

Enhancement of direct positron annihilation due to temporal capture by an atom

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We investigate direct positron annihilation during a collision in a gas; this is physically separable from indirect annihilation, i.e., via formation of positronium (Ps) and its decay. The cross section of direct annihilation is calculated from the evolution of a wave packet with a time width much shorter than the Ps lifetime, where the direct contribution is extracted through the time dependence of the survival probability. The cross section obtained for a hydrogen atom is found to be enhanced in energies around 10 eV well above the Ps formation threshold. This enhancement is shown to be due to temporal positron capture with electronic excitation in the atom.

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I. INTRODUCTION

A positron, which is the antiparticle of an electron, annihilates, emitting two or three gamma photons, when it collides with an electron. The interaction of a positron with matter causes interesting phenomena associated with its annihilation, such as lifetime spectra characteristic of the target matter [1], large annihilation cross sections sensitive to atomic and molecular structures [2], and dissociative ionization of molecules [3]. Understanding of positron annihilation is of great importance for scientific and technological applications to studies of biomolecular structure [4], medical imaging [5], and characterization of materials [6].

The mechanism of positron annihilation is classified into *direct* and *indirect* processes. Direct annihilation occurs during a collision with an atom or a molecule (X), $e^+ + X \rightarrow X^+ + 2\gamma$. Indirect annihilation occurs via formation of para-positronium (Ps), $e^+ + X \rightarrow X^+ + \text{Ps}$, followed by its decay, $\text{Ps} \rightarrow 2\gamma$, as it travels away from the residual ion (X^+). In the direct process, the positron annihilates promptly in a collision time $\tau_{\text{col}} \sim 10^{-17}$ sec, while in the indirect process, it annihilates slowly with the lifetime $\tau_{\text{Ps}} \geq 10^{-10}$ sec of para-Ps. Thus, positron annihilation is subject to two processes with different time scales. Although leading from a common initial state to a common final state, the two processes are physically distinguishable with virtually no interference between them because of the time scales.

The net positron annihilation in a gas should be described in different ways according to the target density. For a target of rarefied gas such that the mean free time τ_{MFT} of a positron, or of a Ps, is much longer than τ_{Ps} , all positron annihilation is described as a single elementary process. The annihilation rate is thus given by a single cross section [7–9],

where the direct and indirect processes are inseparable. However, this is not the case with a dense gas. In fact, experiments are usually performed with a target pressure comparable to, or larger than, 1 atm (equivalent to a number density of $N = 2.7 \times 10^{25} \text{ m}^{-3}$), so that the mean free time is as short as $\tau_{\text{MFT}} \leq 10^{-12}$ sec, satisfying $\tau_{\text{col}} \ll \tau_{\text{MFT}} \ll \tau_{\text{Ps}}$. Hence, although the target density is still low enough to ensure binary collisions, a para-Ps produced will meet secondary collisions before it decays; the Ps may be scattered, excited, and even broken up. Under such conditions, the Ps should be treated as an independent species in matter [10]; the number of net annihilation events in a gas will thus be given by solving a set of coupled rate equations including many different elementary processes, where the effects of the direct and indirect annihilation are separately incorporated. The direct annihilation is still described as a single elementary process, but the indirect annihilation as a sequence of two or more elementary processes via formation of a Ps. Consequently, the annihilation probability of a positron incident on a gas with a velocity v behaves for an infinitesimal time t as $v\sigma_{2\gamma}Nt + (1/2)v\sigma_{\text{Ps}}Nt^2/\tau_{\text{Ps}}$, where $\sigma_{2\gamma}$ and σ_{Ps} denote the cross sections of the direct annihilation and the Ps formation, respectively. This means that the annihilation probability is given on a time scale of $\tau_{\text{col}} \ll t \ll \tau_{\text{MFT}}$ by the sum of contributions from the direct and indirect processes, which both depend on the gas density as proportional to N but differently on time as proportional to t and to t^2 , respectively. This clearly shows that the number of net annihilation events is *not* expressed by the sum of the cross sections $\sigma_{2\gamma} + \sigma_{\text{Ps}}$.

Thus, direct positron annihilation is not part of the total annihilation but an independent elementary process in its own right, though never addressed so far, to our knowledge. Its cross section $\sigma_{2\gamma}$ should be derived from the evolution of wave packets with a time width τ much shorter than the Ps lifetime, because a particle in a dense gas behaves as a wave packet with $\tau \sim \tau_{\text{MFT}}$ [10]. Note that automatic application of the ordinary steady-state scattering theory as in Ref. [9]

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would be misleading, because it is justified only when $\tau_{\text{MFT}} \gg \tau_{\text{Ps}}$ [10].

In the present paper, we elucidate the mechanism of direct positron annihilation in collisions with a hydrogen atom. The effect of the direct process is extracted from the evolution of wave packets with a time-dependent coupled-channel (TDCC) method [11]. It reveals remarkable enhancement of the cross section in energies around 10 eV. By analyzing the time delay of the wave packets, this enhancement is shown to be caused by temporal positron capture with electronic excitation in the atom.

II. ANNIHILATION CROSS SECTION

We derive the spin averaged cross section of direct positron annihilation into two gamma photons. The evolution of a wave packet Ψ^J with total angular momentum J is described by a time-dependent Schrödinger equation (TDSE)

$$i\frac{\partial}{\partial t}\Psi^J(\mathbf{R},\mathbf{r},t)=[H_0-iW(\mathbf{R},\mathbf{r})]\Psi^J(\mathbf{R},\mathbf{r},t) \quad (1)$$

with the three-body scattering Hamiltonian H_0 and the imaginary absorption potential [8]

$$-iW(\mathbf{R},\mathbf{r})=-i\frac{1}{2}\pi r_0^2 c\delta(\mathbf{R}-\mathbf{r}). \quad (2)$$

Here, \mathbf{R} and \mathbf{r} denote the position vectors of the positron and the electron relative to the proton, while $r_0=e^2/mc^2$ is the classical electron radius and c is the speed of light so that $r_0^2 c=\alpha^3$ in atomic units (a.u.) with the fine-structure constant α . The continuity equation for probability conservation is derived as

$$\frac{dP^J(t)}{dt}=-2\int d\mathbf{R}d\mathbf{r}|\Psi^J(\mathbf{R},\mathbf{r},t)|^2W(\mathbf{R},\mathbf{r}), \quad (3)$$

where

$$P^J(t)=\int d\mathbf{R}d\mathbf{r}|\Psi^J(\mathbf{R},\mathbf{r},t)|^2. \quad (4)$$

Hence, the time-dependent annihilation probability is calculated as

$$1-P^J(t)=2\int_0^t dt'\int d\mathbf{R}d\mathbf{r}|\Psi^J(\mathbf{R},\mathbf{r},t')|^2W(\mathbf{R},\mathbf{r}). \quad (5)$$

The direct annihilation cross section is given by the annihilation probability $1-P_\infty^J$ during a collision as

$$\sigma_{2\gamma}=\frac{\pi}{k^2}\sum_J(2J+1)(1-P_\infty^J) \quad (6)$$

with $k=\sqrt{2E}$ for positron energy E . The probability P_∞^J is determined through the time dependence as

$$1-P^J(t)=1-P_\infty^J+O(t) \quad (7)$$

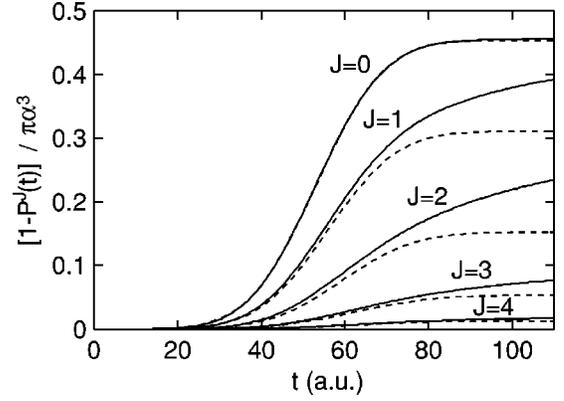


FIG. 1. Time dependence of the probability $1-P^J(t)$ for partial waves $J=0-4$ at a positron energy of 10 eV; the solid lines represent results of the TDCC calculation; the broken lines, contributions from the direct annihilation process, $\int_0^t dt' f_d(t')$, extracted with a fitting procedure (see the text).

in the region of $\tau \ll t \ll \tau_{\text{Ps}}$, where the time width τ of the wave packet is taken as $\tau_{\text{col}} \ll \tau \ll \tau_{\text{Ps}}$. The term of $O(t)$, originating from decay of the para-Ps formed, is irrelevant to the direct annihilation because it contributes after the wave packet has passed away asymptotically from the scattering center. Note that the annihilation cross section is often written in the conventional form $\sigma_{2\gamma}=\sigma_0 Z_{\text{eff}}$ with the effective annihilation parameter Z_{eff} , using the free pair-annihilation cross section $\sigma_0=\pi r_0^2(c/v)$ at relative velocity v [1].

III. NUMERICAL METHOD

The TDCC wave function is expanded [11] as

$$\Psi^J(\mathbf{R},\mathbf{r},t)=\frac{1}{Rr}\sum_{LI}\psi_{LI}^J(R,r,t)\mathcal{Y}_{LI}^J(\hat{\mathbf{R}},\hat{\mathbf{r}}), \quad (8)$$

where \mathcal{Y}_{LI}^J is the angular momentum eigenfunction, and L and l the orbital angular momenta associated with $\hat{\mathbf{R}}$ and $\hat{\mathbf{r}}$, respectively. As the incident wave packet, we take at $t=0$

$$\psi_{LI}^J(R,r,0)=g_{kL}(R)\phi_{1s}(r)\delta_{LJ}\delta_{l0} \quad (9)$$

with the ground-state wave function ϕ_{1s} of the hydrogen atom and a Gaussian form for the positron:

$$g_{kL}(R)=\frac{1}{(w^2\pi)^{1/4}}\exp\left[-\frac{(R-R_0)^2}{2w^2}\right]h_L^-(kR), \quad (10)$$

where R_0 and w are the center and the width of the wave packet, h_L^- being an asymptotic Hankel function. The position width is taken as $w=4\pi/k$; hence the time width is $\tau=2\pi/E$. The wave packet (10) has an energy width of $\Delta E/E=2\Delta k/k \approx 16\%$ with $\Delta k=1/w$. The TDSE (1) is solved with the Hamiltonian H_0 by using the numerical techniques developed in Ref. [11].

Figure 1 shows results for the time-dependent probability $1-P^J(t)$ for partial waves $J=0-4$; it rapidly increases during a collision ($40 \leq t \leq 80$ a.u.) and further linearly increases

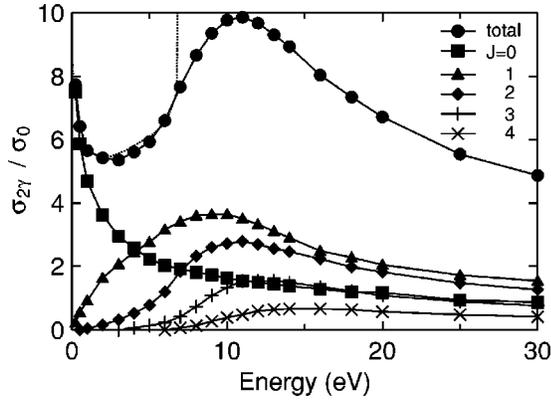


FIG. 2. Direct annihilation cross sections $\sigma_{2\gamma}$ scaled by the free pair-annihilation cross section $\sigma_0 = \pi r_0^2 (c/v)$. The solid lines with symbols represent the present result. Also plotted is a previous result (dotted line) for the total annihilation cross section obtained with a variational calculation [7].

after the collision is completed ($t > 80$ a.u.). Note that the probability could diverge, if the calculation were extended up to $t = \infty$. We extract the contribution of the direct process from the time-dependent probability as follows. The direct annihilation rate $f_d(t)$ is expected to be proportional to the probability density at the scattering center. The indirect annihilation rate $f_i(t)$ is given by the accumulated probability of Ps formation, which in turn is proportional to $\int dt f_d(t)$, because the Ps formation rate has similar time dependence to $f_d(t)$. Thus, taking account of the shape of the wave packet (10) and replacing R_0 by vt , we fit the probability in the form of

$$1 - P^J(t) = \int_0^t dt' [f_d(t') + f_i(t')], \quad (11)$$

where

$$f_d(t) = \frac{a}{(s^2 \pi)^{1/2}} \exp\left[-\frac{v^2}{s^2}(t-t_0)^2\right] \quad (12)$$

and

$$f_i(t) = \frac{b}{2} \left\{ 1 + \operatorname{erf}\left[\frac{v}{s}(t-t_0)\right] \right\}. \quad (13)$$

For $t \gg \tau$, the first contribution converges to a finite value, while the second contribution increases linearly with t . Four fitting parameters a , b , s , and t_0 are determined from the results of the TDCC calculation (see Fig. 1). The direct annihilation cross section is given by the probability $1 - P_\infty^J = \int_0^\infty dt f_d(t)$ obtained in this way.

IV. RESULTS AND DISCUSSION

The results are shown in Fig. 2 for the total and partial ($J=0-4$) cross sections of direct positron annihilation. Convergence of the total cross section with respect to J is apparent in the figure. Well below the Ps formation threshold (6.8

eV), the present result is in good agreement with the previous results of a variational calculation [7] (see Fig. 2), close-coupling calculations [8,9], and a TDCC calculation [12]. The agreement among all the calculations is better than 5%.

It is seen from Fig. 2 that the direct annihilation cross section does not indicate any singular behavior near the Ps formation threshold. This contrasts sharply with some of the previous results [7,8], where spurious divergence appears at the threshold (see Fig. 2). This divergence has been understood as caused by not taking full account of the absorption potential $-iW$ but taking only the first order effect [8,9]. Hence, the origin of the divergence is nothing but the term of $O(t)$ in Eq. (7), which we have eliminated to define the cross section. Recently, Gribakin and Ludlow [13] derived a modified threshold law by taking account of the finite energy width \hbar/τ_{Ps} coming from the Ps lifetime. In their result, the annihilation cross section is continuous at the threshold, and merged into the Ps formation cross section well above the threshold. Although continuous, the cross section increases steeply with energy in the narrow region of $\hbar/\tau_{\text{Ps}} \sim 10^{-5}$ eV. This behavior was further confirmed in an accurate calculation [9], which demonstrates the inseparability in principle of pair annihilation and Ps formation in the narrow region that is experimentally unresolved. On the other hand, the direct annihilation cross section in Fig. 2 does not exhibit such a steep increase near the threshold, nor any sign of the presence of the threshold. This behavior manifests the physical separability between the direct and indirect annihilation processes.

It is further seen from Fig. 2 that the cross section is enhanced around 10 eV, well above the Ps formation threshold. The enhancement comes from the partial waves of $J \geq 1$. For instance, the P -wave ($J=1$) contribution peaks at 9 eV, close to the $n=2$ excitation energy. Contributions from higher partial waves ($J \geq 2$) also have a peak, the position of which shifts toward higher energy with J . These behaviors are understood as follows. The positron is decelerated by dissipating the kinetic energy in electronic excitation, and thereby is temporally captured by the atom. Thus, the annihilation cross section is enhanced around the excitation threshold energies, where the kinetic energy is most effectively dissipated. In fact, the temporal capture is directly proved by looking into the time delay of the wave packet, which is calculated as the difference of mean passage times with and without the collision interaction. As shown in Fig. 3, the time delay is elongated around 10 eV for the partial waves of $J \geq 1$. It is seen from the comparison of Figs. 2 and 3 that the annihilation cross section has parallel energy dependence to the time delay; the longer the time of capture, the larger the probability of annihilation. We conclude from these observations that the enhancement of direct annihilation is due to the temporal positron capture by the hydrogen atom while the electron is thereby excited to the $n \geq 2$ states. Note that the enhancement is not appreciable in the S -wave contribution (see Figs. 2 and 3), because the $1s$ to ns excitation is forbidden in the first order perturbation by the dominant dipole interaction and also because the $1s$ to np excitation is suppressed near the threshold by the centrifugal barrier.

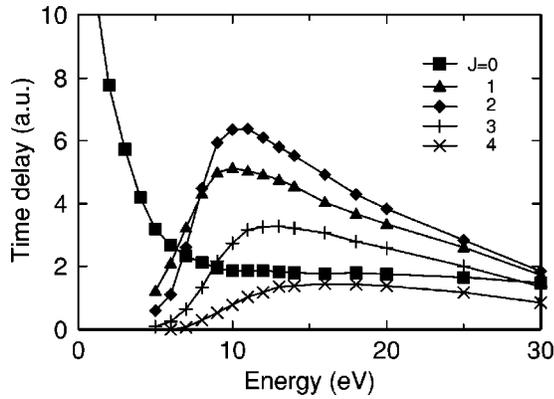
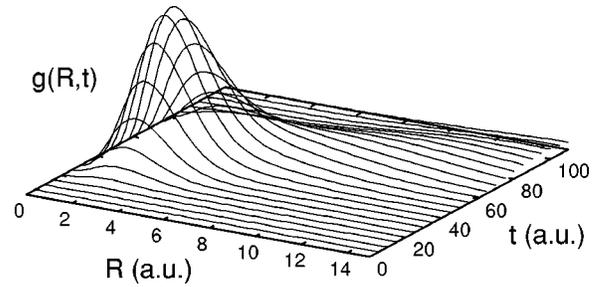


FIG. 3. Time delay of positron wave packets.

We get further insight into the mechanism of the enhancement around 10 eV by investigating the evolution of the radial distribution

$$g(R,t) = \sum_J 2(2J+1) \int d\mathbf{R} d\mathbf{r} |\Psi^J(\mathbf{R},\mathbf{r},t)|^2 W(\mathbf{R},\mathbf{r}) \quad (14)$$

of annihilation position [14]. As seen in Fig. 4, the distribution calculated at 10 eV has a peak around $R=2$ a.u. This comes from the direct annihilation, because the peak grows and diminishes in height but its position does not move while the wave packet is passing the target atom ($40 \leq t \leq 80$ a.u.). The peak is located outside the peak radius (1 a.u.) of the ground-state ($1s$) electron cloud, which is consistent with the conclusion derived in the last paragraph that the electron is excited to the $n \geq 2$ states. Note that the distribution exhibits an expanding skirt, which eventually turns to a broad ridge after the dominant peak has diminished. This comes from the indirect annihilation via Ps formation, because it remains even after the wave packet has passed away

FIG. 4. Evolution of the radial distribution $g(R,t)$ of annihilation position at 10 eV [14].

($t > 80$ a.u.). It would reach $R \geq 10^{-4}$ m within the Ps lifetime.

V. SUMMARY

In the present paper, we have investigated direct positron annihilation with a hydrogen atom in terms of the evolution of the wave packet. It was found that the cross section is enhanced around 10 eV. It was shown through the time delay of the wave packet that the enhancement is due to temporal positron capture with electronic excitation in the target. The enhancement through this mechanism is expected to appear universally in atoms and small molecules, irrespective of whether the energy is above or below the Ps formation threshold.

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