PHYSICAL REVIEW A VOLUME 21, NUMBER 5 MAY 1980

# Electron-capture collisions at keV energies of boron and other multiply charged ions with atoms and molecules. II. Atomic hydrogen

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(Received 29 November 1979)

Single electron-capture cross sections have been measured for boron ions $B^{q+}$ with initial charges $q = 2$, 3, and 4 incident on an atomic hydrogen target. The cross sections $\sigma_{21}$ and $\sigma_{43}$ are large and show little dependence on the ion energy for the range studied, $6q$ to $23q$ keV. The cross section $\sigma_{32}$ increases with increasing energy in this same range. Also reported is a survey of single electron-capture cross sections for ions of carbon ($2 \leq q \leq 4$), nitrogen ($2 \leq q \leq 5$), and oxygen ($2 \leq q \leq 5$) at the single energies $8q$ keV also incident on atomic hydrogen.

## I. INTRODUCTION

In a previous paper[1] (herein denoted I) we reported measured values for electron-capture cross sections for keV energy, multiply charged ions of boron, carbon, nitrogen, and oxygen incident on helium and argon, and, in some cases, on molecular hydrogen. In this paper we report results for these ions incident on atomic hydrogen. Preliminary cross-section values based on a small part of this work were first presented by the present authors in 1976.[2]

Electron-capture studies using thermally dissociated hydrogen as a target gas began in 1960 with the work of Fite, Stebbings, Hummer, and Brackman,[3] who used protons as incident ions. This work was subsequently extended over a wide range of proton energies by McClure,[4] Bayfield,[5] and by Wittkower, Ryding, and Gilbody.[6] The first work with a multiply charged incident ion, $He^{++}$, was done by Fite, Smith, and Stebbings,[7] and was followed by the later studies of Shah and Gilbody,[8] and of Bayfield and Khayrallah.[9]

In the past several years there has been increased interest in the measurement of electron-capture cross sections for multiply charged ions incident on atomic hydrogen. This has been spurred primarily by the controlled nuclear fusion effort and by the realization that electron capture can be both an important energy-loss mechanism in tokamak reactors[10,11] as well as a potential particle loss mechanism for the neutral beam heating scheme for magnetically confined plasmas.[12,13] Thus, since our first work in 1976,[2] experiments have been carried out by Phaneuf, Meyer, and McKnight[14] for ions of carbon, nitrogen, and oxygen, by Gardner *et. al.*[15] for ions of iron, and by Kim *et al.* for ions of tantalum, tungsten, and gold.[16] These three works used "fast" ions, i.e., those with relative velocities larger than or comparable to $v_0$, the characteristic velocity of the electron in the hydrogen ground state. Experimental results have been reported by Crandall *et al.*[17] using "slow" ions of boron, carbon, nitrogen, and oxygen. Results have recently been published by Nutt, *et al.*[18] for slow $C^{2+}$ and $Ti^{2+}$ ions. New studies carried out by Shah and Gilbody[19] and by Olson *et al.*[20] are evidence for renewed interest in the $He^{++} + H$ system.

Published theoretical treatments of slow-electron-capture collisions are few in number; they have generally been limited to systems in which only a few final electronic states need to be considered. The work has, therefore, been limited primarily to one-electron systems such as $H^{+} + H$, $C^{6+} + H$, etc. The calculations performed by Olson, Shipsey, and Browne for $C^{4+}$ and $B^{3+}$ incident on H, and by Harel and Salin[21] for $C^{4+}$, however, are directly applicable to the present work and will be discussed further with the presentation of the data.

## II. EXPERIMENTAL METHOD

Cross sections were measured using a standard beam-gas target method. Ions with known charge-to-mass ratio $q/m$ and kinetic energy were extracted from the Oak Ridge Penning Ion Source Test Stand (ORNL-PIG).[22,23] After leaving the source, these ions were magnetically reanalyzed in order to eliminate unwanted lower charge states, $q-1$, $q-2$, etc., produced by electron-capture collisions in the background gas. The purified beam was then highly collimated and passed through the atomic hydrogen target gas cell. Product ions were selected by charge state $q'$ with a large parallel-plate electrostatic analyzer and were counted individually with use of a Johnston MMI particle multiplier and its accompanying electronics. Details of the experimental apparatus, gas handling system, and electronics may be found in paper I.

The atomic hydrogen was produced in an indirectly heated tungsten gas cell by thermal dissociation of molecular hydrogen at temperatures near 2500 °K. Details of the vacuum furnace construction are given in Ref. 24. The heating was obtained by passing a large (~600 A) pulsed dc current through a thin wall tantalum cylinder mounted coaxially around the tungsten gas cell. The heating current was pulsed at a frequency of 10 Hz and had a 67% "on" duty factor. The large mass of the tungsten gas cell prevented appreciable variation of its temperature during the "off" part of the heater cycle. To eliminate any spurious ion-beam steering effects associated with the magnetic fields caused by the heater current, beam ions were gated through the target only during the off part of the heater cycle.

The following procedure was used to take data. First, with the cell at operating temperature, a given beam of ions was transmitted through the apparatus. Then argon gas was admitted to the gas cell through a Varian leak valve from a reservoir maintained at a pressure near 1000 Torr. To make measurements of the intensities of the initial-charge-state component $q$, the single-electron-capture component $q-1$, and the double-electron-capture component $q-2$, the parallel-plate analyzer voltage was stepped electronically among the three transmission peaks while synchronously routing pulses from the particle multiplier to a multichannel scaler. (Details of the electronic system required to perform this stepping can be found in I.) To evaluate the fraction of the scattering signal produced by background gas, the incident and product ion intensities were remeasured with the target gas flow bypassing the scattering cell and flowing directly into the surrounding vacuum chamber. Next, without changing the leak valve, the reservoir was evacuated to below $10^{-3}$ Torr and refilled with molecular hydrogen to the same absolute pressure as before. Following the procedure outlined above, the intensities of the charge state components $q$, $q-1$, and $q-2$ were measured both for molecular hydrogen entering the gas cell and for it bypassed into the vacuum system. This complete cycle was repeated several times.

The scattering signals for hydrogen and argon were combined to form the following ratios:

$$R_1^Y \equiv \frac{S^Y_{q,q-1}(\mathrm{H},\mathrm{H}_2)}{S^Y_{q,q-1}(\mathrm{Ar})} \quad \text{and} \quad R_2^Y \equiv \frac{S^Y_{q,q-2}(\mathrm{H},\mathrm{H}_2)}{S^Y_{q,q-1}(\mathrm{Ar})}\,, \tag{1}$$

where $S^Y_{q,q-\Delta q}(X)$ is the signal (gas-bypass corrected) for $Y^{q+} + X \rightarrow Y^{(q-\Delta q)+} + X^{\Delta q+}$ collisions. Since the final state of the target species $X$ was not analyzed after the collison, $X^{\Delta q+}$ refers to all possible configurations of internal degrees of freedom, including the continuum. The signals $S_{q,q-\Delta q}$ for hydrogen are written with both H and $\mathrm{H}_2$ in parentheses because the gas call contained a small amount of undissociated $\mathrm{H}_2$. Under single-collision conditions the signal $S^Y_{q,q-2}(\mathrm{H},\mathrm{H}_2)$ can be produced only by residual $\mathrm{H}_2$, and, if the cross section $\sigma^Y_{q,q-2}(\mathrm{H}_2)$ is known, it is a measure of the amount of $\mathrm{H}_2$ present. Assuming molecular flow conditions, conservation of mass flow, and equal temperatures for Ar, H, and $\mathrm{H}_2$ target particles, one can easily show[25] that

$$\frac{\sigma^Y_{q,q-1}(\mathrm{H})}{\sigma^Y_{q,q-1}(\mathrm{Ar})} = \frac{1}{\sqrt{2}}\left(\frac{R_1^Y - F\sigma^Y_{q,q-1}(\mathrm{H}_2)/\sigma^Y_{q,q-1}(\mathrm{Ar})}{1-F}\right), \tag{2}$$

where $F$ is given by

$$F = R_2^Y \sigma^Y_{q,q-1}(\mathrm{Ar})/\sigma^Y_{q,q-2}(\mathrm{H}_2). \tag{3}$$

Since $F$, the fraction of undissociated $\mathrm{H}_2$, is independent of incident ion specie and charge state, $F$ is written without subscripts and superscripts. One is free, therefore, to choose the ion species and charge state that allow the best measurement of $F$. For the present experiments, $F$ was measured for every incident ion with initial charge larger than two. Having relatively large cross sections for double-electron capture from $\mathrm{H}_2$, $\mathrm{B}^{3+}$, and $\mathrm{C}^{3+}$ ions produced the most accurate values for $F$, which was measured four times at approximately equal intervals during the accumulations of the data. Values of 0.089 ± 0.048, 0.147 ± 0.084, 0.140 ± 0.029, and 0.153 ± 0.069 were obtained sequentially, with $C^{3+}$ used for the second one and $B^{3+}$ used for the other three. Since these values are consistent with each other, their error-weighted average value $\bar{F}=0.13 \pm 0.02$ was used to correct all the final atomic hydrogen cross sections for residual undissociated $H_2$. Though consistent with this value, values of $F$ obtained using incident ions other than $B^{3+}$ and $C^{3+}$ were of such poor statistical quality that they were not used in the computation of cross sections.

In order to use Eq. (2) to calculate $\sigma^{Y}_{q,q-1}(\mathrm{H})$, it is clear that a knowledge of the cross sections $\sigma^{Y}_{q,q-1}(\mathrm{Ar})$ and $\sigma^{Y}_{q,q-1}(\mathrm{H}_2)$ is required. The cross sections $\sigma^{Y}_{q,q-1}(\mathrm{Ar})$ required for normalization of the present data were reported in paper I. Additional required cross section ratios $\sigma^{Y}_{q,q-2}(\mathrm{H}_2)/\sigma^{Y}_{q,q-1}(\mathrm{Ar})$ were measured using the techniques described in paper I.

If the assumption of equal temperatures for the Ar, H, and $H_2$ particles that was used in the derivation of Eq. (2) is not made, then the result

$$\frac{\sigma_{q,q-1}(\mathrm{H})}{\sigma_{q,q-1}(\mathrm{Ar})}=\frac{1}{\sqrt{2}}\left(\frac{T(\mathrm{Ar})}{T(\mathrm{H})}\right)^{1/2} R_1 \tag{4}$$

is obtained, where 100% $H_2$ dissociation has been assumed for convenience. Clearly the cross sections reported here will be too large if $T_{\mathrm{H}} < T_{\mathrm{Ar}}$. We now discuss this possibility. Atoms at temperature $T_i$ striking a surface at temperature $T_w$ rebound with a temperature $T$ given, on the average, by[34]

$$T=T_i+\alpha(T_w-T_i), \tag{5}$$

where $\alpha$, the dimensionless quantity known as the thermal accommodation coefficient, depends on the type and temperature $T_w$ of the surface material and on the type of atoms striking the surface. It is easily shown that after $N$ bounces, an atom will have a temperature $T_N$ given by

$$T_N=(1-\alpha)^N T_i+[1-(1-\alpha)^N]T_w\,. \tag{6}$$

For Ar on a clean tungsten surface at 2335 °$K$, $\alpha$ has been measured[26] to be about 0.33. The gas cell used in our experiments was made entirely of tungsten and was of a double-wall design. A typical atom had to strike the cell walls an estimated 200 times before entering the region where it could interact with beam ions. Using $\alpha=0.33$ and $N=200$ in Eq. (6) leads to a value of $T_N$ quite equal to $T_w$. One predicts, therefore, that the argon atoms were probably "thermally accommodated", that is, nearly in thermal equilibrium with the ~2500 °K gas cell. Direct evidence for *equal* accommodation in the present gas cell of Ar and $H_2$ at both 300 °K and about 1900 °K can be found in Fig. 30 of Ref. 27. For temperatures above 1900 °K, the $H_2$ begins to dissociate. The thermal accommodation coefficient for H has not been measured. However, if one assumes only 100 bounces, then $\alpha$ for H on tungsten must be less than 0.015 in order to introduce a 10% or larger error in the reported cross sections. Such a low value for $\alpha$ is quite unexpected but cannot be completely ruled out at present.

The influence of leaks in the gas feed lines and in the scattering cell itself are correctly discussed in Ref. 33. If leaks existed in the relatively cold gas feed lines, then the analysis shows that the cross sections reported here would be at most 40% too *large*. This can only occur if there were large leaks at cold places near the scattering cell itself. Such a leakage conductance is effectively cold if hydrogen does not dissociate at its temperature. As time went on, one significant crack did develop in the outer tungsten wall of one of our two scattering cells. The crack was in a region operating close to the full target temperature; the reader is referred to the target design details and discussion of temperature profiles in Ref. 24. The scattering cells used by other investigators are single walled, rather than double walled. The single-walled cells are found to dissociate $H_2$ adequately, providing strong evidence that the leakage conductance due to our cell crack was very likely hot, resulting in no significant error. In addition, a last portion of our data was taken with a different cell having no significant cracks, the original cell of Ref. 24. The results reproduced our earlier $B^{3+}$, $B^{4+}$, and $He^{++}$ results to within ±15%.

The measured cross sections for $B^{2+}$, $B^{3+}$, and $B^{4+}$ ions incident on atomic hydrogen are plotted vs incident ion energy in Fig. 1. The data for $\sigma^{\mathrm{B}}_{43}$ and $\sigma^{\mathrm{B}}_{21}$ show little dependence on the energy for the range studied. The data for $\sigma^{\mathrm{B}}_{32}$, however, show a rise with energy. Note that the cross sections do not increase monotonically with the in-

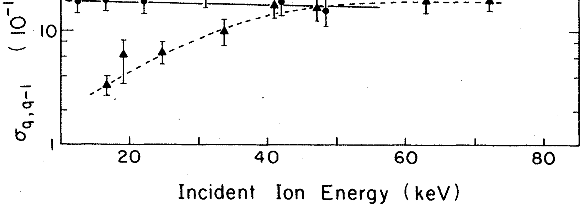

FIG. 1. Electron-capture cross sections for boron ions incident on H. The squares are the present data for $B^{4+}$, the triangles for $B^{3+}$, and the circles for $B^{2+}$.

cident ion charge: $\sigma_{32}^{B} < \sigma_{21}^{B}$ for energies at least as high as about 40 keV.

Energy defects for capture of the H (1*s*) electron into certain final states of the boron ions are listed in Table I. Also included are the approximate crossing distances at which molecular potential-energy curves are computed to cross using the methods of Bates and Moiseiwitsch[28] (see I). Polarizabilities of the final-state ions have been neglected in the calculation of the crossing distances. Since the polarizability of ions increases rapidly with the degree of excitation,[32] the crossing distances may be in error by 2 to 3 a. u. for final states with large principal quantum numbers.

TABLE I. Energy defects $\Delta E$ in eV and initial-state–final-state crossing distances $R_c$ in atomic units for capture into various states in the collision system $B^{q+} + H$. Crossing distances were estimated using the methods of Bates and Moiseiwitsch [a] and are tabulated in units of Bohr radii. Energy defects were calculated using the data of Moore [b] for $B^{2+}$ and $B^{3+}$ and of Eidelsburgh [c] for $B^{4+}$. The units are electron volts.

| | $B^{2+}(1s^22s\,^2S)$ incident | |
|---|---|---|
| Final ion | $\Delta E$ | $R_c$ |
| $B^{+}(1s^22s^2\,^1S)$ | 11.6 | 3.1 |
| $B^{+}(1s^22s2p\,^3P)$ | 6.9 | 4.4 |
| $B^{+}(1s^22s2p\,^1P)$ | 2.5 | 11.2 |
| $B^{+}(1s2s3s\,^3S)$ | −4.5 | |
| $B^{+}(1s2s3s\,^1S)$ | −5.3 | |

| | $B^{3+}(1s\,^1S)$ incident | |
|---|---|---|
| Final ion | $\Delta E$ | $R_c$ |
| $B^{2+}(1s^22s\,^2S)$ | 24.3 | 3.1 |
| $B^{2+}(1s^22p\,^2P)$ | 18.3 | 3.6 |
| $B^{2+}(1s^23s\,^2S)$ | 2.0 | 28.0 |
| $B^{2+}(1s^23p\,^2P)$ | 0.4 | 140.0 |
| $B^{2+}(1s^23d\,^2D)$ | 0.02 | |
| $B^{2+}(1s^24s\,^2S)$ | −5.10 | |

| | $B^{4+}(1s\,^2S)$ incident | |
|---|---|---|
| Final ion | $\Delta E$ | $R_c$ |
| $B^{3+}(1s^2\,^1S)$ | 245.8 | 1.5 |
| $B^{3+}(1s2s\,^3S)$ | 47.2 | 2.8 |
| $B^{3+}(1s2s\,^1S)$ | 43.0 | 2.9 |
| $B^{3+}(1s2p\,^3P)$ | 42.9 | 2.9 |
| $B^{3+}(1s2p\,^1P)$ | 40.2 | 3.0 |
| $B^{3+}(1s3s\,^3S)$ | 12.4 | 6.9 |
| $B^{3+}(1s3s\,^1S)$ | 11.2 | 7.5 |
| $B^{3+}(1s3p\,^3P)$ | 11.2 | 7.5 |
| $B^{3+}(1s3d\,^3D)$ | 10.6 | 7.9 |
| $B^{3+}(1s3d\,^1D)$ | 10.6 | 7.9 |
| $B^{3+}(1s3p\,^1P)$ | 10.4 | 8.0 |
| $B^{3+}(1s4s\,^3S)$ | 0.7 | 110.0 |
| $B^{3+}(1s4s\,^1S)$ | 0.3 | |
| $B^{3+}(1s4p\,^3P)$ | 0.3 | |
| $B^{3+}(1s4d\,^3D)$ | 0.02 | |
| $B^{3+}(1s4d\,^1D)$ | 0.01 | |
| $B^{3+}(1s4f\,^3F)$ | 0.01 | |
| $B^{3+}(1s4f\,^1F)$ | 0.01 | |
| $B^{3+}(1s4p\,^1P)$ | −0.05 | |
| $B^{3+}(1s5s\,^3S)$ | −4.5 | |

[a] Reference 28.

[b] Reference 29.

[c] Reference 30.

Table I shows that in the $B^{4+} + H$ case, there are numerous crossings near 7 to 8 a. u. for capture into the $n = 3$ states of the final $B^{3+}$ ion. An absorbing sphere approximation[31] in the many curve crossing limit leads to an estimate for $\sigma_{43}^{B}(H)$ of about $\pi(7.5a_0)^2 \sim 50 \times 10^{-16}$ cm$^2$, in remarkably good agreement with the measured value.

In the $B^{2+} + H$ system there are three reasonably well-separated initial-state–final-state crossings. For the present range of ion–atom velocities, one expects that the two inner crossings at $3.1a_0$ and $4.4a_0$ are most important. The large magnitude of the cross section $\sigma_{21}^{B}$, however, may imply that the outer crossing at $11a_0$ is also important.

The $B^{3+} + H$ collision system has been treated theoretically by Olson, Shipsey, and Browne[21] in a molecular-orbital, close-coupling approximation. Their results, along with the recent experimental results of Crandall *et al.*,[17] are compared with the present data in Fig. 2. A reasonable degree of agreement between the two data sets and the theory is evident.

Table I shows that there are only two final states, $B^{2+}(1s^22s\,^2S)$ and $B^{2+}(1s^22p\,^2P)$, which cross the initial state at "reasonable" internuclear separations. Since the $n = 3$ states of $B^{2+}$ cross at very large separations, one suspects that these states are probably relatively unimportant for the present velocity range. However, Olson *et al.*[31] report that the magnitude of the cross section $\sigma_{32}$ at the higher energies studied here is due primarily to a noncrossing interaction between the initial state and these $n = 3$ states. Thus one may conclude that the simple curve crossing picture is inadequate in some cases.

Table II lists the cross sections for ions of

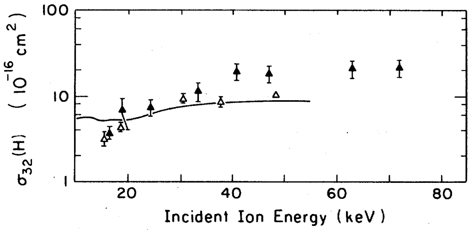

FIG. 2. Comparison of values for the cross section for $B^{3+}$ incident on H. The closed triangles are the present data, and open triangles are the data of Crandall *et al.* (Ref. 17). The solid curve is the theoretical prediction of Olson *et al.* (Ref. 21).

TABLE II. Electron-capture cross sections for various multicharged ions incident on H.

| | Present data | | Crandall, Phaneuf, and Meyer | |
|---|---|---|---|---|
| Incident ion | Energy (keV) | Cross section ($10^{-16}$ cm$^2$) | Energy | Cross section |
| $B^{2+}$ | 16 | 19.2 ± 5.0 | 13 | 14.2 ± 1.2 |
| $B^{3+}$ | 24 | 7.2 ± 2.5 | 24 [a] | 6.8 ± 0.6 |
| $B^{4+}$ | 32 | 38 ± 11 | 25 | 26.5 ± 3.9 |
| $C^{2+}$ | 16 | 6.5 ± 2.6 | | |
| $C^{3+}$ | 24 | 17.7 ± 6.1 | 33 | 16.3 ± 1.8 |
| $C^{4+}$ | 32 | 50 ± 12 | 33 | 29.3 ± 2.6 |
| $N^{2+}$ | 16 | 4.2 ± 1.8 | | |
| $N^{3+}$ | 24 | 36 ± 13 | 24 | 19.3 ± 0.8 |
| $N^{4+}$ | 32 | 46 ± 18 | 30 | 29.2 ± 0.6 |
| $N^{5+}$ | 40 | 56 ± 20 | 38 | 26.7 ± 1.2 |
| $O^{2+}$ | 16 | 2.2 ± 1.0 | | |
| $O^{3+}$ | 24 | 61 ± 21 | | |
| $O^{4+}$ | 32 | 50 ± 17 | | |
| $O^{5+}$ | 40 | 65 ± 24 | 44 | 34.8 ± 2.0 |

[a] This value is an interpolation between measured points.

boron, carbon, nitrogen, and oxygen incident on atomic hydrogen. Notice that for fixed incident ion charge the cross section increases with atomic number $Z$ for $q = 2$ but decreases with atomic number for $q \geq 4$. With use of a curve-crossing model and computed energy defects,[25] this behavior can be understood qualitatively. In particular, assume that in the case of the incident $X^{2+}$ ions the important crossings are those with the $n = 2$ states of the corresponding $X^+$ ions. In most cases capture is most favorable for $5 \lesssim R_c \lesssim 15$ a.u.; for larger $R_c$ coupling matrix elements are too small. Thus one finds that in going from boron to oxygen these crossings move to smaller values of $R_c$ because the energy defects increase in this same order. Thus, the cross sections $\sigma_{21}$ should decrease in this same order, as is observed. Similarly, if one assumes that for the $X^{3+}$ ions the $n = 3$ states of the final $X^{2+}$ ions are the important ones, then the crossings which occur at quite large separations for boron move to progressively smaller separations while going from boron to oxygen. Thus, the cross sections $\sigma_{32}$ should increase in this same order. Again this is observed. For the incident $X^{4+}$ ions, however, the $n = 3$ states of the corresponding $X^{3+}$ ions have energy defects that are approximately equal, independent of atomic species. Since the crossings with these states will occur at approximately equal internuclear separations, independent of atomic species, one expects that the cross sections $\sigma_{43}$ will be approximately independent of atomic species, as is observed experimentally.

Also listed in Table II are values for the cross sections measured by Crandall, Phaneuf, and Meyer.[17] There is considerable disagreement between the two data sets, the present data being on average 50% higher than those of Crandall *et al.* The reason for this discrepancy remains unknown. Such disagreement was not found for the cold target gases studied in paper I. The atomic hydrogen targets used in the two experiments both produce atomic hydrogen by thermal dissociation of molecular hydrogen, but similarities generally end at this point. The target of Crandall *et al.*, based on a design of McClure,[4] uses a resistively heated tungsten tube with $H_2$ admitted directly into the scattering region. The indirectly heated, double-walled target used in the present experiments is thoroughly described in Ref. 24.

If the targets are different, so are the data normalization procedures. Crandall *et al.*[17] normalize directly to the cross section $\sigma_{10}$ for protons on atomic hydrogen. The present data for a given ion is normalized to the cross section for that ion incident on argon. Our normalization procedures were established as accurate to ± 20% in paper I. Although the present method of normalization for atomic hydrogen studies is a little more complicated, we can find no systematic effects in the procedure that can account for the differences in the two data sets.

## III. SUMMARY

Cross sections for electron capture by multiply charged boron ions incident on atomic hydrogen have been measured as a function of energy. The cross section $\sigma_{32}$ is in reasonably agreement with the close-coupling calculation of Olson *et al.*[21]

Electron-capture cross sections were also measured for $C^{q+}$ $(2 \leq q \leq 4)$, $N^{q+}(2 \leq q \leq 5)$, and $O^{q+}(2 \leq q \leq 6)$ incident on atomic hydrogen. These data were taken at collision energies of $8q$ keV.

The trends of these cross sections with atomic number for a fixed incident charge can be understood qualitatively using a simple curve-crossing model.

## ACKNOWLEDGMENTS

This work was primarily supported by the Chemical Sciences Section, Division of Physical Research, U. S. Department of Energy. The Oak Ridge National Laboratory is operated by Union Carbide Corp., also for the U. S. Department of Energy. University of Tennessee participation was partially supported by the U. S. National Science Foundation. The authors acknowledge the skillful assistance of J. Hale and useful conversations with M. Mallory.

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