Low-energy electron capture by C^{4+} ions from atomic hydrogen

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The total-electron-capture cross section for collisions of C^{4+} with ground-state hydrogen (deuterium) is measured in the energy range 6–1000 eV/u using the merged-beam technique. The fraction of C^{4+} metastable ions present in the ion beam is measured to be 5%, which results in a correction to the cross section of only a few percent. The independently absolute measurements are generally in good agreement with previous measurements for total electron capture; however, the reduced uncertainties of these measurements allow a more detailed comparison with theory. Our observations show that existing fully quantal molecular-orbital calculations that include rotational coupling overestimate the cross section by approximately 25%. Better agreement is found with a recent semiclassical impact-parameter coupled-channel calculation. While all available theoretical data predict that the cross section varies smoothly with energy around the maximum, a sharp structure is observed around 400 eV/u. [S1050-2947(97)04107-3]

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I. INTRODUCTION

Low-energy electron capture by highly charged ions from atomic hydrogen continues to generate considerable interest not only because of practical applications in studies of astrophysical plasmas [1,2] and particle transport in the divertor region of thermonuclear fusion devices [3] but also because of the fundamental atomic physics processes that occur during these low-energy collisions. Most of the fully quantum mechanical models describing these one-electron or quasi-one-electron systems consist of a procedure to decompose the time-dependent electronic wave function in either atomic or molecular orbitals. Different theoretical approaches often show large discrepancies, e.g., up to 30% for the C^{4+} + H collision system. Experimental data, when existing, often are not accurate enough to discriminate between theories. Therefore accurate and systematic experimental data are necessary to establish a benchmark for theoretical predictions.

The present state-of-the-art experimental merged-beam technique provides accurate absolute cross sections over an extended energy range enabling a detailed investigation of electron capture processes. In this paper the absolute total-electron-capture cross section for the following reaction is presented:

$$C^{4+} + H(D) \rightarrow C^{3+} + H^{+}(D^{+}). \quad (1)$$

The absolute cross section has been measured using the Oak Ridge National Laboratory ion-atom merged-beam apparatus [4,5] in the energy range between 6 and 1000 eV/u. Numerous theoretical studies have been performed on this collision system, which can be treated as having effectively only one active electron (the C^{4+} ion has a $1s^2$ closed shell). Early semiclassical close-coupling calculations were performed by Olson, Shipsey, and Browne [6] and later by Hanssen *et al.* [7]. Other calculations include a modified two-center atomic-orbital (AO^{+}) expansion performed by Fritsch and Lin [8] and fully quantal molecular-orbital calculations by Gargaud and McCarroll [9] (MO4) and Bottcher and Heil [10] with a limited basis set. Gargaud, McCarroll, and Valiron [11] have also used an extended basis set (MO7) with more accurate molecular potentials and include rotational couplings and electron translation factors. The MO7 calculation shows that the effect of rotational coupling is strong and leads to an increase of up to 30% in the cross section in the energy range 20–500 eV/u. Using a diabatic formalism, a calculation by Andersson and McCarroll [12] agrees with the results of the MO7 calculation. A recent semiclassical impact-parameter calculation by Saha [13] uses a molecular basis with rotational coupling. It shows reasonable agreement with the MO7 calculation, except at the peak in the total cross section, where the cross section is significantly lower. A more in-depth discussion of the various theories and approximations can be found in Ref. [13].

A number of experimental works have also been performed. Both state-selective photon emission spectroscopy and total-capture measurements down to 1000 eV/u have been performed by Dijkkamp *et al.* [14] using a crossed-beam configuration with a partially dissociated hydrogen beam effused from a radio-frequency discharge source. By deceleration of the C^{4+} ion beam, this technique was extended to 50 eV/u by Hoekstra *et al.* [15]. Using a cool source of ions from a laser-produced plasma and a hydrogen furnace as a target, Phaneuf *et al.* [16] were able to extend the total-capture measurements to 15 eV/u. Generally, the total-cross-section measurements are not of sufficient accuracy to distinguish between the various theories, but do show differences with the MO7 calculation in the energy range 100–1000 eV/u. The state-selective measurements of Hoekstra *et al.* [15] indicate that the (MO7) molecular-orbital calculation agrees best with the experimental observations for capture into the individual l subshells. However, discrepan-

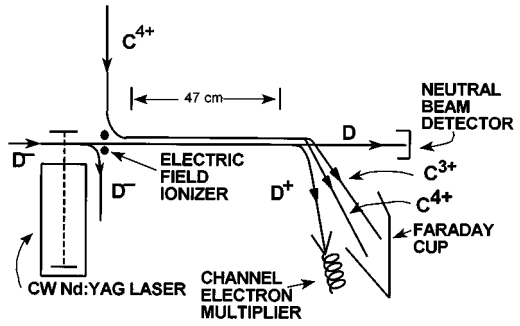

FIG. 1. Schematic of the ion-atom merged-beam apparatus.

cies remain. For example, the MO7 calculation and the more recent calculation by Saha [13] predict that the cross section for capture into the $3d$ state increases strongly below 0.1 keV/u. This is also observed in the measurements of Baptist *et al.* [17]. However, the measurements of Hoekstra *et al.* show a decrease of the population of the $3d$ state down to 50 eV/u. A further investigation of this collision system is therefore warranted and presented in this paper.

II. EXPERIMENTAL METHOD

A. Merged-beam technique

The total-electron-capture cross section is measured using the merged-beam technique, which will only be described briefly here. For more experimental detail the reader is referred to the literature [4,5]. A merged-beam configuration provides a large dynamic range of collision energies and is the only technique available to access thermal energies with good energy resolution. Relatively fast (keV) beams of multicharged ions and neutral H or D are merged, resulting in relative velocities tunable over a very large range. The collision energy $E_{\rm rel}$ in eV/u is given by

$$E_{\rm rel}=\frac{E_1}{m_1}+\frac{E_2}{m_2}-2\sqrt{\frac{E_1E_2}{m_1m_2}}\cos\theta, \tag{2}$$

where E_1 and m_1 correspond to the energy (eV) and the mass (a.u.) of the neutral beam and E_2 and m_2 to those of the multiply charged ion beam. In most cases, the merge angle θ is small and can be set equal to zero. At thermal energies, though, the merge angle limits the lowest center-of-mass energy that can be reached [18].

The C^{4+} beams are produced in the ORNL CAPRICE electron cyclotron resonance (ECR) ion source and electrostatically merged with a neutral H(D) beam. A schematic diagram of the apparatus is depicted in Fig. 1. The neutral beam is a 99.98% pure ground-state H(D) beam, which is produced by photodetachment of an 8-keV $H^-(D^-)$ beam as it passes through an optical cavity of a 1.06-μm Nd:YAG laser (where YAG denotes yttrium aluminum garnet). The small fraction of H(D) atoms in Rydberg states is produced by collisional stripping of $H^-(D^-)$ on the background gas. As shown in Fig. 1, the neutral beam passes through a strong-electric-field (30 kV/cm) ionizing excited H(D) atoms whose electrons are in high-n shells and Stark mixing followed by radiative quenching lower-n excited states. The correction [19] to the cross section is thereby reduced to less than 10%. The merged beams interact in a field-free region of 47 cm, after which the beams are separated magnetically from each other. The product or ''signal'' $H^+(D^+)$ beam is focused by a pair of einzel lenses positioned before and after the magnetic dispersion, is deflected out of the plane of dispersion by electrostatic deflectors, and is then detected by a channel electron multiplier (CEM) operating in a pulse counting mode. The C^{3+} product beam is collected together with the C^{4+} beam in a Faraday cup. The neutral beam intensity is measured by the secondary electron emission from a stainless-steel plate.

The 50–90 keV C^{4+} beam, with an intensity of 1–3 μA in the merge path, has a typical divergence of less than 0.5°. The beam diameter is typically on the order of 6–8 mm full width at half maximum (FWHM). The 50-(particle) nA neutral beam of H(D) atoms is nearly parallel having a diameter of 2–4 mm FWHM and a divergence less than 0.2°. 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.

The total-electron-capture cross sections are determined absolutely by measuring the rate of $H^+(D^+)$ ions produced by the beam-beam interaction over the merge length L. The cross-section value is determined by

$$\sigma=\frac{R}{\epsilon}\frac{\gamma qe^2}{I_1I_2}\frac{v_1v_2}{v_r}\frac{1}{\Omega L}, \tag{3}$$

where R is the $H^+(D^+)$ count rate, ϵ is the efficiency of the CEM for detecting the $H^+(D^+)$, γ is the secondary electron emission coefficient for the neutral beam detector, q is the charge state of the ion, e is the electronic charge, I_1 and I_2 are the measured intensities of the two beams, v_1 and v_2 are the velocities of the two beams, and v_r is the relative velocity between the beams. Ω is the effective form factor and is a measure of the spatial overlap of the beams at three different positions along the merged-path L. The numerical value for γ was determined *in situ* [4].

The $H^+(D^+)$ product ions are detected by a CEM with a diameter of 2.54 cm operating in pulse counting mode. A voltage of -3000 V is applied to the front of the detector to accelerate the positive ions further before they strike the CEM. The total detection efficiency ϵ for both the electronics and the detector is estimated [4,20] to be 0.97. The signal rate 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 H(D) beam stripping on the background gas in the merged path. Although the pressure in the merge path is typically on the order of 1×10^{-8} Pa, this still results in a target thickness for producing background that is a factor of 1000 times higher than the effective target thickess for producing signal. Additional background on the order of 80–100 Hz was a result of photons emitted by the C^{4+} in the Faraday cup.

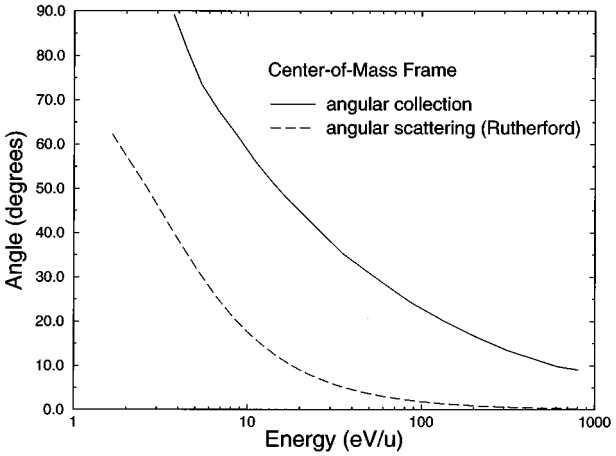

FIG. 2. Angular collection and an angular scattering estimate (see the text) as a function of collision energy in the center-of-mass frame.

B. H^+ signal collection

Electron capture onto C^{4+} from atomic hydrogen is an exoergetic reaction with excess energy denoted by Q. With reaction products positively charged, there can be significant angular scattering in the center-of-mass frame, especially at low collision energies. The angular scattering $\theta_{\text{c.m.}}$ is ''compressed'' in the laboratory frame where the observed scattering θ_{lab} is related to $\theta_{\text{c.m.}}$ by the relation

$$\tan(\theta_{\text{lab}}) = \frac{\frac{\mu}{m_1} v_r^f \sin\theta_{\text{c.m.}}}{V_{\text{c.m.}} + \frac{\mu}{m_1} v_r^f \cos\theta_{\text{c.m.}}}, \tag{4}$$

where $V_{\text{c.m.}}$ is the velocity in the center-of-mass frame, v_r^f is the final relative velocity after the collision, m_1 is the mass of the faster collision partner, and μ is the reduced mass. In the denominator the plus sign is replaced by a minus sign when m_1 is the slower collision partner. v_r^f is larger than the incident relative velocity due to the increase in energy of the reaction Q. The angular acceptance of the current apparatus θ_{lab} is 2.3° and has been estimated by particle trajectory simulations and verified by comparison of data with theory for the O^{5+} + H(D) system (see Ref. [21]). Using Eq. (4) one can estimate the angle over which the D^+ can be emitted and still be detected by the CEM. For this collision system the value for Q is averaged over capture into all $3l$ subshells and estimated to be 11.8 eV at an internuclear separation of $7.5a_0$. Figure 2 shows the maximum angular acceptance in the center-of-mass frame as a function of collision energy. In order to estimate the angular scattering, the ''half-Coulomb'' Rutherford minimum scattering (see Ref. [22]) is also shown in Fig. 2 for an 8-keV D beam for the C^{4+} system. It is clear that all the D^+ product ions are collected over the complete energy range under investigation, unless the real angular scattering is much larger than the Rutherford scattering estimate. Above 400 eV/u the data were taken with H rather than D. For collisions with H (not shown), the scattering is only slightly greater while the collection is only slightly less.

TABLE I. Binding energies for $3l$ states with different core configurations.

State	$1s^2$	$(1s2s)^1S$	$(1s2s)^3S$
$3s\ ^2S$	26.93	29.19	27.70
$3s\ ^4S$			29.84
$3p\ ^2D$	24.81	27.16	26.31
$3p\ ^4P$			27.32
$3d\ ^2D$	24.2	25.54	24.85
$3d\ ^4D$			25.70

C. Metastable ion fraction

The cross section for capture into an ion with an excited core can differ by orders of magnitude from capture into an ion in the ground state [23]. Therefore, it is very important to know the exact fraction of metastables present in the ion beam when a comparison is made with theory for a projectile in a ground-state core configuration. For the collision system investigated, the following metastable states are involved: the C^{4+} $(1s2s)^1S$ state and the C^{4+} $(1s2s)^3S$ state. The lifetimes of these states are calculated [24] to be 3.3 μs and 112 s, respectively. Since the flight time from the ECR source to the merging section is on the order of 12 μs, the effect of the C^{4+} $(1s2s)^1S$ can be neglected.

The metastable fraction of the incident C^{4+} ion beam was determined using the ORNL electron-ion crossed-beam apparatus [25]. The cross section for electron-impact single ionization of C^{4+} was measured from 100 eV to 1500 eV using the same ion beam (similar ion source conditions) as for the capture experiment. The measured cross sections were least-squares fit with a sum of two one-parameter Lotz [26] functions representing the ionization of C^{4+} metastable $(1s2s)$ and ground $(1s^2)$ configurations with thresholds of 93.13 eV and 392.08 eV, respectively. The metastable fraction of the C^{4+} ion beam was thereby determined to be 0.05 ± 0.01, which is in good agreement with previous studies [27].

To *estimate* the cross section for capture onto this metastable core, a multichannel Landau-Zener calculation was performed using the Olson-Salop-Taulbjerg coupling-matrix elements [28–30] and asumming straight-line trajectories. The binding energies for the $3l$ electrons with a C^{4+} $(1s2s)^1S$ and C^{4+} $(1s2s)^3S$ core have been calculated with the COWAN code [31] using a statistical distribution over all J states, i.e., weighting coefficients by $2J+1$. For completeness, the singlet and triplet energies are shown in Table I.

The effect on the measured cross sections can now be estimated by the ratio of the cross sections for capture by a ground state core ion and an ion with an excited core (see Fig. 3). As a cross-check, the same calculation for the C^{4+} + H_2 system has been performed and found to compare favorably with the results of Guillemot *et al.* [32]. From Fig. 3 it is clear that for C^{4+} + H, the cross section for capture with a ground-state core is always larger than for capture by an ion with an excited core. At higher energies, though, the ratio is only on the order of 1.25. The measured cross section, then, taken with an ion beam with 5% metastable states, is estimated to be low by only 2% at the highest energies and

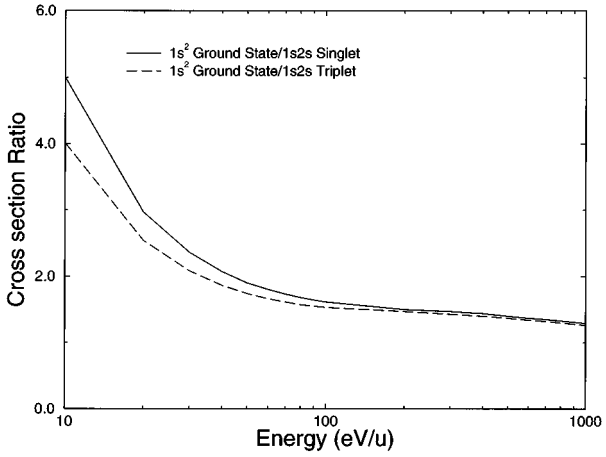

FIG. 3. Ratios of the cross section for capture by a ground state $C^{4+}(1s^2)$ core and an ion with an excited core, $C^{4+}(1s2s)^1S$ or $C^{4+}(1s2s)^3S$. The cross sections were calculated using a multichannel Landau-Zener calculation (see the text).

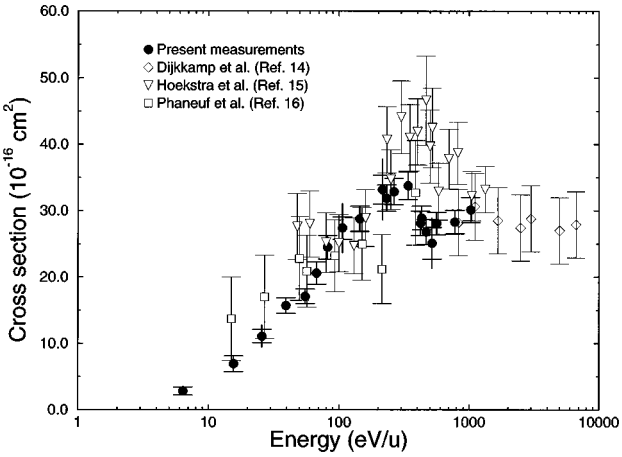

FIG. 4. Present total-electron-capture measurements (filled circles) are compared with other experimental measurements. The relative error bars are plotted at a 90% confidence level and are denoted by error bars with caps. The total uncertainties are denoted at a few energies by vertical error bars that extend beyond the caps.

4.5% at the lowest energies. These small corrections were applied to the data.

III. RESULTS AND DISCUSSION

The experimental results for the absolute total electron capture cross sections for C^{4+} + H(D) are given in Table II. The results include measurements with both hydrogen and deuterium. While there can be a difference in the cross section for collisions with H and D due to the kinematic isotope effect [18,33], no significant difference between H and D is expected at these energies [34]. Deuterium was used to provide a maximum angular collection at the lower energies, while hydrogen provides access to the higher collision energies. The total uncertainties correspond to the quadrature sum of the statistical and absolute uncertainties (12%) estimated at a 90% confidence level. The experimental results are compared with other measurements (Fig. 4) and theory (Fig. 5). The relative error bars are plotted at a 90% confidence level and are denoted by error bars with caps. For a few collision energies (26, 107, 216, and 517 eV/u), the total error bars are plotted at a 90% confidence level and are denoted by vertical error bars that extend beyond the caps. The

TABLE II. Total-electron-capture cross sections for C^{4+}+H(D).

Neutral	Collision energy (eV/u)	Cross section (10^{-16} cm^2)	Relative uncertainty (10^{-16} cm^2)	Total uncertainty (10^{-16} cm^2)
D	6.38	2.9	0.5	0.6
D	15.6	7.0	1.2	1.4
D	25.8	11.1	1.0	1.7
D	39.2	15.7	1.2	2.2
D	55.1	17.1	1.1	2.3
D	67.5	20.6	1.7	3.0
D	82.1	24.5	1.8	3.4
D	107	27.4	1.7	3.7
D	144	28.8	1.9	3.9
D	216	33.2	2.2	4.6
H	232	31.9	2.0	4.3
D	265	32.9	2.0	4.4
D	340	33.8	2.1	4.6
H	422	28.1	1.8	3.8
D	431	28.9	1.8	3.9
H	466	26.8	1.9	3.7
D	517	25.1	2.4	3.8
H	562	28.1	1.7	3.8
H	776	28.3	1.8	3.8
H	1013	30.1	1.9	4.0

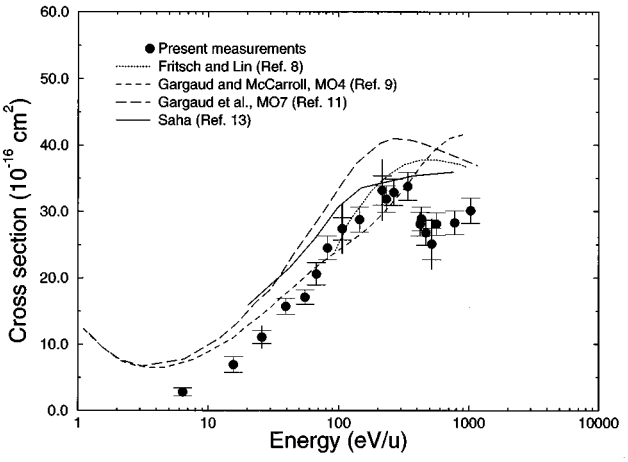

FIG. 5. Present total-electron-capture measurements (filled circles) are compared with various theories. Error bars are the same as in Fig. 4.

data are corrected a few percent for the presence of metastable states in the ion beam as discussed above.

Comparing the experimental results with previous measurements, one can see that at the highest energies around 1 keV/u our measurements show excellent agreement with the total-capture measurements of Dijkkamp *et al.* [14] At intermediate energies good agreement is found with the measurements of Phaneuf *et al.* [16] and Hoekstra *et al.* [15]. In the energy range 400–1000 eV/u a dip in the total-charge-transfer cross section is observed that is unresolved in the total-electron-capture measurements of Hoekstra *et al.* Since these total cross sections are obtained by summing over the cross section for individual l-subshells, the structure in the total cross section is probably washed out. The state-selective measurements do, however, support the position of the structure: It is in this energy range that the cross section for capture to the dominant $3p$ level is sharply decreasing, while capture to the $3s$ is increasing. By 1-keV/u capture to the $3s$ level becomes comparable to capture to $3p$. Capture to the $3d$ level constitutes about 25% of the total cross section but is relatively constant in the vicinity of the structure, as verified by the state-selective measurements of Hoekstra *et al.* within experimental uncertainty.

Comparison with theory

In Fig. 5 a comparison of the experimental results with theory is depicted. It is interesting to compare the two calculations of Gargaud and McCarroll (MO4) [9] and Gargaud, McCarroll, and Valiron (MO7) (MO7) [11]. While the MO4 calculation includes only the Σ states neglecting rotational coupling and electron translation factors, it seems to agree best with the present measurements below 300 eV/u. However, as previously argued by Hanssen *et al.* [7], agreement with the MO4 calculation is most likely fortuitous. Above 300 eV/u, the calculation shows that the cross section continues to increase. Using a more accurate model potential, ETF, and including an expanded basis set with rotational coupling to Π states, the MO7 calculation should be more accurate. It shows good agreement with the close-coupling semiclassical calculations (not shown) of Olson, Shipsey, and Browne [6] and Hanssen *et al.* [7], which extend down to 100 eV/u. A comparison with the present measurements, though, shows that the MO7 calculation overestimates the cross section by 25%.

The most recent calculation is by Saha [13] using a semiclassical impact-parameter, coupled-state method using all $n=3$, Σ and Π states. At the lowest energies this calculation agrees with the MO7 calculation. Compared to our data, though, it would seem that both tend to overestimate the cross section. At these energies the state-selective measurements of Hoekstra *et al.* suggest that the calculations overestimate the cross section for capture into the $3d$ state.

At the maximum, the predicted cross section by Saha is in good agreement with the measured data, but 15% lower than the MO7 calculation. The difference between the two calculations was argued [13] to be due to a difference in the coupling matrix elements and core potentials. Fritsch and Lin [8] used a 16-state two-center atomic orbital expansion to investigate this collision system. Over a limited energy range the calculation is in good agreement with our data and previous measurements. However, the observed sharp drop in the cross section is not seen in any of the calculations that overestimate the cross section by approximately 40% around 500 eV/u. It is unlikely that the structure in the cross section comes from the so-called Stuckelberg oscillations caused by destructive interference of different channels leading to capture since these kinds of processes are, in principle, included in the calculation of Fritsch and Lin (the model of Fritsch and Lin includes all states up to the $n=5$ level).

It is interesting to compare these measurements with the recent ORNL merged-beam measurements [19] for N^{4+}. As in the present measurements, a sharp structure is observed at the peak in the total cross section (for N^{4+}, around 100 eV/u). Semiclassical molecular-orbital calculations by Shimakura *et al.* [35] predict a similar structure, but at a slightly different energy than that observed [19]. Because many channels are coupled in this energy domain, the oscillatory structure is sensitive to the details of the molecular potentials and couplings included. Recent fully quantal molecular-orbital calculations by Zygelman *et al.* [36] also show oscillations near the peak in the N^{4+} cross section.

IV. CONCLUSION

Total-electron-capture cross sections for C^{4+} + H(D) have been measured in the energy range between 6 and 1000 eV/u by using the merged-beam technique. To estimate the contribution due to the presence of metastable states in the ion beam, the metastable fraction is measured using electron-impact ionization below the ground-state threshold and the effect on the total-charge-transfer cross section is estimated by using a multichannel Landau-Zener calculation. It is estimated that the metastable fraction of 5% results in a correction of only a few percent. Within the absolute uncertainties, the measurements generally agree well with previous experimental investigations. The measurements also compare favorably with theory, although the MO7 calculation of Gargaud, McCarroll, and Valiron tends to overestimate the cross section. The measurements agree best with the calculation by Saha, probably due to the more accurate potentials and cou-

plings used. The most striking observation is the sharp structure in the cross section around the maximum, which is not predicted by any of the available models for the total cross section. Such a structure is supported by the energy dependences for the $3s$, $3p$, and $3d$ state-selective cross sections observed by Hoekstra *et al.* [15]; however, the total cross section calculated from the sum does not have sufficient accuracy to resolve the structure. A similar structure was seen in N^{4+} total-electron-capture measurements, but the exact mechanism that is responsible is unclear.

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