Spin conversion induced by spin-orbit interaction in positronium collisions

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Abstract. The positronium spin conversion reaction induced by spin-orbit interaction is investigated. We obtain the reaction rates during positronium-Xe and positronium-Kr collisions by using the Zeeman mixing of positronium states. At thermal energies corresponding to room temperature, the reaction rate for spin conversion due to spin-orbit interaction is found to be almost twice that for the positronium pick-off reaction. We also study the energy dependence of the reaction rate. The mean energy of positronium is controlled by changing the gas temperature and using positronium in thermal equilibrium. We found that the reaction rate increases with the collision energy.

1. Introduction
Positronium (Ps) atoms are known to react with surrounding atoms and molecules [1–3]. Recently, Mitroy and Novikov [4] predicted a new reaction mechanism induced by spin-orbit interaction, which we derived experimentally [5]. In this reaction, ortho-Ps ($s=1$) colliding with a heavy singlet atom is converted into para-Ps ($s=0$) by spin-orbit interaction.

This reaction mechanism has been suggested as the answer to an unresolved question about Ps formation [4, 6-9]. It has long been known that the Ps formation fraction in heavy rare gas, which is estimated from the intensity of long-lived o-Ps component, is very low. The reason, however, has not been elucidated. In figure 1, the Ps formation fraction is shown by closed circles. The experimental value for He is 0.23; the values increase up to 0.33 from He to Ar with increasing atomic number. The experimental values for Kr and Xe are, however, 0.11 and 0.03, respectively, which are anomalously lower than those for He, Ne, and Ar. The expectation values from a simple theory considering energy conservation, usually called the Ore model, are also shown by bars in the figure. One can see that the theoretical values for Kr and Xe contradict the experimental results.

Mitroy and Novikov [4] performed a simulation assuming that ortho-Ps is quenched by this new reaction. They also assumed that the quenching rate depends on the Ps energy and that it is much higher in its early stage. The results of the simulation reproduced the anomalously low Ps formation fraction. For quantitative verification of their simulation, experimental study is necessary. Moreover, experimental evidence for the Ps energy dependence is lacking. In this paper, we describe an experimental study for obtaining the reaction rate for the Ps spin conversion reaction induced by spin-orbit interaction. We also report preliminary results on a study of its energy dependence.
Figure 1. Ps formation fraction in rare gas. The closed circles are experimental values estimated from the intensity of long-lived o-Ps component. The lines represent the upper and lower limit values obtained from the Ore model.

2. Principle of the experiment
We used the Zeeman mixing of Ps states to determine the reaction rate for the Ps spin conversion reaction due to spin-orbit interaction. Ps outside a magnetic field has four spin states: \( |0,0>\), \( |1,0>\), \( |1,1>\), and \( |1,−1>\), where the state \( |s,m>\) has spin \( s\) whose projection on the Z axis is \( m\). Transitions among these states are induced by the spin-orbit interaction, as shown in figure 2(a). The probabilities for transitions A, A’, A”, B, and B’ are equal to each other. Transitions B and B’ do not affect the lifetime of these states, because the mean lifetimes of \( |1,0>\), \( |1,1>\), and \( |1,−1>\) are identical.

![Diagram](image-url)

Figure 2. Transitions among four positronium eigenstates. (a) Without magnetic field. (b) Under a magnetic field of 1.0 T. Numbers of nanoseconds and picoseconds represent the lifetimes of these states.

When we apply a magnetic field of 1.0 T to this system, the states and transitions change as shown in figure 2(b). The \( |1,0>\) state turns into the \( |+>\) state, and \( |0,0>\) turns into \( |->\), where
\(| + \rangle = \frac{1}{\sqrt{1 + y^2}} |1,0\rangle - \frac{y}{\sqrt{1 + y^2}} |0,0\rangle \quad \text{and} \quad | - \rangle = \frac{y}{\sqrt{1 + y^2}} |1,0\rangle + \frac{1}{\sqrt{1 + y^2}} |0,0\rangle ,

\[
y = x/(\sqrt{1 + x^2} + 1) \quad \text{with} \quad x = 4\mu_e B/\hbar \omega_0 ; \quad \mu_e \text{ is the magnetic moment of the electron and } \hbar \omega_0 \text{ is the hyperfine splitting of Ps. The major difference between (a) and (b) is that in (a) the lifetime of } |+\rangle \text{ is only 6.6 ns, whereas in (b) that of } |1,0\rangle \text{ is 142 ns. Therefore, transitions D and D' reduce the mean lifetime of } |1,1\rangle \text{ and } |1,-1\rangle . \text{ This effect is used to determine the Ps spin-conversion reaction rate due to spin-orbit interaction. The transition probabilities for C, C', C'', D, and D' are not identical to those without a magnetic field. This effect is taken into account in the analysis.}

3. Experimental procedure

The sample chamber contains a \(^{22}\text{Na}\) positron source of 50 kBq, silica aerogel (\(\rho=0.1 \text{ g/cm}^3\)) as a Ps converter, and the sample gas. The formation probability of Ps is about 45%. Non-Ps positrons annihilate quickly with the material’s electrons. A magnetic field was applied using a conventional electromagnet. The flux density was set to 1.0 T and 0 T. The \(\gamma\) rays are detected by two BaF\(_2\) scintillators coupled to photomultiplier tubes. The 1.275 MeV nuclear \(\gamma\) rays from \(^{22}\text{Na}\), emitted immediately following positron emission, were detected to determine the time of positron injection into the sample, and the \(\gamma\) rays from the annihilation of positrons were detected to determine the time of positron annihilation. The signals of the photomultiplier tubes were processed by a digital positron lifetime spectrometer [10,11].

4. Results

Figure 3 shows positronium lifetime spectra in Xe for B=0 T and B=1.0 T. Long-lived components due to ortho-Ps are obvious. The long-lived component for B=0 T results from \(|1,0\rangle, |1,1\rangle, \text{ and } |1,-1\rangle, \text{ while that for } B=1 \text{ T results from } |1,1\rangle, \text{ and } |1,-1\rangle \text{ because } |+\rangle \text{ does not contribute to this component. Figure 3(a) clearly shows that the slope for Xe at 1.0 T is larger than that for Xe at 0 T. To demonstrate the change in the spectrum more clearly, the ratio } I(1\text{Tesla})/I(0\text{Tesla}) \text{ is also plotted in figure 3(b), where } I(B) \text{ is the spectrum for } B \text{ Tesla. The ratio in Xe obviously decreases with time, while that for vacuum is almost constant.}
Figure 3. (a) Positronium lifetime spectra in silica aerogel. Spectra labeled as vacuum are obtained without Xe and are shown by a continuous line (B=0 T) and dots (B=1 T). The spectra obtained in Xe gas (1 atm) are shown by crosses (B=0 T) and triangles (B=1 T). (b) The ratio of the spectra obtained without Xe (line) and in Xe gas (dots).

Table 1. Lifetime values of Ps

| Pressure/atm | \( \tau_{\text{long}} (0T)/\text{ns} \) | \( \tau_{\text{long}} (1T)/\text{ns} \) |
|--------------|----------------|----------------|
| vacuum       | 132.9 ± 0.5    | 131.4 ± 0.6    |
| 0.5          | 123.6 ± 0.6    | 119.1 ± 0.9    |
| Xe           | 118.4 ± 0.5    | 109.0 ± 0.6    |
| 1.0          | 112.1 ± 0.5    | 102.1 ± 0.6    |
| 1.5          | 119.5 ± 0.3    | 115.7 ± 0.4    |
| Kr           | 123.9 ± 0.7    | 124.6 ± 1.0    |
| Ar           |                |                |
The lifetime values of Ps are obtained using nonlinear least-squares fitting for the data between 200 ns and 400 ns after Ps formation, where the thermalization is expected to be almost complete and Ps atoms have a mean energy of $\frac{3}{2}k_BT$. The values are summarized in Table 1. Figure 4 shows the decay rate of long-lived Ps against the Xe pressure. As expected, a linear relationship exists between the pressure and decay rate.

The total decay rate ($\lambda_{\text{total}}$) without a magnetic field is the sum of the pick-off annihilation rate ($\lambda_{\text{po}}$) and the Ps spin-conversion rate due to spin-orbit interaction ($\lambda_{\text{so}}$). One can obtain $\lambda_{\text{po}}$ and $\lambda_{\text{so}}$ based on the discussion in section 2. Because transitions occur as shown in figure 2, the populations of the four positronium states obey the following differential equations:

$$\frac{dN_1}{dt} = [-\lambda_0 - \lambda_{\text{so}}(\xi + 2) - \lambda_{\text{po}}]N_1 + \lambda_{\text{so}}\xi N_2 + \lambda_{\text{so}}\eta N_3 + \lambda_{\text{so}}\zeta N_4,$$

$$\frac{dN_2}{dt} = [-\lambda_0 - \lambda_{\text{so}}(\xi + 2) - \lambda_{\text{po}}]N_2 + \lambda_{\text{so}}\xi N_1 + \lambda_{\text{so}}\zeta N_3 + \lambda_{\text{so}}\eta N_4,$$

$$\frac{dN_3}{dt} = [-\lambda_0 - 2\lambda_{\text{so}} - \lambda_{\text{po}}]N_3 + \lambda_{\text{so}}\eta N_1 + \lambda_{\text{so}}\xi N_2,$$

$$\frac{dN_4}{dt} = [-\lambda_0 - 2\lambda_{\text{so}} - \lambda_{\text{po}}]N_4 + \lambda_{\text{so}}\xi N_1 + \lambda_{\text{so}}\eta N_2,$$

where $N_i(t)$, ($i=1, 2, 3, 4$) denote the populations of the states $|+\rangle$, $|-\rangle$, $|1, 1\rangle$, and $|1, -1\rangle$, respectively, $\lambda_0$ is the self-annihilation rate of ortho-Ps [12,13], $\lambda_{\text{po}}$ is the pick-off annihilation rate, and $\lambda_{\text{so}}$ is the ortho-para conversion rate. The transition probability between the Ps states is $\lambda_{\text{so}}\xi$, $\lambda_{\text{so}}\eta$, and $\lambda_{\text{so}}\zeta$.
\[ \lambda_{SO} \eta \], or \[ \lambda_{SO} \zeta \], where \[ \xi = (1-y^2)/(1+y^2) \], \[ \eta = (1-y)^2/(1+y^2) \], and \[ \zeta = (1+y)^2/(1+y^2) \] [5]. Using these equations, \[ \lambda_{PO} \] and \[ \lambda_{SO} \] that reproduce the lifetime values of Ps are obtained uniquely. The rates \[ \lambda_{PO} \] and \[ \lambda_{SO} \] are converted into the dimensionless Ps annihilation parameter \[ \frac{1}{Z_{\text{eff}}}(\text{pick-off}) = \frac{\lambda_{PO}}{4\pi r_0^2 cn} \] and \[ \frac{1}{Z_{\text{eff}}}(\text{spin-orbit}) = \frac{\lambda_{SO}}{4\pi r_0^2 cn} \], where \( r_0 \) is the classical electron radius, \( c \) the speed of light, and \( n \) the gas number density. \( \frac{1}{Z_{\text{eff}}} \) is obtained using \( \frac{1}{Z_{\text{eff}}} = \frac{1}{Z_{\text{eff}}}(\text{pick-off}) + \frac{1}{Z_{\text{eff}}}(\text{spin-orbit}) \). The results are summarized in Table 2. \( \frac{1}{Z_{\text{eff}}} \) values are in agreement with those from previous works. It is found that for Ps–Xe collisions at room temperature, about 60% of \( \frac{1}{Z_{\text{eff}}} \) is due to spin-orbit interaction.

### Table 2. Annihilation parameters of Ps at room temperature.

|       | \( \frac{1}{Z_{\text{eff}}} \) (total) | \( \frac{1}{Z_{\text{eff}}} \) (pick-off) | \( \frac{1}{Z_{\text{eff}}} \) (spin-orbit) |
|-------|------------------|------------------|------------------|
| Xe    | 1.25 ± 0.04      | 0.48 ± 0.12      | 0.77 ± 0.12      |
| Kr    | 0.46 ± 0.03      | 0.36 ± 0.05      | 0.10 ± 0.05      |
| Ar    | 0.30 ± 0.03      | 0.37 ± 0.07      | -0.07 ± 0.07     |

5. Discussion

Experimentally obtained \( \frac{1}{Z_{\text{eff}}} \) is plotted against atomic number in figure 5. It seems to increase more rapidly than the fourth power of \( Z \), as shown by the solid line in the figure.

The spin-orbit Hamiltonian for an electron, \( V_{so} \), can be written [14] as

\[
V_{so} = \alpha l \cdot s = \frac{\hbar^2}{2m^2 c^2 r} \frac{dU}{dr} l \cdot s,
\]

where \( r \) is the electron coordinate, \( U(r) \) is the field, and \( l \) and \( s \) are the orbit and the spin operator, respectively. The main contribution of \( V_{so} \) comes from the region \( r < r_{\text{Bohr}} = \frac{\hbar^2}{Ze r m c^2} \), where the field of the nucleus is not screened, and \( |U(r)| = \frac{Ze^2}{r} \). The value of the coefficient \( \alpha \) for this region is roughly estimated as

\[
\alpha = \frac{\hbar^2}{2mc^2 r} \frac{Ze^2}{r^2} = \frac{\hbar^2 U}{mc^2 r_{\text{Bohr}}^2} = Z^4 \left( \frac{e^2}{\hbar c} \right)^2 \frac{me^4}{r^2},
\]

which is proportional to \( Z^4 \). For outer-shell electrons, the probability that the electron exists in the region \( r < r_{\text{Bohr}} \) is proportional to \( 1/Z^2 \) [14]. Therefore, spin-orbit energy splitting is proportional to

\[
\alpha = \frac{Z^2}{Z^4} = Z^{-2}.
\]

In the present experiment, the \( \frac{1}{Z_{\text{eff}}} \) (spin-orbit) that we measure is the transition rate from ortho-Ps to para-Ps. In first-order perturbation theory, the transition rate has the form of the square of the matrix element \( < f | H | i > \) corresponding to the energy. Therefore, one can expect the transition rate to have \( Z^2 \) dependence. It is interesting that the experimental results are almost in agreement with this simple argument. At present, one cannot discuss the discrepancy between the data and the \( Z^4 \) rule. More precise measurement and theory are needed to allow further discussion.
6. Energy dependence study
The significant effect of spin-orbit interaction during Ps–Xe collisions strongly suggests that this interaction is the cause of the anomalously low Ps fraction in Xe gas. Measurements of the energy dependence of $1Z_{\text{eff}}$(spin-orbit) are necessary to confirm this suggestion; the experiments are now in progress. The mean kinetic energy of Ps atoms is controlled by changing the temperature of the gas and using Ps atoms in thermal equilibrium. The rest of the apparatus is almost the same as that for the room-temperature measurement. Details will be described elsewhere. Tentative results are shown in figure 6. $1Z_{\text{eff}}$(spin-orbit) is found to increase with energy, as expected by Mitroy and Novikov [4].

Figure 5. $1Z_{\text{eff}}$(spin-orbit) for Ar, Kr, and Xe are plotted against the atomic number. The continuous curve shows $Z^4$ dependence.
Figure 6. Annihilation parameter $\gamma_{\text{eff}}(\text{spin-orbit})$ for several temperatures. Mean kinetic energies of Ps $\left(\frac{3}{2}k_B T\right)$ are also indicated.

7. Conclusion
A new Ps reaction, the Ps spin conversion reaction induced by spin-orbit interaction, is investigated experimentally. The reaction rate is unexpectedly large. For Ps–Xe collisions at thermal energies corresponding to room temperature, about 60% of the total quenching rate is found to be a result of this reaction. The reaction rate increases approximately as $Z^4$, as expected on the basis of a simple argument. A study of the energy dependence of the reaction rate is now in progress. The rate is found to increase with collision energy, strongly suggesting that spin-orbit interaction is the cause of the anomalously low Ps formation fraction in Xe gas.

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