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Spin asymmetry in electron-impact ionization

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We investigate electron-impact single ionization of He-like ions of charge state $q = 0, \dots, 6$ in the initial ground ($1\,^1S$) and metastable ($2\,^3S$) states. Good agreement between theory and experiment is established for the total cross sections, and then the scaling behavior with q is considered as a function of the ratio $u = E_i/E_I \leqslant 10$ of the incident electron energy E_i and the ionization threshold E_I. While the expected E_I^2 scaling of the cross sections is confirmed, we also find that at each scaled energy u the spin asymmetry (for the $2\,^3S$ state) converges rapidly to a nonzero constant with increasing q. This indicates that, despite E_I increasing with q^2, exchange effects remain undiminished on the broad scaled-energy range considered. We suggest that this physical behavior is more universal than just for the He-like ions.

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I. INTRODUCTION

Electron-impact single ionization of atoms and ions is a rather mature field with benchmark cross-section data available for those targets that are readily modeled by one or two valence electrons above an inert core. The last long-standing discrepancy between theory and experiment, for the e-He($2\,^3S$) system [1], has been recently resolved by new measurements [2]. Today, the interest has shifted to more complicated processes such as ionization with excitation [3,4], and excitation of double-K-vacancy states associated with autoionization [5], for example.

With confidence in the accuracy of the calculations, and an abundance of supporting experimental data, it is also interesting to determine trends for targets with a specific electronic configuration as a function of ionic charge q. With increasing q the ionization threshold E_I tends to grow as q^2, and, therefore, rapidly larger incident-electron energies are required to ionize the target. Questions we would like to address are how do the ionization cross sections scale, and how important is the inclusion of electron exchange in the calculations with increasing q. Cross-section scaling has been addressed extensively for quite some time (see Burgess and Rudge [6], Burgess *et al.* [7], and Younger [8], for example). However, so far the effect of q on the contribution of electron exchange has escaped similar scrutiny. In part this is due to the fact that there are relatively few measurements of the ionization cross-section spin asymmetries. However, those that do exist, such as for e-H($1\,^2S$) [9,10], e-He($2\,^3S$) [11], and e-alkali [12] have been instrumental in establishing the utility of the convergent close-coupling (CCC) theory [13], and were helpful in the analysis of the cross-section threshold behavior [14].

For (nonrelativistic) targets that have a nonzero total electron spin s_i for the initial state, such as H- and Li-like ions ($s_i = 1/2$), or He-like ions in the metastable $2\,^3S$ state ($s_i = 1$), there are two independent total electron spins $S = s_i \pm 1/2$ contributing to the overall electron-impact collision process. The resulting spin-dependent total ionization cross sections may be labeled as $\sigma_{s_i \pm 1/2}$. The total ionization cross section and its spin asymmetry are then respectively given by

$$\sigma = \frac{s_i \sigma_{|s_i-1/2|} + (s_i+1)\sigma_{s_i+1/2}}{2s_i+1}, \tag{1}$$

$$A = \frac{s_i}{2s_i+1}\,\frac{\sigma_{|s_i-1/2|} - \sigma_{s_i+1/2}}{\sigma}, \tag{2}$$

where the use of $|s_i - 1/2|$ allows the formulas to be trivially valid for $s_i = 0$ targets, such as He($1\,^1S$), as then $S = 1/2$ is the only possible total electron spin.

The spin asymmetry A is a measure of the importance of electron exchange, and for the higher energies $A \approx 0$ due to $\sigma_{|s_i-1/2|} \approx \sigma_{s_i+1/2}$. Note, however, for $s_i = 0$ we always have $A = 0$, but this does not mean that electron exchange is not important. The spin asymmetry parameter A can also be written in terms of the ratio $r = \sigma_{s_i+1/2}/\sigma_{|s_i-1/2|}$. However, r has an infinite range, whereas A is always finite. At the maximum $A = 1$, which occurs whenever $\sigma_{|s_i-1/2|} \gg \sigma_{s_i+1/2}$. At the minimum $A = -s_i/(s_i+1)$, which occurs whenever $\sigma_{|s_i-1/2|} \ll \sigma_{s_i+1/2}$.

II. EXPERIMENT

We begin our investigation by considering electron-impact ionization of He-like ions. Over several years total ionization cross sections have been measured for various two-electron

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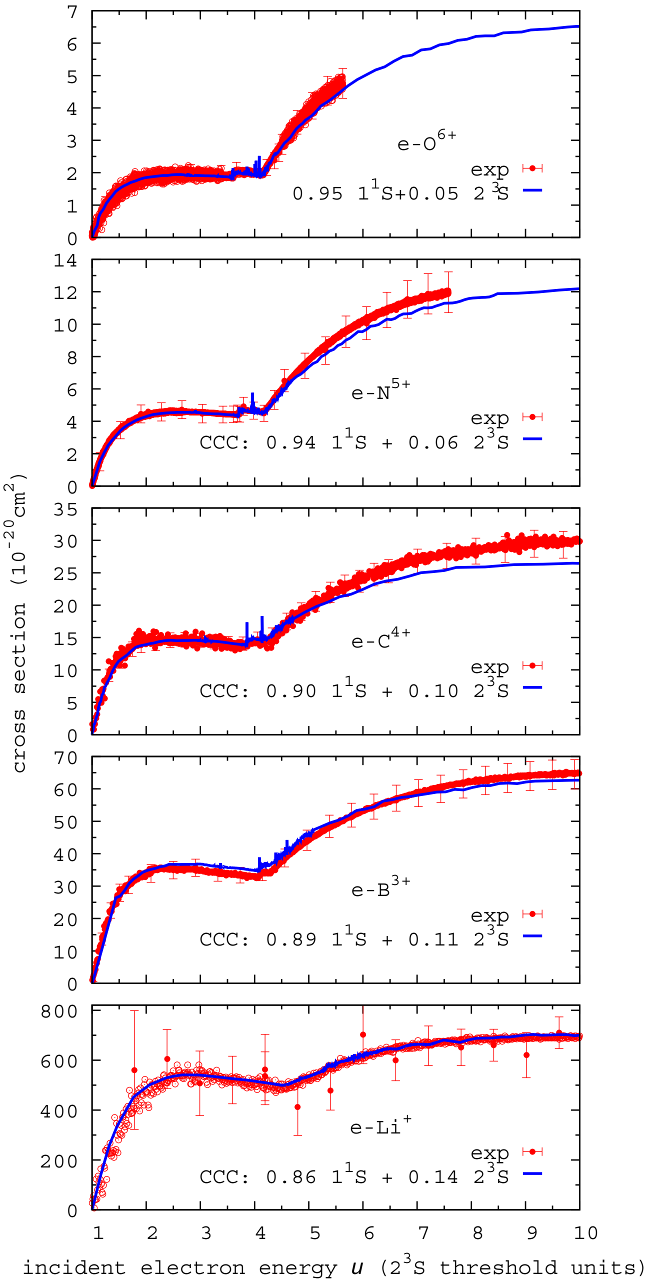

FIG. 1. Total cross sections for electron-impact single-ionization for the specified He-like ions. The experimental target beams were mixtures of the ground $1\,^1S$ and metastable $2\,^3S$ states (see text). The fraction of the states has been determined by best visual fit with the CCC-calculated cross sections. Note, u is the incident electron energy divided by the calculated ionization threshold of the corresponding $2\,^3S$ state given in Table I.

ions. Fairly intense, low-energy beams of multiply charged He-like ions for electron-impact ionization experiments can be produced by electron cyclotron resonance ion sources. Inevitably, beams extracted from such a hot-plasma ion source contain mixtures of ions in the $1\,^1S$ ground and metastable $2\,^3S$ states. Only for experiments with Li^+ ions pure ground-level beams can be readily produced. Results have been published for Li^+ ions in the $1\,^1S$ ground level and for Li^+ ion beams containing a mix of ions in the $1\,^1S$ ground and metastable $2\,^3S$ states [15]. Absolute total single-ionization cross sections were also measured for B^{3+} [16] and N^{5+} [17] ions. In order to provide experimental data for an extended range of charge states we have additionally measured absolute cross sections for electron-impact ionization of two-electron C^{4+} and O^{6+} ions. These data complete the picture for the He-like isoelectronic sequence in the region of low atomic numbers Z. All absolute cross-section measurements were carried out using the animated crossed-beams technique and the data reduction procedures as described in detail previously [5,15]. All experimental cross sections for electron-impact ionization of He-like Li^+, B^{3+}, C^{4+}, N^{5+}, and O^{6+} ions are shown in Fig. 1 together with the results of our theoretical calculations which are discussed next.

TABLE I. Ionization thresholds E_I (eV) for specified initial states of He-like ions as obtained in the CCC calculations in comparison with the data compiled in the NIST Atomic Spectra Database [24].

	$1\,^1S$		$2\,^3S$	
Target	NIST	CCC	NIST	CCC
Li^+	75.64	74.8	16.62	16.6
B^{3+}	259.37	258.2	60.81	60.8
C^{4+}	392.09	390.9	93.13	93.0
N^{5+}	552.07	550.9	132.27	132.1
O^{6+}	739.33	737.6	178.34	178.1

III. CONVERGENT CLOSE-COUPLING THEORY

The targets considered here are ideal for the CCC theory [18], which is applicable to light and heavy projectiles colliding with atomic and molecular targets [19–21]. Following its success in calculating the total ionization cross section and spin asymmetry for the e-H case [13] it has also been applied to fully differential ionization processes that led to the reanalysis of formal scattering theory [22] and the connection to computational methods [23]. Briefly, the foundation of CCC is the complete Laguerre basis which is used to diagonalize the target Hamiltonian to yield N square-integrable target states. These are then used to form close-coupling equations in momentum space for the projectile-target scattering system. Convergence in the results of interest, to the required precision, is obtained by systematically increasing N. With increasing N the negative-energy states converge to the true discrete eigenstates, while the positive-energy states provide an increasingly dense discretization of the target continuum. Ionization processes are associated with excitation of the positive-energy states. In the calculations presented here around $N_l = 30 - l$ Laguerre-based functions were taken for each $l \leqslant 4$. Due to the earlier interest in excitation-autoionization processes [5,15,17] the number of states generated was around 500. New CCC calculations were performed when required using similar principles. Due to the high-energy resolution in the experiments, to identify the various autoionizing states, the calculations were performed on an even finer energy grid.

Calculating the target structure is the first step of the CCC calculations. In the table the CCC-calculated and benchmark [24] ionization threshold energies E_I are presented for the He-like ions considered. Comparison shows good agreement with deviations of typically much less than 1% for the ground state and 0.1% for the $2\,^3S$ metastable level.

IV. RESULTS

In Fig. 1 we compare the absolute measurements with CCC calculations of electron-impact total ionization cross sections for the specified He-like ions. To make the presentations on a similar scale of incident-electron energy E_i we use threshold units $u = E_i/E_I$. The given relative fractions of the ground and metastable states in the beam have been determined by best visual fit using the corresponding components in the CCC theory. The autoionization resonances are visible around $u = 4$, with some detailed analysis provided previously [5,17]. Such good agreement between theory and experiment allows us to examine with confidence the scaling of the calculated cross sections with increasing q. This also has the capacity to identify any systematic discrepancies in the calculations, which are done independently at incident energies increasing rapidly with q.

In the top two panels of Fig. 2 we present the scaling with q of the CCC-calculated cross sections for the two initial states considered. Following previous work [6–8] we multiply the cross sections by E_I^2. Where available, good agreement is found with similarly scaled experimental data. In the bottom panel, the spin asymmetry, which is nonzero only for the $2\,^3S$ initial state, is plotted without any scaling factors. We see that the cross sections scale well for both considered initial states, being almost indistinguishable for $q \geqslant 5$. This is as would be expected. However, despite the increasing incident energies with q^2, the spin asymmetries not only increase with q, but converge rapidly to a constant at a given energy ratio u. In fact, the asymmetries are barely distinguishable for $q \geqslant 3$. The only available experiment, for He($2\,^3S$) [11], is in excellent agreement with the corresponding calculations, and these $q = 0$ results are quite similar to those for all q considered. Apart from the differing onset of the contribution to the total ionization of the autoionizing states the convergence to a constant at each u appears over the entire energy range considered.

Faced with spin asymmetry convergence, with increasing q, to a constant at each u for the case of metastable He-like ions we wonder how general this phenomenon might be. Accordingly, we revisit the calculations of electron-impact ionization of Li-like ions, for $q = 0, \ldots, 5$ [26]. In Fig. 3 we plot the spin asymmetries for the specified Li-like ions in the ground $2\,^2S$ initial state as a function of u. Once again we see rapid convergence at each u with increasing q, from threshold through to higher energies, with the only available experiment [Li($2\,^2S$) [12]] being representative of the spin asymmetries for all q considered. Consequently, we suggest that, with increasing ionic charge and fixed energy ratio u, the rapid convergence to a constant of the ionization cross-section spin asymmetries is a common feature not previously identified.

The results presented here raise some "why" questions. Why is $\sigma_{|s_i-1/2|} > \sigma_{s_i+1/2}$, but never overly dominant, over the considered energy range? Why does A, and therefore $r = \sigma_{s_i+1/2}/\sigma_{|s_i-1/2|}$, rapidly converge to a constant with increasing q for a given E_i/E_I? There are no simple answers to these questions. We already considered the former in the context of the near threshold behavior of the cross section

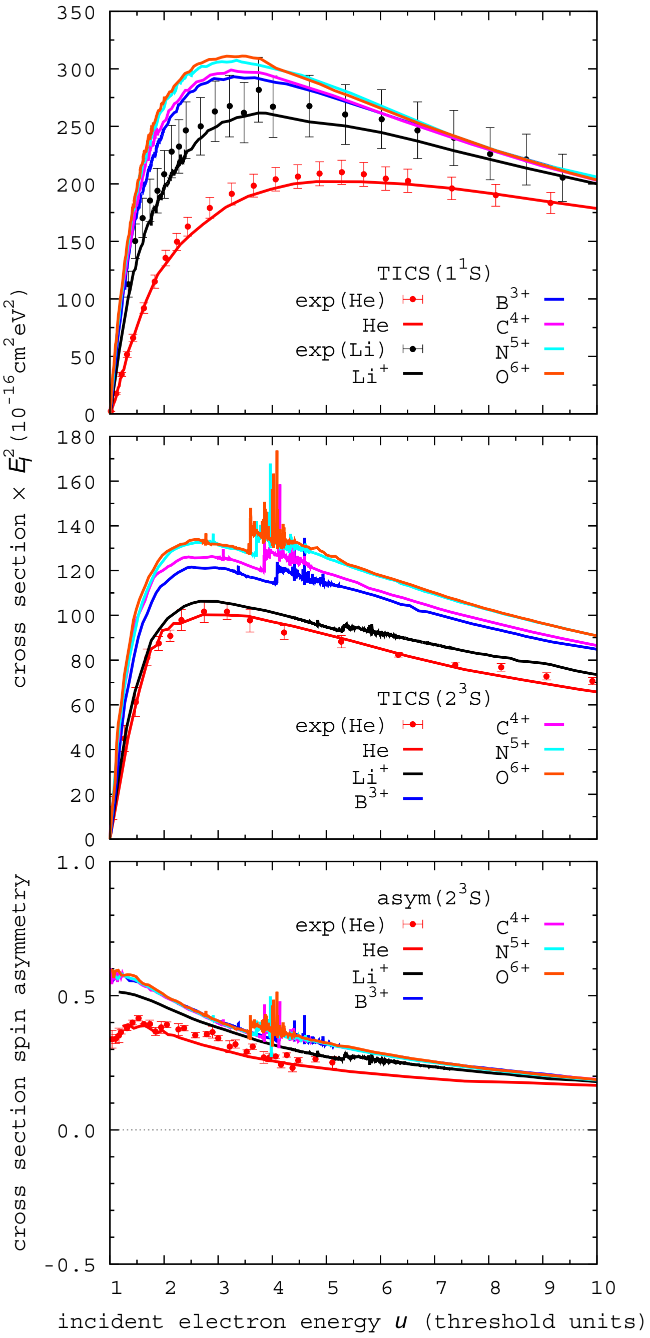

FIG. 2. Scaled (by E_I^2) total cross sections for electron-impact single ionization of the specified He-like ions in either the ground $1\,^1S$ or metastable $2\,^3S$ states. For the latter initial state, total ionization cross-section spin asymmetries are also presented. The data is plotted against u, the incident-electron energy divided by the appropriate ionization threshold energy. The helium cross-section data for $1\,^1S$ is due to Rejoub *et al.* [25], while for $2\,^3S$ the cross sections and spin asymmetries are due to Génévriez *et al.* [2] and Baum *et al.* [11], respectively. The spin-asymmetry range has been determined by the minimum and maximum values of Eq. (2). The Li$^+$ data for the ground state were obtained by Borovik Jr. *et al.* [15].

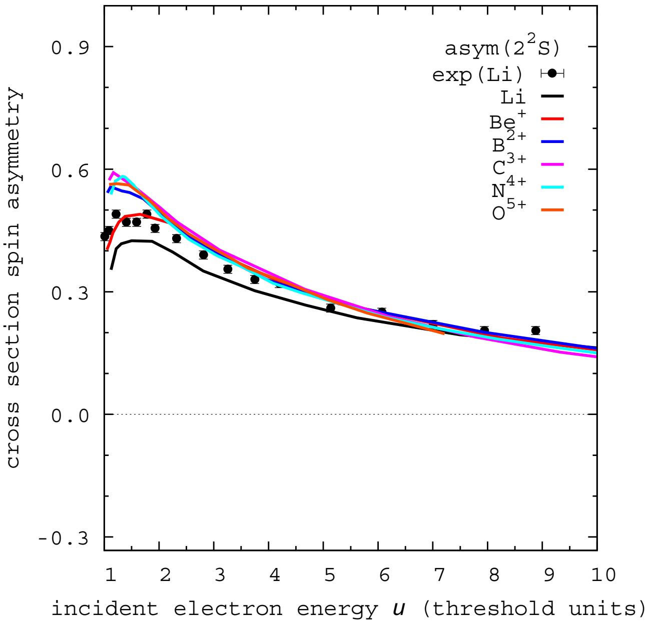

FIG. 3. Spin asymmetries of the total cross sections for electron-impact single ionization of specified Li-like ions plotted against u, the incident electron energy divided by the ionization threshold energy of the corresponding $2\,^2S$ state. The range has been determined by the minimum and maximum values of Eq. (2). The CCC calculations are from Ref. [26]. The e-Li experiment is due to Baum *et al.* [12].

for the e-H system [14]. In the CCC equations for $s_i = 1/2$ quasi-one-electron targets (H- and Li-like) the matrix elements for the two total electron spins $S = 0$ and $S = 1$ are a sum and a difference of the direct and exchange terms, respectively. We suggest that this is responsible for $\sigma_{|s_i-1/2|} > \sigma_{s_i+1/2}$ generally. As exchange contributions diminish with increasing energy (for a given ion) we see the trend toward $A = 0$ with increasing energy. In the case of $s_i = 1$ quasi-two-electron targets (He-like) the essential difference between the two cases of $S = 1/2$ and $S = 3/2$ is that in the latter the contribution to total ionization comes only from $s = 1$ target states (triplet-triplet), whereas for $S = 1/2$ both $s = 0$ (triplet-singlet) and $s = 1$ (triplet-triplet) states contribute. Hence again $\sigma_{|s_i-1/2|} > \sigma_{s_i+1/2}$ generally. With increasing energy the exchange (triplet-singlet) contribution goes to zero and the two direct contributions converge to each other leading to $A \approx 0$. Hence, both one- and two-electron targets display similar spin asymmetry behavior: being substantially positive at threshold and diminishing uniformly toward zero with increasing u.

The question of convergence with q of the spin asymmetries at a specified u is particularly interesting. Burgess *et al.* [7] used reduced coordinates to demonstrate q^4 scaling of the electron-impact excitation cross sections for H-like ions. This is equally applicable here as the total ionization cross section is obtained from summing the CCC-calculated cross sections for exciting the positive-energy states. Essentially, these arguments relate to the diminishing size of the ion and the associated increase in the excitation threshold with increasing q. While the rate of convergence for the scaled individual spin components cannot be determined, the reasoning of Burgess *et al.* [7] applies in the same way irrespective of the total electron spin, or energy of the excited state. So it is not too surprising that the ratio of the two components converges at each u faster with q than the scaled individual components. The generality of the scaling arguments is also why we believe the observed behavior is general in its nature and applicable to all targets for ionization or excitation. While other collision systems may show a different convergence rate in the spin asymmetries, it is clear that electron exchange remains a fundamental requirement in calculations in the energy region below ten times the ionization (or excitation) threshold, irrespective of how high q might be.

Lastly, we note that for the two very different collision systems, presented in Figs. 2 and 3, the (high-q) spin asymmetries are $A \approx 0.6$ at threshold and then diminish in a similar way for both, He-like and Li-like, systems. However, due to $s_i = 1$ and $s_i = 1/2$, Eq. (2) is different for the two cases. Hence r is different for the two systems over most of the energy range, and the similarity of A is a coincidence.

V. CONCLUSIONS

We compared measurements of total ionization cross sections for electron impact on He-like ions of charge $q \leqslant 6$ with corresponding calculations using the CCC theory, and found good agreement. With confidence in the accuracy of the CCC calculations we examined the cross-section scaling when considered as a function of u, the incident electron energy divided by the ionization threshold, and thereby confirmed the $q^4 \equiv E_I^2$ predictions of Burgess *et al.* [7]. Turning attention to the spin asymmetries we found them to have the same qualitative behavior with increasing energy for all of the targets considered, irrespective of q. Furthermore, at a given u, the spin asymmetries converged with increasing q to a constant even faster than the underlying scaled spin-dependent cross sections. Qualitative explanations for both observations were made with reference to the detailed analysis of Burgess *et al.* [7] and the spin coupling within the CCC theory. As these arguments are very general in nature, and apply to excitation just as much as to ionization processes, we expect them to hold very broadly. The most practical conclusion of the present study is that exchange contributions cannot be neglected at energies below ten times the threshold, even for very high q. Additionally, the fact that cross sections that vary by orders of magnitude with vastly different ionization thresholds have much the same spin asymmetries (when plotted against u) is another major conclusion of this work. It will be very interesting to study ionization (and excitation) spin asymmetries for a much broader range of collision systems and initial states.

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