Charge Transfer to Multicharged Recoil Ions in a Penning Trap

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Multicharged neon recoil ions at electronvolt energies are produced inside a Penning ion trap by fast, stripped–heavy-ion impact on neon atoms. The stored recoil ions capture electrons in collisions with the parent neon gas. The measured storage times are used with mean energy determinations and neon gas densities to determine charge-transfer rate coefficients. A significant dependence on ion charge ($3 \leq q \leq 6$) is observed.

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Low-energy multicharged ions can be produced with useful cross sections in a wide range of charge states by impact of fast, stripped heavy ions with target atoms.[1,2] Such recoil ions, extracted in a beam from the collision region, have been used to study charge transfer to multicharged ions at kiloelectronvolt energies.[3] Large-angle scattering—sometimes arising from orbiting—at low collision energies,[4] as well as low-energy ion-beam focusing problems, leads to consideration of an ion-confinement measurement technique for extension of such measurements to still lower energies. Toward this goal, as well as to implement potentially interesting precision spectroscopic investigations on low-energy multicharged ions, we have initiated the study of recoil ions confined at electronvolt energies in a Penning ion trap. In the Penning configuration, ions are confined radially by a uniform axial magnetic field $\vec{B}$, and axially by a dc quadrupole potential.[5] We chose this confinement alternative for the following reasons: (1) Since the recoil of the low-energy ions is essentially perpendicular to the fast stripped-ion beam, there is a high probability of radial confinement by the magnetic field for a heavy-ion beam directed along $\vec{B}$. (2) The harmonic motion of the confined ions, with frequencies dependent on the charge-to-mass ratio q/m, allows the detection of specific ion charge states and selective removal of unwanted ions—e.g., those that could feed lower charge states by cascade charge exchange—by application of resonance excitation at the appropriate cyclotron frequency. (3) The stable confinement of ions at velocities down to zero permits the investigation of very-low-energy collisions; it may be feasible to cool the stored ions by coupling to external circuits. (4) The relatively long confinement times can be used to advantage in the study of metastable levels, which may change charge with appreciably different rates than do ground-state ions. (5) The small confinement volume is well suited to spectroscopy experiments.

Although charge-transfer studies on two and three times ionized atoms, produced by electron impact ionization, had been performed in a Penning trap,[6] at the time this research was initiated the feasibility of higher-charged recoil ion confinement was still in question. Since that time, recoil ions up to ten times ionized have been confined and studied in a Kingdon trap, in which they orbit a central wire in a logarithmic dc potential.[7] Our complementary method, which differs in significant respects, provides a first check on these highly interesting pioneering measurements.

Rate coefficients for charge transfer are useful at low energies since these collisions may significantly alter the ionization equilibrium of astrophysical[8] and laboratory[9] plasmas. The study of Ne^{+q}-Ne charge transfer is of interest because of the variety of collision processes that may occur. Collisions of inert gas atoms with doubly charged ions have been extensively investigated by flow-drift tube methods, yielding charge transfer rate coefficients k which vary over 5 orders of magnitude, depending on the ion state (ground or metastable) and the species of collidant neutral.[10] Transfer ionization and double charge transfer may dominate over single charge transfer in certain cases. The extension of low-energy measurements to higher charge states, possible with UHV technique, potentially permits a study of the q dependence of such collisions. Practically, studies of collisions of interest in plasmas (e.g., Ne^{+q} with He) require knowledge of the rate coefficients of the ions with the parent gas.

Neon recoil ions were generated by collisions with a 1.4-MeV/u Cl ion beam, foil stripped to a mean charge state near $10e$. The Oak Ridge National Laboratory's model EN tandem accelerator produces this beam at relatively high intensity ($\geq$ 100 nA through the ion trap), focused to a 2-mm spot, decisive for the use of a relatively small ($\approx$2-cm-diam) trap electrode structure. This beam is ideal for the production of Ne^{+q} ion numbers suitable for charge transfer studies having $q \leq 6$, although higher confined charge states were found (see Fig. 1) and could easily be enhanced by using more highly charged heavy-ion beams.

The fundamental parameters necessary for the determination of a charge-transfer rate coefficient k are the target density n and the time constant τ for charge changing to occur; they are related by $k = (n\tau)^{-1}$. The neon target gas (natural isotope abundance) was leaked into the collision chamber at a constant rate through a piezoelectrically controlled valve from a baked UHV gas-handling system. Residual gas analyzer (RGA) readings showed negligible entry by other gases. The collision chamber was evacuated by a nominal 1000-l/sec cryopump. Pressures were measured with a capacitance-manometer-calibrated nude ion gauge (NIG). The RGA measured relative gas concentrations. Experimental considerations dictated that the neon gas be introduced near the ion trap; this resulted in a significant pressure gradient through the vacuum system which was calibrated by controlling the flow rate at constant pressure, and also by pressure comparisons between a NIG placed at the trap site and a NIG at the pump orifice, under constant flow conditions. Differences in these calibrations admit a systematic pressure error as high as 30%.

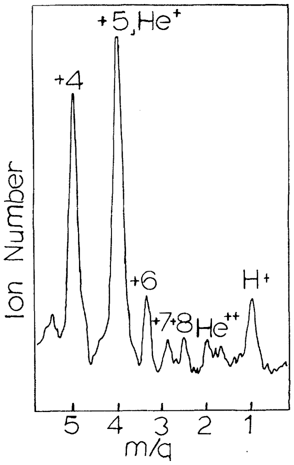

FIG. 1. Analog Ne^{+q} ion signals vs dc potential, calibrated in mass-to-charge ratio. Structure due to $^{22}\mathrm{Ne}$ can be observed on low charge peaks. The $m/q = 4$ peak consists of both Ne^{+5} and He^{+} ions, due to trace helium in the residual gas during these measurements.

The heavy-ion beam was pulsed by voltages applied to electrostatic deflection plates, and stripped by 20-μg/cm^2 foils. Following termination of the beam pulse, the recoil ions were stored for times ranging from 40 to 1500 msec. Cyclotron excitation was applied during the entire measurement cycle. Following the storage interval, the relative ion number was measured by an analog resonance method as in previous Penning-trap measurements.[6] Results are shown in Fig. 1. The Ne^{+6} signal corresponds to ≈ 65 ions, on the basis of the cross sections measured by Cocke,[2] illustrating the sensitivity of the detection method. The trap quadrupole polarity was temporarily reversed to dump any residual ions, and the cycle was repeated until an adequate signal-to-noise ratio was obtained for the delay curve of ion number versus storage time. No evidence for loss of ions to causes other than charge transfer was found. The time constant τ is obtained when data is least-squares fitted by an exponential decay.

All of the Ne^{+q} ions with $3 \leq q \leq 6$ have metastable excited states: These states have various mean lives, ranging from ≈ 10 msec to hundreds of seconds. The presence of long-lived levels can be detected if they change charge at a rate significantly different from the ground-state rate. The least-squares fits of the data by

$$N(t) = N_1 \exp(-t/\tau_1) + N_2 \exp(-t/\tau_2)$$

indicate that ion loss for each charge state is best characterized by a single exponential. Either the metastable states are not highly populated in the collision that creates the ions, or the states change charge with the same rate coefficient as the ground state. Since the latter possibility is strongly species dependent, measurements with other target gases may resolve this question.

Comparison of ion storage measurements on the parent ions using no cyclotron excitation, to others in which the next higher charge state was removed, showed no significant differences in the ion-number decrease with time, i.e., $N(t) = N(0)\exp(-t/\tau)$. Fits of the data by

$$N(t) = A_1 \exp(-t/\tau) - A_2 \exp(-t/\tau_2),$$

where A_1 and A_2 are products of ion numbers and time-constant expressions, had higher χ^2 values than fits by single exponentials, and converged to nonphysical values of the parameters. Thus "feeding" from charge-changing collisions of higher charge states was not significant. Of course, there were relatively fewer ions with higher charge, but the product ions may also receive sufficient recoil energy to leave the trap in the dissociation of the transient molecular-ion state. This result is observed in other Penning-trap measurements[6]; its complete explanation is still open. Following these observations, subsequent measurements were made with all charge states $\geqslant 3$ present. An exception to single-exponential decay occurred during a measurement sequence with residual helium present in the vacuum system. He^+ as well as Ne^{+5} ions contributed to the $m/q = 4$ peak (see Fig. 1), leading to decay curves which were not useful sources of data. Helium was excluded from the vacuum in other measurements.

At any given neon gas pressure, reactions also occur with residual gases such as H_2O. Since rate coefficients for Ne^{+q}-H_2O charge transfer may be an order of magnitude larger than the Ne^{+q}-Ne rate coefficients, serious systematic errors are possible. These can be quantified by performing measurements with neon target-gas densities ranging over a factor of 50. If we use $\tau^{-1} = n_n k_n + \sum_i n_i k_i$, where the subscript n refers to neon and i to other gases, plots of τ^{-1} vs the neon atom density yield a slope equal to the neon charge-changing rate constant for that charge state and an intercept characterizing the charge changing with constant-pressure residual gases. Examples are shown in Fig. 2, where it can be seen that residual-gas effects are 1% at the lowest neon pressure.

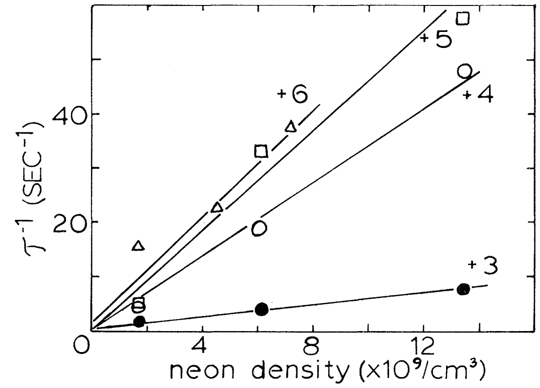

FIG. 2. Plots of reciprocal storage time constant τ^{-1} vs neon target gas density. Less than 1% foreign gas was present during the $3 \leqslant q \leqslant 5$ measurements.

To gain information about the ion energy distribution and mean energy, we measured the number of ions stored in the trap versus the trapping electrostatic potential V (or axial well depth $D = V/2$). The ion number was independent of V until $D < 4.5$ V, below which the number stored decreased smoothly as D was dropped to 1 V. Below 1 V no ions were detected. We repeated these measurements with storage times from 20 to 170 msec. No evolution of the distribution was observed, although charge transfer decreased the absolute ion number. Since there is no reason to assume equal axial and radial energies at the time an ion is produced, this stationary distribution implies that equilibrium has been reached during the $\simeq$ 1-sec ion creation pulse. This plausibly occurs as a result of the combination of elastic ion-ion and ion-atom collisions, and perhaps of anharmonicities in the trap potential. It is generally observed under equilibrium conditions that ions stored in harmonic traps assume a mean energy $E \simeq qD/10$. This mean energy is thought to result from rapid cooling of the charge cloud by loss of a relatively few high-energy ions from the axial well during the ion-production interval.[5]

In the rate-constant measurements reported here, no attempts were made to reach the lowest ion energies, but rather to operate the trap in a way to minimize ion loss from an (originally unknown) recoil energy distribution. At a depth $D = 20$ V, the mean ion energy $\simeq 2q$ eV applies to the measured rate coefficients listed in Table I.

Our rate-coefficient data in Table I show a large increase in k from $q = 3$ to 4, with slower increases for higher q. These rate coefficients apply to charge changing due to any collision process. Statistical uncertainties $\leqslant 20\%$ (highest for the +5 and +6 charge states) arise from errors in the determination of the storage time constants, and from the pressure calibrations. As discussed earlier, there is a possible systematic error as large as 30% in the absolute pressure at the ion trap. Rate coefficients in Table I derived

TABLE I. Charge-changing rate coefficients for the collisions $Ne^{+q} + Ne \rightarrow$ products.

Charge state	k (cm^3 sec^{-1}) [a]	k (cm^3 sec^{-1}) [b]
Ne^{+3}	5.1×10^{-10}	5.4×10^{-10}
Ne^{+4}	3.4×10^{-9}	2.5×10^{-9}
Ne^{+5}	4.5×10^{-9}	3.2×10^{-9}
Ne^{+6}	5.7×10^{-9}	6.2×10^{-9}

[a] This measurement.
[b] Kingdon-trap data, see Ref. 7.

from the newly published Kingdon-trap data[7] are within the expected uncertainties and tend to confirm ion mean energy estimates in these disparate trap types.

The measurements reported here are an essential component of the study of collisions of Ne ions with other targets, since both neon and the target gas must be present in the ion trap. Determinations of rate coefficients as a function of mean ion energy can be performed by varying the axial well depth of the trap; the lower limit of feasibility of this method awaits determination. This demonstration of recoil ion storage in harmonic traps may lead to important developments in areas other than charge transfer.

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