Configuration interaction calculations of positron binding to Be(\(^3\text{P}^0\))

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Abstract

The configuration interaction method is applied to investigate the possibility of positron binding to the metastable beryllium (1s\(^2\)2s2p \(^3\text{P}^0\)) state. The largest calculation obtained an estimated energy that was unstable by 0.00014 Hartree with respect to the Ps + Be\(^+\)(2s) lowest dissociation channel. It is likely that positron binding to parent states with non-zero angular momentum is inhibited by centrifugal barriers.

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1. Introduction

In the last several years there has been substantial progress studying the physics of positron binding to atoms. As late as 1997, there was no rigorous evidence for existence of positron–atom bound states [1], although the possibility of positron binding had been often invoked to explain the existence of very large annihilation rates for positrons annihilating in gases [2–6]. Towards the end of 1997, positron binding to atomic lithium was established in two independent calculations [7,8]. Following these initial studies, positron binding to a further 10 elements of the periodic table was demonstrated [9]. One of the features of these positronic atoms is that all the atoms binding a positron have ionization potentials close to 6.80 eV (the Ps binding energy) and the binding energies are generally larger for the atoms with their ionization energies closest to 6.80 eV.

A related question is whether positrons can form bound states with excited electronic states. There is one excited atomic state known to bind a positron, namely the metastable He(\(^3\text{S}^0\)) state [10,11]. The stability of e\(^+\)He(\(^3\text{S}^0\)) state was established with the stochastic variational method (SVM) [12,13,7] following an initial inconclusive study by Drachman et al. [14].

The answer to this question is relevant to positron–atom collision physics [15], since such states may manifest themselves as Feshbach resonances in the elastic or excitation cross sections. There has been a suggestion that the rich resonant structures prominent in electron–atom scattering [16] are largely absent from the positron–atom spectrum [17,9,15].

This work investigates the possibility of positron binding to the low-lying triplet odd state of neutral beryllium, i.e. the Be(1s\(^2\)2s2p \(^3\text{P}^0\)) system. This state has been identified as one of the most likely excited states to bind a positron [18,9]. This conjecture has largely been based upon the fact that its ionization potential (IP) of 6.598 eV [19] is quite close to the Ps binding energy. This IP is between that of magnesium and calcium, both of which bind a positron with a binding energy of \(\approx 0.4\) eV [9,20,21]. Furthermore, the dipole polarizability of Be(\(^3\text{P}^0\)) is \(\alpha_d = 39.02\alpha_0^3 [22]\), greater than that of ground state Be (\(\alpha_d = 37.7\alpha_0^3 [23]\)).

This metastable state warrants investigation for another reason; the parent atom has an angular momentum greater...
than zero with the loosely bound 2p electron carrying the angular momentum. The quantitative impact that the angular momenta has on the ability to bind a positron is largely unknown. Angular momentum coupling considerations suggests that the existence of a repulsive centrifugal barrier will act to inhibit positron binding by making it less energetically favorable to form a Ps-cluster. Either the electron–positron pair will be in a relative p-state with a smaller energy gain than the Ps(1s) state, or the total angular momentum of the Ps cluster relative to the nucleus will be greater than zero.

2. Details of the calculation

Since the ionization energy of the Be(2s2p3p0) system is just less than 6.80 eV, the threshold for a stable positron complex is that for the Be+(2s) + Ps(1s) dissociation channel. This energy was −0.9192086 Hartree for the present model potential. An energy level diagram showing the ground and low-lying metastable states of Be, and some negative ion and positronic ion energies are shown in Fig. 1.

The CI method as applied to positron–atomic systems with two valence electrons and a positron has been discussed previously [24,25,20], but a short description is worthwhile. The model Hamiltonian is initially based on a Hartree–Fock (HF) wave function for the neutral atom and the parameters of the core-polarization potentials are added to the potential field of the HF core SVM calculation [45]. The threshold for e+ binding to Be(3P0) is 0.91888 Hartree [27]. The exchange potential (\(V_{\text{ex}}\)) of the Be+(2s) + Ps(1s) dissociation channel. This energy was −1.0523 a0 e−1 with respect to the Be 2+ threshold. The one-body polarization potential (\(V_{\text{p1}}\)) was defined to be

\[
V_{\text{p1}}(r) = -\frac{x_d g^2(r)}{2r^4}.
\]

The factor \(x_d\) is the static dipole polarizability of the core and \(g^2(r)\) is a cutoff function designed to make the polarization potential finite at the origin. The same cutoff function has been adopted for both the positron and electrons. In this work, \(g^2(r)\) was defined to be

\[
g^2(r) = 1 - \exp(-\rho / \rho^0),
\]

where \(\rho\) is an adjustable cutoff parameter. The core dipole polarizability was set to 0.0523 a0 while \(\rho\) was set to 0.95 a0 [25]. The two-body polarization potential (\(V_{\text{p2}}\)) is defined as

\[
V_{\text{p2}}(r_i, r_j) = \frac{x_d}{r_i r_j} (r_i \cdot r_j) g(r_i) g(r_j).
\]

This model potential gives a Be(2s2p3p0) binding energy of 0.91147 Hartree with respect to the Be2+ threshold. The experimental binding energy with respect to this threshold is 0.91888 Hartree [27].

The CI basis was constructed by letting the two electrons and the positron form all the possible total angular momentum \(L_T = 1\) configurations, with the two electrons in a spin-triplet state, subject to the selection rules, namely

\[
\max(\ell_\text{e}, \ell_1, \ell_2) \leq L_{\text{max}},
\]

\[
\min(\ell_1, \ell_2) \leq L_{\text{int}},
\]

\[
(-1)^{(\ell_0 + \ell_1 + \ell_2)} = -1.
\]

In these rules \(\ell_0, \ell_1\) and \(\ell_2\) are respectively the orbital angular momenta of the positron and the two electrons.

Our two-electron–positron calculations with non-zero total angular momentum were first validated against the previous \(L_T = 1\) and 2 PsH calculations of Tachikawa [28]. Using their Gaussian-type orbitals we reproduced their reported energy and annihilation rates. Note that the PsH states with \(L_T = 1\) and 2 are unbound [29,28,30].
For the e⁺Be⁺(1P⁰) calculations, the Hamiltonian was diagonalized in a CI basis constructed from a very large number of single particle orbitals, including orbitals up to \( \ell = 12 \). There was a minimum of 12 radial basis functions for each \( \ell \). The largest calculation was performed with \( L_{\text{max}} = 12 \) and \( L_{\text{int}} = 3 \) and gave a CI basis dimension of 498,750. The resulting Hamiltonian matrix was diagonalized with the Davidson algorithm [31], and a total of 1376 iterations were required for the largest calculation.

### 3. Results

Various e⁺Be⁺(1P⁰) expectation values, such as energy and mean distance of the electron and positron from the nucleus are given in Table 1. The 2\( \gamma \) annihilation rate [9] for annihilation with the core and valence electrons are denoted \( \Gamma_c \) and \( \Gamma_v \), respectively. The calculations shown in Table 1 have a minimum of 12 radial basis functions for each \( \ell \) with \( L_{\text{max}} = 12 \) and \( L_{\text{int}} = 3 \). The largest calculation remains unbound by \(-0.0021904\) Hartree.

To demonstrate that \( L_{\text{int}} = 3 \) is sufficient for this system, also shown in Table 1 is the result of a \( L_{\text{max}} = 12 \) and \( L_{\text{int}} = 2 \) calculation. The \( L_{\text{max}} \) parameter needs to be large since it determines the extent to which electron–positron correlations are incorporated into the wavefunction. However, \( L_{\text{int}} \) is largely concerned with electron–electron correlations and, for example, setting \( L_{\text{int}} = 3 \) for the PsH and e⁺Be ground states recovered 99.4% and 98.5%, respectively of the Ps and positron binding energies for a given \( L_{\text{max}} [25] \).

The main problem afflicting CI calculations of positron–atom interactions is the slow convergence of the expectation values with \( L_{\text{max}} \). The largest calculation was performed with \( L_{\text{max}} = 12 \) and \( L_{\text{int}} = 12 \), and \( 1376 \) iterations were required for the largest calculation. This can be seen in Table 1, where the power law factor \( p_E \) relating successive \( E(L_{\text{max}} - 2) \), \( E(L_{\text{max}} - 1) \) and \( E(L_{\text{max}}) \) are tabulated. Applying a simple power law extrapolation with \( p_E = 2.078 \) leads to a prediction of binding \((E = -0.9207021)\), but not much credence should be placed in this since fixing \( p_E \) at its \( L_{\text{max}} = 12 \) value of 2.078 will result in an extrapolation that will overestimate the contribution of the orbitals with \( L > L_{\text{max}} [39,34] \).

An improved method, borrowing from existing work in atomic structure physics [40,41,21], is to estimate the \( L_{\text{max}} \to \infty \) limit for the energy by assuming that

\[
\Delta E_L \approx \frac{A_E}{(L + \frac{1}{2})^5} + \frac{B_E}{(L + \frac{1}{2})^3} + \frac{C_E}{(L + \frac{1}{2})^2}.
\]

The factors \( A_E, B_E \) and \( C_E \) are determined from four calculations at successively larger values of \( L_{\text{max}} \). Applying Eq. (8) to the data in Table 1 at \( L_{\text{max}} = 12 \) one finds \( A_E = -34.601 \), \( B_E = 477.71 \) and \( C_E = -1829.3 \), with \( E = -0.919070 \) Hartree as our present best energy estimate. This is 0.0001390 Hartree higher than the threshold for binding at \(-0.9192086\) Hartree.

Usage of an inverse power series restricted to the first two-terms of Eq. (8) results in \( A_E = -21.875 \) and \( B_E = 172.29 \), and an e⁺Be⁺(1P⁰) energy of \( E = -0.9188267 \), which is also unbound. As has been seen in other CI calculations of positron–atom systems, even when the three-term and the two-term coefficients are significantly different, their extrapolated energies can still lie relatively close together [34].

### Table 1

Results of CI calculations for e⁺Be⁺(1P⁰) for a series of \( L_{\text{max}} \) with fixed \( L_{\text{int}} = 3 \) (the row denoted 12⁺ was computed with \( L_{\text{int}} = 2 \)).

<table>
<thead>
<tr>
<th>( L_{\text{max}} )</th>
<th>( N )</th>
<th>( N_{C1} )</th>
<th>( \varepsilon )</th>
<th>( \langle r_c \rangle )</th>
<th>( \langle r_v \rangle )</th>
<th>( \langle r_v^2 \rangle )</th>
<th>( \Gamma_c )</th>
<th>( \Gamma_v )</th>
<th>( p_E )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>32</td>
<td>11</td>
<td>-0.9049669</td>
<td>-0.014217</td>
<td>2.70529</td>
<td>31.8236</td>
<td>11.59699</td>
<td>0.00020</td>
<td>0.000521</td>
</tr>
<tr>
<td>2</td>
<td>45</td>
<td>32</td>
<td>-0.9105271</td>
<td>-0.0086815</td>
<td>2.70601</td>
<td>27.4961</td>
<td>920.4328</td>
<td>0.000087</td>
<td>0.004302</td>
</tr>
<tr>
<td>3</td>
<td>57</td>
<td>65</td>
<td>-0.9112555</td>
<td>-0.0079531</td>
<td>2.72410</td>
<td>21.5600</td>
<td>619.5881</td>
<td>0.000284</td>
<td>0.020168</td>
</tr>
<tr>
<td>4</td>
<td>69</td>
<td>108</td>
<td>-0.9120770</td>
<td>-0.0071316</td>
<td>2.76099</td>
<td>16.3218</td>
<td>378.4203</td>
<td>0.000571</td>
<td>0.053933</td>
</tr>
<tr>
<td>5</td>
<td>81</td>
<td>153</td>
<td>-0.9129784</td>
<td>-0.0062302</td>
<td>2.80833</td>
<td>13.2441</td>
<td>250.9955</td>
<td>0.000816</td>
<td>0.097421</td>
</tr>
<tr>
<td>6</td>
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<td>-0.9138427</td>
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<td>2.85800</td>
<td>11.6135</td>
<td>189.6602</td>
<td>0.000977</td>
<td>0.141967</td>
</tr>
<tr>
<td>7</td>
<td>105</td>
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<td>-0.9146093</td>
<td>-0.0045993</td>
<td>2.90693</td>
<td>10.6898</td>
<td>157.1977</td>
<td>0.001076</td>
<td>0.184291</td>
</tr>
<tr>
<td>8</td>
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<td>300</td>
<td>-0.9152671</td>
<td>-0.0039415</td>
<td>2.95389</td>
<td>10.1253</td>
<td>138.1528</td>
<td>0.001133</td>
<td>0.232404</td>
</tr>
<tr>
<td>9</td>
<td>129</td>
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<td>-0.9158238</td>
<td>-0.0033848</td>
<td>2.99833</td>
<td>9.75778</td>
<td>125.9701</td>
<td>0.001166</td>
<td>0.259160</td>
</tr>
<tr>
<td>10</td>
<td>141</td>
<td>399</td>
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<td>-0.0029160</td>
<td>3.03992</td>
<td>9.50665</td>
<td>117.6386</td>
<td>0.001183</td>
<td>0.291698</td>
</tr>
<tr>
<td>11</td>
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<td>449</td>
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</tr>
<tr>
<td>12</td>
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<td>-0.0021904</td>
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<td>9.20197</td>
<td>107.2937</td>
<td>0.001193</td>
<td>0.347951</td>
</tr>
<tr>
<td>12⁺</td>
<td>165</td>
<td>334</td>
<td>-0.9169903</td>
<td>-0.0022183</td>
<td>3.11398</td>
<td>9.20391</td>
<td>107.3440</td>
<td>0.001192</td>
<td>0.347880</td>
</tr>
</tbody>
</table>

\( \infty \)

\( \approx 0.9190697 \) is identical and denoted by \( N \). The three-body energy of the e⁺Be⁺(1P⁰) system, relative to the energy of the Be⁺⁺ core, is denoted by \( E \) (in Hartree). The threshold for binding is \(-0.9192086\) Hartree, and \( \varepsilon \) gives binding energy (in Hartree) against dissociation into Ps + Be⁺(2s). The mean electron–nucleus distance \( \langle r_c \rangle \), the mean positron–nucleus distance \( \langle r_v \rangle \), and the mean electron–positron separation \( \langle r_v^2 \rangle \) are given in units of \( a_0 \). The \( \Gamma_c \) and \( \Gamma_v \) columns give the valence and core annihilation rates, respectively (in units of \( 10^7 \) s⁻¹). The \( p_E \) column gives the power-series exponents from the \( L_{\text{max}} \to L_{\text{max}} - 1 \), \( L_{\text{max}} - 2 \) energies. The results in the row \( \infty \approx \) add Eqs. (8) and (9) to estimate the \( L_{\text{max}} \to \infty \) correction.
There are two aspects where convergence can be incomplete. Besides the size of $L_{\text{max}}$, the number of LTOs for each value of $\ell$ still needs to be further increased. This is best seen in Fig. 2 which depicts the energies as a function of $L_{\text{max}}$. Two sets of $e^+\text{Be}(^3\text{P}^0)$ data are shown, the first shows the $e^+\text{Be}(^3\text{P}^0)$ energies from Table 1 which used 12 LTOs, and their extrapolated estimates using Eq. (8). The second set shows the results from initial exploratory calculations also with $L_{\text{max}} = 12$ and $L_{\text{max}} = 3$, but only with a minimum of 8 LTOs per $\ell$. Even a cursory glance at Fig. 2 suggests that the CI prognosis for variationally establishing the existence of a bound state is not promising. Also shown in Fig. 2 are the $\text{Be}(^3\text{P}^0)$ energies using the underlying $e^+\text{Be}(^3\text{P}^0)$ basis without the positron (the 12 LTOs basis).

Even though the valence annihilation rate from the largest calculation is only $0.348 \times 10^9 \text{s}^{-1}$, the slow convergence of $\Gamma_v$ with $L_{\text{max}}$ suggests the presence of a well defined Ps cluster. Fitting the four largest $\Gamma_v$ calculations in Table 1 to the three-term form,

$$\Delta \Gamma_L \equiv \frac{A_L}{(L + \frac{1}{2})^2} + \frac{B_L}{(L + \frac{3}{2})^4} + \frac{C_L}{(L + \frac{5}{2})^6}, \quad (9)$$

results in an extrapolated annihilation rate of $\Gamma_v = 0.805 \times 10^9 \text{s}^{-1}$. However, this value should be regarded as only notional. After all, if this system is indeed unbound, $\Gamma_v$ should approach $\Gamma_{\text{Ps}} = 2.008 \times 10^9 \text{s}^{-1}$ in the limit of an infinite radial basis [42].

It is also noticed that $\langle r_p \rangle$ in Table 1 steadily decreases as $L_{\text{max}}$ increases. This also should not be taken as an indicator of binding since the behavior of $\langle r_p \rangle$ with $L_{\text{max}}$ is not straightforward. For example, the PsH $\langle r_p \rangle$ decreases when $L_{\text{max}}$ changes from 0 to 3, but then starts increasing as $L_{\text{max}}$ increases from 3 to 9 [25,34]. Indeed, this is a trend noticed in CI calculations of positronic atoms with IP < 6.8 eV (e.g. $^3\text{Li}$ [43], $^4\text{Ca}$ and $^4\text{Sr}$ [20,38]), which have shown an initial $\langle r_p \rangle$ decrease to a minimum, which then increases as $L_{\text{max}} \rightarrow \infty$. The behavior of $\langle r_p \rangle$ with $L_{\text{max}}$ for an unbound system is unknown.

4. Conclusions

A large-scale CI calculation of the $e^+\text{Be}(^3\text{P}^0)$ ground state has been performed. The largest calculation gave an energy that was 0.00219 Hartree above the threshold for binding. Using an extrapolation method to estimate the contribution from the higher-partial waves gave an energy that was 0.00014 Hartree above threshold. By its very nature the present calculation is unable to give definitive proof that the $e^+\text{Be}(^3\text{P}^0)$ system does not have a bound state. While a variational calculation of the present kind can be used to give proof of binding, it cannot be used to give proof of non-binding. A converged calculation of low energy $\text{Be}^–\text{Ps}(1s)$ scattering in the $^4\text{P}^0$ channel would be needed to establish lack of binding.

What can be inferred from the present calculation is that a $e^+\text{Be}(^3\text{P}^0)$ bound state (assuming one existed) would have a structure similar to positronic lithium, i.e. it would consist of a Ps-like object weakly bound to the $\text{Be}^+(2s)$ core. The size of the CI expansion required to establish binding would be enormous. For example, it is necessary to include orbitals with $L_{\text{max}} \approx 30$ to get an energy that is lower than the Ps + Li$^+$ threshold [43]. Another conclusion is that the existence of centrifugal barriers does seem to inhibit the binding of positrons to parent atoms with non-zero angular momentum.

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