Resonances of helium in a DC field

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Abstract
A saddle-point complex-rotation method is used to study electric-field effects on the doubly excited resonances of He. By using a 892-term wavefunction, a seven-angular-symmetry calculation (i.e. $L_{\text{max}} = 6$) is performed for a comprehensive study around the He (2, 6a) $^1P^o$ and (2, 6a) $^1D^e$ resonance pair in the presence of an external electric field. A general $M$-state sum rule is shown which suggests the sum of total energy or total width of $M$-state system is not affected by the presence of a weak static field. The $M$-state sum rule is used to investigate the sharing of resonance energies and widths for the locally isolated resonance groups around the He (2, 6a) $^1P^o$ and (2, 6a) $^1D^e$ resonance pair in the small-field region.

1. Introduction
Recently, the $B$-spline-based complex-rotation (BSCR) method by Fang and Ho [1] has been successfully applied to the study of the electric field effects on the autoionization states of He. Their work has stimulated some experimental activities [2] in investigating such a physical phenomenon. In the presence of external electric fields, theoretical calculations by Fang and Ho [1] show clearly that the cross section of the (2, 6a) $^1D^e$ resonance increases for increasing electric field strength, while the cross section for the (2, 6a) $^1P^o$ resonance decreases for increasing electric field strength. In addition to the sharing of cross sections, the electric field also affects the widths of the resonances. The width of both the (2, na) $^1P^o$ and $^1D^e$ resonances decreases initially when the field strength is turned on. There is a corresponding increase in the widths of other singlet states. The identification of those other singlet states was not included in the earlier investigation by Fang and Ho [1] since they have only focused their attention on the energy region very close to the (2, na) $^1P^o$ and $^1D^e$ resonance pair. In the energy region outside the (2, na) $^1P^o$ and $^1D^e$ resonance pair, there are a large number of other $^1L^\pi$ singlet states which are strongly mixed due to the presence of the external electric field. The investigation of those singlet states outside the energy region of the (2, na) $^1P^o$ and $^1D^e$ resonance pair is complicated and more involved. Although a 3680-term wavefunction...
was used by Fang and Ho [1], they are not sufficient for the purpose of including more angular symmetries or more extended energy regions in the calculation.

The purpose of this paper is to perform a more complete and comprehensive study for the behaviour of the resonances of helium in the presence of a static electric field. Experimentally, the available field intensity in the laboratory is of the order of $10^{-5}$ au. This field is too small to create an appreciable effect for low-energy resonances. For this reason, special attention will be given to the $(2, 6a)^{1}P^{o}$ and $^{1}D^{o}$ resonance pair because the required field strengths for the phenomenon predicted in our work are possible in present day laboratories. In this paper, instead of using the BSCR method, we use the well established saddle-point complex-rotation (SPCR) method [3, 4]. The advantage of the SPCR method is that only a smaller-size wavefunction (892 terms) is needed to produce highly accurate resonances for each symmetry. Figure 5 in Fang and Ho [1] is obtained with a wavefunction of 3680 configurations. To illustrate such an advantage in this work, the same four-symmetry (i.e. $L_{\text{max}} = 3$) SPCR calculation for the $(2, 6a)^{1}P^{o}$ and $^{1}D^{o}$ resonance pair only takes a basis size of 623 configurations. In addition, we find the convergence to be excellent for all electric fields tested. Highly accurate energies are also given by this wavefunction for other resonances.

Since we are interested in the study of $(2, 6)$ resonances, a more complete calculation should include angular momentum states of $^{1}G^{e}$, $^{1}H^{o}$ and $^{1}F^{o}$ symmetries in addition to the $^{1}S^{o}$, $^{1}P^{o}$, $^{1}D^{e}$, and $^{1}F^{o}$ states. Using the SPCR method allows us to carry out such a calculation with a reasonable size wavefunction. In this work, the complete seven-symmetry calculation is carried out with a 892-term basis function. More than 60 resonances are calculated. About 60 electric field strengths ranging from 0 to $7 \times 10^{-5}$ au are tested. In the next section, the theory will be given. The results and discussions will be given in section 3. Section 4 gives a brief summary.

2. Theory

In the presence of an external electric field, the Hamiltonian for a two-electron atom is [5]

$$H = H_{0} + V = H_{0} + \vec{F} \cdot (\vec{r}_{1} + \vec{r}_{2})$$

(1)

where $\vec{F}$ is a uniform external electric field. $H_{0}$ is the field-free Hamiltonian. Atomic units are used. The field strength in atomic units is $1 \text{ au} = 5.14 \times 10^{9} \text{ V cm}^{-1}$.

The computational procedures in a SPCR method are outlined in detail elsewhere [3, 4]. The main difference between the SPCR method and the usual complex-rotation method is that in the saddle-point wavefunction, the closed-channel segment and open-channel segment are well separated. A proper vacancy is built into the closed-channel segment explicitly. When complex scaling is carried out, the radial coordinates in $H$ given by equation (1) are not complex scaled as $r \rightarrow re^{i\theta}$. Instead, only the radial coordinate of the outgoing electron is complex scaled with an inverse rotation, i.e. for an $N$-particle system [4]:

$$\Psi(\theta) = \Phi_{ad}(R_{N}) + A \sum_{i} \psi_{i}(R_{N-1}) U_{i}(r_{N}e^{-i\theta})$$

(2)

where $U_{i}(r_{N})$ is the radial wavefunction for the outgoing electron.

In Bryant et al [6], a two-state sum rule is shown which suggests that the sum of total energy or total width of a two-state pair is not affected by the presence of a static field. In the following, we will show that this is a general property for an $M$-state system of arbitrary $M$. That is, given a set of basis functions, the total sum of all the complex eigenvalues $E_{i}$ is invariant with respect to the variation of static field strength. This explains why increasing the
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Figure 1. A four-symmetry calculation and comparison with Fang and Ho [1] for the He (2, 6a) \(^1\text{D}^e\) and (2, 6a) \(^1\text{D}^e\) resonance pair in a static field.

Figure 2. The energy spectrum from a saddle-point complex-rotation calculation in the field-free case. Seven angular symmetries and 892 terms are used in the wavefunction. Our calculated widths are very stable when the angle \(\theta\) is varied from 0.3 to 0.6 rad.

Auger width for a certain resonance is usually accompanied by a decreasing Auger width of other neighbouring resonances.
Let us choose an arbitrary set of basis functions. Using these basis functions, we can construct a secular equation for \( H_0 \). We diagonalize this secular equation and obtain a set of complex-eigenvalue \( E_i \) and corresponding eigenfunctions \( \phi_i \), \( i = 1, 2, \ldots, M^L \). \( L = 0, 1, 2, \ldots, l_{\text{max}} \). \( L \) represents the different angular symmetry states. Let \( \sum_L M^L = M \). Note that each of these \( \phi_i \) has a definite parity which is either even or odd.

Next, we use these \( \phi_i \) as basis functions to construct a secular equation for the total Hamilton with the static electric field:

\[
\begin{vmatrix}
    H_0^{(0)} - \Lambda & V_{01} & 0 & \cdots \\
    V_{10} & H_0^{(1)} - \Lambda & V_{12} & \cdots \\
    0 & V_{21} & H_0^{(2)} - \Lambda & \cdots \\
    \vdots & \vdots & \vdots & \ddots
\end{vmatrix}_{M \times M} = 0 \quad (3)
\]

where \( H_0^{(L)} - \Lambda \) is the block-diagonal matrix of the field-free Hamiltonian \( H_0 \) for the symmetry with angular momentum \( L \), i.e.

\[
H_0^{(L)} - \Lambda = \begin{pmatrix}
    E_1^{(L)} - \lambda & 0 & 0 & \cdots \\
    0 & E_2^{(L)} - \lambda & 0 & \cdots \\
    0 & 0 & E_3^{(L)} - \lambda & \cdots \\
    \vdots & \vdots & \vdots & \ddots
\end{pmatrix}_{M^L \times M^L} \quad (4)
\]

The block matrix \( V_{ij} \) is the coupling matrix between the two symmetries \( L_i \) and \( L_j \) due to the perturbation potential \( V \). Since \( V \) is an odd-parity operator, there is no \( \lambda \) dependence in \( V_{ij} \).
We can now expand equation (3) into an $M$th-order polynomial equation of $\lambda$. It is easy to see that in this polynomial the leading $\lambda$ terms are

$$(-)^M \lambda^M + (-)^{M-1} \sum_{i,L} E^{(L)}_i \lambda^{M-1} + \cdots.$$ 

If we have a $M$th-order polynomial equation:

$$\sum_{i=0}^M c_i \lambda^{M-i} = 0 \quad (5)$$

an elementary theorem suggests that the sum for the roots of this equation must be equal to $-c_1/c_0$. In the case of equation (3), this is just $-\sum_{i,L} E^{(L)}_i$ which is the total sum of negative eigenvalues before the field is turned on. Equation (3) suggests that this sum is not affected by the presence of the static field. The effect of this field is just to redistribute the real energy and width among the resonances. In practice, this redistribution is most apparent between the neighbouring states whose energies lie close together.
3. Results and discussion

In Fang and Ho [1], a 3680-term four-symmetry \((L_{\text{max}} = 3)\) BSCR calculation is carried out for the \((2, 6a) 1^S^e, 1^P^o\) and \(1^D^e\) resonance pair. As a first test of the compact wavefunction used in this work, we carry out a four-symmetry SPCR calculation using a 623-term wavefunction. The results are compared with those of [1]. Since this is a relatively small calculation, much smaller field increment steps are taken in this paper. It is interesting to find that the results from the 623-term wavefunction agree well with the result of Fang and Ho [1]. A comparison of the two calculations is given in figure 1. The Auger widths of these two states are very small. Even so, the agreement between the two calculations persists as the field varies from 0 to \(5.6 \times 10^{-5}\) au.

For a more complete study around the \((2, 6)\) resonances, we have carried out a seven-symmetry calculation. 892 terms are used in the wavefunction for the states with the \(1^S^e, 1^P^o, 1^D^e, 1^F^o\).
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Figure 7. The mixing and sharing of energy and width between the He resonances. (a) is the sum of (2, 6a) $^1$Se, $^1$Po, $^1$De, $^1$Fo, $^1$Ge, $^1$Ho, and (2, 7a) $^1$Se. (b) is the sum of (2, 5b) $^1$Se, (2, 6b) $^1$Se, $^1$Po, $^1$De, $^1$Fo, $^1$Ge, and (2, 7b) $^1$Po. (c) is the sum of (2, 5c) $^1$Po, (2, 6c) $^1$Po, $^1$De, $^1$Fo, and $^1$Ge. The resonance energy of the subgroup (a) $E^{(a)}$ is lowered by $-2.572$ au for a better presentation.

$^1$G*, $^1$H* and $^1$F* symmetries. For the field-free case, our calculated resonance energies and widths are all in excellent agreement with the existing accurate theoretical results. For S, P and D states, they agree closely with those of Chen [7], Ho [8] and Oza [9]. For G and F states, they agree well with those of Oza [10]. For example, the resonance energy and width for the (2, 6a) $^1$P* state are $E_r = -0.514722$ au and $\Gamma = 3.80 \times 10^{-5}$ au. They are $E_r = -0.51472$ au and $\Gamma = 3.8 \times 10^{-5}$ au in Oza [9]. The resonance energy and width for the (2, 6a) $^1$D* state are $E_r = -0.515439$ au and $\Gamma = 6.83 \times 10^{-5}$ au. They are $E_r = -0.515409$ au and $\Gamma = 6.8 \times 10^{-5}$ au in Oza [9].

In the SPCR calculation, the open and closed channels are well separated. This leads to solutions which can be well distinguished as either belonging to the open-channel continuum or to the closed-channel resonances. As an illustration, we show the energy spectrum in the complex energy plane with figure 2. The continuum states correspond to the $l$ se $\ell$ open channels rotated from the horizontal axis by twice the angle $\theta$ used in equation (2). The complex poles which are independent of $\theta$ and lying below the real axis are the resonances. The complex resonance energy, $E_r - i\Gamma/2$, appears to be on the horizontal axis because of the small width of these resonances. They are actually below the horizontal axis as seen in the expanded frame. These widths are very stable when the angle $\theta$ is varied from 0.3 to 0.6 rad.

Figure 3 shows the variations of the resonance energies $E_r$ in the presence of an external electric field in the energy region around the (2, 6a) $^1$P* and $^1$D* resonance pair for a seven-symmetry calculation. The full lines represent the resonance energies $E_r$, and the two broken lines on top and below each full line represent the energies $E_r + \Gamma/2$ and $E_r - \Gamma/2$, respectively. From this figure, the variations of the widths versus the static field for these resonances can be visualized directly. The spectra for the higher resonances in figure 3 lie close together. The enlarged scale spectra for these resonances are shown in figure 4.

As the external field increases, all the resonance energies and widths of the (2, na), (2, nb) and (2, nc) states are changed due to Starking mixing. Nevertheless, in the small-field region,
Figure 8. The energy and width of He (2, na), (2, nb) and (2, nc) resonances in a static field in the energy region around the (2, 5a) 1Po and 1De pair. The full curves are the real energy $E_r$, the broken curves are $E_r \pm \Gamma_1/2$. The upper figure gives the energy spectra of higher resonances on an expanded scale.

The sum rule still applies to those locally isolated resonance groups. For example, the resonance group consisted of (2, 5c) 1Po, (2, 5b) 1S, (2, 6b) 1P and (2, 6a) 1S states, is almost isolated from other resonances in the small-field region as shown in figure 3. If we separate this resonance group into three subgroups, i.e. subgroup (a) (2, 6a) 1S, subgroup (b) (2, 5b) 1S, (2, 6b) 1P and subgroup (c) (2, 5c) 1P, the sharing of resonance energies and widths between the subgroups (a) and (b) + (c) indeed verifies the sum rule in the small-field region up to $F = 2 \times 10^{-5}$ au. This is shown in figure 5. The resonance energy of the subgroup (a) $E_r^{(a)}$ is lowered by $-1.0361$ au for a better presentation. One can see that the sum of the four resonance energies or widths is essentially a constant as the field increases. A detailed breakdown of individual contributions to $\Gamma_1^{(a)}$, $\Gamma_1^{(b)}$ and $\Gamma_1^{(c)}$ is shown in figure 6. Similar results are also found in the resonance group consisting of (2, 6c) 1P, (2, 6b) 1S, (2, 7b) 1P and (2, 7a) 1S states in the small-field region up to about $F = 1 \times 10^{-5}$ au. It should be emphasized that our configuration designation, (2, nx) 1L, is only accurate for the field-free case. As the field increases, especially when an avoided crossing occurs, significant mixing may result. We still use the same terminology for these states mainly for convenience of discussion.
We now apply the sum rule to identify those singlet states which are responsible for the losing autoionization intensities of the (2, 6a) $^1P^0$ and $^1D^e$ resonance pair in the small-field region. For up to about $F = 1 \times 10^{-5}$ au, the resonances around the (2, 6a) $^1P^0$ and $^1D^e$ resonance pair can also be grouped into three subgroups (a), (b) and (c). Subgroup (a) consists of (2, 6a) $^1S^e$, $^1P^0$, $^1D^e$, $^1F^e$, $^1H^o$, $^1F$ and (2, 7a) $^1S^e$. Subgroup (b) includes (2, 5b) $^1S^e$, (2, 6b) $^1S^e$, $^1P^0$, $^1D^e$, $^1F^e$, $^1G^e$, $^1H^o$ and (2, 7b) $^1P^0$. And subgroup (c) consists of (2, 5c) $^1P^0$, (2, 6c) $^1P^0$, $^1D^e$, $^1F^e$ and $^1G^e$. The sum rule for the subgroups (a) and (b) + (c) is shown in figure 7. The resonance energy of the subgroup (a) $E_{(a)}^r$ is lowered by $-2.572$ au for a better presentation. Figure 7 clearly shows the losing autoionization intensity of the subgroup (a) to the other singlet states in subgroups (b) and (c).

We have focused our discussion on the (2, 6) states mainly because the field effects for the (2, 6a) $^1P^0$ and $^1D^e$ resonance pair are more prominent with laboratory-attainable field intensities. In contrast, the field effects are much weaker for the corresponding states in the (2, 4) and (2, 5) series. In figures 8 and 9 the spectra for these states are shown. It is clear from these figures that the DC field effects on the (2, 4a) $^1P^0$, (2, 4a) $^1D^e$, (2, 5a) $^1P^0$ and (2, 5a) $^1D^e$ states are very small.
4. Summary

In conclusion, we have shown a general $M$-state sum rule that the total resonance energy or total width is a constant in the presence of an external electric field. The effect of the electric field is just to redistribute the resonance energies and widths among the resonance states. In Fang and Ho [1], it is found that the autoionization widths of the helium $(2, 6a)^1P^o$ and $^1D^e$ resonance pair decreases as the field strength increases. In this work, we have carried out a seven-angular-symmetry calculation for helium and we have identified those singlet states which are sharing the autoionization intensity of the $(2, 6a)^1P^o$ and $^1D^e$ resonance pair in the small-field region. We hope that our data and findings may stimulate some new experimental activities in the near future.

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References