Excited States of the Positronium Molecule

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Despite the early theoretical prediction of its existence, the Ps_2 molecule has not been experimentally found to date. The difficulty stems from the fact that this system is neutral and therefore it cannot be separated from the positronium atoms (Ps) and its primary decay mode, the annihilation by two-photon emission, is exactly the same as that of the Ps atom. The energy of the photons arising from the annihilation is different in principle: The photons carry 1.02 MeV energy due to the annihilation plus the binding energy of the corresponding system. The binding-energy difference is, however, less than 1 eV and adding it to 1.02 MeV, the energy of the photons coming from the Ps atom or Ps_2 molecule cannot be experimentally distinguished. The experimental observation of the biexcitons can be considered as an indirect indication of the existence of Ps_2.

In our recent study [1, 2] we have predicted the existence of a bound excited-state of the Ps_2 molecule, and investigated its possible decay modes with a special emphasis on the electric dipole ($E1$) transition to the ground state. It was shown that the probability of the $E1$ transition is comparable to that of the annihilation. The unique energy of this transition may possibly be utilized as a sign for the experimental identification of Ps_2.

Since the Hamiltonian of the Ps_2 is invariant under eight permutations which are isomorphic to those of the point group D_{2d}, the eigenstates of Ps_2 can be classified according to the representations μ of the D_{2d} group, $\mu = A_1, A_2, B_1, B_2, E_{11}, E_{12}, E_{21}, E_{22}$. In order for an excited state to be bound against autodissociation, its symmetry has to be different from that of any configuration which can be reached by the Ps-Ps channel lying lower in energy than the state. Two excited states with $L^\pi = 0^+$, found in [3], have symmetries of B_2 and E which are different from that of the ground state (A_1).

To search for all possible bound states of the Ps_2, we have performed a stochastic variational method [4] calculation for states with $L^\pi = 0^\pm, 1^\pm$, and $2^\pm$. The trial function is given by combining correlated Gaussian basis with the angular functions. The basis function is written by

$$\psi = \begin{cases} |\mathbf{v}|^{2K+L} \exp\{-\frac{1}{2}\mathbf{x}A\mathbf{x}\} Y_{LM}(\hat{\mathbf{v}}) & \text{for natural parity state,} \\ |\mathbf{v}_1|^{2K+L} |\mathbf{v}_2| \exp\{-\frac{1}{2}\mathbf{x}A\mathbf{x}\} \left[Y_L(\hat{\mathbf{v}}_1) Y_1(\hat{\mathbf{v}}_2)\right]_{LM} & \\ \qquad \text{for unnatural parity state with } L \neq 0, & \\ (\mathbf{x}_1 \times \mathbf{x}_2) \cdot \mathbf{x}_3 \exp\{-\frac{1}{2}\mathbf{x}A\mathbf{x}\} & \text{for unnatural parity state with } L = 0, \end{cases} \tag{1}$$

where we use relative coordinates $\mathbf{x} = \{\mathbf{x}_1, \mathbf{x}_2, \mathbf{x}_3\}$, A is an $(N-1)\times(N-1)$ positive-definite, symmetric matrix and $\mathbf{v}$ called 'global vector' is defined by $\mathbf{v} = \sum_{i=1}^{N-1} u_i \mathbf{x}_i$. A_{ij} and u_i are dealt with as variational parameters. This approach greatly simplifies the calculations for non-spherical systems by replacing the partial wave expansion with a much simpler representation of the angular motion.

The results are shown in Table 1 that the lowest accessible thresholds and the energies of the various states of Ps_2 are listed in. If the energy of Ps_2 is lower than the lowest accessible threshold, the corresponding state is stable. Therefore, We concluded that only four states predicted already to be stable, $0^+(A_1)$, $0^+(B_2)$, $0^+(E)$ and $1^-(B_2)$, are bound. Figure 1 plots the correlation function of two-particle, defined by $C(\mathbf{r}) = \langle\Psi|\delta(\mathbf{r}_i - \mathbf{r}_j - \mathbf{r})|\Psi\rangle r^2$, for three

excited states. e^--e^+ correlation functions have a peak and a shoulder reflecting the fact that the basic structure is a weakly coupled system of Ps atom in $1S$ state and another Ps atom in $2S$ or $2P$ spatially extended state. Particularly, $0^+(B_2)$ state that is bound slightly against the corresponding threshold clusterize obviously to two Ps atoms.

Table 1: The lowest accessible channel and energy for various states of the Ps_2. Atomic units is used.

	L^π	accessible channel	$-E(Ps_2)$	L^π	accessible channel	$-E(Ps_2)$
A_1	0^+	Ps($1S$)+Ps($1S$)	0.5160037	0^-	Ps($2P$)+Ps($3P$)	0.09
A_2		Ps($1S$)+Ps($1P$)	0.309		Ps($2P$)+Ps($3P$)	0.09
B_1		Ps($1S$)+Ps($1S$)	0.492		Ps($2P$)+Ps($3P$)	0.09
B_2		Ps($1S$)+Ps($2P$)	0.31449		Ps($2P$)+Ps($3P$)	0.09
E		Ps($1S$)+Ps($2S, P$)	0.33016		Ps($2P$)+Ps($3P$)	0.09
A_1	1^-	Ps($1S$)+Ps($2S$)	0.3114	1^+	Ps($1S$)+Ps($3D$)	0.267
A_2		Ps($1S$)+Ps($2P$)	0.3114		Ps($1S$)+Ps($2P$)	0.309
B_1		Ps($1S$)+Ps($2S$)	0.3110		Ps($1S$)+Ps($3D$)	0.267
B_2		Ps($1S$)+Ps($2P$)	0.34408		Ps($1S$)+Ps($2P$)	0.309
E		Ps($1S$)+Ps($1S$)	0.496		Ps($1S$)+Ps($2P$)	0.312
A_1	2^+	Ps($1S$)+Ps($1S$)	0.490	2^-	Ps($1S$)+Ps($3D$)	0.276
A_2		Ps($1S$)+Ps($2P$)	0.308		Ps($1S$)+Ps($2P$)	0.310
B_1		Ps($1S$)+Ps($1S$)	0.492		Ps($1S$)+Ps($3D$)	0.276
B_2		Ps($1S$)+Ps($2P$)	0.310		Ps($1S$)+Ps($2P$)	0.301
E		Ps($1S$)+Ps($2S, P$)	0.310		Ps($1S$)+Ps($2P$)	0.310

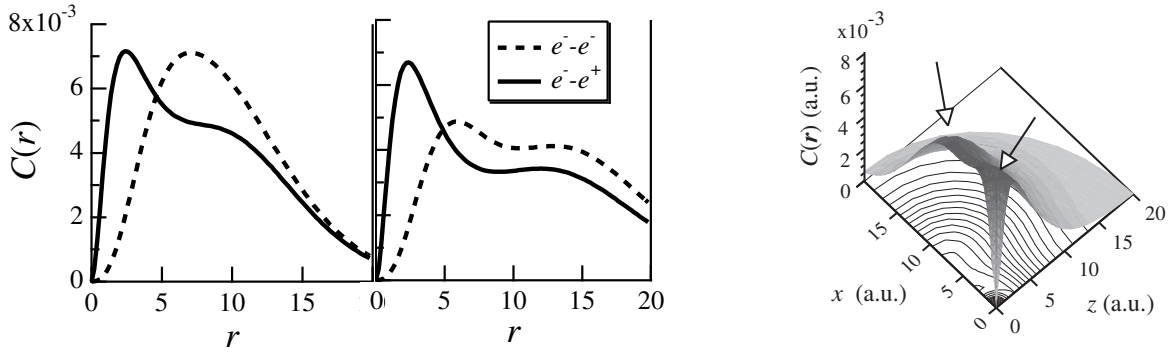

Figure 1: The correlation function of two-particle for the excited states. The left figure is for $0^+(E)$ state, the center is for $0^+(B_2)$ and the right is for e^--e^+ correlation of $1^-(B_2)$ with M=1. The correlation function sliced on xy plane is drawn in the figure for $1^-(B_2)$ state since the correlated function is axially symmetric around z axis. Atomic units are used.

References

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