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Energy Loss of Positrons below the Excitation Threshold in Ar Gas

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We performed positron age-momentum correlation measurements to investigate the positron slowing down process in Ar gas at 7.5 MPa. Increase in the *S* parameter was observed up to 1 ns, and then stayed on the same level. By comparing with the calculated energy loss of positrons via elastic scattering, it was concluded that the increase in the *S* parameter corresponded to the positron slowing down from 11.6 eV to 2.5 eV. The positron slowing down process just below the first electronic excitation energy of Ar gas can be observed using the *S* parameter.

1. Introduction

A positron, the antiparticle of an electron, having the same mass and opposite sign of charge to the electron has been extensively studied in atomic physics as an exotic atom and molecule [1–7]. The positron annihilation rate changes depending on the local electron density. This phenomenon has been used for probing materials [8–12]. For studies of atomic physics and material sciences, the positron kinetic energy should be as low as chemical reactions occur. On the other hand, positrons are obtained from high energy reactions, and the initial positron kinetic energy is a few orders of magnitude higher than suitable energies mentioned above. Energetic positrons injected into a substance slow down by ionization and excitation of surrounding molecules. During the slowing down process in gas, some of the positrons form positronium. Positronium (Ps) is a bound state of a positron and an electron, and has two spin eigenstates depending on the total spin angular momentum quantum number (S): a spin-singlet state ($S = 0$, *para*-positronium, *p*-Ps) and a spin-triplet state ($S = 1$, *ortho*-positronium, *o*-Ps). *p*-Ps self-annihilates with an intrinsic lifetime of 125 ps, and *o*-Ps annihilates with electrons in surrounding atoms with a lifetime of several tens ns which is shorter than the intrinsic lifetime of 142 ns. Other positrons immediately thermalize and then directly annihilate with electrons in surrounding atoms which is known as the free positron annihilation lifetime. Thus, a typical positron annihilation lifetime spectrum (PALS) can be fitted by a summation of exponential decay functions corresponding to the various annihilation processes.

In the case of Ar gas, however, a characteristic shoulder structure that could not be reproduced by a summation of exponential functions appeared in a PALS [13]. Since the slope of the PALS spectrum corresponds to the annihilation rate, increase in the free annihilation rate was observed at the end of shoulder structure. Falk *et al.* applied an electric field to Ar gas, and found out that the increase in the annihilation rate disappeared and the shoulder structure faded [14], because the annihilation rate of free positrons accelerated by the electric field was higher than that of thermalized positrons. Thus, the shoulder structure in the PALS was caused by a change in the annihilation rate during the slowing down process. The free positron annihilation rate of thermalized positron becomes larger than that of epi-thermal positrons. The positron thermalization time can be observed as the end point of the shoulder structure where the annihilation rate becomes a constant value. The positron immediately loses its kinetic energy by inelastic scattering with the Ar atom down to 11.6 eV which is the first electronic excitation energy of the Ar atom, and then gradually loses its kinetic energy down to the thermalization energy only by elastic scattering whose momentum transfer is extremely low [15].

An AMOC (Age-MOmentum Correlation) measurement for positron annihilation in gas can be a powerful technique to investigate scatterings a low energy positron/Ps to atoms/molecules [16, 17]. In the AMOC technique, the Doppler broadening of annihilation γ rays is obtained as a function of positron age. The momentum is usually characterized by the *S* parameter which is defined by the number of counts in the central region of the annihilation γ-ray 511-keV peak normalized by the total number of counts in the peak. If the annihilation γ rays have a large Doppler broadening then the *S* parameter is decreased and vice versa. In the present paper, we performed AMOC measurements and investigated the positron slowing down process just below the first electronic excitation energy of Ar gas of 11.6 eV by determining the *S* parameter as a function of positron age.

2. Experimental

Positron lifetime spectra were recorded with a fast-fast coincidence system by determining the time interval between the detection of a 1.27-MeV γ ray from the β^+ decay of a ^{22}Na source and the detection of one of the 0.511-MeV annihilation γ ray by two plastic scintillation detectors. The size of the plastic scintillators for start and stop signals were ϕ4 cm × 5 cm (diameter × height) and ϕ4 cm × 3 cm, respectively. The energy of the other annihilation γ ray was measured with an HPGe (high purity-Ge) detector with a crystal size of ϕ5.4 cm × 5.2 cm.

As a positron source, ^{22}Na (0.5 MBq) was sandwiched between two sheets of 5-µm thick nickel foil (purity > 99 %). The source was placed in the center of a cylindrical high-pressure vessel with a diameter of 2.8 cm and a height of 5 cm, and the Ar gas (> 99.9999 %) was kept at 7.5 MPa at 293 K. The pressure was high enough so that no positrons annihilate in the walls of the vessel [18]. The start detector was placed as close as possible to the high-pressure vessel. The distance between the HPGe detector and the source was set to be 16 cm to reduce dead time. The stop detector was placed on the opposite side and the distance was determined so as to cover the same solid angle as the HPGe detector.

Pairs of signals of the time interval and energy were stored in a two dimensional multichannel analyzer. The time and energy resolution of the AMOC system used in this study were 300 ps at the full width at half maximum (FWHM) and 1.23 keV at FWHM, respectively. Standard γ-ray sources ^{60}Co, ^{85}Sr, and ^{133}Ba were used to determine an instrument response function of the HPGe detector. The HPGe detector was calibrated with ^{152}Eu and ^{22}Na sources, and the energy resolution was 0.34 keV per channel. To evaluate source corrections in PALS, the lifetime and intensity of the nickel foil were measured with thick kapton films whose lifetime was determined elsewhere. The Doppler broadening of the annihilation γ rays in the nickel foil was measured with thick nickel sheets. The count rate of the system was 0.8 cps and the total measuring time was 16 days.

3. Results and Discussion

Figure 1 shows the accumulated positron annihilation lifetime spectrum in Ar gas at a pressure of 7.5 MPa. One can see a characteristic shoulder structure after the prompt peak due to the slowing down of positrons. The end of the shoulder structure corresponds to a positron thermalization time of 5 ns, which is in good agreement with the previous report [19]. The lifetime and intensity of *o*-Ps were estimated to be 40 ns and 18 %, respectively, from the tail region of the spectrum.

Figure 2 shows the Doppler broadening of annihilation γ rays in Ar gas at t = 0.4 ns. The *S* parameter was defined as the ratio of the central area ($|\Delta E| < 0.5$ keV) to the whole annihilation peak area ($|\Delta E| < 5.0$ keV) and the *S* parameter was found to have a value of 0.344 at t = 0.4 ns.

Figure 3 shows the time dependence of the *S* parameter. Increase in the *S* parameter was observed up to 1 ns and then stayed on the same level around 0.358. We estimated the intensity of *p*-Ps from

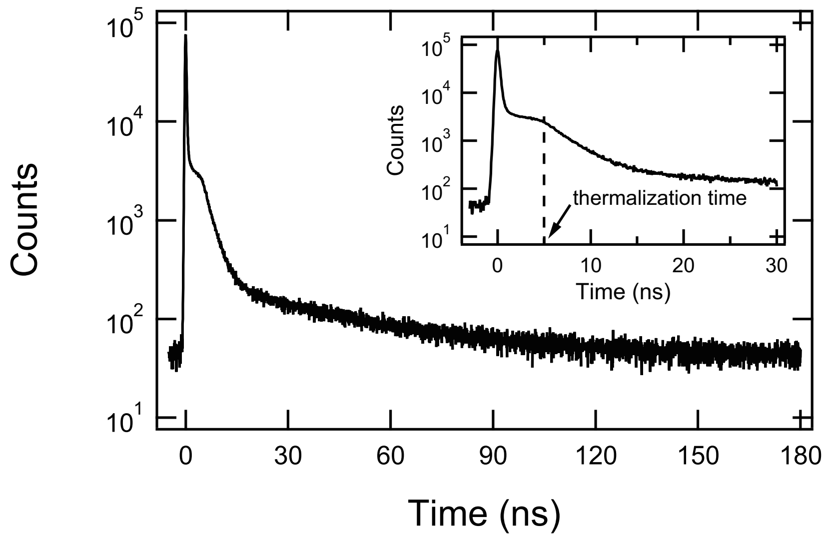

Fig. 1. Positron annihilation lifetime spectrum in Ar gas at 7.5 MPa. The inset shows the enlarged view near t = 0. The shoulder structure can be seen up to 5 ns.

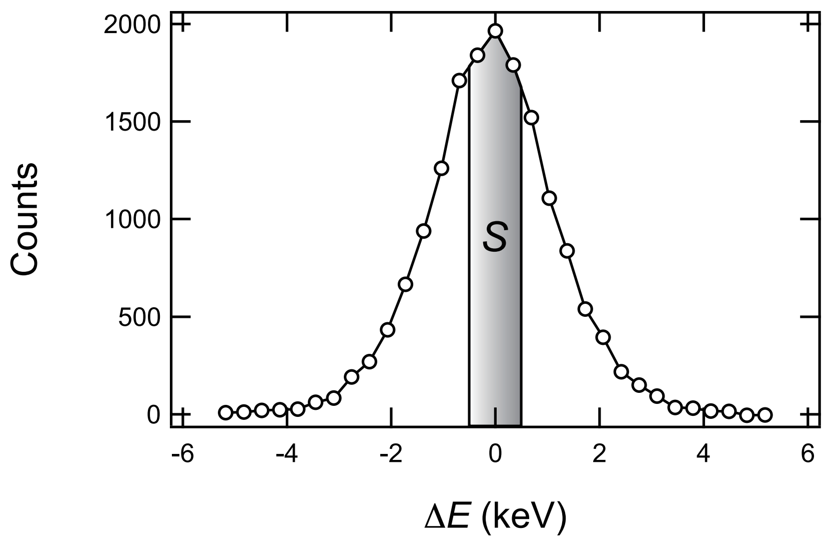

Fig. 2. Doppler broadening of annihilation γ ray in Ar gas for t = 0.4 ns.

that of *o*-Ps and concluded the contribution of *p*-Ps component in the energy spectrum at t = 0 was 30 %. Without the *p*-Ps component, the S parameter at t = 0 ns becomes 0.286 which is smaller than the current value. The increase of the S parameter indicated a reduction in the positron kinetic energy.

Previously, Iwata *et al.* measured the Doppler broadening of the γ rays from the annihilation of positrons with electrons in Ar atom with the positron energy much lower than the electron energy, and estimated that to be 2.30 keV at FWHM [20]. In this work, by deconvolving the measured spectra with the instrument response function, the FWHM of Doppler broadening at t = 5 ns was found to be 2.25 keV ± 0.04 keV. The similarity of this result to the previously measured value suggests that the positron energy at 5 ns should reach the thermalized energy.

When a positron enters the Ar gas, it immediately loses its kinetic energy by inelastic scattering

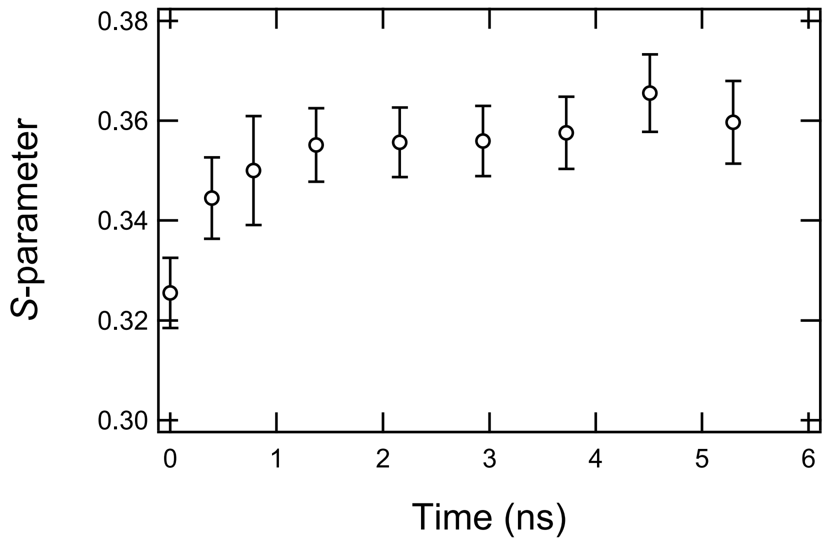

Fig. 3. Positron annihilation time dependence of the *S* parameter. The *S* parameter increased for the first 1 ns and then stayed on the same level after that. This trend suggests that the free positron kinetic energy decreased with time.

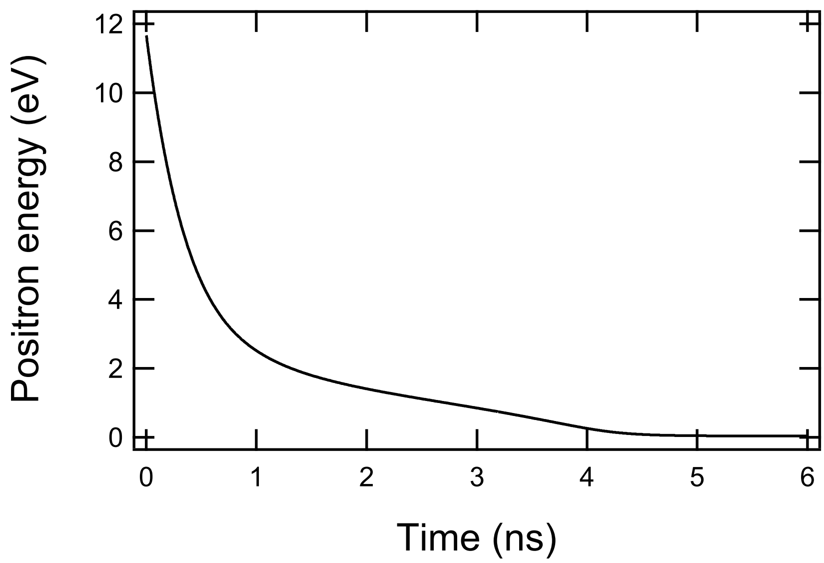

Fig. 4. Calculated (from Eq. 1) time dependence of positron kinetic energy in Ar gas at 7.5 MPa.

with Ar atoms down to 11.6 eV, and thereafter gradually loses its kinetic energy only by elastic scattering whose momentum transfer is extremely low [15]. The energy loss of positron by elastic scattering is written by

$$\frac{dE_{e^+}(t)}{dt} = -\frac{2n_{Ar}\sigma_m M_{Ar}}{(M_{Ar} + m_{e^+})^2}\sqrt{2m_{e^+}E_{e^+}(t)}\left(E_{e^+}(t) - \frac{3}{2}k_B T\right), \tag{1}$$

where $E_{e^+}(t)$ is the time dependence of positron kinetic energy, σ_m the positron-Ar momentum transfer cross-section, M_{Ar} the Ar mass, m_{e^+} the positron mass, n_{Ar} the number density of Ar, k_B the

Boltzmann constant, and T the temperature [21, 22]. Using the energy dependent $\sigma_{\rm m}$ calculated by McEachran [23] and an initial positron kinetic energy $E_{\rm e^+}(0)$ of 11.6 eV, the time dependence of positron kinetic energy below the inelastic threshold is calculated from Eq. (1) and shown in Fig. 4.

The calculation shows that the positron kinetic energy decreases from 11.6 eV to 2.5 eV within 1.0 ns, and then gradually decrease down to the thermalization energy of 0.039 eV near 5 ns. The thermalization time obtained from this calculation is consistent with that estimated (5 ns) from the lifetime spectrum (Fig. 1).

4. Summary

We performed an AMOC measurement to investigate the positron slowing down process in 7.5 MPa Ar gas at room temperature. A comparison of the measurement of the annihilation γ ray spectrum at 5 ns with a previous experiment suggested that the positrons had thermalized at this time. This result was consistent with a calculation of the elastic energy loss process below the inelastic threshold. The calculation also suggested that the observed increase in the S parameter up to 1 ns corresponded to the slowing down of positrons from 11.6 eV to around 2.5 eV. The positron slowing down process just below the first electronic excitation energy of Ar gas was observed via the time dependence of the S parameter.

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