

Collisional cooling of trapped ions with cold atoms: results and insights

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Abstract. We present a series of recent experimental and numerical results which allow us to propose and explain the mechanisms for collisional cooling of a trapped ion by sequential binary collisions with atoms. Our experiments in hybrid atom-ion traps study the cooling of ions when the reservoir of the coolant atoms is spatially localized at the centre of the ion trap. Under these conditions the widely and long held expectation that for the ion to collisionally cool, the atom must be of lighter mass (i.e. $m_I > m_A$) is overturned. Instead we show ion cooling for $m_I \leq m_A$ in addition to $m_I > m_A$, and explain why earlier work establishing mass ratios does not apply to the experiments in hybrid traps. Further, for cooling of an ion A^+ by the parent atom A, the mechanism of resonant charge exchange (RCE) allows for extremely efficient cooling of the ion A^+ . This mechanism is demonstrated by comparing the cooling rate of an ion with a localized ensemble of its parent atoms and with a lighter atomic species, where in the latter case the RCE mechanism does not exist. The difference in measured cooling rates for a given number density of the atoms and the theoretically calculated difference between elastic and RCE cross sections are used to show that the cooling efficiency per collision with RCE is much greater than the cooling by elastic collision. We conclude with some perspectives and prospects of future experiments.

1. Introduction

Ever since the pioneering trapping experiments with ions [1], a primary objective has been to cool them for precision measurements [2]. As typical ion trap potentials are very deep, the trapped ion can be very energetic, which is not useful for precision work. It has therefore always been a simultaneous requirement to have a cooling mechanism in place, wherein the energetic ions can be cooled or thermalized to a fraction of the trap depth. In the era before laser cooling and other trap specific cooling developed, the available cooling mechanism was multiple sequential two-body collisions of a single trapped ion with neutral atoms or even molecules, which were at much colder temperature. Typically, the ion trap volume was flooded with a neutral cool gas, such that the ion trap was immersed in the cold gas. The resulting collisional cooling of ions by atoms, by flooding the ion trap with a neutral atom gas was called buffer gas cooling [3].

Subsequently other ion cooling methods have been developed, such as; resistive cooling with and without feedback, radiative cyclotron or synchrotron cooling and the very versatile and effective laser cooling [4]. While direct laser cooling is convenient only for a small number of ions with suitable electronic structure, its utility is greatly expanded by sympathetic cooling of



ions that are hard to laser cool [4]. The effectiveness of this cooling depends on the ratio of the mass of the two ions.

With the expansion of laser cooling to neutral atoms [5], trapped dilute gas ensembles of localized atoms were readily prepared, as a Magneto-Optical Trap (MOT) or a degenerate quantum gas of Bose or Fermi particles. Science with cold trapped ions and atoms progressed independently and rapidly for approximately two decades. Since the mid 2000's a number of new experiments developed, which combined trapped ions with cold and ultracold atoms, in hybrid trap atom-ion experiments [6–14]. The hybrid traps combine radio frequency Paul traps with atom traps. Penning traps for ions have been avoided as the magnetic field required to trap ions causes nonlinear perturbation of atomic states, which hampers the laser cooling of atoms. The focus in these experiments shifted from precision manipulation of each individual system to understanding the interactions between atoms and ions at the coldest temperatures. The study of collisions and reactive processes in these systems is of contemporary interest.

Here we consider the case of an atom-ion collision, when both the atoms A and ion B⁺ are trapped. In particular, the atoms A are laser cooled and localized at the ion trap centre. Both A and B are alkali atoms. For all collision energies elastic collisions between the atoms and ions occur. Further, when the ion species is the same as the atom, resonant charge exchange (RCE) collisions also occur. We consolidate several of our previous experimental and numerical results on the collisional cooling of ions with atoms. The experiments are performed at collision energies above the temperature equivalent of 10 K. The ion cooling mechanisms are studied numerically and explain ion cooling consistently all the way upto mK equivalent collision energies. In the results discussed below, two cooling mechanisms manifest for this system. The first is that localized atoms at the ion trap centre will always cool trapped ions in collisions through elastic collisions. This is true even if the trapped ion has significantly lighter mass than the trapped atom, which is different from the commonly held views originating from the buffer gas cooling experiments and analysis. Further we demonstrate that the process of resonant charge exchange (RCE) is an extremely effective cooling mechanism for homonuclear atom-ion pairs at collisional energies equivalent to room temperature.

2. Two-body atom-ion interaction

In hybrid atom-ion traps figure 1, it is the low energy atom-ion collision, which is of interest. This implies that the long range part of the molecular ion potential plays a critical role in the collision. As the long range force is attractive, for an atom and ion in the ground state, the potential energy in this limit is $V(R)|_{R \gg R_e} \propto -\alpha_d/(2R^4)$, where R_e is the distance of minimum potential energy and α_d is the dipole polarizability. For excited states, the smallest contributing multipole will determine the long range interaction. As the collision energy decreases, lower partial waves contribute maximally to the collision. The total collision cross section for the polarizability interaction as a function of collision energy E , varies as $E^{-1/3}$ over a large range of collision energies with deviations at the few partial wave limit (very low energies) and at the very high energy end [15].

3. Collisional cooling of ions with a buffer gas

For a system of co-trapped atoms and ions, the ion number density is many orders of magnitude lower than atom number density enabling multiple collisions of each trapped ion with atoms. The collisional cooling of the trapped ions depends on how energy is exchanged between the ions and the atoms. The process of collisional cooling of ions with atoms is complex, because the ion is dynamically trapped in a Paul trap, which combines both static and oscillatory electric fields to fashion an effective trap for the charged particles. In such a trap, the trapping and energy of the ion post collision (PC) depends on how the PC velocity relates to the instantaneous rf field configuration. If the trapped ion maintains its synchronization PC with the field, it will

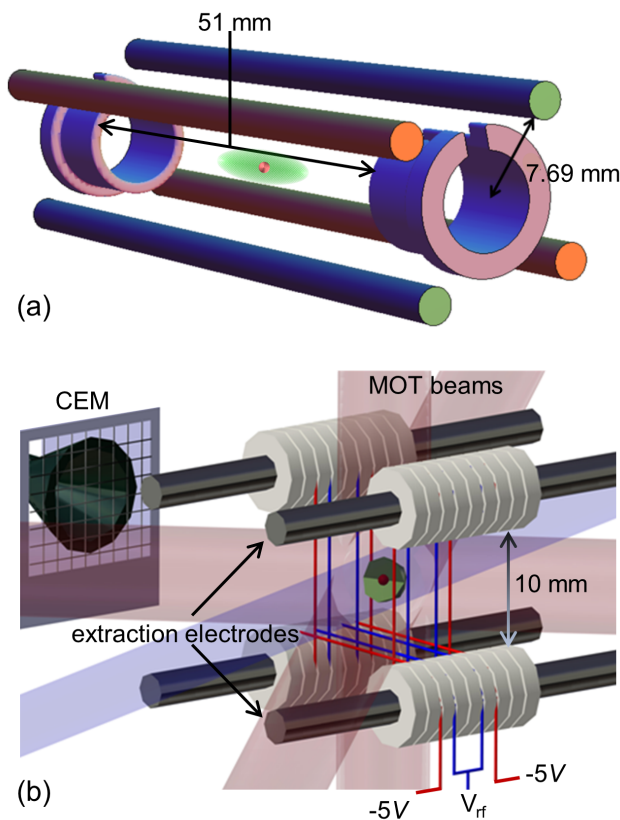


Figure 1. (a) Linear Paul trap with a MOT at the centre. The centred ellipsoid represents the volume within which the ion is trapped. Diagonal electrodes are equipotential and the two pairs of diagonal electrodes are 180° out of phase. The CEM is downstream from the end cap, along the trap axis. In (b) the spherical quadrupole trap with rf potentials on the two central wires and dc potentials on the outer pair of wires is shown. Grey represents the trapped ion volume extent and the dot in the centre is the MOT. Switching Voltage on the quadrupole rods extracts the trapped ions into the CEM, perpendicular to the trap.

stay trapped, but if the velocity and phase synchronization changes significantly, then the PC ion can heat and even exit the trap.

While buffer gas cooling makes relatively modest technical demands, there are subtleties in the cooling outcomes. When the direction of the ion velocity changes significantly per collision, after multiple collisions it will heat out of the ion trap, even if the atoms are at zero kinetic temperature. Kinematically, relatively large changes in the ion velocity will occur when the mass of the atom (m_A) is comparable to or greater than the mass of the ion (m_I). Indeed this is observed in many experiments [3], and therefore the practical operational condition for buffer gas ion cooling is when $m_A/m_I \ll 1$. Under these conditions the ion direction post collision suffers slight changes and so the ion remains trapped and cools rapidly. This gives rise to a critical mass ratio of the atom to ion mass, R_C above which it is theoretically impossible to cool a trapped ion with a buffer gas. Several theoretical analysis of trapped ion-atom collisions have arrived at different values of R_C ($= 1$ [3], 1.55 [16], 1.47 [17], 0.95 [18]) for buffer gas cooling (with our K^+ ion trap parameters).

4. Collisional cooling of ions in hybrid trap experiments: elastic collisions

Hybrid trap experiments differ from the buffer gas as the cooled and trapped atoms are tightly localized and co-centred with the ion trap, and so atom-ion collisions can occur only at the bottom of the ion trap. In the region around the trap centre, the trap field tends to zero and the motion of the ion approximates a force free trajectory near the trap centre. Under these circumstances, we experimentally show that ion cooling occurs irrespective of the mass of the atom. In particular, we show ion cooling when $m_A/m_I \geq R_C$ [12; 19].

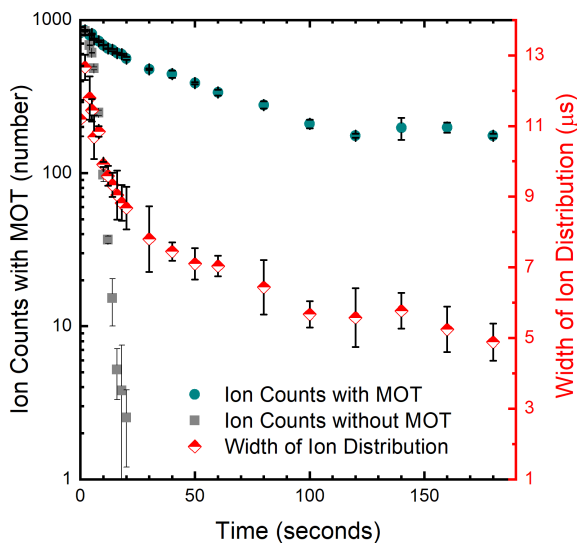


Figure 2. Rb^+ lifetime with MOT (blue, circle) and without MOT (grey, square) is shown as a function of time. The width of the extracted ion arrival time distribution is shown by the red diamonds referenced to the right vertical axis. Figure adapted from data in Ravi et. al. [12] with linear Paul trap of figure 1.

4.1. Cooling of ions by atoms of equal mass

In early experiments [12; 20], we showed that Rb^+ ions can be trapped for several minutes, in the presence of a Rb MOT. The laser cooled MOT atoms are precisely co-centred with the ion trap and the spatial extent of the MOT atoms is much less than of the trapped ions ($V_{\text{MOT}} \ll V_{\text{IT}}$). This is true of both the experimental geometries illustrated in figure 1, and results predominantly in atom-ion collisions proximate to the trap centre. In real traps, there is ion heating due to trap imperfections, which results in loss of the trapped ions. The lifetime of the trapped Rb^+ ion is extended dramatically in the presence of the Rb MOT (figure 2), whereas in the absence of the MOT the trapped ion population decays in the order of 10 seconds due to ion heating [12; 20]. Similar results were also seen in Na- Na^+ experiments [13]. The increase in lifetime is a signature of cooling of ions by atoms of equal mass. This observation is in departure from Major and Dehmelt's work [3], where $R_C = 1$, and thus the ion is expected to neither heat nor cool on the average, when in collision with the buffer gas atoms of equal mass.

The experiment is performed by creating the ^{85}Rb MOT, while the ion trap is ON. The ionizing light is switched ON briefly and then either the MOT is kept ON or switched OFF. The trapped ions are held for variable duration of time in each case and are extracted out of the ion trap to a channel electron multiplier (CEM) with appropriate electrostatic lensing [21] to determine the number of trapped ions at different hold times. The rapid loss of trapped ions in the absence of the MOT allows the determination of the trapped ion heating rate. The persistence of trapped ions at long times, created under the same conditions, but held in presence of the MOT, then allows the determination of a cooling rate for the trapped ions in collision with the localized and trapped atoms. Further, when the trapped ion number stabilized between two and three minutes of hold time [12], the arrival time distribution of ions counted by CEM pulses continued to shrink, allowing the determination of ion temperature by application of the virial theorem. These results show unambiguously that trapped ions of Rb^+ are very efficiently cooled by localized and centred Rb MOT atoms, which have equal mass. While classically the maximum transfer of energy in an elastic collision is for equal masses, most collisions are glancing and therefore, a new cooling mechanism was invoked to explain the extremely efficient cooling of Rb^+ . This mechanism is resonant charge exchange (RCE) for homonuclear atom-ion collisions. The discussion of cooling by RCE is deferred to section 5.

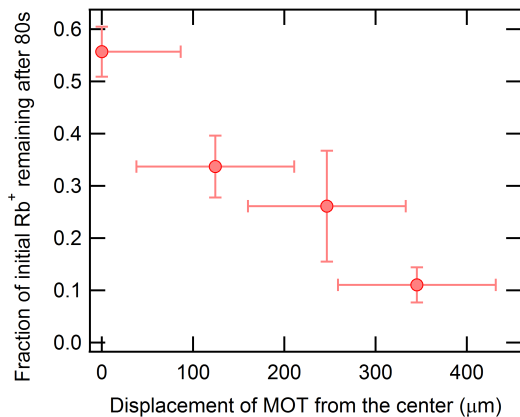


Figure 3. Rb^+ trapped fraction at a hold time of 80 s, as a function of displacement of the Rb MOT from the ion trap centre. The horizontal error bars are due to uncertainties in the determination of the precise displacement and MOT distortion when displaced.

In figure 3 we show how the precise centering of the MOT affects the trapped ion lifetime. This experiment has been performed in the spherical Paul trap shown in figure 1 (b). The MOT is displaced with respect to the ion trap centre, by shifting the magnetic centre for the MOT. The fraction of surviving Rb^+ ions is measured after a hold time of 80 seconds for different MOT displacement. The change in the effectiveness of the ion cooling is clearly seen in the dramatic fall in trapped ion lifetimes as a function of MOT displacement from the co-centred configuration. It is therefore standard experimental procedure to first centre the traps by maximizing the trapped ion lifetime and then take measurements.

4.2. Cooling of ions by atoms of higher mass

Given that $V_{\text{MOT}} \ll V_{\text{IT}}$ in hybrid traps where localized atoms are placed at the centre of the ion trap, the experiment to test the validity of the critical mass paradigm requires a heavier atom MOT with a lighter ion. In figure 4, we show cooling of: $^{85}\text{Rb}^+$ with ^{133}Cs MOT ($m_A/m_I = 1.565$), $^{133}\text{Cs}^+$ with ^{133}Cs MOT ($m_A/m_I = 1$) and $^{133}\text{Cs}^+$ by ^{85}Rb MOT ($m_A/m_I = 0.639$). These results are clearly consistent with cooling by localized and centred

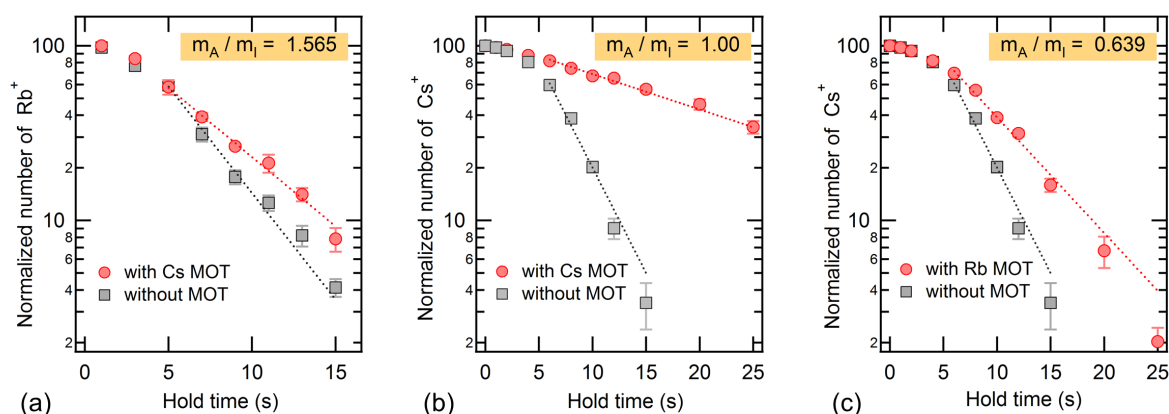


Figure 4. Panel (a) shows Rb^+ lifetime with (red, circle) and without (grey, square) Cs MOT. Panel (b) shows Cs^+ lifetime with (red, circle) and without (grey, square) Cs MOT. Panel (c) shows Cs^+ lifetime with (red, circle) and without (grey, square) Rb MOT. Figure adapted from data in Dutta et. al. [19] with linear Paul trap of figure 1. The equal mass case clearly differentiates from the unequal masses. In all cases, ion cooling is observed.

atoms $\forall m_A/m_I$, which stands it apart from buffer gas cooling [12; 22; 23].

In all two species experiments first a MOT is created, the ion trap is switched on and the MOT atoms are ionized from their excited states by briefly turning on a blue light, creating trapped ions. The MOT cooling light is turned off, which empties the first MOT within milliseconds (this instant is taken as $t = 0$). Then the MOT light for a different species is turned on (or not). The new MOT loads in 5 seconds to steady state. The trapped ions are held for a predetermined amount of time and then are extracted to a channel electron multiplier (CEM). The lifetimes of the trapped ions in the presence of and absence of MOT atoms is measured. The longer lifetime of ions in presence of MOT atoms indicate the cooling of ions by the MOT atoms.

To further check localized cooling, another experiment was performed with $^{39}\text{K}^+$ ions and a MOT of ^{85}Rb atoms, in the spherical Paul trap configuration of figure 1 (b). Once again, enhanced lifetimes were measured for the $^{39}\text{K}^+$ ion in the presence of the ^{85}Rb MOT as seen in figure 5(a), for an even higher ratio of $m_A/m_I = 2.179$. Here the cooling is present but less effective, than in the $^{85}\text{Rb}^+ - ^{133}\text{Cs}$ ion-atom case. The contribution of localization of cold atoms to ion cooling is tested by noting that displacement of the ion trap centre by even a few 100 microns from the ion trap centre completely eliminates the ion cooling. For the heteronuclear atom - ion experiments, no charge exchange collisions were seen in the control experiments.

4.3. Discussion

An intriguing observation is that while ion collisional cooling occurs, all the trapped ions eventually escape the trap, so there is significant heating of the trapped ion as well. This occurs because of the co-existence of two reservoirs of atoms with which the trapped ions collide and due to trap imperfection related losses. The localized cold atom reservoir cools the ion and the background vapour has the action of the buffer gas in the regular paradigm. In the case of $\text{Rb}^+ - \text{Cs}$ ($\text{K}^+ - \text{Rb}$) experiment, the dominant background gases are Rb and Cs (K and Rb) i.e. one of the background gases is heavier than the trapped ion, and therefore leads to heating of the ion. The result is that there is both collisional heating (due to background atoms) and cooling (due to MOT atoms) present simultaneously, which results in the eventual emptying of the ion trap. This is further understood in a simulation of the experiment shown in figure 5.

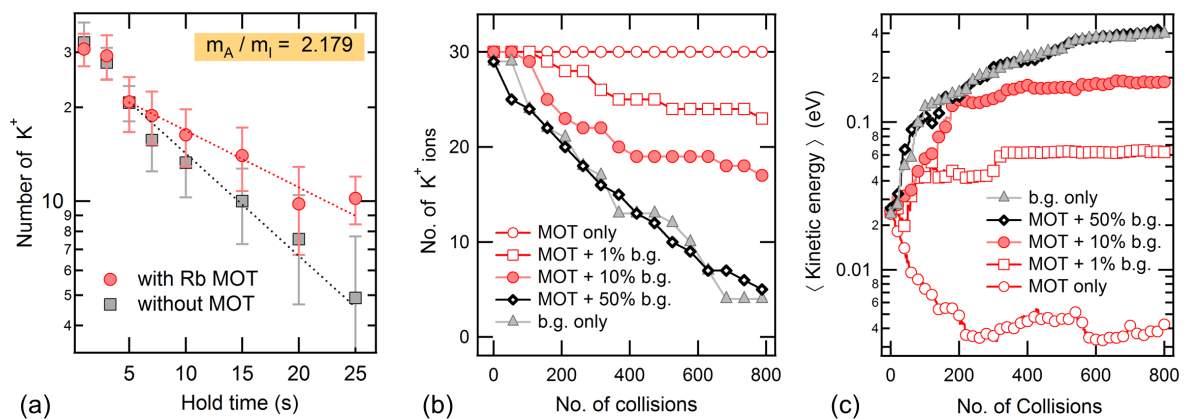


Figure 5. Experimentally measured K^+ ion lifetime with Rb MOT (red, circle) and without MOT (grey, square) is shown as a function of time in panel (a). The simulated lifetime of the same system as a function of number of ion-atom collisions is in (b) and the consequent heating or cooling is shown in (c). The background vapour plays a critical role in the eventual fate of the ion. Figure adapted from data in Dutta et. al. [19].

Here the trapped ion lives forever in the ideal trap when there is no heavy atom background. For any reasonable pressure of the higher mass atom, the ion trap empties out.

The important question which arises is, assuming a Gaussian density of cold atoms at the centre of a perfect ion trap, what is the final temperature and state of the ion, in the complete absence of the heavier mass background gas? Once again from simulations we can determine that, given a small but finite size of the cold atom distribution with full width at half maximum (FWHM) d , the final steady state temperature of a trapped ion will approximately be determined by the trap depth at a distance d even if the atoms are much colder than the kinetic temperature of the ion. This is so because at the centre of the ion trap, the ion effectively sees a buffer gas of heavier atoms, and only upon travelling radially outwards, the localization starts influencing the ion behaviour. The collisions at the trap centre do not heat the ion out of the trap as opposed to a uniform buffer gas which heats the ion in the outer reaches of the trap when the atom is heavier.

The ion can be put in thermal equilibrium with the heavy MOT atoms, if the ion trap depth at d is smaller than the atom temperature T_{MOT} . Under this situation the atom and ion can equilibrate, under the approximation of classical elastic collision, in the simulation.

5. Cooling of trapped ions by resonant charge exchange

We now focus on the homonuclear ion-atom collisions in figures 2 and 4 (b). In any collision, the probability of a head-on collision is negligible and a glancing collision is more likely. Thus irrespective of the condition of equal masses, the average (typical) elastic collision between ion and atom does not transfer much energy from the ion to the atom. The strength of the ion cooling, shown in figure 2 above, signals something else at work. The cooling by resonant charge exchange (RCE) was therefore proposed as a cooling mechanism. RCE is only active when the colliding atom-ion pair have a parent-daughter relation, i.e. the daughter ion is one electron removed from being the parent atom [3; 15]. In such a scenario, in a single glancing collision where little kinetic energy is exchanged, the electron from the cold atom can transfer to the incoming fast ion (which presents an identical ion core as that of the atomic nucleus), resulting in a fast atom leaving the trap and a very cold ion at the ion trap centre. Such cooling is called RCE or “swap” cooling and occurs when the atom and ion are identical species, since then the states available for the electron with the ion core do not extract an energy cost when the electron exchanges from atom to ion. While this mechanism was proposed and simulated to explain Rb^+ ion cooling with Rb MOT [12; 20], experimentally there was no discrimination between the elastic and RCE mechanisms.

To determine whether the RCE mechanism results in cooling of the trapped ion, we compare the collisional cooling of the Cs^+ ion with MOTs of Cs and Rb atoms. In the case of the Cs^+ -Cs homonuclear ion-atom pair RCE is possible along with elastic collisions. On the other hand, for Cs^+ -Rb the collisions are exclusively elastic. On considering only elastic collisions between the trapped Cs^+ ion and an atom of Rb and Cs, the cooling efficiency for the trapped Cs^+ ion with a localized Rb MOT is greater than for an identical Cs MOT.

To measure the cooling of trapped ions via RCE, we first load a Cs MOT, turn on the ion trap, ionize the MOT atoms as described earlier and then either (a) hold the Cs MOT, or (b) empty the Cs MOT and load the Rb MOT, followed by counting the ion number in the trap as a function of trap hold time in both cases. It can be seen easily in figure 6(a) that the same lifetime of trapped ions is achieved for a Cs MOT with 1/2 the peak atomic density and the same spatial volume as the Rb MOT. In addition, we also see (figure 6(b)) that the temporal width of the arrival time distribution of the extracted ions decreases in a nearly identical manner as the hold time increases in both these cases. This implies that the cooling rate for Cs^+ ions with Cs atoms k_{Cs} ($\equiv k_{Cs}^e + k_{Cs}^\times$), is equal to twice the Cs^+ cooling rate with Rb atoms k_{Rb} ($\equiv k_{Rb}^e$), i.e. k_{Cs} ($\equiv k_{Cs}^e + k_{Cs}^\times$) $\approx 2k_{Rb}^e$, where the superscript e and \times refer to elastic and RCE respectively.

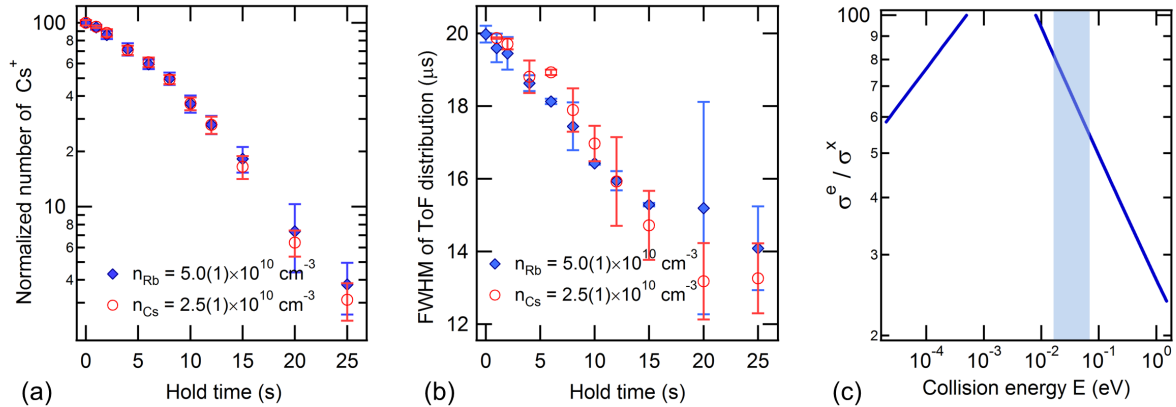


Figure 6. Panel (a) shows the Cs⁺ ion lifetime with Rb and Cs MOT, where the MOT number density is controlled such that the lifetimes are well matched. In panel (b) the corresponding temporal width of the arrival time distributions that are also well matched is shown. The ratio of the elastic to RCE cross sections in the energy range of the experiment, marked in blue is shown in (c). Figure adapted from data in Dutta and Rangwala [24].

At energies much larger than s-wave collision energies, the cross sections for elastic and resonant charge exchange can be treated independently to a good approximation. The ratio of the elastic (σ^e) and RCE (σ^\times) cross sections is illustrated in figure 6(c). Clearly in the energy range of operation (shaded region in figure 6(c)) of the present experiment $\sigma^e \gg \sigma^\times$. Identifying $\delta E_{Cs}^e (\delta E_{Rb}^e)$ as the effective energy lost by a Cs⁺ ion per elastic collision with a Cs (Rb) atom, we use the simulations of Höltkemeier et al. [23; 25] to estimate that for a cold uniform buffer gas $\delta E_{Cs}^e = 0.5\delta E_{Rb}^e$ and that in the limit of a very tightly confined MOT at the trap centre, $\delta E_{Cs}^e = 1.04\delta E_{Rb}^e$. The total cooling rates relate the cross sections and the energy lost per collision by the ion as [24]

$$\frac{\delta E_{Cs}^\times}{\delta E_{Cs}^e} = \frac{\sigma^e}{\sigma^\times} \times \frac{k_{Cs}^\times}{k_{Cs}^e}.$$

Given that $\sigma^e \gg \sigma^\times$, which implies that the elastic collision is much more probable than RCE, and $k_{Cs}^e \gg k_{Cs}^\times$, the experimental data in conjunction with above expression implies that $\delta E_{Cs}^\times \gg \delta E_{Cs}^e$ [24]. From figure 6 above, the ratio of the cooling efficiency between RCE and elastic collision was determined to be

$$39 \leq \frac{\delta E_{Cs}^\times}{\delta E_{Cs}^e} \leq 154.$$

The upper limit of the range represents the case of a uniform buffer gas and the lower limit denotes a spatially localized gas of infinitesimal extent at the trap centre. As the MOT in the experiment is of finite but small extent, the effectiveness of energy transfer in a single collision is contained within the range above. This underlines that ion cooling is very efficient when RCE is active.

6. Conclusions

Long held views on collisional cooling of trapped ions have been modified by the fact that in hybrid trap experiments, the location and distribution of the atomic cloud allows hitherto veiled mechanisms to manifest. Ion cooling by the localized and centred MOT uncovers that different parts of the reservoir of atoms have different effects on the energetics of the trapped and colliding

ion. It opens up the possibility of an ion as a probe for finite and small systems, by measurement of the ion properties. Remarkably whether the ion will attain a steady state or equilibrate with a gas of atoms turns out to be position and size dependent. The ubiquitous atom-ion mass ratios for ion cooling are no longer the discriminant between collisional heating and cooling of the ion. A new cooling mechanism by resonant charge exchange, which is a purely quantum and symmetry enabled exchange mechanism, manifests in a dramatic manner so as to dominate the otherwise primary cooling channel mediated by elastic collisions. Further developments prime this class of experiments as an important and new method for probing quantum systems of both ions and neutrals.

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