Quenching collisions of low-energy metastable multiply charged argon ions

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Quenching rates have been measured for selected metastable levels of Ar^{q+} ions ($q=2$, 3, 9, and 10) stored in a Kingdon ion trap, with mean energies of $262q$ eV and $181q$ eV. Effective quenching cross sections derived from these rates are found to be comparable to electron-capture cross sections of Ar^{q+}-Ar collisions studied independently using ion-beam techniques. This implies that quenching is dominated by electron-capture collisions which change the ion charge state.

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I. INTRODUCTION

An increasing amount of work has been recently done on the collisions of multiply charged ions which have kinetic energies less than 1 a.u. [1]. These collisions are significant processes in astrophysical and controlled fusion plasmas, and have recently become necessary for the measurement and analysis of the lifetimes of metastable levels of highly charged ions confined in ion traps [2]. Electron capture and transfer ionization are the most important collision processes which change the charge of an ion at these energies, but other collisions such as charge stripping also occur with much lower cross section [3]. In basic terms, electron transfer has been successfully analyzed using extended forms [4,5] of the classical overbarrier model [6] and much work has been done on two-electron targets such as He and H_2 [1]. For the many-electron targets used in the recent metastable quenching studies, fewer measurements have been completed, but a basic understanding has nonetheless emerged. Vancura *et al.* [7] have studied the Ar^{q+}-Ar electron-transfer collision for $8 \le q \le 16$, and this collision was earlier investigated by Klinger, Müller, and Salzborn [8] for $2 \le q \le 7$, Aubert *et al.* [9] for $2 \le q \le 12$, and Bárány *et al.* [4] for $4 \le q \le 8$. Typically, only one or two electrons are gained by the projectile ions, although highly charged recoil ions are produced [10]. The cross section for single-electron capture is two to three times that for double-electron capture, and, for example, the total capture cross section for Ar^{8+} on Ar was about 10^{-14} cm^2, with a weak energy dependence between $0.4q$ and $2.7q$ keV [11]. Work by Hoekstra *et al.* [12] investigated the question of the influence of the projectile core electrons on single-electron capture. Using a Li target, fully stripped B^{5+}, H-like C^{5+}, and He-like N^{5+} ions all had the same single-electron-capture cross section between 2 and 8 keV/u. The cross section for Be-like Ne^{6+} differed from that of fully stripped C^{6+}, but no explanation for the difference was offered.

At very low energies, electron transfer is analyzed in terms of the potential-energy surfaces of a quasimolecule formed during the collision [13]. Under these conditions, a metastable state may have a very different electron-capture cross section from a ground state. since the energy-level differences are comparable to the kinetic energy of the collision. At higher projectile energies, more product channels open, and eventually the classical overbarrier model becomes appropriate. Studies of collisions in which the initial level is identified are useful in verifying level independence, or in determining situations when level dependence exists. Such studies have been carried out with Ar ions on inert gas targets using translational energy spectroscopy [14–16] and analyzed using Landau-Zener theory, based on the crossings of potential-energy levels of the quasimolecular collision system. This analysis has also proved effective in the study of angular distributions of Ar^{q+}-Ar collisions [17]. Total cross sections for electron capture in Ar^{4+} + Ar and Ar^{6+}-Ar collisions were obtained from these angular distributions [18].

The metastable states of interest in the lifetime measurements are excited levels of the ground term. These levels will have significant populations in most collision measurements, since they will probably be statistically populated in the ion source, and typically have lifetimes of milliseconds or more, long compared to typical beam transport times. From this, one can conclude that measured cross sections of processes like electron transfer are usually averaged over the populations of these levels, particularly at the higher collision energies. On the other hand, when depopulation of a specific metastable level by collisions is studied, charge transfer is only one of several processes which may destroy the level population. Electron transfer is often endothermic in collisions of many singly charged ions, and at low energies fine-structure mixing by ion–neutral-species collisions plays an important role [19]. Stark quenching has been observed in He^+ metastable lifetime measurements [20]. Although electron-transfer collisions are expected to dominate, it appears useful to verify the primary source of metastable level quenching of highly charged ions, under measurement conditions involving ion storage.

Using a Kingdon ion trap [2,21], we have studied the quenching of metastable levels of argon ions having states $q=2$, 3, 9, and 10 at mean energies of $262q$ eV and in some cases at $187q$ eV. The quenching occurred in collisions with Ar target gas. Previously, a Kingdon trap had been used to obtain the cross sections for electron

transfer from Ne and Xe atoms to Ne^{q+} ions ($3 \leq q \leq 10$) at energies of a few q eV [22].

II. TECHNIQUE AND MEASUREMENTS

The general technique used in these measurements has been previously described [3,21,23] and only a brief synopsis is appropriate here. Metastable- and ground-state ions extracted from an electron cyclotron resonance ion source (ECRIS) were selected by charge-to-mass ratio, and focused through the midplane of a Kingdon electrostatic ion trap. The trap consisted of a cylinder and axially concentric wire, terminated with two end plates. Four symmetrically placed apertures in the cylinder midplane permitted transmission of the ion beam, and the detection of photons and released ions at right angles to the ion-beam direction.

The cylinder potential V_0 was near the extraction potential of the ECRIS V_{ext}, either 3.5 or 2.5 kV in these measurements. The end plates were operated at a potential 400 V higher, to confine the ions near the trap center, and the wire potential V_w was initially near the cylinder potential. When V_w was pulsed rapidly ($\approx$300 ns) to zero, ions inside the trap structure were captured. After a predetermined storage time, the wire potential was allowed to slowly ($\approx$10 ms) increase back to V_0. During the storage interval, photons emitted in the decay of a metastable level, and collected by a quartz optical system, were selected in wavelength with an interference filter, and detected using a photomultiplier tube. Also during the storage interval, some ions were observed to escape the trap to the particle detector. The signals were averaged over many cycles.

To determine the rate of metastable quenching, the photon signal was collected vs storage time over a range of fixed pressures of Ar target gas, which was admitted through a leak valve from a bakable gas-handling system. The photon decay data were fitted to a sum of two exponentials plus a constant background. There was an initial decrease in intensity with a short time constant, observed in all the data, associated with stabilization of the ions in the trap. The longer time constant decay was due to the decay of the metastable level by the transition selected with the interference filter. If the initial channels were eliminated from the data, this longer decay was well fitted by a single exponential plus constant background. The time constant for this decay varied inversely with the pressure of the target gas. The slope of a plot of the inverse of the measured decay time constants vs target gas pressure was the rate coefficient $k=\sigma\bar{v}$ for quenching collisions.

For the logarithmic potential near the trap midplane, the virial theorem provides the relationship $\bar{v}^2 = qeV_0/m \ln(a/b)$ for an ion with charge qe and mass m, confined by the potential difference V_0 in a trap with the ratio of cylinder to wire radii a/b. In the potential away from the trap midplane, the more general relation

$$\bar{v}^2=(qe/m)\mathbf{r}\cdot\nabla V(r,z)$$

applies [24], but is much more difficult to evaluate. In electron-transfer studies, carried out earlier in a Kingdon trap [22], the velocity $(\bar{v}^2)^{1/2}$ was used with the rate coefficient for electron transfer $k_c=\sigma_c\bar{v}$ to obtain the effective cross section for charge transfer $\sigma_{c0}=k_c/(\bar{v}^2)^{1/2}$. The effective cross section for quenching can be calculated in a similar way, for comparison with cross sections measured in other ways. Prior, Marrus, and Vane [22] studied the distribution of velocities of ions near the midplane of a similar Kingdon trap, and found agreement with the predictions of the virial theorem to within 10%.

III. ANALYSIS

A charge-changing collision of a multicharged ion with a neutral atom results in the formation of two product ions, one fast (the incident ion with reduced charge q') and one slow (with charge q_2). The kinetic energy of the slow product ion can be estimated from the repulsive potential energy of the charged products at the capture radius R_x:

$$V(R_x)=\frac{q'q_2e^2}{R_x} ,$$

which is shared approximately equally between the ions if their masses are similar. If capture occurs from a molecule, dissociation into slow product ions is also possible, where the energies now depend in part on the equilibrium separation of the nuclei of the neutral molecule. For both cases, the energies are multiples of electron volts. This can be compared to the mean initial kinetic energy of the original ion:

$$E_{\mathrm{kin},i}=m\bar{v}^2/2=qeV_0/2\ln(a/b)=P_0/2=262q \text{ eV}$$

for $V_0=3.5$ kV. Product ions must have sufficient angular momentum relative to the central wire to remain confined. The highly eccentric orbits of the slow products are likely to lead to rapid ion loss by collision with the central wire, due to small perturbations by weak collisions with other ions or atoms. Johnson [24] has expressed the condition on the initial ion energy for stability in the trap as

$$E_{\mathrm{kin},i} > P_0 \ln(r/b)/[(r/b)^2\sin^2\theta-1] ,$$

where $P_0=qeV_0/\ln(a/b)$ and θ is the angle between the initial ion velocity vector and the radius. This applies to an ion formed at radius r. Since $P_0 > E_{\mathrm{kin},i}$, this condition is likely to be satisfied only at large r, and then most likely for $\theta\approx\pi/2$.

The mean kinetic energies of the fast product ions from electron transfer only exceed their initial kinetic energies slightly, due to the relatively small amount of kinetic energy transferred. Energy gains less than 15 eV are typically observed [15]. However, the charge q of the ion has been reduced to q' so it moves in a lower potential energy. Approximating the initial kinetic energy of the new ion as the original kinetic energy, then the sum of the new kinetic and potential energies must be less than the radial well depth for continued stable confinement. This condition can be written

$$P_0/2+P_0'\ln(r/b)<q'eV_0 ,$$

and, using the definitions of P_0 and P_0', the equation reduces to

$$q/q'<2\ln(a/r)$$

in terms of the initial charge state q and final charge state q'. This results in a limitation on the radius for which the new ion might remain stable: $r<ae^{-(q/2q')}$. Johnson [24] has expressed this condition in a different way, taking into account the angle θ defined above:

$$E_{\mathrm{kin},i}<P_0'\ln(r/a)/[(r/a)^2\sin^2\theta-1] ,$$

which reduces to

$$r<ae^{-(q/2q')[1-(r/a)^2\sin^2\theta]} .$$

From either analysis, only fast ions that change charge at a relatively small radius in the trap (which depends on the initial and final charge states) will remain confined.

These conditions for stability limit, but do not forbid, the storage of product ions following electron transfer to highly charged ions. Based on a straightforward analysis of our observations, in which ions remaining in the trap at the end of the storage cycle were detected regardless of their charge-to-mass ratio, few, if any, product ions were confined. The observed ion number vs storage time decreased as the storage time was lengthened [2,21]. At higher residual gas pressures, the rate of decrease of ion number was faster. If all product ions were confined, one would expect the detected ion number to increase with storage time. If only the fast products remained confined the observed ion number should be constant. Clearly, most product ions were not confined.

The ions in these measurements were detected by letting the wire potential rise adiabatically toward the cylinder potential. The angular momentum of the ions $L=r(mP_0)^{1/2}$ is an adiabatic invariant under these conditions. Consequently, the mean radius of the ion orbits increases as $(V_0-V_w)^{-1/2}$. The observed ion peaks plotted vs release time showed no structure. As $V_w(t)$ approaches V_0, modeling shows that a weak saddle potential forms about the trap midplane near $r=0.7$ cm, due to the higher potential $V_c>V_0$ of the end caps. However, the adiabatic expansion of the charge cloud should permit all charges to escape radially. The observed sample of ions from the trap midplane is expected to be representative of the whole stored sample. Within the available signal-to-noise radios, the ion number signals vs storage time were fitted to a single exponential plus constant background. The time constant for the exponential loss varied inversely with target gas pressure. These observations imply essentially no product ion storage. A background of ions is observed with the particle detector during the storage time before the trap is dumped [21]. This background decreases with time, but is not fitted well by a single exponential, as might be expected for ions formed at different times, rather than at a single time such as an ion injection. This background signal is thought to be due to unstable fast product ions formed by electron transfer, escaping the trap.

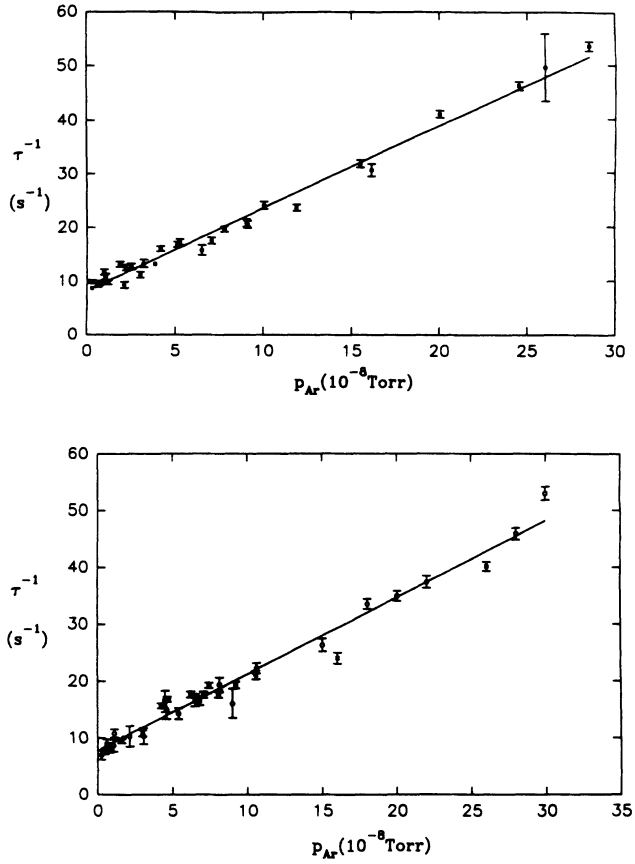

FIG. 1. (a) Plot of the reciprocals of measured metastable decay times of the Ar^{2+} $3p^4\,{}^1S_0$ level vs argon gas pressure. The slope of the plot is the quenching rate coefficient k_{qu}. The mean ion energy was 524 eV. (b) A similar plot at a mean ion energy of 374 eV.

In the earlier Kingdon trap, electron-transfer measurements of Prior, Marrus, and Vane [22], the ions were detected using charge-to-mass selection provided by a quadrupole mass analyzer. Stored product ions accumulating in any charge state would have caused a departure from a single-exponential ion loss rate, which was not ob-

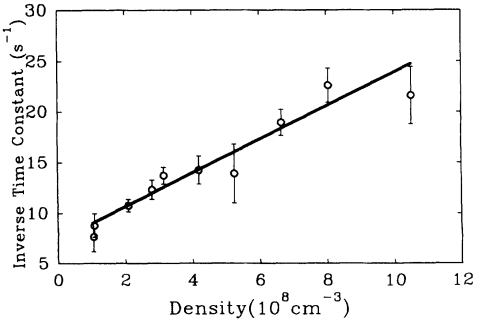

FIG. 2. Plot of the reciprocals of measured metastable decay times of the Ar^{3+} $3p^3\,{}^2P_{3/2}$ level vs argon gas pressure. The slope of the plot is the quenching rate coefficient k_{qu}.

TABLE I. Measured reaction rates for quenching k_{qu} and derived cross sections σ_{qu} for argon ions in metastable levels colliding with Ar.

Collision	Energy (keV)	Rate ($cm^3 s^{-1}$)	Cross section (cm^2)
$Ar^{2+}(^1S_0)+Ar$	0.37	$3.74\pm0.2\times10^{-9}$	$8.6\pm0.5\times10^{-16}$
	0.52	$4.2\pm1.1\times10^{-9}$	$8.3\pm2.1\times10^{-16}$
$Ar^{3+}(^2P_{3/2})+Ar$	0.79	$1.66\pm0.25\times10^{-8}$	$2.7\pm0.4\times10^{-15}$
$Ar^{9+}(^2P_{1/2})+Ar$	1.7	$8.3\pm0.35\times10^{-8}$	$9.1\pm0.4\times10^{-15}$
	2.4	$8.5\pm0.5\times10^{-8}$	$7.9\pm0.5\times10^{-15}$
$Ar^{10+}(^3P_1)+Ar$	2.6	$10.1\pm0.6\times10^{-8}$	$8.8\pm0.5\times10^{-15}$

served. All of these observations indicate that the formation of significant numbers of stored metastable or ground level ions during the observation period is unlikely.

The rate coefficients for metastable level quenching were obtained directly from linear fits to the data in the Stern-Volmer plots of the measured reciprocal metastable decay time constants vs target atom pressure [23]. Examples of such plots for Ar^{2+} and Ar^{3+} appear in Figs. 1 and 2. Similar plots for Ar^{3+} in N_2, Ar^{9+} in N_2, and Ar^{10+} in Ar appear in Refs. [23,25,2], respectively. Effective mean cross sections for quenching in Ar were computed from the rate coefficients using the relation $\sigma_{qu}=k_{qu}/(\bar{v}^2)^{1/2}$. The measured rates and cross sections are summarized in Table I.

IV. DISCUSSION

Electron transfer from Ar to Ar^{q+} has been measured at several energies, most of which lie above the energy range of collisions studied here. At the lower energies, Biedermann *et al.* [18] measured cross sections for capture in collisions of Ar^{4+} with Ar at energies between 100 and 800 eV, obtaining

$$\sigma=64(20)\times10^{-16}\ \text{cm}^2\ ,$$

where the maximum variation in this energy range is stated (rather than the overall measurement uncertainty, which was 30%). For $Ar^{6+}+Ar$ collisions, the total capture cross section was $54(13)\times10^{-16}$ cm^2 between 300 and 800 eV, where again the maximum variation is given. Although these two cross sections do not apply directly to the ions studied in our work, the results indicate that for similar charge states and energies, the cross section varies little with energy. Bárány *et al.* [4] present electron-capture cross-section data at $1.8q$ keV for Ar^{4+} to Ar^{8+} in collision with Ar; for the cases of Ar^{4+} and Ar^{6+}, the cross sections at these higher energies differ little from the low-energy results.

Total cross sections for transfer of up to three electrons were obtained by Vancura *et al.* [7] at $2.3q$ keV for $q=9$ and 10 (as well as higher charge states). Their results are

$$\sigma_9=10.6\pm1.1\times10^{-15}\ \text{cm}^2$$

and

$$\sigma_{10}=9.9\pm1.0\times10^{-15}\ \text{cm}^2\ .$$

The single-electron transfer cross sections

$$\sigma_{9,8}=9.1\pm0.9\times10^{-15}\ \text{cm}^2$$

and

$$\sigma_{10,9}=8.1\pm0.8\times10^{-15}\ \text{cm}^2$$

are about 20% smaller. Aubert *et al.* [9] studied single capture from Ar to Ar^{q+} vs energy, for $q=2$ and 3. At 1.2 keV,

$$\sigma_{2,1}=2.6\times10^{-16}\ \text{cm}^2$$

and at 4.5 keV,

$$\sigma_{3,2}=2.2\times10^{-15}\ \text{cm}^2\ .$$

The Ar^{2+} cross section increased slowly to three times this quoted value near 6 keV, while the Ar^{3+} cross section remained essentially constant up to 42 keV. They also measured

$$\sigma_{9,8}=6\times10^{-15}\ \text{cm}^2$$

and

$$\sigma_{10,9}=5.5\times10^{-15}\ \text{cm}^2$$

at 30 keV. Measurements by Klinger, Müller, and Salzborn [8] are at still higher energies. The energy-gain measurements [15] indicate that below 1 keV, the different metastable levels of, e.g., Ar^{3+} and Ar^{4+} change charge with neutral targets at somewhat different rates, resulting in energy gains with different event totals. Nevertheless, the total cross section is relatively independent of energy over a considerable range, as discussed above. It is instructive to compare the quenching cross sections from Table I with the electron-transfer cross sections, even though the energies are somewhat different, due to the relatively small ($\approx50\%$) variations of electron-transfer cross section with projectile energy in this range.

For Ar^{10+}, the quenching cross section is 8.8×10^{-15} cm^2 at 2.6 keV, compared to single-electron capture cross sections of 8.1×10^{-15} cm^2 at 23 keV and 5.5×10^{-15} cm^2 at 30 keV, and a total capture cross section of 9.9×10^{-15} cm^2 at 30 keV. For Ar^{9+}, the effective quenching cross section is 7.9×10^{-15} cm^2 at 2.4 keV and 9.1×10^{-15} cm^2 at 1.7 keV, while the total-electron-capture cross section is 10.6×10^{-15} cm^2 at 20.7 keV and the single-electron capture cross sections are 9.1×10^{-15} cm^2 at 20.7 keV and 6×10^{-15} cm^2 at 30 keV. For Ar^{3+}, the quenching cross section is 2.7×10^{-15} cm^2 near 800 eV, compared to the single-electron transfer cross section of 2.2×10^{-15}

cm^2 at 4.5 keV. The close agreements of these quenching and electron-transfer cross sections, despite the energy differences for three charge states plus the minimal measured variations of the cross sections with energy for multicharged ions in the energy range, e.g., Ar^{4+} and Ar^{6+}, permit the conclusion that quenching of metastable levels in these high charge state ions is dominated by electron-transfer collisions which change the charge of the ions. These findings are in accord with the data analysis of Vancura *et al.* [7], who found good agreement of the charge state dependence of their data with an absorbing sphere model of Olson and Salop [26], and with the observations of the independence of capture cross sections of the presence of core electrons [12]. In the classical overbarrier model, the electrons are transferred at relatively large distance into excited states of the ion, which depend on the charge state but are not strongly dependent on the core configuration.

The $Ar^{2+}(^1S_0)$ results remain to be considered. The quenching cross sections are 8.64×10^{-16} cm^2 at 375 eV and 8.2×10^{-16} cm^2 at 500 eV, considerably smaller than the Ar^{3+} cross sections. These are both significantly higher than a measured single capture cross section of 2.6×10^{-16} cm^2 at 1.2 keV but a clear energy dependence was observed for this collision [9]. At these lower energies, both the quenching and electron-transfer cross sections are likely to be energy and level dependent. The level dependences of the Ar^{2+} charge-transfer collisions at still lower energies have been studied [27]. Previously measured $Ar^{2+}(^1S_0)$ quenching rates of 5.5×10^{-10} cm^3s^{-1} at 1.3 eV and 2.1×10^{-9} cm^3s^{-1} at 20.2 eV are noted [28], which lead to a cross section near 1.5×10^{-15} cm^2, about twice as high as our results near 400 eV.

Finally, we compare the total loss rates of ions from the trap, as measured using the ion detector, with the quenching rates. For Ar^{2+} in Ar, the loss rate coefficient was

$$6.9\pm4.7\times10^{-9}\ cm^3s^{-1}\ ,$$

and for Ar^{9+} in Ar, the rate was

$$1.78\pm0.4\times10^{-8}\ cm^3s^{-1}\ .$$

The large uncertainties reflect the large fluctuations in the data points in this type of measurement. Comparing these numbers with the rate data for quenching in Table I, the Ar^{2+} ion loss rate is similar to the quenching rate, as might be expected if charge transfer were a major source of ion loss, but for Ar^{9+}, the quenching rate was 8.5×10^{-8} cm^3s^{-1}, exceeding by a factor of 4 the measured ion loss rate from the trap. Since the quenching rate has already been established as equal to the electron-capture rate for Ar^{9+}, the measured loss rate from this trap is low. The cause may be that some product ions were being trapped in the Ar^{9+} charge-transfer collisions, distorting the measured loss rate. Further work on this question is planned, since it may have implications for corrections to the measured metastable level lifetime [23].

V. CONCLUSION

The quenching of metastable levels of multicharged ions stored in a Kingdon ion trap at energies near 1 keV has been studied. It is noted that total cross sections for electron capture vary little as a function of energy for Ar^{q+}-Ar collisions near 1 keV. The metastable quenching cross-section results reported here have been found to be in good agreement with these independently measured total cross sections for electron capture, for Ar^{3+}, Ar^{9+}, and Ar^{10+}. This indicates that quenching of metastable levels in these high charge states is within measurement uncertainties due to change of the ion charge state by electron capture. Differences in measured quenching and charge-transfer cross sections for Ar^{2+} were observed. These differences may be associated with energy and level dependences of charge transfer in this collision, since measurements close to the low energy of this collision were not available. Quenching cross sections for $Ar^{2+}(^1S_0)$ have been measured to be twice as large at still lower energies, supporting this interpretation.

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