM BEAMS

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Abstract: After half a century of work studying the interaction of positrons with ordinary matter, there remain several interesting fundamental questions that are uniquely suitable for investigation using antimatter, and several instances where interesting views of the world could be obtained using positrons. In this paper we examine the possibilities for imaging single molecules with positrons and for looking at a possible zero-temperature or quantum phase transition associated with the sticking of slow positronium at a cold surface. We also discuss prospects for future developments of LINAC positron beams using a rare gas moderator and for detecting positronium molecules.

1.   OVERVIEW

The field of positron physics includes several diagnostic techniques that have become useful, although not widely acclaimed, in certain niches under the skilful hands of some talented experimenters [1,2]. The positron is a strongly perturbing probe in many cases, but after much work we have good theories of positron interactions with matter and the computers necessary for calculations corresponding to realistic experimental conditions are available. The successful niches include areas such as defects in solids, plasma physics and surface physics that are justified in their existence by good connections to other active fields of science and technology. But for a lack of intensity, beams of low energy positrons might join their cousins (photon, neutron and electron beams) as valuable and unique probes of materials. The invention of brightness enhancement and various proposals for new positron facilities including a nano-ampere positron microprobe for the Stockpile Stewardship Program at the Lawrence Livermore National Laboratory, continue to cause dreams of a new era

in antimatter physics. In this contribution we examine some potentially interesting experiments that would be fun to do some day.

2. IMAGING OF SINGLE MOLECULES

Examining individual molecules at an atomic length scale is one of the most challenging and interesting problems facing the physics community. X-rays, which are now available in the form of microbeams, would seem to be a natural candidate for such measurements, but the cross sections are much too small. Electrons and positrons are other possible probes. Although there has been very little thought given to positrons for imaging individual molecules, we believe they have some definite advantages and would like to discuss a possible scenario.

The configuration we envisage is shown in Fig. 1. The molecules of interest are to be deposited randomly, one roughly every fraction of a micron, onto an appropriate metal foil having a negative affinity for positrons. Nickel would be a good choice for e^+. The Ni foil should be about 10^3Å thick. A few keV, focused (1 μm), pulse of e^+ containing 10^6 positrons is incident on the area containing the molecules of interest. As we know the positrons stop and rapidly slow down to thermal energies. They then diffuse to the surface where roughly ten percent, because the e^+ work function for Ni is negative, are remitted from the surface.
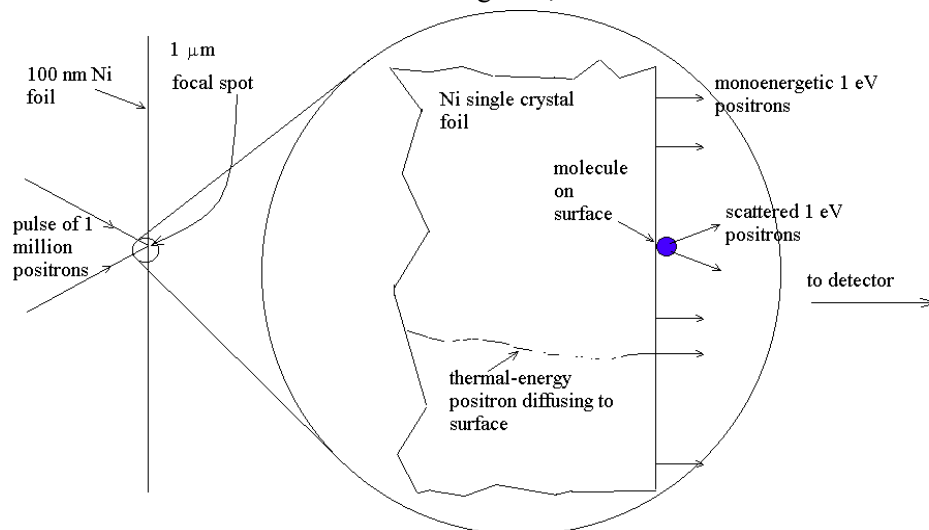

Figure 1. Experimental configuration for observing the structure of single molecules.

When there is no molecule on the surface, roughly one quarter of the e^+ arriving at the surface rolls out from the solid emerging with the maximum kinetic energy ($\cong$1ev for Ni). At the temperature of interest, i.e. a few degrees Kelvin, these e^+ have a transverse coherence length of at least 100Å. Another

quarter scatters inelastically by exciting electron hole pairs in the metal and emerges from the solid with a broad spread of energies.

When a molecule is present, the elastic laterally coherent beam of remitted e^+ will be scattered from the atoms that make up that molecule. The resulting speckle pattern can be displayed on a screen placed a few centimeters from the foil. The speckle pattern will have all the information necessary to reconstruct, with a bit of computation, the full 3D image of adsorbed molecule and whatever else is on the surface in the roughly 1μ spot illuminated by the e^+ reemission beam.

To see if any of this is realistic, we first need some estimates of count rates, i.e. of the time necessary to accumulate a sufficient number of counts for image reconstruction. A one eV e^+ has a wave vector $k=5.12\times10^7$ cm^{-1}, i.e. $\lambda_{dB}/2\pi=1.95$Å. Experiments by Canter and co-workers [3] have demonstrated that e^+ phase shifts are large and fairly universal. This means that the $l=0$ e^+ atomic elastic scattering cross sections at 1ev are at most $\sigma_{EA}=4\pi/k^2 = 4.79\times10^{-15}$ cm^2. For a molecule containing $N \cong 200$ atoms, we need to measure the speckle pattern at roughly 10^3 points with about one percent statistical accuracy. This means we need about 10^7 total counts. The total geometric cross section of the molecule $\sigma_{ET}\approx10^{-13}$ cm^2 then implies that a pulse with a radius of 10^{-4}cm, producing 10^5 elastically emitted cold 1 eV positrons will yield roughly one e^+ scattered into the entire speckle pattern. If there are 10^3 pulses/sec we will accumulate our 10^7counts in 10^4 sec. This, of course, assumes we use an efficient large solid angle detector.

While the time, the geometry, etc., all seem reasonable, we still have to concern ourselves with the fact that the molecule may be destroyed before the 10^4 seconds has elapsed. Its well known that the cross section for 1ev e^+ annihilation with the outer valence electrons is about 10^{-19}cm^2, i.e. about 10^{-4} of σ_{EA}. This means that each atom in the molecule would suffer about 10 annihilations in the course of the 10^4 seconds of intense bombardment. Under normal circumstances this number of annihilations would completely destroy the molecule. However, we believe that in this case it will not happen because of the close proximity to the metal. A suddenly annihilated valence electron leaves us in an electronically and vibrationally excited state of the molecule. If the electronically created hole is in fact filled in a time short compared to some vibration time, $\cong10^{-11}$ second, of the molecule, then the molecule will not be destroyed. More precisely we have returned back to the neutral molecule without exciting many groundstate vibrational modes and the molecule remains intact.

We believe this is indeed what happens. Since the suddenly produced state is clearly not an eigenstate, it will evolve in time. This means it will sample all atomic locations in a time determined, roughly speaking, by the spread of energies in the discrete set of valence levels near the last occupied ones. This spread is probably volts, but surely no smaller than 0.1 eV which corresponds to a cycle time of 10^{-13} to 10^{-14} sec.

When the hopping hole excitation gets close, perhaps 5 Å, to the metal, electrons from the metal will immediately tunnel in and heal the molecule. An

estimate of the tunnel time is obtained by noting that the attempt frequency of an electron at the Fermi energy is roughly $10^{-15} - 10^{-16}$ sec^{-1}. The penetration under a barrier a few eV high and 5 Å wide is perhaps 10^{-2} to 10^{-3}. All this implies that the time to fill the hole is 10^{-13} to 10^{-14} sec, i.e. fast enough to keep the molecule from being damaged. Of course, roughly the same mechanism exists for electrons, x-rays, light, etc. A bulk piece of metal thus seems to prevent damage to simple molecules of reasonable size on its surface. Of course it is possible that the hole becomes localized, i.e. the molecule distorts and traps the hole somewhere removed from the metal surface, but this too takes a vibration time. In such cases we will see speckle patterns from the distorted molecule. It will be interesting to see if this type of annealing actually works.

3.   QUANTUM STICKING

In the past decade, low-energy surface adsorption has been a subject of renewed interest. The central issue remains to determine the behavior of the sticking probability at low incident energies. Any classical description of sticking, i.e. a ball rolling down hill with some dissipation, says the sticking coefficient must be unity as the energy of the particle approaches zero. The quantum description is completely different. When the de Broglie wavelength λ_{dB} of the particle is large compared to the characteristic length λ_c of the surface potential, the amplitude of a noninteracting particle's wavefunction at the surface is proportional to $k_\perp$, the component of the particle's momentum perpendicular to the surface. Thus as E→0, the particle reflects from the surface. This happens even for rather smoothly varying surface potentials because of the wavelength mismatch inherent in the E=0 limit.

This quantum reflection suppresses the sticking probability even assuming there is a *weak* coupling to some inelastic channel (phonons, particle-hole pairs, etc.). High-order virtual processes renormalize the wave function near the surface, increase the effective mass of the particle, and generally enhance the possibilities for sticking. However the sticking probability still goes to zero, albeit more slowly than before, as $E \to 0$. In fact, it has recently been suggested that for a dimensionless coupling, to the inelastic channel, of order unity there is a "phase transition" from perfect reflection to perfect sticking [4,5]. This phase transition, if it exists, would be an extremely interesting example of a zero-temperature quantum critical point. Fluctuations near such a critical point, the values of the critical exponents and the magnitude of the coupling constant are all open questions. Needless to say it would be interesting to experimentally investigate such questions. To do so it is necessary to find a simple physical system with $\lambda_{dB}/\lambda_c \gg 1$ and with strong coupling to low-energy inelastic channels.

Carraro and Cole [6] show that quantum reflection may in fact cease for certain long range attractive potentials and particle energies below a threshold

energy such that the WKB approximation holds, i.e. the adiabatic condition | $dk(z)/dz| \ll k^2(z)$ holds. The elastic potential considered is

$$V(z) = -c_n/z^n, \ z>a,$$

where *a* is a small distance cutoff at the surface. For *n<2*, the adiabatic condition is always satisfied and there is no quantum reflection, irrespective of the strength of the potential. However, for *n>2*, Carraro and Cole find that "as the incident energy is lowered towards zero, quantum reflection will always set in, although [the] energy threshold may be extremely small." Indeed, for ordinary atoms with masses at least as great as that of atomic hydrogen, the threshold energy is less than a few neV. However, the case of positronium seems to be qualitatively different because of its small mass.

We now estimate using Carraro and Cole's formulae, the magnitude of the energy threshold for positronium atoms incident upon a metal surface. The Ps-surface van der Waals potential has the form of the above equation with *n=3* and is roughly given by $V(z) = 2e^2 a_{Ps}^2/z^3$, where a_{Ps} is the positronium Bohr radius and the length unit *a* is taken to be 2 a_{Ps}. The unit of energy is 1/16 a.u. = 1.700 eV and the coefficient $c_3=8$. The Carraro and Cole criterion for quantum reflection is that the incident wave vector be smaller than a characteristic value, which for *n=3* is

$$k_c = 4^5 3^{-6} c_3^{-1} = 0.175$$

The corresponding energy threshold below which quantum reflection sets in is

$$E_c = k_c^2 = 52 \text{ meV}.$$

The experiments of Mills et al. [7,8] show that the energy spectra of positronium with energies less than 50 meV emitted from cold Al(111) surfaces in fact have an exponential dependence that is consistent with the sticking coefficient of Ps being a non-zero constant, in contradiction with the Carraro and Cole energy threshold E_c.

Since λ_{dB} depends inversely on the square root of the product of the mass of the particle and the energy, in order to ensure that we are in the quantum regime of surface adsorption, since $\lambda_c \cong 1\text{Å}$, we have a choice between two possibilities: we can either use atoms, and cool them to ultralow temperatures, or work with much lighter particles (electrons), at reasonable temperatures $k_B T \approx 100K$. In the scattering experiments of Nayak et al. [9], they chose the first option, using ^{4}He atoms incident on a ^{4}He fluid surface. They observed a dramatic increase of the specular reflection coefficient upon reducing the normal component of the incident particle momentum. Berkhout et al. [10] chose the same option, this time using spin polarized atomic hydrogen atoms on a concave mirror coated with ^{4}He, at temperatures of the order 0.01 K. Adsorption coefficients as low as 0.2 were measured for the first time in a direct scattering experiment. In these

experiments estimates show that the dimensionless coupling to inelastic channels is very weak, so the lack of sticking is not surprising.

Estimates suggest that slow Ps reflecting from metallic surfaces coated with a partial monolayer of rare gas atoms could be an ideal system for such experiments. Because of its light mass, i.e. four orders of magnitude smaller than that of He, energies in the range of 10-100 K are sufficient to achieve λ_{dB}/λ_c ≈100. In addition there is very good evidence that coupling to the inelastic channel, i.e. electron hole pairs in the metal, is in the strong coupling limit. The potential between the Ps atom and surface at large distance is clearly Van der Waals. At short distances, Ps completely loses its identity. Experiments show that within a few angstroms of the surface we can think of the e^+ either as a bare particle bound by several eV or as a Ps atom bound by a fraction of eV. This duality implies strong coupling. Preliminary experiments involving the desorption of Ps indirectly from clean and partially oxidized Al (111) surface were interpreted as evidence for unity sticking coefficients [7,8].

We propose to measure the Ps sticking coefficient directly using a beam of cold Ps atoms directed at well-characterized surfaces at cryogenic temperatures. Possible configurations for this experiment are indicated in Figs 2 and 3. In the first experiment slow positronium atoms emitted from a cold Al target crystal travel several mm to a clean and cold Al sticking surface. If the positrons stick to the surface, they quickly equilibrate as a positron surface state. This state has a 0.5 nsec mean lifetime [11] determined by the annihilation rate of the surface positron. A few percent of the time the positron annihilates with an Al core electron, which thus results in the emission of an Auger electron with a kinetic energy of 68 eV [12]. The 1 kG solenoidal magnetic induction is sufficient to confine the Auger electrons to a helical orbit 5.6 mm diameter. The Auger electrons are quickly attracted to the surface of a position sensitive channel electron multiplier array detector. The time of arrival at the detector relative to the time of the pulse of 10^5 positrons at the cold Al target crystal tells us the positronium time of flight to reach the Al sticking surface. The position information from the detector may then be used to infer the positronium velocity and angle of incidence on the sticking surface. The second configuration (Fig 3) uses an electrostatic positron beam and gamma ray detection.

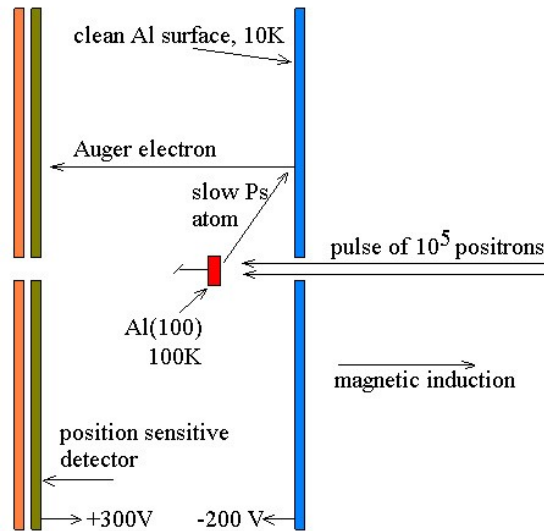*New experiments with bright positron and positronium beams*

Figure 2. Configuration for measuring the slow positronium sticking coefficient as a function of incident angle and positronium kinetic energy. Slow positronium that sticks to the clean Al surface [30 mm diameter] will annihilate a few percent of the time with a core electron, giving rise to an Auger electron. The electron is swiftly transported to a position-sensitive detector whose output signal is used to infer the time of arrival and position of the positronium at the Al surface.
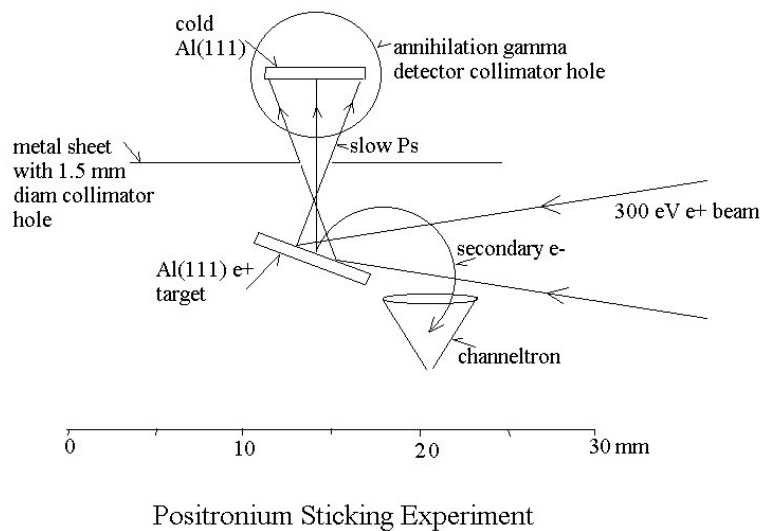

Positronium Sticking Experiment

Figure 3. Alternate configuration for measuring the slow positronium sticking coefficient in a magnetic field-free region.

If the results of our preliminary experiments are verified, we will also measure the sticking coefficent of Ps as the surface is slowly covered by a submonolayer of rare gas atoms. The randomly placed rare gas atoms will gradually weaken the Ps interaction with the surface until it is no longer able to support the sticking effect. It will be of the greatest interest if we can observe a sudden transition from sticking to non-sticking at a threshold gas density. A measurement of the sticking coefficient as a function of angle could also reveal the velocity dependence of the Ps interaction with the surface.

The proposed Ps experiments will be able to examine a fundamental nonperturbative quantum phenomenon that is inaccessible in ordinary experiments. Thus, while cold atoms may be produced with neV kinetic energies, it would be very difficult to engineer a controllable many body surface potential that would be expected to cause quantum sticking. For example, a short ranged interaction can be set up by evanescent light waves at the vacuum interface of a dielectric medium, but the inelastic part of the interaction does not have the polaronic character necessary for a quantum sticking effect. On the other hand, a reactive atom interacting with a metal surface might satisfy the latter requirement, albeit without a propitious channel for momentum transfer to the surface. Furthermore, it will be equally difficult to arrange target surfaces that have matching low temperatures in the μK range that would allow an unambiguous demonstration or refutation of quantum sticking to be established.

4.   POSITRON MACROPULSES

4.1   Introduction

The basis for many new experiments using positrons would be the availability of large bunches of positrons compressed to a small volume and occurring in a time short compared to the annihilation lifetime in the relevant system. We define a macro positron pulse to be a nsec burst containing more than 10^6 positrons focused to a microscopic spot. The development of a macro-pulse positron capability would open the door to the science of many positron effects and to high precision atomic physics experiments on positronium. It would also enable single-shot measurements to be made on transient phenomena in highly stressed solids such as laser ablated or exploding targets and photoactivated chemical reactions occurring near surfaces.

To obtain suitable macropulses one could equip a LINAC positron source with a solid rare gas slow positron moderator to enable the production of more intense bursts of positrons than presently possible. The experiments would also require the installation of various positron traps, bunchers and brightness enhancement stages for making intense bursts of highly focused positrons. One could then study positronium (Ps) molecules such as Ps_2 and measure the free-fall acceleration of positronium. In conjunction with the latter, a high precision

measurement of the 1S-2S interval of positronium would also be possible. Eventual extension of this work to a high intensity polarized positron facility would permit us to study the positronium Bose-Einstein condensate, discussed elsewhere in this volume. It might be best to do such an experiment using a N^{13} source, which gives polarized positrons, instead of a LINAC.

4.2   LINAC Target

The first step in making slow positrons is to make a shower by irradiating a high-Z converter target with 150 MeV electrons from the LINAC [13]. For this energy of primary electrons, the target should be about three radiation lengths of a material such as tungsten or a high-Z liquid metal. A relatively non-toxic alloy such as the binary eutectic 55.5%Bi-44.5%Pb with a melting point of 124 ºC might be a good choice for the latter if it is necessary.

We assume a LINAC power of 10 kW and a pulse rate of 100 pulses/sec. Each 100 J pulse will then contain 4×10^{12} electrons. After three radiation lengths, half the pulse energy is absorbed by the converter and the other half is in a 50 J shower containing about eight times more particles than in the primary pulse, or 3×10^{13} particles, equally distributed amongst electrons, positrons and photons with energies of order 10 MeV [14]. At the exit face of the converter the specific energy loss rate for a 1-cm diameter beam is 10 J/g which only raises the temperature of room temperature W by 100 ºC per pulse. The problems are to dissipate the average power of 5 kW and to minimize the damage caused by the acoustic shock waves at the target surface.

4.3   Slow positrons

Slow positrons at a LINAC are currently obtained using a tungsten vane moderator [15] that is very robust and hence well suited to the severe conditions at a high intensity target. In other applications the tungsten vane moderator is about one hundred times less efficient than is a solid rare gas moderator [16]. Using reports from the LLNL LINAC of more than 10^7 slow positrons per pulse [17], we estimate a yield of at least 10^8 slow positrons per pulse could be obtained from a solid rare gas moderator.

The slow positron moderator will be an 8 g, 1 cm diameter, 1 mm thick gold cup coated with solid Ar [18] at the end of a thin-wall stainless steel tube shown in Fig 4. The vapor pressure of solid Ar is objectionably high above 45 K and the
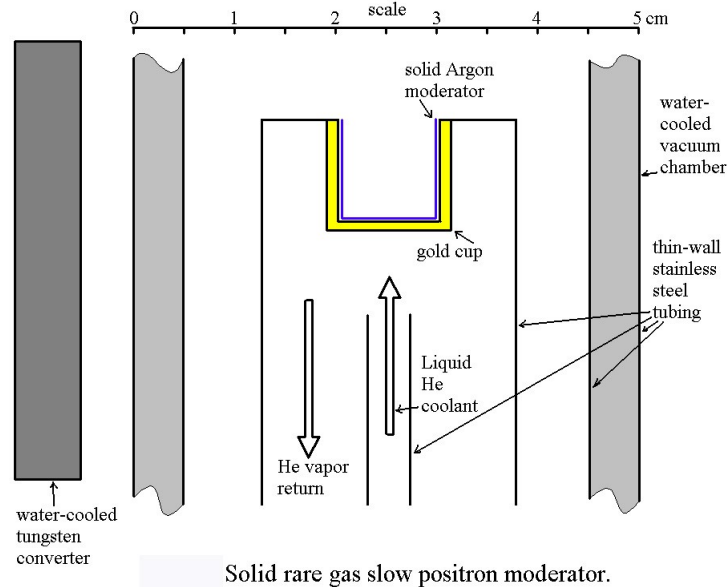

Solid rare gas slow positron moderator.

Figure 4. Solid rare gas moderator suitable for use with a LINAC.

enthalpies of Au and solid Ar are 1 J/g at 43 K and 15 K respectively. We must therefore limit the energy absorption of the moderator to less than about 1 J/g to prevent its temperature from rising above 45 K. To limit the energy absorption to1 J/g per pulse, the 50 J shower must be about 3-cm diameter at the position of the moderator. A 1 kW He liquifier circulating about 500 liters per hour can remove the roughly 10 J per pulse.

4.4 Positron macro-pulses

We suppose we are given 100 bunches per sec in 10 nsec pulses containing 10^8 positrons each. The mean positron kinetic energy is assumed to be 1 keV and the beam confined to a 1-cm diameter by a 100-G solenoidal magnetic induction. To improve its phase space density, the positron beam is brightness-enhanced [19,20] in two stages as follows. First the positrons are accelerated to 10 keV, focused on the back of a few mm diameter self-supporting field excluding 1000 angstrom-thick film of niobium at a temperature below its superconducting transition. The foil is supported over a hole in a field-terminating Permalloy sheet, the thickness of which increases proportionally to the radius. The other side of the film is coated with a layer of Ne or Ar that remoderates the positrons in a 2-mm diameter spot with 30% efficiency. The positrons are now in a magnetic field free region ready for a second stage of brightness enhancement.

The positrons are accelerated again to 5 keV and remoderated in a 300 μm spot by a second brightness enhancement remoderator, a cold (100 K) single crystal Ni(100) surface [21]. The reemitted particles are now in pulses of 6×10^6 positrons having an energy spread of about 30 meV. The positrons are accelerated to 3 keV and may now be focused to a surface density of 2×10^{12} positrons per cm^2. The focused spot may be either 20 μm diameter or a line focus of vertical width equal to 1 μm and horizontal width 300 μm.

5.   FORMATION OF POSITRONIUM MOLECULES

5.1   Introduction

When positrons are present at a high enough density near a solid surface, they will interact significantly with each other and the solid, revealing the new physics of the many positron-many electron system. As envisioned in 1946 by Wheeler [22], the polyelectron states having several positrons and electrons are known to form stable bound states up to the positronium molecule Ps_2 [23]. Positronium was first observed in 1951 [24] and the positronium negative ion in 1981 [25]. Due to advancing techniques, including positron beam brightness enhancement and the availability of intense pulses of positrons, we are now in a position to form and study the di-positronium molecule Ps_2. The necessary experimental conditions of high positron density will also allow us to fill a small cavity with Ps atoms at a density such that Bose-Einstein condensation could occur at room temperature if polarized positrons were available [26]. The positronium super fluid will be a complex system with possible distinct phases associated with the triplet and singlet positronium ground states. It seems unlikely that the fluid will condense in the absence of a container, and thus that Wheeler's vacuum polyelectron series may perhaps terminate with the Ps_2 molecule or its dimers.

5.2 Ps_2 formation

The binding energy of Ps_2 relative to two free Ps atoms is $\Delta E = 0.411$ eV relative to two free Ps atoms [27]. Since the activation energy for thermally desorbing Ps from its surface state on a metal is typically about 0.5 eV, Ps_2 emission will occur at temperatures below those needed for the thermal desorption of Ps atoms. The unknown factor in the formation rate of Ps_2 will be the accommodation or sticking coefficient S_{Ps2} for Ps_2 absorption at the surface. The Ps_2 formation rate [28,29] per surface positron is proportional to the positron surface density since two positrons are involved in the reaction:

$$z_{Ps_2} = (hmn_+ / m_+^2)S_{Ps_2}[\theta\{2E_a - \Delta E\}\exp\{(\Delta E - 2E_a)/kT\} + \theta\{\Delta E - 2E_a\}]$$

Here, m_+ is the surface positron effective mass, m is the free electron or positron mass, n_+ is the surface density of positrons, S_{Ps2} is the thermally averaged Ps_2 sticking coefficient, E_a is the activation energy for the thermal desorption of surface positrons to form positronium and $\Theta\{x\}$ is the Heaviside unit step function. The ratio of the rates for forming Ps_2 *vs*. Ps is [29]

$$z_{Ps2}/z_{Ps} = (n_+/n_0)(S_{Ps2}/S_{Ps})\exp\{(\Delta E - E_a)/kT\} ,$$

provided $E_a \geq \Delta E/2 = 0.205$ eV. The characteristic surface density is $n_0 = 8\pi m_+ kT/h^2 = 2.5\times10^{13}$ cm^{-2} at room temperature and with $m_+=m$. An Al(111) sample surface desorbs Ps with an activation energy [30] $E_a=0.34$ eV and about 50% of the maximal thermal Ps yield at a temperature of about 500 K. At that temperature, the Ps_2 yield would be equal to the thermal Ps yield (each being 25% of the maximal thermal Ps yield) for $n_+ = 8\times10^{12}$ cm^{-2}. A lower requirement on n_+ would result from treating the Al(111) surface with oxygen [7,31].

5.3 Ps_2 detection

Ps_2 annihilates predominantly into two sets of two photons with a rate per pair of photons very close to the spin-averaged Ps annihilation rate of 2 nsec^{-1}, with a branching probability into two sets of three photons of about 0.26%. There will also be a small probability for decay into two annihilation photons plus a free electron-positron pair having kinetic energies on the order of a few eV. One may thus discern the occurrence of Ps_2 formation by measuring the ratio $R_{3\gamma}/R_{2\gamma}$ of three-photon to two-photon annihilations *vs*. positron surface density, or equivalently positron focus spot size. The ratio $R_{3\gamma}/R_{2\gamma}$ may be found from the relative pulse heights from two scintillation detectors, one of which is shielded from the lower energy 3γ events by a few mm of Pb, and the other of which is thin and mostly sensitive to only the 3γ events. The counters may be placed as close to the positron target as possible, since one wishes to detect multiple events due to the near simultaneous annihilation of a burst of many positrons.

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