Positronium spin conversion during collisions with Xe and its application for measuring the kinetic energy

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Abstract. Positronium (Ps) can undergo ortho–para spin conversion reaction during Ps–Xe collisions due to spin–orbit interaction. We have investigated energy dependence of this reaction rate and found it is nearly proportional to $T^{2.1}$, where $T$ is the temperature (300–623 K), while the pick-off annihilation rate is proportional to $T$. The strong temperature dependence of the former annihilation rate is attributed to a nature of $p$-wave scattering because this reaction is forbidden in $s$-wave scattering. In addition, a new method for measuring Ps kinetic energy has been developed with a high resolution and a high sensitivity by utilizing the strong temperature dependence as a “thermometer.” Analyzing the obtained time evolution of Ps kinetic energy, we have determined the momentum transfer cross section during Ps–Xe collisions at a very low energy (40–60 meV) to be 12(2)×10^{-16} cm^{2}.

1. Introduction
In 2006, a new reaction of positronium (Ps), i.e., Ps spin conversion reaction, was found [1]. Ps has four ground states denoted as $|S,M\rangle$, where $S$ is the total spin and $M$ is the z-projection. The triplet states are $|1,1\rangle$, $|1,0\rangle$ and $|1,-1\rangle$, while the singlet state is $|0,0\rangle$. These four states can be exchanged during Ps scattering with a high-Z atom such as Xe due to spin–orbit interaction. When this reaction occurs, an increase in the two-photon annihilation rate is observed because of a large difference in the lifetime between the triplet states and the singlet state.

Even when a magnetic field is applied, two of the three triplet states, $|1,\pm 1\rangle$, remain the same. On the other hand, the last triplet state, $|1,0\rangle$, and the singlet state, $|0,0\rangle$, are mixed each other to make two new ground states, which are denoted as $|+\rangle$ and $|-\rangle$. Under the magnetic field of 0.82 T, the mixture of $|0,0\rangle$ and $|1,0\rangle$ states is 1%, i.e., $|+\rangle = 0.994|1,0\rangle + 0.111|0,0\rangle$ and $|-\rangle = -0.111|1,0\rangle + 0.994|0,0\rangle$ [2]. It should be noted that the Ps lifetime in $|+\rangle$ state is considerably shorter than that in the original triplet states because two-photon annihilation is possible due to the small admixture of the singlet state. Under 0.82 T, the lifetime of $|+\rangle$ state is 9.5 ns.

We used this magnetic effect to evaluate the spin conversion reaction rate discriminating from pick-off annihilation rate. If there is no spin transitions, the lifetime of $|1,\pm 1\rangle$ states remains the same when a magnetic field is applied. However, if there are some spin transitions, in other words, the ar-
rows in figure 1 exist, the lifetime of $|1,\pm 1\rangle$ states is reduced by the transitions to $|+\rangle$ state. We evaluated the reaction rates from the changes in the lifetime.

\[ \text{Figure 1. Ground states of Ps and the corresponding lifetimes in vacuum under a magnetic field of (a) 0.00 and (b) 0.82 T. The arrows between the states indicate possible spin transitions.} \]

2. Temperature dependence of spin conversion annihilation

2.1. Experimental

We have two unknown constants: the spin-conversion annihilation rate and the pick-off annihilation rate. To evaluate them separately, we measured the long lifetime components under two magnetic conditions: 0.00 and 0.82 T. The temperature was kept at 300, 423, 540, 588, and 623 K. In addition, the lifetime without Xe gas was also measured to evaluate effect of silica-aerogel (SAG; 0.1 g/cc) used as a Ps creation medium by which positron-source of $^{22}$Na (2 $\mu$Ci) was sandwiched. The SAG decelerated the positron, supplied an electron to the positron, annihilated the free positrons, and took place Ps–Xe interactions in the inter-grain spaces. The measurement system was a digital-oscilloscope-based positron lifetime spectrometer described in our previous reports [3].

2.2. Results and discussion

As shown in figure 2, for example, the measured lifetime spectra exhibited a prompt peak followed by a long lifetime tail. The peak was due to self-annihilation of the singlet Ps and direct annihilations of positrons at the source, the SAG, chamber walls, etc. The tail was due to the triplet Ps and the lifetime was 128.7(3) ns, which was shorter than that in vacuum, i.e., 142.1 ns, because of the pickoff annihilation with the SAG. Next, Xe gas was introduced at the pressure of 250 kPa and the lifetime was reduced to 85.8(3) ns. This reduction was because of both the spin-conversion annihilation and the pickoff annihilation due to the Xe gas. To distinguish them, the third measurement was done under a magnetic field of 0.82 T, and the lifetime was further reduced to 70.4(5) ns. This further reduction was solely because of the spin-conversion from $|1,\pm 1\rangle$ state to $|+\rangle$ state.

The followings are simultaneous rate equations for number of Ps in the four ground states, $|+\rangle$, $|\pm\rangle$, $|1,1\rangle$ and $|1,-1\rangle$:

\[
\frac{d}{dt} \begin{pmatrix}
N_1 \\
N_2 \\
N_3 \\
N_4
\end{pmatrix} = \begin{pmatrix}
-k\lambda - k(\xi + 2) - \lambda_{po} & k\xi & k\eta & k\zeta \\
k\xi & -k\lambda - k(\xi + 2) - \lambda_{po} & k\zeta & k\eta \\
k\eta & k\zeta & -2k - \lambda_{po} & 0 \\
k\zeta & k\eta & 0 & -2k - \lambda_{po}
\end{pmatrix} \begin{pmatrix}
N_1 \\
N_2 \\
N_3 \\
N_4
\end{pmatrix}, \tag{1}
\]

where the off-diagonal elements correspond to the spin conversion with the reaction rate of $k$ and magnetic field dependent constants of $\xi = 0.952$, $\eta = 0.780$ and $\zeta = 1.22$ at 0.82 T ($\xi = \eta = \zeta = 1.00$ at 0.00 T) [1,2], and the diagonal elements also have the terms of the self-annihilation with the rates of
\[ \lambda_+ = 1.06 \times 10^8 \text{ s}^{-1}, \quad \lambda_- = 7.94 \times 10^7 \text{ s}^{-1} \quad \text{and} \quad \lambda_c = 7.04 \times 10^8 \text{ s}^{-1} \] for \( |+\rangle, \quad |\rangle, \quad |1, \pm\rangle \) states, respectively, as well as the pickoff annihilation with the rate of \( \lambda_{po} \) for all the states.

\[ \lambda_{po} \] is the classical electron radius, \( c \) is the velocity of light, and \( n \) is the number density of the Xe gas atoms. Similarly we determined the annihilation rates at other four temperatures as shown in figure 3. \( \lambda_{po} \) increased nearly proportional to the temperature \( T \). On the other hand, \( \lambda_{sc} \) increased proportional to \( T^2 \). No other gases have exhibited such a strong temperature dependence [4]. This discrepancy can be understood by considering a nature of \( p \)-wave scattering whose cross section is proportional to \( T^2 \) at low energy because \( s \)-wave scattering is forbidden for the spin-conversion reaction [5].

Assuming that the spin-conversion occurs only in \( p \)-wave scattering and that thermalized Ps atoms follows a Maxwell-Boltzmann distribution, we successfully explained the temperature dependence of

**Figure 2.** Ps lifetime spectra at 623 K. The measurement conditions of the Xe gas pressure and the magnetic field strength are (a) 0 kPa and 0 T, (b) 250 kPa and 0 T, and (c) 250 kPa and 0.82 T.

**Figure 3.** Temperature dependence of the spin-conversion annihilation rate (circles) and the pick-off annihilation rate (triangles) during Ps–Xe collisions. The solid lines are fitting results against power functions of temperature \( (300 \leq T \leq 623) \), and the dashed line is calculation result assuming the \( p \)-wave scattering and the Maxwell-Boltzmann distribution.
$Z_{\text{eff}}^1$ as shown in figure 3 with a dashed line. The $p$-wave scattering phase shift ($\delta_p$) is described as $k^2 \cot \delta_p(k) = -1/a + r k^2$/[2], where $k$ is the wave number, $a = 6.8$ is the scattering length and $r = -4.1$ is the effective range [5]; then, the $p$-wave scattering cross section ($\sigma_p$) is described as $\sigma_p(k) = 12 \pi \sin^2 \delta_p/k^2$, and the spin conversion annihilation rate ($\lambda_{\text{sc}}$) is $\lambda_{\text{sc}} = nf \int \rho(v) \sigma_p(v) v dv / \int \rho(v) dv$, where $f = 5.3 \times 10^{-4}$ is the spin conversion annihilation rate per collision and $\rho(v)$ is a Maxwell–Boltzmann distribution at $T$. This model is in a good agreement with the experimental results and shown in figure 3 as $p$-wave model with the dashed line.

3. Application of the spin-conversion reaction for measuring Ps energy

Because of the strong temperature dependence of $Z_{\text{eff}}^1$, we can estimate Ps energy ($E$) by measuring the two-photon annihilation rate at the time of $t (\lambda(t))$. Thermalization of Ps takes a considerably long time because the mass is very small, and it is a major source of uncertainty in precise measurement of Ps hyperfine splitting as a test of quantum electrodynamics [6]. As theoretical calculation is very difficult even for a simple system such as Ps–He scattering, high accuracy data is required [7,8].

However, conventional methods, namely, angular correlation of positron annihilation radiation (ACAR) and Doppler broadening (DB) do not have enough resolution especially at a very low energy near thermalization [8-10]. DB measures energy deviation from 511 keV, and ACAR measures angular deviation from 180 degrees. Both deviations are proportional to $\sqrt{E}$. On the other hand, $Z_{\text{eff}}^1$ is proportional to $T^{2.1} \propto E^{2.1}$ as described in section 2. Then, we can expect the resolution about 80 times higher than those of the conventional methods. In addition, the two conventional methods are sensitive only to the two-photon annihilation events emitting back-to-back ca. 511 keV gamma-rays so that one needs a magnetic field for Zeeman-mixing under which only Ps atoms in $|+\rangle$ state provide effective information. On the other hand, our new method employs both two- and three-photon annihilation events of all the triplet Ps atoms as described later so that we can expect the sensitivity more than six times larger than those of the conventional methods.

The measurement system is similar to a digital age-momentum correlation positron annihilation spectrometer [11] but we use two kinds of digitizers; the triple coincidence events were recorded by a digital oscilloscope (8 bit and 5 GHz sampling) for timing information from two BaF$_2$ scintillation detectors and by a digitizer (14 bit and 100 MHz sampling) for energy information from a high-purity Ge detector. All waveforms were analyzed after the measurement by a PC.

3.1. Analysis and discussion

Figure 4 (a) is the energy spectrum having a full-energy peak at 511 keV and the Compton edge at about 340 keV. Using this energy information, we make two kinds of lifetime spectrum; One is two-gamma lifetime spectrum ($I_2(t)$) belonging to the full-energy peak (509–515 keV), and the other is three-gamma lifetime spectrum ($I_3(t)$) belonging to the valley between the full-energy peak and the Compton edge (410–473 keV) where two-gamma events are considered less detected as shown in figure 5 (b, c), respectively.

Next, $I_2(t)$ is divided by $I_3(t)$ to obtain a decreasing function that is proportional to $\lambda_2(t)$ as

$$\frac{I_2(t)}{I_3(t)} = \frac{\epsilon_2 N(t) \lambda_2(t)}{\epsilon_3 N(t) \lambda_3} = \frac{\epsilon_2}{\epsilon_3} \lambda_2(t),$$

(2)

where $\epsilon_2$ ($\epsilon_3$) is two- (three-) gamma event detection efficiency, $\lambda_3 = 7.04 \times 10^6$ s$^{-1}$ is three-gamma annihilation rate constant, and $N(t)$ is the total number of the triplet Ps. Because lifetimes of the tails
of $I_3(t)$ and $I_4(t)$ are the same value of 101.4 ns. \[\left[I_2(t)/I_3(t)\right] \lambda_3 \] converges at $1.15 \times 10^7$ s$^{-1}$ and \[\lambda_4(t)\] converges at $2.85 \times 10^6$ s$^{-1}$. Therefore, $c_2/c_3 = 4.04$ because equation (2) should be kept at any time. As all the constants are fixed, we obtain the absolute value of the time evolution of the two-gamma annihilation rate as $\lambda_2(t) = 1.74 \times 10^6 \times \left[I_2(t)/I_3(t)\right]$, which can be converted into time evolution of Ps average kinetic energy as shown in figure 5 (the outer y-axis), because the relationship between $\lambda_2(t)$ and $T$ has been obtained as described in section 2.

Figure 4. (a) Energy spectrum measured using a high-purity Ge detector, (b) three-gamma lifetime spectrum, and (c) two-gamma lifetime spectrum.

Figure 5. Time evolutions of the two-gamma annihilation rate (the inner y-axis) and the kinetic energy (the outer y-axis). The solid lines are the fitting result and the thermal energy.
The time evolution of Ps kinetic energy \((E(t))\) is described as a hyperbolic faction assuming classical elastic collisions: 
\[
E(t) = E_{th} \coth^2 \left( \alpha + \beta t \right),
\]
where \(E_{th}\) is the thermal energy. \(\alpha\) is a parameter corresponds to the vertical range and \(\beta\) is one corresponds to the decay time [10]. \(\beta\) can be divided into two components corresponding to the Xe gas and the SAG as follows:

\[
\beta(n) = \beta_{Xe}(n) + \beta_{SAG} = \sigma_m n v_{thXe} \frac{m_{Ps}}{M_{Xe}} + \frac{v_{thPs}}{L} \frac{m_{Ps}}{M_{SAG}},
\]

where \(\sigma_m\) is the momentum transfer cross section during Ps-Xe collision, \(v_{th}(T)\) is Ps velocity at thermal energy \((8.26 \times 10^4 \text{ m/s at 300 K})\), \(m_{Ps} = 1.82 \times 10^{-30} \text{ kg}\) is the mass of a Ps atom, \(M_{Xe} = 2.18 \times 10^{-25} \text{ kg}\) is the mass of a Xe atom, \(L = 7.0 \times 10^{-8} \text{ m}\) is the mean distance between the SAG grains [10], and \(M_{SAG}\) is the effective mass of SAG atoms at the collisions. \(\sigma_m\) and \(M_{SAG}\) are unknown constants to be determined.

Least squares fittings for \(E(t)\) at two gas densities yield \(\beta^{-1} = 89(2) \text{ ns}\) for \(n = 6.06 \times 10^{25} \text{ m}^{-3}\) (250 kPa at 300K) and \(\beta^{-1} = 121(6) \text{ ns}\) for \(n = 2.42 \times 10^{25} \text{ m}^{-3}\) (100 kPa at 300K). Therefore, we obtain \(\sigma_m = 12(2) \times 10^{-16} \text{ cm}^2\) and \(M_{SAG} = 110(20) \text{ u}\) in the energy range between 40–60 meV. This \(\sigma_m\) is not far from the cross section of \(\sigma_w = \pi r_w^2 = 14.7 \times 10^{-16} \text{ cm}^2\), where \(r_w\) is van der Waals radius.

4. Conclusion

The spin conversion annihilation rate during Ps-Xe collisions exhibits a strong temperature dependence, which can be well explained by a nature of \(p\)-wave scattering because the reaction is forbidden in \(s\)-wave scattering. By utilizing this temperature dependence as a method for measuring Ps kinetic energy, we successfully obtained its time evolution and the momentum cross section for Ps-Xe collisions at a very low energy about 40-60 meV with a high sensitivity and a high resolution. The spin conversion reaction would open a new way to study low-energy atomic/molecular collisions [12,13].

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