

Low-energy electron capture by C^{3+} from hydrogen using merged beams

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Measurements of absolute total cross sections for electron capture by C^{3+} in collisions with ground-state hydrogen and deuterium are reported in the energy range 0.3–3000 eV/u. In general, good agreement is obtained with published experimental measurements at the lower (10–110 eV/u) and higher (> 1 keV/u) collision energies. However, the improved accuracy and the large energy range of these measurements made possible by the merged-beams technique indicate an energy dependence different than was suggested by interpolating previous published and unpublished measurements and by a theoretical calculation that attempted to reconcile the previous low- and high-energy total-capture cross-section data. The present measurements above 100 eV/u show excellent agreement with a more recent 22-state molecular-orbital calculation that predicts slight structure in the cross section at collision energies between 1000 and 2000 eV/u. Below 100 eV/u, the present measurements, which were performed with deuterium, deviate from an energy dependence suggested by earlier hydrogen measurements and by fully quantal calculations.

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I. INTRODUCTION

Electron-capture processes at low collision energies involving neutrals and multicharged ions are characterized by typically large cross sections ($> 10^{-15}$ cm²) and are therefore important in any environment containing multicharged ions and neutral species. For example, energy loss in high-temperature plasmas is affected by the radiation from impurity ions that have undergone capture to excited states. Carbon ions are one of the major impurities in magnetic fusion plasma devices. Low-energy (eV/u) measurements of charge transfer are also important for astrophysics, especially where, as is the case in the C^{3+} ion, the cross section is predicted [1,2] to sharply increase at low (near-thermal) energies. In the energy range of interest here, total-electron-transfer cross-section measurements are generally available in the keV/u energy range where a beam-gas-target method is applicable and where the atomic hydrogen target is typically formed by thermal dissociation of H_2 ; e.g., see Refs.

[3,4]. Generally, at the higher energies with collision velocity v , $0.1 < v < 1$ a.u., the total cross section is similar for ions of like charge; e.g., C^{4+} , O^{4+} , and Si^{4+} are predicted to have similar cross sections at keV/u collision energies. The behavior of the total-capture cross section has been successfully parametrized in terms of ionic charge, target binding energy, and collision energy (e.g., see the review by Gilbody [5]). However, at these energies, charge transfer to specific states is electron core dependent and has been investigated by both photon-emission [6] (PES) and translational-energy [7,8] (TES) spectroscopy.

At lower energies cross-section measurements are not extensive, and even total cross sections are known to be affected by the ionic core [3]. For example, calculations by Gargaud and McCarroll [9] predict that the total-capture cross section for Si^{4+} ions is a factor of about 20 larger than that for O^{4+} ions at 1 eV/u. Even though measurements of capture to specific states are not as available, total-capture cross sections at these energies do provide important information. Fully quantal molecular state calculations, which are generally assumed to be the most accurate at the lower energies, are difficult to perform. They are often sensitive to fine details in the method, and have only been tested on a few systems. At these energies, fewer states are involved in the capture. Only (avoided) crossings between initial and final states of the adiabatic potential curves at large internuclear separa-

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rations are active, and can lead to discernable structure even in the total-capture cross section [10–12]. Observation of these structures in the total-capture cross-section measurements is a sensitive test of theory.

The C^{3+} system is of interest for several reasons, some of which have already been mentioned. The ion is Li like, and therefore no long-lived metastables are known to exist [13], which obscure comparison with theory. Since we are also producing H (D) in the ground state the initial state of both collision partners is well characterized. There have been several experimental and theoretical investigations of the $C^{3+} + H$ collision system in the past. Total-electron-transfer cross sections have been measured by Cirić *et al.* [6] at collision energies of 0.92, 1.39, and 1.85 keV/u, by Gardner *et al.* [14] at 2.0 keV/u, and by Crandall, Phaneuf, and Meyer [15] at 2.8 keV/u. More extensive measurements above 5 keV/u have been performed by Phaneuf, Meyer, and McKnight [4] and Goffe, Shah, and Gilbody [16]. At low energies (10–110 eV/u) there have been measurements by Phaneuf *et al.* [3] using low velocity C^{3+} ions extracted from a laser-produced plasma. However, up until now there have been no measurements at intermediate collision energies to connect these data and provide details of the energy dependence. Calculations [1,2,17–19] have been performed based on the molecular curve-crossing model at lower energies. In an early attempt to span the energy range of interest here, distorted-wave calculations [19] extended molecular-orbital calculations [1,19] to 1–5 keV/u. These calculations seemed to reconcile the energy dependence suggested by the measurements at low and high energies. Unpublished total-capture measurements [20] by Yousif and Geddes, included in Ref. [8] and more recently in Ref. [21], show excellent agreement with these calculations. However, a more recent 22-state molecular-orbital calculation by Errea *et al.* [12] predicts a different energy dependence in the total cross section which, up until now, has not been observed in total-electron-capture measurements.

The lack of extensive total cross-section measurements for the C^{3+} system has hampered critical comparison of experimental and theoretical predictions for capture to specific states. Capture to specific states has been investigated by both photon-emission spectroscopy (PES) [6] at 0.7–4.6-keV collision energies, and by translational-energy spectroscopy (TES) [7,8] at 50–1500-eV/u collision energies. However, even though the two measurements for capture to the dominant channel ($1s^2 2s 3s$) 3S agree reasonably well within their limited energy overlap, the PES measurements compare best with the calculations of Errea *et al.* at higher energies, while the TES measurements agree with the calculations of Bienstock *et al.* at the lower energies. The curve crossing for the dominant channel is at a relatively large internuclear separation of $11.5a_0$. Comparison between measurements and theory is obscured by the fact that the TES measurements are normalized to the total-capture cross sections of Bienstock *et al.*, and cannot fully resolve different product states. In addition, the PES measurements have uncertainties of 30%. For capture to the dominant channel, the PES state-selective measurements [6] show a

much flatter energy dependence than that predicted by the *ab initio* calculations of Bienstock *et al.* These calculations also differ from the model-potential calculations of Opradolce *et al.* [18] which only considered contributions from the 3S and 3P product states but investigated the importance of translational factors above 50 eV/u. Their results indicate that the calculations of Bienstock *et al.* overestimate the total cross section due to the neglect of translational effects. However, these calculations only include a limited number of states and could not resolve the observed 3D contribution in the TES measurements. This was later attributed to a ladder mechanism by the calculations of Errea *et al.*, which include 22 molecular states. Up until now, measurements of total- and state-selective-capture cross sections were not sufficient to verify these calculations. A more detailed discussion is found in the literature [12,18,21].

The merged-beams method has successfully been used to measure total-electron-capture cross sections in the energy range between 0.1 and 1000 eV/u for various multicharged ions with ground-state hydrogen and deuterium (see Ref. [22] and references therein). In this paper we report on the absolute electron-capture cross section for $C^{3+} + H$ (D) collisions in the energy range between 0.3 and 3000 eV/u. With this one apparatus we are able to accurately measure total-electron-capture cross sections over such a large energy range that we not only extend measurements to lower energies but are able to connect previous high- and low-energy data and compare with the different theories needed to span such a large energy range.

II. EXPERIMENTAL METHOD

A. Merged-beams technique

Total-electron-capture cross sections are measured using the merged-beams method. Only a brief description will be presented here; for more details the reader is directed to Ref. [23]. In this technique, beams of neutral atoms and multicharged ions having energies in the keV range are merged, resulting in a relative velocity of the two beams that can be tuned over a very large range. The collision energy E_{rel} in eV/u, corresponds to the relative interaction energy of the two beams divided by the reduced mass and is calculated from

$$E_{\text{rel}} = E_1/m_1 + E_2/m_2 - 2\sqrt{(E_1 E_2)/(m_1 m_2)} \cos\theta, \quad (1)$$

where E_1, E_2 , and m_1, m_2 correspond to the energies (eV) and masses (a.u.) of the neutral and multicharged ion beams, respectively. The angle θ is the merge angle of the two beams which, to first order, is equal to zero.

Figure 1 is a simplified schematic of the apparatus. The C^{3+} ion beam is produced by the Oak Ridge National Laboratory (ORNL) Electron Cyclotron Resonance (ECR) ion source [24], and is merged electrostatically with a neutral H or D beam. The merged beams interact in a field-free region for a distance of 47 cm, after which the primary beams are magnetically separated from each other and from the product or signal H^+ (D^+) ions. The C^{2+} product of the reaction is not measured separately, but is collected together with the primary C^{3+} in a large

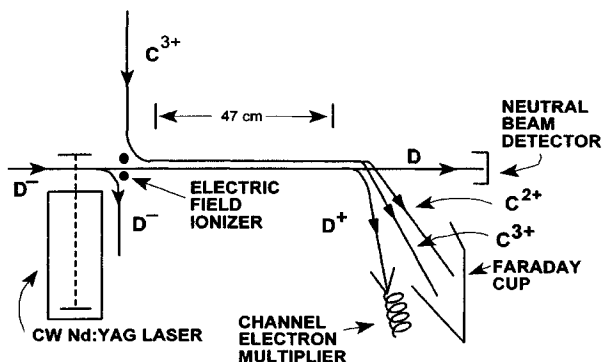


FIG. 1. Schematic of the ion-atom merged-beams apparatus for the $C^{3+} + D$ collision system.

Faraday cup. The neutral beam intensity is measured by secondary-electron emission from a stainless-steel plate, and the signal H^+ (D^+) ions are recorded by a channel electron multiplier operated in a pulse-counting mode. A 99.98% pure ground-state beam of H or D atoms is produced by passing a 6- to 9-keV beam of H^- or D^- ions through the optical cavity of a 1.06- μm continuous-wave (cw) Nd:YAG (yttrium aluminum garnet) laser, where up to 600 W of continuous power circulates and typically 0.5% of the negative ions undergo photodetachment. An electric-field ionizer is used to quench the excited H (D) atoms whose electrons are in high- n shells and which are produced by collisional stripping of H^- (D^-) on background gas. A nearly parallel beam of H (D) atoms is produced having a diameter of 2–4-mm FWHM (full width at half maximum) and an intensity of 10–20 (particle) nA. The divergence of this beam is typically less than 0.2° . A 50- to 90-keV, 2- to 5- μA beam of C^{3+} ions is produced by the ORNL ECR source with a typical diameter of 6–8-mm FWHM in the merge path and a divergence of less than 0.5° . The finite divergence of the primary beams results in a distribution of merging angles, creating a small absolute shift and energy spread in the collision energy (see below).

Electron-capture cross sections are determined absolutely by measuring the rate of H^+ (D^+) ions produced by the beam-beam interaction over the merge path. The cross-section value is determined at each velocity from directly measurable parameters by the following formula:

$$\sigma = \frac{Rqe^2\gamma v_1 v_2}{I_1 I_2 \epsilon \langle F \rangle L v_r}, \quad (2)$$

where R is the signal count rate, q the charge of the ion, e is the electronic charge, γ is the secondary electron emission coefficient of the neutral beam detector, I_1 and I_2 are the intensities of the two beams, ϵ is the efficiency for detecting the product H^+ or D^+ , $\langle F \rangle$ is the average form factor which is a measure of the overlap of the beams over the merge-path of length L , v_1 and v_2 are the velocities of the beams, and v_r is the relative velocity between beams. The integrated three-dimensional form factor is determined from two-dimensional measurements of the overlap at three different positions along the merge

path. The secondary electron emission coefficient γ was measured *in situ* [23] by modulating the laser beam and comparing the decrease in the negative ion beam to the measured increase in the neutral beam. γ was found to be 1.78 ± 0.06 for measurements with 8.6-kV H, and 1.45 ± 0.03 for measurements with 8.6-kV D. These values are somewhat higher than what was determined previously for other collision systems. It was found that the magnitude of γ depended on the C^{3+} multicharged ion beam being present in the demerger chamber. Apparently, the additional C deposited on the stainless-steel surface of the neutral beam detector caused an increase in γ , the number of electrons emitted per incident neutral atom.

The H^+ or D^+ product ions are detected in pulse-counting mode using a 1-in-diameter channel electron multiplier (CEM) with an extended cone. A voltage of -3000 V is applied to the front of the detector to further accelerate the positive ions before they strike the CEM surface. The total counting efficiency (electronics plus detector efficiency) is estimated [23,25] to be 0.97. The signal rate R is separated from the background by using a two-beam modulation technique. Backgrounds on the order of 10 kHz were produced by the fast neutral beam H (D) stripping on background gas in the merged path where pressures were on the order of 1×10^{-10} Torr. Backgrounds on the order of 80 Hz were a result of the C^{3+} collection in the Faraday cup. Signal rates on the order of 30 Hz were observed at the higher collision energies, the signal decreasing to a few Hz at the lower energies due mainly to the number of collisions in the merge path decreasing as v_r .

B. H (D) and C^{3+} beam purity

As has been previously observed [22] in merged-beams studies of $O^{5+} + H$ (D) at collision energies below 100 eV/u, a very small (0.02%) Rydberg population in the H (D) beam can result in significant beam-beam signal compared to that due to capture from the ground state. The excited states of H (D) are created by collisional stripping of the H^- (D^-) beam on background gas. Some of the excited H (D) have trajectories which allow the atoms to merge with the multicharged ions and, if electron loss occurs, produce a beam-beam H^+ (D^+) signal. The cross section for electron removal from an excited state of H with an electron in, e.g., an n shell of 20 is typically a factor of 10^5 greater than the capture cross section from the ground state [26]. Thus contributions from excited states can be a significant fraction of the beam-beam signal, and have been studied in some detail using this apparatus [26]. To correct for the signal due to the excited states, the beam-beam signal was measured with the laser on and then off, the appropriate difference between the signals corresponding to the signal due to the ground state. To reduce the Rydberg population, the H (D) beam is made to pass through a field ionizer (see Fig. 1) which ionizes the higher n levels of the excited H (D), causing them to be swept out of the neutral beam. The electric field required to field ionize a particular n shell in H or D is approximated by the semiempirical relation (see Ref. [26])

$$E = \frac{6.25 \times 10^5 \text{ kV/cm}}{n^4} \quad (3)$$

For some of the measurements reported here, highly excited states of H and D were ionized down to $n = 12$ by applying an electric field of 30 kV/cm. This reduction in the excited component of the H (D) beam led to significantly smaller beam-beam signal corrections and reduced the time for measurements. Depending on such variables as vacuum conditions, the efficiency of photodetachment, and the strength of the signal being measured, it was observed that the applied electric field is able to reduce the component of signal due to the excited states from typically 20–30 % to a few percent.

It was also found that the C^{3+} beam from the ORNL ECR had an O^{4+} (same m/q) contaminant due to the background O_2 gas present in the ion source. The O^{4+} contaminant comprised $7 \pm 3\%$ of the measured intensity of the C^{3+} beam, as estimated from an average of the O^{3+} and O^{5+} analyzed beam intensities. Since the O^{4+} beam velocity was identical to the C^{3+} velocity [which resulted in the same collision energy with the H (D)], and since the cross section [27] for total capture with O^{4+} has been measured using the ORNL ion-atom merged-beams apparatus, corrections to the data can be made. In addition, for comparison, a few measurements were performed with ^{13}C whose m/q did not overlap with any other ion from the source.

C. D^+ signal collection

Since the low-energy electron-capture collisions under study are exoergic, and since both products are positively charged, significant angular scattering of the D^+ or especially the H^+ products can occur in the center-of-mass frame [28]. However, due to the kinematic frame transformation, this angular scattering is significantly compressed in the laboratory frame where the products are collected. The angular acceptance of the apparatus in the lab frame is 2.3° , as determined by ray tracings and verified by comparison of data to theory for the O^{5+} system (see Ref. [22]). From this estimate one can determine the maximum angle into which the product D^+ can be emitted in the center-of-mass frame and still be collected. This maximum angular acceptance is a function of collision energy, the velocity of the center of mass, and the exoergicity of the capture process. Figure 2 shows this maximum angular acceptance in the center-of-mass frame in the forward direction as a function of collision energy for capture into the $C^{2+}(1s^2 2s 3s)^3S$, $C^{2+}(1s^2 2p^2)^1S$, and $C^{2+}(1s^2 2p^2)^1D$ states with exoergicities [21] of 4.72, 11.6, and 16.2 eV, respectively. For scattering in the backward direction the angular acceptance is similar, since only the velocity component perpendicular to the beam leads to a loss of signal. The figure was constructed for an 8.6-kV D beam, the neutral beam used in the measurements below 600 eV/u. To access the higher collision energies, where angular scattering is not expected to be a problem [29], an 8.6-kV H beam was used. For any given capture channel, the maximum angular acceptance improves as the collision energy decreases, due to the decrease in the initial relative ve-

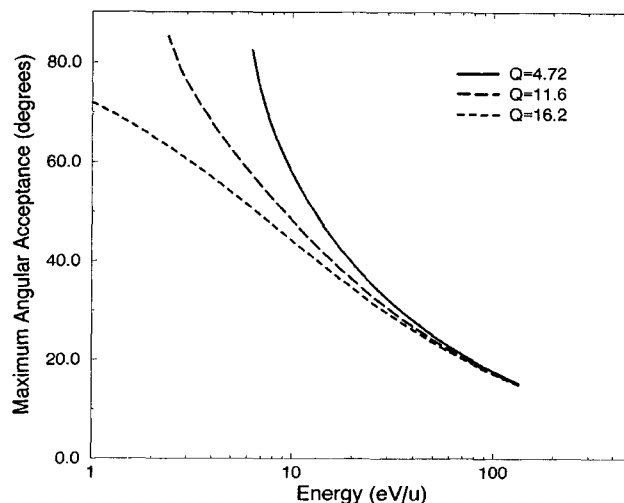


FIG. 2. Angular acceptance in the forward direction in the center-of-mass frame as a function of collision energy for measurements of the $C^{3+} + D$ system, using a 8.6-kV D beam. Angular acceptance in the backward direction is similar and not shown.

locity that is assumed (in these calculations) to scatter off axis. Also, since the final velocity is a sum of this initial velocity and the velocity kick provided by the exoergicity of the reaction, the capture channel with the lowest exoergicity, Q , has the larger angular acceptance. One can see from the figure that for the 3S capture channel ($Q=4.72$ eV) below 6 eV/u the angular acceptance approaches 90° ; hence all the product D^+ is collected. For capture to the nondominant channels, e.g., the 1D with $Q=16.2$, any D^+ product scattered by more than about 45° at a collision energy of 10 eV/u would not be collected. However, this channel accounts for only 1% of the total cross section at 100 eV/u with this percentage predicted [12,19] to decrease for lower collision energies. For capture to the 1S state, there is a slightly higher angular acceptance (see Fig. 2); however, this capture channel is predicted [19] to account for approximately 20% of the cross section at 10 eV/u.

D. Relative and absolute uncertainties

The voltage dividers that measure the accelerating potentials of the two sources are calibrated to each other to within a relative accuracy of 2 V. The absolute voltage is determined to within 0.05%. These accelerating potentials are modified by the unknown plasma space potential of both the ECR and duoplasmatron ion sources. Estimates of $+20 \pm 10$ V per charge for the ECR was obtained by measuring the voltage applied to the merger (which is a parallel plate analyzer) vs the voltage on the ECR ion source. From the positive intercept one obtains the estimate for the ECR plasma potential. A plasma shift of +10 V was estimated for the duoplasmatron. Both these plasma potentials are used in Eq. (1) to estimate the absolute shift in center-of-mass collision energy as a result of these potential shifts and are presented in

TABLE I. Collision energy shifts due to the ECR and duoplasmatron source plasma potentials, and the nonzero merge angle of the two beams.

Center-of-mass collision energy (eV/u)	Center-of-mass energy shift due to $(q \times 20)$ V (ECR source) (eV/u)	Center-of-mass energy shift due to 10 V (duoplasmatron) (eV/u)	Center-of-mass energy shift due to merge angle (eV/u)	Center-of-mass total-energy shift (eV/u)
0.1	-0.02	-0.02	0.16	0.12
0.5	-0.05	-0.05	0.16	0.06
1.0	-0.08	-0.07	0.01	0.01
5.0	-0.18	-0.17	0.15	-0.2
10.0	-0.25	-0.24	0.15	-0.34
50.0	-0.60	-0.54	0.14	-1.0
100	-0.9	-0.76	0.13	-1.53
500	-2.6	-1.7	0.1	-4.2

Table I. Also included in the table is the shift in collision energy due to nonzero merging angles. The range of merging angles is estimated from the range of trajectories within the envelope of the larger multicharged ion beam. The envelope is estimated by the FWHM of the measured horizontal and vertical profiles, which resulted in a maximum possible merging angle to be on the order of 0.7° . Therefore, it is estimated that the bulk of collisions occur with a merge angle of $0.35^\circ \pm 0.35^\circ$. As can be seen from the table, for energies of 1 eV/u and greater, the resultant total absolute shift in collision energy, which is sum of all the contributions, is of little significance when compared to the collision energies. For the measurements reported here, only the collision energy below 1 eV/u is corrected.

There is also a spread in collision energy due primarily to the spread in merging angle. Like the shifts in energy this is only of significance below 1 eV/u. The spread in energy is comparable to the shift in energy due to nonzero merge angles, whose values are presented in the fourth column of Table I.

Relative uncertainties in the measured signal are mostly due to the statistical uncertainties in separating the beam-beam signal from the backgrounds. Occasionally spatial instabilities in the beams whose effect on the beam-beam overlap was beyond our ability to monitor added an additional component which was combined in quadrature. Absolute uncertainties in the cross section were measured to be 12% at the 90% confidence level. A detailed discussion of these uncertainties can be found in Ref. [23].

III. RESULTS AND DISCUSSION

The measured absolute total-electron-capture cross sections for $C^{3+} + H(D)$ are represented in Table II, and include measurements with ^{12}C and ^{13}C ions, and with H and D neutrals. Performing measurements with D provides increased angular collection at the lower energies, while measurements with H provide a larger dynamic range in collision energy [23]. The total uncertainties correspond to a quadrature sum of the relative and absolute uncertainties estimated at the 90% confidence level. Estimated absolute and relative uncertainties in the collision energy, as discussed in Sec. II, were applied only to

the lowest energy where the corrections are of significance.

Figure 3 shows a plot of the present results compared with other measurements and theory over an energy range from 0.1– 10^5 eV/u. All error bars on the present measurements correspond to the relative uncertainties estimated at a 90% confidence level, as presented in Table II. Total uncertainties that include the absolute errors are only slightly larger than the relative errors and are not indicated in the figure. The cross-section values reported at 2.7, 14.8, and 312 eV/u were performed with ^{13}C instead of ^{12}C . As discussed previously, due to the 7% O^{4+} contamination of the C^{3+} ion beam, corrections were made to the cross section using previous O^{4+} data [27].

As can be seen from Fig. 3, the previous measurements at the higher energies show the characteristic maximum in the cross section which occurs when the collision ve-

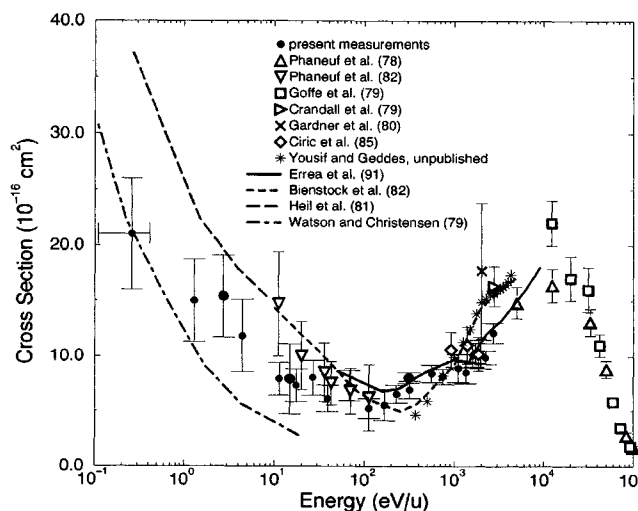


FIG. 3. Comparison of merged-beams data for electron capture in $C^{3+} + H(D)$ collisions with previous measurements and theory. All vertical error bars denote relative uncertainties estimated at 90% confidence level. The measurements at 2.7, 14.8, and 312 eV/u are performed with ^{13}C instead of ^{12}C .

TABLE II. Total-electron-capture cross sections for $C^{3+} + H(D)$. All uncertainties are estimated at the 90% confidence level. The total uncertainty represents the quadrature sum of the relative and absolute uncertainties.

Ion	Neutral	Collision energy (eV/u)	Cross section (10^{-16} cm 2)	Relative uncert. (10^{-16} cm 2)	Total uncert. (10^{-16} cm 2)
^{12}C	D	0.26 \pm 0.15	21.0	5.0	5.6
^{12}C	D	1.3	15.0	3.7	4.1
^{13}C	D	2.7	15.4	3.7	4.1
^{12}C	D	4.4	11.8	3.3	3.6
^{12}C	D	11.3	7.9	1.5	1.8
^{13}C	D	14.8	7.9	3.2	3.3
^{12}C	D	17.4	7.3	1.5	1.7
^{12}C	D	26.7	8.0	1.6	1.9
^{12}C	D	39.0	6.1	1.2	1.4
^{12}C	D	68.0	7.3	1.4	1.6
^{12}C	D	112	5.2	0.9	1.1
^{12}C	D	167	5.5	1.4	1.6
^{12}C	D	230	6.5	0.8	1.1
^{13}C	D	312	8.0	0.5	1.1
^{12}C	D	318	6.9	0.8	1.2
^{12}C	D	562	8.4	0.8	1.3
^{12}C	H	747	8.1	0.7	1.2
^{12}C	H	1099	8.9	1.3	1.7
^{12}C	H	1341	8.5	1.0	1.4
^{12}C	H	1601	10.1	1.1	1.7
^{12}C	H	1700	10.3	0.9	1.5
^{12}C	H	1958	10.2	1.3	1.8
^{12}C	H	2210	9.9	0.6	1.3
^{12}C	H	2714	12.1	0.9	1.7

locity corresponds to the velocity of the electron in H ($v=1$ a.u.). On the low-energy side of this maximum one can see that the present data show a different energy dependence compared to what is suggested by some of the earlier published measurements. At a collision energy of 2700 eV/u our measurements are 25% lower than a measurement by Crandall, Phaneuf, and Meyer [15], and at around 2 keV/u our measurements are almost a factor of 2 below that of Gardner *et al.* [14]. The present data are consistent with the three data points of Ćirić *et al.* [6]; however, these measurements have relatively large uncertainties which preclude detailed comparison. At lower energies between 10 and 100 eV/u, our measurements agree well with the previous measurements by Phaneuf *et al.* [3]; however, those measurements tend to suggest a more rapid increase in the cross section as the collision energy decreases below 100 eV/u. Our measurements do not show such a sharp rise in the cross section until the collision energies are below 10 eV/u.

As discussed above, the previous low- and high-energy total-capture cross-section measurements suggest a steep energy dependence above 100 eV/u that was reconciled by the calculations of Bienstock *et al.* [19]. In order to span such a large energy range, though, their calculations use a coupled-channel method at the lower energies and a distorted-wave method at the higher energies. As can be seen in Fig. 3, the unpublished total-electron-capture measurements by Yousif and Geddes [20] show excellent agreement with the calculations of Bienstock *et al.*, and have led to the latter's use in normalization of the TES

measurements, as recently reviewed in Ref. [21].

Our present measurements, in contrast, show excellent agreement with the 22-state molecular-orbital calculation of Errea *et al.* [12], even reproducing the structure in the total cross section between 1000 and 2000 eV/u. Comparison between the present measurements and those of Yousif and Geddes shows differences in the energy dependence of the cross section (as conveyed by the plotted relative uncertainties). However, when the total uncertainties of the measurements are considered (the measurements of Yousif and Geddes have a total uncertainty of 8%; the total uncertainties of the present measurements are reported in Table II), the discrepancy seems less dramatic. Below 10 eV/u the cross section of the present measurements is observed to increase with the general slope predicted by both Watson and Christensen [2], who only considered capture to the $C^{2+}(2s3s)^3S$ final state, and to Heil, Butler, and Dalgarno [1]. There are several factors to consider at these low energies which may lead to a lower measured cross section. Angular scattering beyond the angular acceptance of the apparatus would tend to make our measurements underestimate the cross section. In fact, using the estimate of the angular acceptance as presented in Fig. 2 at 10 eV/u, the deviation of the observed cross section from the calculations of Bienstock *et al.* could be accounted for by assuming isotropic emission. At eV/u energies, though, the angular collection for the dominant channels dramatically increases, and therefore angular collection should not be a factor. Another possibility for a loss in signal is due

to the isotope effect [22]. Our low-energy measurements are performed with D instead of H, which may tend to reduce the trajectory-effect enhancement in the cross section. This enhancement is a result of the ion-induced dipole attraction affecting the trajectory of the reactants, which results in a sweeping of the initial impact parameter to smaller internuclear minimum separations. This isotope effect has been estimated on the basis of Landau-Zener calculations [22] to be on the order of 20% for the $O^{5+} + H$ (D) system at a collision energy of 0.1 eV/u. This effect, though, is dependent on the ion charge and the specifics of the collisions system.

IV. CONCLUSIONS

The total-electron-capture cross section for the $C^{3+} + H$ (D) system is of importance, and has been studied in detail both experimentally and theoretically for

several years by different techniques. Experimentally, since both the C^{3+} and H (D) can be produced in the ground state, there are no metastable components to interfere with a detailed comparison between theory and experiment. The present measurements extend over four orders of magnitude in collision energy, and are of sufficient accuracy to provide a benchmark for comparison with theory. The present measurements show excellent agreement with the more recent calculations of Errea *et al.*, and even show evidence for the predicted feature in the total cross section between 1000 and 2000 eV/u.

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- [1] T. G. Heil, S. E. Butler, and A. Dalgarno, *Phys. Rev. A* **23**, 1100 (1981).
 - [2] W. D. Watson and R. B. Christensen, *Astrophys. J.* **231**, 627 (1979).
 - [3] R. A. Phaneuf, I. Alvarez, F. W. Meyer, and D. H. Crandall, *Phys. Rev. A* **26**, 1892 (1982).
 - [4] R. A. Phaneuf, F. W. Meyer, and R. H. McKnight, *Phys. Rev. A* **17**, 534 (1978).
 - [5] H. B. Gilbody, *Adv. At. Mol. Phys.* **22**, 143 (1986).
 - [6] D. Ćirić *et al.*, *J. Phys. B* **18**, 3629 (1985).
 - [7] R. W. McCullough, F. G. Wilkie, and H. B. Gilbody, *J. Phys. B* **17**, 1373 (1984).
 - [8] F. G. Wilkie, R. W. McCullough, and H. B. Gilbody, *J. Phys. B* **19**, 239 (1986).
 - [9] M. Gargaud and R. McCarroll, *J. Phys. B* **21**, 513 (1988).
 - [10] B. Zygelman *et al.*, *Phys. Rev. A* **46**, 3846 (1992).
 - [11] N. Shimakura, M. Itoh, and M. Kimura, *Phys. Rev. A* **45**, 267 (1992).
 - [12] L. F. Errea, B. Herrero, L. Méndez, and A. Riera, *J. Phys. B* **24**, 4061 (1991).
 - [13] R. F. Welton, T. F. Moran, and E. W. Thomas, *J. Phys. B* **24**, 3815 (1991).
 - [14] L. D. Gardner *et al.*, *Phys. Rev. A* **21**, 1397 (1980).
 - [15] D. H. Crandall, R. A. Phaneuf, and F. W. Meyer, *Phys. Rev. A* **19**, 504 (1979).
 - [16] T. V. Goffe, M. B. Shah, and H. B. Gilbody, *J. Phys. B* **12**, 3763 (1979).
 - [17] R. J. Blint, W. D. Watson, and R. B. Christensen, *Astrophys. J.* **205**, 634 (1976).
 - [18] L. Opradolce, L. Benmeuraim, R. McCarroll, and R. D. Piacentini, *J. Phys. B* **21**, 503 (1988).
 - [19] S. Bienstock, T. G. Heil, C. Bottcher, and A. Dalgarno, *Phys. Rev. A* **25**, 2850 (1982).
 - [20] R. W. McCullough (private communication).
 - [21] H. B. Gilbody, *Adv. At. Mol. Opt. Phys.* **32**, 149 (1994).
 - [22] C. C. Havener, F. W. Meyer, and R. A. Phaneuf, in *Physics of Electronic and Atomic Collisions*, edited by W. R. McGillivray, I. E. McCarthy, and M. C. Standage (AIP, New York, 1992), p. 381.
 - [23] C. C. Havener *et al.*, *Phys. Rev. A* **39**, 1725 (1989).
 - [24] F. W. Meyer, *Nucl. Instrum. Methods Phys. Res. Sec. B* **9**, 532 (1985).
 - [25] D. H. Crandall and J. A. Ray, *Rev. Sci. Instrum.* **46**, 562 (1975).
 - [26] C. C. Havener *et al.*, in *Physics of Highly Charged Ions*, edited by P. Richard, M. Stockli, L. Koch, and C. D. Lin, AIP Conf. Proc. No. 274 (AIP, New York, 1993), p. 43.
 - [27] C. C. Havener, M. P. Nesnidal, M. R. Porter, and R. A. Phaneuf, *Nucl. Instrum. Methods* **56/57**, 95 (1991).
 - [28] R. E. Olson and M. Kimura, *J. Phys. B* **15**, 4231 (1982).
 - [29] L. R. Andersson, M. Gargaud, and R. McCarroll, *J. Phys. B* **24**, 2073 (1991).