

## Strong Electric-Field Effects on the Doubly-Excited $2s^2\ ^1S^e$ , $2s2p\ ^1P^o$ and $2p^2\ ^1D^e$ States of He

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The method of complex-coordinate rotation is used to investigate electric-field effects on the doubly-excited  $2s2p\ ^1P^o$  and  $2p^2\ ^1D^e$  states of He, the two neighboring states with a field-free separation of 0.017 Ryd. Strong electric-field strengths up to  $F = 0.02$  Ryd are used in our present study. Products of Slater orbitals are used to represent the two-electron wave functions, with  $l_{max} = 8$  being employed for each individual electron. Block matrices with up to  $L_{max} = 6$  (I-states) are used to investigate the convergence behavior for the resonance parameters (resonance energy and width). When the external electric field is turned on, "classic" Stark effect is observed for the  $M=0$  components of these two neighboring states. Results for electric-field effects on the lowest lying  $2s^2\ ^1S^e$  state are also given. Comparisons are made with other calculations when available.

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### I. Introduction

This work presents a theoretical investigation of electric-field effects on the doubly-excited states of He below the  $\text{He}^+$  ( $N = 2$ ) threshold. The states under the present study are the  $2s^2\ ^1S^e$ ,  $2p^2\ ^1D^e$  and  $2s2p\ ^1P^o$  states. From the experimental side, the  $^1P^o$  states have been observed in laboratories using synchrotron radiation light sources [1,2]. Electric-field effects on such P-wave states have not been investigated, even though the experimental study of the field effects on their counterparts in  $\text{H}^-$  has a long history [3-7]. On the theoretical side, Nicolaides and Themelis [8] have recently carried out a calculation of DC field-effects on the He  $2s^2\ ^1S^e$  and  $2s2p\ ^3P^o$  states. In light of the theoretical and possible experimental interest, we now present a theoretical investigation of DC field-effect, on the doubly-excited  $2s^2\ ^1S^e$ ,  $2p^2\ ^1D^e$  and  $2s2p\ ^1P^o$  states of He. The method of complex-coordinate rotation [9-11] is used in the present study. This method was used by Reinhardt and coworkers to examine the Stark effect of hydrogen [12,13], and on the  $N = 2\ ^1P^o$  shape resonance in  $\text{H}^-$  [14]. Recently, we have carried out investigations of the electric-field effects on the doubly-excited states of  $\text{H}^-$  by using a method of complex-coordinate rotation [15-19].

## II. Theory and wave functions

The Hamiltonian of an atom in an external field is

$$H = T + V + \bar{F} \cdot \bar{r}, \quad (1)$$

where  $F$  is the external field, and  $T$  and  $V$  are the usual kinetic and potential operators, respectively. For the He system,  $T$  and  $V$  are given by

$$T = -\nabla_1^2 - \nabla_2^2, \quad (2)$$

and

$$V = -\frac{2Z}{r_1} - \frac{2Z}{r_2} + \frac{2}{r_{12}}, \quad (3)$$

where  $r_1$ , and  $r_2$  are the coordinates of the electrons with respect to the nucleus and  $r_{12} = |\bar{r}_1 - \bar{r}_2|$ .  $Z$  is the charge of the helium nucleus and  $Z = 2$ . Atomic units are used in our work with energy in Rydberg units. The field strength is in Rydberg units, and 1 Ryd =  $2.57 \times 10^9$  V/cm.

In the method of complex-coordinate rotation, the radial coordinates are transformed by

$$r \rightarrow r e^{i\theta}, \quad (4)$$

and the Hamiltonian can be written as

$$H(\theta) = T e^{-2i\theta} + V e^{-i\theta} + \bar{F} \cdot \bar{r} e^{i\theta} \quad (5)$$

Complex eigenvalues are obtained by diagonalizing the transformed Hamiltonian,

$$E = \langle \Phi | H(\theta) | \Phi \rangle / \langle \Phi | \Phi \rangle, \quad (6)$$

and the complex resonance energy is given by

$$E_{res} = E_r - i\Gamma/2, \quad (7)$$

where  $E_r$  gives the shifted energy position, and  $\Gamma$  the resonance width with which the Stark effect can be studied.

In the present work, we use products of Slater orbitals to represent the two-electron wave functions. The products of Slater orbitals are the following:

$$\Phi = A \sum_{la,lb} \sum_{ij} C_{a_i,b_j} \eta_{a_i}(r_1) \eta_{b_j}(r_2) Y_{la,lb}^{LM}(1,2) S(\sigma_1, \sigma_2) \quad (8)$$

where

$$\eta_{a_i}(r) = r^{n_{a_i}} \exp(-\xi_{a_i} r). \quad (9)$$

In equation (8),  $A$  is the antisymmetrising operator,  $S$  is a two-particle spin eigenfunction and the  $\eta$  are individual Slater orbitals.  $Y$  is the eigenfunction of the total angular momentum  $L$ ,

$$Y_{l_a, l_b}^{LM}(1, 2) = \sum_{m_{l_a}} \sum_{m_{l_b}} C(l_a, l_b, L; m_{l_a}, m_{l_b}, M) Y_{l_a, m_{l_a}}(1) Y_{l_b, m_{l_b}}(2) \quad (10)$$

with  $C$  being the Clebsch-Gordan coefficients.

### III. Calculations and results

For the individual electron we use Slater-type orbitals of 9 s-type, 8 p-type, 7 d-type, 6 f-type, 5 g-type, 4 h-type, 3 i-type, 2 k-type, and 1 l-type. The highest  $l$  value for the individual electron is  $l_{max} = 8$ . These orbitals would couple and form two-electron basis of 165, 240 and 316 terms for  $^1S^e$ ,  $^1P^o$  and  $^1D^e$  states, respectively. For the field-free case, we need to use basis functions that are capable of producing reasonably accurate resonance parameters of  $E_r$  and  $\Gamma$ . The resonance parameters for the  $2s2p^1P^o$  state obtained using such a basis set are  $E_r = -1.3860952$  Ryd and  $\Gamma = 0.0026165$  Ryd (see Table I). These are compared with results of  $E_r = -1.3862699$  Ryd and  $\Gamma = 0.0027466$  Ryd, that were obtained by using the method of complex-coordinate rotation with Hylleraas functions [20]. As for the  $2p^2^1D^e$  state, the use of wave functions constructed from Slater orbitals described above leads to a 316-term basis set. The energy of the  $2p^2^1D^e$  state is determined as  $E = -1.4037261$  Ryd and the width  $\Gamma = 0.0046230$  Ryd. They are compared with the complex-coordinate results of  $E_r = -1.4038914$  Ryd and  $\Gamma = 0.0047244$  Ryd [21]. In Table I we summarize the resonance parameters ( $E$ , and  $\Gamma$ ) for the states under the present investigation.

Using wave functions described above, we calculate the field effects on the  $M = 0$  components of the  $^1D^e$  and  $^1P^o$  states as the field strength is increased from zero. The energy levels for the  $2p^2^1D^e$  and the  $2s2p^1P^o$  state exhibit the "classic" Stark effect for these two neighbouring states, as they start to repel each other for increasing field strength. The results are shown in Table II and in Fig. 1. Resonance parameters for field strengths up to  $F = 0.02$  Ryd ( $\approx 51$  MV/cm) are calculated in our work. For high electric-field strengths, we employ up to the I-states ( $L_{max} = 6$ ) for field strengths greater than  $F = 0.015$  Ry. The number of bases for the  $^1S^e$ ,  $^1P^o$ ,  $^1D^e$ ,  $^1F^o$ ,  $^1G^e$ ,  $^1H^o$ , and  $^1I^e$  states are 165, 240, 316, 322, 332, 292, and 191 respectively. A total of 1858 terms are used for the high field cases. In general, when the field strength is increased, states with higher angular momentum would become more important since more channels are open by the external field. For the field strength of 0.020 Ryd, we also carry out calculations for  $L_{max} = 3, 4, 5$ , and 6. From such a test we estimate that the uncertainties in energies and widths are within 1%. The uncertainties are somewhat larger for stronger field strengths, and smaller for weaker field strengths. For practical purposes we have not extended the basis sets beyond  $L_{max} = 6$  in our present calculation. For electric-field strengths of  $F < 0.015$  Ryd, we employ angular momentum states of up to  $L_{max} = 5$ . From the convergence test we conclude that states beyond  $L_{max} = 5$  are not needed if the accuracy for the resonance parameter are set to within 1%.

TABLE 1. Doubly excited states of He below the  $N = 2$  He+ threshold.

Ref.	$2s^2 \ ^1S^e$	$2p^2 \ ^1D^e$	$2s2p \ ^1P^o$
		$E_r$ (Ryd)	
(a)	-1.5555753	-1.4037261	-1.3860953
(b)	-1.555736		
(c)	-1.55607	-1.40385	
(d)		-1.4038914	
(e)			-1.3862699
		$\Gamma$ (Ryd)	
(a)	0.0092338	0.0046230	0.0026165
(b)	0.00906		
(c)	0.009187	0.00536	
(d)		0.0047244	
(e)			0.0027466

(a) present calculations.

(b) Ho (1981, 1986): complex-coordinate calculations, Hylleraas functions [22,23].

(c) Bhatia and Temkin (1975): Feshbach projection method [24].

(d) Ho and Bhatia (1991): Complex-coordinate rotation, Hylleraas functions [21].

(e) Ho (1991): Complex -coordinate rotation, Hylleraas functions [20].

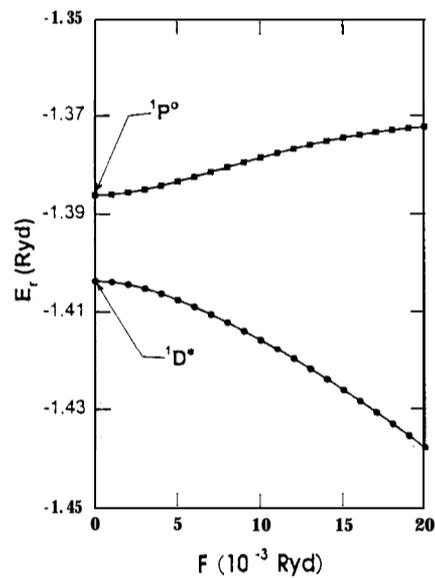


FIG. 1. Stark effect for the doubly-excited  $2p^2 \ ^1D^e$  and  $2s2p \ ^1P^o$  states

TABLE II. Electric-field effects on the  $2s2p\ ^1P^o$  and  $2p^2\ ^1D^e$  states.

$F(10^{-3}\ \text{Ryd})$	$2p^2\ ^1D^e$		$2s2p\ ^1P^o$	
	$E_r(\text{Ryd})$	$\Gamma(\text{Ryd})$	$E_r(\text{Ryd})$	$\Gamma(\text{Ryd})$
0	-1.4037261	0.0046230	-1.3860953	0.0026165
1	-1.4039040	0.0046011	-1.3859636	0.0026356
2	-1.4044223	0.0045411	-1.3855839	0.0026873
3	-1.4052416	0.0044559	-1.3849963	0.0027587
4	-1.4063123	0.0043587	-1.3842508	0.0028367
5	-1.4075870	0.0042590	-1.3833959	0.0029121
6	-1.4090262	0.0041619	-1.3824724	0.0029800
7	-1.4105996	0.0040697	-1.3815122	0.0030389
8	-1.4122846	0.0039831	-1.3805400	0.0030889
9	-1.4140645	0.0039021	-1.3795746	0.0031314
10	-1.4159274	0.0038267	-1.3786310	0.0031685
11	-1.4178644	0.0037570	-1.3777210	0.0032932
12	-1.4198690	0.0036937	-1.3768547	0.0032389
13	-1.4219366	0.0036375	-1.3760407	0.0032807
14	-1.4240636	0.0035899	-1.3752866	0.0033346
15	-1.4262475	0.0035529	-1.3745993	0.0034088
16	-1.4284864	0.0035291	-1.3739845	0.0035135
17	-1.4307787	0.0035217	-1.3734471	0.0036612
18	-1.4331234	0.0035348	-1.3729907	0.0038671
19	-1.4355194	0.0035729	-1.3726171	0.0041486
20	-1.4379657	0.0036416	-1.3723257	0.0045248

In Fig. 2 we show the movement of the resonance poles for the  $^1D^e$  and  $^1P^o$  states ( $M = 0$ ) when the external electric field strength is increased from zero. For the field-free case, the  $^1D^e$  state lies at a position of 0.017 Ryd lower than the  $^1P^o$  state. The width for the former state is 0.0046 Ryd, compared to 0.0026 Ryd for the latter state. When the external electric field is turned on, the interaction between these two resonant poles is evident, as they start to repel each other in the complex-energy plane. At the same time, the width for the  $^1D^e$  state decreases for increasing field strength, while the width for the  $^1P^o$  state increases when the field strength is increased. In Fig. 3 we show the change in widths as a function of the external electric-field strength  $F$ . It is clear that below  $F = 0.016$  Ryd approximately, the widths for these two states tend to average each other out, a kind of sharing the intensity for autoionization. This explains why the width for the  $^1D^e$  state decreases, but that for the  $^1P^o$  state increases for field strength up to about  $F = 0.016$  Ryd. It is further noted that when the external electric-field strength is greater than 0.016 Ryd approximately, the width for the  $^1D^e$  state starts to increase while

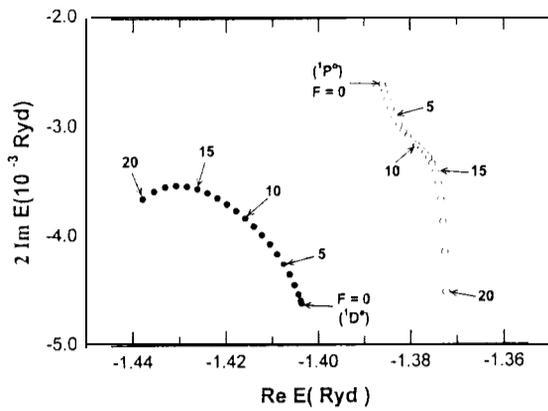


FIG. 2. Movements of the complex resonance poles for the  $2p^2\ ^1D^e$  and  $2s2p\ ^1P^o$  states ( $M = 0$ ) as the external electric-field strength changes. The closed circles are for the  $2p^2\ ^1D^e$  state and the open circles are for the  $2s2p\ ^1P^o$  state. Shown next to the selected poles are the field strength in units of  $10^{-3}$  Ryd. The poles are calculated in the increment of field strength of 0.001 Ryd.

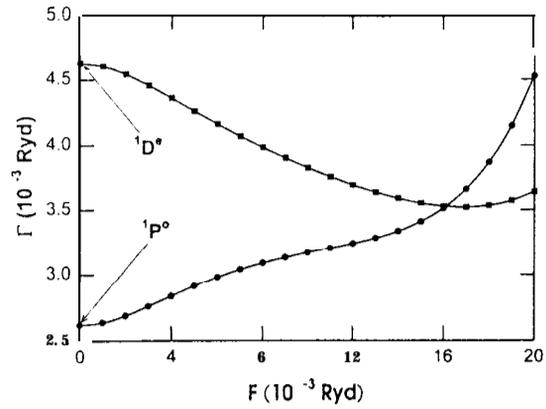


FIG. 3. Electric-field effects on the widths for the  $2p^2\ ^1D^e$  and  $2s2p\ ^1P^o$  states.

the width for the  $^1P^o$  state increases quite rapidly. At the high-field region, the effective potential barrier, formed by the combination of the atomic potential and the external DC field, would become narrower as the external electric-field strength is increased further. The time required for an electron to tunnel out of the potential barrier would therefore be shorter, resulting in the broadening of the autoionization width in the high-field region. In the present work, it is found that for  $F > 0.016$  Ryd approximately, the width for the (field-free)  $^1P^o$  state overtakes that for the (field-free)  $^1D^e$  state. Our present finding is very much similar to the field effects on the widths for the two nearly-degenerate ' $P$ ' (1) and  $^1S^e(2)$  states in  $H^-$  below the  $H$  ( $N = 2$ ) threshold [16]. They are also consistent with the analysis done by Bryant et al. [4] who used a perturbation treatment on two nearly-degenerate autoionization states in a model problem.

In this work we have also investigated the field effects on the lower-lying doubly excited  $2s^2\ ^1S^e$  state. The electric-field effect on this state was investigated recently by Nicolaides and Themelis [8] in which contributions up to the F-states ( $L_{max} = 3$ ) were included. In our present work we use a 231-term wave function to produce the field-free resonance parameters of  $E_r = -1.555753$  Ryd and  $\Gamma = 0.0092338$  Ryd. These are compared with  $E_r = -1.555736$  Ryd and  $\Gamma = 0.00906$  Ryd obtained by using Hylleraas functions [22,23]. Our results are shown in Table III. Fig. 4 shows the field effects on the width of the  $2s^2\ ^1S^e$  state, together with the results given by Nicolaides and Themelis [8] of which the field-free width of 0.00998 Ryd was used. It is seen that the field effects on both investigations

TABLE III. Electric-field effects on the  $2s^2 1S^e$  states.

$F(10^{-3} \text{ Ryd})$	Present		Nicolaides and Themelis [8]	
	$E_r(\text{Ryd})$	$\Gamma(\text{Ryd})$	$E_r(\text{Ryd})$	$\Gamma(\text{Ryd})$
0	-1.5555753	0.0092338	-1.555546	0.009980
1	-1.5555940	0.0092316		
2	-1.5556521	0.0092280	-1.555622	0.009972
3	-1.5557490	0.0092221		
4	-1.5558848	0.0092138	-1.555848	0.009960
5	-1.5560603	0.0092042		
6	-1.5562744	0.0091911	-1.556228	<b>0.009936</b>
7	-1.5565280	0.0091755		
8	-1.5568214	0.0091575	-1.556760	0.009900
9	-1.5571548	0.0091369		
10	-1.5575287	0.0091138	-1.557450	0.009856
11	-1.5579428	0.0090869		
12	-1.5583988	0.0090585	-1.558296	0.009800
13	-1.5588968	0.0090274		
14	-1.5594371	0.0089935	-1.559306	0.009732
15	-1.5600211	0.0089578		
16	-1.5606476	0.0089170	-1.560482	0.009652
17	-1.5613192	0.0088743		
18	-1.5620361	0.0088285	-1.561828	0.009564
19	-1.5627991	0.0087794		
20	-1.5636100	0.0087281	-1.563352	<b>0.009456</b>

show similar results. Both widths decrease for increasing field strength. Furthermore, our widths are consistently smaller than those of Nicolaides and Themelis [8] by approximately the same amount as that for the field-free case. Table III also shows a comparison between the field effects on our resonance position and that of Nicolides and Themelis [8]. It is seen that the agreement is quite good.

The decrease in width for the  $2s^2 1S^e$  state for increasing field strength in the low-field region is a result of interference coming from the higher-lying intershell  $1S^e$  states ( $2sns 1S^e$ ,  $2pnp 1S^e$ , etc., for  $n > 3$ ). The widths for the latter intershell states would be broadened while that for the former intrashell state is decreased. When the external electric field strength is sufficiently large, we expect the width for the  $2s^2 1S^e$  state would also be increased, as we have discussed such phenomenon for the high-field case earlier in the text. In the present work, we have not extended the field strength to beyond  $F = 0.02$  Ryd. Up to the maximum field strength being considered here, we have not yet found the width for the  $2s^2 1S^e$  state "turned around" to increase. For higher field strength, one

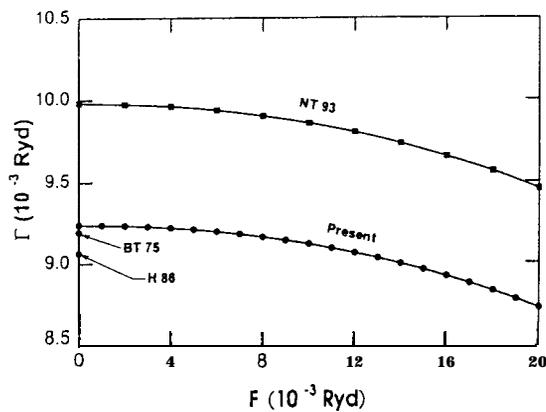


FIG. 4. Electric-field effects on the width for the  $2s^2 \ ^1S^e$  state. NT93 represents the results from Nicolaïdes and Themelis [8]. For the field-free widths, H86 is from Ho [23], BT75 is from Bhatia and Temkin [24].

would need to include more high-angular-momentum states to achieve convergence. To include additional high-angular-momentum states beyond  $L_{max} = 6$  is outside the scope of our present investigation. Nevertheless, we should point out that the field strength at which the width for the  $2s^2 \ ^1S^e$  state starts to increase, if it does increase at all, is an outstanding question, and should be addressed in the future.

In summary, we have employed the method of complex-coordinate rotation to investigate electric-field effects on the states of He below the  $\text{He}^+$  ( $N = 2$ ) threshold. We have observed Stark effect for the  $2s2p \ ^1P^o$  and  $2p^2 \ ^1D^e$  states. Results for field effects on the  $2s^2 \ ^1S^e$  state are also given. Our resonance parameters given as a function of electric-field strengths are useful references for future theoretical and experimental works.

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