PHYSICAL REVIEW A VOLUME 28, NUMBER 3 SEPTEMBER 1983

Electron capture by slow Fe^{q+} ions from hydrogen atoms and molecules

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(Received 12 April 1983)

Total electron-capture cross sections have been measured for collisions of Fe^{q+} ions ($3 \leq q \leq 14$) with H and H_2 at energies in the 10—95 eV/amu range. The cross sections increase approximately linearly with increasing ionic charge q, and, for atomic hydrogen, can be represented by the empirical formula: $\sigma = q \times 10^{-15}$ cm^2 with an rms deviation of $\pm 26\%$. For Fe^{5+} and Fe^{6+}, for which higher-energy data exist, the cross sections are found to be essentially energy independent over the range $17 < E < 2000$ eV/amu. The cross sections for an atomic H target average 26% larger than for H_2. Agreement with generalized theoretical models is within a factor of 2 or better.

I. INTRODUCTION

A better understanding of the behavior of impurities in magnetically confined plasmas is recognized as a prerequisite to the design and operation of a successful fusion reactor.[1] Electron capture in low-energy (< 100 eV/amu) collisions of multiply ionized plasma impurities with hydrogen atoms plays a particularly important role in the cooler edge-plasma region and in the operation of magnetic divertors for impurity control.[2,3] Accurate theoretical calculations of electron-capture cross sections for heavy multiply charged ions at such low energies require a quasimolecular model,[4,5] or even fully quantal calculations,[6] and require detailed potential-energy curves for the specific interacting system. Electron capture is represented by transitions between the appropriate energy levels of the quasimolecular ion. Collision systems containing more than a few bound electrons are characterized by several transitions or "curve crossings" which are favorable for capture and hence a number of possible product states. As a result, the total capture cross sections for such systems are expected to be relatively large ($> 10^{-15}$ cm^2), to increase more or less uniformly with ionic charge, and to be rather insensitive to the velocity of the collision.

Another more generalized class of theory seeks to establish for such systems approximate scaling laws in which the cross sections are parametrized by the ionic charge, collision velocity, and target ionization potential. An example is the absorbing-sphere model of Olson and Salop[7] in which the multiple curve crossings are treated in a modified Landau-Zener approximation. In the low-energy theory of Grozdanov and Janev,[8] the electron-capture process is treated as a tunneling effect caused by the strong attractive Coulomb field of the multicharged ion. Both models predict an approximately linear increase of the total capture cross sections with ionic charge q and a gradual rise with decreasing collision velocity. Ryufuku *et al.*[9] have employed a unitarized distorted-wave approximation based on hydrogenic atomic orbitals and also a simple classical model in which the electron to be captured is required to have sufficient energy to overcome the Coulomb potential barrier between the ion and target nucleus. For partially stripped ions, both of their models use an effective ionic charge based on spectroscopic information to account for the fact that the level structure of the product ion is not hydrogenic, and both predict a rather strong oscillatory dependence of the capture cross section with ionic charge in the 10^7 cm/s velocity range. At very low velocities corresponding to thermal energies, an orbiting or Langevin model[4] may be used to estimate electron-capture cross sections. In this model, the dynamics of the long-range interaction of a point charge with a charge-induced dipole is treated classically, and the assumption is made that all trajectories which surmount the centrifugal barrier lead to capture. One of the aims of current experiments with highly charged ions is to test the accuracy and establish the range of validity of such approximate methods in the energy ranges in which they are applicable.

A more comprehensive but somewhat generalized approach for highly charged, partially stripped projectiles is the "screened-potential" method[6] which treats such interactions in a one-electron approximation. The role of the core electrons on the partially stripped ion is to relax the molecular symmetries, permitting transitions to occur between levels of the quasimolecule which are not allowed in the pure one-electron case, and making possible coupling to united-atom potential curves. The capture cross sections are predicted to increase as the size of the ionic core increases, a result which is analogous in concept to the absorbing-sphere model.

In spite of considerable encouragement from the fusion research community, experimental data for electron capture from hydrogen atoms by typical heavy, multiply charged plasma-impurity ions have not heretofore been reported at energies below 1 keV/amu. This derives mainly from the practical difficulty of producing well-characterized beams of multiply charged ions at such low energies. In the present investigation, a pulsed-laser-produced plasma has been used as a source of a collimated beam of multiply charged iron ions which were directed through an atomic or molecular hydrogen-gas target cell. Total cross sections have been measured for electron capture in $Fe^{q+} + H$ and $Fe^{q+} + H_2$ collisions at energies in the range 10—95 eV/amu and for ionic charges q ranging from 3—14.

II. EXPERIMENTAL

The apparatus and time-of-flight technique used for electron-capture cross-section measurements have been described recently.[10,11] A 2J, 60-ns pulse of CO_2 laser radiation is focused in vacuum onto a ^{56}Fe target. A series of apertures collimate a beam from the expanding plasma which enters an electrostatic analyzer. A beam emerges from the analyzer with selected energy per charge, passes through a calibrated thermal-dissociation atomic hydrogen target, is once more charge analyzed by electrostatic deceleration, and detected by an electron multiplier. Charge separation is effected by time-of-flight analysis. Electron-capture cross sections are deduced by measuring the variation with target thickness of the net fraction of ions which capture an electron in the hydrogen target. The atomic and molecular hydrogen target thicknesses and dissociation fraction (0.87 ± 0.03) were determined in an auxiliary experiment using a probe beam of 20-keV protons.[12] A target of 99.9% pure ^{56}Fe was used because of complications in the time-of-flight spectra for natural Fe caused by the fractional abundances of the isotopes ^{54}Fe (5.8%), ^{57}Fe (2.2%), and ^{58}Fe (0.3%). Owing to their differences in mass, these components coincided temporally with the electron-capture signals for the higher q, where the charge-state resolution was diminished (i.e., $q-1\approx q$).

III. RESULTS AND DISCUSSION

The experimental cross sections for $Fe^{q+}+H$ and $Fe^{q+}+H_2$ collisions are collected in Table I along with the random and estimated total experimental uncertainties. Systematic uncertainties are essentially as outlined in Ref. 11, with the exception that those uncertainties associated with relative ion-detection efficiency were progressively lower for the higher q ions of this investigation, since the relative change in q decreases. For $Fe^{q+}+H$, the estimated absolute systematic uncertainty at good-confidence level varied from ±22% for $q=3$ to ±14% for $q=14$. The corresponding values for H_2 were ±20% and ±12%.

A. Atomic hydrogen results

The present results for $Fe^{5+}+H$ and $Fe^{6+}+H$ are compared in Fig. 1 with higher-energy data obtained by Crandall *et al.*[13] using ion beams from the Oak Ridge National

TABLE I. Experimental total electron-capture cross sections for $^{56}Fe^{q+}+H$ and $^{56}Fe^{q+}+H_2$ collisions.

q	Energy eV/amu	Velocity 10^6 cm/s	$\sigma_{q,q-1}(H)$	Random[a] uncertainty (10^{-16} cm^2)	Total[b] uncertainty	$\sigma_{q,q-1}(H_2)$	Random[a] uncertainty (10^{-16} cm^2)	Total[b] uncertainty
3	10.4	4.48	44	±3	±11			
3	20.4	6.27	44	3	11	28	±5	±10
4	13.8	5.16	25	2	6			
4	27.2	7.24	17	4	9	18	4	8
5	17.3	5.78	48	3	10			
5	34.0	8.10	48	3	10	52	5	13
6	20.8	6.33	57	4	13			
6	40.8	8.87	58	7	16	52	5	12
7	24.2	6.83	70	9	20			
7	47.6	9.58	81	4	16	56	6	14
8	27.7	7.31	90	14	28			
8	54.4	10.24	77	8	19	62	5	13
9	61.2	10.87	108	6	20	83	8	19
10	68.0	11.45	117	8	23	80	11	22
11	74.8	12.01	119	11	26	92	14	28
12	81.6	12.55	68	11	22	56	20	36
13	88.4	13.06	112	15	31	105	17	32
14	95.2	13.55	128	23	44			

[a] Reproducibility at 1 s.d.

[b] The quadrature sum of random uncertainties at 90% confidence level and total systematic uncertainties (including absolute uncertainty) at comparable level of confidence.

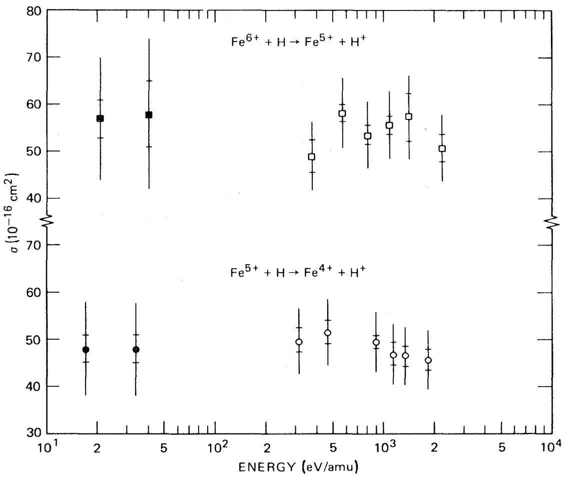

FIG. 1. Experimental total electron-capture cross sections vs collision energy for $Fe^{5+}+H$ (circles) and $Fe^{6+}+H$ (squares). Solid points are present results, and open points are data of Crandall *et al.* (Ref. 13). Inner flags designate statistical reproducibility (standard deviation) and outer flags represent estimated total experimental uncertainty at good-confidence level.

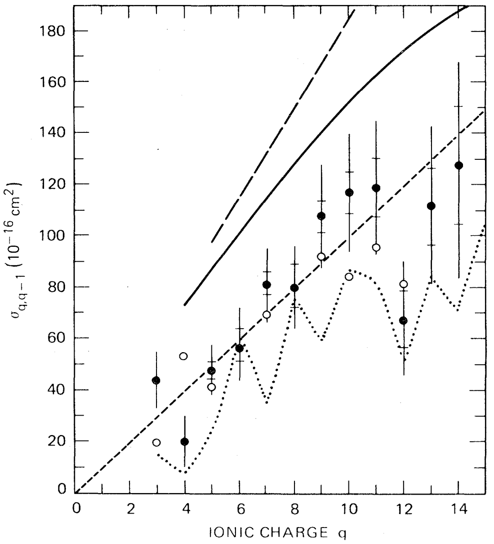

FIG. 2. Total electron-capture cross sections plotted vs ionic charge for $Fe^{q+}+H$ collisions (solid circles are present results) and for $Xe^{q+}+H$ (open circles, from Ref. 13). Flags are as defined for Fig. 1. For $Xe^{q+}+H$ data, $v=4\times10^7$ cm/s, and for $Fe^{q+}+H$ data, $v=(0.6\text{--}1.3)\times10^7$ cm/s. Solid curve is absorbing-sphere model of Olson and Salop (Ref. 7), and long-dashed curve is tunneling model of Grozdanov and Janev (Ref. 8). Both refer to a collision velocity of 10^7 cm/s. Dotted curve is over-barrier classical model of Ryufuku *et al.* (Ref. 9). Short-dashed curve represents the empirical relation: $\sigma=q\times10^{-15}$ cm^2.

Laboratory Penning ion gauge (ORNL-PIG) multiply charged ion source. In both cases, the measured cross sections at the higher and lower energies are of comparable magnitude and indicate the absence of a strong energy dependence. At these energies, as noted earlier, the insensitivity of the total cross sections to collision velocity is an expected result for multiply charged collision systems ($q\geq4$) containing many bound electrons. Such behavior has also been demonstrated experimentally in the 10^7 cm/s velocity range for $Xe^{q+}+H$ and $Ar^{q+}+H$ collisions,[13] and for noble-gas ions colliding with noble gases.[14–16] The present results contrast the behavior of few-electron systems of comparable ionic charge, such as $C^{6+}+H$ and $O^{5+}+H$, which are characterized by a single or, at most, a few favorable curve crossings and which exhibit strong velocity dependences at these low energies.[11]

The variation with charge q of the total capture cross sections for $Fe^{q+}+H$ collisions is shown in Fig. 2 along with the predictions of the absorbing-sphere model of Olson and Salop,[7] the tunneling model of Grozdanov and Janev,[8] and the classical model of Ryufuku *et al.*[9] The $Fe^{q+}+H$ data are not all taken at exactly the same collision velocity, but vary from 0.6×10^7 cm/s for Fe^{3+} to 1.3×10^7 cm/s for Fe^{14+}. These differences are considered to be insignificant, since the velocity dependence is expected to be weak in these regions, as demonstrated for Fe^{5+} and Fe^{6+}. Also plotted for comparison in Fig. 2 are the data of Crandall *et al.*[13] for $Xe^{q+}+H$ at $v=4\times10^7$ cm/s. The $Fe^{q+}+H$ cross sections increase with ionic charge in an approximately linear fashion with notable deviations at $q=4$ and 12. This general increase with ionic charge is in contrast to recent results for the few-electron multiply ionized systems, $C^{q+}+H$ and $O^{q+}+H$, where such behavior is not observed.[11] Both the absorbing-sphere and tunneling models predict a monotonic increase of the capture cross section with ionic charge in this velocity range, but overestimate present measurements on the average by 50–80%. The classical and distorted-wave (not shown) calculations underestimate the measurements by about the same amount, but predict a strong oscillatory dependence on ionic charge.

Not unexpectedly, the magnitudes of the Fe^{q+} cross sections are very comparable to those for Xe^{q+}, and perhaps coincidentally, both exhibit an unexpectedly small cross section for $q=12$. From the perspective of a curve-crossing model, such behavior is surprising since several crossings should exist which are favorable for capture. Unfortunately, the energies of excited levels of Fe^{11+} or Xe^{11+} which are probable final states for electron capture are not readily available in the literature, complicating even a qualitative analysis of the situation. The other apparent anomaly in the present data is the relatively small cross section for $Fe^{4+}+H$. It is interesting to note that the largest dips in the classical prediction of Ryufuku *et al.*[9] occur for $q=4$ and 12 (although there appears to be, in fact, an anticorrelation for the other predicted "oscillations"). For the present application of their model, effective charges for the various charge states of Fe^{q+} were deduced by best fitting a hydrogenic model to spectroscopic energy levels according to their prescription. Since only the low-lying levels are tabulated[17] for Fe^{q+} for $q>5$, the deduced q_{eff} were plotted versus q, and a linear least-squares fit was used.

Since the corresponding $Fe^{12+}+H_2$ cross section shows a similar anomaly, a search for systematic problems in the

Fe^{12+} case was made, with possible contamination of the Fe^{12+} beam by some impurity being most suspect. Potential impurities with the same $m/q = 4.67$ are N^{3+} and Si^{6+}. However, neither N nor Si impurity ions of other charges were identified in any of the time-of-flight spectra, and plots of the measured ion intensity from the laser source versus q varied smoothly with ionic charge, showing no anomalous increase at $q = 12$. Furthermore, no ions at this m/q were detected when the laser power was reduced by a factor of ~ 2 in order to eliminate the Fe ions of higher q. At this reduced laser power, N^{3+} or Si^{6+} ions would have been expected to be readily produced if either nitrogen or silicon were a contaminant in the target. It is thus judged unlikely that a contaminant in the ion beam is responsible for the anomalously small Fe^{12+} cross sections. The fact that the Fe^{13+} and Fe^{14+} cross sections are large suggests that undetected systematic problems at higher q are also unlikely. Of course, depending upon the atomic structure, the likely presence of metastable excited ions in the incident beam is always an important consideration when interpreting such experimental results. Given the potentially large number of final states favorable for capture in a system of ionic charge 12 containing 15 bound electrons, however, it seems rather unlikely that the capture cross section from a long-lived metastable-state would be significantly smaller than that from a ground-state ion.

The present $Fe^{q+} + H$ data can be represented by the empirical formula: $\sigma = q \times 10^{-15}$ cm^2 with an rms deviation of $\pm 26\%$. This is indicated by the short-dashed curve in Fig. 2.

B. Molecular hydrogen results

The variation of the capture cross sections with ionic charge for molecular hydrogen is qualitatively similar to that measured for atomic hydrogen and is shown in Fig. 3 along with the prediction of the absorbing-sphere model. In this case the model underestimates the experimental data. As noted earlier, the cross sections for $q = 4$ and 12 appear to be anomalously low for both H and H_2 targets. Experimental data for $Xe^{q+} + H_2$ collisions[13] are included in Fig. 3 for comparison. As in the atomic hydrogen case, the cross-section exhibits the same apparent anomaly as that for $Fe^{12+} + H_2$.

The measured cross sections for atomic hydrogen exceed those for molecular hydrogen in magnitude by a mean factor $\sigma(H)/\sigma(H_2) = 1.26 \pm 0.06$ standard deviation (s.d.) for Fe ions. This result is consistent with the ratio 1.36 ± 0.02 s.d. measured by Crandall *et al.*[13] for a variety of heavy multiply charged ions colliding with H and H_2 at velocities in the 10^7 cm/s range. Both the absorbing-sphere and tunneling models predict that at low energies the cross sections should decrease as the ionization potential of the target increases. The measured ratio for Fe^{q+} ions is consistent with the factor of 1.32 which results from the scaling of the capture cross sections with the inverse square of the target ionization potential predicted theoretically by Presnyakov and Ultantsev,[18] and demonstrated experimentally for noble-gas-ion—noble-gas collisions by Müller *et al.*[19] and Bliman *et al.*[15]

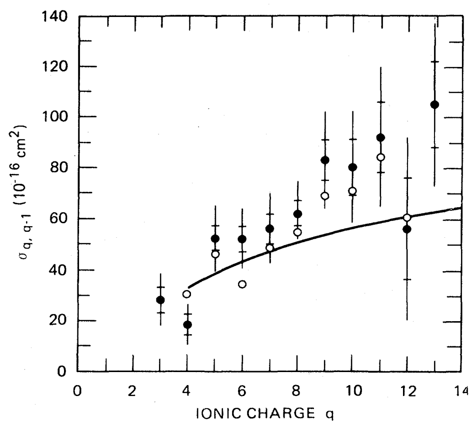

FIG. 3. Total electron-capture cross sections plotted vs ionic charge for $Fe^{q+} + H_2$ collisions (solid circles are present results) and for $Xe^{q+} + H_2$ (open circles, from Ref. 13). For $Xe^{q+} + H$ data, $v = 4 \times 10^7$ cm/s, and for $Fe^{q+} + H_2$ data, $v = (0.6\text{—}1.3) \times 10^7$ cm/s. Flags are as defined in Fig. 1. Solid curve is absorbing-sphere model of Olson and Salop (Ref. 7) for a collision velocity of 10^7 cm/s.

IV. SUMMARY

Experimental total cross sections for electron capture in $Fe^{q+} + H$ collisions at $E < 100$ eV/amu and for $3 \leq q \leq 14$ have been found to scale in an approximately linear fashion with ionic charge q and may be represented to $\pm 25\%$ by the empirical formula: $\sigma = q \times 10^{-15}$ cm^2. The magnitudes of the capture cross sections and this general scaling with q are expected to be representative of any multicharged-ion atomic hydrogen collision system which contains more than a few bound electrons. Generalized theoretical models which attempt to parametrize such processes in terms of ionic charge, target ionization potential, and collision velocity are found to be reliable to within their predicted uncertaintites of a factor of 2. Cross sections for $Fe^{5+} + H$ and $Fe^{6+} + H$ have been demonstrated to be essentially velocity independent over the range $v = (0.6\text{—}6) \times 10^7$ cm/s. Again, for systems containing at least several bound electrons, this behavior is expected and should be representative of cross sections for higher q as well. For ions of the same charge q, the apparent saturation of the cross sections as the number of bound electrons is increased has been demonstrated at low energies for a number of heavy ions by Crandall *et al.*[13] A reasonable criterion for such behavior is that the number of bound electrons in the system should exceed the ionic charge, i.e., $(Z - q) > q$.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge the advice and encouragement of D. H. Crandall, valuable suggestions concerning the manuscript from C. Bottcher, and helpful discussions with R. E. Olson. This research was supported by the Fundamental Interactions Branch, Division of Chemical Sciences, Office of Basic Energy Sciences of the U.S. Department of Energy under Contract No. W-7405-eng-26 with the Union Carbide Corporation.

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