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Article

Quantum Entanglement and Shannon Information Entropy for the Doubly Excited Resonance State in Positronium Negative Ion

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Abstract: In the present work, we report an investigation on quantum entanglement in the doubly excited $2s^2\,{}^1S^e$ resonance state of the positronium negative ion by using highly correlated Hylleraas type wave functions, determined by calculation of the density of resonance states with the stabilization method. Once the resonance wave function is obtained, the spatial (electron-electron orbital) entanglement entropies (von Neumann and linear) can be quantified using the Schmidt decomposition method. Furthermore, Shannon entropy in position space, a measure for localization (or delocalization) for such a doubly excited state, is also calculated.

Keywords: quantum entanglement; von Neumann entropy; linear entropy; Shannon information entropy; position space; positronium negative ion; Hylleraas functions; Schmidt-Slater decomposition method

1. Introduction

Considerable research activities have been carried out on investigations of quantum entanglement in model, artificial, and natural atoms [1–8]. Quantum entanglement plays an important role in related topics such as quantum information, quantum computation, quantum teleportation, and quantum cryptography [9]. Recently, our group has also carried out quantification of entanglement entropies in model and natural atoms [10–19]. For model systems, we have calculated the von Neumann entropy and linear entropy in the ground and excited states of two spinless bosons in 1D harmonic traps and with two-body delta interactions [10]. For the N-particle Moshinsky model, we have investigated the

entropic uncertainty principle between a subgroup with an arbitrary p particles and the other subgroup of $N-p$ particles, and have also investigated the classical and quantum correlations for such a bipartite system [11]. For natural atoms, our recent works include studies on spatial (electron-electron orbital) entanglement in highly correlated two-electron systems such as the helium atom, the hydrogen and positronium negative ions [12–16]. The von Neumann entropy and linear entropy were quantified by calculating the eigenvalues of the one-particle reduced density matrix for such bi-particle systems. We have employed configuration interaction (CI) with B-spline basis to construct wave functions for the ground and excited states of the two spin-$1/2$ fermions (electrons) in the helium iso-electronic sequence [12,13]. By using highly-corrected Hylleraas-type wave functions to represent the ground and singly-excited states of such systems, we have established benchmark values for von Neumann and linear entropies in the two-electron atom/ions [14–16]. Furthermore, quantum entanglements for doubly-excited (resonance) states [17], and for shape resonances in two-electron ions when the nuclear charge is decreased below the critical value as the system becoming unbound, were also investigated [18]. Our ongoing works include quantification of entanglement entropies for atoms embedded in Debye plasmas in which the Coulomb potentials for two-body interactions are replaced by screened Coulomb (Yukawa-type) potentials [19]. In the present work, we report quantification of entropies (von Neumann, linear, and Shannon information) for the doubly excited $2s^2\,{}^1S^e$ state in the positronium negative ion. Studies of quantum entanglement on resonance states in natural atoms [17,18,20] and in artificial atoms like quantum dots were recently reported in the literature [21].

In the field of quantum information, the measurement of a pure state bipartite system is usually by quantifying von Neumann entropy S_{vN}, or linear entropy S_L, the approximation of von Neumann entropy. The definition of the von Neumann entropy is:

$$S_{vN} = -Tr\left(\rho_{red}\ \log_2 \rho_{red}\right) \tag{1}$$

where ρ_{red} is the one-particle reduced density matrix after tracing out all the degrees of freedom of one of the particles, and Tr has the usual meaning of the trace of a matrix, and the linear entropy is the first order term of the expansion of the log of Equation (1). The linear entropy is defined as

$$S_L = Tr(\rho_{red}(1-\rho_{red})) = 1 - Tr\left(\rho_{red}^2\right) \tag{2}$$

The positronium negative ion, Ps^-, is a three-lepton system consists of two electrons and a positron, and interacting with Coulomb potential. Ever since the early work of Wheeler [22] who showed that this three-body system forms a bound state, there have been continuous interest in theoretical studies of various properties for such ion [23–27], and the activities have been highlighted in several reviews [28–30]. On experimental side, Mills first reported observation of this positronic system [31], and other experimental activities were summarized in a recent review [32]. The $2s^2\,{}^1S^e$ resonance in Ps^- was first calculated in 1979 [33], and since then there have been considerable research works on such state reported in the literature [28–30,34,35]. The doubly excited state in Ps^- is also of recent experimental interest [36]. In the present work, we first use the stabilization method and employ Hylleraas basis wave functions to determine the resonance energy and width, as well as the wave function for this resonance state. Once the wave function is obtained, we use Schmidt decomposition method to calculate von Neumann and linear entropies to probe the quantum correlation effects. The resonance wave function is

also used to calculate the Shannon information entropy in positon space to probe the classical correlation of the doubly excited state in positronium negative ion. Atomic units are used in the present work.

2. Theoretical Method

2.1. The Stabilization Method

The non-relativistic Hamiltonian (in atomic units) for the three-body atomic system, assuming the nucleus is infinitely heavy, is given by

$$H = -\frac{1}{2m_1}\nabla_1^2 - \frac{1}{2m_2}\nabla_2^2 - \frac{1}{2m_3}\nabla_3^2 - \frac{Z}{r_{13}} - \frac{Z}{r_{23}} + \frac{1}{r_{12}} \tag{3}$$

where 1 and 2 denote the electron 1 and electron 2, respectively, and 3 for the positron, and r_{ij} is the relative distance between the particle i and j. For S-states we use Hylleraas-type wave functions to describe the system, with

$$\Psi_{kmn}\left(\mathbf{r_{13}}, \mathbf{r_{23}}\right) = \sum_{kmn} C_{kmn}\left\{r_{12}^k r_{13}^m r_{23}^n e^{-\alpha(r_{13}+r_{23})} + (1 \leftrightarrow 2)\right\} \tag{4}$$

where $k+m+n \leq \omega$ and ω, k, m, and n are positive integers, or zero. In the present work we use wave functions up to $N = 252$ terms, with $\omega = 12$. The ground state and the lowest-lying doubly excited state of Ps^- ion are of singlet-spin states, denoted as $1s^2\,{}^1S^e$ and $2s^2\,{}^1S^e$, respectively. As the spin parts of these ions are antisymmetric, the spatial parts are hence symmetric, as shown in Equation (4). For calculations of resonance states we apply the stabilization method [37–40]. In this method, by varying the non-linear parameter α in the wave function, we can plot the stabilization graph. The resonance wave function shows a stable behavior with respect to the change of α, resulting in a plateau near the resonance energy in the energy *vs.* α plot. From the stabilization plot, we can calculate the density of states by evaluating the inverse of the slope [38–40],

$$\rho_n = \frac{\alpha_{n+1} - \alpha_{n-1}}{E_{n+1} - E_{n-1}} = \left|\frac{dE}{d\alpha}\right|^{-1}_{E(\alpha)=E'} \tag{5}$$

The resonance energy and width can then be determined by fitting ρ_n to the Lorentz function,

$$\rho_n(E) = \frac{a(\Gamma/2)}{(E - E_r)^2 + \Gamma^2/4} + b \tag{6}$$

where E_r and Γ are the resonance energy and the resonance width respectively. We choose the α value at the peak of the density of states to construct the wave function for this particular doubly excited resonance state, and then use the wave function to quantify entanglement entropies such as von Neumann entropy and linear entropy by using the Schmidt decomposition method.

2.2. Von Neumann and Linear Entropies

The quantum entanglement of an atomic system can be quantified with entropies. The reduced density matrix, shown in Equations (1) and (2), can be expressed as

$$\rho_{red}\left(\mathbf{r}_1, \mathbf{r}_2\right) = \int \left[\Psi\left(\mathbf{r}_1, \mathbf{r}'\right)\right]^* \Psi\left(\mathbf{r}', \mathbf{r}_2\right) d\mathbf{r}' \tag{7}$$

To calculate eigenvalues of the reduced density matrix, we use the Schmidt decomposition method, and the detail of this computational scheme for Hylleraas functions was presented in [15]. A two-electron wave function can be decomposed into a sum of products of one-particle functions by partial wave expansion in a series of Legendre polynomials, as

$$\Psi\left(\mathbf{r}_1, \mathbf{r}_2\right) = \sum_{l=0}^{\infty} \frac{f_l\left(r_1, r_2\right)}{r_1 r_2} P_l\left(\cos\theta\right) \tag{8}$$

and the coefficients f_l are used for construction of reduced density matrix, with a maximum value $l_{max} = 40$ is used in the present work. The function f_l can be expended by the Schmidt decomposition:

$$f_l\left(r_1, r_2\right) = \sum_{n=0}^{\infty} \lambda_{nl} u_{nl}(r_1) u_{nl}(r_2) \tag{9}$$

where the u_{nl} is a set of one-particle orthonormal basis, and the λ_{nl} can be expressed as an eigenvalue problem in a form of integral equation

$$\int_0^{\infty} f_l\left(r_1, r_2\right) u_{nl}(r_2) dr_2 = \lambda_{nl} u_{nl}(r_1) \tag{10}$$

Once the elements of the density matrix for a given partial wave are determined, eigenvalues λ_{nl} can be obtained by diagonalization of the partial wave reduced density matrix. From Reference [15], it can be shown that λ_{nl} is related to Λ_{nl}, as

$$\Lambda_{nl} = \left(\frac{4\pi\lambda_{nl}}{2l+1}\right)^2 \tag{11}$$

The von Neumann entropy for spatial entanglement is then expressed as

$$S_{vN} = -\sum_{nl} \left(2l+1\right) \Lambda_{nl} \log_2 \Lambda_{nl} \tag{12}$$

and the linear entropy for spatial entanglement as

$$S_L = 1 - \sum_{nl} \left(2l+1\right) \Lambda_{nl}^2 \tag{13}$$

To test the accuracy of our calculations, we can use the following criteria for the sum of eigenvalues,

$$\sum_{nl} \left(2l+1\right) \Lambda_{nl} = 1 \tag{14}$$

In order to make sure that the numerical results are accurate enough for our purpose, we compare the calculated value from Equation (14) with its theoretical value and estimate the numerical errors. Equation (14) is based on the normalization condition. According to such normalization condition, the trace of a density matrix should be one. As a result, in this case, the sum of all diagonal elements in the reduced density matrix would always be one. The $(2l+1)$ weight in Equation (14) is needed to handle the degeneracy regarding the magnetic quantum number m. The readers are referred to Reference [15] for more details about this issue. Here, we should also mention that we emphasis on the spatial entanglement (the electron-electron orbital entanglement) of the positronium negative ion. For entanglement due to the spin part, readers are referred to some earlier publications (Reference [13] and references therein) for detailed discussions.

2.3. Shannon Information Entropy

In this work, we also calculate Shannon information entropy in position space for the doubly excited $2s^2\,{}^1S^e$ state in Ps^-. Shannon entropy was first introduced by Claude E. Shannon [41] in 1948 to quantify the average amount of information in a source and to evaluate probability distribution of a sample, and it also shows statistical correlations between particles [42,43]. Investigations on Shannon information entropy of atomic systems have also been reported in the literature [44]. In our case for the positronium negative ion, Shannon entropy provides information about the localization or delocalization of this two-electron system. The von Neumann entropy, and its linear approximation, the linear entropy, are measures of quantum correlations for the said ion. For a multi-electron system, Shannon information entropy in position space, denoted as S_r, is defined by the one-electron charge density $\rho(r)$, after averaging out the rest of other electrons, as

$$S_r = -\int_0^\infty \rho\,(r)\ln\rho\,(r)\,4\pi r^2 dr \tag{15}$$

Here, in our present case for a two-electron system, the function $\rho(r)$ is shown as

$$\rho(r_1) = \frac{2\pi}{r_1}\int_0^\infty \int_{|r_1-r_2|}^{r_1+r_2} |\Psi|^2 r_2 dr_2 r_{12} dr_{12} \tag{16}$$

The integral in Equation (15) is solved by the numerical Gauss-Laguerre quadrature integration. The convergence of the numerical calculation is tested by the normalization condition:

$$\int_0^\infty \rho(r) 4\pi r^2 dr = 1 \tag{17}$$

We should mention that in the present work we only calculate the Shannon entropy in position space, S_r. In order to test the entropic uncertainty principle [45] for an atomic state in three dimensional space, the Shannon entropy in corresponding momentum space, S_p, should also be calculated, with

$$S_p = -\int_0^\infty \gamma(p)\ln\gamma(p) 4\pi p^2 dp \tag{18}$$

where $\gamma(p)$ denotes the momentum density, normalized to unity. Defining S_T as the entropy sum, the entropic uncertainty principle is expressed as

$$S_T = S_r + S_p \geq 3(1+\ln\pi) \tag{19}$$

The entropy sum can also be used to test a stronger version of the Heisenberg's uncertainty principle for an N-electron system [45], with

$$S_T \geq 3N(1+\ln\pi) - 2N\ln N \tag{20}$$

It would be an interesting and challenging problem to quantify Shannon entropy in momentum space for two-electron states using highly correlated wave functions, and such a calculation is outside the scope of our present investigation.

3. Results and Discussion

3.1. Resonance Energy, Width and Wave Function

In determining the resonance states with the stabilization method, we vary the non-linear parameter α in the Hylleraas-type wave functions to calculate the energy spectrum, and from which stabilization plots are constructed. As shown in Figure 1, there are three stabilization plateaus for the doubly-excited $2s^2\,{}^1S^e$ resonance state below the $N = 2$ ($2s$ and $2p$) thresholds of the positronium atom. For each plateau, we convert it to the density of states ρ_n using Equation (5) and fit the calculated density to the Lorentzian profile, Equation (6), to determine the resonance energy and width. Figure 2 shows the fitting of density of states for plateau "I" to the Lorentzian profile for the $2s^2\,{}^1S^e$ state. Among the different stabilization plateaus for a given state, the one with the best r^2 (In statistics, the coefficient of determination, denoted by R^2 or r^2, shows the goodness of the fit) value closer to 1.0, implying it is the best fit, is chosen to construct the wave function for the state. The results of the three plateaus for the resonance states are listed in Table 1, and it shows that plateau "I" gives the best fit as its r^2 is closest to the ideal value of 1.0. Therefore, we conclude that the best values for the resonance energy and width in our present work obtained using $N = 252$ terms are $E_r = -0.07603006$ a.u. and $\Gamma/2 = 0.00004310$ a.u.. In Table 1, we also compare our present stabilization results with those of earlier results for the resonance energy $E_r = -0.07603044$ a.u. and $\Gamma/2 = 0.00004303$ a.u. [46] that were obtained by using complex-scaling method [47]. It shows that the present results for the resonance parameters (energy and width) agree quite well with those obtained by using the complex-scaling method.

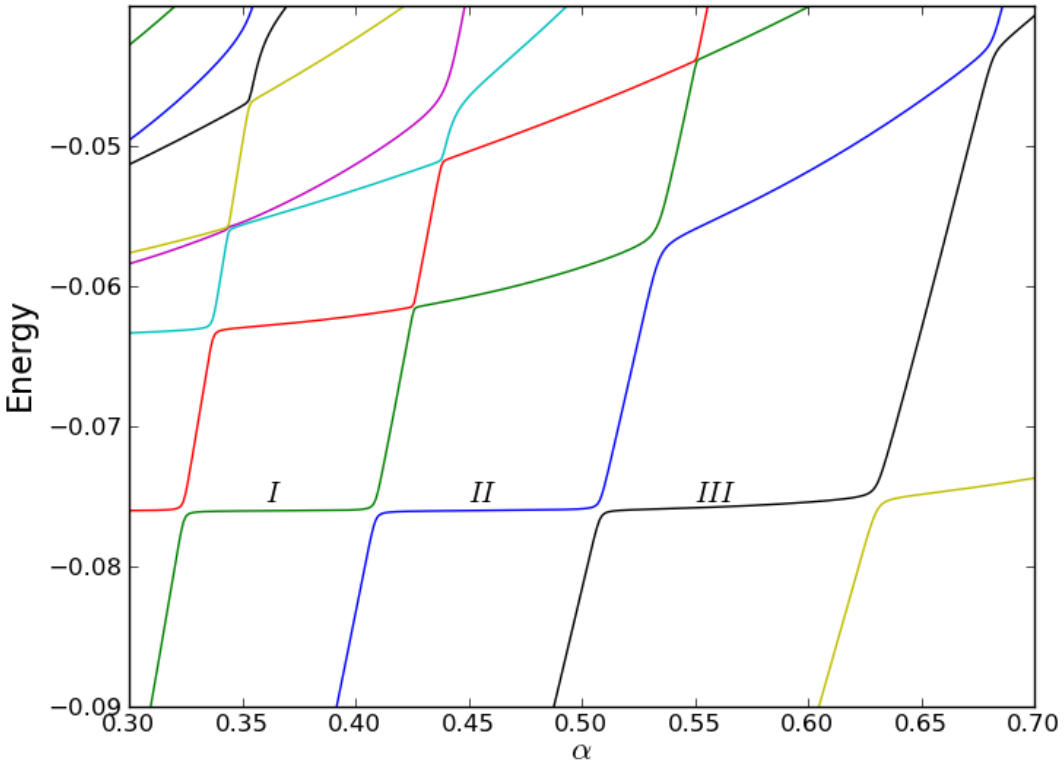

Figure 1. Stabilization plot for the doubly excited $2s^2\,{}^1S^e$ state in Ps^-.

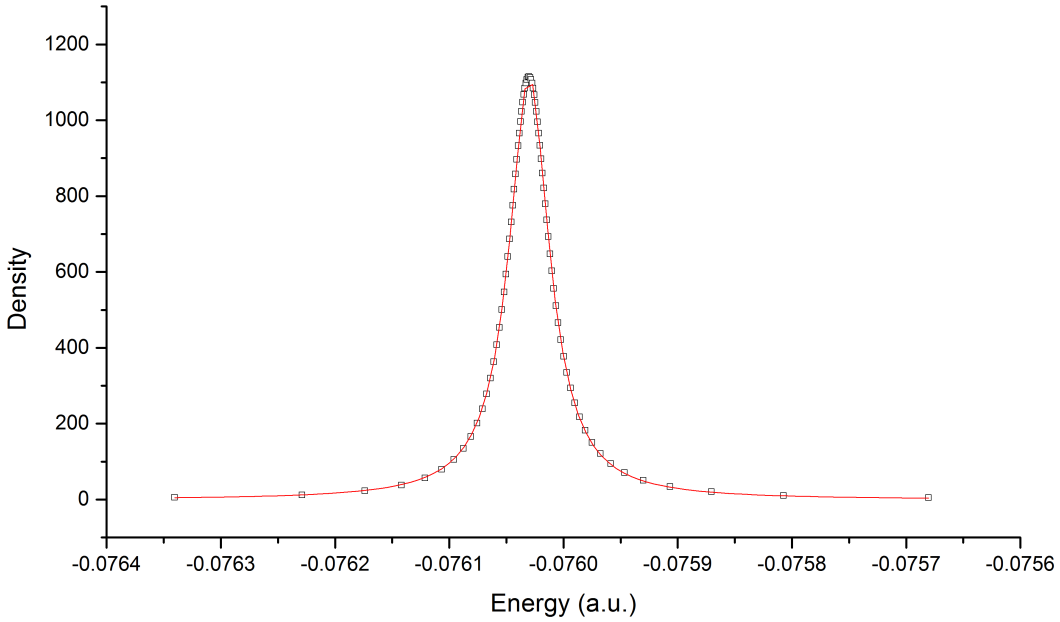

Figure 2. Fitting of density of resonance state to the Lorentzian profile.

Table 1. Resonance energy, width, entanglement entropies for the doubly excited $2s^2\,{}^1S^e$ state in Ps^-, obtained by using $N = 252$ term wave functions.

Wave function	E_r **(a.u.)**	$\Gamma/2$ **(a.u.)**	r^2	S_L	S_{vN}
Reference [46]	−0.07603044	0.00004303			
Section I	−0.07603006	0.00004310	0.9999715	0.516517	1.684376
Section II	−0.07602959	0.00004421	0.9997358	0.516568	1.684917
Section III	−0.07602008	0.00008226	0.9831704	0.517067	1.686998

3.2. von Neumann Entropy and Linear Entropy

Next we employ the resonance wave function at the center of plateau "I" to calculate the von Neumann and linear entropies as given in Equations (4)–(6) using the Schmidt decomposition method [15]. Table 2 summarizes our results obtained by using the 252-term ($\Omega = 12$) wave function. As for the doubly excited resonance state in the Ps^- ion, there are no published results for entanglement entropy in the literature, to the best of our knowledge. We summarize our results in the form of a (S_L, S_{vN}) pair. The entropy pair for the $2s^2\,{}^1S^e$ state is determined as $(0.516517, 1.684376)$. For completeness, we also show in Table 2 the entanglement entropies for the ground $1s^2\,{}^1S^e$ state of the Ps^- ion [16,18]. It shows that S_L and S_{vN} for the doubly excited state are in general larger than those for the ground state, indicating that entanglement would increase when the system is changed from the ground state to the doubly excited state. Such phenomenon can be explained as following; While the ground ${}^1S^e$ state is denoted as $1s^2$ in the single particle approximation, it is a mixture of ss, pp, dd, *etc.*, components. As for doubly excited states below the $N = 2$ threshold, the $2s^2$ state is actually mixed by configurations interaction of various components with $2sns$, $2pnp$, dd, ff, *etc.* In general, the correlation for a doubly excited state that is attributed to configuration interaction between the abovementioned components

would have a larger correlation effect than that for the ground state [48,49]. Furthermore, as the correlation is directly related to the spatial (electron-electron orbital) entanglement [12], we conclude that the stronger correlation effect to a state (such as the present doubly excited state), the larger value is to its spatial entanglement entropy.

Table 2. Entropies in the ground and doubly excited states of Ps^-.

State	Energy (a. u.)	S_r	S_L	S_{vN}
$1s^2\,{}^1S^e$	−0.262005069 [a]	7.955301 [b]	0.120796 [a]	0.412527 [a]
$2s^2\,{}^1S^e$	−0.07603006	10.848235	0.516517	1.684376

[a] [16]; [b] [18].

3.3. Shannon Entropy for Doubly Excited State of Ps^-

In our present work, we also calculate the Shannon information entropy in position space, S_r, for the positronium negative ion, so as to investigate correlations of charge density distribution between the two electrons in this system. Again, we use the wave function obtained by using the stabilization method described above, and carry out numerical integrals on the one-electron density distribution to calculate S_r, by using Equations (13)–(15). Our result is shown in Table 2, giving the value of $S_r = 10.848235$. In comparing with that of the ground state value of $S_r = 7.955301$ (Reference [18]), it is seen that the doubly excited state in Ps^- is more delocalized than that for the ground state. Such results are consistent with our intuition that as the two electrons in a doubly excited state are further away from the nucleus than when they are in their ground state, the doubly excited state hence exhibits a more defused character.

4. Conclusions

We have carried out an investigation of quantum spatial (electron-electron orbital) entanglement on doubly excited $2s^2\,{}^1S^e$ resonance state in Ps^- using Hylleraas functions to take into account of the correlation effects. The resonance wave function is obtained by employing the stabilization method, and once such wave function is obtained the Schmidt decomposition method is subsequently applied to quantify entanglement entropies, *i.e.*, von Neumann entropy and linear entropy. We have also used the resonance wave function to calculate Shannon information in position space for such resonance state, calculated for the first time in the literature, to the best of our knowledge. Finally, we should mention that at present, we are not aware of any experimental scheme to observe entanglement for these two-electron states. Nevertheless, as our calculations of entanglement entropies of two-electron states are quite accurate, it is hoped that our recent results would stimulate experimental investigations to observe entanglement entropies for atomic states, and that our results would play a useful reference for future investigations of quantum information in quantum chemistry and atomic physics.

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Author Contributions

Yew Kam Ho set the research direction, contributed to development of the theory, and managed the overall progress of this project. Chien-Hao Lin carried out the analytic and numerical calculations for the results presented in the present work. Both authors contributed to analyzing the numerical data and provided physical interpretation of the present results. Both authors contributed to writing the paper, have read and approved the final manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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