

State Conversion of Positronium by Oxygen

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Using a silica aerogel sample filled with oxygen gas as a variable energy positronium source, the effect of the positronium motion in oxygen has been investigated with a time selecting energy spectrometer. The positronium state conversion in an inelastic collision between the fast positronium atom and oxygen molecule has been verified and the conversion cross sections are measured as $2.1 \times 10^{-17} \text{ cm}^2$ and $6.6 \times 10^{-18} \text{ cm}^2$ respectively at the kinetic energies of positronium near two thresholds. For the slow positronium, the conversion cross section due to the exchange of electrons in the elastic collision is found to be proportional to $1/\sqrt{v}$, where v is the mean velocity of positronium. This process would provide a clean para-positronium source for other precise experiments.

1. INTRODUCTION

Oxygen is a para-magnetic material, the ground state of O_2 molecule is triplet $X^3\Sigma_g^-$, while the two of the outer electrons are unpaired and have parallel spins. The first and the second excited states are singlet, $a^1\Delta_g$ and $b^1\Sigma_g^+$, which are located at 0.977 eV (E_a) and 1.62 eV (E_b) above the ground state respectively. Deutsch and Berko [1] pointed out that an inelastic collision with spin exchange may occur between the positronium (Ps) atom and the O_2 molecule while the kinetic energy of Ps exceeds the excited state energies of O_2 , resulting that O_2 is excited and Ps is converted from ortho-positronium (o-Ps) to para-positronium (p-Ps), the so called o-p conversion, or para to ortho (p-o conversion). Kakimoto *et al.* [2] studied this inelastic process at different pressures of O_2 by using a high resolution 1D ACAR. In addition, for the slow Ps atoms, the state conversion (specially o-p conversion) can also take place in the elastic collision through a electron exchange (see Ferrell [3]) between Ps and O_2 . So far, the elastic process was restricted to a rough study in a temperature changing experiment [4] because no variable energy of the Ps beam was available. In the investigation work [5] of Ps motion in silica aerogel at vacuum, we found that [2] Ps atoms were emitted with a

Table 1
The annihilation parameters of positron in
Silica Aerogel filled with Oxygen gas

| O ₂ (atm) | 0 | 0.2 | 0.4 | 0.6 | 0.8 | 1.0 | Number of Annihilation |
|------------------------|----------------|-----------------|-----------------|------------|--------------|--------------|------------------------|
| Intensity and lifetime | | | | | | | |
| $I_1, \%$ | 68.7 | 73.9 | 74.3 | 74.5 | 75.2 | 75.1 | 2 γ |
| $I_2, \%$ | 2.8 | 2.3 | 2.2 | 3.2 | 3.2 | 3.3 | 2 γ |
| $I_{31}, \%$ | 0.3 | 2.9 | 4.3 | 5.4 | 5.9 | 6.3 | 2 γ |
| $I_{32}, \%$ | 1.6 | 10.1 | 12.2 | 12.4 | 12.4 | 12.5 | 2 γ |
| $I^{3\gamma}, \%$ | 26.6 | 11.7 | 7.0 | 3.7 | 3.7 | 3.1 | 3 γ |
| $I_{O-Ps}, \%$ | 31.3 | 26.1 | 25.7 | 25.5 | 24.8 | 24.9 | 2 γ +3 γ |
| τ_1, ns | 0.54 | 0.54 | 0.57 | 0.54 | 0.53 | 0.57 | |
| τ_2, ns | 2.6 | 2.8 | 2.8 | 2.6 | 2.6 | 2.8 | |
| τ_{31}, ns | 17 | 24.6 | 18.9 | 17.1 | 15.0 | 12.2 | |
| τ_{32}, ns | 134 ± 2 | 80.0 ± 2 | 54.8 ± 1 | 46 ± 1 | 38.9 ± 1 | 32.2 ± 1 | |

higher kinetic energy from the surface of the fine SiO₂ grains of silica aerogel to the free space between the grains, are slowed down and then thermalized through a sequence of collisions with the grain particles. The gradual loss of the Ps kinetic energy seems a variable energy Ps source, we have used it to carry out a new observation presented here for the state conversion of Ps by O₂.

2. EXPERIMENT

2.1 Sample and Positron Source

Two pieces of low density (0.1 g/cm³) silica aerogel with the size $\phi 14$ mm \times 10 mm are used as the sample. For the long lifetime component (> 10 ns) measurement, the positron source is 1 μ Ci of ²²Na, which is dropped onto a plastic scintillator foil and then sandwiched by the sample. The sample is put into a Al tube, for which one end is transparently coupled with a photomultiplier to form a β^+ trigger counter and the other end is connected to a gas (oxygen) system. For the short lifetime component (\sim ns) measurement, the positron source is 5 μ Ci of ²²Na, which is dropped onto a thin Al foil and then sandwiched by the sample. The sample with the tube is put into a big well-type plastic scintillator which is the start detector to detect 1.28 MeV γ -ray of ²²Na.

2.2 Arrangement

The experimental equipment is a Time Selecting Energy Spectrometer (TSES) which consists of a Ge γ -ray energy spectrometer and a positron lifetime spectrometer, and can also be used for the measurements of lifetime spectrum and Doppler broadening spectrum separately. The TSES has

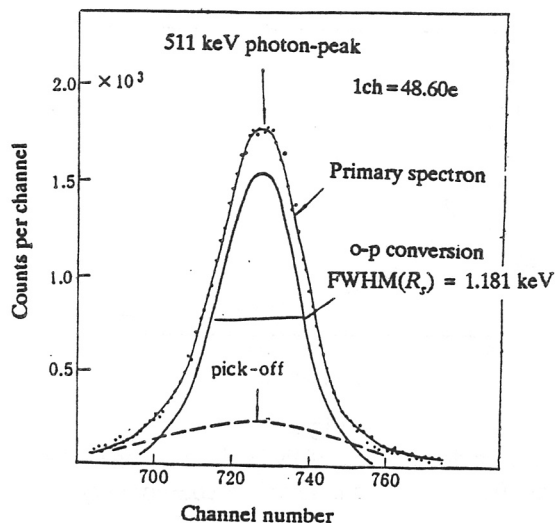


Fig.1

The Doppler spectrum of the annihilation 2γ of p-Ps from o-p conversion, the age of o-Ps is 9.8 ns--12.2 ns, sample filled with 0.2 atm O_2 .

8 time windows which simultaneously collect 8 Doppler spectra corresponding to the different ages of Ps. For the long lifetime measurement, two detectors are used in TSES and the electronics is same as that in Ref. [5]. The γ -ray energy detector is HPGe ($\phi 44$ mm \times 15 mm) with energy resolution $FWHM = 1.089$ keV for 512 keV γ -ray of ^{106}Ru . The start detector in the lifetime system is the β^+ trigger counter and the stop detector is also the HPGe. The time resolution has been now improved to $FWHM = 2.4$ ns because the magnetic field is not needed due to the direct observation of o-p conversion. For the short component measurement, three detectors are used and the electronics is similar to that in Ref. [6], in which the start detector is the well-type plastic scintillator, the stop one is a BaF_2 counter and the time resolution is $FWHM = 0.4$ ns.

2.3 Measurement of the Lifetime Spectra

The lifetime spectra of positron annihilation have been measured at the O_2 pressure of 0, 0.2, 0.4, 0.6, 0.8 and 1.0 atm respectively. Each of the spectra has been resolved to three components: The short component represents essentially the positron free annihilation and the p-Ps decay, its lifetime $\tau_1 = 0.54$ ns keeps constant at various pressure of O_2 but the intensity I_1 noticeably changes. The medium component represents the pick-off process of o-Ps inside the SiO_2 grains of the silica aerogel and both the lifetime $\tau_2 = 2.7$ ns and intensity $I_2 \approx 2.8\%$ do not change with O_2 pressure. For the long component, which represents the o-Ps quenching by the collision with O_2 which filled in the free space between the grains, both the lifetime and intensity change noticeably with oxygen pressure. Furthermore, τ_3 splits into two parts, τ_{31} and τ_{32} , due to the non-thermalized effect [5]. The "peak method" [7] has been used to get the 3γ intensity which is essentially involved in τ_3 . The method of 2γ lifetime spectrometry [5] has been used to get the fraction of 2γ events in every component. All of the annihilation parameters measured are presented in Table 1.

2.4 Measurement of the Time Selecting Doppler Spectra

8 Doppler spectra are obtained in one run of the TSES measurement. For example, one of the spectra is shown in Fig.1, for which the sample is the silica aerogel filled with 0.2 atm high purity O_2 and the time window is 9.8--12.2 ns (a mean age of 11 ns for o-Ps). After subtraction of the random and 3γ continuous background, a very clear peak of the annihilation radiation appears at

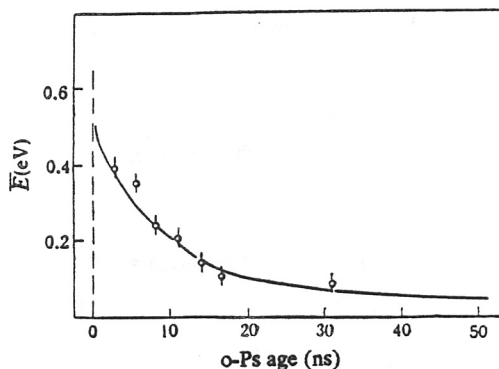


Fig.2

The slowing down curve of Ps in silica aerogel filled with 0.2 atm O_2 (long life time component).

ϕ is experiment point;
 — is a calculated curve based on the elastic collision model.

511 keV. Referring the response function of HPGe (for 512 keV γ of ^{106}Ru , the response curve is nearly a gaussian with FWHM = 1.089 keV in a short time measurement, FWHM = 1.096 keV for a longer time measurement and the ratio of the peak to valley 130:1), the spectrum in Fig.1 is resolved to two parts; the wider one is similar to the spectrum measured at vacuum, which is caused by the pick-off process of o-Ps and the narrower one is like a gaussian and its width FWHM(Rs) = 1.181 keV is slightly wider than that of the mono-energy γ -ray, which is the p-Ps peak caused by

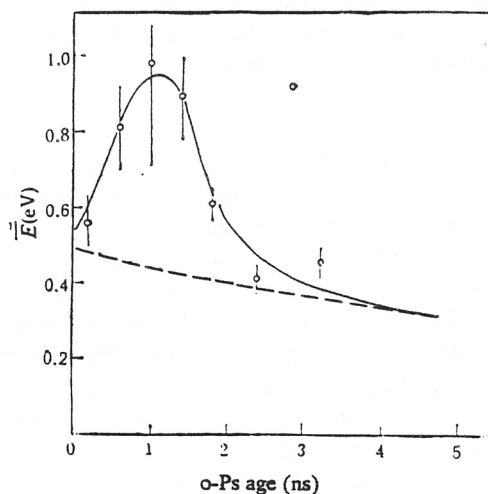


Fig.3

The kinetic energy curve of Ps in silica aerogel filled with 0.2 atm O_2 (the short component).

ϕ is the experiment point;
 — is a fitting curve;
 — is a calculated curve based on the elastic collision model.

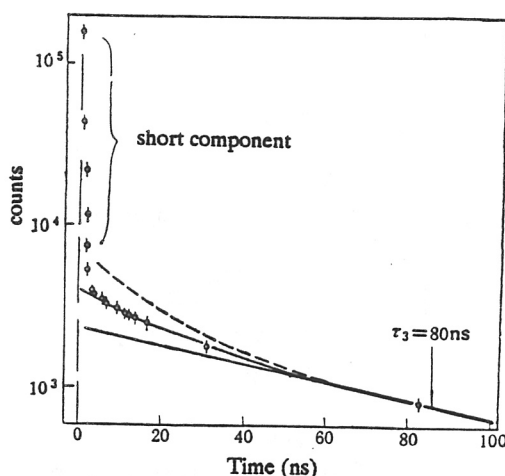


Fig.4

The lifetime spectrum of o-p conversion,
silica aerogel filled with 0.2 atm O₂.

- ϕ experiment point;
- calculated curve, assuming $\sigma_{o-p} = \text{constant}$;
- - - calculated curve, assuming $\sigma_{o-p} \propto 1/\sqrt{v}$;
- · - calculated curve, assuming $\sigma_{o-p} \propto 1/\sqrt{v}$

the o-p conversion at the collision of o-Ps with O₂. Two run (at 0.2 atm and 1.0 atm of O₂) TSES measurements have been performed and so a series of p-Ps peaks and FWHM(Rs) are obtained.

For the silica aerogel Ps source, the initial kinetic energy of Ps is a distribution from 0--4 eV with its maximum at ~ 1 eV [2]. It is roughly like a Boltzmann distribution. Because the kinetic energy eventually becomes a real Boltzmann distribution of room temperature due to the thermalization of Ps atoms, we conceive that the kinetic energy of Ps atoms at any age resembles a Boltzmann distribution but with a different temperature. According to Ref. [6], the Doppler spectrum of the annihilation radiation for Ps atoms with Boltzmann distribution is a Gaussian, we assign its width as FWHM(Ps). Obviously, the relation between FWHM(Ps) and the measured FWHM(Rs) is

$$(\text{FWHM(Rs)})^2 - (\text{FWHM(Ge)})^2 \quad (1)$$

where FWHM(Ge) refers to the width of the response function of HPGe, which can be obtained from a measurement of ¹⁰⁶Ru. According to a formula in Ref. [6], the root-mean-square kinetic energy \bar{E} of Ps atoms at any age is

$$\bar{E} = (1.0289 \text{FWHM(Ps)})^2 \quad (2)$$

The relation between the Ps kinetic energy and Ps age for the long component, which is obtained from the measured values FWHM(Rs) and equation (1) and (2), is shown in Fig.2. A calculated curve based on the elastic collision model [5] is also shown in Fig.2 for comparison. A good agreement in Fig.2 means that the energy loss of Ps is through the elastic process, so that \bar{E} in equation (2) is the p-Ps kinetic energy after o-p conversion, which is very close to the kinetic energy of o-Ps before the conversion.

The measured curve of the kinetic energy \bar{E} for the short component and a calculated curve (dash line, elastic collision model) are shown in Fig.3. The disagreement in 0--2 ns indicates that the

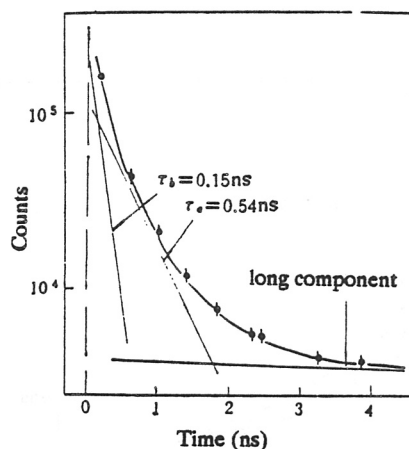


Fig.5

The lifetime spectrum of o-p conversion (short component),
silica aerogel filled with 0.2 atm O_2 .

o-p conversion occurs through inelastic collision with O_2 . In the figure, \bar{E} means the kinetic energy of p-Ps after o-p conversion, so the kinetic energy of o-Ps before conversion should be the sum of E and the excitation energy of O_2 . The fact, $E \sim 0.6$ eV at $t = 0$ and $E \sim 0.9$ eV at $t = 1$ ns, reveals that the rate of the o-p conversion in the case that O_2 transforms from its ground state to the second excited state ($E_b = 1.62$ eV) is much higher than the rate of the first excited state ($E_a = 0.97$ eV). The former process is ended earlier and in which the o-Ps atom consumes more kinetic energy and leaves a lower energy to the converted p-Ps and this is why $E(t = 0) < E(t = 1$ ns). However, all of the o-Ps with energy less than E_a or the o-Ps produced from p-o conversion will lose energy slowly through a sequence of elastic collisions.

All of the peak areas of the converted spectra (e.g. the narrower peak in Fig.1) have been counted, and normalized. The result at the 0.2 atm O_2 condition (so called the lifetime spectrum of o-p conversion) is shown in Fig.4. The spectrum can be divided into two parts: the short component corresponds to the o-p conversion with the inelastic collision process and the long component to the elastic process. Drawing the short component in a smaller time scale (see Fig.5), it can be seen that the curve (after a subtraction of the long component) seems to be composed of two exponentials: $\tau_b \approx 0.15$ ns and $\tau_a \approx 0.54$ ns. We think they correspond to two o-p conversion processes, which are related to the O_2 molecules excited to E_a and E_b respectively. The long component in Fig.4 is also not a mono-exponential. Neglecting the very weak pick-off process (the rate $\lambda_{\text{pick-off}}$) and considering the constant o-Ps decay rate λ_0 , the non-mono-exponential decay may be attributed to the dependence of the $\lambda_{\text{o-p}}$ conversion rate on the o-Ps mean velocity v . For further discussion we name $\sigma_{\text{o-p}}$ as the cross section of o-p conversion. Three calculated curves are shown in Fig.4: 1) assuming $\sigma_{\text{o-p}} \propto 1/v$, the rate $\lambda_{\text{o-p}} \propto v \cdot \sigma_{\text{o-p}}$, where $v \propto \bar{v}$ is the collision frequency of o-Ps with O_2 , so $\lambda_{\text{o-p}} = \text{constant}$ and the total annihilation rate $\lambda_3 = \lambda_0 + \lambda_{\text{o-p}} + \lambda_{\text{pick-off}} \approx \text{constant}$, the curve is a mono-exponential, which is in disagreement with the experiment; 2) assuming $\sigma_{\text{o-p}} = \text{constant}$, $\lambda_{\text{o-p}} \propto v \propto \bar{v}$, putting the values of v (we know it from Fig.2) in calculation, the calculated curve is a dash line shown in Fig.4, which is also in disagreement with the experiment; 3) assuming $\sigma_{\text{o-p}} \propto 1/\sqrt{v}$, the calculated curve fits the experimental points.

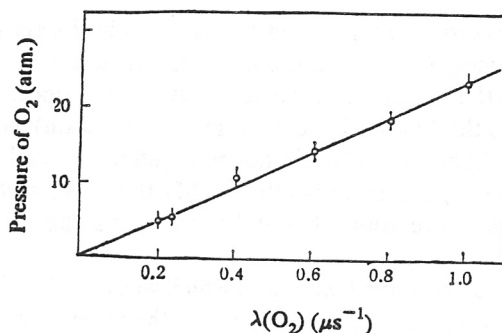


Fig.6

The annihilation rate of the thermalized o-Ps in O₂.

3. CALCULATION OF THE CROSS SECTION OF o-p CONVERSION

It is well known that the relation between the annihilation rate λ and the annihilation cross section σ is expressed as:

$$\sigma = \frac{\lambda}{vn} \quad (3)$$

For the interaction of Ps with O₂ we consider here, n in equation (3) refers to the molecule number of O₂ in unit volume and v is the mean velocity of Ps.

At room temperature, the mean velocity of the thermalized Ps atoms is $v(\text{therm}) = 0.7545 \times 10^7$ cm/s, and the molecule number of 1 atm O₂ is $n = 2.687 \times 10^{19}$ cm⁻³, then $\sigma_{o-p}(\text{therm})$, the cross section of o-p conversion for the thermalized o-Ps atoms, can be obtained if we know $\lambda_{o-p}(\text{therm})$ which is the o-p conversion rate of the thermalized o-Ps atoms at room temperature. From the τ_{32} data in Table 1, $\lambda_{32} = 1/\tau_{32}$ is the total rate of the thermalized o-Ps in the sample (silica aerogel + O₂). Subtracting the rate in silica aerogel ($1/\tau_{32}$ in vacuum), the total rates in pure O₂ at various O₂ pressures, $\lambda(\text{O}_2)$, are obtained and presented in Fig.6. $\lambda(\text{O}_2) = 23.60 \mu\text{s}^{-1}$ for 1 atm O₂ is found. Because $\lambda_{o-p}(\text{therm}) = \lambda(\text{O}_2) - \lambda_{\text{pick-off}}(\text{O}_2)$, where $\lambda_{\text{pick-off}}(\text{O}_2)$ refers to the pick-off rate of the thermalized o-Ps in 1 atm O₂ and its value of $0.96 \mu\text{s}^{-1}$ is obtained from a comparison between the Doppler spectra (like in Fig.1) of the thermalized o-Ps at different pressures of O₂, the value $\lambda_{o-p}(\text{therm}) = 22.64 \mu\text{s}^{-1}$ is obtained and the cross sections are:

$$\begin{aligned} \sigma_{o-p}(\text{therm}) &= (1.1 \pm 0.1) \times 10^{-19} \text{ cm}^2 \\ \sigma_{\text{pick-off}}(\text{O}_2) &= (4.6 \pm 0.6) \times 10^{-21} \text{ cm}^2 \end{aligned} \quad (4)$$

where the experimental errors are essentially from the errors in λ measurements. The cross section for any o-Ps velocity can be deduced according to the $1/\sqrt{v}$ relation.

The annihilation rates of the two inelastic processes are obtained as $\lambda_a = 1/\tau_a = 1852 \mu\text{s}^{-1}$ and $\lambda_b = 1/\tau_b = 6667 \mu\text{s}^{-1}$ (see Fig.5). The mean velocity of Ps is estimated from Fig.3, then the o-p conversion cross sections in the inelastic collisions of o-Ps near the two excited thresholds of O₂ molecule are:

$$\begin{aligned} \sigma_{o-p}(E_b) &\sim 2.1 \times 10^{-17} \text{ cm}^2 \\ \sigma_{o-p}(E_a) &\sim 6.6 \times 10^{-18} \text{ cm}^2 \end{aligned} \quad (5)$$

4. DISCUSSION

1. Regarding the o-p conversion cross section $\sigma_{o-p} \propto 1/\sqrt{v}$ in the elastic process, a qualitative interpretation is given as following: Ps atom on the whole is a neutral particle, σ_{o-p} depends on the "stay time" of Ps atom around the O_2 molecule, which is inversely proportional to the velocity v of o-Ps, so that $\sigma_{o-p} \propto 1/\sqrt{v}$ (like the $1/v$ law in neutron nuclear reaction) would be expected. But in fact, Ps atom is an electrical dipole, the electric-magnetic effect excited by the passing of o-Ps through the electron clouds of O_2 molecule would modify the cross section σ_{o-p} close to $1/\sqrt{v}$ dependence. A quantitative interpretation should be based on the quantum electrodynamics calculation.

2. Considering the deceleration curve (see Fig.2) which was calculated by using a free collision model (collision of a free o-Ps with O_2 and SiO_2 molecules), the agreement between calculation and experiment indicates that no bound state of o-Ps has been formed with the molecules of O_2 and SiO_2 . This is also supported by the fact that the annihilation rate of o-Ps in O_2 varies linearly with oxygen pressure (see Fig.6). The kinetic energy shown in Fig.2 is the centre of mass kinetic energy of the converted p-Ps, which is only 0.2 eV at $t = 10$ ns. From equation (4) we know that the intensity of the pick-off process (as a background) is only 1/20 of that in o-p conversion process, so the silica aerogel filled with oxygen is a clean p-Ps source which is suited for a precise experiment such as the annihilation 2γ energy measurement.

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