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Doubly Excited Resonance States of Helium Atom: Complex Entropies

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Abstract We provide a diagonal form of a reduced density matrix of S-symmetry resonance states of two electron systems determined under the framework of the complex scaling method. We have employed the variational Hylleraas type wavefunction to estimate the complex entropies in doubly excited resonance states of helium atom. Our results are in good agreement with the corresponding ones determined under the framework of the stabilization method (Lin and Ho in Few-Body Syst 56:157, 2015).

1 Introduction

Last few years produced bulk of studies on quantum entanglement of few-body systems because of the development of new experimental techniques that have opened up perspectives for their applications in quantum information technology [1]. In particular, there has been a remarkable increase of interest in exploring properties of various quantum composite systems [2–17]. In recent years, both the entanglement and Shannon Entropy in helium and helium-like ions have also accelerated a great research attention [18–33]. However, studies on the correlation in resonance states have not yet drawn much attention. Pont et al. [34] proposed to associate the entanglement of a resonance state with a real part of a linear entropy computed from complex-valued eigenvalues of a reduced density matrix \( \hat{\rho} \) determined in the framework of the complex scaling method (CSM) [35–38]. Within the framework of this approach, the properties of two-electron one-dimensional (1D) quantum dots near the autoionization thresholds have recently been investigated in [39]. The latest researches performed by Lin et. al. treated the quantum entanglement for doubly excited resonance states in helium and in the positronium negative ion under the framework of the stabilization method, [28] and [29], respectively.

A recent study [40], performed by one of us, has made a detailed discussion on the diagonal representation of the reduced density matrix for resonance states by using the CSM. Following to [35,41], the real and imaginary parts of an expectation value of a given complex-scaled operator give the average value of the quantity under consideration, and its uncertainty, respectively. In a strict mathematical sense, the value of the von Neumann (linear) entropy can be given as the expectation value of the operator \( \log_2 \hat{\rho} (\hat{1} - \hat{\rho}) \). However, it must be stressed that the entropy cannot rigorously be treated as the expectation value of a quantal observable [42]. Therefore, in a resonance case, the interpretation of the real and imaginary parts of the expectation value of \( \log_2 \hat{\rho} (\hat{1} - \hat{\rho}) \) as the entropy and its uncertainty, respectively, is problematic. Despite of this fact, we call the average values of the operators \( \log_2 \hat{\rho} \) and \( \hat{1} - \hat{\rho} \) as the complex-valued von Neumann and linear entropies, respectively, and, following to [34,39,40], use them to characterize the correlation in resonance states. The
purpose of this brief paper is to determine the complex entropies for doubly excited resonance states of helium, \(2s^2 1S^e\) and \(2p^2 1S^e\), and compare the results obtained with the corresponding ones determined in different ways [27,28].

This paper is organized as follows. In Sect. 2, we derive a diagonal representation of the complex reduced density matrix for the singlet S-symmetry resonance states. In Sect. 3, we determine the complex linear and von Neumann entropies for doubly excited resonance states \(2s^2 1S^e\) and \(2p^2 1S^e\) and some concluding remarks are left for Sect. 4.

2 Method

For the system under consideration here, the complex-scaled Hamiltonian is of the form [43]

\[
\hat{H}^\theta = \sum_{j=1}^{2} \left[ -\frac{e^{-2i\theta}}{2} \nabla_j^2 - \frac{Ze^{-i\theta}}{r_j} \right] + \frac{e^{-i\theta}}{r_{12}},
\]

(1)

where \(\theta\) is the so-called scaling angle. A key point of the CSM is that the complex-scaled Hamiltonian is a non-Hermitian operator and the inner product is defined as

\[
\langle \psi | \varphi \rangle = \int \psi(x)\varphi(x)dx.
\]

(2)

In the case of the resonance states, the density operator is defined as

\[
\hat{\rho} = |\psi^R\rangle \langle \psi^L|,
\]

(3)

where \(|\psi^R\rangle\) and \(|\psi^L\rangle\) are right and left eigenstates of \(\hat{H}^\theta\), which are determined by the following eigenequations \(\hat{H}^\theta |\psi^R\rangle = E_i(\theta)|\psi^R\rangle\) and \(\langle \psi^L | \hat{H}^\theta = \delta_{ij} |\psi^L | \psi^R\rangle\), respectively, \(\langle \psi^L | \psi^R\rangle = \delta_{ij}\). The parameters of a given resonance level are determined at the angle \(\theta = \theta_{opt}\) at which

\[
\left. \frac{dE_i(\theta)}{d\theta} \right|_{\theta=\theta_{opt}} = 0.
\]

The complex reduced density matrices for particles 1 and 2 are given by \(\hat{\rho}_{12} = T_{r21}[|\psi^R\rangle \langle \psi^L|]\). An essential point is that the Hamiltonian (1) does not change upon permutation of the coordinates of particles, so that its right and left eigenfunctions can be expressed in an identical form \(\langle \psi^L | \mathbf{r} \rangle = \langle \mathbf{r} | \psi^R \rangle = \psi\), \(\langle \psi | \psi \rangle = 1\) [40,44]. Here we restrict our study to the singlet S-symmetry states, the wavefunctions of which depend only on the radial coordinates \(r_1, r_2\) and the inter-electronic angle coordinate \(\theta_{12}\), \(\psi(r_1, r_2) = \psi(r_1, r_2, \cos \theta_{12})\). First we derive a diagonal form of the corresponding complex reduced density matrix with respect to the inner product (2). For the sake of brevity, we start with the Fourier-Legendre series representation of \(\psi\), namely

\[
\psi(r_1, r_2, \cos \theta_{12}) = \sum_{l=0}^\infty \frac{f_l(r_1, r_2)}{r_1 r_2} P_l(\cos \theta_{12}),
\]

(4)

where

\[
f_l(r_1, r_2) = r_1 r_2 \frac{2l + 1}{2} \int_0^\pi \psi(r_1, r_2, \cos \theta_{12}) P_l(\cos \theta_{12}) \sin \theta_{12} d\theta_{12}
\]

\[= r_1 r_2 \frac{2l + 1}{2} \int_{-1}^1 \psi(r_1, r_2, t) P_l(t) dt.
\]

(5)

Further we proceed similarly as in the case of a two-boson wavefunction in [40]. Here we expand the function \(f_l\) into the basis of orthonormal Laguerre basis functions, namely

\[
f_l(r_1, r_2) = \sum_{ij} a_{ij}^{(l)} L_i(r_1)L_j(r_2),
\]

(6)
with
\[
L_n(r) = \sqrt{\omega}e^{-\frac{r}{2\omega}} F_1(-n; 1; r\omega),
\]
where the coefficient \(a_{ij}^{(l)}\) are given by the following integrals
\[
a_{ij}^{(l)} = \frac{2l + 1}{2} \int_0^\infty \int_0^1 r_1 r_2 \psi(r_1, r_2, t) P_l(t) L_l(r_1)L_j(r_2) dt d r_1 d r_2.
\]

Since the function \(f_l\) is symmetric under the permutations of coordinates, the matrix \(A_l = [a_{ij}^{(l)}]\) is symmetric, which means that its eigenvector matrix \(V\) (\(V^{-1} = V^T\)) and the corresponding eigenvalues \(D = \text{diag}(d_0, d_1, \ldots)\) satisfy \(A_l = V D V^T\). After rewriting this formula as
\[
a_{ij}^{(l)} = \sum_n (V_l)_{in} d_{nl}(V_l)_{jn},
\]
and substituting it into (6), we obtain
\[
f_l(r_1, r_2) = \sum_n d_{nl} v_{nl}(r_1)v_{nl}(r_2),
\]
where
\[
v_{nl}(r) = \sum_k (V_l)_{kn} L_k(r).
\]

Hence, the substitution of Eq. (10) together with the well-known formula:
\[
P_l(\cos \theta_{12}) = \frac{4\pi}{2l + 1} \sum_{m=-l}^{m=l} Y_{lm}(\varphi_1, \theta_1) Y_{lm}^*(\varphi_2, \theta_2),
\]
into (4) gives
\[
\psi(r_1, r_2) = \sum_{m=-l}^{m=l} \sum_{m=-l}^{m=l} k_{nm} u_{nlm}(r_1) u_{nlm}^*(r_2),
\]
where \(u_{nlm}(r) = \frac{v_{nl}(r)}{r} Y_{lm}(\varphi, \theta)\) and \(k_{nm} = \frac{4\pi d_{nl}}{2l + 1}\). Thus, \(\langle \hat{u}_{n'l'm'}^* \rangle \hat{u}_{nlm} \rangle = \delta_{nn'} \delta_{ll'} \delta_{mm'}\). As a result, the reduced density matrix for particles 1 or 2, takes a form
\[
\hat{\rho}_{12} = \sum_{m=-l}^{m=l} \sum_{m=-l}^{m=l} \lambda_{nm} \hat{u}_{nlm} \hat{u}_{nlm}^*,
\]
where \(\lambda_{nl} = k_{nl}^2\). The vectors \(\langle \hat{u}_{n'l'm'}^* \rangle \hat{u}_{nlm} \rangle\) are nothing else that the left and right eigenstates of \(\hat{\rho}_{12}\), respectively, with the complex eigenvalues \(\lambda_{nl}\), \(\langle \hat{u}_{n'l'm'}^* \rangle \hat{u}_{nlm} \rangle = \lambda_{nl} \delta_{nn'} \delta_{ll'} \delta_{mm'}\). Moreover, all of them, but the ones with \(l = 0\) are 2l + 1-fold degenerate so that the normalization condition gives
\[
\sum_{nl} (2l + 1) \lambda_{nl} = 1.\]
From here on, we call \(\lambda_{nl}\) complex occupancies and in terms of them, the von Neumann and linear entropies have the forms [7]
\[
S = -\sum_{nl} (2l + 1) \lambda_{nl} \log_2 \lambda_{nl},
\]
and
\[
L = 1 - \sum_{nl} (2l + 1) \lambda_{nl}^2,
\]
respectively. Following to [40], we associate the real and imaginary parts of the complex entropy as the amount of correlation and its uncertainty, respectively.
3 Results and Discussion

To compute the S-state energies, the widths and resonance eigenfuctions as well, we have used the Hylleraas basis functions

$$\psi_{Hy} = \sum_{nmp} c_{nmp} e^{-\alpha x^2} y^m u^p,$$

(16)

with \(0 \leq n + m + p \leq N\), where \(s = r_1 + r_2, t = r_1 - r_2, \alpha = r_1^2 + r_2^2 - 2r_1r_2 \cos \theta_{12}\), which leads to the eigenproblem:

$$[H^{(0)} - ES]C = 0,$$

(17)

where \(H^{(0)} = \langle n'm'p'|\hat{H}^{(0)}|nmp\rangle\) and \(S = \langle n'm'p'|nmp\rangle\) are the Hamiltonian matrix and the overlap matrix, respectively. To determine the energies of resonance states \(2s^2 1S^e\) and \(2p^2 1S^e\) and their corresponding wavefunctions, we used an expansion (16) given by a 364-term \((N = 11)\). Our results obtained at \(\theta_{opt} = 0.3, \alpha = 1\): \(E_{2s^2 1S^e} = -0.7778678 - 0.00227061i, E_{2p^2 1S^e} = -0.62192722 - 0.00107813i\) reproduce the corresponding literature values determined with a very high accuracy [45], that is, \(E_{2s^2 1S^e} = -0.777867636 - 0.002270653i, E_{2p^2 1S^e} = -0.621927254 - 0.00107818i\), with to at least 7 digits. The calculation of the complex entropies requires determination of the coefficients \(d_{nl}\). As described in Sect. 2, for a fixed value of \(l\), the \(n_{th}\) order approximations to the \(n_m\) lowest coefficients \(d_{nl}\) can be obtained by diagonalizing the matrix \(A_l = [d_{ij}]_{n_m \times n_m}\), the elements of which are generated by the three-dimensional integrals (8), which we treat here numerically. Having the coefficients \(d_{nl}\) determined in that way for \(l \leq l_m\), we obtain thus the approximations to the true values of \(S\) and \(L\) as \(S_{ap} = -\sum_{n,l} n_{m,l}^{-1}(2l + 1) \lambda_{nl} \log_2 \lambda_{nl} \) and \(L_{ap} = 1 - \sum_{n,l} n_{m,l}^{-1}(2l + 1) \lambda_{nl}^2\), respectively, where \(\lambda_{nl} \equiv (\frac{\sigma d_{nl}}{2l + 1})^2\). The adjustable parameter \(\omega\) appearing in (7) can be used to improve the convergence. A good criterion for convergence is the closeness of the sum of approximate complex occupancies to the theoretical value 1. We have verified by chance that a good rate of convergence is achieved at a parameter value \(\omega = 2.5\) for both cases under consideration.

Table 1 Linear and von Neumann complex entropies for the state \(2s^2 1S^e\) computed at \(\omega = 2.5\)

| \(n_m\) | \(l_m\) | \(L_{ap}\) | \(S_{ap}\) | \(\sum n_{m,l}^{-1}(2l + 1) \lambda_{nl}\) |
|--------|--------|-----------|-----------|----------------------------------|
| 10     | 0      | 0.477362  + 0.00176401i | 0.337396  + 0.000928007i | 0.7227374  + 0.000207261i |
| 1      | 0.451993  + 0.00133491i | 1.298414  + 0.00011255i | 0.999573  + 0.00002117i |
| 2      | 0.451993  + 0.00133491i | 1.304087  + 0.000731422i | 0.999991  + 4.52956 * 10^{-6}i |
| 3      | 0.451993  + 0.00133491i | 1.304263  + 0.000737337i | 0.999999  + 3.27358 * 10^{-6}i |
| 4      | 0.451993  + 0.00133491i | 1.304294  + 0.000735341i | 1.000000  + 2.99857 * 10^{-6}i |
| 5      | 0.451993  + 0.00133491i | 1.304303  + 0.000734536i | 1.000000  + 2.92017 * 10^{-6}i |
| 15     | 4      | 0.451994  + 0.00133551i | 1.304295  + 0.000735802i | 0.9999999  + 5.33902 * 10^{-7}i |
| 5      | 0.451994  + 0.00133551i | 1.304305  + 0.000735984i | 1.000000  + 4.58637 * 10^{-7}i |
| 20     | 4      | 0.451994  + 0.00133543i | 1.304293  + 0.000735533i | 0.9999994  + 8.59354 * 10^{-7}i |
| 5      | 0.451994  + 0.00133543i | 1.304303  + 0.000735716i | 0.9999999  + 7.84988 * 10^{-7}i |

Table 2 Linear and von Neumann complex entropies for the state \(2p^2 1S^e\) computed at \(\omega = 2.5\)

| \(n_m\) | \(l_m\) | \(L_{ap}\) | \(S_{ap}\) | \(\sum n_{m,l}^{-1}(2l + 1) \lambda_{nl}\) |
|--------|--------|-----------|-----------|----------------------------------|
| 10     | 0      | 0.903817  + 0.002901117i | 0.773731  - 0.00071258i | 0.319393  + 0.005675030i |
| 1      | 0.777520  - 0.00071459i | 2.192127  - 0.00318651i | 0.967059  - 0.000226222i |
| 2      | 0.777321  - 0.00071258i | 2.429632  - 0.00416282i | 0.999129  - 0.000388452i |
| 3      | 0.777321  - 0.00071258i | 2.443242  - 0.00421897i | 1.000160  - 0.000392804i |
| 4      | 0.777321  - 0.00071258i | 2.445819  - 0.00423267i | 1.000320  - 0.000393569i |
| 5      | 0.777321  - 0.00071258i | 2.446525  - 0.00423974i | 1.000350  - 0.000393096i |
| 15     | 4      | 0.777501  - 0.00088995i | 2.445411  - 0.00375428i | 0.999367  + 5.24362 * 10^{-7}i |
| 5      | 0.777501  - 0.00088995i | 2.446149  - 0.00375853i | 0.999767  + 3.16617 * 10^{-7}i |
| 20     | 4      | 0.777500  - 0.00088971i | 2.445416  - 0.00375074i | 0.999367  + 1.26021 * 10^{-7}i |
| 5      | 0.777500  - 0.00088971i | 2.446156  - 0.00375410i | 0.999777 - 4.65872 * 10^{-7}i |
Table 3  Comparison of complex eigenenergy, linear and von Neumann complex entropies for $2s^2 \, S^e$ and $2p^2 \, S^e$ states of helium atom

|            | $Re[E]$       | $Im[E]$      | $Re[L_{ap}]$ | $Re[S_{ap}]$ |
|------------|---------------|--------------|--------------|--------------|
| Ref. [27]  |               |              |              |              |
| $2s^2 \, S^e$ | $-0.778775$   |              | $0.882$      |              |
| $2p^2 \, S^e$ | $-0.622559$   |              | $1.346$      |              |
| Ref. [28]  |               |              |              |              |
| $2s^2 \, S^e$ | $-0.7778583$  | $-0.0022875$ | $0.461704$   | $1.378501$   |
| $2p^2 \, S^e$ | $-0.6219259$  | $-0.000105$  | $0.777653$   | $2.450665$   |
| Ref. [45]  |               |              |              |              |
| $2s^2 \, S^e$ | $-0.77786736$ |              |              |              |
| $2p^2 \, S^e$ | $-0.62192754$ |              |              |              |
| This work  |               |              |              |              |
| $2s^2 \, S^e$ | $-0.7778678$  | $-0.00227061$| $0.451994$   | $1.304303$   |
| $2p^2 \, S^e$ | $-0.62192722$ | $-0.00010781$| $0.777500$   | $2.446156$   |

In Tables 1 and 2 our results obtained for $L_{ap}$ and for $S_{ap}$ together with the results for the sum of approximate occupancies are presented as functions of the cut-offs $n_m$ and $l_m$, for the $2s^2 \, S^e$ and $2p^2 \, S^e$ states, respectively. Note that already at $n_m = 10$, fairly stable results were achieved at $l_m = 1$ in the former case and at $l_m = 2$ in the latter one. We can see that the real and imaginary parts of the sum of approximate occupancies tend to one and zero, respectively, as $n_m$ and $l_m$ are increased, which confirms the correctness of our calculations. It is evident from the tables that in the case of $2s^2 \, S^e$, the terms higher than $l = 0$ in (4) contribute very little, which means that the angular correlation effects are expected to be very poor in this case. As expected, the situation is different in the case of $2p^2 \, S^e$, where, in contrast, the most of electrons correlation comes mainly from the first angular level $l = 1$, which means that the angular correlations contribute importantly to the total correlation of the state. In Table 3 our results are compared with the ones obtained by two other methods [27,28], where, for the sake of completeness, we also added the reference values for the energy. We find that the real parts of the complex entropies coincide fairly well with the results obtained by C.H. Lin et al. by the stabilization method [28]. Both the results obtained by the Lin et al. and our results differ considerably from the ones obtained by Restrepo Cuartas et al. [27]. We suspect that the reason for this lies in the fact that probably the J.P. Restrepo Cuartas et al. have not taken into account in their calculations the degeneracies with respect to the magnetic quantum number. Note that the imaginary part of the complex linear entropy is positive for the $2s^2 \, S^e$ state and negative for the $2p^2 \, S^e$ state. To understand the physical meaning of the sign of the imaginary part of the complex entropy, more constructive analysis is needed, and this will be a subject of our further research.

4 Summary

In summary, we derived the diagonal representation of the complex reduced density matrix of the singlet $S$-symmetry states based on the Fourier-Legendre series. Subsequently, we have computed the complex entropies for doubly excited resonance states of helium atom. Our results for the real parts of the complex entropies are in a very good agreement with those determined under the framework of the stabilization method by C.H. Lin et al. [28]. The approach presented in this paper can be an alternative to the one based on the stabilization method when determining the real parts of the linear and von Neumann entropies of $S$-symmetry resonance states.

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