Electron-impact single ionization of atomic ions in the Ga isonuclear sequence

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Theoretical distorted-wave calculations and experimental crossed-beam measurements are carried out for the electron-impact ionization of atomic ions in the Ga isonuclear sequence. For Ga^{+} and Ga^{2+} the indirect process of $3d$ subshell excitation followed by autoionization is calculated to make a significant contribution to the total ionization cross section near threshold. For Ga^{3+}, Ga^{4+}, and Ga^{5+} the total ionization cross section is calculated to be dominated by direct ionization of the $3d$ subshell. For Ga^{6+}, Ga^{7+}, Ga^{8+}, and Ga^{9+} the indirect process of $3p$ subshell excitation followed by autoionization is calculated to make a sizable contribution to the total ionization cross section. The distorted-wave predictions for the Ga ions are generally confirmed by the reasonable agreement found between theory and experiment along the isonuclear sequence.

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I. INTRODUCTION

An important process in the modeling of hot astrophysical and laboratory plasmas is the electron-impact ionization of atoms in various charge states. Gallium ($Z=31$) is the element with the widest range in the liquid state: it melts at 29.7 °C and it boils only at 2204 °C [1]. Therefore, it is not only an important metal in certain industrial plasmas but is also under consideration for forming a liquid curtain in front of the divertor plates in future fusion devices [2].

In this paper, we carry out both theoretical distorted-wave calculations and experimental crossed-beams measurements for the electron-impact ionization of the nine lowest-charge states of atoms in the Ga isonuclear sequence. Although Ga^{+} has been studied both theoretically [3] and experimentally [4,5], there exists no previous work on the electron-impact ionization of Ga^{2+} through Ga^{9+}. The Ga^{+} and Ga^{2+} ions are low-charge members of the Zn and Cu isoelectronic sequences. Large contributions from the indirect process of $3d$ subshell excitation followed by autoionization to the total ionization cross section have been found in detailed jjJ level resolved distorted-wave calculations for higher-charge Zn- and Cu-like atomic ions [6,7]. The ground configuration of the outer subshells for Ga^{3+} through Ga^{9+} is given by $3p^6 3d^w$ ($w=10$ to 4). We expect that a major contribution to the total ionization cross section for these ions should come from direct ionization of the $3d$ subshell. However, the relative contribution made by $3p$ subshell excitation followed by autoionization to the total ionization cross section should grow as the $3d$ subshell occupation number w decreases. Moores and Reed [8] give a comprehensive review of the strong $3p \rightarrow nl$ excitation-autoionization contributions to the total ionization cross section calculated and observed for atomic ions with configurations $3p^6 3d^w$ ($w=1$ to 3).

The remainder of this paper is organized in the following manner: in Sec. II, we review the distorted-wave theory applied to electron-impact ionization of atomic ions, in Sec. III we outline the experimental method, and in Sec. IV we compare experiment and theory for Ga^{+} through Ga^{9+}. We conclude with a brief summary.

II. DISTORTED-WAVE CALCULATIONS

The electron-impact single-ionization of an atomic ion can occur by two different nonresonant processes: direct ionization,

$$e^- + A^{q+} \rightarrow A^{(q+1)+} + e^- + e^-, \tag{1}$$

and excitation autoionization,

$$e^- + A^{q+} \rightarrow (A^{q+})^* + e^- \rightarrow A^{(q+1)+} + e^- + e^-, \tag{2}$$

where A is an arbitrary ion with charge q. For low-charge ions, the branching ratio for autoionization is approximately one, so that total ionization is simply the sum of the direct ionization and inner-shell excitation processes.

The direct ionization and the inner-shell excitation cross sections may be calculated in a configuration-average distorted-wave (CADW) approximation [9]. The threshold energies and the bound radial orbitals for the Ga configurations are calculated using the relativistically corrected Hartree-Fock atomic structure code of Cowan [10,11] where the mass-velocity and Darwin terms are included in the radial Schrödinger equations. The continuum radial orbitals are calculated as distorted-wave solutions of the relativistic radial Schrödinger equation using configuration-average Hartree and semiclassical exchange potentials [12]. The first-order scattering amplitude for either the ionization or excitation process is averaged over all states of an initial configuration and summed over all states of a final configuration. The excitation amplitude is evaluated using incident and scattered electrons calculated in a V^N potential, where N

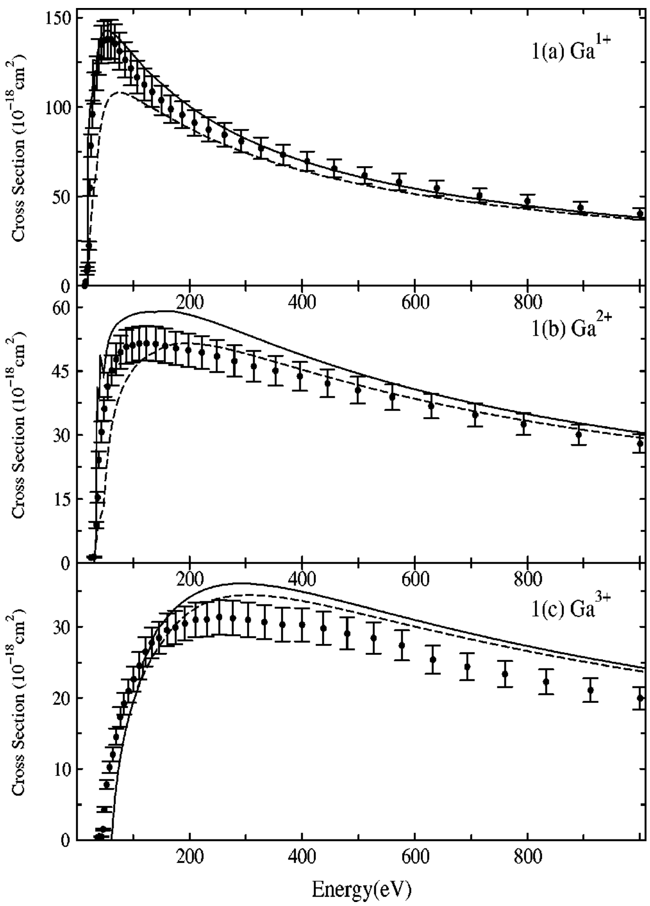

FIG. 1. The calculated total ionization cross sections (solid lines) are compared to the experimental cross sections (dots with error bars) for Ga^{+} (a), Ga^{2+} (b), and Ga^{3+} (c). The direct ionization cross section from the calculations is also shown (dashed line).

is the number of target electrons. The ionization amplitude is also evaluated using incident and scattered electrons calculated in a V^{N} potential, while the bound and ejected electrons are calculated in a V^{N-1} potential [13,14]. The configuration-average distorted-wave method has been successfully applied to study, in an efficient manner, the variations in the electron-impact ionization cross section along the Ti, Fe, Ni, Kr, Mo, and W isonuclear sequences [15–20].

III. EXPERIMENT TECHNIQUE

The measurements were performed at the Giessen electron-ion crossed-beams facility that has been described in detail earlier [22,23]. The Ga ions were produced by a 10-GHz electron cyclotron resonance ion source [24]. A newly developed high-temperature oven was used to evaporate Ga atoms into the plasma of the ion source. Employing this technique, the ion source could be held in operation for about 20 h before the oven had to be refilled and ion currents of several μA of $^{71}Ga^{+}$ up to $^{71}Ga^{9+}$ were extracted at 10 kV acceleration voltage. After magnetic analysis of the desired mass-to-charge ratio, the ion beam was collimated to typically 2×2 mm^2, which led to a reduction of the current by a factor of about 0.01. While passing the interaction region, the ion beam was crossed with an intense electron beam.

Two electron guns were employed for the present measurements. One electron gun [25] produces a ribbon-shaped electron beam with energies from 10 to 1000 eV and currents up to 450 mA. This gun was used for the lower-charged ions with charge states $q=1,\ldots,7$. For the higher charge states $q=8$ and 9 another electron gun [26] designed for energies from 50 to 6500 eV and currents up to 430 mA was employed.

After the interaction, the ionization products and the parent ion beam were separated by a second double-focusing 90° magnet. Whereas the incident ion beam was collected by a wide Faraday cup inside the magnet chamber, the product ions were detected by a channeltron-based single-particle detector [27].

Absolute cross sections were obtained by employing the dynamic crossed-beam technique [28], where the electron gun is moved up and down through the ion beam with simultaneous registration of the ionization signal and both actual beam currents. The total experimental uncertainties of the measured cross sections are typically 8% at the maximum resulting from the quadrature sum of the nonstatistical errors of about 7.8% (ion detection efficiency $\pm 3\%$, ion and electron currents $\pm 5\%$, ion and electron velocity $\pm 1\%$, and channel width $\pm 1\%$), and the statistical error at 95% confidence level. Typical measurement times for one data point were between 1 and 30 min.

IV. COMPARISON OF THEORY AND EXPERIMENT

The electron-impact single ionization cross sections for Ga^{+} through Ga^{9+} are shown in Figs. 1–3. The error bars show the total experimental uncertainty.

A. Influence of 3*d* subshell excitation on the total ionization of Ga^{+} and Ga^{2+}

The current calculations of the total ionization cross section for Ga^{+} are consistent with the results in Ref. [3], and since the calculations for the higher ionization stages of Ga use the same procedure, this gives us a valuable check on our calculations. The CADW total ionization cross section for Ga^{+} agrees remarkably well with the experiment in Fig. 1(a). For low-charge-state ions the CADW ionization cross section usually overestimates the total cross section by as much as a factor of 2 due to neglect of correlation between the outgoing scattered electron and the ejected electron [18]. The very good agreement seen in Fig. 1(a), where the peak of the cross section near energies of 50 eV is only 10% above the experimental data points, is somewhat fortuitous. The excitations from the $3d^{10}4s^2$ to the $3d^94s^24\ell$, $3d^94s^25\ell$, and $3d^94s^26\ell$ autoionizing configurations just above the ionization threshold enhance the total ionization cross section. The experimental ionization threshold for Ga^{+} is 20.5 eV [21] and all autoionizing states of the form $3d^94s^2n\ell$ above this threshold lead to single ionization. The main excitations are to $\ell=0-2$ and excitations to higher ℓ are small. The excitation cross sections at threshold decrease

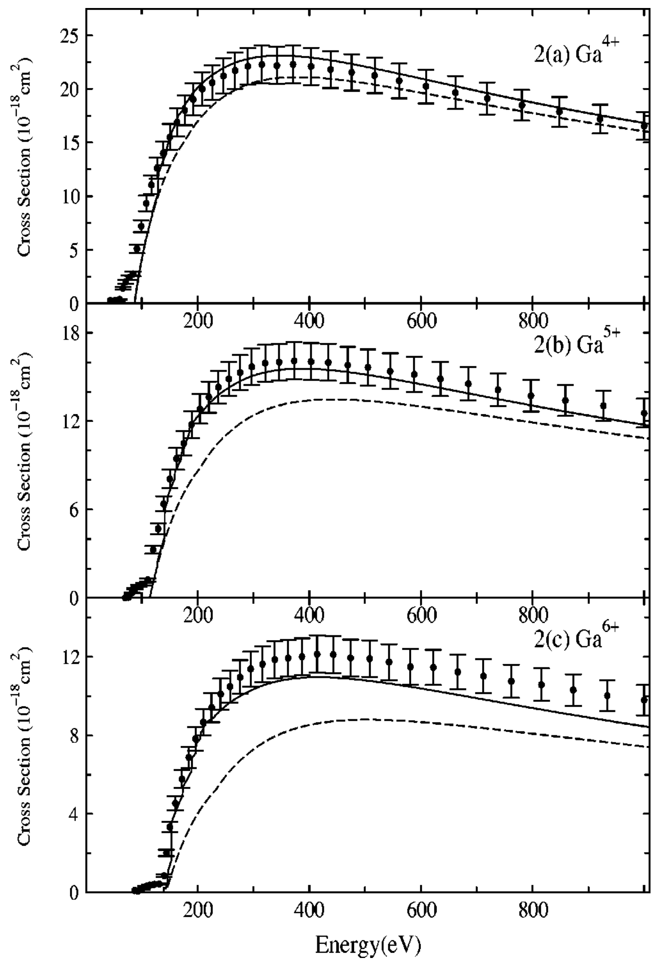

FIG. 2. The calculated total ionization cross sections (solid lines) are compared to the experimental cross sections (dots with error bars) for Ga^{4+} (a), Ga^{5+} (b), and Ga^{6+} (c). The direct ionization cross section from the calculations is also shown (dashed line).

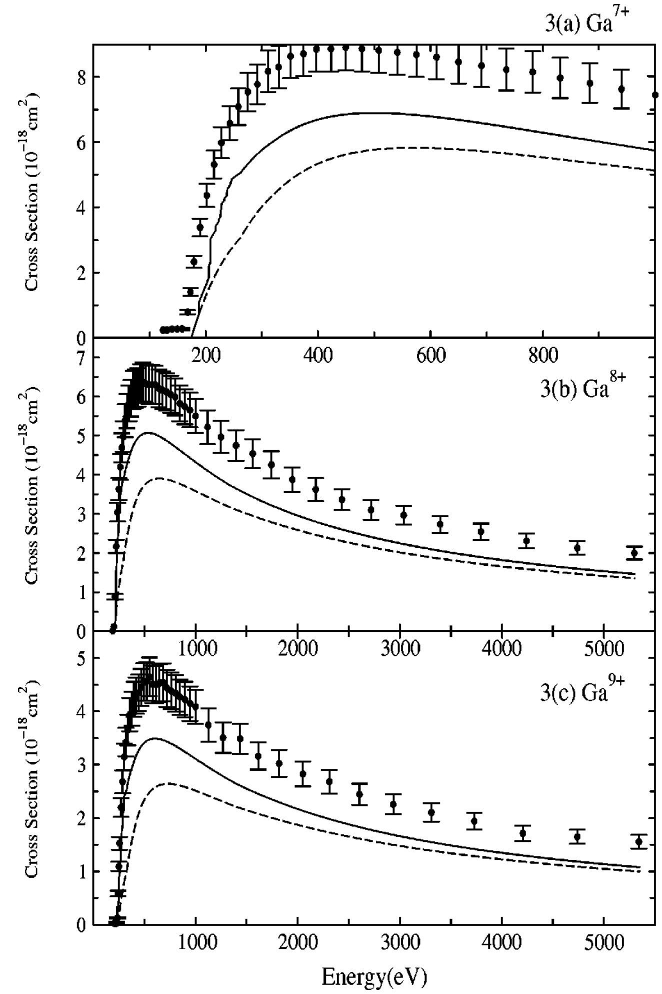

FIG. 3. The calculated total ionization cross sections (solid lines) are compared to the experimental cross sections (dots with error bars) for Ga^{7+} (a), Ga^{8+} (b), and Ga^{9+} (c). The direct ionization cross section from the calculations is also shown (dashed line).

rapidly with increasing n, so for most higher ionization stages $n=5$ was the highest level included in the excitation-autoionization calculation.

The calculations for Ga^{2+} were essentially the same as the calculation for Ga^{+}. The total cross section for Ga^{2+} is shown in Fig. 1(b). In Ga^{2+} the dominant autoionizing states were excitations from the $3d$ shell to the 4ℓ and 5ℓ shells. The total cross section at the peak of the cross section for Ga^{2+} is about 20% above the experimental data points, still surprisingly good agreement.

B. Dominance of $3d$ subshell direct ionization on the total ionization of Ga^{3+}, Ga^{4+}, and Ga^{5+}

The CADW total ionization cross sections are in very good agreement with the experimental cross sections for these ions. The distorted-wave approximation becomes better as the ion charge increases. The total cross sections are shown in Figs. 1(c) through 2(b).

For these ions, the direct ionization contribution to the total cross section is larger than the excitation-autoionization contribution. This is not to say that excitation-autoionization is negligible, but the number of electrons available for direct ionization out of the $3d$ subshell is larger than the number of electrons available for excitation out of the $3p$ subshell for these ions. The onset of excitation ionization occurs at energies well above the direct ionization threshold, so the direct ionization is the only process leading to ionization just above this threshold. As the ionization stage increases excitation autoionization contributions should become stronger and begin to occur at energies closer to the direct ionization threshold. This is confirmed when we compare Fig. 1(c) for Ga^{3+} to Fig. 2(b) for Ga^{5+}.

In contrast to Ga^{+} and Ga^{2+} where the excitation from the $3d$ shell dominates the excitation-autoionization contribution to the cross section, in Ga^{3+} through Ga^{5+}, the $3d$ excitations are below the first ionization threshold, so the dominant autoionizing excitations are from the $3p$ subshell to the 4ℓ and 5ℓ subshells. Excitations from the $3s$ subshell to the 4ℓ and 5ℓ subshells were examined, but these only made negligible contributions to the total cross section.

There are significant shifts in the experimental ionization thresholds to energies lower than the theoretical ionization thresholds for these ions. For example, the threshold for single ionization for Ga^{3+} in this experiment, around 45 eV, occurs at energies well below the theoretical threshold, 61.9 eV, for ionization out of the $3d^{10}$ configuration. The theoretical threshold is in reasonable agreement with the known threshold of 64.2 eV [21]. This indicates the existence of a metastable fraction of Ga^{3+} ions in the $3d^9 4s$ configuration. A CADW calculation of the direct ionization from this configuration shows a predicted threshold at 44.3 eV. Since the metastable fraction is unknown, Fig. 1(c) only shows the ionization from the ground state $3d^{10}$ of Ga^{3+}.

The total cross section at the peak of the cross section for these ions is about 20% above the experimental data points for Ga^{3+}, but the theoretical cross section is always within the experimental error for Ga^{4+} to Ga^{5+} except for the shifts near threshold due to the existence of metastable ions in the experiment.

C. Influence of 3*p* subshell excitation on the total ionization of Ga^{6+}, Ga^{7+}, Ga^{8+}, and Ga^{9+}

The excitation autoionization out of the $3p$ subshell in Ga^{6+}, Ga^{7+}, Ga^{8+}, and Ga^{9+} dominates the total ionization cross sections—see Figs. 2(c) through 3(c). In the calculation for Ga^{6+} shown in Fig. 2(c), excitations from the $3p$ shell to the $n\ell$ shell, $n=4-7$ and $\ell=0-5$, and also excitations from the $3s$ shell to the 4ℓ shell, $\ell=0-3$ were included. The highest-energy excitations included were around 200 eV. These excitation-autoionization contributions cause the steep rise in the cross section for Ga^{6+} just above threshold. The cumulative effect of the many weak excitations to the higher *n* that occur closer and closer in energy is to give an almost smooth continuumlike contribution to the total ionization cross section. Above 200 eV, the theoretical cross section is below the experimental data points by about 10%. The inclusion of higher *n* excitation autoionizing levels would increase the theoretical total ionization cross section and this may explain part of the shortfall at higher energies. The higher levels would have a larger probability of deexciting radiatively, however, violating the assumption that autoionization occurs from all excited states above the ionization threshold energy, so there are physical reasons as well as computational reasons to cut off the calculated contributions from the higher *n* levels. The ionization thresholds in the experiment are again shifted from the theoretical ground-state ionization thresholds because of a fraction of metastable ions in the experiment.

The agreement between the CADW total ionization cross sections and the experiment is relatively poor for Ga^{7+}, Ga^{8+}, and Ga^{9+}. The contribution to the total cross section from the excitation autoionizations from the $3p$ and $3s$ subshells is large in the theoretical calculations [see Figs. 3(a), 3(b), and 3(c) in the energy range from threshold to around 300 eV]. The highest final *n* levels considered for the excitation-autoionization calculations out of the $3p$ and $3s$ shells were $n=7$. The theoretical cross sections are 25–30 % below the experimental measurements. Since the distorted-wave approximation usually overestimates the cross sections and taking the branching ratio to ionization for the excited states to be unity gives the maximum enhancement to the cross section, this underestimate by the theory is surprising. The existence of an unknown metastable fraction might account for the higher experimental cross sections.

V. SUMMARY

The total ionization cross sections for Ga^{q+} ions where $q=1$ to 9 have been calculated in the configuration-average distorted-wave approximation and compared to experiment. The agreement is generally good, even surprisingly good in some cases, for $q=1$ to 6. The excitation-autoionization contribution to the total cross section for these ions varies predictably with the ionization stage—depending on the relative occupation number of the subshells from which direct and indirect processes can occur. The agreement for $q=7$, 8, and 9 total cross sections is not as good even though one might expect the distorted-wave approximation used to be more accurate as the ion charge increases. Part of the difficulties in comparing the experiment to theory is the unknown fraction of ions in metastable states and this may explain some of the discrepancies between theory and experiment.

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