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Study of Positronium formation in nano-channelled silicon as a function of sample temperature

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Abstract. Oxidized nanochannel in silicon have been demonstrated to be suitable for positronium (Ps) formation and cooling also at low sample temperature. To investigate the Ps yield and to clarify the Ps formation mechanism we studied, by Positron Annihilation Spectroscopy (PAS), nanochanneled Si p-type samples in the 150-430 K temperature range. Ps yield was found to be constant in the 150-300 K temperature range, then it increases up to ~50% of its value from 350-400 K. This effect is associated to a decrease of the fraction of positrons annihilating in Si and in the SiO₂ layer on the nanochannels surface. This finding is compatible with the thermal decrease of the positive charge distribution at the Si/SiO₂ interface limiting e⁺ reaching the SiO₂ layer and to a charge rearrangement at the SiO₂ surfaces.

1. Introduction

Production of large amounts of Positronium (Ps) is required for many advanced experiments in fundamental physics. Confinement of $\sim 10^{17}$ Ps/cm³ in nanometric cavities at temperature $T < 30$ K would allow one to achieve the Ps Bose Einstein Condensation (Ps-BEC) [1]. Ps-BEC would open the route for linked experiments as making a plane wave monoenergetic Ps beam and a Ps annihilation γ ray laser [2]. The production of Ps beams at $T < 160$ K are required for high precision spectroscopy studies [3] and anti-hydrogen formation by charge exchange process with antiprotons [4, 5].

In recent years, porous [6] and channeled [7] insulating materials have attracted much attention concerning Ps production. These kind of materials are promising for two reasons: i) they efficiently produce Ps also at low sample temperatures and ii) provide a cooling mechanism [8–10]. Ps formed in the bulk is emitted with an energy of the order of few eV [11] into pores, then it cools down reducing its kinetic energy by collisions with the walls of the pores reaching the sample temperature [8]. Indeed when a network of connected open pores is present, Ps can escape into the vacuum after a hopping diffusion. For a review about Ps formation and cooling mechanism see Ref. [12].

Ps formation, cooling and emission into the vacuum from ordered nanochannels etched into a Si substrate and subsequently oxidized have been recently investigated [7, 8]. It was found that up to

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27% of implanted e^+ at 7 keV escapes into vacuum as Ps and a fraction of this Ps is emitted with thermal energy in the 150-300 K sample temperature range [8]. The high Ps amount was inferred to be due to the high probability of thermalized e^+ in Si to diffuse to the Si/SiO₂ interface due to the small distance among nanochannels (few nanometers) with respect to the diffusion length (more than 200 nm). The e^+ then pass into the SiO₂ because this is energetically favored [13]. Here Ps can be formed and emitted into the nanochannels.

In this work we present a study of Ps formation mechanism in nano-channelled Si by investigating the Ps yield in the 150-430 K temperature range by using 2γ - 3γ annihilation ratio of Ps (3γ -PAS) and Positron Annihilation Lifetime Spectroscopy (PALS).

2. Experiment

Two Si samples with oxidized nanochannels were synthesized by electrochemical etching according to the procedure described in Ref. 7 and 8. p-type Si (100) with resistivity 0.15–0.21 Ω cm was etched in an aqueous solution at 48 % of HF with added ethanol in volume ratio of 1:3=HF:ethanol. The etching current and time were $I=10$ mA and $t=13$ min, respectively. The samples were subsequently oxidized in air at 100°C for 2h and then subjected to 2 cycles of re-etching in HF for 60 s with a successive 2h annealing in air at 100°C in order to tune the nanochannels size between 10 and 18 nm [7]. One sample was capped with ~30 nm of TiO₂ deposited by RF reactive sputtering (13.56 MHz) from a pure TiO₂ target, at a pressure of 6.6 Pa. The sputtering gas was Ar (30 sccm). The sample was mounted on a rotating support and not heated during deposition. The cathode was maintained at -850 V dc self-bias, corresponding to an effective load power of approximately 84 W.

o-Ps 3γ annihilations were measured in the capped and un-capped samples by 3γ -PAS with the Trento variable slow positron beam [14]. As a first approximation, the mean e^+ implantation depth z can be related to the e^+ implantation energy E through the equation $z = 40/\rho E^{1.6}$ with z in nanometers when the material density ρ and E are expressed in g/cm^3 and keV, respectively [15]. The ρ of the present samples was estimated to be 1.9 g/cm^3 [7]. The annihilation γ rays were detected by two high purity germanium detectors (HpGe) with 45% efficiency and 1.4 keV energy resolution at 511 keV [16]. The distribution of γ -ray energy (E_γ) was subdivided in the 511 keV peak area (P), attributable prevalently to 2γ annihilation ($|511 - E_\gamma| \leq 4.25$ keV) and in the valley area (V), due to o-Ps 3γ annihilation ($410 \leq E_\gamma \leq 500$ keV). The 3 - 2γ ratio, $R(E)=V(E)/P(E)$ parameter was calibrated by measuring the Ps formation in a Ge crystal as a function of T [17, 18]. The fraction of e^+ annihilating as o-Ps is defined

as $F_{3\gamma}(E) = \frac{3}{4} \left[1 + \frac{P_1(R_1 - R(E))}{P_0(R(E) - R_0)} \right]^{-1}$ where R_1, R_0, P_0, P_1 are the R and P values corresponding to 100%

and 0% o-Ps formation respectively [19].

Information about the e^+ and Ps pick-off annihilations were obtained by PALS measurements carried out on the capped sample. PALS was done with apparatus PLEPS (Pulsed Low Energy Positron Beam) [20] at NEPOMUC (Neutron induced POSitron source MUniCh) [21]. The detector plus pulsing system time resolution was 330 ps. Both 3γ -PAS and PALS measurements were carried out by while varying T from 150 to 430 K.

3. Results and Discussion

The $F_{3\gamma}(E)$ o-Ps fraction was measured at $E= 2, 3.3, 5$ and 7 keV in the un-capped sample and at $3.3, 5$ and 8 keV in the capped one (Fig.1). E was chosen to be higher in the capped sample in order to implant e^+ deeper than the TiO₂ layer ($\rho \sim 4$ g/cm^3) of ~30 nm, corresponding to $E \sim 2$ keV. In both samples, $F_{3\gamma}$ is roughly constant between 150 and 300 K at each E . Then it increases reaching a maximum value at T=350-400 K. At higher T, $F_{3\gamma}$ remains approximately constant at the reached

value. In the un-capped sample, the decrease of the absolute $F_{3\gamma}$ values, at a given T increasing E , is mainly due to o-Ps pick-off annihilations. In nanochannels the o-Ps diffusion length is very large, of the order of 1400 nm [7], thus o-Ps formed by e^+ implanted with $E=2$ keV have a high probability to reach the vacuum and to annihilate via 3γ . Instead, o-Ps formed deeper in the sample undergoes to many interactions with the nanochannels inner surface and the pick-off probability increases. In the capped sample, as o-Ps formed at each depth is expected to overtake roughly the same number of interactions with the channel surface, the pick-off probability should be the same. The observed $F_{3\gamma}$ decrease from $E=3.3$ keV to $E=8$ keV is due to a lower o-Ps formation at high E as an increasing fraction of e^+ annihilate in the bulk silicon underlying the nanochannels.

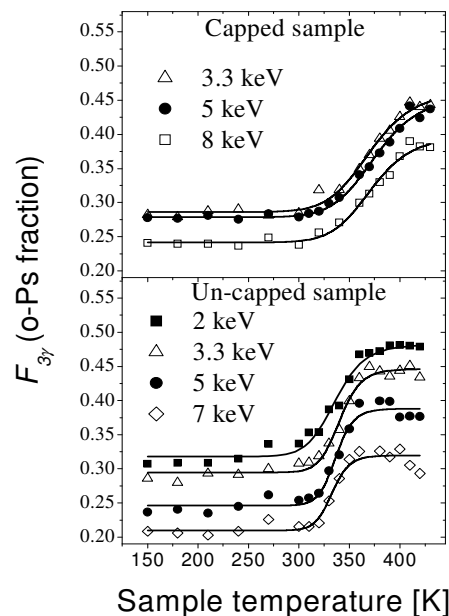


Figure 1. $F_{3\gamma}$ (o-Ps fraction) vs. sample temperature measured at different positron implantation energy in capped (upper panel) and un-capped sample (lower panel). The continuous lines are best fit to the data (see text)

Both in capped and un-capped samples, the maximum values of $F_{3\gamma}$ at $T > 350$ K are ~50% higher than the values measured in the same sample and at the same E at $T < 300$ K. This increase is sensitively higher than the increment due to the thermal activated detrapping of o-Ps from the silica surface that is expected to be less than 10% in the investigated temperature range [22]. Moreover, fitting the data of the capped and un-capped samples with the equation of thermal activated detrapping

$$F_{3\gamma}(T) = A \left(1 + B e^{-\frac{E_b}{k_b T}} \right) / \left(1 + C e^{-\frac{E_b}{k_b T}} \right) \quad (\text{where } k_b \text{ is the Boltzmann constant and the parameters } A, B \text{ and } C$$

are defined as $F_{3\gamma}(T=0) = A$ and $F_{3\gamma}(T=\infty) = \frac{A(1+B)}{1+C}$) an activation energy $E_b = 0.7 \pm 0.3$ eV, up to

eight times higher than the E_b values of 0.11-0.16 eV estimated on a single crystal SiO_2 surface [23] and in porous silica [22], is found. These findings point out that the observed $F_{3\gamma}$ increase can not be entirely attributed to thermal Ps detrapping.

To understand the origin of the observed Ps increase, PALS measurements were carried out on the capped sample. In Fig.2 the three achieved lifetimes are reported as a function of E . Beyond the TiO₂ capping layer, $E > 3$ keV, the lifetimes are constant: τ_1 , due to an average of p -Ps self-annihilation and e^+ free annihilations in Si, results $\cong 200$ ps [24], τ_2 , coming from the annihilation of e^+ trapped in the bulk silica layer at the nanochannels surface, is $\cong 420$ ps [25] and τ_3 , due to o-Ps pick-off annihilation, is $\cong 48$ ns. At $E < 3$ keV τ_3 is due to Ps annihilating in open volume in the TiO₂ capping layer while τ_1 , that reaches $\cong 250$ ps, is compatible with e^+ lifetime in amorphous TiO₂ [26].

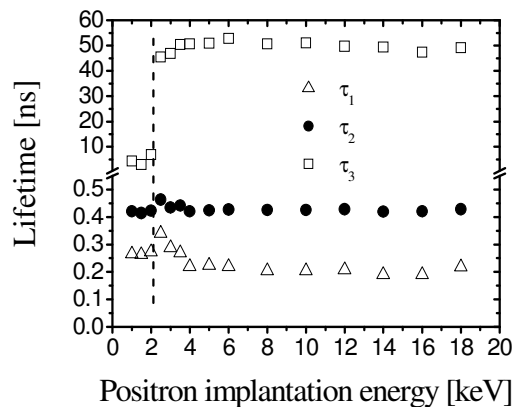


Figure 2. Positron lifetimes vs. positron implantation energy in the capped sample. Errors are inside the symbols. The vertical dashed line marks the border of the TiO₂ capping layer.

The lifetimes in the nanochannels measured as a function of T at $E=5$ keV (Fig.3), result approximately constant in the considered temperature range. Only a very small decrease of τ_3 from $\cong 48$ ns to $\cong 46$ ns is observed at $T > 350$ K.

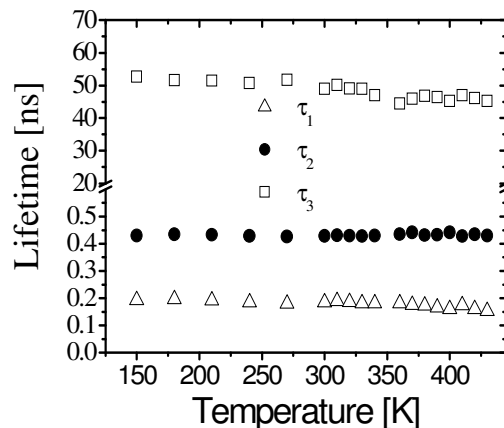


Figure 3. Capped sample: lifetimes vs. sample temperature at $E = 5$ keV. Errors are inside the symbols.

The intensities I_i , corresponding to lifetimes of Fig.3, have been normalized to take into account the o-Ps fraction annihilating via 3γ and not detected by PALS. The normalized intensities to the total number of implanted e^+ , $I_i^* = I_i [1 - F_{3\gamma}(E = 5\text{keV})]$ with i from 1 to 3, are reported in Fig.4.

$I_1^* \cong 20\%$, $I_2^* \cong 40\%$ and $I_3^* \cong 15\%$ as the total fraction $F_T = F_{3\gamma}(E = 5\text{keV}) * 100 + I_3^*$ of formed o-Ps are constant below $T = 300\text{ K}$ (see Fig. 4). Above 300 K it is observed : i) an increase of F_T mainly due to the increase of $F_{3\gamma}$, ii) a strong decrease of I_2^* pointing out the decrease of e^+ annihilation in SiO_2 , iii) a small decrease of I_1^* and I_3^* , the last due to the decrease in o-Ps pick-off. F_T was fitted by using the equation of the thermal activated detrapping (see above) in order to find a more accurate value of E_b not altered by pick-off (Fig.4). The E_b pointed out by this fitting procedure is $0.64 \pm 0.15\text{ eV}$.

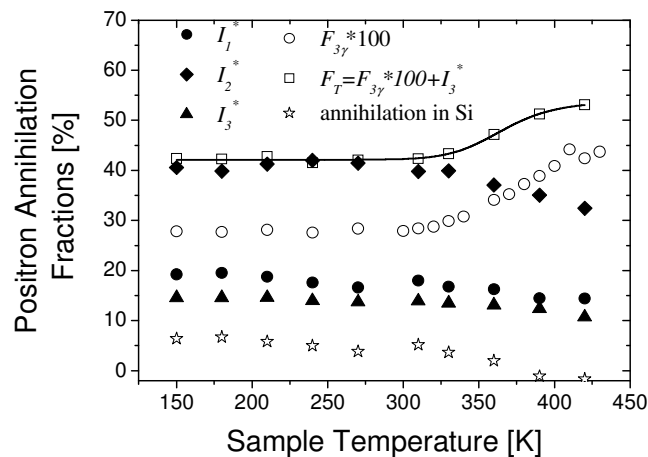


Figure 4. e^+ and Ps annihilation fractions vs. sample temperature. The fractions have been estimated according to the procedure described in the text for the capped sample at $E = 5\text{ keV}$. The continuous line is the best fit of the total o-Ps fraction formation (see text).

The fraction of e^+ annihilating in Si can be estimated as $I_1^* - 1/3 * F_T$ where $1/3 * F_T$ is the p-Ps fraction taking into account the p-Ps:o-Ps ratio 1:3. As appreciable by Fig. 4, this fraction is around 5-7 % at $T < 300\text{ K}$ and it approaches to zero at higher T.

In light of the data presented in Fig. 4, we can conclude that the observed increase of Ps is related to: (a) a decrease of e^+ annihilating in SiO_2 and (b) an increment of e^+ that reach SiO_2 layer coming from Si. Both this effects could be ascribed to a modification of the charged defects at the Si/SiO₂ interface and in the SiO₂ layer where it changes the electronic structure at the walls of the nanochannels. At room temperature the p-type Si /SiO₂ interface is positively charged due to donor's levels above the Fermi level and it limits e^+ reaching the surface [27]. The effect of the temperature appears to be that of lowering the positive charge promoting more positrons reaching SiO₂. Positrons in the thin SiO₂ layer and/or at the SiO₂ surface must be also favored to form Ps. This could occur with the formation of thermally generated negatively charged dangling bonds [28].

4. Conclusion

In the present paper, we have studied the o-Ps formation as a sample temperature (range 150-430 K) performed by 3γ -PAS and PALS on Si samples with oxidized nanochannels synthesized by electrochemical etching. The measurements point to an increase of Ps formation between 300 and 400 K. This increase of Ps yield is associated to a decrease of the fraction of e^+ annihilating in silica and the disappearing of e^+ annihilation in the Si substrate. These combined effects has been interpreted as a thermal charge rearrangement at the Si/SiO₂ interface and at the SiO₂ nanochannels surface at high T that contrasts the positive electric field present in Si p-type. Future measurements in similar samples

etched in n-type Si will be clarifying. In fact, if the above hypothesis is correct, a higher Ps formation at $T \leq 300$ K would be expected in n-type Si where at room temperature e^+ diffusion towards the Si/SiO₂ interface is favored [27]. As a consequence, the Ps increase at $T > 300$ K would be less pronounced.

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