Reevaluation of electron-capture cross sections in C^{4+} + H collisions

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We calculated the total electron-capture cross section for collisions of C^{4+} ions with atomic hydrogen in the energy range of 10–2000 eV/nucleon using the close-coupling two-center atomic orbital expansion method. Various basis sets have been used to check the convergence of the calculation and the results are shown to be in general agreement with existing experimental data. However, we found no evidence of a dip in the total electron-capture cross sections near 500 eV/nucleon as reported by Bliek *et al.* [Phys. Rev. A **56**, 526 (1997)]. [S1050-2947(98)07507-6]

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I. INTRODUCTION

Electron-capture cross sections for collisions between C^{4+} ions with atomic hydrogen have been measured in many experiments since the earlier 1980s. Using ions from laser-produced plasma, Phaneuf *et al.* [1] obtained total electron-capture cross sections down to about 15 eV/nucleon. Using state-selective photon emission spectroscopy, electron-capture cross sections to $3s$, $3p$, and $3d$ states have been measured by Dijkkamp *et al.* [2] down to 1000 eV/nucleon and by Hoekstra *et al.* [3] down to about 50 eV/nucleon. These early experiments often have large error bars. To amend this situation, recently Bliek *et al.* [4] used the state-of-the-art merged-beam ion-collision facility at Oak Ridge National Laboratory to determine accurate electron-capture cross sections in the 6–1000 eV/nucleon region. Their measured total electron-capture cross sections above 200 eV/nucleon disagree noticeably with the earlier experimental data. Furthermore, their data showed a distinct dip in the energy region near about 500 eV/nucleon.

On the theoretical side, this collision system has been studied in a number of papers since the 1980s. Calculations using molecular orbitals (MO) as basis functions often were performed with different numbers of molecular functions, and with or without electron translation factors [5–10]. The earlier calculations with a small number of basis functions are less reliable but recent calculations with larger basis functions are expected to be adequate. An alternative approach is to perform semiclassical calculations using atomic orbitals on the two collision centers as basis functions. This has been used by Fritsch and Lin [12], where cross sections for total capture and for capture to the dominant $n=3$ subshells were calculated for energies down to 100 eV/nucleon. Comparing the calculations with existing experiments before 1994, one can say that the theoretical results of Fritsch and Lin (to be called FL herafter) are in general agreement with experiments [2,3] above 200 eV/nucleon and the calculations of Saha [10] and of Gargaud *et al.* [8] are in general agreement with the experiment of Phaneuf *et al.* [1] and of Hoekstra *et al.* [3] in the lower-energy region. However, all of these experiments have relatively large error bars and it is not possible to discriminate the various theories that do show non-negligible differences.

The new experimental data of Bliek *et al.* gave smaller error bars. Comparing with the earlier data, the major discrepancy is in the higher-energy region above 200 eV/nucleon. In particular, the total cross section of Bliek *et al.* shows a distinct dip near 500 eV/nucleon. The existence of such a dip is quite surprising. While partial cross sections to individual subshells are known to change rapidly with collision energies, the total cross section for charge transfer in general is known to vary smoothly with collision energies.

The discrepancy between the results of Bliek *et al.* with the more sophisticated close-coupling calculations in the higher energy region is also quite surprising. It is our experience that these close-coupling calculations should be capable of producing reliable cross sections for the dominant channels. The discrepancy, which is about at the 50% level, is not expected for the total electron-capture cross section. Thus we decided to undertake a more careful reexamination of the present collision system, to check whether existing calculations were at fault. Our results are in good agreement with the earlier close-coupling calculations of FL above 200 eV/nucleon. We have also extended the calculations to the low-energy region down to 10 eV/nucleon. In the low-energy region below 80 eV/nucleon simple estimate indicates that accounting for the curved trajectories can affect the calculated total cross section. When the trajectory effect is considered we are able to obtain total electron-capture cross sections in good agreement with the data of Bliek *et al.* below about 100 eV/nucleon. Our low-energy results are also in reasonably good agreement with the MO calculations of Gargaud *et al.* [8] and of Saha [10]. However, as we will show later, the agreement among the various experiments and theories are only qualitative. In the low-energy region, precise experimental data are still needed in order to unravel the limitation of the various theoretical models. We also check the partial electron-capture cross sections with previous measurements and the agreement is also quite good over the whole energy range except that the experimental error bars are quite large.

The theoretical method used in this calculation is essentially the same as the one used in Fritsch and Lin in 1984 although the calculation was done using a different computer code that uses different primitive basis functions. In Sec. II we describe the model potential and the basis functions used

TABLE I. Comparison of cross sections (in 10^{-15} cm^2) for electron tranfer into C$^{3+}(n)$ subshells (σ_n) and into all states (σ_{tot}) in C^{4+}+H collisions obtained by Fritsch and Lin [12] and from the present calculation. $a[b]$ stands for $a \times 10^b$.

	Fritsch and Lin					Present work				
E (keV/nucleon)	σ_{3s}	σ_{3p}	σ_{3d}	$\sigma_{n=4}$	σ_{tot}	σ_{3s}	σ_{3p}	σ_{3d}	$\sigma_{n=4}$	σ_{tot}
2.0	1.34	0.94	0.84	0.24	3.41	1.31	1.01	0.85	0.23	3.43
1.0	1.17	1.44	1.20	0.19	3.69	1.16	1.42	0.88	0.20	3.67
0.5	0.79	2.00	0.79	0.16	3.77	0.79	2.12	0.85	0.15	3.91
0.2	0.23	2.72	0.29	0.09	3.33	0.40	3.17	0.37	0.08	4.00
0.1	0.02	2.19	0.09	0.04	2.35	0.14	2.94	0.20	0.06	3.34

in the calculation. In order to extend the calculation to the lower-energy region, we describe how we account for the curved trajectory in a heuristic way. The results are shown and analyzed in Sec. III. A short summay is given in Sec. IV.

II. THEORETICAL METHODS

We used the semiclassical approximation where the heavy particle is moving along a straight-line trajectory for each impact parameter b. The time-dependent electronic wave function is expanded in terms of traveling atomic orbitals (AO) on the two collision centers. Such a two-center AO expansion approach has been used extensively in the past two decades for many collision systems. It has been shown that the cross sections for the major channels, which are dominated by collisions at large impact parameters, in general can be obtained accurately using the two-center AO expansion method [13]. For the present system, we used the same model potential as in FL but the basis functions used in the present work are generated from the even-tempered orbitals [14]. In either approach, the bound excited states of the C^{3+} ions and of the H atom are accurately represented. For collision energy in the 10–2000 eV/nucleon region, electron capture to the $n=3$ states is the dominant process, with a very minor contribution from capture to the $n=4$ states. These results are already clear from the earlier work of FL.

In this study we employed several sets of basis functions, but only those from two sets will be shown. In the first basis set, we include the $n=3$ and $n=4$ atomic orbitals of the C^{3+} ion and the $n=1$ and $n=2$ atomic orbitals of the H atom. This minimum basis set is expected to be adequate since electron capture occurs mostly in the large impact parameter region (between 4 and 8 a.u.) where expansion of wave functions in terms of atomic orbitals should be adequate. However, in view of the discrepancy between FL and the new experimental data, we decided to perform a number of calculations with a larger basis set. In addition to the orbitals included in the first set, we also include some pseudostates on the C^{4+} center. These pseudostates allow for a better representation and flexibility of the wave function at small internuclear separation. We include $\ell=0$, 1, 2, and 3 states on the C^{4+} center and for each ℓ there are a few pseudostates in addition to the bound states. All together, in the second set there are 61 states on the C^{4+} center, together with the four states on the H center. As we will show in the next section, the resulting electron-capture cross sections from the two calculations differ by less than a few percent.

The difference in the probabilities occurs only in the small impact parameter region, which contributes little to the total cross section. We have also performed calculations using different pseudostates generated from different primitive basis functions but obtained essentially the same electron-capture cross sections. Thus we believe that the results reported below are converged to a few percent.

The standard two-center atomic orbital close-coupling calculations are performed in general in the near-velocity matching region. However, for the collision between multiply charged ions and hydrogen atom, electron capture occurs at large internuclear separation. Thus we expect that the atomic orbital expansion method to describe the time-dependent wave function reasonably well even at lower energies. However, at low collision energies the straight-line trajectory approximation is not valid. Instead, a curved trajectory describing the relative motion between the two heavy particles is needed. This trajectory is to be computed from some averaged internuclear potential. Within the semiclassical approach, this averaged potential is not precisely defined. Thus we account for the trajectory effect in the following heuristic manner. Since the incoming path is between a neutral atom and an ion, the trajectory in the outer part is essentially a straight line. On the outgoing path after charge exchange, the two heavy particles have charges +3 and +1 for C^{3+} and H$^+$, respectively. Thus the trajectory is mostly Coulombic. To account for the trajectory effect, we make the following ansatz: Calculate the distance of closest approach r_c assuming that the internuclear potential is due to the Coulomb force between C^{3+} and H$^+$ for each impact parameter b. Since the incoming path is a straight line, the distance of closest approach is approximated by $b+(r_c-b)/2$. We then interpret the probability calculated using the straight-line trajectory for impact parameter b to be the same as the probability for a curved trajectory, which has the distance of closest approach given by $b+(r_c-b)/2$. Clearly for the repulsive Coulomb interaction this has the effect of reducing the total cross section. This simple model of accounting for the trajectory effect is definitely not a rigorous treatment but it allows us to make an estimate of the trajectory effect on the total cross section for collisions at lower energies without the full quantum treatment of the motion of the heavy particles, which has other limitations of its own. A similar method has been used previously to calculate the neutralization cross sections between positive and negative ions at low energies and the electron-impact detachment of negative ions

TABLE II. Cross sections (in 10^{-15} cm^2) for electron tranfer into C$^{3+}(n)$ subshells (σ_n) and into all states (σ_{tot}) in C^{4+}+H collisions at energies below 0.08 keV/nucleon from the present calculations using straightline vs curved trajectories. $a[b]$ stands for $a \times 10^b$.

E (keV/nucleon)	Straight line					Curved trajectory				
	σ_{3s}	σ_{3p}	σ_{3d}	$\sigma_{n=4}$	σ_{tot}	σ_{3s}	σ_{3p}	σ_{3d}	$\sigma_{n=4}$	σ_{tot}
0.08	0.10	2.60	0.23	0.11	3.04	9.13[−2]	2.40	0.20	8.22[−2]	2.77
0.06	0.03	2.12	0.27	0.12	2.54	2.83[−2]	1.91	0.23	8.37[−2]	2.28
0.04	0.02	1.73	0.34	0.15	2.24	1.88[−2]	1.47	0.28	5.51[−2]	1.82
0.02	3.81[−3]	0.87	0.52	0.13	1.56	6.25[−4]	0.53	0.39	2.53[−3]	0.92
0.01	1.43[−3]	0.35	0.61	0.13	1.10	9.24[−7]	0.06	0.25	0.00	0.31

[15]. The results obtained using such an approach are in good agreement with experiments.

III. RESULTS AND DISCUSSION

We have calculated electron-capture cross sections using the two basis sets described in the preceding section. In the low-energy region electron-capture probabilities oscillate rapidly with impact parameters. We checked to make sure that calculations were performed with sufficient dense meshes of impact parameters in obtaining the total cross section.

In Table I we present the calculated total electron-capture cross sections and partial cross sections to individual $3s$, $3p$, $3d$, and $n=4$ states. These results were obtained with the first basis set. At energies above 0.1 keV/nucleon, they are compared to the earlier results of FL, which used a larger number of basis functions. At the higher energies the present calculations essentially reproduce the results from FL to better than a few percent. At the two lower-energy points, 0.1 and 0.2 keV/nucleon, the two calculations have large differences. The discrepancy is probably because of the insufficient number of impact parameters used in FL, where the meshes were dense enough for the higher energies but not enough for the lower energies.

At energies below 0.1 keV/nucleon, we present the cross sections calculated using the straightline trajectories and those calculated using the curved trajectories following the model discussed in the preceding section. The results are shown in Table II. The trajectory effect was found to be negligible above 0.1 keV/nucleon, but very significant at the low-energy end.

In Fig. 1(a) we compare the total electron-capture cross sections in the 10–1000 eV/nucleon region obtained from different theoretical calculations with the experimental data of Bliek et al. [4]. Besides the present calculations, we show the recent MO calculation of Gargaud et al. [8], where the dominant seven molecular states were used and the motion of the heavy particles is treated quantum mechanically such that the curved trajectory effect is implicitly included. We also show the other MO calculation done by Saha [10], where essentially identical MO's were used but the trajectory effect was not accounted for. Clearly none of the theoretical results shows any dip in the cross section near 500 eV/nucleon in contrast with the experiment. However, the two MO calculations do not agree at the higher energies above 100 eV/nucleon. This may be a reflection of the different

electron translational factors used in the two MO calculations. The present AO calculation agrees well with the MO result of Gargaud et al. [8] at the higher energies. In Fig. 1(a) the present AO results below 80 eV/nucleon shown are those calculated including corrections due to the curved trajectories. They are in good agreement with the data of Bliek et al. However, from Table II we note that the present results using straight-line trajectories are much closer to the other two MO results. In other words, the present AO calculations using straight-line trajectories are very close to the MO calcula-
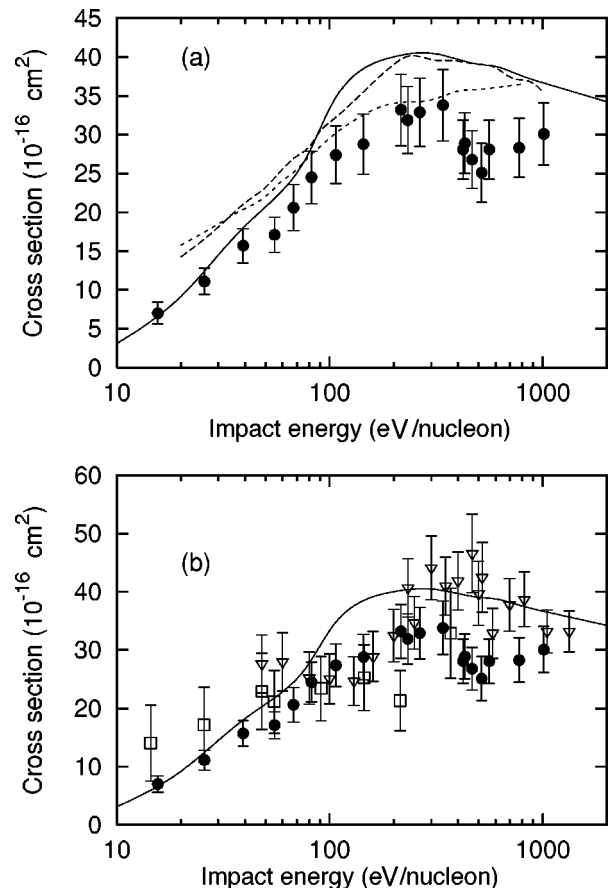

FIG. 1. (a) Comparison of total electron-capture cross sections for C^{4+} on H collisions. Solid line, present calculation; dashed lines, Gargaud et al. [8]; dotted lines, Saha [10]. The recent experimental results from Bliek et al. are also shown for comparison. (b) Comparison of the present theoretical result with the data of Hoekstra et al. [3] (triangles), of Phaneuf et al. [1] (open squares), and the recent data of Bliek et al. [4] (solid circles).
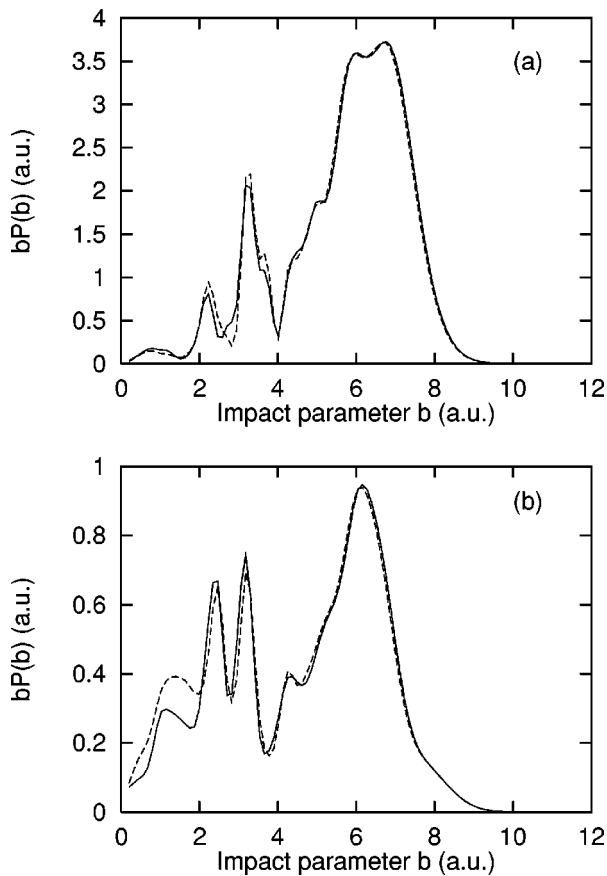
FIG. 2. Impact-parameter dependence of $bP(b)$ for electron capture to (a) $3p$ at impact energy of 0.5 keV/nucleon and (b) $3d$ at 0.3 keV/nucleon calculated with two different basis sets as explained in the text. The solid line is from the large basis set calculation and the dashed one, from the small basis set calculation.
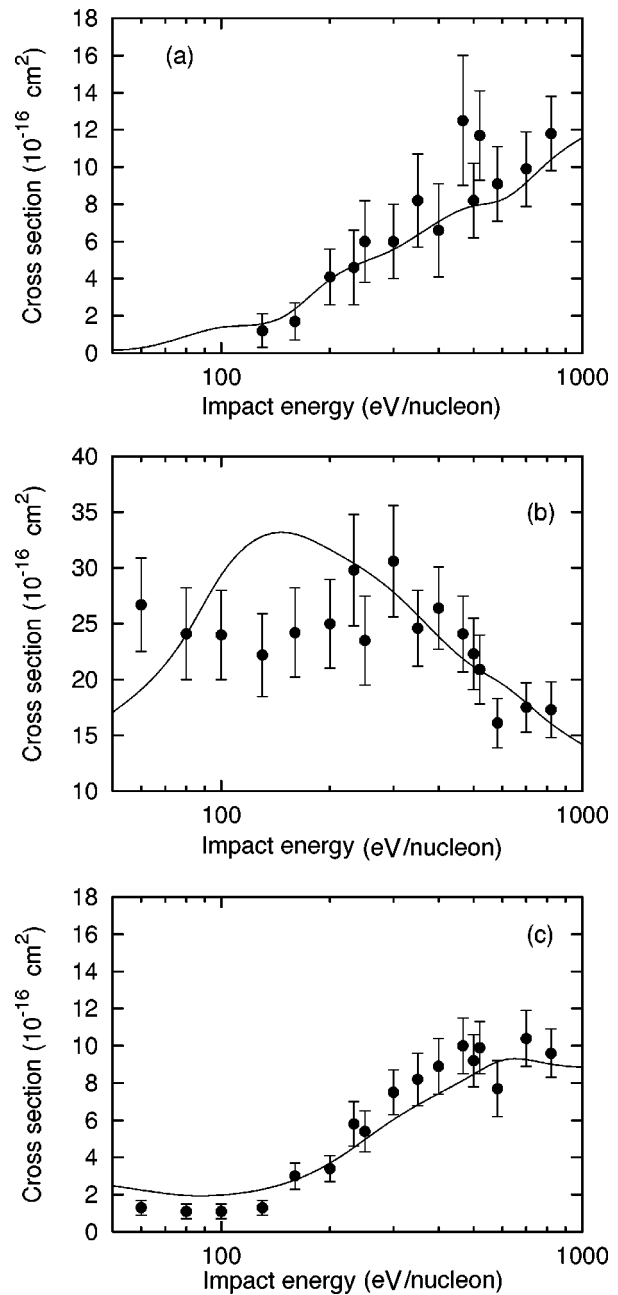

FIG. 3. Comparison of calculated and experimental partial electron-capture cross section to (a) $3s$, (b) $3p$, and (c) $3d$ states of C^{3+} ions. The experimental data are from Hoekstra et al. [3] .

tions of Gargaud et al. [8] over the whole energy range shown. If the calculations of Gargaud et al. [8] eventually turn out the correct answer, then they would point out the deficiency of the ad hoc procedure adopted in the present method for accounting for the trajectory effect. In that case, it would also mean that the measured total electron-capture cross sections by Bliek et al. [4] are too small in the energy region shown. However, no such conclusion can be drawn at this time.

In Fig. 1(b), we compare the present calculated results with available experimental data. Our curved trajectory results agree with the data of Bliek et al. [4] below 80 eV/ nucleon but the straight-line trajectory results actually agree better with the data of Phaneuf et al. [1]. At higher energies above 200 eV/nucleon our results as well as the earlier results of FL, agree with the data of Hoekstra et al. [3]. In the 80–200 eV/nucleon region, both our result and the calculations of Gargaud et al. are all higher than the experimental data by about 20–35%.

It is appropriate to make some comments on the trajectory effects found in this calculation. In general, the AO expansion method is not applied to such slow collision. The procedure we adopted is an attempt to estimate the effect of the deflection of the heavy particles on the total electron-capture cross sections. The deflection will be different if a deuterium target atom is used. We have estimated the curved trajectory effect on the total electron-capture cross sections for C^{4+} on D collisions at 80, 60, 40, 20, and 10 eV/nucleon and the

results are 2.91, 2.39, 2.09, 1.21, and 0.63 in units of 10^{-15} cm^2, respectively. These numbers are about 10–30 % higher than the results for H target shown in Table II. This isotope dependence occurs at higher energies than the Si^{4+} on H system [16] but this can be explained by the fact that electron capture occurs at smaller impact parameters for the C^{4+} on H system. In the present model we did not include the effect due to the attractive induced dipole potential in the incoming path. This induced dipole potential will favor increasing cross sections for the H target. The combined influence of the attractive induced dipole potential on the incoming path and the repulsive Coulomb potential on the outgoing path may reduce the overall trajectory effect on the total electron-capture cross sections and thus the estimate we made here may be an upper bound.

To illustrate that we believe the reported cross sections

are essentially converged, we show the impact parameter dependence of $bP(b)$ for the probability of electron capture to $3p$ at 0.5 keV/nucleon in Fig. 2(a) and electron capture to $3d$ at 0.3 keV/nucleon in Fig. 2(b). The calculations were performed using the two different basis sets explained in the preceding section. Clearly the results are essentially independent of the basis set used to better than a few percent. We have also checked the calculations using other basis functions and found identical results. We thus believe that the results are converged. We comment that total electron capture is dominated by transitions occurring at large impact parameters where the atomic orbital expansion method is expected to be valid.

In addition to the total cross sections, partial cross sections or the ratios of electron capture to $3s$, $3p$, and $3d$ states have also been determined in a number of experiments [2,3,11]. In Fig. 3(a), we compare the present result for capture to $3s$ with the data of Hoekstra et al. [3]. The general agreement is very good. We mention that the results from Gargaud et al. and from Saha are also in general agreement with the data. In Fig. 3(b) we compare the electron-capture cross section to the $3p$ state obtained from the present calculation with the experimental data of Hoekstra et al. [3]. This is the dominant channel. There is a general good agreement except that the experiment has large error bars. In Fig. 3(c) we compare the electron-capture cross section to the $3d$ state obtained from the present work with the experimental data. The agreement with Hoekstra et al. is quite good above 150 eV/nucleon. Below this energy our results are slightly higher than the data of Hoekstra et al. but in agreement with the ratios measured by Baptist et al. (not shown). It is interesting to note that electron capture to the $3d$ state increases with decreasing collision velocity below 100 eV/nucleon. This result is identical to the calculations obtained by Gargaud et al. and of Saha. In terms of molecular potential curves this increase results from the crossing between the entrance channel and the $3d$ exit channel at $R=7.9$ a.u. At higher energies this crossing is diabatic but it becomes more effective in populating $3d$ states at lower energies, as expected from a typical Landau-Zener model.

IV. SUMMARY AND CONCLUSIONS

In this paper we reexamined the electron-capture cross sections for collisions between C^{4+} ions with H atoms in view of the new experimental data from Bliek et al. where a distinct dip in the cross section was found near 500 eV/nucleon. We have performed careful calculations based on the two-center atomic orbital expansion method and confirmed the earlier results of Fritsch and Lin for energies above 200 eV/nucleon. In both calculations we did not find any dip, which also agrees with the MO calculations. We have also extended the AO calculations to the lower energy region and adopted a heuristic method to account for the trajectory effect for collisions at low energies. When such a trajectory effect is accounted for, we can obtain absolute cross sections in agreement with experiment of Bliek et al. However, this would also imply that the trajectory effect is important for the present collision system at a rather high energy. If the present account of the trajectory effect is correct, then we would expect a significant isotope effect, which can be tested in future merged-beam experiments.

In conclusion, we believe that the observed dip near 500 eV/nucleon in the total electron-capture cross sections is questionable. On the other hand, there remain large discrepancies even in the total cross sections between existing experiments and among the different theories at low energies. While the total electron-capture cross sections for ion-atom collisions may be perceived to be well understood, the reality is that there are still significant discrepancies among results from the state-of-the-art experiments and the most sophisticated theoretical calculations, especially in the lower energy region. Until very precise measurements become available, it would be difficult to assess the accuracy of most of the present theoretical approaches and expose the limitation of each method.

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