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Homogeneous porous silica for positronium production in AEgIS

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Abstract. Positronium (*Ps*) formation measurements in homogeneous porous silica (Xerogel of 85 mg cm^{-3} and swollen MCM-41 of 390 mg cm^{-3}) were performed at different temperatures (8K-293K) by means of a variable energy positron beam equipped with a Ge detector. The results indicate that Xerogel and swollen MCM-41 samples have a high *Ps* production, which is independent on the temperature. An estimation of the ortho-*Ps* mean diffusion length was obtained by measuring samples capped with an Al film (~110 nm). An efficient formation of cooled *Ps* atoms is a requisite for efficient production of antihydrogen, with the aim of a direct measurement of the Earth gravitational acceleration of antimatter, a primary scientific goal of AEgIS (Antimatter Experiment: gravity, Interferometry, Spectroscopy, CERN). Porous materials with open pores at the surface 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 before emerging in the free space outside the target. The results indicate that Xerogel and swollen MCM-41 are good candidates for an efficient formation of cold *Ps* atoms within the framework of the AEgIS project.

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 has been approved at the CERN Antiproton Decelerator to directly measure the gravitational acceleration g of a beam of cold antihydrogen [2-4]. Gravitational interaction between matter and antimatter has never been tested experimentally.

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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 the order of 10^4 m/s to allow *Ps* laser excitation to a Rydberg state (Ps^*) [5, 6] and for efficient $\overline{H}$ formation, which requires that the relative velocity of antiprotons 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 between Ps^* and cold $\overline{p}$

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

with a residual electron (more details in Refs. 4 and 7).

The porous materials tested in the present work (Xerogel and swollen MCM-41) were characterized by means of a variable energy positron beam with a high purity germanium detector. The results indicate that Xerogel and swollen MSM-41 are appropriate candidates for efficient formation of cold *Ps* atoms for the AEgIS project.

2. Experimental procedure and methodology

Silica Xerogel samples were fabricated using a procedure developed for particle capture collectors of the NASA Stardust project [8]. The sample density was controlled by mixing different amounts of acetonitrile into the sol. The solution was then dried at room temperature, 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. The density of the prepared Xerogel sample is 85 mg cm^{-3} with a porosity of about 96%. MCM-41 samples were prepared according to Refs. 9 and 10. The MCM-41 samples were synthesized at 115°C by using cetyltrimethylammonium bromide (CTAB, Aldrich), 1,3,5-trimethylbenzene (TMB, Aldrich), pyrogenic silica (Aerosil 200 Degussa), sodium hydroxide (Prolabo), and deionized water in molar ratios 1 SiO_2/0.26 NaOH/0.035 $NaAlO_2$/0.1 CTAB/20 H_2O/x TMB. The material was then filtered, washed with water, and dried at 80°C for 24 h. The solids were then calcined in air at 550°C for 8 h. The density of the prepared swollen MCM-41 samples is 390 mg cm^{-3} with a mesoporous and macroporous volume of 1.65 cm^3 g^{-1} and 0.5 cm^3 g^{-1}, respectively.

The measurements of *Ps* formation yield were performed by means of a monoenergetic positron beam using the well-known "3γ method" [11-13]. The positron implantation energy was variable from 0.9 keV up to 18 keV. The measurements were performed at different temperatures from 8 K up to room temperature (RT$\equiv$ 293 K) with a vacuum level between 10^{-8}-10^{-10} mbar. The samples were put in contact with the cold-finger of a cryostat. 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:

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

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* ($f = 1$). The calibration for 0% and ~100% of *Ps* in vacuum was performed using a Ge single crystal (100) at 1000 K [12]. In all other cases we report

the result obtained from Eq. (3) as "3γ fraction" F_{3g} ($f(E) = F_{3g}$). We warn however that this nomenclature, which is adopted by other authors [14,15], is somewhat misleading, since the true fraction of positrons annihilated in 3γ is smaller by a factor ¾. Nevertheless F_{3g} 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.

3. Results and discussion

Figure 1 shows the 3γ fraction F_{3g} as a function of the positron implantation energy in Xerogel and MCM-41 samples (full symbols) obtained at room temperature (RT). The *Ps* yield is very high at all implantation energies. Similar results were found in Aerogel with a density of 150 mg cm^{-3} [16]. The decrease of F_{3g} observed in Fig. 1 (full symbols) at low implantation energies is symptomatic of *Ps* escape in the vacuum. This is an instrumental effect related to a decreased efficiency of detection of ortho-*Ps*.
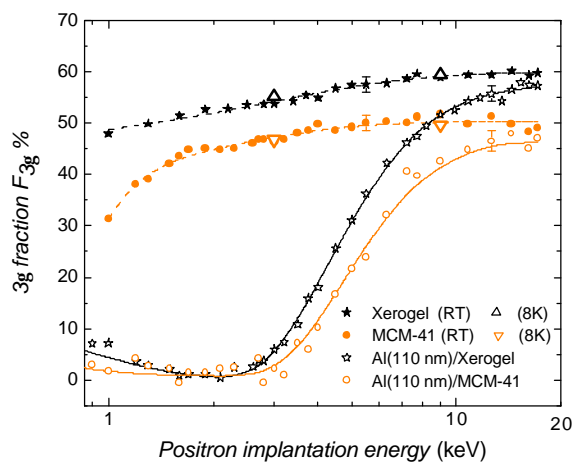
Figure 1. Positronium 3γ fraction F_{3g} as a function of the positron implantation energy in Xerogel and MCM-41 in homogeneous and capped samples. The dashed lines are only a visual guide. The continued lines are fits of the VEPFIT model. Error bars are shown for one point only in each evolution.
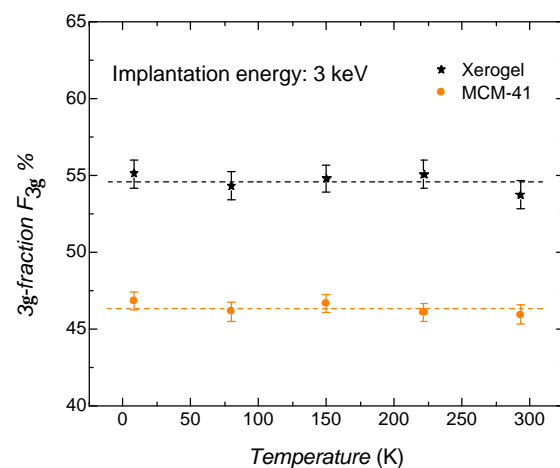
Figure 2. Positronium 3γ fraction F_{3g} as a function of the temperature for positron implanted inside the sample at 3 keV in Xerogel and swollen MCM-41 samples.

The porous samples were capped with an Al film of 110 nm to avoid the *Ps* escape (open symbols in Fig. 1). *Ps* formation at about 2 keV is almost zero in capped samples. In this case about 100% of positrons are implanted inside the Al film where *Ps* formation is not expected. The experimental data were fitted by VEPFIT [17] with a two-layer model (Al/porous material). VEPFIT takes into account the positron implantation distribution as a function of the implantation energy and the material density. The continuous line through the experimental data in Fig. 1 corresponds to the best-fit values of *Ps* diffusion lengths. The ortho-*Ps* diffusion lengths were estimated in (3.5 ± 0.3) μm and (0.56 ± 0.05) μm in Xerogel and MCM-41 samples, respectively. This model is based on the assumption that the diffusion coefficient does not depend of the *Ps* temperature during cooling. A full quantum mechanical treatment is required to properly describe the regime in which *Ps* is confined in a system whose dimensions are of the same order as the *Ps* de Broglie wavelength. However, these high diffusion lengths are consistent with a high ortho-*Ps* emission outside the material also when positrons are implanted at relatively high energies. *Ps* atoms are emitted from the pore walls with high kinetic energy (1-2 eV). Low *Ps* kinetic energies are desirable for AEgIS. The mean kinetic energy of ortho-*Ps* was estimated in Xerogel at room temperature. A value of 36 meV was obtained by means of Coincidence Doppler Broadening with and without the application of a static magnetic field (~0.3 T)

[18] (with a procedure similar to that used by Nagashima et al. [19]). This result indicates that ortho-*Ps* cooling in homogeneous porous silica is feasible. Measurements of the ortho-*Ps* emission energy in vacuum for these materials using *Ps* time of flight (*Ps*-TOF) are underway in homogeneous silica samples. *Ps*-TOF results were recently reported in Refs. 20 and 21 in silicon and silica ordered nano-channels films. Mills et al. [22] observed ortho-*Ps* thermalization also at low temperature in SiO_2 compressed powder (180 mg cm^{-3}). The results of Ref. 22 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 materials with low energies), which is a condition easily met with homogenous porous samples. 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. Low-density silica homogeneous materials are promising target candidates under two aspects: high formation and long survival of ortho-*Ps* inside the pores.

The temperature dependence of the *Ps* yield was studied since the AEgIS experiment will be performed at very low temperature (100 mK). Figure 1 shows that at 3 and 9 keV the values of F_{3g} measured at RT and at 8 K (open triangles) in Xerogel and MCM-41 samples are independent on the temperature within the experimental errors. More detailed information of the *Ps* yield as a function of the temperature in both materials for positrons implanted in the sample at 3 keV is presented in Fig. 2. These results indicate that in homogeneous silica-based materials the high *Ps* production does not depend on temperature. A similar result was found in silica films [21].

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