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Resonant width and energy determined by photoionization from excited divalent atoms

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Explicit examples are presented to illustrate the presence of a more symmetric resonant structure in spectra from bound excited states of a divalent atom. Comparison between the theoretical ground-state and bound excited-state photoelectron spectra shows that both the resonant energy E_r and the resonant width Γ of a doubly excited autoionization state of a divalent atom (e.g., alkaline-earth-metal atom) can be determined experimentally from photoionization originated from a bound excited state more effectively.

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The photoionization spectrum dominated by the doubly excited autoionization states due to the strong interaction between two outer electrons of a divalent atom (e.g., an alkaline-earth-metal atom) represents perhaps one of the most direct and unambiguous atomic processes for a detailed quantitative study of the many-electron effects in atomic transitions. Theoretically, the resonant structure of an *isolated* autoionization state located at resonant energy E_r can be described by the Fano formula [1] in terms of a set of resonant parameters, which includes the resonant width Γ, the asymmetry parameter q, and the nonresonant background cross section σ_b. The peak photoionization cross section $\sigma_{\max}$, located at $E_{\max}=E_r+\frac{1}{2}(\Gamma/q)$ according to the Fano formula, equals $\sigma_b(1+q^2)$, and the cross section is also expected to reach a zero at an energy $E_{\min}=E_r-\frac{1}{2}(\Gamma q)$. Physically, the resonant profile, which is determined by the asymmetry parameter q, measures qualitatively the interference between transitions from an initial state to the bound and continuum components of the final-state wave function. The q parameter is transition dependent, i.e., it varies significantly for transitions originating from different initial states. If the cross section is dominated by the contribution from the transition to the bound component of the final-state wave function, the q value is large and the resonant structure is nearly symmetric. For an intermediate q value, the resonant profile is generally asymmetric, which results from a comparable contribution from transitions to both bound and continuum components of the final-state wave function. If the contribution from the transition to the bound component is very small in comparison with the transition to the continuum background, q is very small and a zero cross section is expected either at or near E_r.

In contrast, the resonant width Γ, which dictates the decay rate or, physically, measures the interaction strength between the bound and continuum components of the state wave function of a doubly excited autoionization state, is *independent* of the transition process. For an autoionization resonance, which is dominated by a doubly excited $np\nu l'\,^{2S+1}L_J$ *bound* component embedded in a singly ionized $ns\epsilon l\,^{2S+1}L_J$ background *continuum*, the resonant width Γ is approximately proportional to $|\langle np\nu l'\,^{2S+1}L_J|(1/r_{12})|ns\epsilon l\,^{2S+1}L_J\rangle|^2$. Experimentally, the width of a doubly excited autoionization state can be determined by the resonant structures originating from any initial states. Except for a few recent experiments [2–4], most of the existing measurements are limited to the photoabsorption spectra from the ground state, which are almost always strongly asymmetric due to the

simultaneous change of electronic orbitals of two outer electrons in a double excitation process [5,6]. For a series of broad resonances with small q values, the standard theoretical interpretations [1,7] for an isolated resonance become less effective, especially when the broad resonance is also closely situated next to a second overlapping doubly excited series. As a result, an accurate experimental determination of Γ is often difficult if not impossible. For a narrow resonance, the width measurement could also be hampered by the lack of an adequate energy resolution.

The purpose of this paper is to demonstrate, based on detailed photoionization calculations for Be and the Be-like C^{2+} ion, that the resonant energies and widths can be determined more effectively from the photoelectron spectra of the excited atoms. Our theoretical calculations are carried out using a B-spline-based configuration-interaction procedure for the *continuum* (CIC) [8]. This CIC procedure has been applied successfully to the photoionization of two-electron atoms [9,10] and the single- and multiphoton ionization of the neutral Mg atom [11].

In Fig. 1 we compare the theoretical photoionization cross sections from the ground state and the bound excited $2s5s\ ^1S$ state of Be-like C^{2+} ions at the energy region dominated by the doubly excited $2pns\ ^1P$ and $2pnd\ ^1P$ autoionization series. In the ground-state photoelectron spectrum, the prominent asymmetric structures can be attributed unambiguously to the $2pnd\ ^1P$ autoionization series and the broad $2pns\ ^1P$ series exhibits a near zero cross section in the vicinity of the resonant energy E_r, which represents a Fano profile of very small q. Our calculation shows that the transition amplitude for the $2s^2\ ^1S \rightarrow 2pns\ ^1P$ photoionization is overwhelmingly dominated by the one-electron $2s^2 \rightarrow 2s\epsilon p$ *bound-to-continuum* transition, and the zero cross section results directly from the sign change of the dipole matrix at E_r due to the sign change of the effective radial function when the scattering phase shift increases rapidly by a total of π across the resonance. Figure 1 also shows that the $2pns\ ^1P$ resonant profile in the photoelectron spectrum from the $2s5s\ ^1S$ bound excited state represents a totally different type of transition, i.e., $2s \rightarrow 2p$, followed by a "shake-up" of the outer ns electron. This is similar to the process involving a two-electron excitation of the core electron studied elsewhere [4]. Since the contribution from the direct $2s5s \rightarrow 2s\epsilon p$ *bound-to-continuum* transition is small, the $2pns\ ^1P$ autoionization state is represented by a nearly symmetric resonant profile corresponding to a large q value.

A second, and perhaps a more elucidating example is shown in Fig. 2 for photoionization from the $2s^2\ ^1S$ ground state and the $2s3s\ ^1S$ bound excited state of Be. Experimentally, the absorption spectrum from the ground state of Be was first examined in detail by

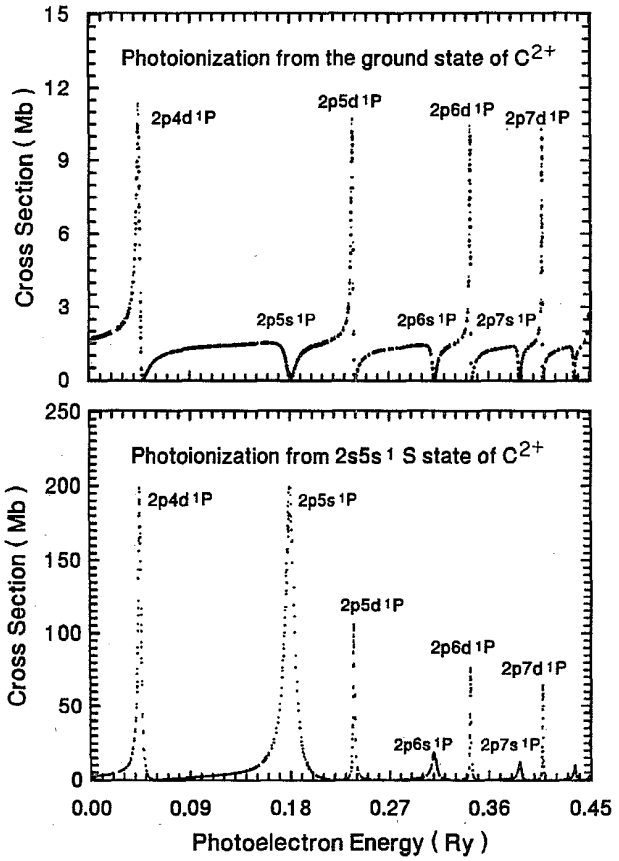

FIG. 1. Photoionization cross sections from the $2s^2\ ^1S$ ground state and the $2s5s\ ^1S$ bound excited state of the Be-like C^{2+} ion.

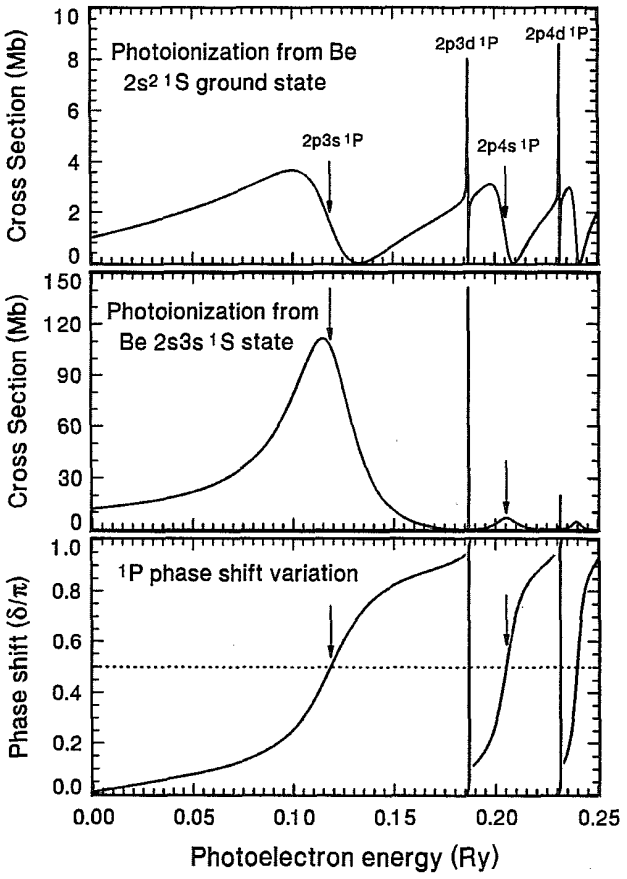

FIG. 2. Photoionization cross sections from the $2s^2\ ^1S$ ground state and the $2s3s\ ^1S$ bound excited state of the Be atom. The background scattering phase shifts are subtracted so that the resonant energy can be identified at a phase shift $\delta = \pi/2$.

TABLE I. The excitation energies (upper entry, in angstroms) of Be $2snl\ ^1P^o$ resonances from the $2s^2\ ^1S^e$ ground state. The theoretical resonance energy E_r is derived from phase shift variation and E_{peak} is the energy corresponding to the peak ground-state photoionization cross section shown in Fig. 2. The effective quantum numbers ν (lower entry) are estimated against the statistically weighted Be^+ $2p$ limited at 1.047 542 Ry [12].

	Theory		Observation		
State	E_r	E_{peak}	E_r[a]	Peak absorption[b]	Three-photon[c]
$2p3s\ ^1P$	1133.8	1158.7	1134	1158	
	2.408	2.294	2.407	2.299	
$2p4s\ ^1P$	1023.6	1030.9	1025	1036	
	3.411	3.287	3.387	3.217	
$2p5s\ ^1P$	985.4	987.9	986.2	989.2	
	4.410	4.299	4.377	4.264	
$2p6s\ ^1P$	967.4	968.9	967.8	970.0	
	5.404	5.293	5.374	5.216	
$2p3d\ ^1P$	1045.0	1045.0	1045.8	1045.2	1045.2
	3.098	3.098	3.088	3.096	3.096
$2p4d\ ^1P$	994.2	994.2	991.6	994.6	
	4.096	4.096	4.180	4.083	
$2p5d\ ^1P$	971.8	971.8	969.5	971.9	971.6
	5.095	5.095	5.250	5.090	5.110

[a]Esteva, Mehlman-Balloffet, and Romand [5].
[b]Mehlman-Balloffet and Esteva [6].
[c]Clark *et al.* [3].

Mehlman-Balloffet and Esteva [6] and Esteva, Mehlman-Balloffet, and Romand [5]. They have reported the resonant energies of the ^1P autoionization series both at the maximum absorption and at the maximum transmission. The narrow $2pnd\ ^1P$ series has also been identified in a three-photon resonant-ionization experiment by Clark *et al.* [3]. In the present calculation, the theoretical resonant energies E_r are derived from the variation of the scattering phase shift shown in Fig. 2. Table I shows a close agreement between our theoretical results and the observed resonant energies. The energies corresponding to the maximum ground-state photoionization cross sections shown in Fig. 2, i.e., E_{peak}, are also listed for comparison. The effective quantum numbers ν are estimated against the Be^+ $2p$ ionization limit. The resonant energies E_r for the narrow $2pnd\ ^1P$ series reported by Esteva, Mehlman-Balloffet, and Romand [5] are probably in error, judging from the unexpected large variation in their quantum defects and the close agreement between the three-photon and the peak absorption results. Since the estimated resonant widths for the $2pnd\ ^1P$ series are significantly smaller than the experimental energy resolution, the energy at peak absorption should represent the *most probable* energy for the narrow resonance. Our theoretical resonant energies, together with the *most probably* observed energies taken from Table I, are compared with other earlier theoretical results in Table II.

Our theoretical photoelectron spectrum from the ground state shown in Fig. 2 is in qualitative agreement with most of the earlier calculations [16–20] as well as

TABLE II. Comparison of theoretical excitation energies (in eV) of Be $2snl\ ^1P^o$ resonances from the $2s^2\ ^1S^e$ ground state with the *most probable* observed resonance energies E_r and E_{peak} taken from Table I.

	E^r						E_{peak}		
State	Expt.[a]	Present	Altick[b]	LV[c]	Lin[d]	Others	Expt.[a]	Present	CHC[e]
$2p3s\ ^1P$	10.93	10.93	10.77	10.70	10.49	10.97[f]	10.71	10.69	10.52
$2p4s\ ^1P$	12.10	12.11	12.07	12.05	11.99		11.97	12.01	11.98
$2p5s\ ^1P$	12.57	12.58	12.60	12.56			12.53	12.54	12.53
$2p6s\ ^1P$	12.81	12.82		12.81			12.78	12.79	12.79
$2p3d\ ^1P$	11.86	11.86	11.86	11.83	11.85	11.93[g]	11.86	11.86	11.92
$2p4d\ ^1P$	12.47	12.47	12.47	12.46	12.47	12.54[g]	12.47	12.47	12.50
$2p5d\ ^1P$	12.76	12.76					12.76	12.76	12.78

[a]Taken from Table I.
[b]Reference [13].
[c]Laughlin and Victor [14].
[d]Reference [15].
[e]Chi, Huang, and Cheng [16].
[f]Norcross and Seaton [7].
[g]Moccia and Spizzo [17].

TABLE III. The resonant energies (in eV) and widths Γ (in $a[b]=a\times10^{b}$ Ry) of selected Be $2snl\ ^1P^o$ resonances.

	E^r				Γ			
State	Expt.[a]	Present	Moores[b]	Serrao[c]	Present	Moores[b]	Serrao[c]	Others
$2p3s\ ^1P$	10.93	10.93	10.99	11.78	3.81[−2]	2.67[−2]	2.36[−2]	3.68[2][d]
$2p4s\ ^1P$	12.10	12.11	12.13	12.90	1.24[−3]	9.57[−3]	8.69[−3]	
$2p5s\ ^1P$	12.57	12.58	12.60	13.57	5.54[−3]	4.45[−3]	3.70[−3]	
$2p6s\ ^1P$	12.81	12.82			2.98[−3]			
$2p3d\ ^1P$	11.86	11.86	11.93		2.80[−5]	3.70[−6]		8.08[−6][e]
$2p4d\ ^1P$	12.47	12.47	12.52		2.05[−5[	2.20[−6]		1.00[−6][e]
$2p5d\ ^1P$	12.76	12.76			1.41[−5]			

[a]Taken from Table I.
[b]Reference [21].
[c]Reference [22].
[d]Norcross and Seaton [7].
[e]Moccia and Spizzo [17].

the observed absorption spectrum [5,6]. Both observed and calculated spectra exhibit a substantial overlap between the broad $2p(n+1)s\ ^1P$ and the narrow $2pnd\ ^1P$ series, in spite of a weak interaction strength between these two series [20]. A qualitative estimate, based exclusively on the observed absorption spectrum from the ground state, would yield a resonant width at least two to three times greater than the earlier theoretical estimates [7,21] for the broad $2pns\ ^1P$ resonances. This discrepancy is easily resolved when we compare the ground-state spectrum with the photoelectron spectrum from the bound excited $2s3s\ ^1S$ state shown in Fig. 2. In fact, the broad and the narrow series are completely separated with a substantially reduced estimated resonant width for the broad resonance. The large energy separations between the E_{peak} and E_r (indicated by the arrows in Fig. 2) also suggest a small q resonant profile. This is consistent with the $q\sim0$ resonant profile shown in Fig. 1 as the nuclear charge Z increases along the Be isoelectronic sequence. Our calculated resonant widths are compared with a few existing theoretical values in Table III.

In conclusion, the presence of a more symmetric resonant profile in spectra from bound excited states suggests that, experimentally, both Γ and E_r of a doubly excited autoionization state can be determined more conclusively from photoionization of excited atoms.

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