Polarization effects in the photoionization of barium 4*d* electrons

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Partial 4*d* photoionization cross sections have been calculated for atomic barium including a polarization potential to evaluate the effects of core polarization. Results of three theoretical models are compared: (i) the relativistic random-phase approximation, (ii) the relativistic random-phase approximation modified to include relaxation, and (iii) the relativistic random-phase approximation modified to include relaxation and core polarization. Theory is also compared with photoelectron spectroscopy measurements. The inclusion of both relaxation and polarization effects in the calculations greatly improves agreement with experiment.

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Photoionization studies above the 4*d* subshell thresholds of atomic barium have sharpened our understanding of the role of many-electron effects in atoms. The broad, delayed peak (so-called "giant resonance") found in the total absorption spectrum [1] showed that a large centripetal barrier was present in the potential for continuum *f* electrons. Photoelectron spectroscopy experiments [2–4] found that approximately half of the total absorption in this region is due to main-line 4d photoionization and half results from satellite channels. A series of RRPA-type calculations [5] showed that it is important to include relativistic effects, interchannel coupling, and relaxation effects in order to obtain an adequate description of the total absorption. Furthermore, it was found that the partial 4*d* cross section is more accurately modeled by theory when the nonorthogonality between orbitals of the frozen-core ground state and the relaxed-core final state is corrected with the inclusion of overlap integrals in the dipole matrix elements. The effect of core polarization was previously studied by Kutzner *et al.* [6] through the addition of a class of second-order diagrams in many-body perturbation theory (MBPT) which were found to dramatically improve agreement with experiment [2–4] for the partial 4*d* photoionization cross section.

Recently, polarization effects have been implemented in calculations of negative ion photodetachment calculations [7–9] using a polarization potential. The combination of relaxation and polarization effects calculated in this way were successfully applied to 4*d* photodetachment of I^- [9]. The purpose of the current study is to apply the polarization potential method for the first time to a neutral species and determine its effect on the 4*d* cross section of atomic Ba.

The polarization potential method has been described by Amusia [10] and consists of adding the term

$$V_{pol}(r) = -\frac{\alpha_d}{2(r^2+h^2)^2} \tag{1}$$

to the potential of the photoelectrons. Here, α_d is the dipole polarizability of the remaining positive ionic core for Ba^+ and h is a cutoff radius (approximately the size of the valence electron cloud) which prevents the potential from becoming unmanageable for the radial coordinate approaching zero. In this particular case, the dipole polarizability for Ba^+ has been calculated by Rozsnyai [11]. The value of the dipole polarizability for Ba^+ ($\alpha=171.4$ a.u.) is very large, even when compared with most neutral atoms [12]. This is accounted for by the presence of a loosely-bound 6*s* electron in the valence shell. The cutoff radius, h, is determined by requiring that $V_{\mathrm{pol}}(0)$ be equivalent to the energy correction of the subshell 4*d* orbitals [10]. This condition may be expressed as

$$V_{pol}(0) = \Delta E_S(4d) - |\varepsilon_{4d}|, \tag{2}$$

where ε_{4d} is the Dirac-Hartree-Fock (DHF) eigenvalue for 4*d* orbitals and $\Delta E_S(4d)$ is the absolute value of the difference between the total ground state self-consistent-field energies of the neutral barium atom and the positive barium ion with a 4*d* electron missing. Equations (1) and (2) are combined to yield the value of the cutoff radius, h_{4d}. Specifically for the barium 4*d* subshell, the cutoff radius was found to be 4.48 a.u. (slightly less than the 5.08 a.u. expectation value of the radius for the DHF 6*s* orbital).

The partial 4*d* cross sections are shown in Fig. 1. The theoretical cross sections shown in Fig. 1 are geometric means of length and velocity. While the strict RRPA calculations are gauge invariant [13], this does not hold when relaxation and polarization effects are included in the potential. Differences in length and velocity partial 4*d* cross sections are approximately 10% at the peak for calculations including only relaxation effects (RRPAR) and approximately 5% for calculations including both relaxation and polarization (RRPARP). The experimental partial cross sections [2–4] are results of photoelectron spectroscopy measurements partitioning absorption cross sections [1]. The partial main-line cross section has about half the total oscillator strength of the total absorption cross section.

The theoretical cross section calculations follow an interesting progression as each many-body effect is included. The calculation with the highest peak and narrowest distribution of absorption is the nonrelativistic random-phase approximation with exchange (RPAE) calculation of Amusia *et al.* [14] which included only 4*d* intrachannel coupling. Including relativistic effects in the relativistic random-phase approximation with only 4*d* intrachannel coupling included [RRPA (4*d*)] [5] has the effect of somewhat broadening and lower-

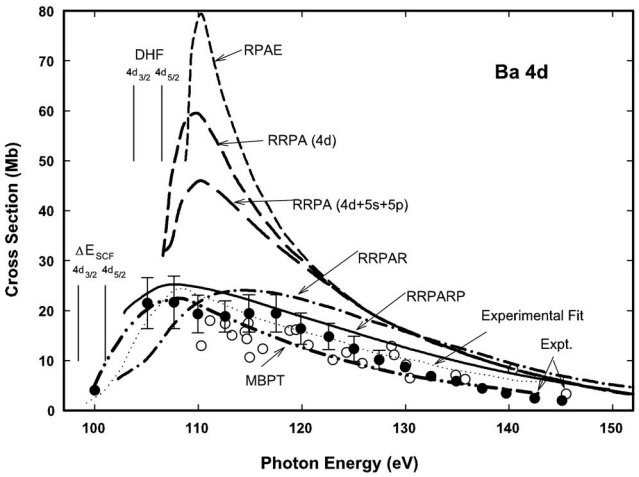

FIG. 1. RPAE is random-phase approximation with exchange calculation of Ref. [14]; RRPA($4d$) is relativistic random-phase approximation with intrachannel $4d$ coupling only, Ref. [5]; RRPA ($4d+5s+5p$) is relativistic random-phase approximation with interchannel coupling among $4d$,$5s$, and $5p$ channels, Ref. [5]; RRPAR is the relativistic random-phase approximation with interchannel coupling and relaxation effects, Ref. [5]; RRPARP is the current calculation utilitzing the relativistic random-phase approximation with interchannel coupling, relaxation, and polarization effects included. Many-body perturbation calculations (MBPT) including relaxation and polarization diagrams, Ref. [6], are represented by the double-dot-dashed line. The photoelectron spectroscopy measurements are indicated by the solid circles (Ref. [2]), dotted line (Ref. [3]), and open circles (Ref. [4]).

ing the peak due to the spin-orbit splitting of the $4d_{3/2}$ and $4d_{5/2}$ thresholds. Interchannel coupling with $5s$ and $5p$ subshells, included in the RRPA ($4d+5s+5p$) calculation [5] further reduces the peak by redistributing oscillator strength away from the $4d$ channels into other single-excitation channels. The inclusion of relaxation effects in the relativistic random-phase approximation modified to include relaxation effects (RRPAR) [5] redistributes the absorption from the threshold to higher energies at the same time reducing the overall oscillator strength by approximately 23% due to the inclusion of overlap integrals between orbitals of the ground state and the orbitals of the final state calculated in the potential of the relaxed ion.

The present calculations (RRPARP) include all of the above many-body effects as well as the effects of core polarization by the outgoing photoelectron through the inclusion of the polarization potential of Eq. (1). The impact of polarization on the photoionization cross section is to draw oscillator strength back toward the threshold, thus partially canceling the effects of core relaxation. The RRPARP result is consistent with Ba calculations evaluating polarization effects via many-body perturbation theory [6]. The effect is also similar in type, but larger in magnitude, to that found for I^- [9]. Inclusion of polarization effects allows the outgoing photoelectron to partially fill the core hole assumed in the relaxed potential model. Whereas assuming core relaxation yields a more diffuse, less attractive photoelectron potential, including a polarization potential enhances the attraction, effectively reducing the kinetic energy of the outgoing photoelectrons and increasing absorption near threshold. The many-body perturbation theory (MBPT) calculation of Kutzner *et al.* [6] which include both relaxation and polarization effects via the inclusion of diagrams is also shown for comparison purposes and provides a fairly complete description of the electron correlation.

In conclusion, it has been found that the partial $4d$ photoionization cross section of atomic Ba is well described when the effects of relativity, interchannel coupling, core relaxation, and polarization are all included. An interesting followup study might investigate the role of relaxation and polarization on the autoionization resonances between and below the $4d$ thresholds of barium and xenon.

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