Low-energy electron capture by N^{3+}, N^{4+}, and N^{5+} from hydrogen atoms using merged beams

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Measurements of absolute total cross sections for single-electron capture in collisions of N^{3+}, N^{4+}, and N^{5+} ions with ground-state atomic hydrogen are reported in the energy region 0.9–1400 eV/amu. Good agreement is obtained with available theoretical calculations and with previous experimental results at the higher energies. None of the systems studied shows a clear signature of the induced-dipole attractive force in enhancing the cross section.

INTRODUCTION

This paper presents the results of experiments in which absolute total cross sections for single-electron capture $\sigma_{q,q-1}$ are measured down to very low relative collision energies using a merged-beams technique. The process which has been studied is

$$N^{q+}+H(D)\rightarrow N^{(q-1)+}+H^{+}(D^{+}) , \quad (1)$$

where $q=3$, 4, and 5. The energy range of these experiments extends from 0.9 to 1400 eV/amu. This study is part of an ongoing research program initiated in our laboratory to study electron-capture collisions of multiply charged ions with neutral atoms from keV to near-thermal energies.

The considerable current interest in such processes arises from their practical application in controlled thermonuclear fusion studies, x-ray laser and radiation research, astrophysics, and multiply-charged-ion source development. These processes are also of fundamental interest and constitute a theoretical problem that may be solved by using restrictive approximation methods.

During the past decade, extensive theoretical and experimental work has been devoted to studies of electron-capture processes involving multiply charged ions. A detailed discussion on the theoretical and experimental progress in this field may be found in the recent review articles by Janev *et al.*[1] and Gilbody,[2] respectively. For partially stripped multicharged ion–hydrogen-atom systems, theoretical calculations of various degrees of sophistication have been carried out by a number of groups. For example, Gargaud and McCarroll[3] have performed quantal perturbed-stationary-state calculations of total and state-selective capture cross sections for collisions of N^{3+} and N^{5+} with atomic hydrogen. The energy region covered in these calculations extends from approximately 1 to 1000 eV/amu. Other low-energy ($E<1$ keV/amu) calculations for N^{q+}($q=3$, 4, and 5) +H systems are described in Refs. 4–11. Although these calculations are generally consistent with one another, discrepancies still exist among them, particularly at the lowest collision energies. One objective of the present program is to provide benchmark absolute measurements to which these calculations may be compared, so that a better understanding of electron-capture processes in multicharged ion–hydrogen-atom collisions can be achieved.

Measurements of total electron-capture cross sections for collisions of fully and partially stripped ions of C, N, O, F, and Ne with atomic hydrogen have been reported by Meyer *et al.*[12] in the energy range 0.18–8.5 keV/amu. Similar measurements of $\sigma_{q,q-1}$ have been performed for a variety of multicharged ions incident on atomic hydrogen by Phaneuf,[13] Crandall *et al.*,[14] Phaneuf *et al.*,[15–17] Panov *et al.*,[18] Bendahman *et al.*,[19] Dijkkamp *et al.*,[20] and Seim *et al.*[21] These experiments employed ion-beam–gas-target techniques to measure $\sigma_{q,q-1}$, but were unable to access the collision-energy region below 100 eV/amu due, among other things, to the difficulty of producing beams of projectile ions in high charge states at low collision velocities.

To address this low-energy region, an apparatus has been developed whereby total electron-capture cross sections can be determined at low energies using the merged-beams technique.[22] In this technique a fast (30–80 keV) multicharged ion beam is merged electrostatically with a 6–9-keV neutral beam traveling at nearly the same speed and the two beams are allowed to interact over an 80-cm path (see Fig. 1). Multicharged ion beams of necessary intensity are produced by the Oak Ridge National Laboratory Electron Cyclotron Resonance (ORNL-ECR) multicharged ion source.[23] The H^+ products of the collision are magnetically separated from the primary beams and are detected. Since the interaction energy is determined by the relative velocity of the beams, a wide range of interaction energies may be accessed under a variety of laboratory conditions using this technique. In the first cross-section measurements with this apparatus, absolute total electron-capture cross sections were determined for a relative energy ranging from 0.9 to 800 eV/amu for the $O^{5+}+H(D)$ systems.[22]

One noteworthy feature of the measurements performed for the $O^{5+}+H(D)$ systems is that at relative energies below 3 eV/amu, the capture cross section $\sigma_{54}(E)$ increases with decreasing energy. This increase in $\sigma_{54}(E)$ strongly resembles the predictions of the classical (Langevin) orbiting model according to which the cross section becomes inversely proportional to velocity at low energies.[24] It is believed[25] that the increase in $\sigma_{54}(E)$

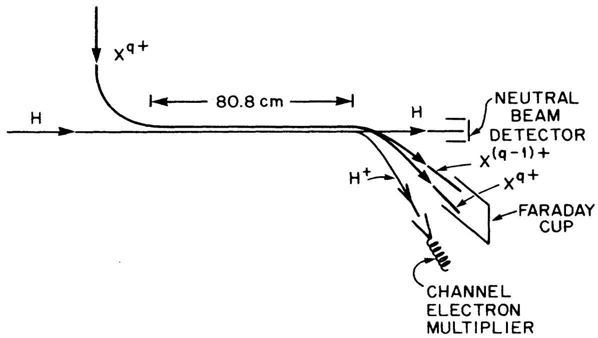

FIG. 1. Simplified schematic of the ion-neutral merged-beams apparatus.

may be caused by such "trajectory effects" which arise because of the ion-induced dipole attraction between the reactants at low energies. This attraction permits collisions to sample, at an increased relative velocity, internuclear separations that are smaller than the impact parameter. If there is a favorable potential-curve crossing at small internuclear separations, this effect may produce an enhancement of the capture cross section in some cases. Calculations for the $N^{3+}+H$ system by Watson and Christensen[6] and by Rittby *et al.*[26] have indicated that such effects enhance the capture cross section for collision energies less than 0.1 eV/amu. So, it is of interest to search for such trajectory effects at the lowest achievable collision energies.

In the following sections, we will briefly describe the experimental apparatus and method, and then present the results for each N^{q+} projectile separately.

EXPERIMENTAL METHOD

The apparatus used and the technique employed in the present experiments have been described in detail previously.[22] Only a brief description of the experimental method will be given here. A ground-state beam of H or D atoms is produced by passing a 6–9-keV beam of H^- or D^- ions through the optical cavity of a 1.06-μm Nd:YAG (YAG is yttrium aluminum garnet) laser where up to 600 W of continuous power circulates. The H^- (D^-) beam is produced by a commercial duoplasmatron ion source. Typically 0.5% of the negative ions in the beam are neutralized by photodetachment. The system is designed to produce a nearly parallel beam of H(D) atoms in the merge path with a diameter of 2–4 mm full width at half maximum (FWHM) and an intensity of 10–20 (particle) nA. The divergence of this beam is typically less than 0.2°. A 50–90-keV, 2–4-μA beam of N^{q+} ions produced by the ORNL-ECR multicharged ion source is merged electrostatically with this neutral beam. Typical FWHM of the multicharged ion beam along the merged path is 6–8 mm with a mean divergence of less than 0.5°. The 80.8-cm-long interaction path of the beams is strongly differentially pumped, and maintained at an average pressure of less than 2×10^{-10} Torr with beams present. In order to quantify the spatial overlap of the beams, two-dimensional intensity profiles of each beam are measured by beam scanners at four different positions along the merge path (see Fig. 2). One of the mechanical scanners used in the present experiments was modified to employ slits rather than knife edges. This reduced the susceptibility of the profile measurements to ion-beam intensity fluctuations and thereby improved the relative accuracy of the cross-section measurements. One of the products of the collision [Eq. (1)], i.e., H^+ (D^+), is magnetically separated from the primary beams and deflected and focused onto a channel electron multiplier where it is counted. At these low relative collision energies, production of H^+ (D^+) by electron capture from N^{q+} is known[1] to be dominant over ionization by several orders of magnitude. The $N^{(q-1)+}$ product of the reaction, then, need not be measured separately and is simply collected together with the primary N^{q+} in a large Faraday cup. As an example of typical operating conditions, 4 μA of N^{3+} merged with 10 (particle) nA of D at an $E_{\rm rel}$ of 5 eV/amu produced 76 Hz of beam-beam signal and 10 kHz of background due to the stripping of H atoms on residual gas. A fast two-beam chopping scheme is used to separate this signal from the background. Since the signal event rate produced by the beam-beam interaction is proportional to the relative velocity of the beams (assuming an energy-independent cross section), the signal-to-background ratio determined the lowest energies at

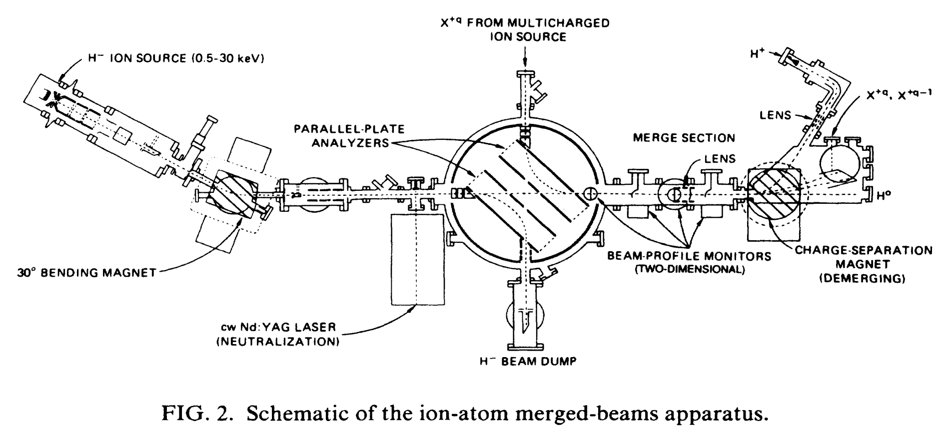

FIG. 2. Schematic of the ion-atom merged-beams apparatus.

which measurements could be made. If the cross section, in fact, also decreases with decreasing energy, measurements become even more difficult at lower energies. The lower-energy limit imposed by the apparatus itself is approximately 0.2 eV/amu, and is a consequence of the finite angular divergence of the interacting beams.

The ECR source used in the present experiments is expected to produce reactant N^{3+} and N^{5+} ions not only in the ground state but also in metastable states. For Be-like N^{3+} ions, the metastable $(2s2p)^3P$ states lie very near in energy to the $(2s^2)^1S$ ground state and are statistically favored by a 9-to-1 ratio. In measurements of electron-impact ionization of Be-like C^{2+} ions produced in a discharge-type source, this metastable fraction was determined to be 90%.[27] A comparably high metastable fraction is expected for beams of N^{3+} beams produced by the ECR ion source.

The He-like N^{5+} $(1s2s\,^3S)$ metastable fraction produced by the ECR source has been determined in an independent experiment[28] to be $(6.2\pm0.6)\%$, and was found to be independent of the source operating conditions. Li-like ions (e.g., N^{4+}) are known to have no long-lived metastable states.[29] The effect of metastable reactant ions on the reaction cross sections will be discussed later. The estimated systematic uncertainties in measured cross-section values are the same as those reported previously.[22]

RESULTS AND DISCUSSION

The data for N^{3+}, N^{4+}, and N^{5+},$+$H(D) systems are listed in Table I. Along with each measurement, the random uncertainty estimated at two standard deviations (90% confidence level) on counting statistics and the total estimated absolute uncertainty are also listed. The total absolute uncertainty is also consistent with a 90% confidence level and is determined by summing the statistical uncertainty and estimated systematic uncertainties in quadrature. The uncertainty in the energy is consistent with a 90% confidence level (see Ref. 22).

N^{3+} + H(D) collisions

Absolute total cross sections for single-electron capture, σ_{32} for collisions of N^{3+} with H(D) are shown in Fig. 3 as a function of relative collision energy expressed in eV/amu. Also shown in the figure are the various calculations and measurements of σ_{32} for this system that are available in the literature in the energy range of 0.1–10 000 eV/amu. As may be seen from the figure, the present highest-energy measurements agree well with the earlier measurements of Seim *et al.*[21] and Crandall *et al.*[14] Furthermore, the agreement between the present results and the theoretical calculations of Gargaud and McCarroll,[3] Bienstock *et al.*,[5] Watson and Christensen,[6] and McCarroll and Valiron[7] is seen to be excellent. More recent calculations by Gargaud,[30] which include electron translation factors, agree with earlier calculations[3] and extend to higher energies (1200 eV/amu). These calculations indicate that electron capture by ground-state N^{3+} occurs primarily into the $(1s^22s^23s)$ state of N^{2+}, i.e.,

$$N^{3+}(1s^22s^2)^1S + H(1s) \rightarrow N^{2+}(1s^22s^23s)^2S + H^+ \; .$$

Contributions from capture to the $(1s^22s^23p)^2P$ (Ref. 30), $(1s^22s2p^2)^2D$, and the $(1s^22s2p^2)^2S$ states of N^{2+} were found to be small (i.e., $<4\%$) in these calculations.[5] State-selective measurements for this system by Wilkie *et al.*[31] based on translational energy spectroscopy confirm the dominance of capture into the 3*s* state in 200–1000-eV/amu energy range, but also indicate increasing population of 3*p* and a number of other minor channels at the highest energies investigated. The metastable N^{3+} fraction is believed[31] to have been small in those measurements.

It was noted earlier that most ($\sim 90\%$) of the N^{3+} reactant ions produced by the ECR source used for the present measurements are expected to be in metastable states of the $1s^22s2p$ configuration. The favorable comparison of the present experimental results with the ground-state N^{3+}+H calculations[3,5–7] suggests that electron capture by metastable N^{3+} may have the same total cross section as that by ground-state N^{3+}.

N^{4+} + H(D) collisions

Measurements of σ_{43} for N^{4+}+H(D) collisions are shown in Fig. 4 along with the experimental measurements of Crandall *et al.*[14] and Seim *et al.*[21] and the theoretical calculations of Feickert *et al.*[8] It is evident from the figure that the agreement between the present results and those of Crandall *et al.*, Seim *et al.*, and Feickert *et al.* is excellent in the energy region in which they overlap.

Electron capture in N^{4+}+H(D) collisions is predicted to occur predominantly into the $n=3$ state of the N^{3+} ion. There are three possible competing channels, i.e., 3*s*, 3*p*, and 3*d*. these channels have potential-energy curve crossings with the incident channel at approximately 6, 7, and 8 a.u., respectively.[8] The calculations of Feickert *et al.* show that the 3*d* final state provides a dominant contribution to σ_{43} for collision energies less than a few tenths of an eV/amu. At the higher energies of the present investigation, contributions from the 3*s* and 3*p* crossings are expected to dominate.

We are aware of no other theoretical calculations for this system in the energy region covered by the present data. Since the Li-like N^{4+} ion beam does not have any metastable components, the measurements presented here correspond to ground-state reactants and are believed to have sufficient accuracy to provide a critical test of any future theoretical calculations.

N^{5+} + H(D) collisions

Measurements of σ_{54} for collisions of N^{5+} with H and D are shown in Fig. 5. Again, the error bars in Fig. 5 represent random uncertainties and the reproducibility of the data at the 90% confidence level. There is considerably more scatter in these data than was observed for the N^{3+} and N^{4+} measurements. It is believed that the origin of this scatter lies primarily in the observed short-term instability of the N^{5+} beam produced by the ion source. This instability gives rise to an additional uncertainty in the accurate determination of the overlap of the two beams and hence the reproducibility of σ_{54}, preventing measurements from being extended to lower energies. Unlike that observed in the N^{3+} and N^{4+} systems, the capture cross section (and hence observed signal rate) is relatively large at the lower energies and would have permitted measurements to be performed below 1 eV/amu.

Figure 5 also shows the various theoretical and other experimental work that has been performed for this system. A comparison of the present work with that of Dijkkamp *et al.*[20] reveals excellent agreement in the energy region in which the measurements overlap.

TABLE I. Experimental electron-capture cross sections for $N^{q+}+H(D)\rightarrow N^{(q-1)+}+H^{+}(D^{+})$, $q=3,4,5$.

Ion	E_{rel} (eV/amu)	ΔE_{rel} (eV/amu)	$\sigma_{q,q-1}$ (10^{-16} cm^2)	Relative uncertainty (10^{-16} cm^2)	Total absolute uncertainty (10^{-16} cm^2)
N^{3+}	0.9	0.3	23.4	5.8	6.5
	1.2	0.3	22.8	5.4	6.2
	1.7	0.3	14.7	4.3	4.7
	2.4	0.3	22.6	5.0	5.8
	3.7	0.4	25.7	4.9	5.9
	6.7	0.4	34.9	7.4	8.7
	12.8	0.5	37.0	5.1	6.5
	14.7	0.5	36.6	4.9	6.3
	19.4	0.6	33.3	2.8	4.6
	51.1	0.9	31.5	5.1	6.2
	97.5	1.2	27.8	2.7	4.1
	164	1.5	30.0	3.0	4.2
	265	2.0	23.6	2.9	3.7
	505	3.1	22.7	2.7	3.5
	902	4.3	19.6	1.8	2.7
	1381	6	19.0	1.1	2.2
N^{4+}	1.2	0.3	9.7	5.4	5.5
	1.8	0.3	13.5	4.8	5.1
	2.5	0.3	11.2	3.5	3.8
	4.1	0.4	20.2	3.3	4.2
	6.2	0.4	19.5	5.6	6.1
	9.1	0.5	22.6	3.6	4.6
	11.5	0.5	22.8	4.1	4.8
	19.4	0.6	28.4	3.5	4.7
	43.0	0.8	31.3	2.6	4.3
	79.8	1.1	37.2	2.3	4.7
	182	1.8	31.6	3.2	4.5
	516	3.0	32.3	1.8	3.7
	672	3.5	30.8	1.5	3.4
	982	4.4	33.8	0.8	3.5
N^{5+}	1.5	0.3	73	19	21
	2.9	0.4	68	11	14
	4.1	0.4	81	10	15
	7.5	0.5	59.4	5.2	9.3
	9.0	0.5	80	19	21
	10.8	0.5	65	17	19
	14.0	0.6	71	13	15
	21.3	0.7	72	17	18
	23.3	0.7	58	11	13
	30.5	0.8	55	12	13
	41.5	0.9	63.3	6.5	9.5
	54.4	0.9	53.8	5.4	8.0
	74.7	1.2	38.7	5.0	6.6
	76.6	1.2	45.0	5.0	6.7
	125	1.4	39.0	7.2	8.2
	152	1.6	36.4	2.6	4.5
	215	1.8	38.9	5.0	6.3
	313	2.5	23.8	2.4	3.4
	570	3.5	23.6	2.4	3.4

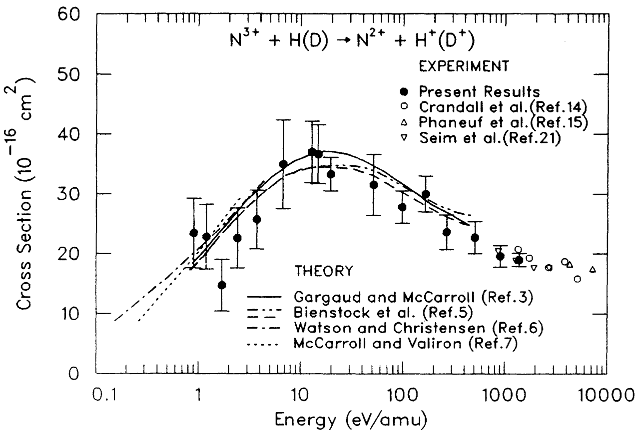

FIG. 3. Comparison of merged-beams data for electron capture in N^{3+}+H(D) collisions with previous measurements and theory. The present data are presented with error bars denoting relative experimental uncertainties, as listed in Table I. The dashed curve of Bienstock *et al.* denotes their calculation for capture into the $(2s^23s)$ state of N^{2+}, while the chain curve of Bienstock *et al.* includes capture into the $(2s^23s)$ and $(2s2p^2)^2S$, ^2D states. Note the calculations are performed for ground-state $N^{3+}(1s^22s^2)$, while the N^{3+} beam used in the present measurements is known to contain a significant fraction of metastable $(1s^22s2p)$. See text for discussion.

Dijkkamp *et al.*, Crandall *et al.*, and Seim *et al.*[21] measured σ_{54} by detecting product N^{4+} ions, in contrast to the present experiment which detects the product $H^+(D^+)$ ions. A factor to be considered in evaluating measurements of σ_{54} is the known presence of 6% metastable N^{5+} in the ion beam used in the present experiments. Single-electron capture by metastable N^{5+} ions will produce autoionizing states of N^{4+},[32] which Auger decay before being analyzed and detected. Thus, σ_{54} determined by the detection of N^{4+} products will be smaller than that determined by the detection of (H^+) products by an amount equal to the metastable fraction

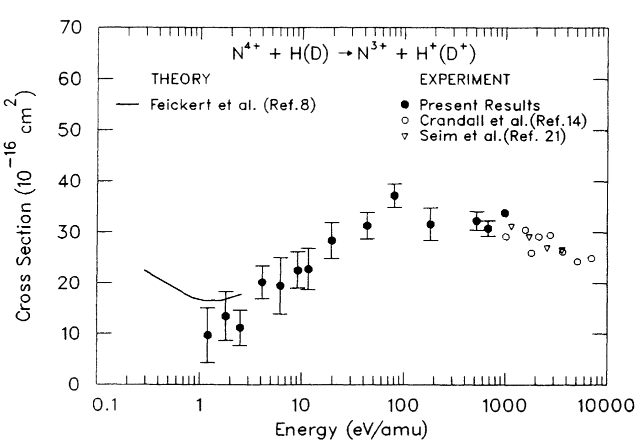

FIG. 4. Comparison of merged-beams data for electron capture in N^{4+}+H(D) collisions with previous measurements and theory.

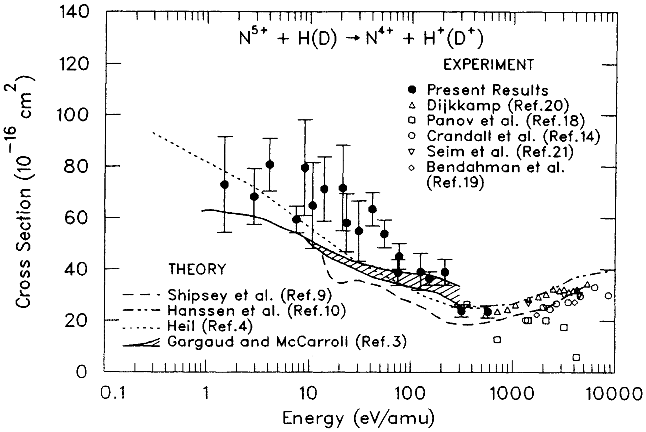

FIG. 5. Comparison of merged-beams data for electron capture in N^{5+}+H(D) collisions with previous measurements and theory. The calculations of Gargaud and McCarroll are denoted by upper and lower bounds which correspond to the origin of coordinates being placed on the N and H nucleus, respectively.

present in the reactant beam times the ratio of the capture cross sections for the metastable and ground states of N^{5+}. Since the relative uncertainties of the individual experiments are greater than the metastable fraction (6%), no definitive conclusion could be reached concerning the relative magnitude of the electron-capture cross section for N^{5+} metastable projectiles.

Theoretical calculations of Gargaud and McCarroll[3] and Shipsey *et al.*[9] indicate that for this system, electron capture occurs primarily into the $(1s^24s)^2S$ state of N^{4+}, i.e.,

$$N^{5+}(1s^2)+H \rightarrow N^{4+}(1s^24s)+H^+ .$$

Capture into the 3*s*, 3*p*, and 3*d* states of N^{4+} is estimated to contribute negligibly[3] (i.e., $<3\%$) to σ_{54} at energies less than 1 keV/amu. More recent calculations by Gargaud,[30] which include electron-translation factors, follow closely the calculations of Heil.[4] The measurements reproduce the general trend of σ_{54} predicted by theory, which indicates a rising cross section as the collision energy decreases. Unfortunately, measurements could not be extended to lower energies where trajectory effects become important and can lead to sharp increases in the cross section.[25]

SUMMARY

Absolute total cross sections for single-electron capture involving collisions of N^{3+}, N^{4+}, and N^{5+} ions with atomic H or D have been measured in the energy range 0.9–1400 eV/amu. Agreement between the present results and available theoretical calculations and other experimental results is good. In view of the good agreement between the ground-state calculations for the N^{3+}+H systems and the present measurements in which the N^{3+} metastable component is large, it is suggested that total electron capture by metastable N^{3+} may have the same cross section as the ground-state N^{3+}.

One objective of the present study was to elucidate the possible role of "trajectory effects" in N^{q+} ion-hydrogen atom collisions at low energies. None of the $N^{q+}+H(D)$ systems studied shows a clear signature of the induced-dipole attractive force in enhancing the cross section in the range of energies studied. For higher ion charge states, such trajectory effects may occur in the energy range accessible to our present merged-beams apparatus. The search for such effects will be a subject of future experiments.

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