

Positron attachment to the He doubly excited states

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The projection method is used to demonstrate the existence of positron attachment to three doubly excited states of helium. The $e^+\text{He}(2s^2\ ^1\text{S}^e)$, $e^+\text{He}(3s^2\ ^1\text{S}^e)$, and the $e^+\text{He}(2s2p\ ^3\text{P}^o)$ states have binding energies of 0.447 eV, 0.256 eV and 0.486 eV respectively. These energies were computed with the stochastic variational method and the configuration interaction method. These states will exist as resonances in the e^+ -He continuum and the $e^+\text{He}(2s^2\ ^1\text{S}^e)$ state could be detectable in the $e^++\text{He}$ collision spectrum. A resonance width of 0.068 eV was computed for the $e^+\text{He}(2s^2\ ^1\text{S}^e)$ state by using the complex rotation method. The existence of a series of $e^+\text{He}(ns^2\ ^1\text{S}^e)$ resonances associated with the $\text{He}(ns^2)$ double Rydberg series is also predicted and an explicit calculation demonstrating the existence of the $e^+\text{He}(3s^2\ ^1\text{S}^e)$ state is reported.

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In this letter the ability of a positron to attach itself to the doubly excited states of helium is demonstrated by explicit calculation using the Feshbach projection operator approach that was used in some of the earliest calculations of the helium doubly excited spectrum [1, 2]. Besides the intrinsic interest in such exotic Coulomb systems, the result provides a pathway to providing experimental confirmation that positrons can be attached to electrically neutral atoms to form bound states.

It is now widely accepted that positrons can form bound states with a variety of atoms [3-5]. While the evidence for positron binding is strong, it is derived from calculation. Binding energies range from 0.0129 eV in the case of $e^+\text{Na}$ [6] to 0.50 eV for the $e^+\text{Ca}$ ground state [7].

There is solid experimental evidence that positrons can form bound states with a variety of molecules. The energy resolved positron annihilation cross sections for a number of molecules (e.g. C_3H_8 , C_6H_{14}) show features that have been identified as Feshbach resonances formed by the trapping of positrons in vibrationally excited states of molecules [8]. This is thought to be the mechanism responsible for the large positron annihilation rates observed for many molecules in gas-phase positron annihilation spectroscopy experiments [9].

While the experimental evidence of positron binding to molecules is good, there is no experimental evidence that could be construed as demonstrating the existence of positron-atom bound states. One possible signature would be the existence of resonant structures associated with atomic excited states in the positron scattering spectrum. Years of experimentation, however, have revealed little evidence for the existence of resonant states in positron-atom scattering spectra [4, 10, 11].

A number of schemes have been put forward to demonstrate the existence of positron-atom bound states [12-15]. The most recent proposal suggested that positron scattering experiments be performed on open shell transition-metal atoms having polarizabilities and ionization energies conducive to binding positrons [15]. Open shell systems are recommended since such systems would have low-lying excited states that could also bind a positron. Positron binding to low-lying excited states would result in Feshbach resonances appearing in the low-energy annihilation cross section. However, the transition-metal atoms most likely to bind a positron represent difficult propositions for experimentation.

The present letter demonstrates that three of the doubly excited states of helium, namely the $\text{He}(2s^2\ ^1\text{S}^e)$, $\text{He}(3s^2\ ^1\text{S}^e)$ and $\text{He}(2s2p\ ^3\text{P}^o)$ states can attach a positron with attachment energies exceeding 0.250 eV. The $e^+\text{He}(2s^2\ ^1\text{S}^e)$ and $e^+\text{He}(3s^2\ ^1\text{S}^e)$ states manifest themselves as resonances in the $e^++\text{He}$ continuum. A positron cannot excite the $\text{He}(2s2p\ ^3\text{P}^o)$ state from the $\text{He}(1s^2\ ^1\text{S}^e)$ ground state since there is no exchange interaction between the positron and electrons. These states can be regarded as analogues the triply excited negative ion resonances seen in the electron-helium spectrum at 57-61 eV incident energy [16-20].

One motivation for the present investigation was the realization that the doubly excited states of helium have energetics very similar to those of the Mg atom which binds a positron with a binding energy of 0.465 eV [28] and also supports a prominent p -wave shape resonance in the elastic scattering channel at 0.096 eV incident energy [28, 29]. The binding energy of the $\text{Mg}^+(3s)$ ground state is -0.55254 a.u. [30] while the $\text{He}^+(2s)$ binding energy

TABLE I: Energies (in a.u.) of some He states given with respect to the He^{2+} threshold. Three sets of helium energies are given. One set, E_{CR} are taken from complex rotation calculations, the two other sets are taken from projection operator calculations. The projection operator energies in the E_{QHQ} column come from calculations that use a Hylleraas basis, while those in the E_{CI} column come from CI calculations as described in the text. There is no complex rotation energy for the $\text{He}(2p^2\ ^3\text{P}^e)$ state since it is a bound state.

State	E_{CR}	E_{QHQ}	E_{CI}
$\text{He}^+(2s)$	-0.500000	-0.500000	-0.500000
$\text{He}(2s^2\ ^1\text{S}^e)$	-0.777818 [21]	-0.778774 [1]	-0.778781
$\text{He}(2s2p\ ^3\text{P}^o)$	-0.760498 [22]	-0.761492 [2]	-0.761492
$\text{He}(2p^2\ ^3\text{P}^e)$		-0.710500 [23]	-0.710500
$\text{He}(2p^2\ ^1\text{D}^e)$	-0.701946 [24]	-0.702817 [25]	-0.702819
$\text{He}(2s2p\ ^1\text{P}^o)$	-0.69314 [26]	-0.692895 [27]	-0.692897
$\text{He}(2p^2\ ^1\text{S}^e)$	-0.621928 [21]	-0.622744 [2]	-0.622736

is -0.50 a.u.. The binding energy of the $\text{Mg}(3s^2)$ ground state with respect to the $\text{Mg}^+(3s)$ threshold is -0.2810 a.u., while the binding energy of the $\text{He}(2s^2)$ resonance with respect to the $\text{He}^+(2s)$ state is -0.2778 a.u. [21]. The respective dipole polarizabilities, calculated with oscillator strength sum rules [31], are $76.2 a_0^3$ for $\text{He}(2s^2)$ and $71.3 a_0^3$ for $\text{Mg}(3s^2)$ [31]. The He energies are listed in Table I and plotted in Fig. (1).

The projection method [1, 2] provides a computational strategy for the identification of resonances. In this method, the electrons are not allowed to occupy those low-lying states that could result in the auto-ionization of the system. The projection method energies, E_{QHQ} , of the helium doubly excited states in Table I, computed using Hylleraas basis sets, differ from those determined by the dynamically complete complex rotation method by less than 0.001 a.u.. The projection method has successfully been applied to calculate the positions of the He^- resonances associated with the He doubly excited states [32]. Here, the Hamiltonian was chosen for the $N = 2$ electron and one positron system to be

$$\hat{H} = -\sum_{i=1}^{N+1} \frac{\nabla_i^2}{2} - \sum_{i=1}^N \frac{2}{r_i} + \frac{2}{r_{N+1}} + \sum_{i<j}^N \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_{i=1}^N \frac{1}{|\mathbf{r}_{N+1} - \mathbf{r}_i|}. \quad (1)$$

Investigation of resonant states requires diagonalizing the Hamiltonian $\hat{Q}\hat{H}\hat{Q}$, where the projection operator $\hat{Q} = (1 - \hat{P})$. For the $n = 2$ helium doubly excited states one can use combinations of the single particle projection operator $\hat{P}_i = |\phi_{1s}(\mathbf{r}_i)\rangle\langle\phi_{1s}(\mathbf{r}_i)| \equiv |1s\rangle\langle 1s|$ where $\phi_{1s}(\mathbf{r}_i)$ is the wavefunction of the $\text{He}^+(1s)$ orbital [32].

Two independent computational methods, the configuration interaction (CI) and the stochastic variational method (SVM) [3, 33], are used to diagonalize $\hat{Q}\hat{H}\hat{Q}$.

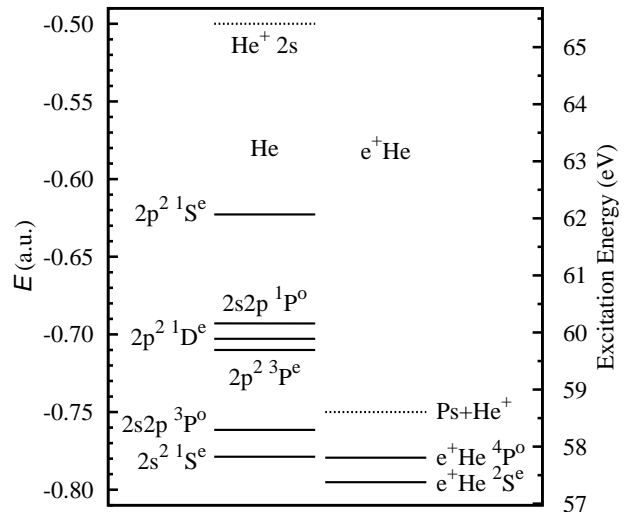


FIG. 1: Energy level diagram showing the positions of the He doubly excited states and the states with an attached positron. The position of the $\text{Ps}(1s)+\text{He}^+(2s)$ threshold is also shown. The axis on the right gives the positron collision energy (in eV) needed to excite these states.

The $\text{He}^+(1s)$ ground state is excluded from the CI wavefunction by Schmidt-orthogonalizing the $\ell = 0$ single-particle electron orbital basis to the $\text{He}^+(1s)$ state. This obviates the need for the inclusion of an explicit projection operator since $\langle 1s|\otimes\langle n\ell||\Psi\rangle = 0$ will automatically be satisfied by the CI basis that is used to diagonalize Eq. (1). The single particle $|n\ell\rangle$ in the present calculations were chosen to be Laguerre type orbitals (LTOs).

The CI method was initially applied to the calculation of the He doubly excited states. The basis included 49 LTOs for $\ell = 0$, and 50 LTOs for the other ℓ 's. The largest ℓ value used in these calculations was $\ell = 8$. The CI energies are given in Table I, and were extrapolated to the $\ell = \infty$ limit using a procedure described shortly. They agree with the E_{QHQ} energies to within 10^{-5} a.u..

The $e^+\text{He}$ CI basis was constructed by letting the two electrons and the positron form all of the possible configurations with a fixed electron-electron spin (S_e), total angular momentum (L_T), and total wavefunction parity (π), subject to the further selection rules, $\max(\ell_0, \ell_1, \ell_2) \leq J$, and $\min(\ell_1, \ell_2) \leq L_{\text{int}}$, and $(-1)^{(\ell_0+\ell_1+\ell_2)} = -1^\pi$. In these rules ℓ_0 , ℓ_1 and ℓ_2 are respectively the orbital angular momenta of the positron and the two electrons, with a maximum single-particle orbital angular momentum of J . The number of LTOs for each ℓ was 15 with the exception of $\ell = 0, 1, 2$, and 3 where 18, 18, 17, and 16 LTOs were used. The parameter L_{int} was set to 4. The largest ℓ in the orbital space was $J = 12$ for the $e^+\text{He}(2s^2\ ^1\text{S}^e)$ state and $J = 9$ for the $e^+\text{He}(2s2p\ ^3\text{P}^o)$ state.

The main technical problem afflicting CI calculations of positron-atom interactions is the slow convergence of the energy with J [4, 34]. One way to determine the

$J \rightarrow \infty$ energy, $\langle E \rangle_\infty$, is to use an asymptotic analysis. It has been shown that successive increments, $\Delta E_J = \langle E \rangle_J - \langle E \rangle_{J-1}$, to the energy can be written as [34–36]:

$$\Delta E_J \approx \frac{A_E}{(J + \frac{1}{2})^4} + \frac{B_E}{(J + \frac{1}{2})^5} + \frac{C_E}{(J + \frac{1}{2})^6}. \quad (2)$$

The $J \rightarrow \infty$ limit, is determined by fitting sets of $\langle E \rangle_J$ values to Eq. (2). The coefficients, A_E , B_E and C_E are determined at a particular J from 4 successive energies ($\langle E \rangle_{J-3}$, $\langle E \rangle_{J-2}$, $\langle E \rangle_{J-1}$ and $\langle E \rangle_J$). Once the coefficients have been determined it is easy to obtain the $J \rightarrow \infty$ limit. Application of asymptotic series analysis to helium has resulted in CI calculations reproducing the ground state energy to an accuracy of 10^{-8} a.u. [36].

The CI energy of the $e^+\text{He}(2s^2)$ state (see Table II) was -0.795058 a.u.. Subtracting this from the $\text{He}(2s^2)$ E_{CI} of -0.778781 a.u. gives a binding energy of 0.016277 a.u.. This binding energy is an underestimate since the energy of the $\text{He}(2s^2)$ state in the CI basis used for the $e^+\text{He}$ calculation was -0.778771 a.u.. The $J \rightarrow \infty$ extrapolation contributed 10% to the binding energy. The $e^+\text{He}(2s^2)$ binding energy is only 4% smaller than the binding energy of the positron to the $\text{Mg}(3s^2)$ ground state, namely 0.01704 a.u. [28].

The $\text{He}(2s2p \ ^3P^o)$ state also binds a positron with a binding energy of 0.017870 a.u.. The surprisingly large binding energy is caused by the relatively small excitation energy of 0.051 a.u. from the $\text{He}(2s2p \ ^3P^o)$ state to the $\text{He}(2p^2 \ ^3P^e)$. This leads to the $\text{He}(2s2p \ ^3P^o)$ state having a static dipole polarizability of $157 a_0^3$. The $e^+\text{He}(2s2p \ ^3P^o)$ positron binding energy is larger than that of the $e^+\text{Be}(2s2p \ ^3P^o)$ state which is only 0.000087 a.u. [37].

The SVM was also used to determine the energy of the resonance state using the projection ansatz. The SVM diagonalizes the Hamiltonian in a basis of explicitly correlated gaussians (ECGs). The non-linear parameters of the ECG basis are optimized by a trial and error process. Such a process is possible since the ECG matrix elements of the Hamiltonian are very easy to compute. The diagonalization of $\hat{Q}\hat{H}\hat{Q}$ is approximated by adding an orthogonalizing pseudo-projector (OPP) [3, 38, 39], to the Hamiltonian to exclude the $\text{He}^+(1s)$ state from being occupied. The modified Hamiltonian is

$$\hat{H}_{\text{OPP}} = \hat{H} + \lambda \hat{P}_{\text{OPP}} \quad (3)$$

where λ is chosen to be a large positive number. The operator \hat{P}_{OPP} is defined as

$$\hat{P}_{\text{OPP}} = |\phi_{1s}(\mathbf{r}_1)\rangle\langle\phi_{1s}(\mathbf{r}_1)| + |\phi_{1s}(\mathbf{r}_2)\rangle\langle\phi_{1s}(\mathbf{r}_2)| \quad (4)$$

where $\phi_{1s}(\mathbf{r}_i)$ again refers to the $\text{He}^+(1s)$ state. Any part of the wavefunction with a non-zero overlap with the $\text{He}^+(1s)$ state tends to increase the energy. The energy minimization inherent to the SVM leads to a ground state wavefunction with a very small overlap with the $\text{He}^+(1s)$ state. The parameter λ was set to 10^6 a.u. for the present calculations. The $\text{He}^+(1s)$ state was expanded as a linear combination of 12 gaussians.

TABLE II: Calculated energies of some $e^+\text{He}$ states. The CI calculations are also given with a $J \rightarrow \infty$ correction as discussed in the text. The binding energies are denoted by ε .

State	Method	J	$\langle E \rangle_J$ (a.u.)	ε (a.u.)	ε (eV)
$e^+\text{He}(2s^2 \ ^1S^e)$	CI	12	-0.793537	0.014756	0.4015
	CI	∞	-0.795058	0.016277	0.4429
	SVM	—	-0.795210	0.016429	0.4471
$e^+\text{He}(2s2p \ ^3P^o)$	CI	9	-0.776306	0.014814	0.4031
	CI	∞	-0.779362	0.017869	0.4863
$e^+\text{He}(3s^2 \ ^1S^e)$	CI	12	-0.468860	—	—
	CI	∞	-0.481643	0.009420	0.2563

The SVM energy of the $\text{He}(2s^2)$ state was -0.778786 a.u., i.e. 5×10^{-6} a.u. below the CI energy. The dimension of the largest SVM calculation of the $e^+\text{He}(2s^2)$ state was 900 ECGs. The SVM binding energy of the positron to the $\text{He}(2s^2)$ state given in Table II was 0.016429 a.u. Examination of the convergence pattern suggests that the SVM energy is within 2×10^{-4} a.u. of the variational limit. The SVM and CI binding energies for this state are in excellent agreement when the respective uncertainties arising from finite size basis sets are taken into consideration.

The $e^+\text{He}(2s^2 \ ^1S^e)$ system is also likely to support a $^2P^o$ shape resonance just above the $\text{He}(2s^2 \ ^1S^e)$ threshold. This is based on the similarity of the He and Mg polarizabilities and the positron attachment energies in the $^2S^e$ channel. The $e^+\text{-Mg } ^2P^o$ shape resonance was located at 0.00351 a.u. above the elastic scattering threshold and had a width of 0.00396 a.u. [28].

It is likely that there will be an infinite series of resonances associated with the set of $\text{He}(ns^2)$ doubly excited states. An investigation of the $(m^{2+}, 2e^-, e^+)$ system revealed that this system remains bound when the mass $m^{2+} \rightarrow 0$ [40]. Decreasing the m^{2+} mass weakens the effective strength of the $m^{2+}-e^-$ interaction and provides an analogue of the $\text{He}^{2+}-ns(e^-)$ interaction. A first test was performed by a CI investigation of the $e^+\text{He}(3\ell, 3\ell')$ systems. In this case the single particle basis was orthogonalized to the $\text{He}^+(1s, 2s, 2p)$ states. The CI energy of the $\text{He}(3s^2)$ state is -0.354562 a.u. Since the removal energy of the electron with respect to the $\text{He}^+(3\ell)$ threshold, -0.132340 a.u., is less than the positronium ground state energy of -0.25 a.u., the threshold for attaching a positron to the $\text{He}(3s^2)$ state is at $-0.47222\bar{2}$ a.u..

The CI calculation for the $e^+\text{He}(3s^2)$ state gave an energy of -0.481643 a.u. The binding energy of this state is 0.009420 a.u. The stability of this system provides strong evidence for an infinite number of $e^+\text{He}(ns^2)$ type resonances. It is likely that the rich resonance structures of the PsH system [41] will be replicated for positron interactions with the doubly excited helium atoms.

Reference can be made to $e^-+\text{He}$ scattering experiments to give a first order estimate on the viability of experimental detection. A number of electron scattering

experiments have demonstrated electron attachment to the He doubly excited states [16–20]. Experiments that detect total cross sections involving ground state atoms and ions probably do not have a sufficiently large signal to background ratio to detect the $e^+\text{He}$ resonances. For example, He^+ ions were detected in the experiment of Quemener *et al.* [17]. There, the cross section for the creation of He^+ varied by only 1% over the width of the $\text{He}^-(2s^22p)$ resonance. Higher signal to background ratios have been achieved in $e^-\text{He}$ experiments that measured differential cross sections [18, 19].

Finally, the widths of the resonances and energy resolution of positron beams need to be considered. Modern trap-based positron beams can achieve a total energy resolution of about 40 meV [9, 42]. An indication of the resonance widths can be made by reference to the widths of their doubly excited parent states. The width of the $\text{He}(2s^2)$ state is $\Gamma = 123$ meV [26] and the $\text{He}(2s2p^3P^o)$ state is $\Gamma = 8.1$ meV [26]. The widths of He^- resonances

based on these parents, that of the $\text{He}^-(2s^22p^2P^o)$ state of 71 meV and that of the $\text{He}^-(2s2p^2^4P^o)$ state of 10.3 meV [43] are reflective of their two electron parents. We performed an SVM complex rotation calculation [44] by augmenting the ECG basis with additional functions representing the $e^+\text{He}$ and He^+Ps continuum. The energy shifted to -0.79484 a.u. and the width was 0.00249 a.u. (68 meV), which is large enough to detect with current positron beam technology. Previously known positron-atom resonances are either too narrow as in the case of hydrogen and sodium [45, 46], or involve atoms which do not naturally exist in gaseous form [15, 28, 46].

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