Origins of positronium emitted from SiO$_2$

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The time-of-flight of positronium emitted from single-crystal and amorphous SiO$_2$ has been measured using a high-intensity pulsed positron beam at KEK. For both the samples, two components of peak energies around 1 and 3 eV have been observed. They are attributed to the Ps formed in the bulk and on the surface of the SiO$_2$, respectively. [S0163-1829(98)03340-2]

It is well known that many solids emit positronium (Ps) when bombarded with low-energy positrons.[1,2] The emission is a surface effect in the case of metals. In the case of insulators, Ps formed in the bulk is also emitted.[3−11] The Ps emission energy reflects the band structure of the insulator, Ps state in the bulk, Ps formation mechanism, and work functions of the electrons and positrons.

The Ps emission energy from SiO$_2$ has been measured by using a time-of-flight (TOF) technique for the single-crystal surface[7] and by the angular correlation of annihilation radiation (ACAR) method for silica aerogel.[6,9,10] In the TOF measurement[7] the sample was bombarded with a slow positron beam through a microchannel plate array (MCP) with a hole. The time interval between the emission of secondary electrons from the sample and the pickoff annihilation of the emitted orthopositronium (o-Ps) on the MCP was measured to obtain the energy distribution. The distribution thus measured is peaked at 3.27 eV.

The ACAR method also provides the Ps emission energy from the SiO$_2$ surface when it is combined with the use of silica aerogel.[6,9,10] This material is an aggregate of amorphous SiO$_2$ fine grains. When positrons from β^+ decay enter this material, they lose energy in the grains and are then emitted as Ps atoms into the free space. Because the part of the parapositronium (p-Ps) energy lost in its short lifetime is only a few percent of the initial energy, the p-Ps component of the ACAR data shows the momentum distribution of the Ps emitted from the grain surface. The Ps emission energy distribution obtained using this method is peaked at 0.8 eV.

In the present work, we have measured the TOF of the o-Ps from a SiO$_2$ surface using an intense, pulsed, monoenergetic positron beam at KEK (Refs. 12 and 13) in order to investigate the discrepancy between these two results.[11] Positrons were obtained from bremsstrahlung pair-production at the beam dump of a 2.5 GeV electron linac. The repetition rate of the linac was 25 Hz and the pulse width of the electron beam was 10 ns. An array of tungsten foils was used to moderate the positrons to eV energies. The positrons were transported to the chamber after being accelerated to an energy of 9 keV in order to minimize pulse widening. The width of the positron pulse was 23 ns and each pulse contained 10^6 positrons.

Details of the TOF chamber are described elsewhere.[11] It is similar to the setups in previous works.[8,14] The beam was collimated by a copper disk with a hole of 10 mm in diameter (e^+ collimator) before entering the chamber. The beam diameter near the sample was measured to be 11 mm using MCP. The sample bias was varied in a range of 0–9 kV to control the incident energy of the positrons using a retarding element (RET) composed of 4 electrodes with a hole of 40 mm in diameter. An aluminum disk with a hole of 30 mm in diameter (Ps collimator) was placed in front of the RET in order to collimate the emitted Ps to within 25 degrees of the normal to the sample surface. The base pressure of the chamber was about 1×10^{-6} Pa.

The samples were SiO$_2$ single crystal (obtained from Toyocom Co.) and amorphous SiO$_2$ (Suprasil P-10 supplied by Shin-Etsu Quartz Products Co. Ltd.). The dimensions of the samples were 20 mm$\times$20 mm$\times$5 mm. The surface of the single crystal was normal to the c axis. We have also measured the Ps emission energy from KI and MgO single crystals. They were obtained from the University of Utah
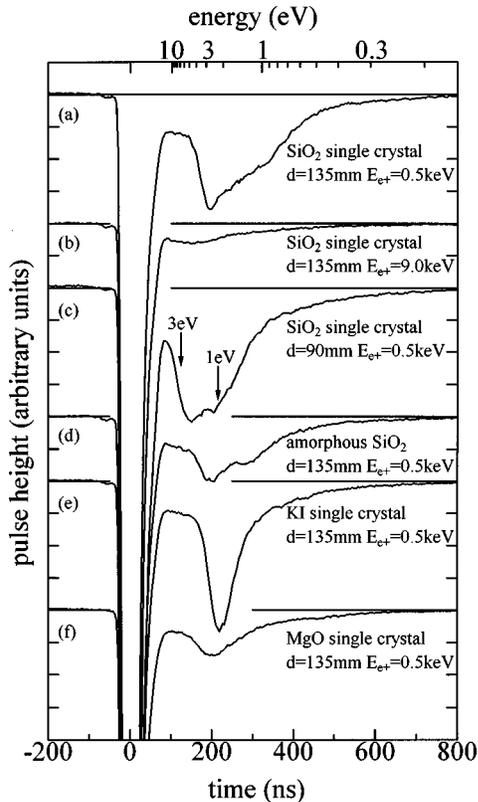
FIG. 1. Time-of-flight spectra. The Ps energy on the top is only for the case $d=135$ mm. For the case $d=90$ mm (c) the arrows indicate the time corresponding to 1 and 3 eV. All the data are normalized to the measurement time.

and Electric Space Products International Ltd., respectively. The dimensions were the same as those of the SiO_2 samples. The surfaces of these samples were normal to the $\langle 100 \rangle$ axis.

The time interval between the linac pulse signal and the detection of the γ ray from o-Ps self-annihilation in flight was measured using digital oscilloscopes with a sampling speed of 5 GS/s (Tektronics TDS-684B). Only the γ rays from the self-annihilation of the o-Ps in the view of annular plastic scintillator were detected through a lead slit of 4.5 mm width. The scintillator was 600 mm in diameter and 10 mm in thickness and was coupled on opposite sides to two photomultiplier tubes (HAMAMATSU H1949) through lucite light pipes. The distance between the sample and the region visible by the γ-ray detector was adjusted by moving the sample holder together with the RET and the Ps collimator. Each anode signal was connected to the 50-Ω input of the two oscilloscopes separately. The pulses from the electron linac were delayed and connected to the trigger inputs. The traces for the 10^4 linac pulses were averaged in the oscilloscopes and transferred to a personal computer. Some of the averaged traces for the same condition were further summed numerically.

Figure 1 shows the averaged traces (TOF spectra). The distance between the sample and the detector slit is denoted by d. The positron incident energy, which is obtained from the difference between the positron transport energy (9 keV) and the voltage of the sample holder, is denoted by E_{e+}. (There might be a small error in the estimation of E_{e+} because of possible charge buildup of the sample surfaces.
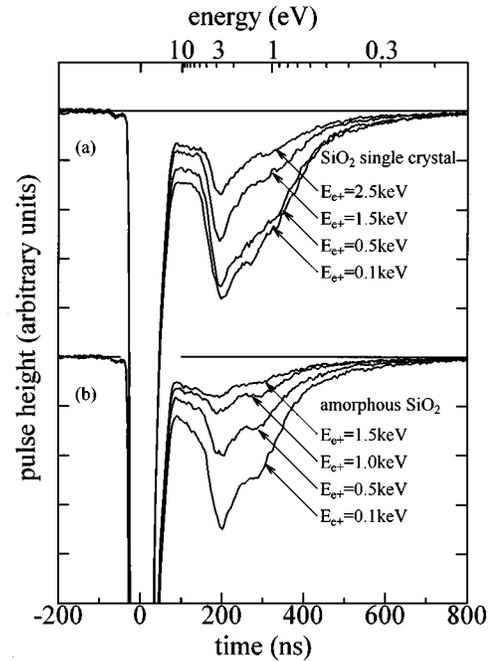

FIG. 2. The positron incident energy dependence of the TOF spectra for (a) SiO_2 single crystal and (b) amorphous SiO_2 (Suprasil P-10). All the data are normalized to the measurement time.

However, it does not affect the following discussion.) All the data were normalized to the measurement time.

The SiO_2 single crystal data for $d=135$ mm and $E_{e+}=0.5$ keV shows two delayed components following a large peak from the prompt annihilation of the positron pulses in the sample. They correspond to Ps energies of about 3 and 1 eV. When E_{e+} is raised to 9 keV, these peaks disappeared. The two peaks also exist in the spectrum taken at $d=90$ mm. The spectra for the amorphous SiO_2 also shows two components. The corresponding energies are close to 3 and 1 eV, though the energy of the lower-energy peak is slightly higher than that for the single crystal.

Only one component was observed in the KI and MgO data. The peak energies are 2.1 and 2.6 eV, respectively. Figure 2 shows the incident energy dependence of the TOF spectra for SiO_2 single crystal and amorphous SiO_2. The data were normalized to the measurement time.

The two components for SiO_2, with the peak energies of 3 and 1 eV, appear to correspond to the observations by Sferlazzo et $al.$[7] and Chang et $al.$,[6,9,10] respectively. The 1-eV component was not observed by Sferlazzo et $al.$[15] This might be because the Ps detection efficiency by the pickoff annihilation on the MCP was too low for the energies lower than 3 eV. The component of 3 eV was not observed by Chang et $al.$ We will discuss this below.

The peak energy for KI is consistent with the measurement by Tuomisaari et $al.$[8] The existence of only one component peaked at the energy different from those for SiO_2 shows that the two peaks for SiO_2 are not due to systematic effects of the experimental setup. The peak energy for MgO is lower than the value reported by Sferlazzo et $al.$[7] The reason for this discrepancy is unknown.

Figure 2 shows that the positrons implanted with higher energy penetrate deeper into the SiO_2 samples and have lower probabilities of diffusing out of the surface before they

annihilate in the bulk. It also shows that the diffusion length for the amorphous is shorter than that for the single crystal. We did not analyze the data to obtain diffusion length for each component because it is difficult to decompose the spectra into the two components. However, it is to be noted that the energy dependences of the components of 3 and 1 eV seem to be different. This implies that the components are from different origins.

One of the two components for SiO_2 can be attributed to the emission of Ps which is formed in the bulk, and the other to the emission of Ps which is formed on the surface. We attribute from the following discussion the 1-eV component to the former and the 3-eV component to the latter.

If the Ps is formed in the bulk and thermalized before emission, then the emission energy is written as

$$T_{bulk} = -\phi_+ - \phi_- + E_G - E_B + 6.8 \text{ eV},\qquad(1)$$

where ϕ_+, ϕ_-, E_G, and $E_B (<6.8\,\text{eV})$ are the positron and the electron work functions, the band gap, and the Ps binding energy in the bulk, respectively. The quantity $\phi_{Ps} = -T_{bulk}$ (<0) may be called Ps work function. If the Ps is formed on the surface from a positron thermalized in the sample, two cases are possible. The emission energy of the Ps that is formed from the valence electron has a maximum of

$$T_{surface}^v = -\phi_+ - \phi_- + 6.8 \text{ eV}.\qquad(2)$$

If, on the other hand, there exist thermalized electrons in the conduction band at the surface, the energy of the Ps formed from such an electron is

$$T_{surface}^c = -\phi_+ - \phi_- + E_G + 6.8 \text{ eV}.\qquad(3)$$

Note that

$$T_{bulk} - T_{surface}^v = E_G - E_B > 0,\qquad(4)$$

because the reported value of E_G for SiO_2 is in the range 9–11 eV.[16,17] This leads to $T_{bulk} > T_{surface}^v$. Note also that

$$T_{surface}^c - T_{bulk} = E_B > 0,\qquad(5)$$

which leads to $T_{surface}^c > T_{bulk}$. Thus our data imply either (i) $T_{bulk} = 1$ eV and $T_{surface}^c = 3$ eV, or (ii) $T_{bulk} = 3$ eV and $T_{surface}^v = 1$ eV.

In the case (i), it is easy to explain why the 3-eV component is not visible in the ACAR data; the Ps emitted from a grain surface with 3 eV penetrates shallowly into and re-emerges from other grains with lower energy. Once the energy reaches 1 eV, it can never enter another grain and hardly loses energy before it annihilates with lifetime of 125 ps. In this case, positron work function can be given from Eq. (3) as

$$\phi_+ = -\phi_- + E_G + 6.8 \text{ eV} - T_{surface}^c \sim 3 \text{ eV}\qquad(6)$$

because the value of $\phi_- - E_G$ is reported to be about 1 eV.[18] The positive value of ϕ_+ is consistent with the positron lifetime spectra for silica powders or aerogel[19] where no component attributable to the slow positrons annihilating in the free space is observed. The value of E_B is estimated from Eq. (5) to be 2 eV, which is smaller than that derived by Sferlazzo et al. (4.8 eV).[7] However, if we assume that the Ps is formed on the surface by electron pickup from excitons or surface states,[8] then E_G in Eq. (3) is replaced by an excitation energy smaller than E_G and the estimated value becomes larger, up to 5 eV (limited by the positiveness of ϕ_+). Furthermore, the value of T_{bulk} is consistent with the effective potential barrier height against o-Ps penetration into the SiO_2 estimated from positron lifetime data for SiO_2 powders.[4]

In the case (ii), the reason for the nonexistence of 3 eV component in the ACAR data cannot be explained easily. The positron work function is obtained from Eq. (2):

$$\phi_+ = -\phi_- + 6.8 \text{ eV} - T_{surface}^v.\qquad(7)$$

This value is estimated to be $-4--6$ eV by using the reported value of ϕ_- (10–12 eV).[16,17] The negative value of ϕ_+ implies that not only Ps but also slow positrons would be emitted from the surface. This is not consistent with the positron lifetime spectra for silica powders or aerogel.

In conclusion, we have measured the TOF of the Ps emitted from single-crystal and amorphous SiO_2 using a high-intensity pulsed positron beam at KEK. The data indicate that Ps is emitted by two mechanisms: emission of Ps formed in the bulk and Ps formation on the surface, with energies 1 and 3 eV, respectively.

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