Instability of tripositronium

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(Received 29 March 2013; published 13 May 2013)

The stability of tripositronium, a system consisting of three electrons and three positrons, has been investigated systematically by varying the repulsion strength between like-charged particles. The possibility of the existence of a Ps_3 bound state that is stable against dissociation appears utterly unlikely based on the results of variational calculations employing all-particle explicitly correlated Gaussian basis functions.

DOI: 10.1103/PhysRevA.87.054501 PACS number(s): 31.15.ac, 31.15.xt, 36.10.Dr

I. INTRODUCTION

Since the early work of Wheeler [1] on quantum mechanical entities composed of electrons and positrons (which he called *polyelectrons*) there has been a significant interest in finding particle-antiparticle systems stable against dissociation. This interest has taken place on both theoretical and experimental sides. While Mohorovičić [2], Ruark [3], and Wheeler [1] envisaged the existence of the positronium atom (Ps), Deutsch [4] provided experimental evidence for it in 1951. In his work Wheeler also predicted the stability of a larger, three-particle system $e^+e^-e^-$ (Ps^- or positronium negative ion). The first experimental observation of Ps^- was made three decades later by Mills [5], who also measured its annihilation decay rate [6]. Soon after the work of Wheeler, Hylleraas and Ore [7] showed rigorously, using the variational method, that even a larger four-particle Ps_2, which is a matter-antimatter analog of the hydrogen molecule where positrons play the role of "nuclei," is also stable. The experimental confirmation of its existence was made just recently when Cassidy and Mills performed experiments with intense positron bursts implanted into a thin film of porous silica [8,9]. This discovery has created a new chapter in the experimental positronium physics and chemistry and invigorated new theoretical studies. Further advances in positron technology may permit much higher density of positrons and raise questions of whether larger polyelectrons can exist in nature [10]. Eventually, one might reach the point where a macroscopic number of Ps atoms can undergo a phase transition to form a Bose-Einstein condensate. This quantum matter-antimatter system would be of great interest for many reasons, in particular for possible production of an annihilation γ-ray laser [8,11]. Because of the very short wavelength such a laser could be used to probe objects as small as atomic nuclei.

Theoretical discovery and investigation of new matter-antimatter systems remains a challenging task [12,13]. This particularly concerns systems composed of a few electrons and positrons. There are several reasons for this. First, these systems (if they exist) are expected to be weakly bound. This is illustrated by the fact that the binding energies of both Ps^- and Ps_2 are only 5% and 3% of their total energies. Second, there is no separation into heavy "nuclei" and light electrons; thus all particles must be treated equally. Finally, traditional quantum chemical ansatzes based on expansions in terms of single-particle atomic orbitals are known to converge extremely slowly for systems containing positrons due to strong attraction between oppositely charged particles and formation of clusters [14–17]. In order to be able to describe such systems accurately it is necessary to approximate the wave function with an expansion that depends explicitly on all interparticle coordinates.

In this work we have performed a systematic study of the stability of Ps_3 ($e^+e^+e^+e^-e^-e^-$ or positronium trimer). In the framework of the variational method complemented with the use of explicitly correlated Gaussian (ECG) basis functions we demonstrated that this system is unlikely to have bound states. While there has been previous work [18] where ECG-type calculations of Ps_3 were attempted, that work did not provide a systematic study of the stability. Due to the weak binding and complex structure of Ps_3 (if it exists) the convergence of the trial wave function in the stochastic variational method (SVM) might have lead to a wrong configuration (a dissociated system) corresponding to a local minimum of the total energy. Capturing the right configuration using a rough initial guess for the nonlinear parameters of ECGs is generally not guaranteed when the system is very close to dissociation. In this work we remedied this issue by slowly varying the Coulomb repulsion strength and maintaining the system in a bound state. As our calculations have shown, in the limit of the exact interaction strength Ps_3 becomes dissociated.

II. METHOD

It has been demonstrated in a number of applications [18–21] that the variational method in conjunction with the use of ECG basis functions provides a powerful framework for studying bound states of Coulomb few-body systems with very high accuracy. In fact, many reliable predictions of the stability of quantum systems containing positrons were first made using ECG expansions [22–25].

In the present work we are concerned with computing the lowest S-state of a six-particle system with unit charges. For definiteness, let us assume that particles 1, 2, and 3 are positrons while particles 4, 5, and 6 are electrons. The ECG basis functions suitable for calculations of zero total orbital angular momentum states have the following form:

$$\phi_k = \exp\left(-\alpha_{12}^k R_{12}^2 - \alpha_{13}^k R_{13}^2 - \cdots - \alpha_{N-1,N}^k R_{N-1,N}^2\right), \quad (1)$$

where $R_{ij} = |\mathbf{R}_i - \mathbf{R}_j|$ are interparticle distances and α_{ij}^k are nonlinear parameters. Superscript k in α_{ij}^k reflects the fact that the nonlinear parameters are unique for each basis function. N is the total number of particles (six in our case).

ECG basis functions explicitly include all possible interparticle distances, and each distance comes with its own parameter. These nonlinear parameters can be tuned in order to achieve a compact and accurate representation of the wave function of the system. It is this flexibility due to a large number of parameters that gives the ECG basis the ability to describe very accurately the correlated motion of particles in essentially arbitrary few-body systems.

Functions (1) are both rotationally and translationally invariant. Effectively, they only describe the intrinsic motion of the particles. The motion of the system as a whole is eliminated from consideration. More details describing technical aspects of calculations with ECG basis functions can be found in Refs. [18–21] and references therein.

Polyelectrons, in particular those consisting of the same number of electrons and positrons, possess very high permutational symmetry. The Hamiltonian of a Ps_n system is invariant not only upon permutations of electrons or positrons but also under charge conjugation (i.e., when all electrons are replaced with positrons and vice versa). A detailed analysis of the symmetry of Ps_2 is presented in Ref. [26]. In our calculations we do not explicitly deal with spin-components of the trial wave function. Instead, we use Young projection operators for both sets of identical particles, electrons and positrons [27]. Imposing the charge conjugation symmetry requires an additional factor in the symmetry projector, $1 \pm P_{14}P_{25}P_{36}$ (here P_{ij} is a permutation of spatial coordinates of particles i and j). For the ground-state calculations with zero angular momentum, we chose the plus sign as such a configuration is more energetically favorable. Thus, the total symmetry projector applied to the basis functions is

$$(1 \pm P_{14}P_{25}P_{36})(1 - P_{46})(1 - P_{13})(1 + P_{45})(1 + P_{12}). \quad (2)$$

A common way to study the stability of weakly bound Coulomb few-body systems is to increase or decrease masses or charges of particles so that the system under study becomes relatively strongly bound. Then, assuming that the energy of that bound state is a continuous function of the masses and charges, they are changed slowly to approach their actual values. At the same time the wave function of the system is evolved gradually by adjusting the shape of the basis functions and increasing, if necessary, the length of the variational expansion. This provides a practical possibility to maintain the boundedness of the trial wave function as long as the Hamiltonian allows for a bound state. Variational calculations with ECGs are particularly suitable in such kind of calculations as they allow very efficient optimization of the basis at each step.

Because of the symmetry of the Ps_3 system, scaling of all charges and (or) masses simultaneously does not change the degree of boundedness. Effectively, it only redefines the units of distance and energy. On the other hand, increasing or decreasing the charge or mass of only one sort of particles (i.e., either positrons or electrons) does not leave the Hamiltonian invariant with respect to charge conjugation. Therefore, in order to preserve the symmetry properties of the system and at the same time be able to vary the boundedness, we split the potential energy operator into two parts, the attractive interaction and the repulsive interaction. The Hamiltonian then looks as follows (atomic units are assumed throughout):

$$H = -\frac{1}{2}\sum_{i=1}^{N} \nabla_{\mathbf{R}_i}^2 + \beta V_{\text{attr}} + \gamma V_{\text{rep}}, \quad (3)$$

where

$$V_{\text{attr}} = -\frac{1}{R_{14}} - \frac{1}{R_{15}} - \frac{1}{R_{16}} - \frac{1}{R_{24}} - \frac{1}{R_{25}} - \frac{1}{R_{29}} - \frac{1}{R_{34}} - \frac{1}{R_{35}} - \frac{1}{R_{36}}, \quad (4)$$

$$V_{\text{rep}} = \frac{1}{R_{12}} + \frac{1}{R_{13}} + \frac{1}{R_{23}} + \frac{1}{R_{45}} + \frac{1}{R_{46}} + \frac{1}{R_{56}}. \quad (5)$$

Parameters β and γ define the strength of attraction and repulsion between particles. In the real Ps_3 system, they both are equal to unity. In our calculations we vary γ, starting with a value $\gamma < 1$ that corresponds to a well-bound state and increasing it gradually up to $\gamma = 1$. Parameter β is kept equal to unity at all times. The bound state of the system could certainly be maintained in the opposite way, i.e., by keeping $\gamma = 1$ and $\beta > 1$.

III. RESULTS

Table I shows the binding energies of both Ps_2 and Ps_3 as functions of the repulsion strength. The calculations for the smaller system, Ps_2, are converged to beyond the seventh significant figure regardless of the value of γ when only 500 basis functions are used. Calculations of Ps_3 are considerably more time-consuming due to higher number of degrees of freedom in that system. In addition, they require the use of larger basis sets. We increased the size of the basis gradually from 1000 for a well-bound state with $\gamma = 0.8$ to 2000 in the case of $\gamma = 1.0$ in order to provide sufficient accuracy for higher values of γ and make sure that the relative error in the binding energy remains small enough. As one can see from the trends of both the total and binding energies in Table I, Ps_2 remains a relatively well-bound system for any γ, while the energy of Ps_3 approaches the dissociation threshold when $\gamma \to 1$. The dissociation of Ps_3 in the limit $\gamma = 1$ is seen even better if we look at the trends of the expectation values $\langle r_{++}\rangle$ and $\langle r_{+-}\rangle$, which stand for the average distance between like particles and oppositely charge particles respectively. One can clearly see that the system gets broken into two subsystems upon γ approaching unity. In fact, recalling that this six-particle Ps_3 system has 15 interparticle distances total (6 corresponding to the like particles and 9 to the oppositely charge particles) it is easy to deduce that the ratio of $\langle r_{++}\rangle/\langle r_{+-}\rangle \approx 1.5$ in the limit of two separated fragments corresponds to the dissociation channel $Ps_3 \to Ps_2 +$ Ps.

Assuming that the $Ps_3 \to Ps_2 +$ Ps dissociation channel indeed takes place, it should be possible to estimate the binding energy as a function of γ with a quick calculation. In this case we effectively have a two-body system with masses 4 and 2 (in atomic units), which interact via Coulomb potential $V(R) = 4(\beta - \gamma)/R$, where R is the separation

TABLE I. The total and binding energies and average interparticle distances of Ps_2 and Ps_3 molecules as a function of the repulsion strength parameter γ. All quantities are in a.u. The values in parentheses are estimates of the remaining uncertainty due to finite size of the basis used in calculations.

	Ps_2					Ps_3				
γ	Basis	E_{tot}	E_{bind}	$\langle r_{++}\rangle$	$\langle r_{+-}\rangle$	Basis	E_{tot}	E_{bind}	$\langle r_{++}\rangle$	$\langle r_{+-}\rangle$
0.8	500	−0.6221182(0)	0.1221182(0)	4.264(0)	3.478(0)	1000	−0.9731482(50)	0.1010299(50)	6.014(0)	4.906(0)
0.9	500	−0.5651750(0)	0.0651750(0)	4.831(0)	3.823(0)	1000	−0.8537477(50)	0.0385727(50)	7.644(2)	6.041(2)
0.95	500	−0.5394609(0)	0.0394609(0)	5.267(0)	4.073(0)	1000	−0.8040834(50)	0.0146224(50)	9.667(10)	7.412(10)
0.98	500	−0.5250743(0)	0.0250743(0)	5.654(0)	4.285(0)	1000	−0.7789411(50)	0.0038668(50)	13.61(10)	10.06(10)
0.985	500	−0.5227641(0)	0.0227641(0)	5.736(0)	4.329(0)	1000	−0.7752591(50)	0.0024951(50)	15.41(10)	11.26(10)
0.99	500	−0.5204813(0)	0.0204813(0)	5.826(0)	4.377(0)	1000	−0.7717932(50)	0.0013119(50)	18.76(10)	13.49(10)
0.993	500	−0.5191254(0)	0.0191254(0)	5.883(0)	4.408(0)	1000	−0.7698473(30)	0.0007219(30)	22.93(20)	16.27(20)
0.995	500	−0.5182274(0)	0.0182274(0)	5.924(0)	4.429(0)	1500	−0.7686265(30)	0.0003991(30)	28.68(50)	20.11(50)
0.997	500	−0.5173342(0)	0.0173342(0)	5.966(0)	4.452(0)	1500	−0.7674843(20)	0.0001501(20)	43.10(1.)	29.72(1.)
0.998	500	−0.5168895(0)	0.0168895(0)	5.988(0)	4.463(0)	1500	−0.7669540(10)	0.0000645(10)	63.19(5.)	43.11(5.)
0.999	500	−0.5164460(0)	0.0164460(0)	6.010(0)	4.475(0)	1500	−0.7664593(8)	0.0000133(8)	125.2(10.)	84.45(10.)
0.9995	500	−0.5162247(0)	0.0162247(0)	6.022(0)	4.481(0)	1500	−0.7662264(6)	0.0000017(6)	207.1(20.)	139.0(20.)
1.0	500	−0.5160038(0)	0.0160038(0)	6.033(0)	4.487(0)	2000	−0.7660021(15)	Unbound	487.2(∞)	325.8(∞)

between two fragments, Ps_2 and Ps. The binding energy is then $E_{bind} \sim \frac{32}{3}(\beta-\gamma)^2$. For $\gamma = 0.999$ and $\gamma = 0.995$ this gives 0.0000107 a.u. and 0.0000027 a.u. respectively, which is in good agreement with the calculated binding energies of 0.0000133 a.u. and 0.0000017 a.u. listed in Table I.

While the results shown in Table I cannot be be considered a rigorous mathematical proof of instability of Ps_3, they provide solid evidence that this system has no bound state with $L = 0$.

One could also think about the possibility of states with $L > 0$. Dipositronium, Ps_2, is known to have a stable excited state with $L = 1$ [23,28]. However, the existence of that state is stipulated by a reduced dissociation threshold. Ps_2 ($L = 1$) cannot decay into two Ps atoms in the ground state due to the parity conservation. The situation is different in Ps_3, however, because this system dissociates into two nonidentical fragments. Nevertheless, we have performed Ps_3 ($L = 1$) calculations for some values of γ and did not observe an indication that there could be a bound state.

We have also investigated the possibility of resonance states around the Ps_2+ Ps threshold using the complex scaling method [21]. No narrow resonances were found in the spectrum. This concerns both the case of pure Coulomb interaction ($\beta,\gamma = 1$) and the case when the repulsion strength is weakened ($\beta = 1$, $\gamma < 1$).

In summary, we have performed a systematic study of the stability of tripositronium by varying the interaction strength between particles. The results of our variational calculations, in which we expanded the wave function of a six-particle Ps_3 system in terms of explicitly correlated Gaussian basis functions, indicate that tripositronium is not stable against dissociation.

ACKNOWLEDGMENT

S.B. and O.V.P acknowledge financial support from the National Science Foundation, Grant CHE-1300118.

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