

**3/3/1'3/1'' states of He<sup>-</sup>**

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Extensive complex-rotation calculations are carried out for eight He<sup>-</sup> 3/3/1'3/1'' states in order to understand the resonance structure of He<sup>-</sup> below the He 3s3p<sup>1</sup>P<sup>o</sup> threshold. It is found that the spectrum is very different from that of the He<sup>-</sup> 2/2/1'2/1'' states. A 3s-3p energy inversion is uncovered. Our result suggests that unlike the resonance structures below the n=2 region, 3s3p<sup>2</sup>S and 3s3p<sup>2</sup>P do form closed-channel resonances below the 3s3p<sup>3</sup>P<sup>o</sup> threshold. The results of this study are used to understand the scattering experiment of Roy [Phys. Rev. Lett. **38**, 1062 (1977)].

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The triply excited resonances of a three-electron atomic system were first reported in an electron-helium scattering experiment in 1965 [1]. In this experiment, only two triply excited resonance structures were observed. They were interpreted to be the 2s<sup>2</sup>2p<sup>2</sup>P<sup>o</sup> and 2s2p<sup>2</sup>D resonance by Fano and Cooper [2]. In the observed spectrum, the <sup>2</sup>P<sup>o</sup> structure is below the He 2s2s<sup>1</sup>S threshold but the <sup>2</sup>D structure is well above this threshold and extremely close to the 2s2p<sup>3</sup>P<sup>o</sup> threshold. This is very different from the triply excited resonances observed in recent hollow lithium experiments where both the 2s<sup>2</sup>2p<sup>2</sup>P<sup>o</sup> and 2s2p<sup>2</sup>D resonances are well below the Li<sup>+</sup> 2s2s<sup>1</sup>S threshold [3,4]. The work of Kuyatt *et al.* [1] and Fano and Cooper [2] has stimulated a great deal of interest in the study of these triply excited systems. Recent synchrotron radiation experiments have also reported the triply excited lithium resonances below the Li<sup>+</sup> 3/3/1' thresholds [5]. However, 35 years after the experiment of Kuyatt *et al.*, there are still relatively few experimental and theoretical studies on the He<sup>-</sup> resonances near the He 3/3/1' thresholds.

In 1977, Roy [6] reported the spectra in an electron-helium collision experiment which revealed several interesting resonancelike structures near the He 3/3/1' thresholds. He found a very steep valley at 68.83±0.04 eV which is followed by a sharp peak at 69.00±0.04 eV. He labeled them the 1 and 1' structures. Above 69 eV, the data points are not very clean but another broad dip which he labeled as 2 is seen around 69.67±0.04 eV. The He 3s3s<sup>1</sup>S threshold is at about 69.38 eV whereas the He 3s3p<sup>3</sup>P<sup>o</sup> threshold is at 69.47 eV. Hence both 1 and 1' are below these two thresholds while 2 is above the second threshold. Roy interpreted the 1 and 1' structure as a He<sup>-</sup> 3s<sup>2</sup>3p<sup>2</sup>P<sup>o</sup> resonance at 68.98±0.07 eV and 2 to be the 3s3p<sup>2</sup>D resonance.

For almost a quarter of a century, to my knowledge, no successful theoretical attempts have been made to interpret the spectra of Roy [6]. Recently, a state specific study was made by Nicolaides and Piangos on the n=3 triply excited He<sup>-</sup> resonances [7]. They used their results to identify the structures seen by Roy [6]. One limitation in this theoretical study is that the calculation is essentially a closed-channel study. The interaction of the closed and open channels is not

included. For triply excited three-electron systems, this interaction is particularly strong, causing a large width and possibly a significant shift between the resonance energy and closed-channel energy. For example, the width of Li 3s<sup>2</sup>3p<sup>2</sup>P<sup>o</sup> is found to be about 0.25±0.05 eV in the experiment and 0.282 eV from theory [5]. Similar results were also obtained by Azuma *et al.* [8]. Hence, it is quite conceivable that the width of He<sup>-</sup> 3s<sup>2</sup>3p<sup>2</sup>P<sup>o</sup> will also be very broad. Morishita and Lin studied the symmetries of n=3 triply excited three-electron systems [9], but they did not present energy and widths for these systems.

The large width of He<sup>-</sup> 3s<sup>2</sup>3p<sup>2</sup>P<sup>o</sup> may have several important implications in regards to the experimental results of Roy [6]. The structures 1 and 1' are reported to be about 0.17 eV apart. If the width of He<sup>-</sup> 3s<sup>2</sup>3p<sup>2</sup>P<sup>o</sup> is well over 0.2 eV, the likelihood that 1 and 1' are two parts of the same resonance will be greatly enhanced. On the other hand, if the width of this triply excited state is less than 0.1 eV, then it is highly unlikely that 1 and 1' belong to the same resonance state. Furthermore, 1 and 1' correspond to a dip and a peak in the observed spectrum. Nicolaides and Piangos [7] interpreted them to be the resonance positions of two triply excited resonances. For a very broad resonance, the true resonance position may differ significantly from either the peak or the dip position in the observed spectrum. Hence, knowing the width of these triply excited resonances is crucial in the understanding of the observed spectrum.

It is very challenging to calculate the widths for these 3/3/1'3/1'' states. There are an infinite number of open channels coupled to these states. A large number of open-channel continua interact strongly with these states. For example, any continuum of a two-electron target state with a 3s or 3p orbital may contribute significantly to the width. There are four such 1s3/1 states and 22 2/3/1' states. Each of these target states may correspond to either one or two open channels, depending on the symmetry. In addition, there could be some open channels of 3/3/1' which make a very large contribution to the width and shift.

The experimental results of Kuyatt *et al.* [1] revealed that there are very few 2/2/1'2/1'' closed-channel resonances below the He 2s2p<sup>3</sup>P<sup>o</sup> threshold. States such as 2s2p<sup>2</sup>S and 2p<sup>3</sup>P<sup>o</sup> are completely absent from the experiment. It would be very interesting to find out whether more closed-channel resonances are possible for the He<sup>-</sup> 3/3/1'3/1'' states.

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TABLE I. Helium target state energies ( $E$ ) and wave functions used in this work (in a.u.)  $N$  is the number of terms used in the wave function.  $M$  represents the  $M$ th lowest member of the resonance series.

$nl n' l' {}^S L^\pi(M)$	$N$	$E$ (this work)	Ref. [13]	Ref. [14]
$1s3s {}^1S(3)$	53	2.061 216		
$1s3s {}^3S(2)$	31	2.068 685		2.068 683
$1s3p {}^1P^o(2)$	56	2.055 122		
$1s3p {}^3P^o(2)$	52	2.058 023		2.058 058
$2s2s {}^1S(1)$	34	0.777 856	0.777 868	
$2p2p {}^1S(2)$	45	0.621 955	0.621 926	
$2s3s {}^1S(3)$	50	0.589 955	0.589 895	
$2p3p {}^1S(4)$	52	0.548 127	0.548 085	
$2p3p {}^1P(1)$	38	0.580 245	0.580 25	
$2s2p {}^1P^o(1)$	44	0.692 649	0.693 135	
$2s3p {}^1P^o(2)$	34	0.596 640	0.597 07	
$2p3s {}^1P^o(3)$	62	0.563 921	0.564 09	
$2p3d {}^1P^o(4)$	65	0.547 093	0.547 1	
$2p3d {}^1D^o(1)$	34	0.563 792	0.563 8	
$2p2p {}^1D(1)$	65	0.702 147	0.701 946	
$2p3p {}^1D(2)$	65	0.569 210	0.569 22	
$2s3d {}^1D(3)$	71	0.556 392	0.556 43	
$2p4p {}^1D(4)$	64	0.536 503		
$2p3d {}^1F^o(1)$	65	0.558 275	0.558 28	
$2s3s {}^3S(1)$	41	0.602 586	0.602 578	0.602 486
$2p3p {}^3S(2)$	49	0.559 744	0.559 747	0.558 841
$2p2p {}^3P(1)$	50	0.710 490	0.710 500	0.710 491
$2p3p {}^3P(2)$	40	0.567 799	0.567 81	0.567 728
$2s2p {}^3P^o(1)$	36	0.760 818	0.760 492	0.760 458
$2s3p {}^3P^o(2)$	77	0.584 812	0.584 67	0.584 580
$2p3s {}^3P^o(3)$	45	0.579 012	0.579 03	0.578 990
$2p3d {}^3P^o(4)$	59	0.548 832	0.548 84	0.548 813
$2p3d {}^3D^o(1)$	34	0.559 310	0.559 33	
$2p3p {}^3D(1)$	47	0.583 784	0.583 78	0.583 669
$2s3d {}^3D(2)$	50	0.560 611	0.560 69	0.560 198
$2p3d {}^3F^o(1)$	55	0.566 212	0.566 20	0.565 928
$3s3s {}^1S(1)$	51	0.353 505	0.353 54	
$3s3p {}^3P^o(1)$	61	0.350 992	0.350 38	

In this work, calculations will be carried out for  $\text{He}^-$   $3l3l'3l''$  resonances for both doublets and quartets. The method we use is the saddle-point complex-rotation method [10]. The saddle-point variation method is most efficient for obtaining a set of compact and optimized basis functions for closed-channel resonances. By including the open-channel continua via the complex-rotation method, the resonance parameters of the quantum systems can be calculated with square integrable basis functions. This method has proved to be highly accurate in predicting the resonance parameters for Li triply excited resonances [11,5].

In the saddle-point complex-rotation method, the resonance energy and width of interest are obtained by solving the complex secular equation from

$$\delta E = \frac{\langle \Psi | H_o | \Psi \rangle}{\langle \Psi | \Psi \rangle} \quad (1)$$

TABLE II. The energies, widths, and Auger branching ratios for  $3l3l'3l'' {}^4L$  states of  $\text{He}^-$ .  $N_{sdl}$  is the number of terms used in the closed-channel part of the wave functions and  $E_{sdl}$  is the corresponding energy.  $E_{res}$  is obtained by coupling the closed-channel segment with a large number of open channels.

	$3s3p^2 {}^4P$	$3p^3 {}^4S^o$	$3s3p3d {}^4F^o$
$N_{sdl}$	284	140	258
$-E_{sdl}$ (a.u.)	0.377 591	0.356 259	0.355 284
$-E_{res}$ (a.u.)	0.377 324	0.356 296	0.354 942
Width (meV)	154	172	258
$-E$ (Ref. [7]) (a.u.)			0.352 753
$nl n' l' {}^S L^\pi(M)$	Approximate Auger branching ratios (%)		
$1s3p {}^3P^o(2)$	0.25		
$2s3s {}^3S(1)$			0.12
$2p2p {}^3P(1)$		0.46	
$2p3p {}^3P(2)$	20.83	94.49	2.57
$2s3p {}^3P^o(2)$	75.82		17.03
$2p3s {}^3P^o(3)$	0.29		0.55
$2p3d {}^3P^o(4)$	0.20		1.51
$2p3d {}^3D^o(1)$	0.83	5.05	
$2p3p {}^3D(1)$	0.36		21.95
$2s3d {}^3D(2)$	1.40		22.65
$2p3d {}^3F^o(1)$			33.37

where  $H_o$  is the nonrelativistic Hamiltonian for  $\text{He}^-$  and  $\Psi$  is given by

$$\Psi = \Psi_{sdl} + A \sum_{o,i} \psi_o(1,2) \phi_{oi}(3). \quad (2)$$

$A$  is the antisymmetrization operator.  $\psi_o$  represents the two-electron open-channel target states and  $\phi_{oi}$  is the corresponding outgoing electron wave function which consists of a linear combination of Slater orbitals.  $LS$  coupling is adopted for these wave functions. In some cases, two  $\phi_{oi}$  of different angular momentum may be possible for each  $\psi_o$ . The  $\Psi_{sdl}$  are the closed-channel wave functions obtained from the saddle-point variation method. The basis functions in both  $\Psi_{sdl}$  and  $\psi_o$  are Slater orbitals. The complex scaling is carried out by inverse scaling for the radial coordinates in  $\phi_o$ . No complex scaling is done for  $H_o$ ,  $\Psi_{sdl}$ , or  $\psi_o$ .

The saddle-point variation method [12] gives a relatively compact wave function using the minimax procedure. In this calculation, we found three quartet resonances  $3s3p^2 {}^4P$ ,  $3p^3 {}^4S^o$ , and  $3s3p3d {}^4F^o$  all below the  $3s3p {}^3P^o$  threshold. They are closed-channel resonances. For the doublets, we found that  $3s^23p {}^2P^o$  and  $3s3p^2 {}^2D$  are below the  $3s3s {}^1S$  threshold, and  $3s3p^2 {}^2S$  and  $3s3p^2 {}^2P$  are below the  $3s3p {}^3P^o$  threshold. The  $3p^3 {}^2P^o$  is above  $3s3p {}^3P^o$  but below the  $3s3p {}^1P^o$  threshold. The energy obtained from the saddle-point method ( $E_{sdl}$ ) includes only the closed-channel effect. To find the actual resonance position, we need to consider the interaction from the open channels.

There are an infinite number of open channels associated with each  $\text{He}^-$   $3l3l'3l''$  state. Since  $\Psi_{sdl}$  couples with the

TABLE III. The energies, widths, and Auger branching ratios for  $3l3l'3l''\ ^2L$  states of He<sup>-</sup>.  $N_{sdl}$  is the number of terms used in the closed-channel part of the wave functions and  $E_{sdl}$  is the corresponding energy.  $E_{res}$  is obtained by coupling the closed-channel segment with a large number of open channels.  $E_T$  is the resonance energy above the helium  $1s1s$  ground state using 1 a.u.=27.207 67 eV.

	$3s^23p\ ^2P^o$	$3s3p^2\ ^2D$	$3s3p^2\ ^2P$	$3s3p^2\ ^2S$	$3p^3\ ^2P^o$
$N_{sdl}$	303	335	294	259	297
$-E_{sdl}$ (a.u.)	0.376 788	0.369 428	0.351 789	0.352 417	0.339 153
$-E_{res}$ (a.u.)	0.377 056	0.370 001	0.352 874	.353 726	0.344 829
Width (meV)	176	177	96	91	208
$E_T$ (eV)	68.74	68.93	69.40	69.38	69.62
$-E$ (Ref. [7]) (a.u.)	0.375 855	0.365 546		0.344 010	0.342 673
$nl'n'l'\ ^SL^\pi(M)$	Approximate Auger branching ratios(%)				
$1s3s\ ^1S(3)$	0.24				
$1s3s\ ^3S(2)$		0.54		.27	
$1s3p\ ^1P^o(2)$	0.37		0.23		
$1s3p\ ^3P^o(2)$	1.11	0.25		.29	0.39
$2s2s\ ^1S(1)$					
$2p2p\ ^1S(2)$	0.33			0.27	0.33
$2s3s\ ^1S(3)$	30.96	2.77		7.83	2.20
$2p3p\ ^1S(4)$	0.24			4.72	2.09
$2p3p\ ^1P(1)$			1.28		0.64
$2s2p\ ^1P^o(1)$	0.16		0.55	0.30	
$2s3p\ ^1P^o(2)$	2.31		1.13	1.14	0.20
$2p3s\ ^1P^o(3)$	6.03	7.64	27.16	15.35	1.63
$2p3d\ ^1P^o(4)$			1.03	.85	0.60
$2p3d\ ^1D^o(1)$			2.50		0.34
$2p2p\ ^1D(1)$					0.36
$2p3p\ ^1D(2)$	2.05	17.51	1.85		14.07
$2s3d\ ^1D(3)$	0.31	1.53	2.38	0.70	1.93
$2p4p\ ^1D(4)$		0.35			
$2p3d\ ^1F^o(1)$			0.39	0.36	0.19
$2s3s\ ^3S(1)$	1.63			1.54	
$2p3p\ ^3P(2)$	4.45	1.27	40.56	0.96	37.65
$2s2p\ ^3P^o(1)$					
$2s3p\ ^3P^o(2)$	43.25	56.17	12.32	57.40	1.74
$2p3s\ ^3P^o(3)$	0.45	0.72	1.26	4.88	0.22
$2p3d\ ^3P^o(4)$				0.38	1.19
$2p3d\ ^3D^o(1)$		2.09	6.48		0.61
$2p3p\ ^3D(1)$	5.00	6.50			3.53
$2s3d\ ^3D(2)$	0.94	1.73	0.56	2.65	
$2p3d\ ^3F^o(1)$		0.73			0.82
$3s3s\ ^1S(1)$					0.15
$3s3p\ ^3P^o(1)$					28.66

open-channel continua via the two-electron operators in  $H_o$ , only those  $\psi_o$  with at least one  $n=3$  orbital will interact strongly with  $\Psi_{sdl}$ . Hence, we consider most of the  $\psi_o$ 's with an  $n=3$  orbital. The  $\psi_o$  included in this calculation are tabulated in Table I. Due the large number of open channels, the complex-rotation calculation can be quite computation intensive. We try to use relatively compact but sufficiently accurate  $\psi_o$ 's. The number of terms in each  $\psi_o$  is given in Table I and the corresponding energy is compared with some of the theoretical results in the literature [13,14]. In a recent

He<sup>-</sup> photoionization calculation, highly accurate two-electron target states were used by Xi and Fischer [14]. The target states used in this work compare well with this reference (see Table I). We have also investigated the contributions from the  $2l2l'$  channels; they are usually small as expected. The  $3s3s\ ^1S$  and  $3s3p\ ^3P^o$  channels are also included in Table I for the study of  $3p^3\ ^2P^o$ .

There are 32 target states in Table I; some of these correspond to two outgoing channels. It is not feasible to include all of them in a complex-rotation calculation. Some of the

channel contributions are very small and they may be neglected in the final calculation. To find out which are the ones that may be neglected, a complex-rotation calculation for each open channel is carried out individually. Those channels with single-channel width result less than 1 or 2 meV are left out in the final calculation. For the final coupled calculation in this work, we use up to 12 target states and up to 17 open channels. The resonance energy and width results for the quartets are given in Table II and those for doublets are given in Table III.

When we compare the complex-rotation calculations for  $\text{He}^-$  with those for lithium, a significant difference is observed. For many of the triply excited lithium resonances [11,5], the width obtained from the fully coupled calculation is very close to the sum of the partial widths obtained from those calculations with one open channel at a time. Hence, the Auger branching ratios can be accurately predicted from the partial widths. In the case of  $\text{He}^-$ , we found that the fully coupled width result is no longer very close to the sum of the single-channel complex-rotation results. In general, the fully coupled results are smaller by about 6–20%. But for  $3p^3^2P^o$  and  $3s3p^2^2S$  the difference is even larger, indicating very significant interference between the different open channels. The reason for this is not entirely clear. We note that the energies of  $\text{He}$  two-electron target states lie much closer together than those of  $\text{Li}^+$  and the Auger energies of  $\text{He}^-$  are much smaller than those of lithium; both may have contributed to the stronger interference. For this reason, the Auger branching ratios of  $\text{He}^-$  are harder to predict with our complex-rotation method. Nevertheless, the relative magnitude of the widths from the single-channel complex-rotation calculation may show which are the important channels in Auger decay. For this reason, we have included these results in the tables as the “approximate branching ratios.” For those single-channel results that are less than 0.1% of the total sum, the branching ratios are omitted from these tables.

The results in Table II and III are quite interesting and unexpected. For example, the energy of  $2s2p^2^4P$  [15] is significantly higher than that of  $2s^22p^2P^o$  for  $\text{He}^-$ . But, for  $3s3p^2^4P$  and  $3s^23p^2P^o$ , the order is reversed. This is a  $3s$ - $3p$  energy inversion. It implies that for  $3s3p^3P^o$  the electron affinity of a  $3p$  electron is actually larger than that of a  $3s$  electron. To my knowledge, this is the first  $3s$ - $3p$  energy inversion found for a three-electron system. A similar  $2s$ - $2p$  inversion has been found in four-electron systems [16]. The physical reason for this inversion was discussed in Ref. [16].

For  $\text{He}^-$ , there is no evidence for the existence of closed-channel resonances  $2s2p^2^2P$  and  $2s2p^2^2S$ . In fact, the  $^2D$  structure observed is extremely close to the  $2s2p^3P^o$  threshold. In this work, we found that  $3s3p^2^2D$  is lower than the

$3s3p^3P^o$  threshold by about 0.54 eV and both  $3s3p^2^2S$  and  $3s3p^2^2P$  are below the  $3s3p^3P^o$  threshold, suggesting that these  $\text{He}^-$  closed-channel resonances exist.

In the saddle-point variation calculation for  $\Psi_{sdl}$ , we have avoided the inclusion of open-channel target states. For example, in the computation for  $3p^3^2P^o$ , we make sure that it is orthogonal to the  $3s3s^1S$  and  $3s3p^3P^o$  targets. The open channels are added via the complex-rotation procedure. In the case of this  $^2P^o$  resonance, the energies of these target states are extremely close to the resonance energy. The interaction between the closed-channel segment and the corresponding open channels is very strong. They cause a very large shift from  $E_{sdl}$  to the resonance energy  $E_{res}$ . This large shift in  $2p^3^2P^o$  is reflected in Table III.

It is very interesting to note that the widths of the two lowest doublet resonances  $3s^23p^2P^o$  and  $3s3p^2^2D$  are both about 176 meV, very close to the energy separation of 1 and 1' ( $0.17 \pm 0.04$  eV) in the experiment [6]. Furthermore, the resonance energy of  $^2P^o$  is lower than that of 1 by  $0.07 \pm 0.04$  eV and  $^2D$  is also lower than that of 1' by  $0.07 \pm 0.04$  eV. Clearly, what was seen in 1 and 1' is a superposition of the two resonances. The steep dip and rise of the cross section is the enhanced result of this superposition.

The experimental position of 2 in Roy [6] is at  $69.67 \pm 0.04$  eV. This is very close to the calculated energy of  $3p^3^2P^o$  predicted at 69.62 eV in this work. Nicolaides and Piangos [7] interpret this 2 to be the  $3p^3^2P^o$  resonance; our result supports their interpretation. The width of this resonance, 0.208 eV, is particularly large. This seems to agree with the very large dip observed in the experiment.

Reference [7] is the only other  $\text{He}^-$   $3/31'31''$  theoretical study available in the literature. While our result support their interpretation in regard to the experimental results of Roy [6], quantitative differences between the two theories are quite substantial. The comparisons are given in Table II and III. In some cases, even the nature of the resonance is different. For example, the result for  $3s3p^2^2S$  in this work is below the  $3s3p^3P^o$  threshold suggesting that it is a closed-channel resonance. On the other hand, this  $^2S$  in Nicolaides and Piangos [7] is higher than the  $3s3p^3P^o$  threshold by approximately 0.17 eV. The large discrepancy between the two calculations highlights the challenge of theoretical study on these triply excited systems.

In conclusion, we have made extensive complex-rotation calculations for eight closed-channel  $3/31'31''$  resonances of  $\text{He}^-$ . These results were used to understand the spectra observed in Roy [6]. We found that the resonance structure of the  $3/31'31''$  states is very different from that of  $2/21'21''$  states. A  $3s$ - $3p$  inversion is uncovered. We hope that these results will be helpful for future experimental analysis.

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