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Abstract. Positronium yield using the "3γ method" and lifetime measurements were performed at cryogenic and room temperature by means of a variable energy positron beam in homogeneous porous silica (Aerogel). An estimation of the positronium (*Ps*) mean diffusion length was obtained by measuring capped samples. 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 the primary scientific goal of AEgIS (Antimatter Experiment: gravity, Interferometry, Spectroscopy; CERN, Geneva). Porous materials are necessary to obtain a high *Ps* yield as well as to thermalize *Ps*. Our results indicate a high *Ps* production, long survival time and diffusion length in Aerogel samples. It will be shown that positronium yield, lifetime and diffusion length are independent on temperature and on the effect of gas adsorption at low temperature. The results indicate that Aerogel is a good candidate for an efficient formation of cold *Ps* for the AEgIS experiment.

1. Introduction

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 [1]. The AEgIS experiment is carried out at the CERN Antiproton Decelerator to directly measure the gravitational acceleration *g* of a beam of cold antihydrogen [2, 3]. Gravitational interaction between matter and antimatter has never been tested.

An efficient formation of cooled positronium (*Ps*) atoms is a requisite for the production of antihydrogen ($\overline{H}$) in the AEgIS experiment. Porous materials are necessary to produce 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 of the order of 10^4 m/s to allow *Ps* laser excitation to a Rydberg state (Ps^*) [4] and for efficient $\overline{H}$ formation, which requires that the relative velocity of antiprotons ($\overline{p}$) and Ps^* must be not higher than the classical orbital velocity of the positron in the Rydberg *Ps* atom.

The formation of cold Rydberg antihydrogen $\overline{H}^*$ will be possible by means of the charge exchange reaction (more details in Refs. 2 and 3). The AEgIS experiment will be performed at cryogenic temperatures (~100 mK). A negative aspect of the cryogenic environment is the formation of ice at the

surface of porous materials, as well in high vacuum condition. This effect could avoid the *Ps* escape in the free space outside the sample.

The Aerogels tested in the present work were characterized at room and cryogenic temperatures. Since the experimental evidences show a high *Ps* production, mobility and long survival time independent on the temperature, it is possible to conclude that the studied Aerogels are good candidates for the AEgIS experiment.

2. Experimental procedure and methodology

Silica aerogel samples were produced using a procedure developed for the particle capture collectors of the NASA Stardust project [5]. The first step for preparing ultra-low density aerogels involves the formation of a silica sol. The sol was prepared from tetraethyl ortho silicate (TEOS) in a way similar to that developed by Tillotson and Hrubesh [6], where the hydrolysis and condensation of a silicon alkoxide forms a suspension of micro-particles. The aerogel precursor mixture was prepared by combining acetonitrile, sol, water, methyl triethoxy silane and ammonium hydroxide. The density of the final aerogel is controlled by varying the ratio of the acetonitrile to that of the sol. The methyl triethoxy silane was added to make the aerogel hydrophobic. The resulting wet gel was dried in an autoclave, where the temperature was ramped up to 568 K at approximately 36 K/h, after pressurizing the system to 54 bar with argon. The vessel was then depressurized at a rate of 2.25 bar/h. Once the temperature had reached ambient pressure, the system was opened and allowed to cool before removing the aerogels. This process of "supercritical solvent extraction" allows the solvent to be removed while the silica network remains in a highly expanded state in the form of a three dimensional array of filaments. The aerogel samples have densities of 85 and 150 mg cm^{-3}. The sample with lower density was partially dried at room temperature in air before supercritical solvent extraction. Some samples were capped with an Al layer of about 110-120 nm. The Al layers were deposited by molecular beam epitaxy.

Both the lifetime and *Ps* formation measurements were performed, in high vacuum conditions (about 10^{-8} mbar), at two different temperatures: cryogenic temperature (18 K) and room temperature (295 K). The measurements of *Ps* formation yield were made by means of a monoenergetic positron beam, with energy up to 18 keV, using the well-known "3γ method" [7-9] and detecting the γ rays with a high purity germanium detector. Lifetime measurements were performed with a fast-fast timing coincidence system with a time resolution (FWHM) of 270 ps. A 15 mCi source of ^{22}NaCl deposited between two thin Kapton foil (7.5 μm) was sandwiched between two identical porous samples. The thickness of the samples, of about 1.5 cm, was sufficient to assert that the positrons annihilate only into the samples. The samples were covered with an Al foil to ensure good thermal contact between the cold finger and the sample. Each spectrum contained about 2×10^6 counts and it was analysed with the POSITROFIT program [10] in four discrete components.

3. Results and discussion

Figure 1.a shows the *Ps* yield as a function of the positron implantation energy in Aerogel 85 and 150 mg/cm^3 measured at room temperature (full symbols). *Ps* yield is very high for any implantation energy. The more promising Aerogel (85 mg/cm^3) was also tested at low temperature (18 K, open symbols). The results indicate that *Ps* yield does not depend on temperature for any implantation energy. A similar result was recently found in silica films [11] and in MCM-41 [12].
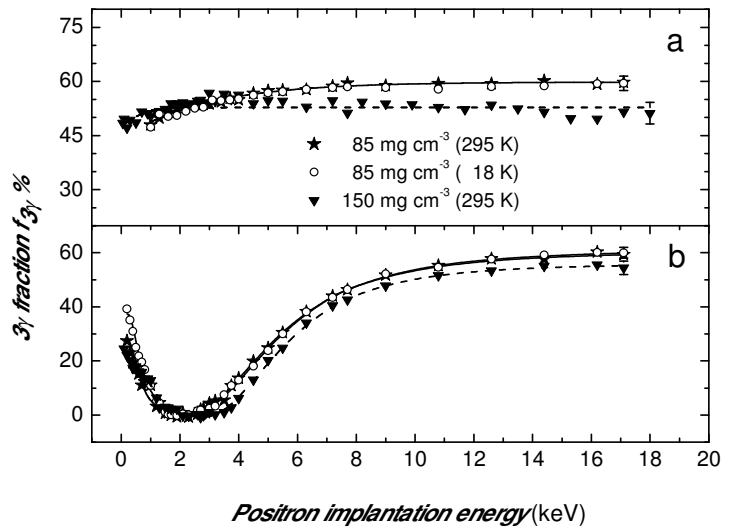
Water absorption by porous SiO$_2$ was ascertained [13] even in ultra-high vacuum conditions, where at low temperatures (below 150 K) ice is formed [14]. The results presented in Fig. 1.a show that the effect of water adsorption at low temperature in the studied porous silica does not influence the *o-Ps* yield. The main reason is attributed to the hydrophobicity of the Aerogels presented here [15].

In order to obtain the positronium diffusion length the mesoporous samples were capped with an Al film to prevent *o-Ps* escape into the vacuum. The obtained results in Aerogel 85 and 150 mg/cm^3 at

room temperature are shown in Fig. 1.b (full symbols). The Aerogel 85 was also measured at 18 K (open symbols) and the results show that the distribution does not depend on temperature. Only at low energy, where a fraction of the positrons form *Ps* at the Al surface, there is a small difference between the results obtained at room temperature and at 18 K due to the formation of ice at the Al surface. *Ps* formation is almost zero for implantation energies of about 2 keV; indeed, all the incident positrons annihilate in the metallic layer where no *Ps* formation is expected. The results of the *Ps* fraction as a function of the positron incident energy obtained in a capped porous sample were analyzed by means of a semi-linear fitting procedure using the VEPFIT program [16]. This program extracts relevant parameters from positron measurements on layered structures. In the present case our attention was focused on the diffusion *Ps* length inside the porous samples. The experimental data were fitted using a

Figure 1. *Ps* fraction $f_{3\gamma}$ as a function of the positron implantation energy in Aerogel 85 and 150 mg cm^{-3} at 18 K and 295 K. In panel b the samples were capped with Al to avoid the positronium escape. Error bars are shown for one point only in each evolution.

Table 1. Lifetime τ_4, mean free path l, thicknesses of the Al capping layer z_{Al} and the interface z_{int} and *o-Ps* diffusion length L_{Ps} in Aerogel 85 and 150 mg cm^{-3} samples. The value indicated with F is a fixed parameter.

Aerogel	T(K)	τ_4 (ns)	l (nm)	z_{Al} (nm)	z_{int} (nm)	L_{Ps} (µm)
85	300	129(3)	23(2)	108(6)	15F	2.30(0.25)
85	18	128(3)	22(2)	108(6)	15F	2.25(0.25)
150	300	122(3)	14(2)	120(5)	15F	1.90(0.20)

model of layers comprised of: surface, an Al layer, an interface and a semi-infinite layer of the porous material. The interface between Al and the porous material used in the analysis model takes into account a possible contamination effect of few nanometers (z_{int}) on the porous material with Al during deposition. The transport of positrons after implantation and thermalization in a solid is approximated by the classical diffusion theory. The depth dependent implantation rate of thermalized positrons is obtained by adopting the implantation-profiles of the so-called Makhov distribution [17]. According to the fitting results the positron diffusion length after thermalization, of about 20 nm, is much lower that the *Ps* diffusion length. In this condition it is possible to assume that the diffusion equation holds only for *Ps*. The adopted model is based on the assumption that the diffusion coefficient does not depend on the *o-Ps* temperature during its cooling. The continuous and dashed lines in Fig. 1.b correspond to the best fit values of the *o-Ps* diffusion length. The most relevant parameters of the fitting procedure are shown in Table 1. These high diffusion lengths are consistent with a high *o-Ps* emission outside the material also when positrons are implanted at relatively high energies.

Positron lifetime measurements were performed in vacuum in the studied materials at room temperature and also at 18 K in Aerogel 85 mg/cm^3. The best fits of the spectra were obtained with four discrete lifetime components. The first shortest is ascribed to *p-Ps* as well as to free positrons which annihilate in the bulk SiO$_2$ (τ_1= 0.2(1)). The second component is ascribed to structural defects (τ_2= 0.6(1) ns, with ($I_1 + I_2$)≈ 80%). The third component probably comes from o-Ps annihilating in structural defects (τ_3= 3(1) ns, I_3≈ 0.80(5)%). The fourth lifetime component τ_4 reported in Table 1 is the most interesting one, since it can be attributed to *o-Ps* annihilations into the pores. The results indicate the presence of a long *o-Ps* lifetime that is independent on the sample temperature (within the experimental uncertainty). The intensity of this component is about 20% in all cases; because this

intensity depends also on the energy window of the stop channel in the lifetime spectrometer, only a fraction of *o-Ps* is revealed with a lifetime measurement.

It is known that the lifetime of *o-Ps* confined into closed pores depends on temperature according to the extended Tao-Eldrup (TE) model [18, 19]. But in silica when the porosity is very high (93-96%) and the pores are large and interconnected, as in the aerogels presented here, there are evidences that the lifetime is independent on temperature, i.e. there is no decrease in the pick-off rate at low temperature compared to room temperature [20-23]. In this case *Ps* is partially confined into the pores and it diffuses long distances during cooling as has been discussed before. The value of the fourth lifetime component allows to estimate the *o-Ps* mean free path *l* [24, 25] between collisions with the pore walls (see Table 1).

The results obtained in Aerogels indicate that these materials are good converters for the AEgIS experiment under the following aspects: high *o-Ps* yield, long survival time of *o-Ps* and very high *o-Ps* mobility independently on temperature and on the effect of gas adsorption at low temperature. These characteristics indicate that *o-Ps* cooling in Aerogel is feasible. In fact, the literature tells that in this homogeneous porous materials *Ps* thermalization and escape in vacuum were ascertained [26-28]. In particular, Mills et al. [27] have observed 2% and 8% of *Ps* thermalized (with respect to the implanted positrons) into the vacuum at 4.2 and 77 K, respectively, in an homogeneous porous silica with a mass density (180 mg cm^{-3}) similar to the materials studied here.

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