Experimental Evidence for Spin-Orbit Interactions in Positronium-Xe Collisions

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(Received 22 March 2006; published 19 December 2006)

Spin-orbit interaction of positronium (Ps) with the surrounding atoms, predicted by Mitroy and Novikov [Phys. Rev. Lett. **90**, 183202 (2003)], has been detected experimentally. We have found that the lifetime of the magnetically *unperturbed* ortho-Ps in Xe gas of 1 atm decreases significantly when a magnetic field of 1.0 T is applied. This decrease is attributed to the Ps spin conversion caused by spin-orbit interaction during Ps-Xe collision. The annihilation cross section of ortho-Ps due to this interaction has been determined to be 3 times as large as that expected by Mitroy and Novikov.

DOI: 10.1103/PhysRevLett.97.253402 PACS numbers: 36.10.Dr, 34.50.−s

An electron and a positron can bind together to form a light atom called positronium (Ps). The intrinsic natures and the interaction of Ps have been studied extensively [1–3]. One of the problems not yet solved is that the measured fractions of long-lived Ps in Kr and Xe gas are anomalously below the prediction of the Ore model [4–7]. To explain this, Wright *et al.* proposed the possibility of a Ps-Xe resonance state [7]. The mechanism for the resonance, however, has not been understood. Recently Mitroy and Novikov [8] have suggested that ortho-Ps to para-Ps conversion reaction induced by the spin-orbit interaction may be the cause of the low fraction of long-lived Ps. This new type of reaction can be understood as a result of singlet-triplet mixing during a collision induced by different energy shifts due to spin-orbit interaction for the electron and the positron in Ps, given that they have different signs of charge and are in different locations. This attractive suggestion has to be supported by experimental evidence before it is accepted. In the present Letter, we have succeeded in observing this effect experimentally and obtaining the reaction rate, by using magnetic field mixing of Ps to overcome the experimental difficulties.

Ps can exist in the para-Ps state ($|S = 0, m = 0\rangle$) or one of the ortho-Ps states ($|S = 1, m = -1, 0, 1\rangle$). The para-Ps in vacuum self-annihilates into 2γ with a lifetime of 125 ps while ortho-Ps, which decays into 3γ, has a much longer lifetime of 142 ns. When a static magnetic field is applied, the Zeeman mixtures of the para-Ps eigenstate $|0, 0\rangle$ and the $m = 0$ substate of ortho-Ps $|1, 0\rangle$ become new eigenstates. The four Ps eigenstates in a magnetic field B are $|+\rangle = (1/\sqrt{1+y^2})|1, 0\rangle - (y/\sqrt{1+y^2})|0, 0\rangle$, $|-\rangle = (y/\sqrt{1+y^2})|1, 0\rangle + (1/\sqrt{1+y^2})|0, 0\rangle$, $|1, 1\rangle$, and $|1, -1\rangle$, where $y = x/(\sqrt{1+x^2}+1)$ with $x = 4\mu_e B/\hbar\omega_0$; μ_e is the magnetic moment of the electron and $\hbar\omega_0$ is the hyperfine splitting of Ps [1,9,10]. Note that $|1, 1\rangle$ and $|1, -1\rangle$ are not affected by the magnetic field. When a magnetic field is not applied, $|+\rangle$ is equivalent to $|1, 0\rangle$, and thus the three ortho-Ps states, $|1, 1\rangle$, $|1, -1\rangle$, $|1, 0\rangle$ contribute to the long lifetime component in the positron lifetime spectrum. The self-annihilation rates of $|+\rangle$ and $|-\rangle$ are given as $\lambda_+ = \frac{1}{1+y^2}\lambda_o + \frac{y^2}{1+y^2}\lambda_p$, $\lambda_- = \frac{y^2}{1+y^2}\lambda_o + \frac{1}{1+y^2}\lambda_p$, respectively, where λ_o is the self-annihilation rate of ortho-Ps ($1/142$ ns $\simeq 7 \times 10^6$/s) and λ_p is that of para-Ps ($1/125$ ps $= 8 \times 10^9$/s). Because of the 3 orders of magnitude difference between λ_p and λ_o, λ_+ is a rapidly decreasing function of B [1,9,10]. For example, if $B = 1.0$ T, the lifetime of $|+\rangle$ is as short as 6.6 ns, while that of $|1, \pm 1\rangle$ remains at 142 ns. The state $|+\rangle$ under a B of 1.0 T thus no longer contributes to the long lifetime component in the lifetime spectrum; the long lifetime component is from the states $|1, \pm 1\rangle$ only.

In the present experiment, we have detected the spin-orbit interaction by measuring the change in the long lifetime of Ps in Xe and Kr when a static magnetic field of 1.0 T is applied. The principle of this experiment is as follows. If the spin transition among the four Ps eigenstates does not exist, the long lifetime τ_{long} of $|1, \pm 1\rangle$ is independent of the magnetic field. If the spin transition exists, the transitions from $|1, \pm 1\rangle$ to $|+\rangle$ will result in decreases in the τ_{long} of $|1, \pm 1\rangle$ on applying the magnetic field, since the lifetime of $|+\rangle$, 6.6 ns, is much smaller than that of $|1, \pm 1\rangle$.

The experimental apparatus is shown schematically in Fig. 1. A ^{22}Na positron source of 50 kBq was used. The 1.275 MeV nuclear γ rays, emitted immediately following the positron emission, were detected to determine the time of the positron injection into the sample, and the γ rays (0–511 keV) from the annihilation of positrons were detected to determine the time of the annihilation of the positron. Ps atoms were formed inside or on the silica grain and emitted into the free space in silica aerogel ($\rho = 0.1$ g/cm^3) supplied by Airglass [11]; the formation probability of Ps is about 45% [12]. Non-Ps positrons stay inside the grain after the thermalization due to its positive work function and they annihilate quickly with the material electrons. The magnetic field was applied using a conventional electromagnet. The flux density was set at ± 1.0 T and 0 T. The sample chamber was kept at 300 ± 1 K during the measurements. The γ rays were detected by two BaF_2 scintil-

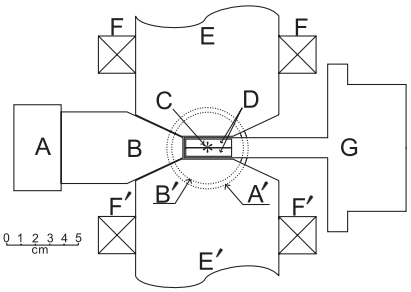

FIG. 1. Schematic diagram of the apparatus. A, A′: Photomultiplier tubes (Hamamatsu H6614Q-mod); B, B′: BaF_2 scintillators; C: ^{22}Na positron source sandwiched by KAPTON foils; D: silica aerogel (0.1 g/cm^3); E, E′: iron pole pieces; F, F′: magnetic coils (not to scale); G: sample chamber.

lators coupled to photomultiplier tubes. The signals of the photomultiplier tubes were processed by a digital positron lifetime spectrometer [13,14]. The sampling rate and the time range of the LeCroy WaveRunner6050 oscilloscope were set to be 1GS/s and 2 μs, respectively.

Figure 2 shows lifetime spectra of Ps in silica aerogel. The spectra labeled as "vacuum," obtained without Xe, are shown by a continuous line ($B = 0$ T) and dots ($B = 1.0$ T). They are normalized against the total count. The spectrum for $B = 1.0$ T is the sum of those for $+1.0$ T and -1.0 T, which were essentially the same. The long lifetime component for $B = 0$ T results from $|1, 1\rangle$, $|1, 0\rangle$ and $|1, -1\rangle$; that for $B = 1.0$ T is due to $|1, 1\rangle$ and $|1, -1\rangle$ because $|+\rangle$ does not contribute to this component. As expected, the data for $B = 0$ T and those for $B = 1.0$ T look parallel, and the intensity for the latter is reduced to 2/3. The lifetime values are determined from least-squares fit to the data between 200 and 400 ns after Ps formation; in

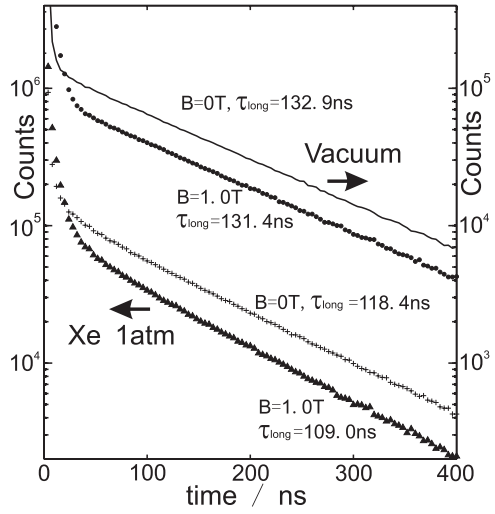

FIG. 2. Positron lifetime spectra in silica aerogel. The spectra labeled as vacuum are obtained without Xe and are shown by a continuous line ($B = 0$ T) and dots ($B = 1.0$ T). The spectra obtained after 1 atm of Xe gas was introduced are shown by crosses ($B = 0$ T) and triangles ($B = 1.0$ T). The random backgrounds have been subtracted.

this time range the thermalization is expected to be almost complete [15] and Ps atoms have a mean energy of $\sim\frac{3}{2}k_BT$ (0.039 eV). The lifetime values obtained, 132.9 and 131.4 ns, are approximately equal to each other. The difference between the intrinsic lifetime of ortho-Ps, 142.05 ns [16,17], and the present values is due to the interaction of Ps with the surfaces of silica grains in the aerogel. The spectra obtained with Xe gas introduced into the aerogel are shown by crosses ($B = 0$ T) and triangles ($B = 1.0$ T). The long lifetime decreases to 118.4 ns when Xe is introduced and further decreases to 109.0 ns when a magnetic field of 1.0 T is applied. The intensity also decreases to less than two thirds on applying the magnetic field. The changes are clearly visible in the figure. The lifetime values for similar measurements with Ar, Kr at 2.5 atm and those with Xe at 0.5 atm and 1.5 atm are also listed in Table I.

The significant change in the Ps lifetime in Xe gas induced by a magnetic field is exactly what we expected above; because the lifetime of $|+\rangle$ in a magnetic field of 1.0 T is 6.6 ns, much smaller than that of $|1, \pm 1\rangle$, the Ps spin transition from the states $|1, \pm 1\rangle$ to $|+\rangle$ has resulted in the reduction of the long lifetime of the former states. A possibility of the change in the target atom by the magnetic field is excluded because of its closed shell electronic structure [18].

Possible origins of the Ps spin transition are the following: (i) the magnetic dipole interaction with target electrons or nucleus, (ii) the paramagnetic electron-exchange reaction [19–22], and (iii) the spin-orbit interaction [8]. The magnetic dipole interaction (i) is very weak [9,19,20]. Its influence on Ps reaction have never been observed and there is no reason that it should be observed with Xe or Kr. The paramagnetic electron-exchange reaction (ii) is possible only when either the initial or final state of the target is nonsinglet. It is not possible in the Ps-Xe collision at thermal energies because the ground state of Xe is singlet and the lowest excitation energy is larger than 8 eV. Therefore the only conceivable mechanism which explains the observed change in the long lifetime is the spin-orbit interaction (iii).

The annihilation rate of Ps due to collisions with gas atoms and molecules is traditionally expressed in terms of a dimensionless parameter $_1Z_{\mathrm{eff}}$ [2,3,23], defined as $_1Z_{\mathrm{eff}} = \lambda_{\mathrm{gas}}/(4\pi r_0^2 cn)$, where λ_{gas} is the annihilation rate of Ps due to Ps-gas interaction in a gas of number density n, with r_0 the classical electron radius and c the speed of light. This parameter corresponds to the annihilation cross section averaged over the distribution of Ps energies at the time of the annihilation. When the Ps speed is v, the annihilation cross section σ is expressed as $\sigma = 4\pi r_0^2 c\ {}_1Z_{\mathrm{eff}}/v$. One can determine the values of the total annihilation parameter $_1Z_{\mathrm{eff}}(\mathrm{total})$ by using

$$\lambda = [(4\pi r_0^2 c)_1 Z_{\mathrm{eff}}(\mathrm{total})]n + \lambda_{\mathrm{vacuum}}, \qquad (1)$$

where λ is the decay rate $(1/\tau_{\mathrm{long}})$ in a gas of density n, and

TABLE I. The lifetime (τ_{long}) values of Ps and the annihilation parameters ${}_1Z_{\text{eff}}$(total), ${}_1Z_{\text{eff}}$(pick-off), and ${}_1Z_{\text{eff}}$(spin-orbit).

System	Pressure/atm	τ_{long}/ns (0 T)	τ_{long}/ns (1.0 T)	${}_1Z_{\text{eff}}$(total)	${}_1Z_{\text{eff}}$(pick-off)	${}_1Z_{\text{eff}}$(spin-orbit)
Vacuum		132.9 ± 0.5	131.4 ± 0.6			
	0.5	123.6 ± 0.6	119.1 ± 0.9			
Xe	1.0	118.4 ± 0.5	109.0 ± 0.6	1.25 ± 0.04	0.48 ± 0.12	0.77 ± 0.12
	1.5	112.1 ± 0.5	102.1 ± 0.6			
Kr	2.5	119.5 ± 0.3	115.7 ± 0.4	0.46 ± 0.03	0.36 ± 0.05	0.10 ± 0.05
Ar	2.5	123.9 ± 0.7	124.6 ± 1.0	0.30 ± 0.03	0.37 ± 0.07	-0.07 ± 0.07

λ_{vacuum} is that measured without the gas. The value of ${}_1Z_{\text{eff}}$(total) for Xe was obtained from a least-squares fit of Eq. (1) to the data for vacuum and the three different pressures. Those for Kr and Ar were obtained from the difference in λ between 2.5 atm and vacuum. Values of ${}_1Z_{\text{eff}}$(total) thus obtained for Xe, Kr, and Ar are 1.25 ± 0.04, 0.46 ± 0.03, and 0.30 ± 0.03, respectively.

It is possible to decompose the total annihilation parameter ${}_1Z_{\text{eff}}$(total) into ${}_1Z_{\text{eff}}$(pick-off) and ${}_1Z_{\text{eff}}$(spin-orbit), which are the contributions from the pick-off annihilation and the ortho-para conversion induced by the spin-orbit interaction, respectively. The fates of the four Ps states in a magnetic field are described by the coupled rate equations [24–26],

$$\begin{aligned}
\dot{N}_1 &= [-\lambda_+ - k(\xi+2) - \lambda_{\text{po}}]N_1 + k\xi N_2 + k\eta N_3 + k\zeta N_4, \\
\dot{N}_2 &= [-\lambda_- - k(\xi+2) - \lambda_{\text{po}}]N_2 + k\xi N_1 + k\zeta N_3 + k\eta N_4, \\
\dot{N}_3 &= [-\lambda_o - 2k - \lambda_{\text{po}}]N_3 + k\eta N_1 + k\zeta N_2, \\
\dot{N}_4 &= [-\lambda_o - 2k - \lambda_{\text{po}}]N_4 + k\zeta N_1 + k\eta N_2, \qquad (2)
\end{aligned}$$

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, λ_o the self-annihilation rate of ortho-Ps [16,17], λ_{po} the pick-off annihilation rate, and k the ortho-para conversion rate. The transition probability between the Ps states is $k\xi$, $k\eta$, or $k\zeta$, where $\xi = (1-y^2)^2/(1+y^2)^2$, $\eta = (1-y)^2/(1+y^2)$, and $\zeta = (1+y)^2/(1+y^2)$. This explicit form comes from Eqs. (3)–(5) in [26] with setting polarizations $P_{\text{positron}} = P_{\text{electron}} = 0$.

Equations (2) are applicable to the Ps spin transition due to the spin-orbit interaction because of the following reason. Since the spin-orbit Hamiltonian $V_{\text{so}} = (\alpha^2 \frac{1}{r_{\text{pos}}} \times \frac{dV_{\text{pos}}}{dr_{\text{pos}}}) l_{\text{pos}} s_{\text{pos}} + (\alpha^2 \frac{1}{r_{\text{elec}}} \frac{dV_{\text{elec}}}{dr_{\text{elec}}}) l_{\text{elec}} s_{\text{elec}}$ [8] is a sum of the positron term and the electron term, the Hamiltonian flips either the spin of the electron or that of the positron in Ps exclusively. Here the subscripts "pos" and "elec" refer to the positron and the electron, respectively. For the spin-flip of the electron the transition probabilities among the four Ps eigenstates are derived in the same way as those for the case of paramagnetic electron-exchange collision [20] and coefficients in transition probabilities (ξ, η, ζ) is the same as those for the electron paramagnetic exchange. For the spin-flip of the positron we also have the same coefficients because of the symmetry. Therefore, by putting $k = k_{\text{elec}} + k_{\text{pos}}$, we have transition probabilities $k\xi$, $k\eta$, and $k\zeta$ for the Ps spin transition due to the spin-orbit interaction. Thus Eqs. (2) do not need any further modification.

The parameters to be determined are $\lambda_{\text{po}}^{\text{gas}}$, k^{gas}, $\lambda_{\text{po}}^{\text{silica}}$, and k^{silica}, where $\lambda_{\text{po}}^{\text{gas}}$ and k^{gas} are the pick-off annihilation rate and the ortho-para conversion rate due to the sample gas, and $\lambda_{\text{po}}^{\text{silica}}$ and k^{silica} are those due to the silica aerogel. The relation between these parameters and τ_{long} is obtained by using the Eqs. (1) through a diagonalization of the 4×4 matrix; thus we have a function f:

$$\tau_{\text{long}} = f(\lambda_{\text{po}}^{\text{gas}}, k^{\text{gas}}, \lambda_{\text{po}}^{\text{silica}}, k^{\text{silica}}, B).$$

Using the four experimental values of τ_{long} for $B = 0$ T and 1.0 T and in the presence and absence of the sample gas, we can obtain simultaneous equations for $\lambda_{\text{po}}^{\text{gas}}$, k^{gas}, $\lambda_{\text{po}}^{\text{silica}}$, and k^{silica}, which can be solved using multidimensional Newton's method. The errors in these parameters are evaluated by the analysis that changes the value of τ_{long}.

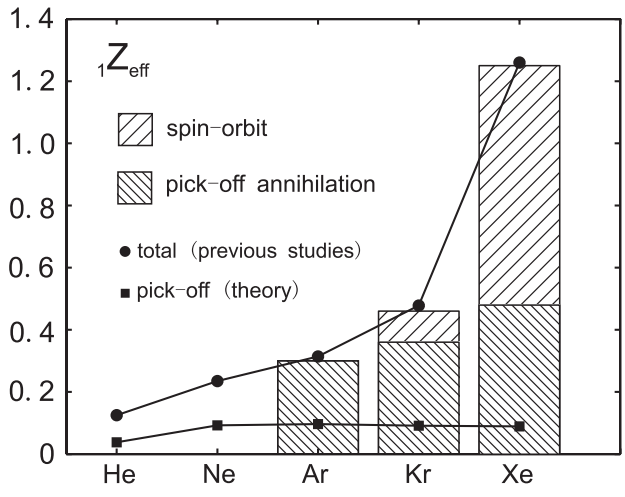

FIG. 3. The annihilation parameter ${}_1Z_{\text{eff}}$ for rare gases. The total heights of the columns represent ${}_1Z_{\text{eff}}$(total) for Xe, Kr, and Ar from the present work. Partial contributions of ${}_1Z_{\text{eff}}$(pick-off) and ${}_1Z_{\text{eff}}$(spin-orbit) for Xe and Kr are also shown by columns with close and wide hatches, respectively. Previous experimental values for ${}_1Z_{\text{eff}}$(total) [2,4,7] and results of theoretical calculations for ${}_1Z_{\text{eff}}$(pick-orbit) [27,28] are shown by solid circles and squares, respectively.

The conversions from these parameters to the partial contributions are made by using the definitions ${}_1Z_{\mathrm{eff}}(\text{pick-off}) = \lambda_{\mathrm{po}}^{\mathrm{gas}}/4\pi r_0^2 cn$ and ${}_1Z_{\mathrm{eff}}(\text{spin-orbit}) = k^{\mathrm{gas}}/4\pi r_0^2 cn$. For Xe, we obtain ${}_1Z_{\mathrm{eff}}(\text{pick-off}) = 0.48 \pm 0.12$ and ${}_1Z_{\mathrm{eff}}(\text{spin-orbit}) = 0.77 \pm 0.12$. For Kr, ${}_1Z_{\mathrm{eff}}(\text{pick-off}) = 0.36 \pm 0.05$ and ${}_1Z_{\mathrm{eff}}(\text{spin-orbit}) = 0.10 \pm 0.05$. For Ar, the contribution from the spin-orbit interaction is negligible within the statistical uncertainty.

In Fig. 3, values of ${}_1Z_{\mathrm{eff}}(\text{total})$ obtained are represented by the total heights of the columns. ${}_1Z_{\mathrm{eff}}(\text{pick-off})$ and ${}_1Z_{\mathrm{eff}}(\text{spin-orbit})$ are shown by the columns with close and wide hatches, respectively. They are also summarized in Table I. ${}_1Z_{\mathrm{eff}}(\text{total})$ corresponds to the quantity simply expressed as ${}_1Z_{\mathrm{eff}}$ in previous works [2,4,7]. These previous values of ${}_1Z_{\mathrm{eff}}$ are plotted with circles in the figure. The agreement between these parameters indicates that their and our experiments are consistent. However, in the previous works, all of ${}_1Z_{\mathrm{eff}}$ was interpreted so as due to the pick-off annihilation.

The present work has shown that 60% of ${}_1Z_{\mathrm{eff}}(\text{total})$ for Ps-Xe collision is due to the spin-orbit interaction. The rapid increase in ${}_1Z_{\mathrm{eff}}$ as the atomic number increases from Ar to Xe was previously attributed to some special effect on pick-off annihilation [2,4,7]. The present work has proved that this is not the case, and that the increase is due to spin-orbit interaction. Mitroy and Novikov [8] proposed a scaling 0.0205:0.265:1 in ${}_1Z_{\mathrm{eff}}(\text{spin-orbit})$ for Ar, Kr, and Xe (roughly proportional to Z^4). This scaling is in agreement with the present results. As for the absolute value, they expected that 20% of ${}_1Z_{\mathrm{eff}}(\text{total})$ for Xe is due to the spin-orbit interaction (not shown in the figure). This is one third of the present result.

The results of the theoretical calculation for ${}_1Z_{\mathrm{eff}}(\text{pick-off})$ [27,28] are denoted by squares in Fig. 3. They are still considerably smaller than the experimental values of pure ${}_1Z_{\mathrm{eff}}(\text{pick-off})$ obtained after decomposing ${}_1Z_{\mathrm{eff}}(\text{total})$. This may be due to short-range electron-positron correlations which are ignored in the calculation.

As for the resolution of the anomalously low fraction of the observed long-lived Ps in Ps-Xe system [2–7], the study of the energy dependence of ${}_1Z_{\mathrm{eff}}(\text{spin-orbit})$ is crucial [8]. Because ${}_1Z_{\mathrm{eff}}(\text{spin-orbit})$ is expected to increase with Ps energy [8], the interaction efficiently quenches fast Ps just after its birth and results in the low fraction of long-lived Ps. In order to obtain experimental evidence, an age-momentum-correlation experiment with HPGe detector [29] and one with angular correlation of annihilation radiation [30] are now in progress.

The authors would like to thank Y. Nagashima for helpful discussions. One of the authors (H. S.) gratefully acknowledges K. Asai, M. Koshimizu, and K. Shibuya for discussions. This research was partially supported by the Ministry of Education, Science, Sports and Culture, Grant-in-Aid for Scientific Research (A), No. 16204029 (2004–2006), grants from Japan Science and Technology Agency, and The Industrial Technology Research Grant program from New Energy and Industrial Technology Development Organization (NEDO) of Japan.

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