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Atomic-structure effects in multiphoton ionization of magnesium

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We present an application of a simple configuration-interaction approach to multiphoton ionization of magnesium, using a finite L^2-basis set constructed from a nearly complete set of one-particle frozen-core Hartree-Fock orbitals. The effects due to multielectron interactions to two- and three-photon processes at energies both near and away from the resonant structures dominated by singly excited intermediate states and doubly excited autoionization states are examined in detail. Our calculation has shown that the multiphoton spectra are mostly dominated by the resonant structures due to intermediate *singly* excited bound states. Such spectra are qualitatively different from the single photoionization spectrum, which often exhibits a strong presence of *doubly* excited autoionization structures. Our calculated results are in close agreement with other existing experimental and theoretical results for single- and two-photon ionizations.

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Recent advances in experimental multiphoton studies of alkaline-earth-metal atoms have led to the possibility of a systematic characterization of higher-L doubly excited autoionization structures, including the even-parity states, which are not accessible to the single-photon process [1,2]. The strong influence of the atomic structure effect to the photoelectron angular distribution for a four-photon nonresonant ionization from the ground state of Mg, due to the multielectron interactions in the ground and intermediate bound excited states, has been demonstrated in a recent combined experimental and theoretical study [3]. The purpose of this paper is to extend the simple configuration-interaction (CI) procedure, which has been applied successfully to the nonresonant multiphoton calculation, to the multiphoton ionization at energies near the highly correlated autoionization states in the continuum. In particular, we have calculated the generalized cross sections for ionizations leading to ^1S and ^1D continuua in two-photon absorption and to ^1P and ^1F continuua in three-photon absorption.

Within the low-field limit, the calculational procedure for *nonresonant* multiphoton ionization of a divalent atom, using a *single final continuum channel* and multiconfiguration state wave functions for the initial and all intermediate states, has been presented in detail in Ref. [3]. To accurately represent the highly correlated final state in the continuum in the present calculation, the single final continuum channel is replaced by a multiconfiguration state wave function calculated with a configuration-interaction method for *continuum spectrum* (CIC) [4]. Briefly, all state wave functions, which represent both bound and continuum states, are expressed in terms of a finite L^2-basis set constructed from B-spline-based frozen-core Hartree-Fock (FCHF) single-particle orbital functions in a simple CI procedure [3–6]. Following the theoretical procedure detailed in Ref. [3] and using a final-state wave function calculated with the CIC procedure presented in Ref. [4], the N-photon generalized ionization cross section σ_N in $\mathrm{cm}^{2N}\mathrm{sec}^{N-1}$, with photon energy $\hbar\omega$ and outgoing electron momentum k, is given by an expression similar to Eq. (2) of Ref. [3]

$$\sigma_N = \frac{4}{kA^2}\frac{ac}{a_0}\left[\frac{\omega}{F_0}\right]^N \sum_{J_f} |D^{(N)}|^2 , \tag{1}$$

where $D^{(N)}$ is the N-photon transition amplitude given by Eq. (3) of Ref. [3] and the amplitude A is determined by the CIC procedure given in Ref. [4].

Our calculated generalized cross sections in 10^{-50} $\mathrm{cm}^4\mathrm{sec}$ for linearly polarized one-color two-photon processes from the ground state of Mg, i.e.,

$$\mathrm{Mg}(3s^2\,{}^1S) + 2\hbar\omega \rightarrow \mathrm{Mg}^{**}(3pnl\ {}^1S \text{ or } {}^1D) \rightarrow \mathrm{Mg}^+(3s\,{}^2S) + e^- , \tag{2}$$

are shown in Fig. 1. The two-photon ionization spectrum (upper figure) leading to the ^1S continuum is dominated by the strong resonant structure due to the $3s3p\ {}^1P$ intermediate state and the $3pnp\ {}^1S$ autoionization series. For the ^1D continuum, only a hint of the $3pnp\ {}^1D$ autoionization series is seen, in contrast to the narrow but visible $3pnf\ {}^1D$ series. The $3s3p\ {}^1P$ intermediate resonant structure remains strong. A few of the final-state autoionization structures shown in Fig. 1, including $3p4p\ {}^1S$, $3p5p\ {}^1\mathrm{S}$, and $3p4f\ {}^1D$ states, have been observed recently by Shao and Charalambidis [7]. The fact that the $3pnp\ {}^1D$ series is not observed experimentally [7] is consistent with the absence of prominent structure for the $3pnp\ {}^1D$ series in our calculated spectrum. The results of our Mg two-photon ionization calculation are in close agreement with an earlier L^2-basis calculation by Moccia and Spizzo [8]. In Fig. 2, we present our calculated cross sections for the direct photoionization from the $3s3p\ {}^1P$ bound excited state of Mg at energies close to the $3p^2\ {}^1S$ autoionization state. Our theoretical result is in excellent agreement with the observed data compiled from an earlier two-color two-step experiment [2]. To present a direct comparison, the experimental results are normalized against the theoretical cross sections at the peak value. Our calculated length and velocity results agree to

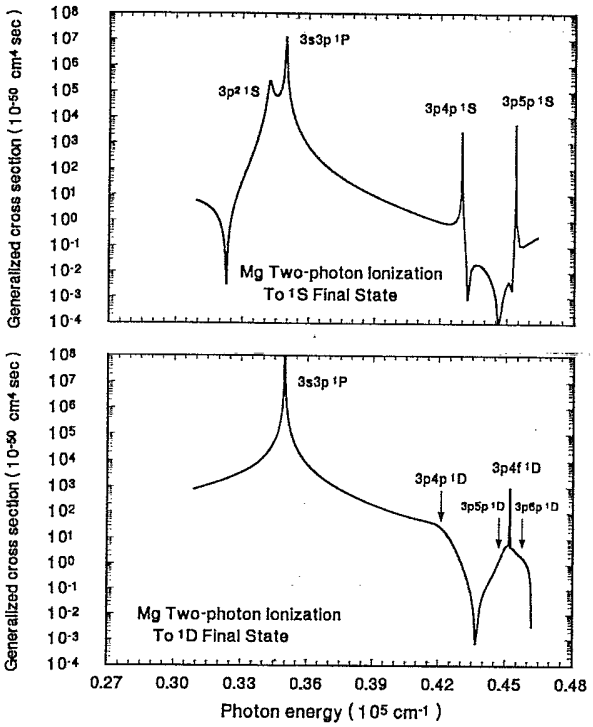

FIG. 1. The calculated generalized cross sections for two-photon ionizations leading to ^1S and ^1D continuua from the ground state of Mg.

better than 5%. Only the length results are shown in Fig. 2. In spite of the excellent agreement, the calculated width is about 10% greater than the experimental value and our earlier theoretical data [9]. In addition, the peak cross section is also noticeably larger than our earlier result [10]. In the present calculation, only 12 configuration series are included. For a more accurate quantitative result, additional configuration series are required in the CIC calculation. A more comprehensive calculation, with

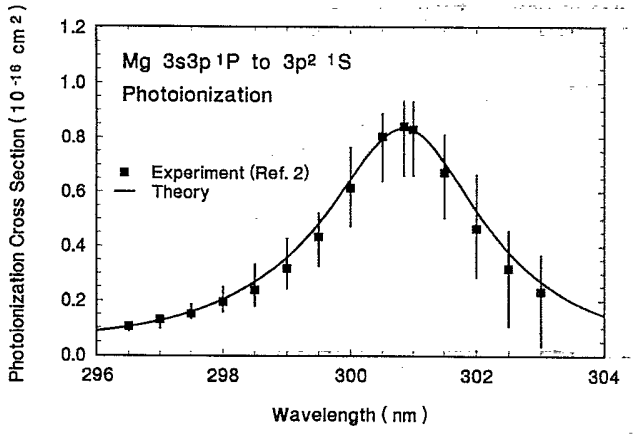

FIG. 2. The calculated photoionization cross section from the singly excited $3s3p\ ^1P$ state of Mg to the doubly excited $3p^2\ ^1S$ autoionization state in the continuum. The experimental data from Ref. [2] are normalized against the theoretical cross sections at the peak value.

expanded basis set, for the width and profile of the autoionization structures is currently in progress.

In Fig. 3, we present the generalized cross sections for linearly polarized one-color three-photon ionization of Mg from ground state that lead directly to the ^1F continuum. The ionization spectrum is dominated by the resonant structures due to the $3snd\ ^1D$ intermediate bound excited states. The strongly correlated $3p3d\ ^1F$ doubly excited autoionization states [11] is less prominent but remains visible on the low-energy side of the $3s6d\ ^1D$ intermediate resonance. Again, a total of 12 configuration series is included in our state wave-function calculation for all bound and continuum states. The length and velocity results are nearly indistinguishable on the logarithmic plot. The effects due to the configuration interaction is illustrated by the substantial difference both in energy and amplitude of the resonant structure between the top and bottom ionization spectra as the total number of configuration series is reduced from 12 to one in that only a singly excited $3sl$ configuration series is included in all wave-function calculations. In particular, the length and velocity results are clearly separated in the single series calculation, which represents a single channel FCHF approximation. We have also examined the structure effect at energy near the $3p3d\ ^1F$ resonance in a separate calculation which includes only four dominant configuration series, i.e., $3sf$, $3pd$, $3dp$, and $3df$ in the ^1F continuum. A comparison between the 4-series and 12-series results is shown in Fig. 4. As expected, the final state CI effect is much stronger at energies in the immediate neighborhood of the $3p3d\ ^1F$

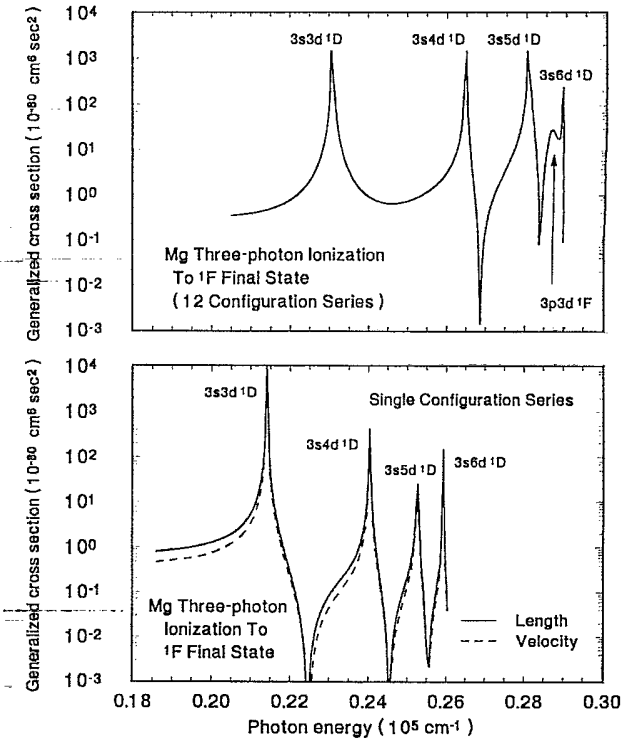

FIG. 3. The calculated generalized cross section for three-photon ionization leading to ^1F continuum from the ground state of Mg.

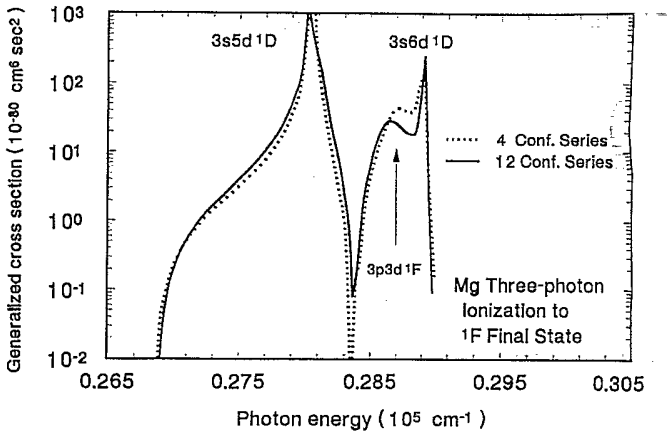

FIG. 4. The calculated generalized cross section for three-photon ionization from the ground state of Mg at energy close to the doubly excited $3p3d\ ^1F$ autoionization structure.

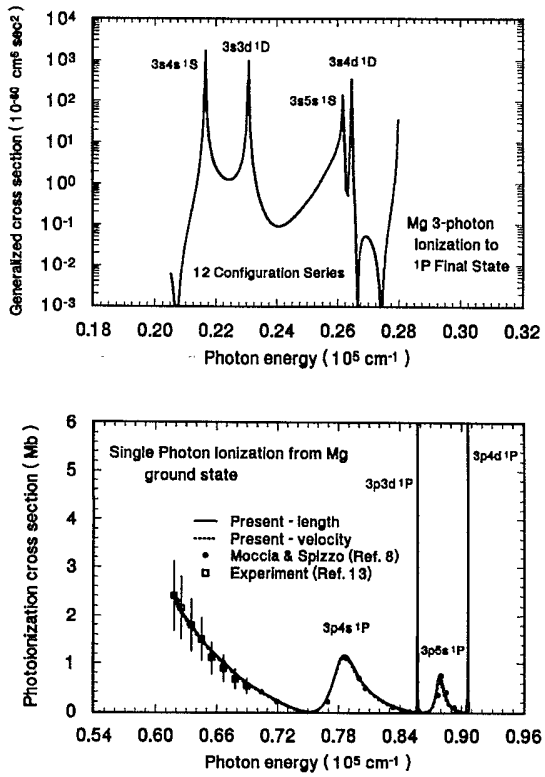

FIG. 5. The generalized cross section for three-photon ionization and the single photoionization cross section leading to ^1P continuum from the ground state of Mg. The experimental data are taken from Ref. [13].

resonance than that close to the $3snd\ ^1D$ intermediate resonant structures.

For the three-photon ionization leading to the ^1P continuum, the prominent doubly excited $3pns\ ^1P$ autoionization states, which dominate the one-photon process, are completely overwhelmed by the $3sns\ ^1S$ and $3snd\ ^1D$ intermediate resonant structures. The photon energies for the three-photon and the single-photon spectra shown in Fig. 5 are scaled to a ratio of 1 to 3 for easy comparison. The broad $3p4s\ ^1P$ structure seen in the single photoionization spectrum is reduced to a slightly broaden shoulder at the low-energy side of the $3s5s\ ^1S$ intermediate resonance in the three-photon spectrum. Again, the length and velocity results for the three-photon ionization are nearly indistinguishable in Fig. 5.

We have also compared in Fig. 5 our calculated one-photon ionization cross sections with the recent theoretical results by Moccia and Spizzo [12] and the most recent *absolute* photoabsorption cross sections measured by Yih *et al.* [13]. The present theoretical results are also consistent with the normalized experimental data by Preses *et al.* [14]. In addition, our results are in close agreement with other earlier theoretical results, including the velocity results by Bates and Altick [15] and the multiconfiguration Hartree-Fock results by Froese Fischer and Saha [16], although all the above mentioned theoretical results are noticeably less than the results from the earlier version of the R-matrix method by O'Mahony and Greene [17] at energy near the ionization threshold.

In conclusion, the present theoretical study has shown that, as the number of photon increases, the energy spectrum of a one-color multiphoton process does not necessarily reveal the atomic structure effects which dominate the final-state multielectron interactions in the continuum, in spite of its ability to reach directly the higher-L states of both odd and even parity. On the other hand, a multicolor multistep process may turn out to be a more efficient physical mechanism in the study of multielectron interactions in the continuum if it is capable of yielding detail photoionization data for transition from a highly correlated bound excited state to a strongly correlated autoionization state such as the $3s3p\ ^1P \rightarrow 3p^2\ ^1S$ transition shown in Fig. 2.

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