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Excitation of the $3p^5 5p$ levels of argon from the $3p^5 4s$ metastables

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ABSTRACT

Measurements have been reported recently of cross sections for electron-impact excitation of the argon $3p^5 5p$ manifold of levels from the $3p^5 4s$ metastable levels. We report results of calculations of the direct excitation cross sections at electron-impact energies from threshold to 500 eV for these transitions. The collision cross sections are calculated in the scaled plane-wave Born approximation (scaled PWB) developed by Kim. This scaling transforms the Born cross sections for dipole-allowed and spin-allowed excitations into reliable cross sections that compare as well with accurate measurements as does the sophisticated and more complex convergent close coupling method. We have used *jj* coupling in a single *LS* configuration Dirac-Fock calculation to describe the target wavefunctions. The optical emission cross section measurements, by Jung et al., include estimated contributions of 25% or more from cascading from higher excited levels and so are larger than direct excitation cross sections. Nevertheless, our cross sections agree reasonably well with the measurements for transitions where the core *j* value remains unchanged, except for the $1s_5$–$3p_8$ excitation. The results do not agree well with the measurements when the core *j* value changes, as similarly seen in the results of recent relativistic distorted wave calculations by Sharma et al.

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1. Introduction

Measurements have been reported recently [1] of cross sections for excitation of argon $3p^5 5p$ manifold of levels from the $3p^5 4s$ metastable levels. In Paschen notation, these are transitions from the $1s_3$ and $1s_5$ metastables to the $3p_n$ ($n = 1$–10) levels. These cross sections are important because, like excitation to the lower set of levels in the $3p^5 4p$ or $2p_n$ manifold, the excitation energy is rather low, on the order of 3–4 eV, and the cross sections can be large, on the order of 1×10^{-20} to 5×10^{-20} m^2. Some of these transitions are in the 395–470 nm range and are useful for optical emission spectroscopy plasma diagnostics [2]. In addition, optical emission spectroscopy of similar transitions have been used to estimate the number densities of argon atoms in the $3p^5 4s$ metastable and resonance states [3]. More generally, cross sections for these transitions are important in computer models of low temperature plasma conditions where the metastable populations can be large.

We have carried out calculations at electron-impact energies from threshold to 500 eV for transitions to the 10 levels of the $3p^5 5p$ $3p_n$ manifold from the $1s_3$ and $1s_5$ metastable levels of argon. We use single configuration *LS* target wavefunctions in *jj* coupling and use the multiconfiguration Dirac-Fock (MCDF) computer program of Indelicato and Desclaux [4] in the optimized level mode. The direct collision cross sections are calculated in the scaled plane-wave Born (PWB) approximation developed by Kim [5]. This method is applicable to spin-allowed and dipole-allowed transitions of neutral atoms and has been shown [5,6] to give good agreement with accurate theoretical and experimental data for excitations in H, He, Li, Be, Na, Mg, K, Ca, Rb, Sr, Cs, Ba, Hg, and Tl. This method allows the use of simple single configuration wavefunctions and yet provides rapid and reliable calculations of direct excitation cross sections for many neutral atoms [5,6].

The scaled plane-wave Born approximation produces only integrated cross sections unlike the R-matrix [7] and convergent close coupling [8] methods which can be used to obtain differential cross sections, Stokes parameters, etc. But the ease of application combined with orders of magnitude less computational effort makes the scaled plane-wave Born approximation very attractive when only integrated cross sections over a large energy range are needed. As is well known, the PWB approximation, or variants thereof, is unable to predict resonance structures near threshold. However, when only averaged integrated cross sections over an energy range near threshold are required such as in modeling of plasmas in lamps, gas discharges, fusion devices and stellar atmospheres, scaled PWB cross sections provide useful estimates. The scaled PWB approximation has been applied to atoms with simple electronic configurations with few energy levels [5]. In the present paper, we study the application of this method to excitation between levels arising from two open shells in argon where electron repulsion and spin-orbit coupling give rise to a manifold of closely spaced levels

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and compare the performance of the method with results from the relativistic distorted wave method.

Jung et al. [1] estimate that cascade effects can be significant in the optical emission spectroscopic measurements of excitation to the 3p manifold. Based on Bethe–Born calculations, they conclude that cascade contributions could be 25% or larger, especially for levels with small direct excitation cross sections (i.e., excitation from the argon metastable levels to the dipole forbidden $3p_1$ and $3p_5$ levels). In the experiment higher levels are excited which cascade down to the upper level that is observed in the optical experiment. In the situation for the $3p_n$ levels, cascading occurs from the $3p^5 4d$ and $3p^5 6s$ levels. These are weakly excited from the metastable $1s_n$ levels, but they decay strongly to the $3p_n$ upper levels because they are close in energy and have good overlap of the excited orbital wavefunctions. Cascading is relatively less important when the direct excitation cross section is large, on the order of 10^{-20} m^2. The measurements of optical excitation functions to the 3p manifold include cascading and are thus "apparent" cross sections rather than "direct" [1].

Calculations of the direct cross sections for these transitions by a relativistic distorted wave method (RDW) have been carried out by Sharma et al. [9] using relativistic wave functions for the bound states and the continuum states of the scattered electron. Their results are at electron-impact energies of 4 eV and higher. They use the GRASP92 computer program [10] to calculate the wave functions of upper and lower levels of the transitions. For the upper levels of the $3p_n$ manifold, they use a multiconfiguration Dirac-Fock approximation with $3p^5 4\underline{p}$, 4p, $5\underline{p}$ and 5p configurations, where $\underline{p}$ indicates $j = 1/2$ and the absence of the underline indicates $j = 3/2$. Their cross section results agree reasonably well with the measurements for allowed transitions where the angular momentum of the $3p^5$ core j_c, does not change. When j_c changes, their results do not agree well with the measurements. They also report results for forbidden transitions, which are very small.

2. Outline of the collision model

The plane-wave Born scaling model has been formulated by Kim [5]. The method starts with the PWB cross section, σ_{PWB}, written as

$$\sigma_{\mathrm{PWB}} = \frac{4\pi a_0^2 R}{T} F\mathrm{PWB}(T)$$

where T is the incident electron energy, a_0 is the Bohr radius (0.529 Å), and R is the Rydberg energy (13.6 eV). The F_{PWB} is a constant times the standard definition of the collision strength.

The scaling, called BE scaling, replaces the incident electron energy T in the denominator of the plane-wave Born cross section by $T+B+E$. Upon changing the incident electron energy in the denominator, we have for the BE scaled cross section, σ_{BE}, the expression where B is the binding energy of the atomic orbital that is excited, and E is the excitation energy. This change in the denominator of the impact cross section is similar to the modification first introduced by Burgess [11] to allow for acceleration of the incident electron as it approaches the target. The scaling reduces the Born cross section

$$\sigma_{BE} = \sigma_{\mathrm{PWB}} \left(\frac{T}{T+B+E} \right)$$

at low energies and shifts the peak of the cross section to higher energies. The scaled cross section approaches the PWB values at high energies above the energy of the peak cross section. Kim [5] has shown many examples in which the BE scaling transformed PWB cross sections for dipole-allowed and spin-allowed excitations into reliable cross sections that compare well with measurements.

Table 1
Our calculated fractional contributions and those of Sharma et al. [9] (in parenthesis) for the *jj* configurations to the argon $3p^5 5p$ wavefunction. ($\underline{p}$ denotes $j = 1/2$, p denotes $j = 3/2$).

Configuration	$3\underline{p}^2 3p^3 5\underline{p}$	$3\underline{p}3p^4 5\underline{p}$	$3\underline{p}^2 3p^3 5p$	$3\underline{p}3p^4 5p$
$J=1$				
$3p_{10}$	0.4443 (0.4368)	0.0047 (0.0039)	0.5129 (0.5197)	0.0381 (0.0315)
$3p_7$	0.5383 (0.5385)	0.0056 (0.0084)	0.4556 (0.4513)	0.0005 (0.0008)
$3p_4$	0.0029 (0.0044)	0.9264 (0.9029)	0.0050 (0.0049)	0.0656 (0.0873)
$3p_2$	0.0151 (0.0167}	0.1029 (0.0841)	0.0204 (0.0197)	0.8616 (0.8778)
$J=2$				
$3p_8$	0.9258 {0.9571)		0.0718 (0.0367)	0.0024 (0.0057)
$3p_6$	0.0742 (0.0354)		0.9229 (0.9616)	0.0028 (0.0023)
$J=3$				
$3p_9$			1.0000 (0.9994)	

These examples strongly suggest that the BE scaling offers a simple but effective way to approximately allow for distortion, polarization and electron exchange effects in the PWB approximation.

3. Target structure and wavefunctions

We use a single *LS* configuration in a *jj* coupled Dirac-Fock calculation to describe the target wavefunctions. The outer orbitals are the 4s orbital for the $1s_3$ and $1s_5$ metastables and the $5\underline{p}$ ($j = 1/2$) and 5p ($j = 3/2$) orbitals for the $3p_n$ manifold. Table 1 shows the contributions of the jj configurations to the $3p^5 5p$ states. Adding the $3p^5 4\underline{p}$ and $3p^5 4p$ configurations to the Dirac-Fock calculation would change the contributions of the $3p^5 5p$ configurations only slightly. To check this we compared our results to the multiconfiguration Dirac-Fock calculations of Sharma et al. [9] who included both the $3p^5 4p$ and $3p^5 5p$ configurations. The $3p^5 4p$ configuration contributions are clearly much smaller and thus negligible. Our configuration weights for the $3p^5 5p$ configurations closely match those of Sharma et al. [9] except for the few contributions that are very small. In the present calculation of oscillator strengths and excitation cross sections, full account is taken of the non-orthogonality of orbitals of the two levels obtained from separate optimization as implemented in the MCDF code [4].

To further establish the validity of our use of simple $3p_n$ wavefunctions, we also calculated the similar simple $2p_n$ manifold wavefunctions and found good agreement with the multiconfiguration Dirac-Fock wavefunctions of Srivistava et al. [12]. Moreover, our $2p_n$ manifold wavefunctions give oscillator strengths that agree with B-spline calculated dipole oscillator strengths of Zatsarinny and Bartschat [13]. We therefore conclude that our simple target wavefunctions are adequate for our present cross section calculations.

Calculated dipole oscillator strengths for the excitations agree reasonably well with spectroscopic values [14], Sharma et al. GRASP values [9], and with the B-spline close coupling calculations of Zatsarinny and Bartschat [13]. Table 2 shows the oscillator strengths and the comparisons. Only the $1s_3$–$3p_{10}$ f value is noticeably too small, owing perhaps to the unusual sensitivity of the overlap integrals that contribute to the oscillator strength. Our oscillator strengths are not expected to be as accurate as those of the B-spline close coupling calculations [13] but our f values compare well enough to verify the utility of our wavefunctions for cross section calculations.

Table 2
Electric dipole *f* values for the excitation transitions.

Final level	Initial level							
	$1s_5$				$1s_3$			
	ZB [13]	NIST [14]	Present	GRASP [10]	ZB [13]	NIST [14]	Present	GRASP [10]
$3p_{10}$	1.92E−4	1.81E−4	1.30E−4	2.59E−4	8.71E−4	8.26E−4	9.83E−7	1.69E−3
$3p_9$	3.42E−3	3.58E−3	4.86E−3	3.79E−3				
$3p_8$	8.09E−4	7.38E−4	1.15E−3	1.34E−3				
$3p_7$	4.57E−4	4.49E−4	6.55E−4	7.33E−4	9.84E−5	6.40E−5	7.97E−5	3.50E−5
$3p_6$	3.91E−3	3.63E−3	6.03E−3	5.94E−3				
$3p_4$	2.52E−7		6.06E−7	3.74E−5	4.06E−3	4.26E−3	8.14E−3	6.63E−3
$3p_2$	7.04E−4	6.39E−4	1.55E−4	1.18E−3	5.09E−3	4.41E−3	4.81E−3	5.71E−3

4. Results

Our direct excitation cross sections are compared in Figs. 1–9 with apparent experimental cross sections [1] and the direct RDW cross section calculations of Sharma et al. [9]. Our calculated cross sections are tabulated in Table 3 at selected energies. We show the combined systematic and statistical uncertainty of the measurements.

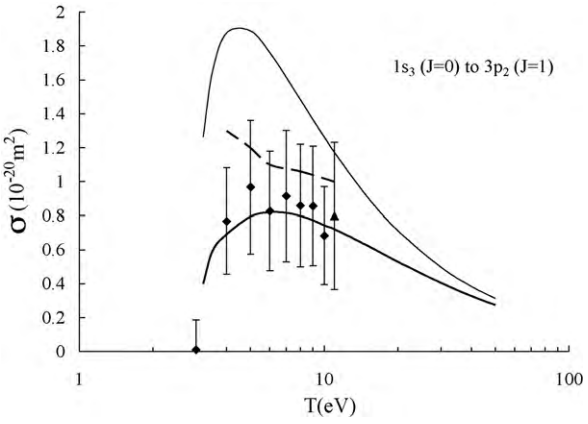

Fig. 1. Argon excitation cross sections from the $1s_3$ ($J=0$) metastable level to the $3p_2$ ($J=1$) level. The thick solid line in each figure is our scaled PWB calculation. The narrow solid line is the PWB approximation and the dashed line is the relativistic distorted wave theory of Sharma et al. [9]. The solid diamonds are the experimental measurements of Jung et al. [1]. The measurement uncertainties include both the relative statistical uncertainty and the ±40% systematic uncertainty quoted by the authors.

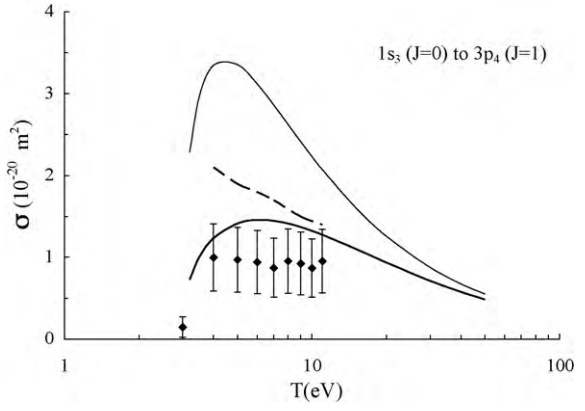

Fig. 2. Argon excitation cross sections from the $1s_3$ ($J=0$) metastable level to the $3p_4$ ($J=1$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is different.

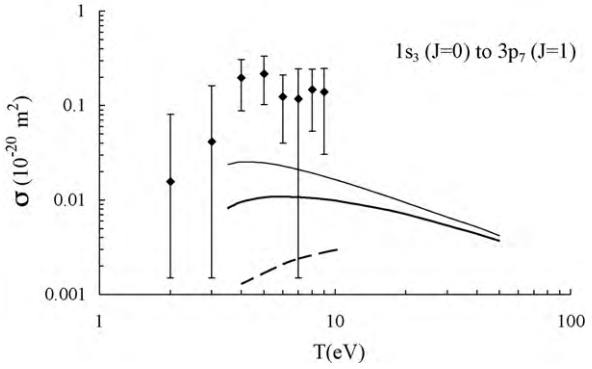

Fig. 3. Argon excitation cross sections from the $1s_3$ ($J=0$) metastable level to the $3p_7$ ($J=1$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is logarithmic.

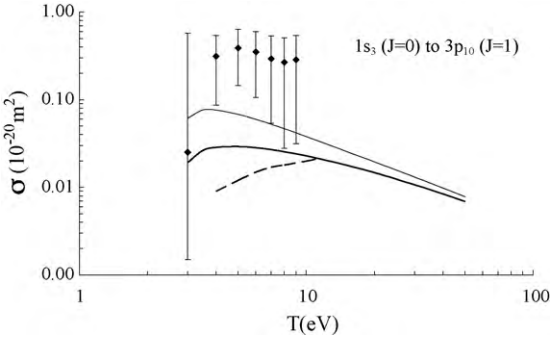

Fig. 4. Argon excitation cross sections from the $1s_3$ ($J=0$) metastable level to the $3p_{10}$ ($J=1$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is logarithmic.

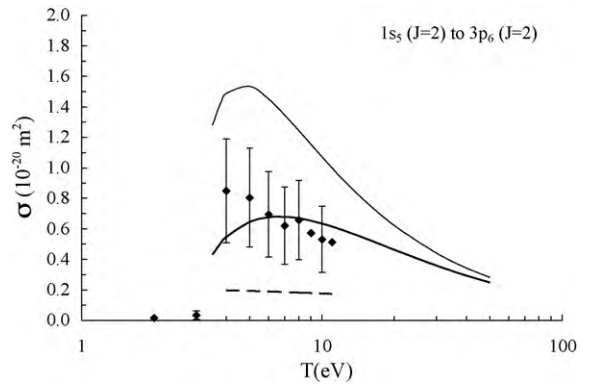

Fig. 5. Argon excitation cross sections from the $1s_5$ ($J=2$) metastable level to the $3p_6$ ($J=2$) level. Identification of the data and the uncertainties is the same as in Fig. 1.

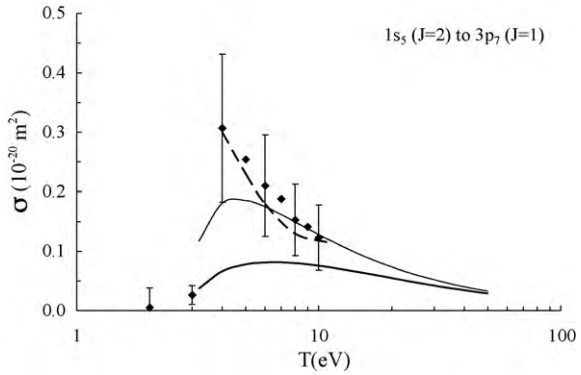

Fig. 6. Argon excitation cross sections from the $1s_5$ ($J=2$) metastable level to the $3p_7$ ($J=1$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is different.

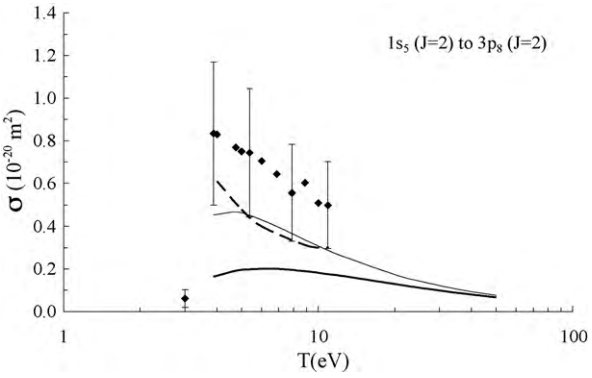

Fig. 7. Argon excitation cross sections from the $1s_5$ ($J=2$) metastable level to the $3p_8$ ($J=2$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is different.

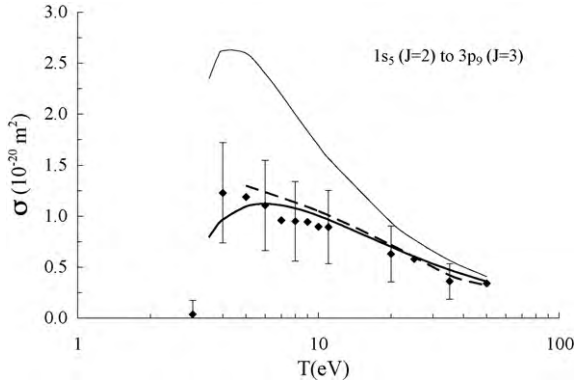

Fig. 8. Argon excitation cross sections from the $1s_5$ ($J=2$) metastable level to the $3p_9$ ($J=3$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is different.

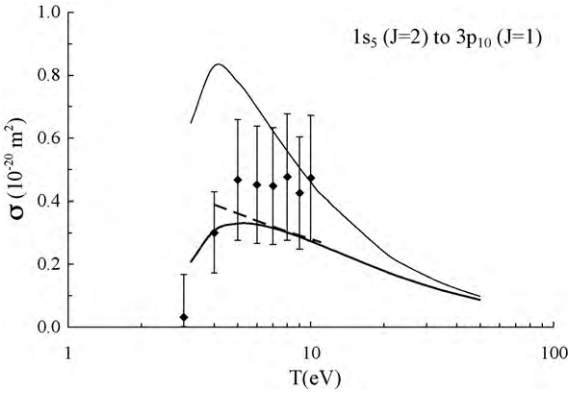

Fig. 9. Argon excitation cross sections from the $1s_5$ ($J=2$) metastable level to the $3p_{10}$ ($J=1$) level. Identification of the data and the uncertainties is the same as in Fig. 1. Note that the ordinate scale is different.

5. Discussion of results

Comparing our results to the measurements of Jung et al. [1] shows reasonable agreement, at least within the measurement uncertainty at 8 eV, for excitation from the $1s_5$ metastable to the $3p_6$, $3p_9$ and $3p_{10}$ levels of the 3p manifold ($J=2$ to $J=2$, 3 and 1, respectively). These are transitions where the $3p^5$ core J value, $J_c=3/2$, does not change in the excitation. Agreement with the measurements is not good for excitation from the $1s_3$ level to the $3p_7$ and $3p_{10}$ levels ($J=0$ to $J=1$) where J_c does change from 1/2 to 3/2 during the excitation. The poor agreement with experimental measurements when J_c changes was seen also in the relativistic distorted wave (RDW) calculations of Sharma et al. [9], and was seen in similar excitation cross sections for transitions from the metastable levels to the $2p_n$ manifold [2,15,16].

It has been observed by Jung et al. [1] that their cross sections for the $3p_n$ manifold of levels at 8 eV do not scale proportionately with the oscillator strength, as expected from the high energy Bethe–Born approximation. Proper scaling had been observed previously for the excitations to the $2p_n$ manifold of levels. We show in Table 4 that our calculated cross sections at 8 eV and 500 eV scale proportionately with f values quite well and better than that of the measurements for most of the transitions. We list the values of σ/f, σ being the present calculated cross section, normalized to the value for the calculated $1s_5$–$3p_9$ excitation cross section and f being the NIST value of the oscillator strengths. If f value proportionality were perfect, all the entries in the σ/f columns would be one. We see instead that most of the ratios for our calculated cross sections range from 0.5 to 2.0, suggesting that proportionality with the oscillator strength is nearly as expected from the Born–Bethe theory valid at high energy. Surprisingly, the proportionality is reasonably good for our calculated cross sections even at 8 eV where the first Born approximation is unrealistic.

Table 3
BE scaled cross sections (in 10^{-20} m^2) at selected energies.

Excitation i–j	4 eV	6 eV	8 eV	10 eV	50 eV	100 eV	500 eV
$1s_3$–$3p_{10}$	0.029	0.028	0.026	0.023	0.007	0.004	0.0005
$1s_3$–$3p_7$	0.010	0.011	0.011	0.010	0.004	0.002	0.0005
$1s_3$–$3p_4$	1.234	1.457	1.413	1.323	0.485	0.272	0.064
$1s_3$–$3p_2$	0.692	0.822	0.799	0.745	0.276	0.155	0.036
$1s_5$–$3p_{10}$	0.307	0.327	0.301	0.274	0.087	0.046	0.010
$1s_5$–$3p_9$	0.968	1.122	1.079	1.005	0.358	0.200	0.046
$1s_5$–$3p_8$	0.168	0.201	0.195	0.183	0.067	0.038	0.009
$1s_5$–$3p_7$	0.067	0.082	0.081	0.076	0.029	0.017	0.004
$1s_5$–$3p_6$	0.548	0.677	0.670	0.635	0.248	0.142	0.034

Table 4
Calculated (σ_{calc}) cross sections (10^{-20} m^2) at 8 and 500 eV and measured (σ_{msrd}) cross sections at 8 eV normalized to the $1s_5$–$3p_9$ values. Complete proportionality with f values as predicted by Bethe–Born theory would lead to values of one for σ/f for all transitions.

Excitation $i \rightarrow j$	f_{ij} NIST	σ_{calc} 8 eV	σ_{calc} 500 eV	σ_{msrd} 8 eV	σ_{calc}/f 8 eV	σ_{calc}/f 500 eV	σ_{msrd}/f 8 eV
$1s_3$–$3p_{10}$	0.000826	0.0255	0.0005	0.2677	0.10	0.05	1.22
$1s_3$–$3p_7$	0.000064	0.0105	0.0005	0.1478	0.54	0.61	8.70
$1s_3$–$3p_4$	.004260	1.4131	0.064	0.9547	1.10	1.17	0.84
$1s_3$–$3p_2$	.004410	0.7985	0.036	0.8602	0.60	0.64	0.74
$1s_5$–$3p_{10}$	0.000184	0.3014	0.010	0.4766	5.44	4.23	9.76
$1s_5$–$3p_9$	0.003580	1.0788	0.046	0.95	1.00	1.00	1.00
$1s_5$–$3p_8$	0.000738	0.195	0.009	0.56	0.88	0.95	2.86
$1s_5$–$3p_7$	0.000449	0.0805	0.004	0.153	0.59	0.69	1.28
$1s_5$–$3p_6$	0.003630	0.6697	0.034	0.6577	0.61	0.73	0.68

The major exception to proportional scaling is the transitions to the $3p_{10}$ level. The σ/f ratio varies greatly from the value one for both metastable initial levels, and is observed in both our calculated results and the measurements. Consistent with this deviation from proportionality, a Bethe/Fano plot (not shown) for this transition does not show straight line behavior proportional to the oscillator strength, indicating that the high energy Bethe–Born theory is not valid for this transition and proportionality cannot be expected. All the other transitions have Bethe/Fano plots that approach the straight line behavior with slopes proportional to their oscillator strengths, suggesting that they have approached the high energy region where the Bethe–Born theory applies. Also, it is interesting to note that the RDW calculations of Sharma et al. [9], not shown in Table 4, exhibit less proportional scaling than our calculations and less proportionality than the measurements. As clearly pointed out in [1], the measurements are of apparent cross sections rather than direct excitation measurements, meaning that cascading to the excited level from higher levels is included. Apparent cross sections are therefore expected to be larger than direct cross sections and, while cascading is not expected to be very large, it may nevertheless be part of the reason that f-scaling is not observed in the measurements. A more detailed explanation of this violation of proportionality for excitations to the $3p_n$ manifold, at low impact energy as compared to the $2p_n$ manifold, based on the validity of the plane-wave Born approximation has been given by Jung et al. [1].

Finally, using our calculated f values instead of the NIST f values in Table 4 gives similar σ/f values to those shown in Table 4. The discussion in the above paragraphs still holds. The Bethe–Born formulation shows dependence on f/E rather than just f, where E is the excitation energy. The values of excitation energy E for these transitions vary only slightly, however, and so it affects the scaling only slightly. Consequently, we have not needed to include the small E dependence in Table 4.

6. Conclusion

The simple single *LS* configuration Dirac-Fock wavefunctions used in our calculations seem to be adequate for the collision cross section calculations. This is evident from the calculated oscillator strengths shown in Table 2 and the contributions of the *jj* configurations shown in Table 1. The scaled plane-wave Born collision model gives reasonable agreement with the measured cross sections for excitations where the core j value does not change, in agreement with results of the RDW model of Sharma et al. [9].

A good calculation of cross sections requires accurate target wavefunctions to describe the electronic structure of the target as well as a suitable collision model. We have simple but reasonably accurate wave functions, which incorporate Coulomb interactions sufficiently well as the excited electron moves in different regions of configuration space outside a tight core in the two levels involved. Our wavefunctions also account for spin-orbit coupling by the use of relativistic wavefunctions. The validity of our wavefunctions is indicated by the *jj* configuration weights in Table 1 and oscillator strengths in Table 2. Our collision model, on the other hand, is a very simple first-order collision model. Our good agreement with measurements suggests that it is more important to use accurate wavefunctions than to use more elaborate collision models. This is consistent with many earlier uses of this model [5,6].

The fairly good agreement that we have with proportionality of the calculated cross section with oscillator strength implies that the lesser proportionality of the measurements is, in fact, caused by cascading contributions in the experiment. This has been suggested by the experimenter's themselves as a partial explanation. It gains more credence from our calculations. However, the severe disagreement between both our BE scaling and the RDW results and experimental results for core changing collisions still remains to be explained.

The RDW method is generally believed to be valid at intermediate and higher energies, and certainly the PWB calculations are valid only at higher energies. Our scaled PWB model [5] is, however, valid at all energies and is simple. These are important arguments in its favor, especially for generating many cross sections rapidly in computer models of plasmas and gases. The model cannot predict scattering resonances at low energies, as is done by more elaborate collision models, because it is a first-order perturbation model based on the plane-wave Born approximation. But it is capable of generating cross sections averaged over the resonance region and can provide useful data.

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