Electron-impact excitation of excited atomic barium

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We present results of integral-excitation and total-ionization cross section calculations from the $6s6p\ ^1P_1$, $6s5d\ ^1D_2$, $6s6p\ ^3P_{0,1,2}$, and $6s5d\ ^3D_{1,2,3}$ states of Ba. The unitarized first-order many-body theory and the close-coupling method were used to obtain the integrated cross sections, while the Born approximation was used to estimate the ionization cross sections. A comparison is given with the semiempirical estimates of excitation cross sections obtained from small-angle differential cross-section measurements.

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I. INTRODUCTION

Establishing a comprehensive database of accurate electron-atom/ion excitation cross sections is of great importance for the kinetic modeling of plasmas. Integrated cross sections (ICS) for electron scattering from Ba are used in a variety of applications, such as modeling of the Ba-vapor lasers [1–4], discharge lamps [5], plasma switches [6], and various planetary ionospheres [7–12].

In our previous work [13] we used the convergent close-coupling method (CCC) and the unitarized first-order many-body theory (UFOMBT) to investigate the ICS for electron impact excitation from the ground state of Ba. The only directly measured ICS is the $6s6p\ ^1P_1$ optical excitation function obtained by Chen and Gallagher [14]. The good agreement between the experimental and theoretical results has encouraged us to carry out similar calculations for other transitions.

The aim of the present paper is to study transitions between excited states of barium. We will present electron impact ICS and total-ionization cross sections for the $6s6p\ ^1P_1$, $6s5d\ ^1D_2$, $6s6p\ ^3P_{0,1,2}$, and $6s5d\ ^3D_{1,2,3}$ states. The choice of the transitions has been made from the analyses of the major features in the energy-loss spectra obtained at 10, 20, and 36.7 eV impact energies at small (5°–20°) scattering angles [15–17]. Transitions with the largest differential cross sections (DCS) were selected. We also took into account the need for cross section data from applications, such as Ba discharge as a ligthing device [18].

There are no direct experimental data available for the ICS involving transitions between excited states of barium. However, DCS have been measured by Li and Zetner [19], Zetner *et al.* [15,17], and Johnson *et al.* [16] for a large number of excitations from $6s6p\ ^{1,3}P_1$ and $6s5d\ ^{1,3}D_2$ states, and good agreement was established with the CCC calculations. The DCS measurements and the theoretical results (CCC and UFOMBT) revealed, in general, a forward-peaking behavior of the DCS. Such behavior is consistent with the breakdown of the nonrelativistic LS coupling approximation for a number of states in Ba, and, in particular, for the $6s6p\ ^{1,3}P_1$ and $6s5d\ ^{1,3}D_2$ states that have been included as initial states in the set of transitions considered here. We, therefore, have some confidence in the reliability of the calculated ICS since forward scattering is the predominant contribution to the ICS. The forward-peaking behavior of the DCS allows us to give a semiempirical estimate of the corresponding experimental ICS, which has been done for a number of transitions.

II. THEORETICAL MODELS

The details of the present theories have already been given in Ref. [13] and references therein. Briefly, the close-coupling calculations have been performed at Flinders University using the CCC method. In this method a set of Ba target states are used to expand the total e-Ba wave function and to form a set of close-coupling (CC) equations. They are solved using the momentum-space close-coupling method [20], in the distorted-wave representation [21,22]. The calculations have been performed in two models. The 55-state CC calculations include all negative-energy states (relative to the Ba^+ ground state) obtained in the structure calculations. The CC(55) model does not include any of the positive-energy states and therefore has no coupling to the ionization channels. The 115-state CCC calculations include a large number of positive-energy states to model the coupling to the ionization channels. The CC(55)-model calculations have been performed at a large number of incident electron energies. The CCC(115) calculations are significantly more time consuming and have been performed at fewer energies. There are, in general, little differences between CC(55) and CCC(115) results for transitions between excited states, which indicates a good rate of convergence of the CC expansion. This can be explained by the fact that channel coupling effects are strongest between closest-lying states. For electron-impact excitations from excited states, there are a large number of target states close in energy to the initial state and these comprise the most important terms in the CC expansion. Nearly all

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such close-lying states have beed included in the CC(55) model.

The UFOMBT method used here has been discussed in general and in particular its implementation for Ba by Clark *et al.* [23] and Zetner *et al.* [15]. The UFOMBT is one version of the distorted wave approximation (DWA). The UFOMBT calculations were performed at Los Alamos using the CATS and ACE codes. The CATS code is a user-friendly version of Cowan's atomic structure code, and ACE is a collisional excitation code for DWA-type calculations. In the DWA scheme, it is envisioned that the wave function of the incident electron is ''distorted'' by the atomic potential. Subsequently, that electron excites the atom into its final state in a one-step process. Finally, the electron leaves and its wave function is distorted again by the atomic potential. In UFOMBT every excitation process is handled separately. Thus, every excitation process is considered independent, and no coupling effects are included. A comparison with the close-coupling approach enables the establishment of the importance of the channel-coupling effects, so long as the underlying target structure is sufficiently similar.

In both CCC and UFOMBT methods the structure of the barium atom has been described by a model of two active electrons above a frozen Hartree-Fock core (see Ref. [24] for details). Barium wave functions exibit strong configuration interaction, which manifest itself in the presence of a large number of doubly excited states in the discrete spectrum. In addition, the situation is complicated further by a breakdown of the nonrelativistic approximation for a number of transitions. The wave-function decomposition and account of the major relativistic effects has been discussed in Refs. [13,17] for the CCC method and in Refs. [15,17] for the UFOMBT method.

In the CCC model the scattering calculations are performed in a nonrelativistic LS-coupling scheme. The account of the major relativistic effect, singlet-triplet mixing in the Ba wave functions, is achieved by transformation of the nonrelativistic scattering amplitudes to the intermediate coupling scheme [13], a procedure similar to the one described by Saraph [25]. In our previous work we found that this method of accounting for relativistic effects proved to be successful in the calculation of differential cross sections for scattering from $6s6p\ ^{1,3}P_1$ and $6s5d\ ^{1,3}D_2$ states [15–17]. We shall refer to the cross sections obtained according to this procedure as semirelativistic. The UFOMBT model adopts a similar approach to the account of relativistic effects. The important difference, though, is that Ba target states are described in the intermediate coupling scheme from the beginning and then the scattering calculations are performed, see Clark [26] and Mann [27] for details.

Neither of the presented theoretical results (CCC and UFOMBT) are able to obtain highly accurate results close to excitation thresholds. The UFOMBT theory is a high-energy approximation that breaks down at low energy. The present CCC results rely substantially on the approximate scheme for the account of relativistic effects. This could make present CCC results inaccurate for energies close to the threshold for some transitions. We, therefore, do not present results for incident electron energies close to excitation thresholds.

III. RESULTS

A. Excitation

Results are presented from UFOMBT and nonrelativistic and semirelativistic CCC(115) and CC(55) calculations for e-Ba excitation for the following transitions:

$$6s6p\ ^1P_1 \rightarrow 6p5d\ ^1D_2, 5d^2\ ^3P_2, 5d^2\ ^1D_2,$$
$$6s6d\ ^1D_2, 6s7p\ ^1P_1, \text{ and } 6s7s\ ^1S_0;$$
$$6s5d\ ^1D_2 \rightarrow 6p5d\ ^3F_2, 6p5d\ ^1D_2, 5d^2\ ^1D_2, 6s7p\ ^1P_1,$$
$$6p5d\ ^3F_3, 5d^2\ ^3P_1, \text{ and } 6s6p\ ^1P_1;$$
$$6s5d\ ^3D_2 \rightarrow 6s6p\ ^3P_2, 6p5d\ ^3F_3 \text{ and } 6p5d\ ^3P_2;$$
$$6s6p\ ^1P_1 \leftarrow 6s6p\ ^3P_{0,1,2}, 6s5d\ ^3D_{1,2,3} \text{ and } 6p5d\ ^1D_2.$$

Figure 1 presents ICS for excitation from the $6s6p\ ^1P_1$ state. The ICS for excitation of the $6p5d\ ^1D_2$ state, given in Fig. 1(a), shows reasonable agreement between the CC and UFOMBT results in the $E_0 > 20$ eV energy range, while for $E_0 < 20$ eV the UFOMBT results are too high by a factor of about 2, indicating the importance of channel coupling. A comparison of nonrelativistic and semirelativistic CC results indicates that the singlet-triplet mixing leads to uniform and relatively small reduction of the ICS. The variation in this mixing between the CC and FOMBT theories is responsible for the 20% or so difference between the two theories at the higher energies.

Figure 1(b) shows our results for excitation of the $5d^2\ ^3P_2$ state. Here we observe very strong effects of singlet-triplet mixing. In a nonrelativistic calculation this transition can happen only via exchange scattering, which leads to a very fast decrease of the nonrelativistic ICS as the incident electron energy increases. The major contribution to the semirelativistic ICS at the higher energies comes from the LS-coupled $5d^2\ ^1D_2$ term that can be excited from the initial $6s6p\ ^1P_1$ state via direct scattering. Comparing the UFOMBT and CC, we observe that the former are about a factor of 10 higher than the CC semirelativistic results. This difference can be attributed only partially to the difference in the $5d^2\ ^1D_2$ term mixing coefficients used in UFOMBT and CC calculations, which are 0.55 and 0.36 correspondingly. The channel-coupling effects must be very important for this transition. Our experimental estimate at 20 eV clearly supports the CC results. The magnitude of the discrepancy between the two theories is indicative of the difficulty of the problem, and the importance of a self-consistent fully relativistic approach to e-Ba scattering.

Figure 1(c) shows the results for excitation of the $5d^2\ ^1D_2$ state. Here essentially all results show good agreement. The agreement between the semirelativistic CC55 calculation results and UFOMBT results improves with increasing energy indicating the decreasing importance of channel coupling

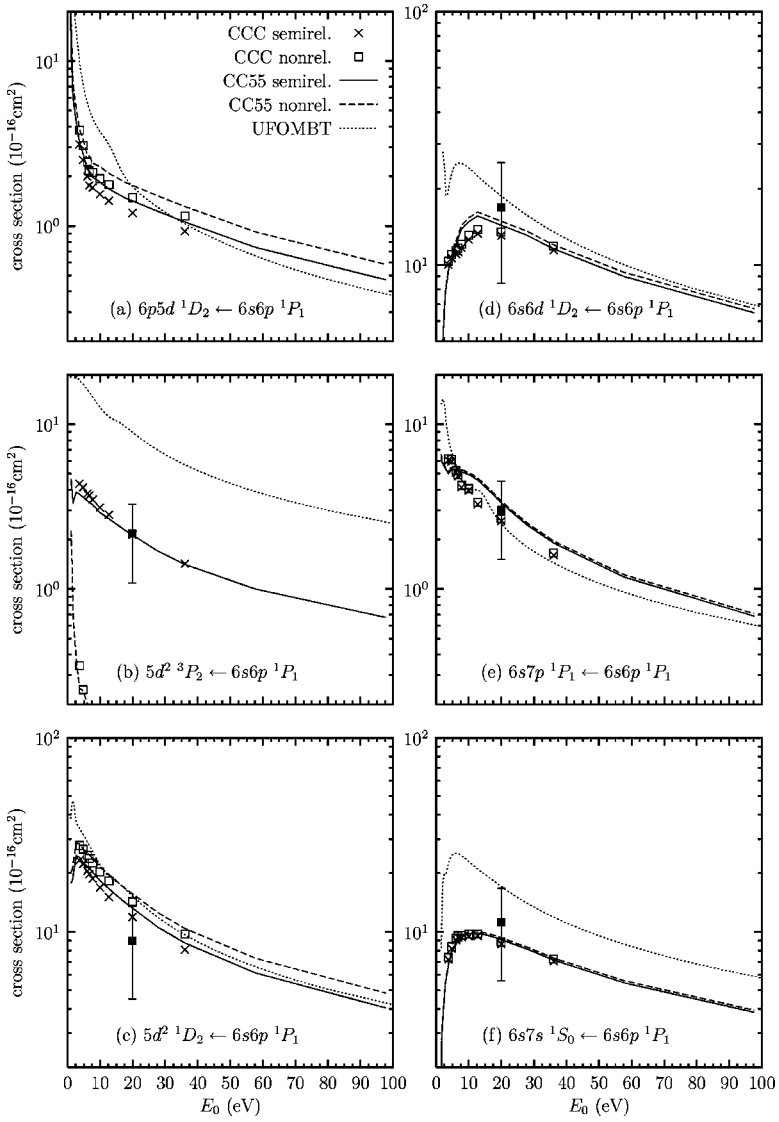

FIG. 1. Integral excitation cross sections for electron scattering by $6s6p\ ^1P_1$ state of barium. The experimental results are given by filled rectangles with the appropriate error bars.

with increasing energy. This also indicates a good agreement between the correponding target wave functions used in the UFOMBT and CC calculations.

Figure 1(d) gives the ICS for excitation of the $6s6d\ ^1D_2$ state. From this figure one can see that in the low-energy region ($E_0<10$ eV) there is strong disagreement between the UFOMBT the CC(55) and CCC results while the latter two agree with each other very well. At 10 eV incident electron energy the UFOMBT results are about 50% higher than the CC results. However, with increasing energy the disagreement essentially disappears. At 100 eV incident electron energy, the UFOMBT results are only about 10% higher then the CC results indicating a slight difference in the underlying wave functions.

Figure 1(e) displays the ICS for excitation of the $6s7p\ ^1P_1$ state. This is a direct excitation process. The figure shows a good agreement between the UFOMBT results and the CCC results for the whole energy range except close to the threshold (for $E_0<5$ eV), where UFOMBT is not expected to be reliable.

Figure 1(f) shows our results for excitation of the $6s7s\ ^1S_0$ state. This is a dipole-allowed transition with a large cross section that falls off slowly with the increasing energy. The UFOMBT results are about 30% larger than the CC(55) and CCC results that, in turn, agree with each other. The general shape of all cross sections is the same. The disagreement should be probably attributed to the different target wave functions used in the UFOMBT and CC calculations. The CC results are in better agreement with our 20-eV experimental estimate.

The $6s5d\ ^1D_2$ and $6s5d\ ^3D_2$ metastable states are populated by radiative decay of the laser-excited $6s6p\ ^1P_1$ state. Electron-impact excitation of these two metastable states to the seven and five higher excited states, respectively, as listed above, can be discussed along the same lines as done for Fig. 1. We find that there is a good agreement between the results obtained from the nonrelativistic and semirelativistic CC calculations for the $6s5d\ ^1D_2$ to $6p5d\ ^1D_2$, $6s7p\ ^1P_1$, $5d^2\ ^3P_1$, $6s6p\ ^1P_1$ and for the $6s5d\ ^3D_2$ to $6s6p\ ^3P_2$, $6p5d\ ^3F_3$, $6p5d\ ^3P_2$ excitations. We can, therefore, conclude that for all these excitations relativistic effects are not important. The experimental estimates for the $6s5d\ ^1D_2$ to $6s7p\ ^1P_1$ and $6s5d\ ^3D_2$ to $6s6p\ ^3P_2$ transition are in very good agreement with the CC results. As ex-

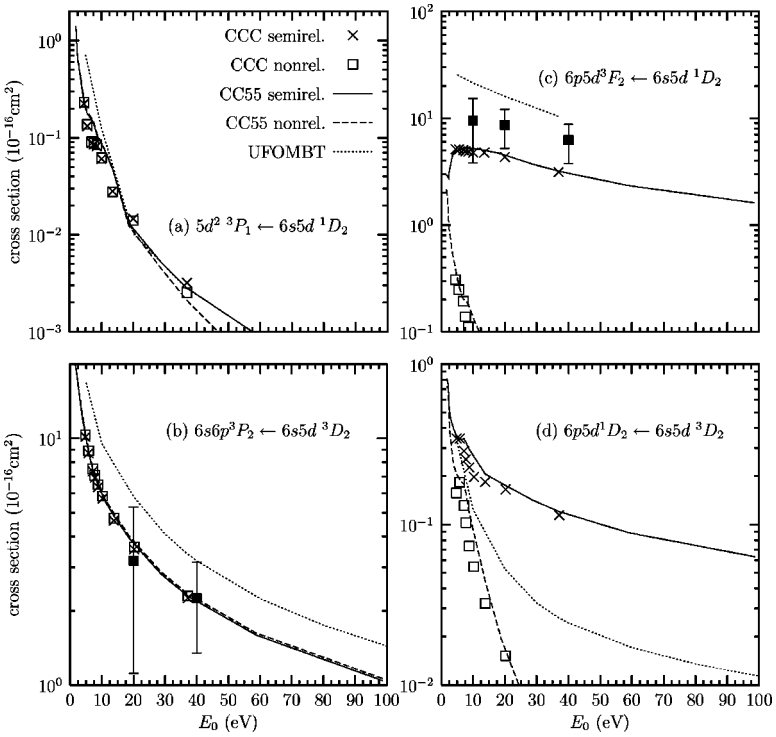

FIG. 2. Integral excitation cross sections for electron scattering by $6p5d\ ^1D_2$ and $6p5d\ ^3D_2$ states of barium. The experimental results are given by filled rectangles with the appropriate error bars.

amples, we show here in Fig. 2(a) the $6s5d\ ^1D_2$ to $5d^2\ ^3P_1$ and in Fig. 2(b) the $6s5d\ ^3D_2$ to $6s6p\ ^3P_2$ excitation results. On the other hand, we find that relativistic effects are very important for the $6s5d\ ^1D_2$ to $6p5d\ ^3F_2$, $6p5d\ ^3F_3$ and $6s5d\ ^3D_2$ to $5d^2\ ^1D_2$, $6p5d\ ^1D_2$ excitations as indicated for two transitions in Figs. 2(c) and 2(d). For this type of transitions we find generally much poorer agreement between CC and UFOMBT results. The origin of such disagreements, at least at high energies, can be traced to the different wave-function composition in CC and UFOMBT methods. Numerical results and/or figures not presented in this paper can be obtained from the first author.

Figure 3 shows our ICS results for an excitation out of the $6s6p\ ^3P_J (J=0,1,2)$ states to the $6s6p\ ^1P_1$ state and Fig. 4 shows our ICS results for an excitation out of the $6s5d\ ^3D_J (J=1,2,3)$ states to the $6s6p\ ^1P_1$ state. The analysis of these figures can be performed as before with similar conclusions.

Although, no experimental integral cross sections are available for the excitations discussed here, some measurements of the corresponding DCS's have been reported by Li and Zetner [19], Johnson *et al.* [16], and Zetner *et al.* [15,17]. In all cases the DCS are limited to scattering angles from about 5° to 35° and to impact energies of 10, 20, 36.7, and 40 eV. The transitions covered by these investigations are from $6s5d\ ^1D_2$ to $6s6p\ ^1P_1$, $6p5d\ ^3F_2$ and $6s7p\ ^1P_1$ states, and from $6s5d\ ^3D_2$ to $6s6p\ ^3P_2$ state. In all cases the DCS decreases from 0° to 20° by two orders of magnitude. An extrapolation of these DCS's to 180° and to 0° and integration yields, therefore, meaningful experimental integral cross sections. Contributions coming from angles higher than 35° are negligible. There are some uncertainties involved with the extrapolations to 0° since the cross sections increase sharply with decreasing angle. However, this uncertainty is damped by the fact that the DCS values are multiplied by $\sin(\theta)$ during the integration and that the DCS values from CCC semirelativistic calculations are available to guide these extrapolations. The calculated and experimental DCS values agree very well in all cases in shape and in most of the cases also in magnitude.

DCS values are also available (Zetner [28]) for excitations from the $6s6p\ ^1P_1$ level to the $5d^2\ ^3P_2$, $6s7s\ ^1S_0$, $6s7p\ ^1P_1$, and $6s6d\ ^1D_2$, as well as to the combined $(5d^2\ ^1D_2 + 6p5d\ ^1D_2)$ levels at 20-eV impact energy and angles ranging from 5.5° to 16°. Our calculations show that at 20 eV for the combined $(5d^2\ ^1D_2 + 6p5d\ ^1D_2)$ level the $5d^2\ ^1D_2$ component is by far dominant. Within rather large experimental uncertainties the DCS for the combined $(5d^2\ ^1D_2 + 6p5d\ ^1D_2)$ level can be assigned to the largest $5d^2\ ^1D_2$ component. The same comments apply to these DCS's as to the ones discussed above for excitations from the $6s5d\ ^1D_2$ and $6s5d\ ^3D_2$ metastable levels.

We carried out the required extrapolations and integrations for the available experimental DCS and some of the resulting integral cross sections are shown in Figs. 1 and 2. In cases where the experimental and calculated DCS values agreed well within the experimental error limits and good agreement in the shape of the DCS's angular behaviors was found, we accepted the calculated integral cross-section values. In cases when the angular behaviors were similar but the absolute values differed, we scaled the calculated integral cross-section values using a factor derived from the DCS. Finally, when neither the shape nor the absolute values agreed well, we estimated the required scaling factors, for these two effects. The error limits were deduced by considering the errors associated with the experimental DCS and

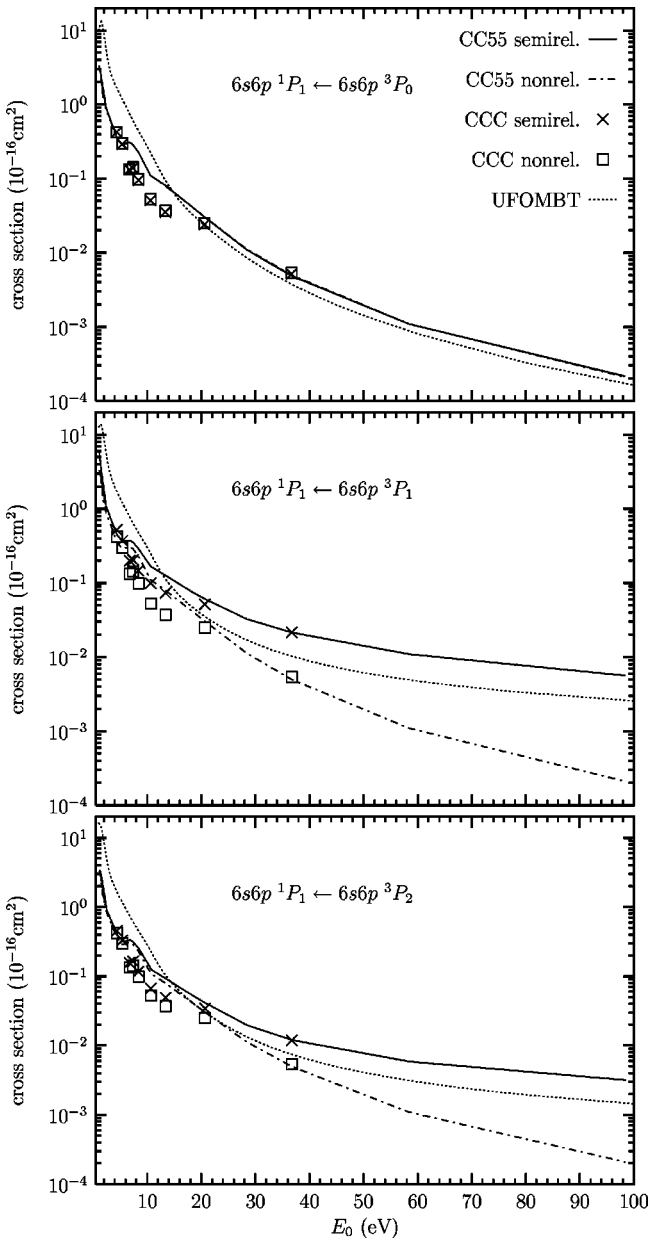

FIG. 3. Integral excitation cross sections of the barium $6s6p\ ^1P_1$ state for electron scattering by $6s6p\ ^3P_{0,1,2}$.

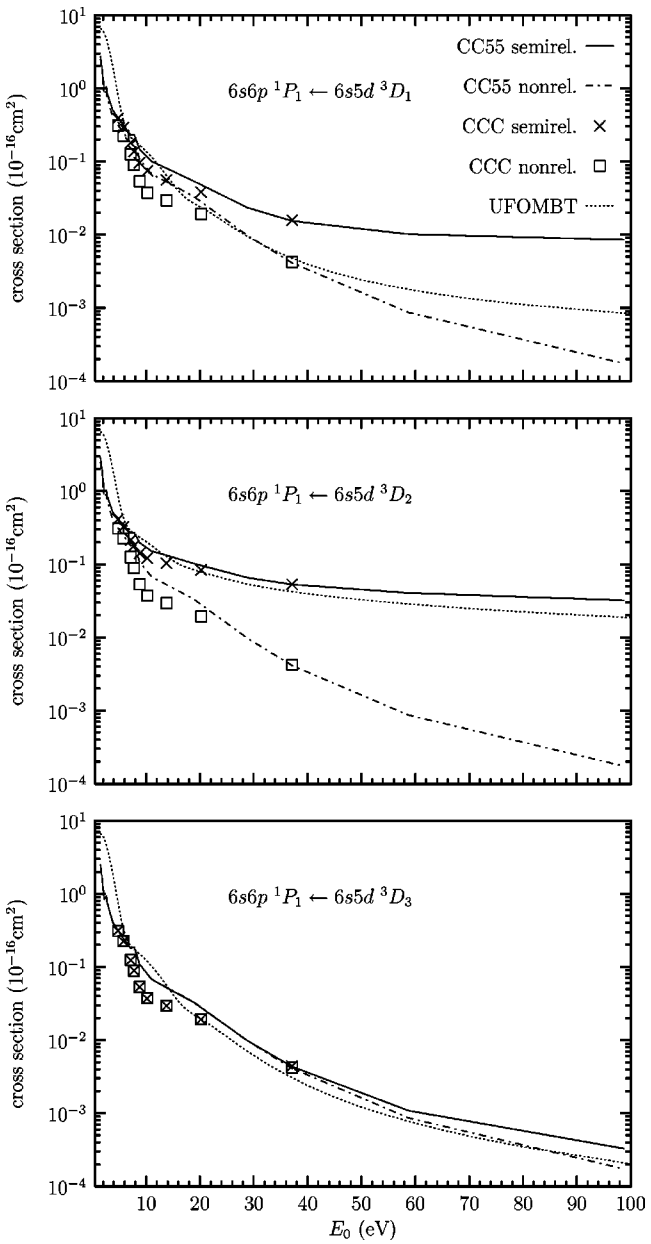

FIG. 4. Integral excitation cross sections of the barium $6s6p\ ^1P_1$ state for electron scattering by $6s5d\ ^3D_{1,2,3}$.

with the uncertainties associated with the extrapolations. The resulting error limits are rather large but no better results can be expected in the near future and for certain modeling purposes these results may be adequate. The agreement with the CCC results is good in all cases except for the $6s5d\ ^1D_2$ to $6p5d\ ^3F_2$ excitation shown in Fig. 2(c).

B. Ionization from the $6s6p\ ^{1,3}P_{0,1,2}$ and $6s5d\ ^{1,3}D_{1,2,3}$ states

In order to obtain a reliable ionization cross section for initial states with nonzero angular momentum l_i, one has to include in the CC expansion a set of target states with large values of l. These states generate a large number of channels and the problem quickly becomes computationally intractable.

In the CCC method the total ionization cross section σ_i^+ is obtained as a sum of excitation cross sections for all positive-energy (relative to the Ba^+ ground state) states

$$\sigma_i^+ = \sum_{n:e_n>0} \sigma_n^{CC}. \tag{1}$$

Note that the inclusion of two-electron excitations in the calculation of Ba target states means that ionization with excitation process is also included, but not ionization of the core, or double ionization. No relativistic effects have been accounted for in the calculation of the total-ionization cross sections as we expect them to be adequately averaged over by the underlying summation over all singlet and triplet state ICS.

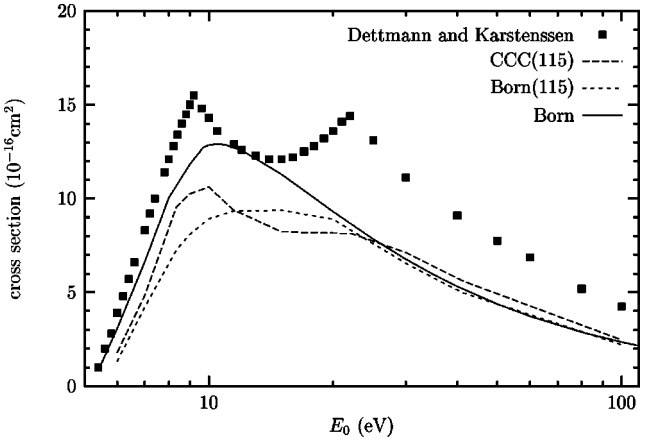

FIG. 5. Ionization cross sections σ_i^+ for electron scattering by the ground state of barium. Experiment is due to Dettmann and Karstenssen [29].

While it is computationally intractable in the CC model to obtain a reliable estimate of the excitation cross sections σ_n^{CC} $(e_n>0)$ and consequently the total-ionization cross section σ_i^+, we can use a number of simpler models to obtain an estimate of the excitation cross sections for positive-energy states. The simplest and computationally most efficient model is the first Born approximation (FBA). We have used FBA to calculate excitation cross sections σ_n^{FBA} for positive-energy states. An estimate of the total-ionization cross section is then obtained by substituting FBA cross sections σ_n^{FBA} into Eq. (1) instead of the CC cross sections σ_n^{CC}.

The error in the total-ionization cross section associated with the replacement of the CCC cross sections with FBA cross sections can be illustrated on the example of ionization from the ground state of barium. In Fig. 5 we presented both the 115-state CCC estimate [labeled CCC(115)] and the corresponding Born estimate [labeled (Born(115)] together with the experimental data of Dettmann and Karstenssen [29]. We find sufficiently good agreement between the CCC and Born results, except the region around first maximum at 10 eV. We have also presented in Fig. 5 the σ_i^+ Born results for a larger number of states. The use of the FBA allows us to take the largest target-space orbital angular momentum $l_{\max}=6$, sufficient to achieve convergence in the Born σ_i^+ values. The substantial difference between the convergent Born and 115-state Born results suggests a lack of convergence in our CCC(115) σ_i^+ values at energies around the first cross-section maximum. At larger energies we observe discrepancy between the theory and the experiment, which is due to the lack of allowance for inner-core ionization processes in the present theory.

In Fig. 6 we present our FBA estimates for the $6s6p\ ^1P_1$, $6s5d\ ^1D_2$, $6s6p\ ^3P_{0,1,2}$, and $6s5d\ ^3D_{1,2,3}$ initial states. In the approximation we use here, the cross sections for ionization from individual fine-structure levels are all the same and equal to the corresponding nonrelativistic (LS-coupling) cross section. All cross sections have a maximum at around 10 eV similarly to the ground-state ionization cross section.

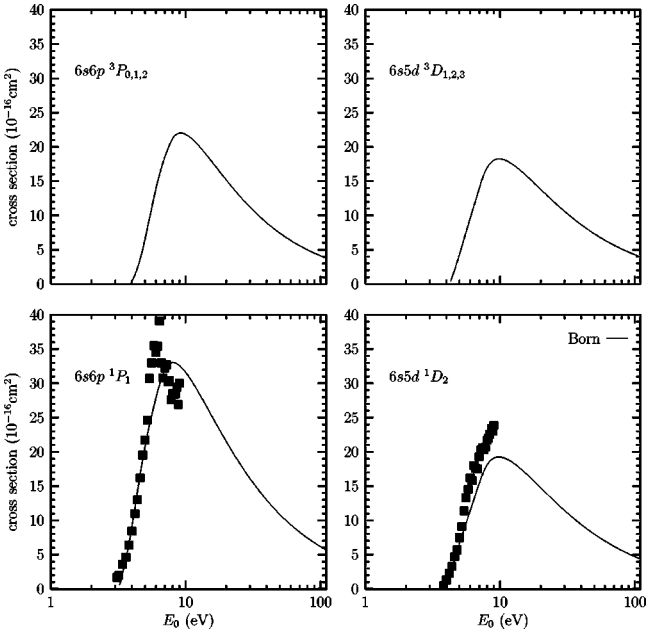

FIG. 6. Ionization cross sections σ_i^+ for electron scattering by the $6s5d\ ^3D_{1,2,3}$, $6s5d\ ^1D_2$, $6s6p\ ^3P_{0,1,2}$, and $6s6p\ ^1P_1$ states of barium. The solid lines represent the theoretical results, while the filled rectangles are the experimental points from Trajmar, Nickel, and Antoni [30].

Experimental σ_i^+ should have a second maximum at around 25 eV due to inner-core ionization (absent in the present model). For ionization from the $6s6p\ ^1P_1$ state we can compare the theoretical results with the experimental data of Trajmar, Nickel, and Antoni [30], which are consistent with that of Bushaw *et al.* [31]. We find good agreement with the experiment except perhaps in the energy region corresponding to the cross-section maximum. We believe that the conclusion, based on the experimental results, that ionization cross section from the $6s6p\ ^1P_1$ state is approximately two times larger than from the ground state is correct. For ionization from the metastable D states we can compare with experimental results of Trajmar, Nickel, and Antoni [30], which were perfomed with a mixture of 70% $6s5d\ ^1D_2$ and 30% $6s5d\ ^3D_2$ states. Due to similarity of the $6s5d\ ^1D_2$ and $6s5d\ ^3D_2$ cross sections we make less error by comparing the experiment with theoretical $6s5d\ ^1D_2$ cross section. A good agreement was found at low energies; however, our results become somewhat lower than the experiment for incident electron energies above 6 eV.

IV. CONCLUSIONS

Excitation and ionization of atomic barium by electron impact has been considered on a broad range of energies for various initial excited states. We find good agreement between CCC(115) and CC(55) calculations for most discrete transitions, which indicates relatively minor importance of coupling to the target continuum. For a number of transitions the effect of singlet-triplet mixing is very large. For some transitions differences between UFOMBT and CC results reveal strong dependence of the excitation cross sections on

the details of the Ba wave functions, and sometimes due to the neglect of interchannel coupling in the former theory. Ionization from the ground and first few excited states has also been estimated using the CCC formalism. The accuracy of these cross sections is somewhat less than those for the discrete transitions, owing to the requirement of very large computer-intensive calculations. The present results, therefore, should be considered as a step in establishing a reliable data base of excitation cross section for transitions involving excited states in Ba. Fully relativistic (Dirac Hamiltonian based) scattering theory should be developed to validate and improve the present results. Such work is underway at Flinders University.

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