GENERAL FORMS OF WAVE FUNCTIONS FOR DIPOSITRONIUM, Ps₂

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ABSTRACT

The consequences of particle interchange symmetry for the structure of wave functions of the states of dipositronium was recently discussed by the author [1]. In the present work, the methodology is simply explained, and the wave functions are explicitly given.

INTRODUCTION

Dipositronium and higher aggregates of positronium are the most symmetrical of all molecules. The symmetry operations that commute with the Hamiltonian include electron interchange and positron interchange as required by the Pauli exclusion principle, as well as electron-positron interchange, which is closely related to the concept of charge conjugation. Other commuting operators include those for spatial inversion and for spin and orbital angular momentum. The first complete and correct treatment of the influence of these operators on the structure of the wave functions was recently given by the author [1]. In that work, arguments for the methods used were given extremely succinctly owing to space limitations; here we explain the methods fully and clearly, and we give all the wave functions explicitly.

Charge conjugation means reversal of the charges for all particles. The Hamiltonian and the wave functions are invariant to this operation. For positronium, Ps, charge conjugation is equivalent to interchanging the electron and positron. In terms of the wave function, we affect this by inverting the spatial coordinates and exchanging the spin coordinates. The parities of these operations in terms of angular momentum quantum numbers are l and s + 1, respectively. The eigenvalue for electron-positron interchange, which we call "r," is thus \((-1)^{l+s+1}\). Electrons and positrons have opposite intrinsic parities [2], so their interchange gives another factor of \((-1)\). The charge conjugation parity \(C\) is thus \(-r\) for Ps in all its states.

That \((-1)^{l+s} = C\) for Ps is a geometrical coincidence – the positions of the two particles and their center of mass happen to be collinear. For Ps₂ the situation is quite different: charge conjugation is still equivalent to electron-positron interchange, but the latter is not expressed in the wave function by coordinate inversion. Thus, for Ps₂, we can no longer relate angular momentum quantum numbers to charge conjugation parity. Electron-positron interchange involves the interchange of both the electrons, each with either positron, thus introducing one more factor of \((-1)\); thus we assume that \(C = r\) for the states of Ps₂. Since photons have intrinsic charge conjugation parity \(-1\), \(C\) can also be written as \((-1)^n\), where \(n\) is the number of photons issued in the complete annihilation of the system. This is true for both Ps and Ps₂ in all their states.

GROUP THEORY AND THE SYMMETRY OF WAVE FUNCTIONS OF Ps₂

Kinghorn and Poshusta [3] were first to use abstract group theory to study the structure of Ps₂ wave functions. They realized that the wave functions of Ps₂ must transform as irreducible representations of some group. They observed that the two operators for the like-particle interchange, the two for the antiparticle interchange operators, and their combinations together with the identity operator constitute a group that is isomorphic with the point group \(D_{2d}\). They used this group to guide the construction of spatial factors of the wave functions while ignoring the role of spin in symmetry considerations. Some errors resulted from this procedure.
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$D_{2d}$ is a prime group. Since the three interchange operators are mutually independent and commuting, we believe that the group controlling the symmetry of wave functions is the direct product of three subgroups, those for electron interchange, positron interchange, and electron-positron interchange. Therefore the correct group is not prime. This is the idea that underlies the present work.

It should be understood that $Ps_2$, as $Ps$, is an atom, and as such it belongs to the full rotation point group as well.

Conventions of Group Theory and Wave Function Nomenclature

To proceed, we need to refine our concepts of points and particles. We consider a 4-dimensional space for each particle: three dimensions for ordinary space and one for spin. Spin space consists of two discrete points, which are variously called “$\pm 1/2$,” “$\uparrow$” and “$\downarrow$,” or “$\alpha$” and “$\beta$.” We consider four points placed arbitrarily in this 4D space, labeled $a$, $b$, $c$, and $d$. We label our particles 1, 2, 3, and 4. There are 4! distinct arrangements of our four particles on these four points. One such arrangement is: 1 on $a$, 2 on $b$, 3 on $c$, and 4 on $d$. We call this the “reference configuration.”

There are two distinct types of particle interchange operators, those that interchange two specified particles regardless of which points they are on [4], and those that interchange any particles that are on two specified points [5]. Each type gives rise to a group theory of its own “flavor.” The two flavors, or conventions, due to Bunker [4] and Wigner [5], respectively, are equally valid and useful, and differ only in bookkeeping details. Usually, one need not even be aware which convention one is working with, but in the present case the distinction is crucial, owing to traditional practices of wave function nomenclature.

3D wave functions are often denoted by Greek letters, sometimes with a subscript to indicate the state involved, for example, $\varphi_{1s}$. The particle of interest and the point in space where the function sits may be indicated by subscripts on the function’s argument, which is a vector in 3D space: $\varphi_{1s}(r_{1b})$. In contrast, the symbols $\alpha$ and $\beta$ commonly but incorrectly called “spin functions,” are in fact not functions but rather the names of points in 1D spin-space, as noted above. The complete 4D wave function for particle 2 in a $1s$-orbital centered on point $b$ with spin “up” is: $\varphi_{1s}(r_{1b})\alpha(2)$. It is important to note the mixed usage of the Greek letters: One defines a function in 3D space; the other, a point in 1D spin-space. When performing particle interchange operations in 4D space, one must be aware of this difference, else one will mix the two conventions of group theory, and errors will result.

In essence, we must devise interchange operators of mixed conventions in order to complement the mixed conventions of traditional wave function nomenclature. Properly done, this will give us operators that faithfully represent particle interchanges, and will provide us with a factorable group for the structure of wave functions of $Ps_2$.

Each of the four particles of $Ps_2$ can have either of two spins, so the total number of spin functions is $2^4 = 16$. The number of spin states must be the number that have $M = 0$, which is $\binom{4}{2} = 6$. The group we seek has order 8 since it is the direct product of three subgroups each of order 2.

Particle Interchange Operators

We find that the following definitions conform to the distinction between the two conventions of group theory, and yield a factorable group that governs the structure of wave functions for the states of $Ps_2$: We define interchange operators of the Bunker-type [4] ($\hat{P}_{12}$, $\hat{P}_{13}$, etc.) and of the Wigner-type [5] ($\hat{Q}_{ab}$, $\hat{Q}_{bd}$, etc.), and two operators that accommodate the nomenclature conventions for wave functions. For the interchange of identical particles $\mu$ and $\nu$,
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\[ \hat{R}_{\mu\nu} = P_{\mu\nu} Q_{[\mu[\nu]} \]  

(1)

for the interchange of antiparticles,

\[ \hat{S}_{\eta} = Q_{[\mu[\eta]} P_{\mu\eta} \times Q_{[\nu]} P_{\lambda\nu} \]  

(2)

Particle numbers inside square brackets denote the points occupied by the indicated particles in the reference configuration. The desired group is \( \{ E, \hat{R}_{\mu\nu} \} \otimes \{ E, \hat{R}_{\eta\eta} \} \otimes \{ E, \hat{S}_{\eta} \} \), which is isomorphic with \( D_{2h} \).

A 4D function of any specified symmetry can be projected from an unsymmetrical function by the application of:

\[ \hat{T}_{pqr} = (1 + r \hat{S}_{\eta})(1 + q \hat{R}_{\eta\eta})(1 + p \hat{R}_{\mu\nu}) \]  

(3)

The symmetry numbers \((p,q,r)\) designate the irreducible representations of \( D_{2h} \). For the states of \( Ps_2 \), \( p = q = -1 \), and \( r = C \).

The utility of the group \( D_{2h} \) is principally that it requires us to correctly craft the interchange operators. We have done this immediately above, and now we turn to the more pedestrian task of deducing resulting general forms for the wave functions. We can do this with only tangential reference to the underlying group theory.

**SPIN DEPENDENCE OF THE WAVE FUNCTIONS**

Molecular hydrogen has two pairs of particles, and the non-identical particles are not related by interchange operators. Consequently, wave functions for its states are factorable into 3D and spin-dependent parts. For \( Ps_2 \), however, the two types of particles are related by an interchange operator, and factorability of its wave functions may no longer be possible for all states. For states with factorable wave functions, each of the symmetry quantum numbers is likewise factorable: \( r = r^{(\text{spin},3D)} \), and similarly for \( p \) and \( q \). In the interest of clarity, we first seek factorable wave functions as we proceed to the spin coupling problem.

We must first consider two distinct schemes for spin coupling: one that first couples identical particles, then couples electron pairs with positron pairs; and another that first couples electron-positron pairs into \( Ps \)-like two-particle functions, then couples these. We denote the first scheme as \((e,e;p,p)\), and the second, \((e,p;e,p)\). We indicate spin functions of the latter scheme with tildes. In both schemes, 1 and 2 denote electrons, and 3 and 4, positrons.

Systems of two particles have four spin functions, a singlet and a set of triplets. In terms of the one-electron spin functions \( \alpha \) and \( \beta \), the triplet functions are:

\[ \sigma_{11}(1,2) = \alpha(1)\alpha(2) \]
\[ \sigma_{10}(1,2) = \frac{1}{\sqrt{2}}(\alpha(1)\beta(2) + \beta(1)\alpha(2)) \]
\[ \sigma_{01}(1,2) = \beta(1)\beta(2) \]  

(4)

and the singlet:

\[ \sigma_{00}(1,2) = \frac{1}{\sqrt{2}}(\alpha(1)\beta(2) - \beta(1)\alpha(2)) \]  

(5)

The subscripts are the spin quantum numbers \( s \) and \( m \). For positronium-like functions, the singlet function, for example, would be:

\[ \tilde{\sigma}_{00}(1,3) = \frac{1}{\sqrt{2}}(\alpha(1)\beta(3) - \beta(1)\alpha(3)) \]  

(6)
Hereafter, we suppress particle labels for simplicity, and rely on tildes or their absence to indicate antiparticle or particle pairs, respectively. Quantum numbers for the two-particle spin functions are given in Table 1.

Spin functions for $\text{Ps}_2$ in the $(e,e';p,p')$-scheme are now given. Functions in the $(e,p,e',p')$-scheme are gotten simply by inserting tildes over all spin functions. For $S=2$,

$$\Gamma_{22} = \sigma_{11}\sigma_{11}$$
$$\Gamma_{21} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{10} + \sigma_{10}\sigma_{11})$$
$$\Gamma_{20} = \frac{1}{\sqrt{6}} (\sigma_{10}\sigma_{1-1} + 2\sigma_{10}\sigma_{10} + \sigma_{1-1}\sigma_{11})$$
$$\Gamma_{2-1} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{1-1} + \sigma_{1-1}\sigma_{10})$$
$$\Gamma_{2-2} = \sigma_{1-1}\sigma_{1-1}$$

Four more wave functions can be expressed in terms of $\sigma_{1m}$. For $S=1$,

$$\Gamma_{11} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{10} - \sigma_{10}\sigma_{11})$$
$$\Gamma_{10} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{1-1} - \sigma_{1-1}\sigma_{11})$$
$$\Gamma_{1-1} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{1-1} - \sigma_{1-1}\sigma_{10})$$

and for $S=0$,

$$\Gamma_{00} = \frac{1}{\sqrt{3}} (\sigma_{10}\sigma_{10} - \sigma_{11}\sigma_{10} + \sigma_{1-1}\sigma_{11})$$

From $\sigma_{1m}$ and $\sigma_{00}$ we can write:

$$\Gamma_{11} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{00} + \sigma_{00}\sigma_{11})$$
$$\Gamma_{10} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{00} + \sigma_{00}\sigma_{10})$$
$$\Gamma_{1-1} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{00} + \sigma_{00}\sigma_{1-1})$$

and

$$\Gamma_{11} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{00} - \sigma_{00}\sigma_{11})$$
$$\Gamma_{10} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{00} - \sigma_{00}\sigma_{10})$$
$$\Gamma_{1-1} = \frac{1}{\sqrt{2}} (\sigma_{10}\sigma_{00} - \sigma_{00}\sigma_{1-1})$$

Finally:

$$\Gamma_{00} = \sigma_{00}$$

Platzman and Mills [6] used the $(e,p,e',p')$-scheme, and denoted their spin functions as $|S,C,P\rangle$, where "P" indicates "parity." The relationships between the spin functions as expressed in the two schemes are significant for us, and are displayed in Table 2, from which the quantum numbers for the states of $\text{Ps}_2$ can be easily deduced: From Table 2, the quantum numbers $p^{(p_{00})}$ and $q^{(p_{00})}$ are read from the left hand side, and $\mu^{(p_{00})}$ from either. $p = p^{(p_{00})}$, $p^{(3D)} = -1$ gives $p^{(3D)}$ and similarly for $q$. $\mu^{(3D)}$ is independent of the other symmetry numbers and apparently can be either $\pm 1$. The relationships $C = r = r^{(p_{00})} r^{(3D)} = (-1)^n$ yields $n$, the number of photons required for complete annihilation. Evidently $n$ can be any non-negative integer. Its value determines that of $r^{(3D)}$, or vice versa. These quantum numbers are collected in Table 3.

**3D DEPENDENCE OF THE WAVE FUNCTIONS**

Table 3 shows that, of the six spin functions, four have a complete set of the symmetry numbers $(p,q,r)$, and therefore qualify as the spin functions of factorable wave functions. For these four functions, general forms of the 3D factors can be written down at once. For the remaining two, the problem is more complicated.
Factorable Cases

A suitable 3D factor $\mathcal{F}_{pq}$ is gotten by projecting from an unsymmetrical function of the four 3D coordinates $f$ with the desired values of the symmetry numbers (see eq. (3)):

$$\mathcal{F}_{pq}(\vec{r}_1, \vec{r}_2, \vec{r}_3, \vec{r}_4) = \hat{T}_{pq}f(\vec{r}_1, \vec{r}_2, \vec{r}_3, \vec{r}_4)$$  \hfill (12)

This is the sum of eight terms, and it can be expressed as the product of the row matrix

$$[1 \ p \ q \ pq \ pr \ qr \ pqr]$$  \hfill (13)

and the column matrix

$$[f(1234) \ f(2134) \ f(1243) \ f(3142) \ f(3412) \ f(4312) \ f(4321)]^T$$  \hfill (14)

For simplicity, we omit all characters in the arguments of $f$ except particle numbers. We further simplify by omitting the column matrix altogether hereafter since it is the same for all states, and designate each wave function by only its row matrix (13). This we can write succinctly by indicating only signs. For example, for the 3D partner of the spin function $\Gamma_{20}$ (see Table 3), $\mathcal{F}_{--r} \equiv \mathcal{F}_{--c}$, is indicated as:

$$[+-- \ r --r --r] \equiv [+-- \ C --C --C C]$$  \hfill (15)

The four factorable wave functions are:

$$\psi_{AC}^{(1)} = \mathcal{F}_{--c} \Gamma_{2M}$$
$$\psi_{AC}^{(2)} = \mathcal{F}_{--c} \Gamma_{1M}$$
$$\psi_{AC}^{(3)} = \mathcal{F}_{--c} \Gamma_{00}$$
$$\psi_{AC}^{(4)} = \mathcal{F}_{++c} \Gamma_{00}$$  \hfill (16)

The angular momentum quantum numbers, and superscripts, the states. It is clear that the first three states are degenerate except for small spin-spin and spin-orbit interactions, which is reminiscent of the states of $\text{Ps}$.

Non-factorable Cases

The fourth function in Table 3, $\Gamma_{10}^s \equiv \Gamma_{10}^s$, and the fifth, $\Gamma_{10}^u \equiv \Gamma_{10}^u$, are clearly not candidates for being parts of factorable wave functions, for the like-particle interchange operators interchange the functions as well as the particles; e.g.,

$$\hat{R}_{12} \Gamma_{10}^s = \Gamma_{10}^s$$  \hfill (17)

We resort to a simple, direct tactic: we project from some unsymmetrical 4D function, say $f(1234)\sigma_{00} \sigma_{00}$, using the operator in eq. (3) with $p = q = -1$ and $r = C$. We arrive at a suitable expression for the wave function:

$$[+-- \ 0000]\sigma_{10} \sigma_{00} + [0000 \ C(+-+-)]\sigma_{00} \sigma_{10}$$  \hfill (18)

We could just as well project from $f(1234)\sigma_{00} \sigma_{10}$, giving:

$$[+-- \ 0000]\sigma_{00} \sigma_{10} + [0000 \ C(+-+-)]\sigma_{10} \sigma_{00}$$  \hfill (19)

These two functions are clearly independent and equally valid. We take a linear combination of them:

$$[a(+-+-) \ bC(+-+-)]\sigma_{10} \sigma_{00} + [b(+-+-) \ aC(+-+-)]\sigma_{00} \sigma_{10}$$  \hfill (20)

where $a$ and $b$ are disposable. We subject them to the constraint $|a|^2 + |b|^2 = 1$ to preserve normalization. Perhaps these parameters will be determined by energy minimization, or to avoid a contradiction for a given value of $C$. In any case, we denote the expression (20) as $\psi_{AC}^{(5,6)}(a_C, b_C)$, and write the last two wave functions as

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\[ \Psi_{\lambda^+}^{(5)}(a_+,b_+) \]
\[ \Psi_{\lambda^-}^{(6)}(a_-,b_-) \]

We see that the 3D and spin factors of these wave functions do not conform to any irreducible representation of the \( D_{2h} \) group, but that the overall functions do.

CONCLUSION

In this note we give general forms of the wave functions of \( P_{52} \). Still to be considered are: orbital angular momentum, annihilation rates, asymptotic forms, and computational strategies.

REFERENCES

Table 1. Quantum numbers for two-particle spin functions; identical particles on top, antiparticles on the bottom.

<table>
<thead>
<tr>
<th>Spin functions</th>
<th>$p^{(\text{spin})}$</th>
<th>$p^{(\text{3D})}$</th>
<th>Intrinsic parity</th>
<th>$j^{(\text{spin})}$</th>
<th>$j^{(\text{3D})}$</th>
<th>$C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{00}$</td>
<td>-1</td>
<td>1</td>
<td>1</td>
<td>1</td>
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<td></td>
</tr>
<tr>
<td>$\sigma_{1m}$</td>
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<td>-1</td>
<td>1</td>
<td>1</td>
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<td></td>
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<tr>
<td>$\bar{\sigma}_{00}$</td>
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<td>-1</td>
<td>(-1)$^y$</td>
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<tr>
<td>$\bar{\sigma}_{1m}$</td>
<td>-1</td>
<td>1</td>
<td>(-1)$^y$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 2. Spin functions of the present work (left side) and the corresponding functions of Platzman and Mills [6] (right side, $|SCP)$; these authors give expressions only for the two $|0++\rangle$ states). For clarity, only functions with $M = 0$ are shown.

<table>
<thead>
<tr>
<th>1,2,3,4 = $e,e'p,p'$</th>
<th>1,2,3,4 = $e,p,e'p'$</th>
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</thead>
<tbody>
<tr>
<td>$\Gamma_0 = \frac{1}{\sqrt{6}}(\sigma_1 \sigma_{1-1} + 2 \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
<td>$\Gamma_0 = \frac{1}{\sqrt{6}}(\sigma_1 \sigma_{1-1} - 2 \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
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<tr>
<td>$\Gamma_{10} = \frac{1}{\sqrt{2}}(\sigma_1 \sigma_{1-1} - \sigma_{1-1} \sigma_{11})$</td>
<td>$\Gamma_{10} = \frac{1}{\sqrt{2}}(\sigma_1 \sigma_{1-1} - \sigma_{1-1} \sigma_{11})$</td>
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<tr>
<td>$\Gamma_{00} = \frac{1}{\sqrt{2}}(\sigma_1 \sigma_{1-1} - \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
<td>$\Gamma_{00} = \frac{1}{\sqrt{2}}(\sigma_1 \sigma_{1-1} - \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
</tr>
<tr>
<td>$\Gamma'<em>{10} = \sigma</em>{00} \sigma_{00}$</td>
<td>$\Gamma'<em>{10} = \sigma</em>{00} \sigma_{00}$</td>
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<tr>
<td>$\Gamma_0' = \frac{1}{\sqrt{3}}(\sigma_1 \sigma_{1-1} + 2 \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
<td>$\Gamma_0' = \frac{1}{\sqrt{3}}(\sigma_1 \sigma_{1-1} + 2 \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
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<td>$\Gamma_{10}' = \frac{1}{\sqrt{2}}(\sigma_1 \sigma_{1-1} - \sigma_{10} \sigma_{10} + \sigma_{1-1} \sigma_{11})$</td>
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<tr>
<td>$\Gamma_{00}' = \sigma_{00} \sigma_{00}$</td>
<td>$\Gamma_{00}' = \sigma_{00} \sigma_{00}$</td>
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</table>
Table 3. Quantum numbers associated with the spin functions of $P_{S_2}$.

<table>
<thead>
<tr>
<th>Spin functions</th>
<th>$p^{(\text{spin})}$</th>
<th>$q^{(\text{spin})}$</th>
<th>$r^{(\text{spin})}$</th>
<th>$p^{(\text{3D})}$</th>
<th>$q^{(\text{3D})}$</th>
<th>$r^{(\text{3D})}$</th>
<th>Intrinsic parity</th>
<th>$C$</th>
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<td>$\pm 1$</td>
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<td>$\pm 1$</td>
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<tr>
<td>$\Gamma'<em>{10}, \tilde{\Gamma}'</em>{10}$</td>
<td>$\Gamma_{10}', \tilde{\Gamma}<em>{10}' - \Gamma</em>{10}'', \tilde{\Gamma}_{10}''$</td>
<td>-1</td>
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