

## Viewpoint

## In tight spaces

Toshio Hyodo

*Slow Positron Facility, Institute of Materials Structure Science, High Energy Accelerator Organization, KEK, 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan*

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*Laser spectroscopy of positronium confined to nanoscale pores is a tool to probe the size of buried cavities and a step toward the long-term goal of a positronium BEC.*

Subject Areas: **Atomic and Molecular Physics**

## A Viewpoint on:

**Cavity Induced Shift and Narrowing of the Positronium Lyman- $\alpha$  Transition**

D. B. Cassidy, M. W. J. Bromley, L. C. Cota, T. H. Hisakado, H. W. K. Tom and A. P. Mills

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Positronium (Ps) is the bound state of an electron and its antiparticle, the positron [1]. It is made purely of leptons and, as such, provides a unique opportunity for studying bound-state, two-body quantum electrodynamics and the effect of virtual annihilation that occurs between a particle and an antiparticle in a bound state.

So far, experimentalists performing precision measurements [2] of the energy levels and the intrinsic lifetime of Ps have made every effort to eliminate the effects of the environment so they could compare their results with theoretical predictions [3]. In a paper appearing in *Physical Review Letters*, David Cassidy and colleagues at the University of California, Riverside, and San Diego State University present a spectroscopic study of the Ps atom in a completely different situation [4]. They measure the line shape of the Lyman- $\alpha$  atomic transition (the 1S-2P transition) of Ps confined within the roughly 5 nm diameter pores of a porous silica film. This is the first time that spectroscopy has been done on atoms inside such small pores, and shows that the 1S-2P transition energy of Ps confined to the nanometer length scale is significantly shifted compared to “free” Ps. One of the long-term goals of studying Ps in confinement is to produce a positronium Bose-Einstein condensate (BEC). With positronium’s small mass (and long de Broglie wavelength), it should be possible to form a Ps BEC at higher temperatures than with more massive atoms like sodium or rubidium [5]. There are, of course, obvious difficulties in doing this, which stem from the complexities of preparing antiparticles and cooling them within their short lifetime of 140 ns. In this context, Cassidy *et al.*’s results are an important proof of principle that optical spectroscopy of the confined Ps atoms can be made before they annihilate.

Silica is well known as a medium for forming Ps. Positrons shot into a silica target can, as they thermalize, interact with electrons in the solid to form Ps. If the tar-

get is porous silica, an aggregate of silica nanoparticles, or ultrafine silica powders, the Ps atoms are likely to diffuse to a surface from which they can be spontaneously emitted into the pore region or free space between the nanoparticles. (Similarly, some of the positrons that diffuse to the surface may also form Ps by picking up an electron.) Once within these “voids,” a Ps atom will not return to the bulk. This is because the work function of Ps for silica is negative, meaning that it costs more energy for the Ps atom to be in the silica than in the vacuum [6]. Perhaps it is worth mentioning that the particle-antiparticle annihilation is a relatively slow process compared to electron and phonon excitations and Ps formation.

Ps is unique among the neutral atoms in that it spontaneously emits gamma rays when it annihilates. Analyzing these gamma rays provides a useful probe of a positronium atom’s interaction with other atoms, molecules, or solid surfaces before it annihilates and is an established technique for studying materials. As a result, there are a number of methods for preparing and measuring Ps that consist essentially of detecting the annihilation gamma rays. But in order to perform accurate spectroscopy of Ps in confinement, Cassidy *et al.* had to first create a very short and dense pulse of positrons to maximize the temporal and spatial overlap of the Ps bunch and the laser beam used in the measurements. Harnessing a series of existing techniques, they first moderated and trapped positrons that were produced by a sodium-22 radioactive source in a so-called Surko trap [7]. They then “dumped” the trapped positrons by applying a parabolic potential along the cavity of the trap to form a pulse with a temporal width of 15–20 ns, which was further compressed (with a “buncher”) into a subnanosecond pulse containing  $\sim 2 \times 10^7$  positrons. The pulse was also spatially compressed with a pulsed magnet [8]. Finally, they injected the positron pulse onto

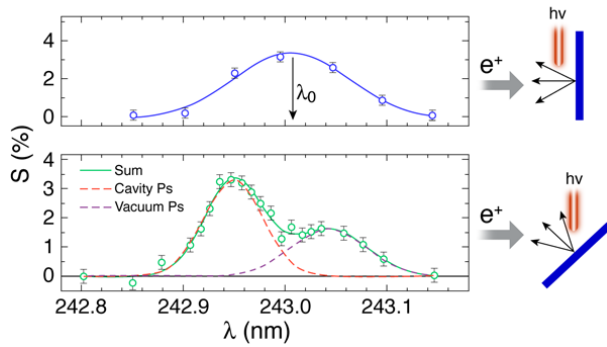


FIG. 1: Two experimental setups allow Cassidy *et al.* to measure and compare the excitation spectra of “free” and confined positronium. (Top plot) The excitation spectrum of positronium that escapes the sample surface is measured using laser light aimed parallel to the sample surface (top right schematic). By rotating the sample 45 degrees (bottom right schematic), it is possible to probe Ps that lies within the silica pores as well as the Ps outside (bottom plot). The spectrum has two components: one shifted to longer wavelength (free Ps, in vacuum) and the other shifted to shorter wavelength (cavity Ps, inside the pores). (Credit: Carin Cain, adapted from Ref. [4])

a porous silica target having an open pore structure. The lifetime of the ortho-Ps (Ps with the electron and positron spins parallel) in the pore is shorter than its intrinsic lifetime but still longer than about 50 ns, allowing some of the Ps atoms to diffuse through the connected pores into the open space in front of the film surface.

The team used a combination of different lasers and specialty optics to both produce light over a range of frequencies near the Ps Lyman- $\alpha$  transition and to photoionize the 2P state of Ps. They irradiated the target in two configurations: one where the target surface was perpendicular to the positron beam but parallel to the laser light, and a second where the surface was rotated by  $\sim 45^\circ$  with respect to the positron beam and the light axes (see schematics, Fig. 1). In the former case, the light did not touch the surface and thus only the Ps coming out of the surface (vacuum Ps) was excited. In the latter, the laser entered the silica and the Ps could be excited while inside the film pore.

To make their principle measurement, the team used a technique they developed in earlier work [9], called single-shot positron annihilation lifetime spectroscopy, which yields the excitation spectrum of Ps. When only those Ps atoms that were in the vacuum were excited, they observed a Doppler broadened line shape centered at the 1S-2P wavelength  $\lambda_0$  (top plot, Fig. 1). The broadening occurs because the Ps atoms are moving with a distribution of speeds. When the target was rotated so that the light passed through the film, they saw two components in the line shape, whose centers were above and below  $\lambda_0$  (bottom plot, Fig. 1). The longer wavelength component corresponds to free Ps in vacuum: the

Doppler broadened line shape is shifted to lower frequencies because the Ps emitted from the tilted target surface is, on an average, moving towards the laser. The shorter wavelength component is due to the transition of Ps atoms inside the pore. The width of this component is narrower than the vacuum component.

Cassidy *et al.* see an energy shift of about 1.3 meV for the confined Ps, which they attribute to energy level shifts of both the 1S and the 2P states. A simple model for the center-of-mass energy level shift that would correspond to changes in the size of the 1S and 2PPs atoms in confinement would give  $\Delta E_0 = 3.6$  meV, almost three times larger than the observed value. By accounting for the repulsive potential the cavity surface exerts on the Ps wave function [10], they show the shift they observe corresponds to a cavity diameter of  $\sim 5$  nm, consistent with the nominal size of the pore of the sample.

The transition width for positronium in confinement is, experimentally, narrower than that of vacuum, but still broader than expected [11]. That said, the line shapes for “free” Ps are already extremely broad because of positronium’s low mass; thus any methods to further narrow them would open up the possibility of, for example, using positronium atoms for gravity interferometry experiments [12]. Since Cassidy *et al.* attribute the residual broadening they see to disorder in the sample, it may be possible to achieve narrower Ps transitions by using a sample with less disorder.

The fact that Cassidy *et al.* are able to perform laser spectroscopy of confined positronium before the atoms annihilate provides a new method for the determination of the size of pores in materials in addition to the already established uses of Ps[13]. It also paves the way for preparing a Ps BEC. Though challenges remain, including finding a way to increase the spatial density of the injected positrons and to effectively cool the Ps atoms, a Ps BEC could be the ultimate source for precision measurements. Though it is an even longer-term goal, there is particular interest in observing stimulated annihilation from a positronium BEC as a first step toward making a gamma-ray laser.

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## About the Author

### Toshio Hyodo



Toshio Hyodo is a Professor at the Institute of Material Structure Science, High Energy Accelerator Research Organization and a Professor Emeritus at the University of Tokyo. He is ex-Chair of the International Advisory Committee on Positron Annihilation and the current President of the Japanese Positron Science Society. He received his Ph.D. from the University of Tokyo in 1979 for his experimental work on positron annihilation in solids. During 1980–1982 he was a visiting researcher at the Physics Department of Queen's University, Canada. His research interests include positronium (Ps) states in solid materials and Ps interactions with molecules and solid surfaces using silica aerogel as a Ps formation medium, as well as slow positron beam instrumentations. He is also actively interested in the improvement of physics education.