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2010 J. Phys.: Conf. Ser. 225 012007

(http://iopscience.iop.org/1742-6596/225/1/012007)

International Symposium on Advanced Science Research 2009 IOP Publishing
Journal of Physics: Conference Series **225** (2010) 012007 doi:10.1088/1742-6596/225/1/012007

Positronium Formation in Porous Materials for Antihydrogen Production

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Abstract. Positronium (*Ps*) formation measurements in several porous materials as: Vycor, germanate Xerogel, Metal-Organic Frameworks MOF-177 and Aerogel with two densities (20 and 150 mg/cm^3), were performed by means of a variable energy positron beam provided with a Ge detector and a positron lifetime spectrometer. An efficient formation of cooled *Ps* atoms is a requisite for the production of antihydrogen, with the aim of a direct measurement of the Earth gravitational acceleration *g* of antimatter, which is a primary scientific goal of AEGIS (Antimatter Experiment: Gravity, Interferometry, Spectroscopy). Porous materials are necessary to form a high yield of *Ps* atoms as well as to cool *Ps* through collisions with the inner walls of the pores. The different materials were characterized and produce *Ps* into the pores. Lifetime measurements give an estimation of the typical pores dimension of the substances. A comparative study of the positron lifetime and the *Ps* fraction values in the above mentioned materials indicates that silica Aerogel, with the appropriate density, is an excellent candidate for an efficient formation of cold *Ps* atoms for the AEGIS project.

1. Introduction

The AEGIS experiment has been approved at the CERN Antiproton Decelerator to directly measure the gravitational acceleration g on a beam of cold antihydrogen [1]. Gravitational interaction between matter and antimatter has never been tested experimentally. Some fundamental questions of modern physics relevant to unification of gravity with the other fundamental interactions, models involving vector and scalar gravitons, matter–antimatter symmetry can be enlightened via experiments with antimatter [2].

Figure 1 shows a schematic drawing of the basic experimental setup that should reach an accuracy of 1% in the measurement of the matter-antimatter gravitational acceleration. The experiment is designed to allow higher precision measurements through cooling of the antiproton ($\overline{p}$) beam [3]. The essential steps leading to the production of antihydrogen ($\overline{H}$) and the measurement of its gravitational interaction in AEGIS with the use of the CERN cold $\overline{p}$ are the following: i) production of Ps; ii) laser excitation of Ps to form Rydberg positronium (Ps^*) and to increase the cross section of the following reaction [4,5]; iii) production of cold Rydberg antihydrogen $\overline{H}^*$ at 100 mK based on the reaction between cold $\overline{p}$ and Ps^*; iv) acceleration of $\overline{H}^*$ beam using inhomogeneous electric fields [3] and v) determination of g in two-grating Moiré deflectometer coupled with a position-sensitive detector [6].

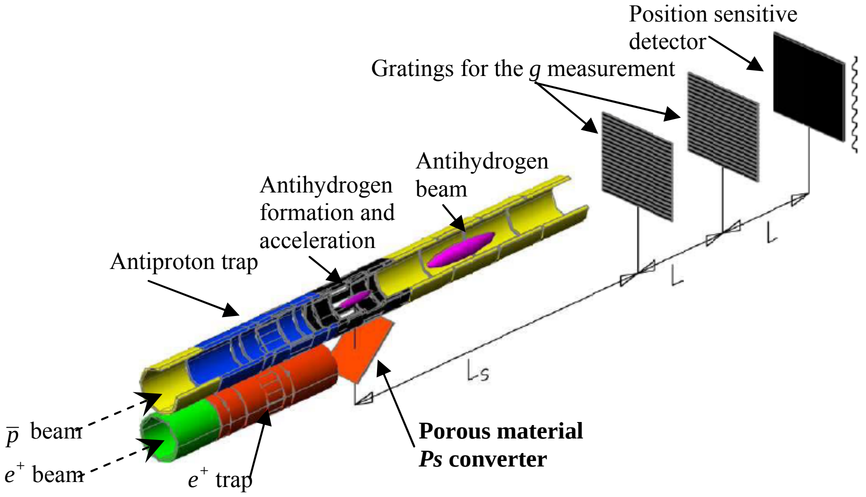

Figure 1. Sketch of the experimental setup region where antiprotons and positrons are manipulated to form and to accelerate antihydrogen. This region is at low temperature (~100 mK) in an axial magnetic field of 1 T (after Ref. 7).

In the present work results regarding Ps formation are reported. The Ps ionization energy of the bound state between a positron and an electron is 6.8 eV. There are several models of Ps formation, which were reviewed in a monograph by Mogensen [8]. The first general theory was published by Ore and Powell [9]. Ground state Ps is formed for 75% as ortho-Ps (spin 1) and for 25% as para-Ps (spin 0). Para-Ps annihilates in two γ rays with 511 keV each and ortho-Ps in vacuum is required to annihilate into three γ rays at least, with a maximum energy of 511 keV each and with a total energy of 1022 keV ($2m_0c^2$). We are interested only in the fraction of Ps emitted by the converter as ortho-Ps with a characteristic lifetime in vacuum of 142 ns, since the lifetime of para-Ps is too short (125 ps) to allow the necessary laser excitation before its decay. The ortho-Ps lifetime can be shortened by collisional pick-off annihilation and ortho-para conversion (both processes lead to annihilation in two γ rays) in ranges typically from a fraction of 1 ns to tens of nanoseconds.

An efficient formation of cooled Ps atoms is a requisite for the production of antihydrogen. Porous materials are necessary to form a high yield of Ps atoms as well as to cool Ps through collisions with the inner walls of the pores. The collisions between Ps and the internal surface of the pores involve a weak coupling to phonons or other surface modes. The velocity distribution of the Ps atoms coming out of the target should be the order of 10^4 m/s to allow Ps laser excitation to a Rydberg state (Ps^*). The formation of cold $\overline{H}^*$ (Fig. 1) will be possible by means of the charge exchange reaction between Ps^* and cold $\overline{p}$

$$Ps^* + \overline{p} \rightarrow \overline{H}^* + e^- , \tag{1}$$

with a residual electron.

The different porous materials tested in this work (Vycor, germanate Xerogel, Metal-Organic Frameworks MOF-177 and Aerogel with two densities) were characterized by means of a variable energy positron beam with a high purity germanium detector and positron lifetime spectroscopy. The results indicate that these materials produce Ps into the pores. Lifetime measurements give an estimation of the typical pores dimension of the substances. A comparative study of the positron lifetime and the Ps fraction values in the above mentioned materials indicates that silica Aerogel, with a selected density, is an appropriate candidate for an efficient formation of cold Ps atoms for the AEGIS project.

2. Experimental procedure and methodology

The composition of the porous materials used in the present work is shown in Table 1.

Table 1. Composition of the materials.

Porous sample	**Composition**
Vycor	$(SiO_2)_{0.55}(B_2O_3)_{0.35}(Na_2O)_{0.1}$
Germanate Xerogel	$(Ge)_{0.3}(SiO_2)_{0.7}$
MOF-177	$Zn_4O(O_2C\text{-})_6$
AerogelC-20	$(SiO_2)_{1-X}(C)_X$, X=0.0067
Aerogel-150	SiO_2

The procedure used to produce a mesopore film in the Vycor glass of about 2 μm in depth [10] was the following: i) the glass sample was annealed at 530°C for 16 h; ii) the sample was next converted into a porous sorbent by leaching in hot (80°C) 3N H_2SO_4 for 15 minutes and then in 0.5N NaOH solution at room temperature for 5 min and iii) the sample was washed out to neutrality with distilled water and dried at 80°C for 24 h. The germanate Xerogel was prepared following the procedure indicated in Ref. 11. After drying, the sample was sintered with the following conditions: the Xerogel was heated from room temperature to 450°C under oxygen flux with a ramp of 6°C/h, and then it was kept at such temperature for 1 day in argon atmosphere to replace oxygen with inert gas. Further heating to the final temperature (680°C) was then carried out in 2% vol. H_2 in Argon. Nitrogen sorption measurements showed the presence of a bimodal distribution of pores, comprised of nanopores (below 2 nm) and mesopores (between 20 nm and 50 nm). The number density of the small pores was strongly dominant (150:1). MOF-177 was synthesized according to Ref. 12 in the form of quasi-cubic grains with a side of about 50 μm. MOF-177 is a porous materials with low density (4.27×10^{-1} g cm^{-3}) and large, regular, accessible cages and tunnels. This material is specially used for gas storage [13]. Before the positron measurements granular MOF-177 was compacted with a charge of 8×10^3 N cm^{-2} into a cylindrical pellet of 1.5 cm in diameter and 7 mm in thickness. Silica Aerogel samples were fabricated using a procedure developed for the particle capture collectors of the NASA Stardust project [14]. The sample density was controlled by mixing different amounts of acetonitrile into the sol. The solution was then dried by supercritical solvent extraction, which allows the solvent to be removed while the network gel remains in a highly expanded state in the form of a three dimensional array of filaments. For this comparative study, two samples were selected with densities 20 mg cm^{-3} (AerogelC-20) and 150 mg cm^{-3} (Aerogel-150) and high porosity (99% and 93%, respectively). AerogelC-20 was doped with 0.4 wt.% (0.67 at.%) of graphite.

The measurements of *Ps* formation yield were performed by means of a monoenergetic positron beam using the well-known "3γ method" [15-17]. The positron implantation energy was variable from 0.05 keV up to 18 keV. The measurements were performed at room temperature with a vacuum level between 10^{-7}-10^{-9} mbar. A high purity germanium (HPGe) detector was used to obtain the annihilation γ spectrum. From the annihilation spectrum it was possible to obtain the *R*(*E*) parameter

$$R(E) = \frac{V}{P}, \tag{2}$$

where *P* and *V* are the integrated counts (after background subtraction) in the peak (511 ± 4.25 keV) and valley area (after the Compton edge, from 350 keV up to 500 keV) of the energy γ spectrum. When there is no pick-off annihilation, i.e. when the *Ps* is emitted in vacuum, the positronium fraction $f(E)$ can be obtained from $R(E)$ using the relationship:

$$f(E) = \left[1 + \frac{P_1\left(R_1 - R(E)\right)}{P_0\left(R(E) - R_0\right)}\right]^{-1}, \tag{3}$$

where P_0 and R_0 are the parameters when the positronium fraction is nil (F=0) and P_1 and R_1 are the parameters when 100% of the positrons becomes *Ps* (F=1). When the *Ps* is formed inside the pores, the pick-off effect reduces the probability of 3γ annihilations by a factor

$$\varepsilon = \frac{\lambda_{3\gamma}}{\lambda_{3\gamma} + \lambda_{p.o.}}, \tag{4}$$

where $\lambda_{3\gamma}$= (142 ns)$^{-1}$ is the 3γ annihilation rate in vacuum and $\lambda_{p.o.}$ the pick-off annihilation rate. Thus the proper expression for the *Ps* fraction becomes:

$$F(E) = \varepsilon^{-1} f(E). \tag{5}$$

In the present work the factor ε^{-1} can be evaluated for high positron implantation energies from the available lifetime measurements (see below). In all other cases we report the result obtained from Eq. (3) as "3γ fraction" ($F_{3\gamma}$= f). We warn however that this nomenclature, which is adopted by other authors [18,19], is somewhat misleading, since the true fraction of positrons annihilated in 3γ is smaller by a factor ¾. Nevertheless $F_{3\gamma}$ is the useful parameter to assess the potential of a material to emit o-*Ps* that has lost kinetic energy by collision while meandering through open pores.

The calibration for 0% and ~100% of *Ps* in vacuum was performed using a Ge single crystal (100) at 1000 K [16]. However the R_0 and R_1 parameters depend on the scattering and absorption of the annihilation gamma rays photons in the sample and on the experimental geometry. Corrections of R_0 and R_1 measured with the Ge sample are thus necessary [20]. In thick samples of porous materials a direct measurement of R_0 is impossible because *Ps* is formed even at high implantation depth. These corrections were thus performed following the procedure indicated in Ref. 20. The contribution of these corrections affects $F_{3\gamma}$ from 1% up to 3%.

Lifetime measurements were performed with the pulsed low-energy positron-lifetime spectrometer PLEPS at the high intensity positron beam facility NEPOMUC at the research reactor Heinz Maier–Leibnitz FRM II of the Technical University Munich [21,22]. The resolution of the spectrometer was about 240 ps. The lifetime measurements were performed with a time-windows of 20 ns periodically repeated. In the presence of pores with a size distribution, the lifetime spectra are expected to be the superposition of infinitely many exponential components. In the present case, an approximate analysis in four components was adopted to obtain four mean lives: ($\tau_1 \ldots \tau_4$) and the corresponding intensities ($I_1, .. I_4$). The intensities I_j of each lifetime component were corrected by multiplying by [1-exp(-20ns/τ_j)] after the subtraction of the random background and subsequently were re-normalized.

3. Results and discussion

Figure 2 shows the 3γ fraction $F_{3\gamma}$ as a function of the positron implantation energy for the different materials under test. At low implantation energy (≤200 eV) the 3γ fraction for all samples is high, between 31% and 50%. In MOF-177, germanate Xerogel and Vycor an increase of the 3γ fraction vs. the implantation energies was found, followed by an asymptotic decrease. This trend, which is observed also in other materials [18,19,23], can be explained by a model that takes into account the energy dependence of the ortho-*Ps* formation and escape [24]. The increase in MOF-177, germanate Xerogel and Vycor, if it occurs, is documented by only one experimental point, which is not enough to obtain the energy dependence of the *Ps* yield using the model of Ref. 24. On the contrary, the asymptotic decrease is clearly observed and can easily be fitted with the mentioned model (dashed line above 100 eV); the corresponding best-fit ortho-*Ps* diffusion lengths are 17 nm, 5 nm and 3 nm for MOF-177, Germanate Xerogel and Vycor respectively. The model cannot be applied to silica Aerogel,

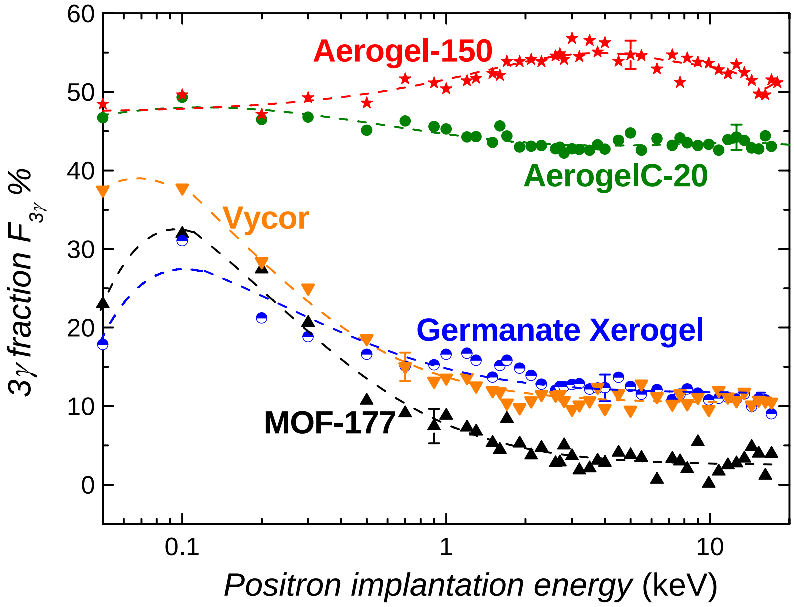

Figure 2. Positronium 3γ fraction $F_{3\gamma}$ as a function of the positron implantation energy in different porous samples: MOF-177, Vycor, germanate Xerogel, and two densities of Aerogel (20 mg cm^{-3} and 150 mg cm^{-3}). The dashed lines upper 0.1 keV in MOF-177, germanate Xerogel and Vycor are fits of the model proposed in Ref. 24. The others dashed lines are only a visual guide. Error bars are shown for one point only in each evolution.

where the low pick-off rate combined with a diffusion length of some microns [25,26] cancels the characteristic decrease of the 3γ yield at high energy. The 3γ fraction in Aerogel samples is very high at all implantation energies, as expected for a material with very large free space between silica particles. The values between 43% and 57% observed above 1 keV are similar or higher than to the maximum values reported in literature [19].

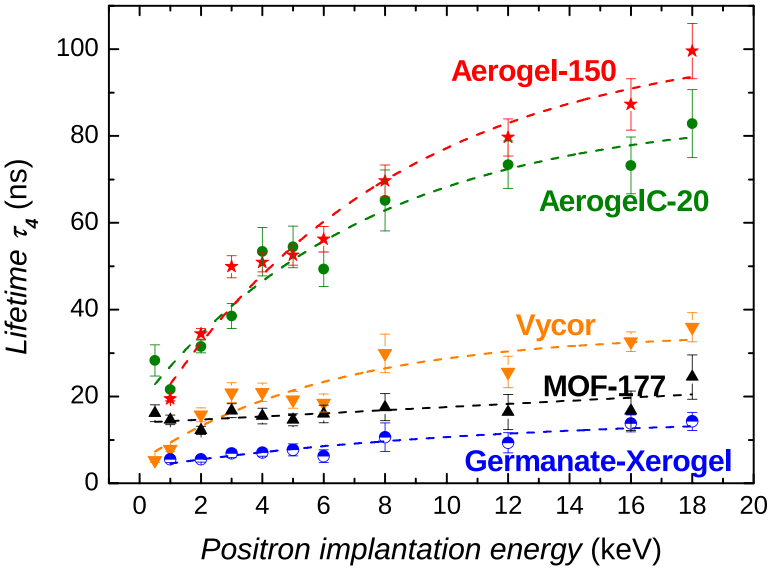

Figure 3. Positron lifetime of the long component τ_4. The dashed lines are only a visual guide.

The relevant results of the lifetime measurements are reported in Figure 3 and Table 2. Figure 3 depicts the implantation energy dependence of the longest lifetime τ_4 isolated in the four-component analysis of the lifetime spectrum. The decrease of τ_4 observed for Aerogel at low implantation energies is symptomatic of *Ps* escape in the vacuum, and is an instrumental effect related to a decreased efficiency of detection of ortho-*Ps* when the pick-off annihilation channel is suppressed.

Table 2. Lifetime and intensity associated to the long component, equivalent pore size for spherical and cylindrical geometries and *Ps* fraction *F* (Eq. 5).

Porous sample	**Lifetime** τ_4 (ns)	**Intensity** I_4 (%)	**Diameter** (nm)		*F* (%)
			Spherical	Cylindrical	
Vycor	35	2.7	2.5	2.2	40
Germanate Xerogel	14	3.7	1.6	1.4	~100
MOF-177	20	3.7	1.8	1.7	20
AerogelC-20	82	27	6.0	5.2	74
Aerogel-150	100	23	9.4	8.4	71

Table 2 includes the lifetime τ_4 measured at the highest implantation energy, which can be taken as the upper limit of the ortho-*Ps* lifetime in the pores of the samples. The data reported in Table 2 for Aerogel are consistent with the results obtained by Mincov et al. [27] and by Chang et al. [28]. The lifetime values can be used to estimate the size of the pores with the extended Tao-Eldrup model [29] for spherical pores and its extension to cylindrical geometry [30]. The diameters obtained for spherical and cylindrical (infinitely long) pores are reported in Table 2. This table includes the intensity I_4, which is to be considered the lower limit of the ortho-*Ps* fraction because ortho-*Ps* also contributes to the shorter components and because the energy window of the lifetime spectrometer is set for 2γ annihilations. A different estimate of the ortho-*Ps* fraction may be obtained as ¾ of *F*, where *F* is the *Ps* fraction, evaluated from Eq. 5 using the experimental lifetime data to calculate the factor ε (Eq. 4).

4. Concluding remarks

In our quest for a promising positron-*Ps* converter to be chosen in the AEGIS experiment, we have tested a series of materials with known open porosity. We have found that the highest formation of *Ps* occurs in germanate Xerogel and Aerogel. However, high formation is not the only important factor. It must be reminded that *Ps* is emitted into the pores with an initial kinetic energy of the order of a few eV. Since our goal is to obtain ortho-*Ps* in the free space with energies of the order of 10^{-2} eV, it is necessary that the *Ps* suffers a large number of collisions with the walls of the pores before emerging in the free space outside the target. This means that the useful ortho-*Ps* fraction must come from the deep regions of the target. This is certainly not the case of germanate Xerogel, where ortho-*Ps* appears to be strongly quenched in the pores. The reason is certainly the presence in this material of a dense distribution of very small pores, probably with a low degree of interconnections, which act as dominant ortho-*Ps* cages. Most probably, this situation also occurs in Vycor. MOF-177 is a new material; there is no previous information of its ability of producing *Ps*. Our experiment shows that, in spite of its very large free volume, this material is not really interesting as a converter. On the contrary, low-density Aerogel is a promising target candidate under both aspects: high formation and long survival time of ortho-*Ps* inside the pores. In the present work it was not possible to measure the kinetic energy of ortho-*Ps* outside the target. The literature tells however that ortho-*Ps* cooling in porous silica is feasible. Nagashima et al. [31] found an average *Ps* energy of 93 meV in Aerogel of 100 mg cm^{-3} at room temperature in vacuum. Mills et al. [25] observed ortho-*Ps* thermalization also at low temperature in SiO_2 powders compressed up to a similar density to the more dense Aerogel that was studied in the present work. The results of Ref. 25 indicate: 2% at 4.2 K, 8% at 77 K and 12% at 300 K of thermal ortho-*Ps* with positron implantation energies of 19 keV, 10 keV and 7 keV, respectively. These high implantation energies require rather thick samples (several microns, for typical Aerogel densities), which is a condition easily met with homogenous Aerogel. However a compromise must be reached between efficient cooling, which requires deep positron implantation, and high ortho-*Ps* yield outside the target, which is favoured by implantation depths not large in comparison with the ortho-*Ps* diffusion length. In the case of Aerogel, the ortho-*Ps* diffusion length is expected to be about 2 μm, as measured for similar material [26] and to implant positrons at 2 μm in a target with mass density of 150 mg cm^{-3} the positron beam energy should be about 3.5 keV. Time-of-flight measurements of the kinetic energy of the emitted ortho-*Ps* are highly desirable for finding the best compromise.

Acknowledgments

We gratefully acknowledge Prof. Gianpaolo Rosati for the compaction of the MOF-177 samples. Work partially supported by Istituto Nazionale di Fisica Nucleare (Italy).

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