Neutralization of H^- in Energetic Collisions with Multiply Charged Ions

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Employing the crossed-beam technique, we have measured absolute cross sections for neutralization of H^- ions in collisions with multiply charged ions Ne^{q+} ($q \leq 4$) and Ar^{q+}, Xe^{q+} ($q \leq 8$) at center-of-mass energies ranging from 20 to 200 keV. It is found that the cross sections are independent of the target ion species. The data are in excellent agreement with quantum calculations. A universal scaling law for the neutralization cross section is given.

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The investigation of neutralization reactions of negative ions in collisions with neutral atoms and molecules is a well established field (see, e.g., the review by Risley [1]). In contrast, only very few data are available on neutralization processes of negative ions in energetic collisions ($E_{\mathrm{c.m.}} > 10$ keV) with positive ions [2,3].

In this Letter we report on a comprehensive study, both experimentally and theoretically, of neutralization of H^- ions in energetic collisions with multiply charged ions. We show that the neutralization cross section σ_{-0} is independent of the target ion species. Quantum calculations outlined below are found to be in excellent agreement with the experimental data. In addition, a universal scaling law for σ_{-0} can be deduced from our theoretical approach.

Employing the crossed-beam technique, we have measured absolute cross sections for neutralization reactions

$$H^- + X^{q+} \rightarrow H^0 + \cdots \qquad (X\colon \text{Ne, Ar, Xe}) \qquad (1)$$

at center-of-mass energies ranging from 20 to 200 keV. Positive ion charge states $q \leq 4$ (Ne^{q+}) and $q \leq 8$ (Ar^{q+}, Xe^{q+}) were used. Reactions of type (1) have lately received particular interest because of possible applications in magnetic-fusion related neutral beam injection [3–5]. Furthermore, the behavior of the loosely bound electron of the H^- ion subject to the strong long-range Coulomb field of its multiply charged collision partner is of intrinsic interest.

Since the principle experimental arrangement [6] and the signal-recovery technique [7] employed for the present measurements have been described in detail previously, only a short account will be given here. Two well collimated and charge-analyzed ion beams of adjustable energies (H^- beam: 9–238 keV, 30–100 nA; X^{q+} beams: $q\times$ 10 keV, 2–150 nA delivered by a 5 GHz electron cyclotron resonance ion source) are made to intersect at an angle of 45° in an ultrahigh vacuum of a few 10^{-11} mbar. Downstream from the interaction region, an electrostatic analyzer is used to separate the neutral hydrogen atoms from the parent H^- ions. The H^0 atoms are detected and counted in a channeltron-based single particle detector. The final charge state of the target ion X^{q-i} ($i = 0, 1$) remains undetermined in this experiment. Even under the prevailing ultrahigh vacuum conditions, background events originating from charge-changing collisions of H^- ions with residual gas particles dominate by typically 2 orders of magnitude. In order to discriminate H^0 atoms formed in collisions (1) from background events, a beam-pulsing technique [7] has been applied. Typical signal rates were between 5 and 50 counts/s; typical measurement times were up to 2 h for one cross section.

The calculations presented are based on the theory of ion-atom collisions [8,9] developed as a generalization of the Keldysh [10] theory of multiphoton ionization. The basis of the theory is as follows: One-electron removal from H^- occurs at large internuclear distances where the positive ion field can be described as the pure Coulomb field. The dynamic part of the problem has an exact solution if we expand the electron-projectile interaction potential over multipoles and retain the monopole and dipole terms. Thus, the physical mechanism of electron removal from the H^- ion, i.e., the combined processes of electron transfer into the target ion and ionization of the electron into the continuum, is formulated as underbarrier and overbarrier electron transmission in the nonstationary potential created by the ion moving in the vicinity of the atom. In this approach, the binding energy of the H^- ion acquires the imaginary part related to the decay of the initial state during the collision, and the wave function is a wave packet of the Volkoff-Keldysh states. The stationary-phase three-dimensional calculations enable us to express all the elements of theory [8,9] (the imaginary part of the binding energy, transition amplitudes, the unitarity of the transition probabilities) in terms of the reduced transition amplitudes given by

$$h(\mathbf{p}) = \int_{-\infty}^{\infty} dt \langle \Phi_{\mathbf{p}}(\mathbf{r}, t) | \mathbf{r} \cdot \mathbf{F}(t) | \Phi_0(r) e^{-i\varepsilon_0 t} \rangle . \qquad (2)$$

Here, $\mathbf{p}$ and $\mathbf{r}$ are the momentum and coordinate of the active electron, $\Phi_0(r)$ is the unperturbed wave function of

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the negative ion, and ε_0 is the real part of the binding energy. The magnitude of the ion field

$$\mathbf{F}(t) = q \frac{\mathbf{R}(t)}{[R(t)]^3} \tag{3}$$

depends on the positive ion charge q and on the time-dependent internuclear distance $\mathbf{R}(t)$. Note that it is possible to omit the monopole term, $q/R(t)$, because its phase contribution is identical for both the initial and final channels and cancels out in Eq. (2). The Volkoff-Keldysh states

$$\Phi_{\mathbf{p}}(\mathbf{r}, t) = (2\pi)^{-3/2} \exp\left[i\mathbf{k}(t) \cdot \mathbf{r} - \frac{i}{2} \int_0^t k^2(\tau)\, d\tau \right],$$

$$\mathbf{k}(t) = \mathbf{p} - \mathbf{A}(t), \qquad \mathbf{A}(t) = -\int_0^t \mathbf{F}(\tau)\, d\tau \tag{4}$$

describe the motion of the unbound electron with definite values of momentum $\mathbf{p}$ in the time-dependent field and satisfy the nonstationary Schrödinger equation

$$\left[i\frac{d}{dt} + \frac{1}{2}\Delta_r + \mathbf{r} \cdot \mathbf{F}(t) \right] \Phi_{\mathbf{p}}(\mathbf{r}, t) = 0. \tag{5}$$

The total probability for electron removal from the negative ion is given by [8,9]

$$W = 1 - \exp\left\{ -\int d\mathbf{p}\, |h(\mathbf{p})|^2 \right\}, \tag{6}$$

and the cross section is then obtained by integrating over the impact-parameter variable.

The problem of choosing the unperturbed negative ion wave function is solved on the basis of a short-range one-electron potential approach. The one-electron representation is natural for the final channel. As for the initial one, two-electron correlation effects are important for calculations of the binding energy of the unperturbed H^- ion. However, if the binding energy is known, the short-range potential model can be very effective for the analysis of bound-free processes; e.g., electron detachment in a static electric field, photoionization, multiphoton ionization, etc. [10–12]. In the present approach, the integral over $\mathbf{r}$ in Eq. (2) has the main contribution from the asymptotic region where the wave function is defined by

$$\Phi_0(r) \approx \Phi_0^{\text{as}}(r) = \frac{B\sqrt{\gamma}}{\sqrt{2\pi}} \frac{e^{-\gamma r}}{r}, \tag{7}$$

$\gamma/2 = -\varepsilon_0 = 0.0275$ a.u. $= 0.75$ eV and $B = 1.56$. The value of B is obtained from electron-density calculations with Pekeris's many-parameter two-electron wave function [13].

The cross section parametrization is a very important item for many applications [14]. For electron removal from the neutral hydrogen atom, a scaling law was established and compared with experimental data [15–17]. In the present case, the Keldysh-type theory [8,9] leads to the following scaling law for the electron removal cross section (charge transfer plus ionization):

$$\sigma = 2\pi \int_0^\infty W b\, db = qQ(v^2/q), \tag{8}$$

where W is given by Eq. (6) and v is the collision velocity. The values of Q were calculated numerically and investigated analytically. On the basis of asymptotic behavior at low and high velocities, the following analytic expressions have been found:

$$\frac{\sigma}{q}[10^{-14}\ \text{cm}^2] = \begin{cases} 0.239 \ln(2.50/y + 0.8), \\ \quad y \le 0.3, \\ (0.129/y)\ln(8.12y + 1.01), \\ \quad y \ge 0.3, \end{cases}$$

$$y = 0.25v^2/q = E/100q, \tag{9}$$

where v is taken in atomic units $v_0 = 2.2 \times 10^6$ m/s and E in keV/amu. It is worth noting that the discrepancy between the two formulas is less than 1% in the region of $0.1 < y < 1$. The analysis [9] shows that the theory presented is valid for $0.02/q^2 < y < 1.5q$. At larger values of y the dipole potential in Eq. (2) should be replaced by the Coulomb potential [9]. The result for $y > q$ can be expressed in the form

$$\frac{\sigma}{q}[10^{-14}\ \text{cm}^2] = \frac{0.129}{y} \ln\left(\frac{8.12y}{\sqrt{1 + 0.557y/q}} + 1.01 \right), \tag{10}$$

which agrees with *ab initio* Bethe-Born calculations [18] in the asymptotic region $y \gg q$ for ionization by bare ion projectiles. Here we observe small deviations from the scaling law (8) and (9) at high but still nonrelativistic energies (see Fig. 1 below). At relativistic energies, the value of y in Eqs. (9) and (10) takes the form

$$y = \frac{E}{100q} \frac{(1 + 0.5E/mc^2)}{(1 + E/mc^2)^2}, \tag{10a}$$

where m is the photon rest mass ($mc^2 = 938 \times 10^3$ keV). The theory discussed is valid for $E < 2 \times 10^6$ keV/amu and for all values of the positive ion charge q.

Experimental cross sections for neutralization reactions (1) are shown in Fig. 2 together with theoretical results at a center-of-mass energy $E_{\text{c.m.}} = 50$ keV for charge states $q \le 4$ for neon ions and $q \le 8$ for argon and xenon ions. The neutralization cross sections σ_{-0} are found to be increasing with q from 4.6×10^{-15} cm^2 for $q = 1$ to $7.2 \times$

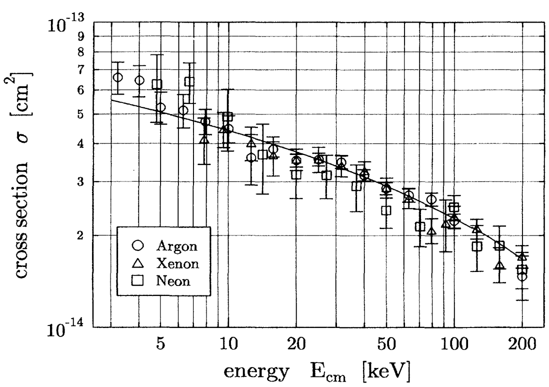

FIG. 1. Energy dependence of cross sections σ_{-0} for $H^- + X^{4+} \to H^0 + \cdots$ [X: Ne(□), Ar(○), Xe(△)] and theoretical calculations (——) according to Eq. (8).

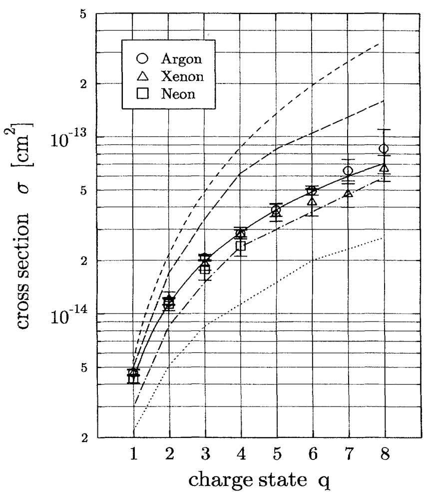

FIG. 2. Cross sections σ_{-0} for $H^- + X^{q+} \rightarrow H^0 + \cdots$ [X: Ne(□), Ar(○), Xe(△)] at $E_{c.m.} = 50$ keV compared to theoretical calculations [—— (this work), - - - Kim *et al.*, 1971]. The long-dashed, dot-dashed, and dotted curves represent CTMC calculations [3,22] with different model potentials.

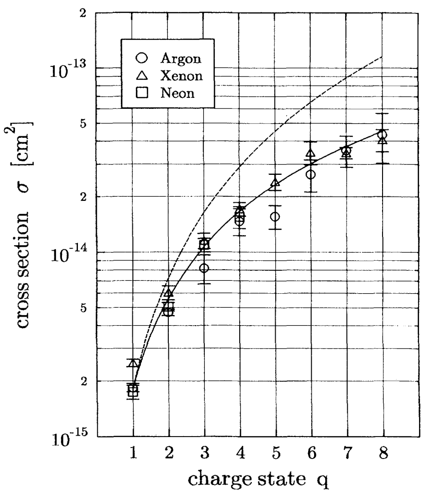

FIG. 3. Cross sections σ_{-0} for $H^- + X^{q+} \rightarrow H^0 + \cdots$ [X: Ne(□), Ar(○), Xe(△)] at $E_{c.m.} = 200$ keV compared to theoretical calculations [—— (this work), - - - Kim *et al.*, 1971].

10^{-14} cm^2 for $q = 8$ and do not exhibit any dependence on the target ion species. The Bethe-Born calculations of Kim and Inokuti [18] (short-dashed line) agree with the data only for $q = 1$. For larger values of q, this approach fails to yield a valid description of the experimental data, since at a center-of-mass energy of 50 keV the neutralization cross section does not yet assume its asymptotic Born-type behavior. The results of classical trajectory Monte Carlo (CTMC) calculations shown in Fig. 2 significantly depend on the choice of the binding potential and are roughly proportional to the square of the classical turning radius. Calculations with a short-range potential, giving the correct H^- binding energy, are a factor of 3 smaller than the experimental values. This is due to the neglect of the tunneling mechanism as well as to the difference between the classical microcanonical distribution and the quantum Wigner distribution function, especially pronounced for short-range potentials. Classical mechanisms, which often give good results for Coulomb interactions, should be replaced by quantum treatments when either underbarrier transitions are important or the WKB approximation is not applicable. Comparison of calculations based on the original CTMC method [19] with experimental results for electron detachment cross sections in collisions of H^- ions with various other negative ions also showed that CTMC results are a factor of 3 below experiment [20].

Theoretical values obtained by the present quantum calculations are in excellent agreement with the data. The cross sections σ_{-0} at $E_{c.m.} = 200$ keV (Fig. 3) are found to show the same behavior. The experimental results for $E_{c.m.} = 20$ and 100 keV (not shown here) are also reproduced by our calculations with the same overall accuracy as in Figs. 2 and 3.

The energy dependence of σ_{-0} for the reactions

$$H^- + X^{4+} \rightarrow H^0 + \cdots \qquad (X\text{: Ne, Ar, Xe}) \qquad (11)$$

is shown in Fig. 1. Again, within the experimental error bars, no dependence on the target ion species exists. The cross section is smoothly declining from approximately 6.5×10^{-14} cm^2 at $E_{c.m.} = 3.2$ keV to approximately 1.5×10^{-14} cm^2 at $E_{c.m.} = 200$ keV. The solid line represents results obtained with the present theoretical method and agrees very well with the experimental data. A survey of cross section energy dependence for other values of target ion charge states q covered in our experiment yielded very similar results.

In Fig. 4, scaled cross sections σ_{-0}/q are shown as a function of scaled energy $E_{c.m.}/q$ together with a curve

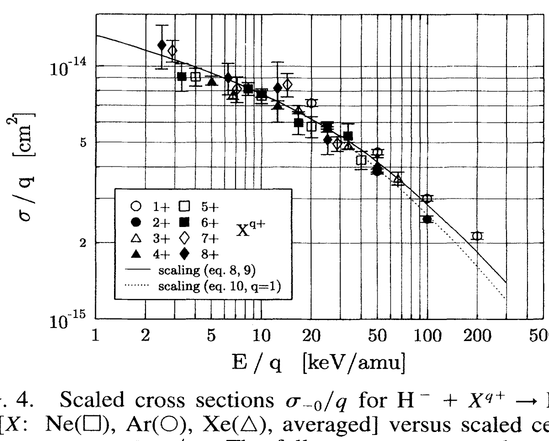

FIG. 4. Scaled cross sections σ_{-0}/q for $H^- + X^{q+} \rightarrow H^0 + \cdots$ [X: Ne(□), Ar(○), Xe(△), averaged] versus scaled center-of-mass energy $E_{c.m.}/q$. The full curve represents theoretical values calculated from Eqs. (8) and (9).

obtained from the scaling law given by Eqs. (8) and (9). The experimental values are cross sections σ_{-0} averaged over all ion species at the same scaled energy $E_{\mathrm{c.m.}}/q$. The theoretically deduced scaling law is valid for all charge states q in the whole energy range covered by our experiment.

In summary, we have presented a comprehensive experimental and theoretical study of H^- neutralization in energetic collisions with multiply charged positive ions. Our data show that the neutralization cross section is independent of the target ion species regardless of the target ion charge state q. This is an experimental verification of Massey's early prediction in 1976 "that various positive ions of the same charge tend 'to look alike' to a distant H^- ion" [21]. Quantum calculations based on the expansion of the nonstationary electron wave function over the set of Volkoff-Keldysh states are in excellent agreement with experiment. Additionally, a scaling law deduced from theory is nicely confirmed by the experimental data. Thus, accurate predictions of cross sections for H^- neutralization in collisions with multiply charged ions in a wide range of collision energies are now available.

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