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# Electron-impact excitation from the $(4p^55s)$ metastable states of krypton

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Theoretical results from multistate semirelativistic Breit-Pauli *R*-matrix calculations and two first-order distorted-wave calculations are presented for electron-impact excitation of krypton from the $(4p^55s)$ $J=0,2$ metastable states to the $(4p^55s)$ and $(4p^55p)$ manifolds. Except for a few cases, in which the method to account for relativistic effects becomes surprisingly critical, fair overall agreement between the predictions from the various theoretical models is achieved for intermediate and high energies. However, significant discrepancies remain with the few available experimental data.

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## I. INTRODUCTION

Electron collisions with noble gases have been a topic of continuous interest for both fundamental and practical reasons. From a purely theoretical point of view, accurate calculations for electron-impact excitation of all the noble gases but helium from their ground state $(np^6)^1S$ have proven to be very challenging (see, for example, Refs. [1–3] and references therein), and agreement with the few available experimental data is not always satisfactory. On the other hand, data for these transitions, as well as for excitation from metastable initial states, are in high demand for modeling applications in the discharge physics associated with gas lasers and the lighting industry [4–6].

Interestingly, describing electron-induced transitions from the metastable levels, $(np^5[n+1]s)^3P_{2,0}$ ($1s_5$ and $1s_3$ in Paschen notation), seems somewhat easier for theory than handling transitions from the ground state. This is mostly due to the much smaller energy transfer associated with these transitions. As a result, many of the important cross sections, particularly for the optically allowed transitions, are several orders of magnitudes larger than the cross sections for the corresponding transitions from the ground state. As a further consequence, one can expect that perturbative approaches will become valid at relatively low *absolute* energies, since the importance of channel coupling is generally determined by the *ratio* of incident energy and characteristic excitation energies. The possibility of successfully combining results from a "low-energy" Breit-Pauli *R*-matrix (close-coupling) theory [1] with a "high-energy" distorted-wave approach was demonstrated by Maloney *et al.* [7] for the case of electron-impact excitation of the $(3p^54s)\rightarrow(3p^54p)$ transitions in argon.

From an experimental point of view, on the other hand, measurements of excitation cross sections from the metastable initial states are generally considered to be even more difficult than those for targets in the ground state. In the *e*-Ar case, for example, experimental data from a Russian collaboration [8,9] differed dramatically from those reported by the Wisconsin group [10,11], with the latter being in much better and actually quite satisfactory agreement with theoretical predictions [1,7]. The experimental difficulties include the preparation of a metastable target, possibly competing signals from excitations of ground-state atoms, cascading, and problems in separating excitation signals originating from the two $1s_5$ and $1s_3$ initial states.

In light of the urgent need for these data in modeling applications for the krypton target, the very few currently available experimental data [12,13] and theoretical predictions [14], on-going experiments in the Wisconsin group [15], and the promise of being able to provide reliable predictions from our theoretical models, we extended our recent work on electron-impact excitation of krypton in the $(4p^6)^1S_0$ ground state [3] to include transitions from the metastable excited states $(4p^55s)^3P_2$ $(1s_5)$ and $(4p^55s)^3P_0$ $(1s_3)$.

## II. SUMMARY OF THE THEORETICAL MODELS

The calculations reported here were performed along the lines described in the recent paper by Dasgupta *et al.* [3] and hence the details will not be repeated here. Two semirelativistic Breit-Pauli *R*-matrix (close coupling) calculations, to be referred to as BP15 and BP51 below, were performed, as well as two independent distorted-wave calculations, to be labeled as DW-1 and DW-2, respectively. Details of these methods can be found in the above paper, as well as the references given therein. Very briefly, the BP51 model coupled 31 physical states with configurations $4p^6$, $4p^55s$, $4p^55p$, $4p^54d$, and $4p^56s$, as well as 20 pseudostates with configurations $4p^5\overline{6p}$ and $4p^5\overline{7p}$, respectively. The principal reason for including the latter states was the fact that the $\overline{6p}$ and $\overline{7p}$ pseudo-orbitals were constructed to improve the target description by effectively allowing for some term dependence in the bound orbitals. In the simpler BP15 calculation, only states with the configurations $4p^6,4p^55s,4p^55p$ were coupled. Finally, relativistic effects were accounted for by including the one-electron terms of the Breit-Pauli Hamiltonian in the diagonalization of both the

 

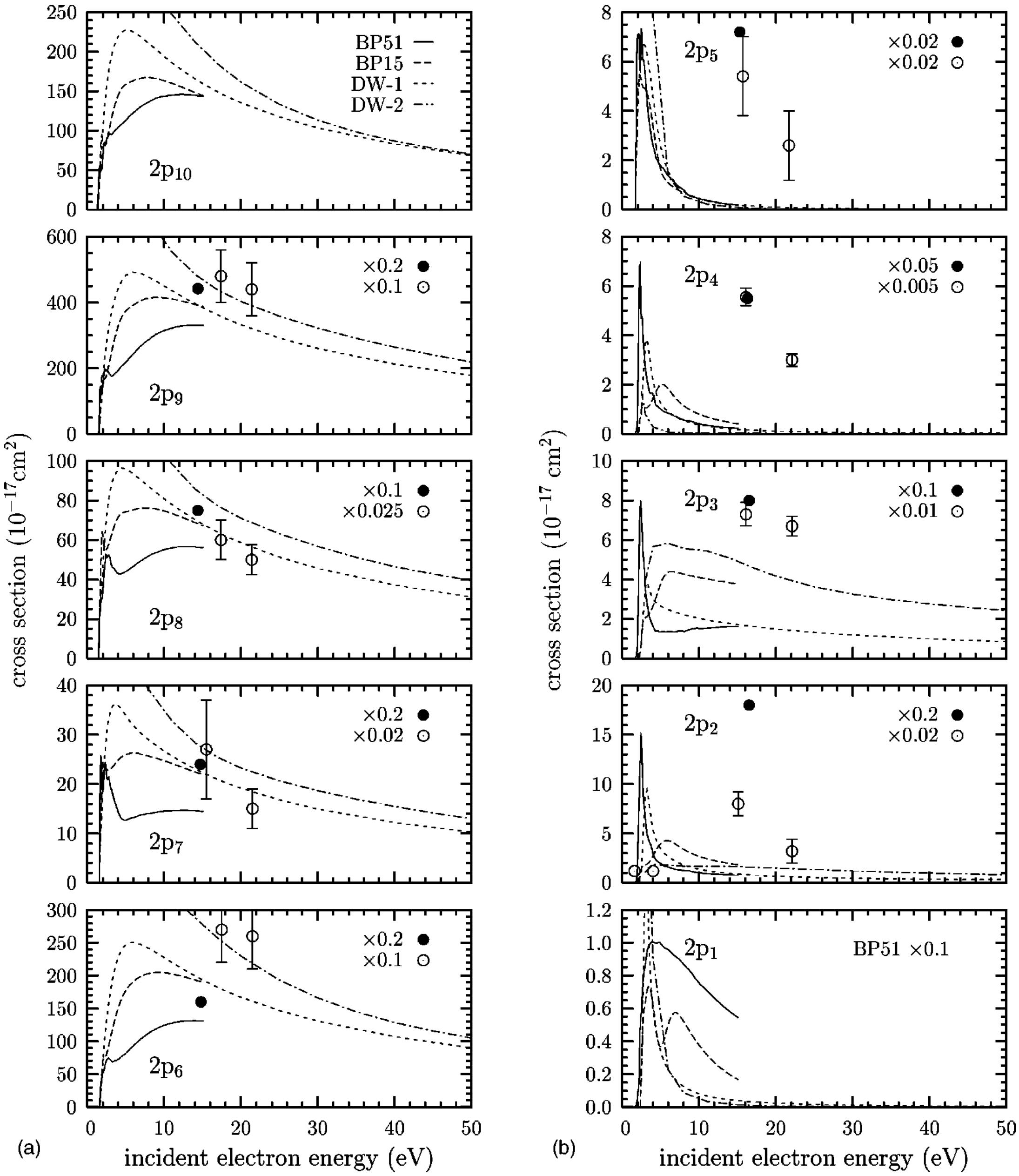


FIG. 1. Cross sections for electron-impact excitation of krypton from the $1s_5$ $(J=2)$ state to the $4p^5 5p$ manifold as a function of the collision energy. The experimental data of Kolokolov and Terekhova [13] (solid circles) and Mityureva *et al.* [12] (open circles) have been multiplied by the factors indicated.

*N*-electron target and the $(N+1)$-electron collision problem.

As described in Dasgupta *et al.* [3], the most important differences between the two distorted-wave approaches are the following: (1) the DW-1 calculation uses a semirelativistic method to calculate bound-state wave functions that are optimized for each final state while the DW-2 calculation uses the same bound-state wave functions as the BP15 calculation; (2) the DW-1 calculation does not include relativistic effects in the calculation of the distorted waves while DW-2 does; and (3) the DW-1 calculation unitarizes the *S*-matrix while DW-2 does not. Note that the lack of unitarization often results in a steep nonphysical increase in distorted-wave cross sections near threshold (see, for example, Fig. 2 of Maloney *et al.* [7]). However, if the ultimate goal is to combine the close-coupling predictions (generally more reliable for low collision energies) with distorted-wave results at higher energies, then this problem is not significant.

## III. RESULTS

Results for the direct excitation cross sections of the states in the $(4p^5 4p)$ manifold from the initial metastable states $1s_5$ $(J=2)$ and $1s_3$ $(J=0)$ are presented in Figs. 1 and 2, as a function of the incident-projectile energy. For excitation from the $1s_5$ state, our predictions are compared with the

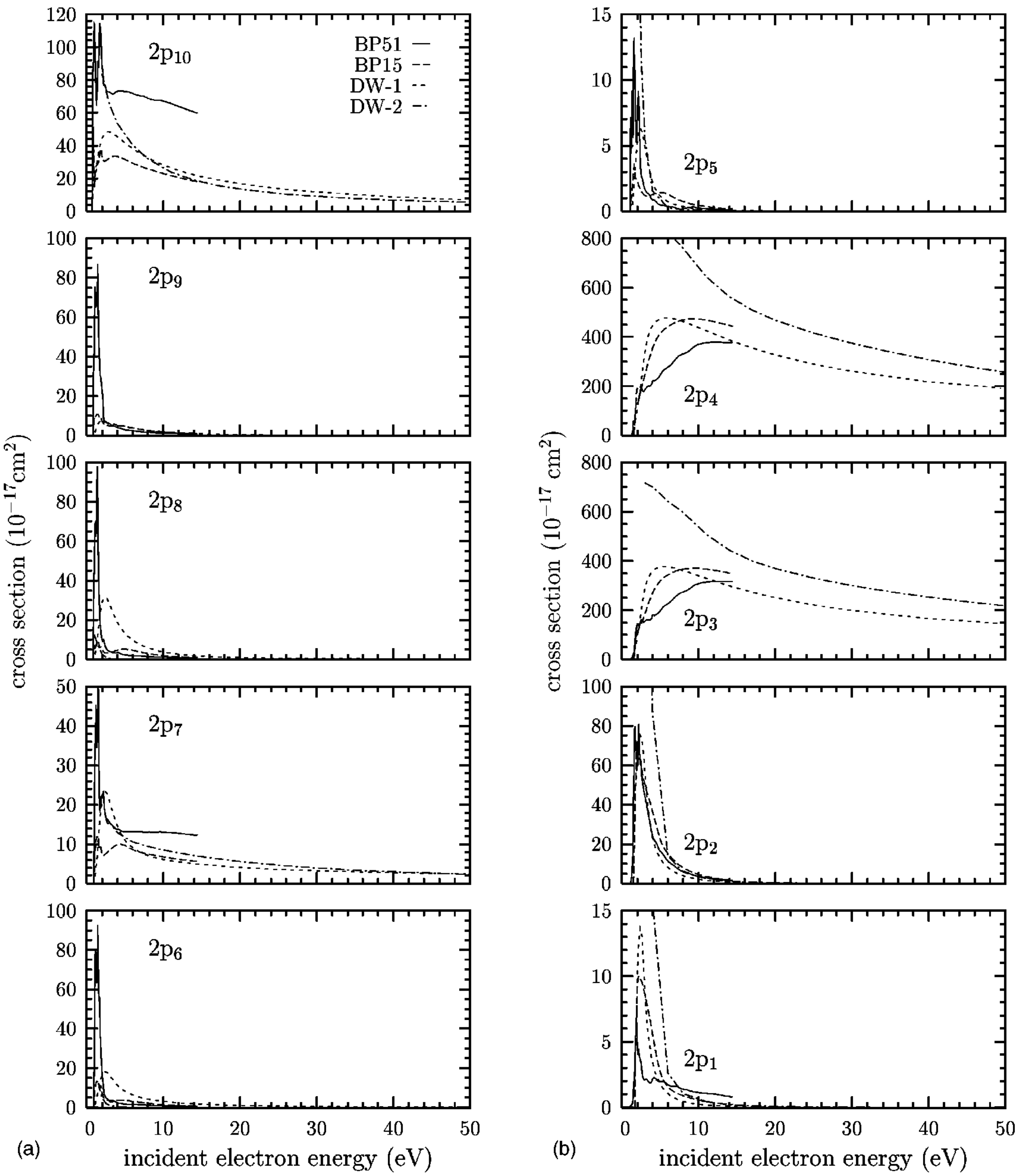


FIG. 2. Cross sections for electron-impact excitation of krypton from the $1s_3$ ($J=0$) state to the $4p^5 5p$ manifold as a function of the collision energy.

experimental data of Mityureva *et al.* [12] and of Kolokolov and Terekhova [13].

As can be seen from Figs. 1 and 2, the agreement between the predicted cross sections from the various theoretical approaches is generally fair, while agreement with the experimental data is virtually nonexistent. In order to even fit the experimental points on the graphs without extending the scale dramatically, the published values had to be reduced by one to two orders of magnitude. However, based on previous experience for electron collisions with metastable argon atoms [10,11,16], this disagreement is not really surprising. In fact, it was to some extent expected and provided motivation for the present work.

Nevertheless, potential problems remain in the theoretical results, particularly for relatively small cross sections. As was already the case for excitation from the ground state [3], the BP51 model predicts significantly different results for excitation of the $2p_1$ state than BP15 and the distorted-wave models. This discrepancy between the various theoretical predictions can be traced back to the difference in the muliconfiguration description of the target state. Fortunately, however, the cross sections for exciting this state are relatively small and, therefore, we do not expect these differences to cause major problems when the present results are being used in modeling applications.

A very interesting point in the theoretical results concerns the excitation of the transitions $1s_3 \rightarrow 2p_9$, $1s_3 \rightarrow 2p_8$, and $1s_3 \rightarrow 2p_6$. Although the predicted cross sections are small

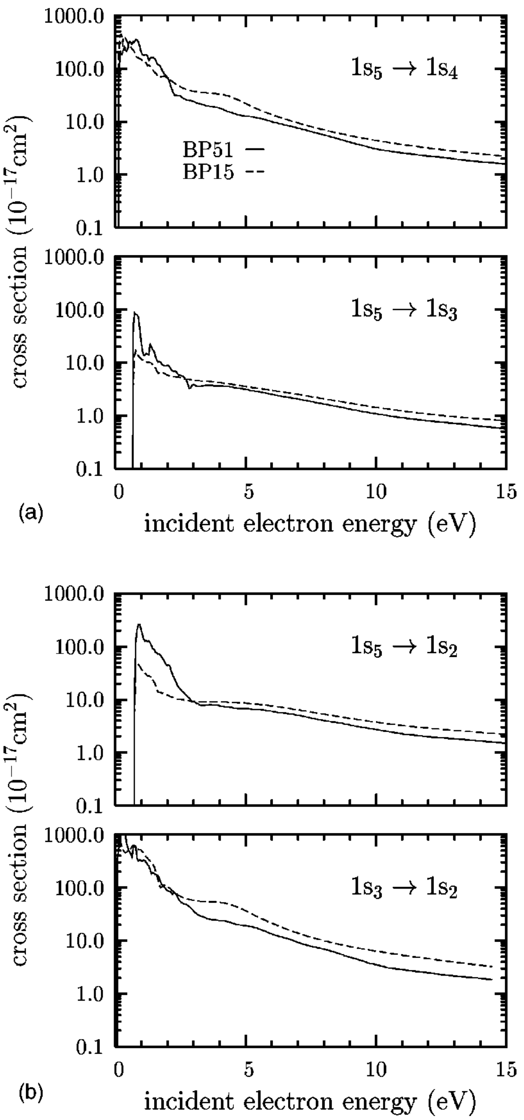


FIG. 3. Cross-section predictions from the BP15 and BP51 models for electron-impact-induced transitions in krypton from the $1s_5$ and $1s_3$ metastables states to other members of the $4p^55s$ manifold as a function of the collision energy.

(see also below), we note that the DW-2 result for $1s_3 \to 2p_9$ ($\Delta J = 3$) is *exactly zero* and the DW-2 predictions for the other two transitions ($\Delta J = 2$) fall off much faster with increasing energy than the DW-1 and the close-coupling results. Our preliminary analysis suggests the following reason for these somewhat surprising differences: In the DW-2 method, it is assumed that the *total electronic angular momentum J* of the target is well defined *during the collision*. In the relativistic treatment of Madison and Shelton [17], the atom therefore undergoes a transition from an initial state with $J_0$ to a final state with $J_1$. The $J$ transfer ($\Delta J$) is composed of orbital angular momentum ($\Delta L$) and spin-change ($\Delta S$) transfers. For the present case, $\Delta L$ must be unity since the active target electron undergoes an $s \to p$ change. Furthermore, $\Delta S$ can be either zero (no spin change) or one (spin change). Consequently, $\Delta J$ is limited to (0,1,2) and thus the $1s_3 \to 2p_9$ transition ($\Delta J = 3$) is strictly forbidden in this coupling scheme. The $1s_3 \to 2p_8$ and $1s_3 \to 2p_6$ transitions ($\Delta J = 2$) are allowed, but only through a spin change. Therefore, they exhibit a decrease proportional to $E^{-3}$ with increasing incident energy $E$ that is typical for exchange cross sections.

In the DW-1 treatment, as well as in the Breit-Pauli *R*-matrix models, relativistic effects are only treated in first-order perturbation theory, together with unitarization (forced in DW-1, automatic in RM) and recoupling from a nonrelativistic $LS$-scheme to a relativistic scheme that distinguishes between different final $J$ values of the target. The latter treatment is often associated with the ''Percival-Seaton'' [18] or ''Rubin-Bederson'' [19] hypothesis (see also Csanak *et al.* [20] for comments), i.e., it is assumed that the collision is so ''fast'' that the $J$ value of the target is only established properly through inner-atomic spin-orbit coupling a long time after the actual collision is over. If this angular-momentum coupling scheme is used, $^3P^o \to {}^3D^e$ transitions are possible through *direct* processes. The latter produce cross sections that decrease with increasing energy as $\log(E)/E$, and this high-energy dependence is clearly seen in the corresponding panels of Fig. 2.

Note that recoupling of nonrelativistic results is a common procedure to predict results for fine-structure resolved transitions. It is often used with great success, but typically is justified by a comparison of recoupled results with those that were calculated in a fully relativistic scheme. It seems as if the procedure could be problematic in the above cases. We plan to further investigate this topic, but note here that these strong differences in the predicted energy dependence might offer an interesting opportunity for an experimental check. The major difficulty would be to fully isolate the initial state as $1s_3(^3P_0^o)$ in order to avoid contamination of the signal originating from excitation out of the $1s_5(^3P_2^o)$ state.

As one might have expected, we also see the dominance of core-preserving over core-changing transitions in the theoretical predictions. Note that the $1s_5$ and the $2p_{10}$-$2p_5$ states are associated with the $(4p^5)^2P_{3/2}$ core of $Kr^+$, whereas $1s_5$ and $2p_4$-$2p_1$ are built from the $(4p^5)^2P_{1/2}$ core. Except for collisions very close to threshold, where the BP51 model sometimes predicts very sharp peaks that are not seen in the other models, the core-preserving transitions $1s_5 \to 2p_{10}, \ldots, 2p_5$ and $1s_3 \to 2p_4, \ldots, 2p_1$ are found to be significantly stronger than the core-changing transitions $1s_5 \to 2p_4, \ldots, 2p_1$ and $1s_3 \to 2p_{10}, \ldots, 2p_6$, respectively.

For the most important transitions shown in Figs. 1 and 2, namely, $1s_5 \to 2p_{10}$, $1s_5 \to 2p_9$, $1s_5 \to 2p_8$, $1s_5 \to 2p_6$, $1s_3 \to 2p_4$, and $1s_3 \to 2p_3$, we actually performed distorted-wave calculations calculation for incident energies up to 200 eV. As expected, the trend in the level of agreement between the predictions from these two models continues beyond 50 eV. The principal reason for the deviations of the two predictions from each other at high energies are the small differences in the intermediate-coupling coefficients used for these states (see Table 1 of Dasgupta *et al.* [3]), as well as differences in the orbitals. The sensitivity of the results to these differences depends on the transition of interest but is relatively small compared, for example, to typical experimental uncertainties in the absolute normalization of total cross sections.

Finally, Fig. 3 presents BP15 and BP51 predictions for transitions between the $(4p^5 5s)$ levels. These transitions, too, may become very important in low-energy plasmas since they allow for the possibility of moving an electron from a metastable state with $J=0,2$ to a state with $J=1$ that can decay radiatively to the ground state. Note that these cross sections are predicted to be extremely large at very low projectile energies, with a rapid drop-off for increasing energy that is typically for such forbidden transitions.

## IV. SUMMARY

To summarize, we have presented results from several sets of calculations for electron-impact excitation of the krypton $(4p^5 5s)$ and $(4p^5 5p)$ states from the metastable $(4p^5 5s)$ $1s_5$ and $1s_3$ levels. Overall, the agreement between the predictions from the different theoretical models, including perturbative and nonperturbative approaches using different target descriptions and approximations to account for relativistic effects, was fair and comparable to the situation for the corresponding transitions from the ground state.

On the other hand, the agreement with the few published experimental data is extremely poor. However, similar work on *e*-Ar collisions already suggested that such discrepancies would be very likely. In light of the difficulties associated with experimental investigations of these transitions, the apparent success of our methods for the argon target, and the somewhat low probability of several independent theories being consistently wrong by several orders of magnitude, we are confident that the present datasets are a valuable addition to the database used for *e*-Kr collisions in the modeling of gas discharges. This confidence is further boosted by comparison with preliminary (unpublished) data of the Wisconsin group [15] that are in much closer agreement with our predictions than the data shown in Fig. 1.

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