Electron-spin amplitudes in the exchange quenching of orthopositronium

Sudha R. Swaminathan,[1] W. G. Merrill,[2] S. Glynn,[2] and J. G. Quattrucci[2]
[1]*Department of Physical and Earth Sciences, Worcester State University, 486 Chandler Street, Worcester, Massachusetts 01602, USA*
[2]*Department of Chemistry, Worcester State University, 486 Chandler Street, Worcester, Massachusetts 01602, USA*
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We study single collisions between orthopositronium beams and targets with unpaired electrons, in which the conversion (quenching) of the long-lived orthopositronium into the short-lived parapositronium occurs as a result of electron exchange. We consider unpolarized and polarized targets with one and two unpaired electrons. Angular-momentum coupling and density-matrix techniques are used to calculate the probabilities of obtaining either para- or orthopositronium after the collisions. The probability of the initial orthopositronium being converted to parapositronium (quenching probability) is given in terms of complex scattering amplitudes labeled with total electron spin. Quenching probabilities are calculated for polarized and unpolarized targets, with and without detection of the spins of the targets after the collisions. Ratios of the probabilities of producing parapositronium to that of obtaining orthopositronium are given in terms of electron-spin amplitudes.

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I. INTRODUCTION

We study elastic collisions between orthopositronium beams and atomic or molecular targets with two outgoing products, a positronium atom and a target atom or molecule. Orthopositronium is the spin-1 bound state of an electron and a positron, which decays into *three* photons after 142.05 ns [1], while parapositronium is the spin-0 bound state, which decays into *two* photons after 0.125 16 ns [2]. We consider collisions between orthopositronium and two types of targets: (i) doublets with one unpaired electron and a total unpaired-electron spin of $1/2$ and (ii) triplets with two unpaired electrons and a total unpaired-electron spin of 1. During the interaction, an electron from the target can be exchanged with the electron in the orthopositronium, and either parapositronium with a spin of 0 or orthopositronium with a spin of 1 can be produced along with targets in ground states having spins of $1/2$ or 1. The conversion of orthopositronium to parapositronium is referred to as the quenching of the long lifetime of orthopositronium. As we will show in the Method section, for singlets with no unpaired electrons and a total unpaired-electron spin of 0, there is no quenching if the target remains in the ground state.

Theoretical studies of the quenching of orthopositronium as well as the results of scattering experiments with positronium beams are extensively reviewed in Ref. [3]. The authors note that for targets with no unpaired electrons (singlets) such as helium, exchange quenching can occur only if the positronium energy is above that of the first triplet excited state, but for targets with one unpaired electron (doublets) such as hydrogen, or targets with two unpaired electrons (triplets) such as oxygen, quenching can occur for any positronium energy [3].

For elastic scattering of positronium with hydrogen, scattering lengths as well as ratios of conversion cross sections to total elastic scattering cross sections have been calculated using a static exchange approximation [4] and through an extraction of s-wave phase shifts from generalized Hylleraas wave functions [5,6]. In Ref. [4], for positronium kinetic energies ranging from 0 to 4.352 eV, the ratios of conversion cross sections to total elastic scattering cross sections ranged from 8.1% to 12.5%, while in Ref. [6], for five positronium energies ranging from 0 to 3.801 eV, the ratios ranged from 3.7% to 4.9%. The effect of electron exchange was studied in positronium-hydrogen scattering using a close-coupling approximation in Ref. [7]. For positronium energies ranging from 0.068 to 4.352 eV, the ratios of conversion cross sections to total elastic scattering cross sections ranged from 8.11% to 14.1% when electron exchange was taken into account [7]. Moreover, the elastic cross section *without* electron exchange was approximately two orders of magnitude less than that *with* electron exchange [7]. Calculations of spin-averaged total cross sections have also been made using the coupled-pseudostate method in the energy range from 0 to 40 eV [8]. The authors noted that when electron exchange was accounted for, there was little variation in the cross sections calculated using different approximations [8]. More recently, positronium-hydrogen scattering has been studied in the 0–5.1 eV range with the Kohn variational method [9] and in the 0–6.5 eV range with the coupled-pseudostate method [10]. We compare our analysis for doublet targets to the differential cross sections presented in Ref. [10].

The quenching of orthopositronium in oxygen has been studied using the angular correlation of annihilation radiation (ACAR) technique [11,12]. With this technique, (i) only two-photon annihilation events are detected [13], (ii) after the conversion from ortho- to parapositronium, an increase in the number of two-photon annihilation events is observed [12], and (iii) in many situations, two-photon events following a conversion are easily distinguished from other two-photon events [12]. The authors concluded that the conversion of orthopositronium to parapositronium has two components: (i) elastic conversion, in which the oxygen molecule is in the ground state with two unpaired electrons (triplet) and (ii) inelastic conversion, in which the oxygen molecule is in an excited state with no unpaired electrons (singlet) [12]. The cross section was measured to be $1.0 \pm 0.3 \times 10^{-23}$ m^2 for the elastic-conversion process and approximately 5×10^{-21} m^2 for the inelastic-conversion process [12]. More recently, the age-momentum correlation (AMOC) technique was used to determine the cross section for the elastic-conversion process to be $(1.16 \pm 0.01) \times 10^{-23}$ m^2 [14].

The conversion of orthopositronium to parapositronium may also be important in the annihilation of positrons from the

center of the Milky Way galaxy [15,16]. The annihilation of the galactic positrons can occur after the formation of positronium in the interstellar medium [17]. The annihilation occurs by either (i) two-photon decays in which the energy of each photon equals the rest mass of the electron (positron), that is, a discrete value of 0.511 MeV, or (ii) three-photon decays in which the energies of the photons can be distributed over a continuous range from 0 to 0.511 MeV [15–17]. Based on analysis of the electron-positron annihilation spectrum from the center of the Milky Way galaxy, the ratio of three-photon fluxes to that of two-photon fluxes was reported to be 3.95 ± 0.32 [18]. Based on spin-counting arguments, one can assume that in collisions between positrons and the interstellar medium, triplet orthopositronium is three times more likely to form than singlet parapositronium. For each positron that forms parapositronium and decays to two photons, three positrons can form orthopositronium, each of which can decay to three photons. The theoretical ratio of the fluxes should therefore equal 4.5 [15]. One explanation for the lower measured ratio of the fluxes is that some of the orthopositronium converts to parapositronium in the grains of interstellar dust *before* its subsequent annihilation to photons [15].

In 1958, Ferrell used the Pauli exclusion principle to point out that when the electron in the orthopositronium is exchanged either with the one unpaired electron of a nitric oxide molecule, or with either of the two unpaired electrons of the oxygen molecule, quenching can be achieved *without* a spin flip of the exchange electrons [19]. An exchange of electrons without a spin flip is referred to as Majorana exchange [20,21]. We assume that interaction terms in the Hamiltonian that lead to a spin flip are negligible, and require that the total spin of all the electrons in the target-positronium system, the spin of the positron, the total spin of the target-positronium system, and the total spin magnetic quantum number are conserved. Ferrell's results are given in terms of scattering amplitudes for direct interactions (D) and exchange interactions (E). Our results are given in terms of scattering amplitudes f^{S_e}, where S_e is the total electron spin. The electron-spin amplitudes f^{S_e} are independent of the positron spin, the total spin, as well as the total magnetic quantum number, and can be expressed in terms of the scattering amplitudes D and E. We have included all the possible spin states in a given collision process, while in Ferrell's paper, only the states relevant to the discussion on quenching were emphasized; for example, he noted a missing term in Fig. 1d of Ref. [19] for oxygen (triplet). There is also a discrepancy between our spin-scattering matrix elements and Ferrell's which arises from the signs for the exchange interactions in Figs. 1b and 1c of Ref. [19] for nitric oxide (doublet).

We use angular-momentum coupling techniques to construct the spin-scattering matrices which contain the amplitudes for the transitions from the spin states of the initial (target-orthopositronium) system to the spin states of the final (target-positronium) system. We construct spin-density matrices for describing the initial and final system from Pauli spin matrices, and polarization vectors and tensors, using techniques described in Refs. [22–24]. In the collisions we study, the incoming orthopositronium beam is unpolarized, but the targets can be either polarized or unpolarized. A beam is considered to be polarized if its spins are aligned along the positive or negative direction of its momentum vector, which is chosen to be along a specific axis. A beam is considered to be unpolarized if it contains a mixture of spins in both directions. Para- and orthopositronium formation probabilities were obtained from spin-density matrices in the study of collisions between spin-polarized positron beams and atomic or molecular targets [25,26]. In this work, we calculate the probabilities of mixed-spin systems such as the target-positronium system being in specific spin states. Specifically, from the diagonal elements of spin-density matrices for the final system, we calculate the probabilities for obtaining both types of positronium, for both polarized and unpolarized targets, with and without detection of the spins of the targets just after the collisions. We present three types of results: (i) total probabilities for obtaining both types of positronium, (ii) quenching probabilities for producing parapositronium after electron exchange, and (iii) ratios of para- to orthopositronium production probabilities.

II. METHOD

In this section we describe the construction of the spin-scattering matrices, M, and the spin-density matrices, ρ_{in}, for the initial system. The spin-density matrices for the final system, ρ_{out}, are calculated from the product $M\rho_{\text{in}}M^{\dagger}$, where $M^{\dagger}$ represents the conjugate-transpose matrices of M. We introduce projection operators $P^{(\text{para,target})}$ and $P^{(\text{ortho,target})}$ in order to select the para- and orthopositronium states which are produced in *coincidence* with particular target states. The probability of producing parapositronium equals the expectation value of $P^{(\text{para,target})}$ which equals the trace of $\rho_{\text{out}}P^{(\text{para,target})}$, while the probability of obtaining orthopositronium equals the expectation value of $P^{(\text{ortho,target})}$ which equals the trace of $\rho_{\text{out}}P^{(\text{ortho,target})}$. Probabilities of obtaining para- or orthopositronium in *coincidence* with particular target states are calculated from appropriate diagonal elements of $\rho_{\text{out}}P^{(\text{para,target})}$ or $\rho_{\text{out}}P^{(\text{ortho,target})}$, respectively.

We express the collisions as $f + c \rightarrow g + d$, where f represents the target in the initial system, c represents the incoming orthopositronium, g represents the target in the final system, and d represents both types of positronium. The target f and orthopositronium c interact to form intermediate states in which electron exchange can occur. After the electron exchange, the intermediate states develop into the target g and either type of positronium d. As the initial system evolves from f and c to g and d, the quantum numbers representing the total electron spin S_e of the target-positronium system, the positron spin $S_p = \frac{1}{2}$, the total spin S_t of the target-positronium system, and the total spin magnetic quantum numbers M_t are all conserved. The intermediate states are labeled with the conserved quantum numbers as $|[S_e\ \frac{1}{2}]S_tM_t\rangle$. We define complex scattering amplitudes f^{S_e} and their complex conjugates $(f^{S_e})^*$ which are independent of S_t, M_t, and the positron spin, as follows:

$$f^{S_e} = \left\langle \left[S_e\ \tfrac{1}{2}\right] S_tM_t \middle| M \middle| \left[S_e\ \tfrac{1}{2}\right] S_tM_t \right\rangle, \tag{1}$$

$$(f^{S_e})^* = \left\langle \left[S_e\ \tfrac{1}{2}\right] S_tM_t \middle| M^{\dagger} \middle| \left[S_e\ \tfrac{1}{2}\right] S_tM_t \right\rangle. \tag{2}$$

The amplitudes for transitions from particular states of f and c to particular states of g and d are given by the elements of the spin-scattering matrices M with rows and columns

TABLE I. Matrix elements $\langle S_g M_g|\langle S_d M_d|M|S_f M_f\rangle|S_c M_c\rangle$ of spin-scattering matrices for the reactions $f + c \to g + d$. The columns are labeled with the initial state basis vectors $|S_f M_f\rangle|S_c M_c\rangle$ where f represents a doublet with $S_f = \frac{1}{2}$, or a triplet with $S_f = 1$. The rows are labeled with the final state basis vectors $\langle S_g M_g|\langle S_d M_d|$ where g represents a doublet with $S_g = \frac{1}{2}$ or a triplet with $S_g = 1$. The incoming orthopositronium spin $S_c = 1$, while the outgoing positronium spins are $S_d = 0$ for parapositronium and $S_d = 1$ for orthopositronium. The matrix elements for doublets are given in terms of D_2 and E_2, with $D_2 = \frac{1}{2}f^0 + \frac{1}{2}f^1$ and $E_2 = \frac{1}{3}f^{1/2} - \frac{1}{3}f^{3/2}$. The matrix elements for triplets are given in terms of D_3 and E_3, with $D_3 = \frac{2}{3}f^{1/2} + \frac{1}{3}f^{3/2}$ and $E_3 = \frac{1}{3}f^{1/2} - \frac{1}{3}f^{3/2}$.

Doublet	$\left\|\frac{1}{2}\frac{1}{2}\right\rangle\|11\rangle$	$\left\|\frac{1}{2}\frac{1}{2}\right\rangle\|10\rangle$	$\left\|\frac{1}{2}\frac{1}{2}\right\rangle\|1{-}1\rangle$	$\left\|\frac{1}{2}{-}\frac{1}{2}\right\rangle\|11\rangle$	$\left\|\frac{1}{2}{-}\frac{1}{2}\right\rangle\|10\rangle$	$\left\|\frac{1}{2}{-}\frac{1}{2}\right\rangle\|1{-}1\rangle$			
$\left\langle\frac{1}{2}\frac{1}{2}\right\|\langle 00\|$		$-\frac{1}{2}E_2$		$\sqrt{\frac{1}{2}}E_2$					
$\left\langle\frac{1}{2}{-}\frac{1}{2}\right\|\langle 00\|$			$-\sqrt{\frac{1}{2}}E_2$		$\frac{1}{2}E_2$				
$\left\langle\frac{1}{2}\frac{1}{2}\right\|\langle 11\|$	$D_2{-}E_2$								
$\left\langle\frac{1}{2}\frac{1}{2}\right\|\langle 10\|$		$D_2 - \frac{1}{2}E_2$		$-\sqrt{\frac{1}{2}}E_2$					
$\left\langle\frac{1}{2}\frac{1}{2}\right\|\langle 1{-}1\|$			D_2		$-\sqrt{\frac{1}{2}}E_2$				
$\left\langle\frac{1}{2}{-}\frac{1}{2}\right\|\langle 11\|$		$-\sqrt{\frac{1}{2}}E_2$		D_2					
$\left\langle\frac{1}{2}{-}\frac{1}{2}\right\|\langle 10\|$			$-\sqrt{\frac{1}{2}}E_2$		$D_2{-}\frac{1}{2}E_2$				
$\left\langle\frac{1}{2}{-}\frac{1}{2}\right\|\langle 1{-}1\|$						$D_2{-}E_2$			
Triplet	$\|11\rangle\|11\rangle$	$\|11\rangle\|10\rangle$	$\|11\rangle\|1{-}1\rangle$	$\|10\rangle\|11\rangle$	$\|10\rangle\|10\rangle$	$\|10\rangle\|1{-}1\rangle$	$\|1{-}1\rangle\|11\rangle$	$\|1{-}1\rangle\|10\rangle$	$\|1{-}1\rangle\|1{-}1\rangle$
$\langle 11\|\langle 00\|$		$-E_3$		E_3					
$\langle 10\|\langle 00\|$			$-E_3$				E_3		
$\langle 1{-}1\|\langle 00\|$						$-E_3$		E_3	
$\langle 11\|\langle 11\|$	$D_3{-}2E_3$								
$\langle 11\|\langle 10\|$		$D_3{-}E_3$		$-E_3$					
$\langle 11\|\langle 1{-}1\|$			D_3		$-E_3$				
$\langle 10\|\langle 11\|$		$-E_3$		$D_3{-}E_3$					
$\langle 10\|\langle 10\|$			$-E_3$		$D_3{-}E_3$		$-E_3$		
$\langle 10\|\langle 1{-}1\|$						$D_3{-}E_3$		$-E_3$	
$\langle 1{-}1\|\langle 11\|$					$-E_3$		D_3		
$\langle 1{-}1\|\langle 10\|$						$-E_3$		$D_3{-}E_3$	
$\langle 1{-}1\|\langle 1{-}1\|$									$D_3{-}2E_3$

representing the final and initial states, respectively. The states are labeled with spin and magnetic quantum numbers and the elements are expressed in terms of the complex scattering amplitudes as follows:

$$\langle S_g M_g|\langle S_d M_d|M|S_f M_f\rangle|S_c M_c\rangle = \sum_{S_t M_t}\langle S_g M_g|\langle S_d M_d|\left|\left[S_e \tfrac{1}{2}\right] S_t M_t\right\rangle f^{S_e}\left\langle\left[S_e \tfrac{1}{2}\right] S_t M_t\right|\,|S_f M_f\rangle|S_c M_c\rangle. \tag{3}$$

To calculate the matrix elements, we (i) couple the initial target and orthopositronium state spins, (ii) transform the coupled initial states to the intermediate states, (iii) transform the intermediate states to the coupled final states, and (iv) uncouple the coupled final states. Clebsch-Gordan coefficients are used in steps (i) and (iv), and the U coefficients of Jahn are used in steps (ii) and (iii) as shown in Eqs. (4)–(7) below. The U coefficients are equal to $\sqrt{(2S_e+1)(2S_c+1)}$ times the standard Racah coefficients W [27].

$$|S_f M_f\rangle|S_c M_c\rangle = \sum_{S_t M_t}\langle S_f M_f S_c M_c|S_t M_t\rangle|[S_f S_c]S_t M_t\rangle, \tag{4}$$

$$|[S_f S_c]S_t M_t\rangle = \sum_{S_e} U\left(S_f \tfrac{1}{2} S_t \tfrac{1}{2}; S_e S_c\right)|[S_e \tfrac{1}{2}]S_t M_t\rangle, \tag{5}$$

$$\left|\left[S_e \tfrac{1}{2}\right] S_t M_t\right\rangle = \sum_{S_d} U\left(S_g \tfrac{1}{2} S_t \tfrac{1}{2}; S_e S_d\right)|[S_g S_d]S_t M_t\rangle, \tag{6}$$

$$|[S_g S_d]\,S_t M_t\rangle = \sum_{M_g M_d}\langle S_g M_g\, S_d M_d\,|S_t M_t\rangle|S_g M_g\rangle|S_d M_d\rangle. \tag{7}$$

The spin assignments are as follows: $S_c = S_d = 1$, $M_c = M_d = 1,\ 0,\ -1$, for orthopositronium and $S_d = M_d = 0$, for parapositronium. For a singlet target with no unpaired

electrons, $S_f = S_g = 0$, and the total spin of the initial system, $S_t = 1$. Due to conservation, the total spin of the final system is also equal to 1, which can occur only if $S_d = 1$. In other words, in a collision between a singlet and orthopositronium in which electron exchange occurs without a spin flip, if the target remains in the ground state after the collision, there will be no quenching of the incoming orthopositronium. For doublet targets with one unpaired electron, $S_f = S_g = \frac{1}{2}$, $M_f = M_g = \frac{1}{2}, -\frac{1}{2}$, and for triplet targets with two unpaired electrons, $S_f = S_g = 1$, $M_f = M_g = 1, 0, -1$. With one electron in orthopositronium, the total electron spin S_e can be 0 or 1 for collisions with doublets and $\frac{1}{2}$ or $\frac{3}{2}$ for triplets. The matrix elements $\langle S_g M_g|\langle S_d M_d|M|S_f M_f\rangle|S_c M_c\rangle$, shown in Table I were calculated using the spin values given above. As mentioned in the Introduction, these matrix elements can be compared to the scattering amplitudes of Ref. [19].

The individual spin-density matrices, $\rho_{\rm in}$, for the incoming orthopositronium and the targets are constructed using the methods of Refs. [24–26]. We choose the momentum direction of the targets to be along the positive z axis. A target is considered to be unpolarized when the beam contains a mixture of spins along positive z and negative z. For unpolarized orthopositronium,

$$\rho_{\rm in} = \tfrac{1}{3}(E), \tag{8}$$

where E is a 3×3 unit matrix. For doublets with only a z component of the vector polarization,

$$\rho_{\rm in} = \tfrac{1}{2}(\varepsilon + P_z\sigma_z), \tag{9}$$

where ε is a 2×2 unit matrix, σ_z is a Pauli matrix, and the polarization vector, $P_z = {\rm Tr}(\rho_{\rm in}\sigma_z) = \pm 1$. For unpolarized targets, $P_z = 0$. For triplet vector and tensor polarizations with only z components,

$$\rho_{\rm in} = \tfrac{1}{3}\left(E_1 + \tfrac{3}{2}P_z S_z + \tfrac{1}{2}P_{zz}S_{zz}\right), \tag{10}$$

where E_1 is a 3×3 unit matrix, S_z is a 3×3 spin matrix, $S_{zz} = 3S_z^2 - 2E_1$, $P_z = {\rm Tr}(\rho_{\rm in}{\rm S}_z)$, and $P_{zz} = {\rm Tr}(\rho_{\rm in}{\rm S}_{zz})$. For $P_z = \pm 1$, $P_{zz} = 1$, for $P_z = 0$, $P_{zz} = -2$, and for unpolarized targets, $P_z = P_{zz} = 0$. The spin-density matrices for the initial doublet-orthopositronium system are calculated by taking direct products of the individual matrices from Eqs. (8) and (9). We get

$$\rho_{\rm in} = \tfrac{1}{6}\left[(\varepsilon \times E) + P_z(\sigma_z \times E)\right]. \tag{11}$$

Similarly, for the initial triplet-orthopositronium system, from Eqs. (8) and (10), we get

$$\rho_{\rm in} = \tfrac{1}{9}\left[(E_1 \times E) + \tfrac{3}{2}P_z(S_z \times E) + \tfrac{1}{2}P_{zz}(S_{zz} \times E)\right]. \tag{12}$$

The projection operators $P^{(\rm para,target)}$ and $P^{(\rm ortho,target)}$ are defined such that for specific S_g and M_g only elements $\langle S_g M_g|\langle S_d = 0\, M_d = 0|\rho_{\rm out}P^{(\rm para,target)}|S_g M_g\rangle|00\rangle$ and $\langle S_g M_g|\langle S_d = 1 M_d|\rho_{\rm out}P^{(\rm ortho,target)}|S_g M_g\rangle|1 M_d\rangle$ are nonzero. The matrix elements, $\rho_{\rm out}$ are as follows:

$$\begin{aligned}
&\langle S_g M_g|\langle 00|\rho_{\rm out}P^{(\rm para,target)}|S_g M_g\rangle|00\rangle \\
&\quad = \sum_{M_f M_c}\sum_{M_f'' M_c''}\langle S_g M_g|\langle 00|M|S_f M_f\rangle|S_c M_c\rangle \\
&\qquad \times \langle S_f M_f|\langle S_c M_c|\rho_{\rm in}|S_f M_f''\rangle|S_c M_c''\rangle \\
&\qquad \times \langle S_f M_f''|\langle S_c M_c''|M^\dagger|S_g M_g\rangle|00\rangle,
\end{aligned} \tag{13}$$

and

$$\begin{aligned}
&\langle S_g M_g|\langle 1 M_d|\rho_{\rm out}P^{(\rm ortho,target)}|S_g M_g\rangle|1 M_d\rangle \\
&\quad = \sum_{M_d}\sum_{M_f M_c}\sum_{M_f'' M_c''}\langle S_g M_g|\langle 1 M_d|M|S_f M_f\rangle|S_c M_c\rangle \\
&\qquad \times \langle S_f M_f|\langle S_c M_c|\rho_{\rm in}|S_f M_f''\rangle|S_c M_c''\rangle \\
&\qquad \times \langle S_f M_f''|\langle S_c M_c''|M^\dagger|S_g M_g\rangle|1 M_d\rangle,
\end{aligned} \tag{14}$$

with $\rho_{\rm in}$ given by Eq. (11) for doublets and by Eq. (12) for triplets. The probabilities of obtaining para- or orthopositronium are calculated from the diagonal elements of $\rho_{\rm out}$, both for specific final states of the targets and for cases in which the spins are not detected.

III. RESULTS

We present below (i) total probabilities for obtaining both types of positronium, (ii) quenching probabilities, and (iii) ratios of para- to orthopositronium production probabilities, for doublet and triplet targets. We consider the time just after the collisions and give our results in terms of complex scattering amplitudes f^{S_e}, which have units of length. As explained in the Method section, the total electron spin $S_e = 0$ or 1 for doublets, and $\frac{1}{2}$ or $\frac{3}{2}$ for triplets.

A. Total probabilities

The total probability of obtaining both types of positronium for either polarized or unpolarized targets, with or without detection of the spins of the targets after the collisions, is labeled T_d for doublets and T_t for triplets, and is calculated to be

$$T_d = \tfrac{1}{4}|f^0|^2 + \tfrac{3}{4}|f^1|^2, \tag{15}$$

and

$$T_t = \tfrac{1}{3}|f^{1/2}|^2 + \tfrac{2}{3}|f^{3/2}|^2. \tag{16}$$

B. Quenching probabilities

The quenching probability is the probability of producing parapositronium from the conversion of the incoming orthopositronium. The quenching probability in collisions with unpolarized or polarized targets in which the spins of the targets are not detected after the collisions is labeled P_d for doublets and P_t for triplets and is calculated to be

$$P_d = \tfrac{1}{16}|f^0 - f^1|^2, \tag{17}$$

and

$$P_t = \tfrac{2}{27}|f^{1/2} - f^{3/2}|^2. \tag{18}$$

C. Ratios

For unpolarized targets with or without detection of the spins after the collisions, as well as for polarized targets without detection of the spins after the collisions, the ratio of the probability of producing parapositronium to that of

obtaining orthopositronium is labeled R_{ud} for doublets and R_{ut} for triplets and is calculated to be

$$R_{ud} = \frac{P_d}{T_d - P_d}, \tag{19}$$

and

$$R_{ut} = \frac{P_t}{T_t - P_t}. \tag{20}$$

For polarized targets, when the spins are detected and $M_g = M_f$ but not zero, the ratio for doublets is

$$R_{pd} = \frac{P_d}{3T_d - 7P_d}, \tag{21}$$

and for triplets is

$$R_{pt} = \frac{P_t}{2T_t - 4P_t}. \tag{22}$$

Quenching probabilities and ratios for unpolarized and polarized targets, with and without detection of the spins of the targets, are given in Table II. We note that for polarized doublet and polarized triplet targets, the quenching probabilities and ratios in the cases in which the spins of the targets are detected are different than those in which the spins of the targets are not detected.

The complex electron-spin amplitudes defined in Eq. (1) are difficult to calculate exactly. However, they can be written in terms of their real and imaginary parts as $f^1 = r^1 e^{i\theta_1}$ and $f^0 = r^0 e^{i\theta_0}$ for doublets, and as $f^{3/2} = r^{3/2} e^{i\theta_{3/2}}$ and $f^{1/2} = r^{1/2} e^{i\theta_{1/2}}$ for triplets. With only two independent equations given by the ratios R_{ud} and R_{pd} or R_{ut} and R_{pt}, we cannot solve for the four unknowns (two real parts, and two imaginary parts). Our goal is to understand the nature of these amplitudes by exploring the dependence of the real parts of the amplitudes on measurable positronium probabilities obtained through cross sections.

We begin by substituting $f^1 = r^1 e^{i\theta_1}$ and $f^0 = r^0 e^{i\theta_0}$ in the expressions for P_d, T_d, R_{ud}, and R_{pd}. Then, we manipulate the expressions for R_{ud} and R_{pd} and obtain a quadratic equation in the ratio $\frac{r^1}{r^0}$, which we solve as

$$\frac{r^1}{r^0} = \frac{-(R_{ud} - 7R_{pd})\cos(\theta_1 - \theta_0) \pm \sqrt{(R_{ud} - 7R_{pd})^2 \cos^2(\theta_1 - \theta_0) - (11R_{ud} - 29R_{pd})(3R_{ud} - 5R_{pd})}}{(11R_{ud} - 29R_{pd})}. \tag{23}$$

Similarly, by substituting $f^{3/2} = r^{3/2} e^{i\theta_{3/2}}$ and $f^{1/2} = r^{1/2} e^{i\theta_{1/2}}$ in the expressions for P_t, T_t, R_{ut}, and R_{pt}, and manipulating the expressions for R_{ut} and R_{pt}, we obtain a quadratic equation in the ratio $\frac{r^{3/2}}{r^{1/2}}$, which we solve as

$$\frac{r^{3/2}}{r^{1/2}} = \frac{-(R_{ut} - 4R_{pt})\cos(\theta_{3/2} - \theta_{1/2}) \pm \sqrt{(R_{ut} - 4R_{pt})^2 \cos^2(\theta_{3/2} - \theta_{1/2}) - (4R_{ut} - 7R_{pt})(7R_{ut} - 10R_{pt})}}{2(4R_{ut} - 7R_{pt})}. \tag{24}$$

The ratios are functions of only two measurable parameters, R_{ud} and R_{pd} for doublets, and R_{ut} and R_{pt} for triplets. Given values for the parameters, the ratios can be plotted for $0 \leqslant (\theta_1 - \theta_0) \leqslant 360^\circ$ and $0 \leqslant (\theta_{3/2} - \theta_{1/2}) \leqslant 360^\circ$ and special cases noted. For example, if orthopositronium is three times more likely than parapositronium to be a product, then 75% of the spin states of the final target-positronium system would be orthopositronium states and 25% would be parapositronium states. Then, for doublets, $P_d = \frac{1}{4}T_d$, $R_{ud} = \frac{1}{3}$, and $R_{pd} = \frac{1}{5}$, and if $\theta_1 - \theta_0 = 180^\circ$, $\frac{r^1}{r^0} = 0$ or 1. For triplets, $P_t = \frac{1}{4}T_t$, $R_{ut} = \frac{1}{5}$, and $R_{pt} = \frac{1}{4}$, and if $\theta_{3/2} - \theta_{1/2} = 180^\circ$, $\frac{r^{3/2}}{r^{1/2}} = 0.065$ or 1.5345.

As mentioned in the Introduction, positronium-hydrogen scattering has been studied with the coupled-pseudostate method by Blackwood *et al.*, and differential cross sections are calculated in terms of electron-spin amplitudes [10]. The results of the spin analysis in Ref. [10] are for collisions between polarized orthopositronium beams and unpolarized hydrogen targets (doublet) in which the final state spins of the hydrogen and positronium are not measured. We consider unpolarized orthopositronium beams and obtain our results from diagonal elements of density matrices. We consider cases in which both types of positronium are produced *with* or *without* detection of the spins of the targets. The probabilities T_d and P_d given by Eqs. (15) and (17) above are consistent with the differential cross sections given as Eqs. (21) and (20), respectively, in Ref. [10], if (i) we assume that the electron-spin amplitudes of Ref. [10] are defined in the same way as Eq. (1) above, and (ii) we multiply our values by the ratio of the momenta $\frac{p_{ab}}{p_0}$ defined in Ref. [10].

The elastic spin-conversion cross section for the quenching of orthopositronium in oxygen given by Shinohara *et al.* in Ref. [14] is proportional to the quenching probability P_t for triplet targets. The total cross section for the scattering of orthopositronium in oxygen in which both orthopositronium and parapositronium are obtained after electron exchange, and the oxygen is in the ground state, would be proportional to the total probability T_t. Given the total cross section and the proportionality constants, we could calculate the ratios R_{ut} or R_{pt}, substitute the values in Eq. (24), and plot specific expressions for $\frac{r^{3/2}}{r^{1/2}}$.

We study orthopositronium-target collisions in which electron exchange occurs and two types of outgoing products are obtained, targets in the ground state and para- or orthopositronium. If λ_o is the rate at which the outgoing orthopositronium decays to three photons and $\lambda_{\rm eq}$ is the rate at which the incoming orthopositronium converts to parapositronium through exchange quenching, then, after the collisions, (i) the fraction of the total number of decays which are two-photon decays equals $\frac{\lambda_{\rm eq}}{\lambda_o + \lambda_{\rm eq}}$ and (ii) the ratio of the

TABLE II. Quenching probabilities and ratios of the probabilities of producing parapositronium to that of obtaining orthopositronium just after collisions in which an orthopositronium beam collides with an unpolarized doublet, a polarized doublet with $M_f = \pm\frac{1}{2}$, an unpolarized triplet, or a polarized triplet with $M_f = \pm 1$ or 0, to form parapositronium with $S_d = 0$ or orthopositronium with $S_d = 1$. The final state spins of the doublet and triplet targets are $S_g = \frac{1}{2}$ and $S_g = 1$, respectively. For doublets, $P_d = \frac{1}{16}|f^0 - f^1|^2$, $T_d = \frac{1}{4}|f^0|^2 + \frac{3}{4}|f^1|^2$, $R_{ud} = \frac{P_d}{T_d - P_d}$, and $R_{pd} = \frac{P_d}{3T_d - 7P_d}$. For triplets, $P_t = \frac{2}{27}|f^{1/2} - f^{3/2}|^2$, $T_t = \frac{1}{3}|f^{1/2}|^2 + \frac{2}{3}|f^{3/2}|^2$, $R_{ut} = \frac{P_t}{T_t - P_t}$ and $R_{pt} = \frac{P_t}{2T_t - 4P_t}$.

Doublet	M_g undetected	$M_g = \frac{1}{2}$	$M_g = -\frac{1}{2}$	
Unpolarized	P_d	$\frac{1}{2}P_d$	$\frac{1}{2}P_d$	
$M_f = \frac{1}{2}$	P_d	$\frac{1}{3}P_d$	$\frac{2}{3}P_d$	
$M_f = -\frac{1}{2}$	P_d	$\frac{2}{3}P_d$	$\frac{1}{3}P_d$	
Unpolarized	R_{ud}	R_{ud}	R_{ud}	
$M_f = \frac{1}{2}$	R_{ud}	R_{pd}	0.5	
$M_f = -\frac{1}{2}$	R_{ud}	0.5	R_{pd}	
Triplet	M_g undetected	$M_g = 1$	$M_g = 0$	$M_g = -1$
Unpolarized	P_t	$\frac{1}{3}P_t$	$\frac{1}{3}P_t$	$\frac{1}{3}P_t$
$M_f = 1$	P_t	$\frac{1}{2}P_t$	$\frac{1}{2}P_t$	0
$M_f = 0$	P_t	$\frac{1}{2}P_t$	0	$\frac{1}{2}P_t$
$M_f = -1$	P_t	0	$\frac{1}{2}P_t$	$\frac{1}{2}P_t$
Unpolarized	R_{ut}	R_{ut}	R_{ut}	R_{ut}
$M_f = 1$	R_{ut}	R_{pt}	0.5	0
$M_f = 0$	R_{ut}	0.5	0	0.5
$M_f = -1$	R_{ut}	0	0.5	R_{pt}

total number of two-photon decays to that of three-photon decays equals $\frac{\lambda_{eq}}{\lambda_o}$. For example, the rate of spin conversion on the oxygen molecule reported as $24.7 \pm 0.2/\mu$s in Ref. [14], is the exchange quenching rate λ_{eq}.

The ratio of three-photon fluxes to that of two-photon fluxes from the annihilation of positronium in the center of the Milky Way galaxy was reported to be 3.95 ± 0.32 [18], a value lower than the expected value of 4.5 obtained from spin-counting arguments [15]. As noted in the Introduction, one explanation for the lower measured value is the quenching of orthopositronium to parapositronium in the grains of the interstellar dust, prior to photon annihilation. Since we do not know (i) the cross section for the conversion of orthopositronium to parapositronium after electron exchange or (ii) the total cross section for the scattering of orthopositronium after electron exchange, we cannot calculate ratios of para- to orthopositronium production probabilities for doublet and triplet targets and incorporate them in Eqs. (23) and (24).

However, we derive an expression for R_γ, the ratio of the total number of photons from three-photon decays of orthopositronium integrated over all time, to that from two-photon decays of parapositronium integrated over all time. The parapositronium is produced through exchange quenching of orthopositronium. Given n positrons that form positronium in some medium, if orthopositronium is three times more likely to form than parapositronium, then $\frac{n}{4}$ positrons can form parapositronium and $\frac{3n}{4}$ can form orthopositronium. The number of two-photon decays after exchange quenching equals $\frac{n}{4} + \frac{3n}{4}(\frac{\lambda_{eq}}{\lambda_o + \lambda_{eq}})$, while the number of three-photon decays after exchange quenching equals $\frac{3n}{4} - \frac{3n}{4}(\frac{\lambda_{eq}}{\lambda_o + \lambda_{eq}})$. With each two-photon decay producing two photons, and each three-photon decay producing three photons,

$$R_\gamma = 4.5\left(\frac{\lambda_o}{\lambda_o + 4\lambda_{eq}}\right). \tag{25}$$

Note that when $\frac{\lambda_{eq}}{\lambda_o + \lambda_{eq}} = 0$, $R_\gamma = 4.5$, as expected in the absence of exchange quenching and when $\frac{\lambda_{eq}}{\lambda_o + \lambda_{eq}} = 1$, $R_\gamma = 0$, as expected in the absence of any outgoing orthopositronium. Furthermore, when triplet orthopositronium is three times more likely to be a final product compared to singlet parapositronium, then the ratio $\frac{\lambda_0}{\lambda_{eq}} = 3$, and $R_\gamma = \frac{13.5}{7} = 1.93$. Eq. (25) can be rewritten so that $\frac{\lambda_0}{\lambda_{eq}} = (\frac{4R_\gamma}{4.5 - R_\gamma})$, and $\frac{\lambda_0}{\lambda_{eq}}$ can be obtained from measured values of R_γ.

IV. SUMMARY

The quenching of orthopositronium by electron exchange is a fundamental process that can occur in laboratories on Earth as well as in the Milky Way galaxy. In one review of experimental measurements of positronium-gas cross sections, the importance of the exchange interaction at low energies and the need to measure partial cross sections was noted [28]. In another review, the production of intense positronium beams for resolving discrepancies in cross-section calculations and measurements at low energies was highlighted [29]. In the literature, spin analysis of exchange quenching is built on Ferrell's work in Ref. [19]. In Ref. [11], Kakimoto *et al.* use probability density arguments similar to those of Ref. [19] to estimate cross sections for nonconversion and conversion scattering of orthopositronium from oxygen. In Ref. [10], Blackwood *et al.* calculate probability densities in collisions between polarized orthopositronium beams and hydrogen targets. Spin-counting arguments are made by Zurek in Ref. [15] to calculate galactic-photon-flux ratios.

We have expanded the spin analysis in two ways. First, in order to include all possible initial and final spin states for polarized and unpolarized targets in a systematic way and reduce inconsistencies in the signs of exchange terms, we use angular-momentum coupling to calculate spin-scattering matrices. Second, in order to describe unpolarized beams and mixed-spin systems accurately, we construct density matrices. This enables us to obtain probabilities for cases in which the spins of the targets *are* or *are not* detected in coincidence with the production of positronium.

We have presented quenching probabilities and total probabilities for producing para- and orthopositronium in single collisions of an unpolarized orthopositronium beam with doublet and triplet targets. Our results are presented in terms of complex electron-spin amplitudes f^{S_e} which are labeled by the total electron spin S_e of the target-positronium system. The calculation of these amplitudes, which are not typical scattering lengths, involves the scattering of the electron in

the positronium with unpaired electrons in the targets. We will postpone such calculations to the future.

In Ref. [10], Blackwood *et al*. point out that (i) orthopositronium beams used in scattering experiments today are spin polarized and (ii) in collisions with unpolarized hydrogen targets in which the spins of the positronium and hydrogen are not detected, the ortho to para conversion cross sections are independent of the polarization of the orthopositronium beam. In Ref. [11], Kakimoto *et al*. give cross sections for the inelastic conversion of orthopositronium to parapositronium in positronium-oxygen scattering. This is important, since the lowest excited state of oxygen is only 0.98 eV above the ground state, and if the initial kinetic energy of the positronium beam is higher than that, both the inelastic-conversion process in which the oxygen is in the singlet state and the elastic-conversion process in which the oxygen is in the triplet state must be considered [13].

In this work, we have not considered excited states of the targets after the collisions or polarized orthopositronium beams. Our next project is to use the techniques described above to calculate quenching probabilities and total positronium production probabilities for excited states and polarized beams, and polarization vectors for outgoing beams.

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