Doubly excited resonance structures in He photoionization from $1s2s\ ^{1,3}S$ metastable states

T. K. Fang and T. N. Chang
Department of Physics and Astronomy, University of Southern California, Los Angeles, California 90089-0484
(Received 28 October 1996; revised manuscript received 20 March 1997)

The resonant structures corresponding to selected He sp, $2n^{\pm}$ and $2pnd\ ^{1,3}P^o$ autoionization series below the He^+ $N=2$ threshold, following photoionization from $1s2s\ ^{1,3}S$ metastable states, are examined in detail. Theoretical photoionization cross sections, calculated using a B-spline-based configuration-interaction approach with a length-velocity agreement better than 1%, are presented. A sign change in Fano profile parameters q in $1s2s\ ^1S \rightarrow sp,2n^+\ ^1P$ transitions along the $2n^+$ autoionization series is identified and analyzed. [S1050-2947(97)05208-6]

PACS number(s): 32.80.Fb, 32.80.Dz, 32.70.Jz, 31.25.Jf

For an isolated resonance at energy E_r with a resonant width Γ, the square of the q parameter in the Fano formula [1] can be expressed in terms of the ratio of the peak photoionization cross section $\sigma_{\max}$ at an energy $E_{\max}=E_r+\Gamma/2q$ to the slowly varying background cross section σ_b, i.e.,

$$q^2+1=\sigma_{\max}/\sigma_b\,. \tag{1}$$

In general, the resonance profile of ground-state photoionization is fairly regular along the doubly excited autoionization series. As the effective principal quantum number ν increases, both the resonant width and transition probability (or the oscillator strength) decrease as a function of ν^{-3}. Since $\sigma_{\max}$ is proportional to the oscillator strength but inversely proportional to Γ [see, e.g., Eq. (46) of Ref. [2]], $\sigma_{\max}$ [and the q parameter according to Eq. (1)] is expected to approach a constant value [3]. The purpose of this paper is to present the results of a B-spline-based configuration-interaction (BSCI) calculation [4,5] that suggest that, instead, the photoionization structure profiles from the *bound excited states* could vary substantially as ν increases along the autoionization series. In particular, our calculation has identified a sign change in the Fano q parameters in the He $1s2s\ ^1S$ photoionization spectra along the $sp,2n^+\ ^1P$ autoionization series.

Similar to an earlier He photoionization calculation in the nonresonant region [6], the two-electron basis functions employed in the present calculation include products of all one-electron functions (i.e., the extended CC-CC *calculation* detailed in [6]) with all possible combinations involving both negative-energy *bound* orbitals and positive-energy *continuum* orbitals. The typical size of the extended BSCI basis in a CC calculation ranges from 7000 to 10 000. The Hamiltonian matrix is diagonalized using a two-step procedure that requires approximately half the size of the matrix [5,6].

For the $1s^2\ ^1S \rightarrow {}^1P$ photoionization, our calculated asymptotic q values of approximately -2.5 and -3.2 (see, e.g., Table I) for the $2n^+$ and $2n^-$ series, respectively, agree very well with the estimated observed values of -2.4 and -3.2 [3]. For the lowest 2^+ state, our calculated q value of -2.68 is also in agreement with the recently revised observed value of -2.75 ± 0.01 [3]. For the 3^- state, our q value of -4.19 is in close agreement with the value of -4.25 from the most recent complex-rotation calculation

TABLE I. Comparison between the calculated resonant energies E_r (in Ry), expressed in terms of the kinetic energy of the outgoing photoelectron, and the widths Γ (in $a[\mu]=a\times10^{\mu}$ Ry) of the He sp, $2n^{\pm}$, and $2pnd\ ^1P$ autoionization series below the $N=2$ threshold (derived from the energy variation of the scattering phase shifts) and the fitted values from the calculated photoionization spectra from the $1s^2\ ^1S$ ground state and the $1s2s\ ^1S$ metastable state. The fitted Fano q parameters and the background cross sections σ_b are also given.

	From phase shift		From $1s^2\ ^1S$				From $1s2s\ ^1S$			
State	E_r	Γ	E_r	Γ	q	σ_b	E_r	Γ	q	σ_b
2^+	2.614 031	2.73[−3]	2.614 046	2.72[−3]	−2.68	1.49	2.614 020	2.75[−3]	85.8	0.07
3^+	2.871 880	5.80[−4]	2.871 879	6.08[−4]	−2.58	1.26	2.871 880	6.05[−4]	−8	0.14
4^+	2.931 294	2.48[−4]	2.931 294	2.59[−4]	−2.52	1.24	2.931 289	2.59[−4]	0.02	0.09
5^+	2.957 000	1.21[−4]	2.957 004	1.27[−4]	−2.49	1.23	2.957 002	1.31[−4]	3.15	0.08
6^+	2.970 538	7.33[−5]	2.970 538	7.66[−5]	−2.48	1.23	2.970 538	7.62[−5]	4.74	0.07
3^-	2.805 853	7.57[−6]	2.805 853	7.69[−6]	−4.19	1.30	2.805 853	7.71[−6]	240	1.27
4^-	2.907 034	3.86[−6]	2.907 034	4.02[−6]	−3.25	1.25	2.907 034	4.02[−6]	137	0.41
5^-	2.945 415	1.78[−6]	2.945 415	1.87[−6]	−3.16	1.24	2.945 415	1.91[−6]	67.3	1.48
6^-	2.964 131	9.66[−7]	2.964 131	1.01[−6]	−3.21	1.23	2.964 131	1.04[−6]	18.3	19.6
$2p3d$	2.905 857	4.11[−8]	2.905 857	4.31[−8]	−16.8	1.25	2.905 857	4.31[−8]	185	37.0
$2p4d$	2.944 792	2.75[−9]	2.944 792	3.01[−9]	−34.8	1.24	2.944 792	3.02[−9]	741	7.49
$2p5d$	2.963 778	1.8[−10]	2.963 778	2.1[−10]	−55.2	1.23	2.963 778	2.1[−10]	801	14.1

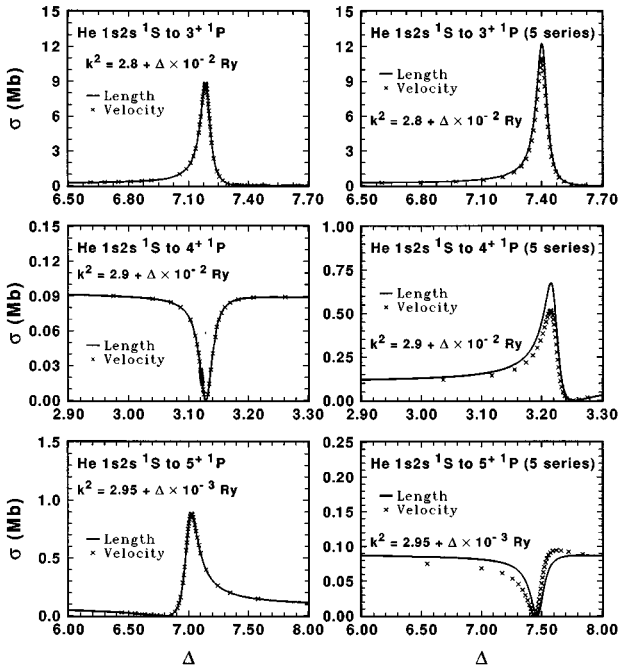

FIG. 1. Variation of the $1s2s\ ^1S \rightarrow ^1P$ photoionization resonance structures along the $sp,2n^+$ autoionization series. The plots on the right-hand side represent the results of a five-series BSCI calculation.

[3]. The theoretical q value of 3^- state is expected to be greater than the experimental one due to the smaller observed peak cross section as the actual resonance width is approximately two orders of magnitude smaller than the experimental energy resolution.

The resonant energy E_r and width Γ of the sp, $2n^{\pm}$, and $2pnd\ ^1P^o$ resonance series, derived directly from the energy variation of calculated scattering phase shifts, are listed in Table I. Also listed in Table I are values of E_r, Γ, q, and σ_b estimated from a fit of the calculated ground-state photoionization spectra of a set of N doubly excited resonances to a composite Fano formula

$$\sigma(E)=\left[\left(\sum_{\nu=1}^{N} g_\nu(E)\right)-(N-1)\right]\sigma_b(E). \tag{2}$$

The resonant spectral function

$$g_\nu(E)=\frac{(q_\nu+\epsilon_\nu)^2}{1+\epsilon_\nu^2} \tag{3}$$

is a function of the reduced energy $\epsilon_\nu(E)=(E-E_\nu)/\frac{1}{2}\Gamma_\nu$, where E_ν and Γ_ν are the resonant energy and width of a state ν. The resonant spectral function $g_\nu(E)\rightarrow 1$ at an energy E far from E_ν (or when $|E-E_\nu| \gg \Gamma_\nu$). Near a resonance μ, i.e., when $E\rightarrow E_\mu$, Eq. (2) reduces to the standard Fano formula as $\Sigma_{\nu=1}^{N} g_\nu(E)\rightarrow g_\mu+(N-1)$ and $\sigma(E)\rightarrow\sigma_b(q_\mu+\epsilon_\mu)^2/(1+\epsilon_\mu^2)$. As expected, the background cross sections σ_b (in Mb) for the $1s^2\ ^1S\rightarrow ^1P$ photoionization varies smoothly and can be parametrized by a function of photoelectron energy $E=k^2$ in Ry units, i.e.,

$$\sigma_b(E)=15.6616-9.5523E+1.5803E^2. \tag{4}$$

The small deviation of the fitted E_r and Γ from the theoretical values obtained from the phase shift variation may be attributed to the fact that only the *second-order* contribution to the interaction energy between the discrete and continuum is included in the derivation of Fano formula [1]. Our calculated ground-state photoionization spectra in the resonant region below the $N=2$ threshold are expressed in terms of Eqs. (2) and (4) with the values of E_r, Γ, and q listed in Table I. The length and velocity results agree to better than 1% at energies both away from and near the resonance peak,

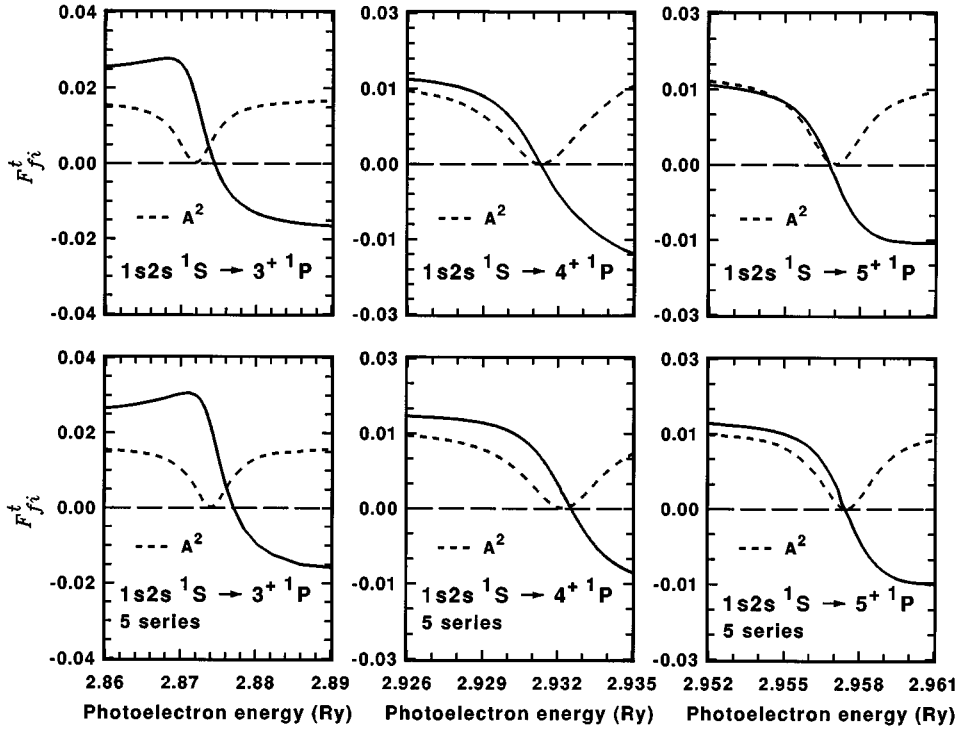

FIG. 2. Energy variation of the total transition amplitudes F^t_{fi} (solid curve) and the square of amplitude A (dotted curve) of the oscillating functions that represent the outgoing photoelectron from the $1s2s\ ^1S\rightarrow sp,2n^+\ ^1P$ photoionization. The results of the five-series calculation are also given for comparison.

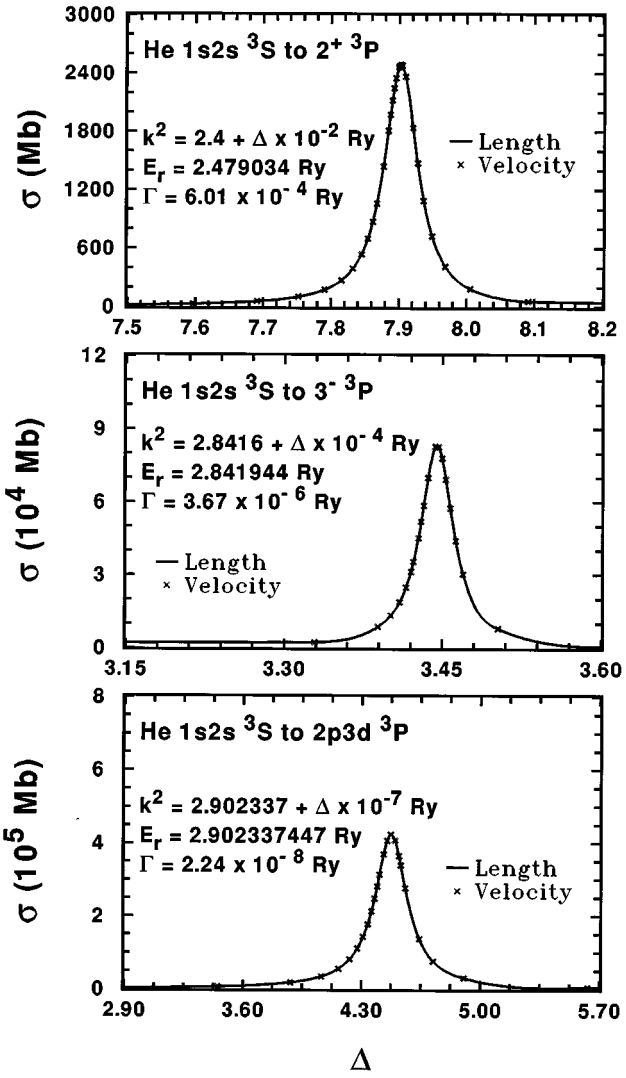

FIG. 3. Comparison between the length and velocity results of He $1s2s\ ^3S \rightarrow {}^3P$ photoionization cross sections σ of a few selected doubly excited resonances. The resonance energy E_r and width Γ are derived from the calculated scattering phase shifts.

even when the cross sections differ by several orders of magnitude across the resonance. A more comprehensive comparison of the existing theoretical and experimental data can be found elsewhere [3,7,8].

The experimental photoionization measurements from $1s2s\ ^{1,3}S$ metastable states were limited to the nonresonant region from the ionization threshold to about 2400 Å [9]. Our earlier qualitative calculation has shown already that the resonance profile of the doubly excited $sp,2n^+$ series varies significantly near the 3^+ and 4^+ states [7]. The results of the present calculation are listed in Table I in terms of a set of fitted E_r, Γ, q, and σ_b from the Fano formula for *each* resonance. The q parameter of the $sp,2n^+$ series changes from a large positive value for the 2^+ state to a negative value of -8 for the 3^+ state. It changes back to a positive value for the 5^+ state after it reaches a near zero value of 0.02 for the 4^+ state, resulting into a *window* resonance shown in Fig. 1. In addition, unlike the ground-state photoionization, σ_b from the $1s2s\ ^1S$ metastable state varies significant. To estimate quantitatively the effects of the configuration interaction on the resonance profiles, we have carried out an additional calculation with a smaller BSCI basis set, including only five configuration series in the BSCI basis, i.e., $1ss$, $2ss$, $2pp$, $3ss$, and $3pp$ in the initial state and $1sp$, $2sp$, $2ps$, $2pd$, and $3sp$ in the final state. The results of this second calculation are compared with the spectra obtained from our more refined calculation in Fig. 1. As expected, in addition to the substantial increase in the difference between the length and velocity results, the resonance energies are also shifted to the higher-energy side in the five-series calculation. Although the variation of the q parameters slows down slightly (i.e., the near-zero q value is shifted from the 4^+ state to the 5^+ state), the overall qualitative feature remains the same.

Except for the $1s2s\ ^1S \rightarrow sp,22^+\ ^1P$ photoionization, which is dominated by a one-electron $1s \rightarrow 2p$ excitation, all other $1s2s\ ^1S \rightarrow sp,2n^+\ ^1P$ transitions involve the change of orbitals of both electrons. As a result, the absolute cross sections for transitions to the 3^+ or higher states are substantially smaller than the ones to the 2^+ state. In fact, the total transition amplitude F^t_{fi} from an initial state i to a final state f is dominated by a direct $2s \rightarrow \epsilon p$ bound-continuum transition. Following the BSCI approach, σ is proportional to $|F^t_{fi}|^2$ and inversely proportional to the square of the amplitude A of the oscillating radial function that represents the outgoing ionized electron [see, e.g., Eq. (20) of Ref. [4]], i.e.,

$$\sigma \sim \frac{|F^t_{fi}|^2}{A^2}. \tag{5}$$

To better understand the variation of the resonance profile along the autoionization series, the total transition amplitudes F^t_{fi} for the $1s2s\ ^1S \rightarrow sp,2n^+\ ^1P$ photoionization (the solid curve) and A^2 (the dotted curve) are plotted against the photoelectron energy in Fig. 2. Also shown in Fig. 2 are F^t_{fi} and A^2 obtained in the five-series BSCI calculation. According to Eq. (5), the peak of the resonance coincides with the minimum in A^2, whereas the zero cross section is directly linked to the zero in F^t_{fi}. When the zero in F^t_{fi} is located on the higher-energy side of the minimum in A^2, the resonance structure is represented by a negative q parameter. In contrast, if the zero in F^t_{fi} is located on the lower-energy side of the minimum in A^2, the q parameter takes a positive value. The window resonance occurs as A^2 approaches its minimum at an energy where F^t_{fi} changes its sign. Although separately the features of both F^t_{fi} and A^2 do not vary significantly, our calculation clearly has shown that a small change in the relative position of the zeros in F^t_{fi} and the minima in A^2 is responsible for the strong variation in structure profile along the $sp,2n^+\ ^1P$ autoionization series. Also, in spite of the small effect due to the configuration interaction to F^t_{fi} and A^2 individually, the small change in the relative positions of F^t_{fi} and A^2 has shifted the window resonance from the 5^+ state to the 4^+ state.

Similar to the photoionization from the ground state, the resonance structure of the $1s2s\ ^3S \rightarrow {}^3P$ photoionization does not vary significantly along the autoionization series. The length-velocity agreement in the present calculation,

TABLE II. Comparison between the calculated resonant energies E_r (in Ry), expressed in terms of the kinetic energy of the outgoing photoelectron, and the widths Γ (in $a[\mu]=a\times10^{\mu}$ Ry) of the He sp, $2n^{\pm}$, and $2pnd\ ^3P$ autoionization series below the $N=2$ threshold (derived from the energy variation of the scattering phase shifts) and the fitted values from the calculated photoionization spectra from the $1s2s\ ^3S$ metastable state. The fitted Fano q parameters and the background cross sections σ_b are also given.

	From Phase shift		From $1s2s\ ^3S$			
State	E_r	Γ	E_r	Γ	q	σ_b
2^+	2.479 034	6.01[−4]	2.479 032	5.98[−4]	−124.8	0.160
3^+	2.830 659	1.58[−4]	2.830 658	1.65[−4]	−18.45	0.103
4^+	2.914 327	6.15[−5]	2.914 327	6.35[−5]	−36.72	0.098
5^+	2.948 577	2.89[−5]	2.948 577	2.94[−5]	−41.69	0.098
6^+	2.965 786	1.49[−5]	2.965 786	1.61[−5]	−43.79	0.097
3^-	2.841 944	3.67[−6]	2.841 944	3.80[−6]	−907.8	0.101
4^-	2.920 885	1.47[−6]	2.920 885	1.58[−6]	−691.3	0.098
5^-	2.952 108	8.02[−7]	2.952 108	8.14[−7]	−623.0	0.097
6^-	2.967 841	4.67[−7]	2.967 841	4.85[−7]	−580.3	0.097
$2p3d$	2.902 337	2.24[−8]	2.902 337	2.32[−8]	−2068	0.099
$2p4d$	2.942 736	9.93[−9]	2.942 736	1.03[−8]	−2157	0.098
$2p5d$	2.962 587	4.92[−9]	2.962 587	5.37[−9]	−2189	0.097

shown in Fig. 3 for a few selected resonances, is again better than 1% over the entire energy region. The resonant energy E_r and width Γ of the sp, $2n^{\pm}$, and $2pnd\ ^3P^o$ resonance series, derived from the energy variation of the calculated scattering phase shifts, are listed in the second and third columns in Table II. Also tabulated are the fitted values of E_r, Γ, q, and σ_b, using our calculated spectra of a set of N doubly excited resonances, to the composite Fano formula given by Eq. (2). The background cross sections σ_b (in Mb) varies smoothly and can be fitted to a function of kinetic energy $E=k^2$ in Ry units, i.e.,

$$\sigma_b(E)=2.3458-1.5110E+0.2538E^2. \tag{6}$$

The peak cross sections are generally very large, in part due to the relatively small resonance widths. The theoretical cross sections for the $1s2s\ ^3S\rightarrow{}^3P$ photoionization are expressed in terms of Eqs. (2) and (6) with the values of E_r, Γ, and q listed in Table II.

This work was supported by the NSF under Grant No. PHY94-13338.

[1] U. Fano, Phys. Rev. **124**, 1866 (1961).
[2] T. N. Chang, Phys. Rev. A **37**, 4090 (1988).
[3] M. Domke, K. Schulz, G. Remmers, G. Kaindl, and D. Wintgen, Phys. Rev. A **53**, 1424 (1996); M. Domke, G. Remmers, and G. Kaindl, Phys. Rev. Lett. **69**, 1171 (1992).
[4] T. N. Chang and X. Tang, Phys. Rev. A **44**, 232 (1991).
[5] T. N. Chang, in *Many-Body Theory of Atomic Structure and Photoionization*, edited by T. N. Chang (World Scientific, Singapore, 1993), p. 213.
[6] T. N. Chang and T. K. Fang, Phys. Rev. A **52**, 2638 (1995).
[7] T. N. Chang and M. Zhen, Phys. Rev. A **47**, 4849 (1993).
[8] T. N. Chang, T. K. Fang, Young Soon Kim, and Baek Il, Nam J. Korean Phys. Soc. (to be published).
[9] R. F. Stebbings, F. B. Dunning, F. K. Tittel, and R. D. Rundel, Phys. Rev. Lett. **30**, 815 (1973).