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Collisional Cooling of Light Ions by Cotrapped Heavy Atoms

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We experimentally demonstrate cooling of trapped ions by collisions with cotrapped, higher-mass neutral atoms. It is shown that the lighter $^{39}K^+$ ions, created by ionizing ^{39}K atoms in a magneto-optical trap (MOT), when trapped in an ion trap and subsequently allowed to cool by collisions with ultracold, heavier ^{85}Rb atoms in a MOT, exhibit a longer trap lifetime than without the localized ^{85}Rb MOT atoms. A similar cooling of trapped $^{85}Rb^+$ ions by ultracold ^{133}Cs atoms in a MOT is also demonstrated in a different experimental configuration to validate this mechanism of ion cooling by localized and centered ultracold neutral atoms. Our results suggest that the cooling of ions by localized cold atoms holds for any mass ratio, thereby enabling studies on a wider class of atom-ion systems irrespective of their masses.

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The cooling and trapping of dilute gases of ions [1,2] and atoms [3] has led to unprecedented precision in spectroscopy [4] and the study of interacting many-particle systems [5]. Cotrapping ions and atoms [6–14] widens the scope of inquiry to the two-particle $1/r^4$ asymptotic interaction. Among the different methods to cool trapped ions, cooling by elastic collisions with cold neutral atoms is arguably the most generic. Indeed, this has been extensively used in buffer gas cooling of relatively heavy ions by collisions with cold lower-mass neutral atoms. However, the complementary phenomenon, that of the cooling of low-mass ions by collisions with heavier neutral atoms, has never been demonstrated experimentally. This is partly because calculations show that trapped ions in a uniform buffer gas will heat up when the ratio of the atom mass (m_a) to the ion mass (m_i) exceeds a critical value [15–18]. In recent years, theoretical studies suggest a possible experimental resolution by using a localized ensemble of ultracold neutral atoms placed precisely at the center of the ion trap [12,19].

In this Letter, we experimentally demonstrate the cooling of low-mass $^{39}K^+$ ions in a Paul trap by localized, heavier ^{85}Rb atoms in a magneto-optical trap (MOT) and validate our experimental results with numerical simulations. We also demonstrate the cooling of trapped $^{85}Rb^+$ ions by ultracold ^{133}Cs atoms in a MOT. The cooling manifests itself through an increase in the lifetime of the trapped ions. Our results support the possibility that the cooling of ions by localized cold atoms may hold for any mass ratio, thereby enabling studies on the other half of ultracold atom-ion systems irrespective of their masses.

The atom-ion mass ratios ($m_a/m_i = 2.179$ for $^{85}Rb - {}^{39}K^+$ and $= 1.565$ for $^{133}Cs - {}^{85}Rb^+$) we demonstrate cooling for exceed the critical mass ratios (CMRs) [15–18] beyond which ion heating is predicted for uniform atomic gas densities. The experiment is consistent with our Monte Carlo (MC) simulations and other theoretical models [19] that consider collisions with centrally localized density (as opposed to uniform density) of cold atoms. We further demonstrate the competing ion heating mechanism due to collisions with the background gas vapor. The dependence of the steady state ion temperature on the size of the cold atomic cloud is also established numerically. The present work lays the foundation for studies on a wider class of sympathetically cooled ion-atom mixtures, and the resulting clarity may enable the realization of the few-partial-wave regime.

Trapping and cooling ions and atoms.—The conditions and mechanisms for trapping atoms and ions are different, because the atom is neutral and the ion charged. Atom traps typically have small trap depths and use a combination of static magnetic and/or optical fields. Ions, on the other hand, are trapped in Penning or Paul traps [20], of which the latter, a dynamic trap with time-varying electric fields, is compatible [15] with overlapped cold atom ensembles, because the perturbation of the Paul trap operation on cotrapped cold atoms is negligible [6,21]. It is for this reason that hybrid traps combine Paul traps for ions [1] with atom traps such as a MOT [7,10–13], magnetic trap [8], or optical dipole trap [8,9,22]. However, due to the dynamic trapping of ions in a Paul trap [1,20], the trapped ions in the absence of laser cooling are more energetic than the atoms [12,13].

Signature of the cooling of trapped ions.—The motion of the dynamically trapped ions is described theoretically by the Mathieu equations, which are parameterized in terms of dimensionless a and q parameters, the values of which characterize the ion trap operation. The ion motion is separable [1] into (i) the micromotion, which is synchronous with the time-varying radio frequency (rf) field and (ii) the secular motion (macromotion), which is the low-frequency oscillation of the ion in the effective trapping potential that the ions experience in the dynamic trapping fields. The micromotion is position dependent, while the secular motion determines the size of the trapped ion's orbit

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and characterizes the trapped ion's temperature. The smaller the trapped ion's orbit, the lower the ion's temperature, and therefore the task of cooling the ion is reduced to arresting the amplitude of the secular motion.

Criteria for ion heating and cooling.—The pioneering work on collisional cooling of ions by neutral atoms, by Major and Dehmelt [15], considers the ion trap volume within a constant density of buffer gas atoms. In this case, it was found that ions collisionally heat when $m_a > m_i$, cool when $m_a < m_i$, and do not change temperature when $m_a = m_i$. The ratio $m_a/m_i = 1$ is then the theoretical CMR differentiating the cooling and heating regimes. In practice, the cooling by a buffer gas has found extensive applications when $m_a \ll m_i$ [23].

In recent years, several independent theoretical analyses for ion cooling by neutral atoms have arrived at limiting CMRs [16–18] that are different from 1 (CMR $= 1.55$ [16], $= 1.47$ [17], and $= 0.95$ [18] for our K^+ ion trap parameters). Exceeding these CMRs results in the heating of ions (and below these cooling occurs) if the ion is assumed to be located within a cold gas of uniform density. However, in hybrid ion-atom traps, the atomic density is localized, for example, in a MOT, and is located at the center of the ion trap [see Figs. 1(a) and 1(b)]. When this localized and precisely centered nature of the atomic density profile is taken into account, the trapped ions are expected to be cooled by elastic collisions with the ultracold atoms irrespective of the atom-ion mass ratio [12,17,19]. At the center of the ion trap, the ion's micromotion is negligible, while the ion's secular speed is the greatest—thus, a collision with an ultracold atom, that is essentially at rest, always results in a reduction in the ion's secular motion and hence in the cooling of the ion [see Figs. 1(b) and 1(c)].

Experimental procedure.—The experimental apparatus is described over several articles [24–27] and is schematically illustrated in Figs. 1(a)–1(c). The ion trap has a depth of $U \approx 0.5$ eV, $q = 0.42$, and $a \approx 0$. For this experiment, we have a vapor-loaded dual MOT of ^{39}K and ^{85}Rb in operation. The ^{39}K and ^{85}Rb MOTs are very well overlapped with the ion trap center, and so the ions created from the MOT are efficiently loaded into the ion trap. The loading of the two MOTs is independently controlled by mechanical shutters placed in the paths of the respective laser beams. The $^{39}K^+$ ions are created by resonant two-photon ionization of atoms in the ^{39}K MOT, where the first photon (cooling laser) results in the $4S_{1/2} \rightarrow 4P_{3/2}$ excitation at 767 nm and a second photon is sourced from a collimated light-emitting diode (LED) with the emission centered at 456 nm. Further experimental details are provided in Supplemental Material [28].

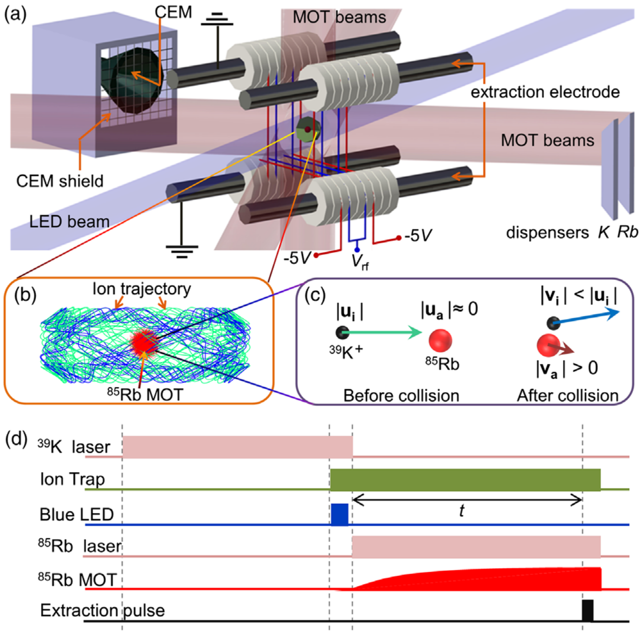

FIG. 1. (a) A schematic of the experimental setup. The rf voltage $V_{\rm rf}$ is fed to the two central wire electrodes, and the two external wire electrodes serve as the end caps. The extraction of the trapped ions to the CEM is done by biasing two of the rod extraction electrodes appropriately after the hold time. (b) Details of overlap volumes of the ^{85}Rb MOT and a calculated, representative ion trajectory. (c) Illustrative diagram for a transfer of momentum from the $^{39}K^+$ ion to the almost stationary ^{85}Rb MOT atom, resulting in a lower kinetic energy of the ion postcollision and a hot atom which usually escapes the MOT. $|\mathbf{u_i}|$ ($|\mathbf{u_a}|$) and $|\mathbf{v_i}|$ ($|\mathbf{v_a}|$) represent the magnitude of the ion (atom) velocity before and after a collision, respectively. (d) The timing sequence for the experiment.

The experiment measures the lifetime of the trapped $^{39}K^+$ ions when held with and without the ^{85}Rb MOT atoms. The experimental sequence is illustrated in Fig. 1(d). As the LED light can also ionize ^{85}Rb MOT atoms in a similar two-photon process, the MOTs are operated sequentially to prevent the ionization of ^{85}Rb atoms. First, the ^{39}K MOT is loaded, ion-trapping fields are switched on, $^{39}K^+$ ions are created by switching on the blue LED briefly, and the ^{39}K MOT light is shuttered off, which empties the ^{39}K MOT. Subsequently, either the ^{85}Rb MOT light is allowed in and the ^{85}Rb MOT loaded or the ^{85}Rb light is kept blocked and no MOT is loaded. The trapped $^{39}K^+$ ions are held for variable hold times and extracted for detection by a channel electron multiplier (CEM), where individual ion arrivals register as individual ~8 ns pulses [24,25]. These pulses are counted, and the surviving ion number in the ion trap determined as a function of the ion hold time (t), in the presence or absence of the ^{85}Rb MOT.

Potassium ion cooling by rubidium atoms.—In Fig. 2(a), we present a representative plot showing that a larger number of $^{39}K^+$ ions survive when held in the presence of the ^{85}Rb MOT. As the ^{85}Rb MOT takes ~5 s to load, initially there is no significant difference between the number of $^{39}K^+$ ions counted when ^{85}Rb MOT is off (square) or on (circle), but by 25 s a clear separation in the

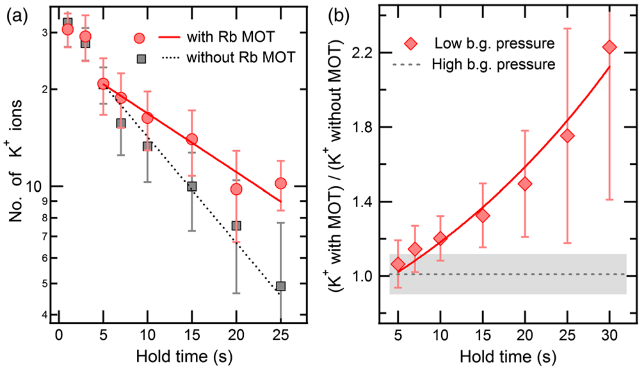

FIG. 2. (a) The number of $^{39}K^+$ ions remaining in the ion trap for different values of the hold time either in the absence (squares) or the presence (circles) of the ^{85}Rb MOT. The dotted and the solid lines are single exponential fits for the respective cases. The increase in the survival probability of trapped $^{39}K^+$ ions in the presence of the ^{85}Rb MOT indicates the cooling of $^{39}K^+$ ions. The error bars represent the width (1 s.d.) of the underlying ion number distribution. (b) The ratio between the number of trapped $^{39}K^+$ ions in the presence and absence of the ^{85}Rb MOT, for low ($\leq 5.9 \times 10^{-10}$ Torr) background partial pressure of Rb (symbols) and for high ($\geq 8.3 \times 10^{-10}$ Torr) background partial pressure of Rb (dotted line). The shaded region represents 1 s.d. for the measurements at high background pressure. At a high background pressure, the cooling of $^{39}K^+$ ions (indicated by ratios > 1) is not experimentally discernible. The solid line is a fit to an exponential for the low background pressure case. The quoted partial pressure of Rb was obtained from the loading rate of the ^{85}Rb MOT [29].

number of surviving ions emerges. The larger survival probability of the trapped $^{39}K^+$ ions in the presence of the ^{85}Rb MOT can be explained only by the cooling of the trapped ions by the ^{85}Rb MOT atoms [12,13]. If, instead, the ions were heating, the survival probability for $^{39}K^+$ ions would be lower when in contact with ^{85}Rb MOT atoms. The experiments were repeated with $^{40}K^+$ ions instead of $^{39}K^+$ ions, and cooling of $^{40}K^+$ ions by ^{85}Rb atoms was also observed. The cooling of $^{39}K^+$ ions occurs because the ultracold ^{85}Rb atoms are localized precisely at the center of the ion trap, and $V_a \ll V_i$, where V_a (V_i) is the volume of the trapped atoms (ions).

The experiment in Fig. 2(a) is repeated nine times—in each case, the ion decay is relatively fast up to $t \sim 5$ s, and two cases, with and without a MOT, are indistinguishable, consistent with the ^{85}Rb MOT loading time. For $t \geq 5$ s, the number of ions $N(t)$ in both cases decays exponentially but with different lifetimes. We fit a single exponential decay of the form $N(t) = N_i e^{-k(t-5)}$ to obtain the ion loss rate without a MOT (k_1) and with a MOT (k_2) for each of the nine data sets. The mean values from the nine experimental runs are $k_1 = 0.089(0.014)\ \mathrm{s}^{-1}$ and $k_2 = 0.056(0.018)\ \mathrm{s}^{-1}$, where the values in the parentheses are the standard deviation (s.d.) of means. The corresponding lifetimes without a MOT and with a MOT are $\tau_1 = 11.2^{+2.2}_{-1.6}$ s and $\tau_2 = 17.8^{+8.2}_{-4.3}$ s, respectively. The increase in the ion lifetime is due to the cooling of $^{39}K^+$ ions by the ultracold ^{85}Rb atoms in the MOT.

In Fig. 2(b), we plot, for $t \geq 5$ s, the ratio between the number of trapped $^{39}K^+$ ions in the presence and absence of the ^{85}Rb MOT, obtained by averaging over the nine experimental runs. The ratio increases with increasing t, and a fit to an expression of the form $Ae^{k_{\mathrm{eff}}(t-5)}$ yields $A = 1.02(\pm 0.04)$ and $k_{\mathrm{eff}} = 0.0293(\pm 0.0024)\ \mathrm{s}^{-1}$. The agreement between k_{eff} and $(k_1 - k_2)$ provides a consistency check on the fits and ascertains the observed increase in ion lifetime. The ion temperature without and with a MOT increases approximately at rates $\approx k_1 U$ and $\approx k_2 U$, respectively, and thus the estimated net cooling rate (R) due to the presence of the MOT is $R \approx k_{\mathrm{eff}}\ U \approx 14.7 \pm 1.2$ meV/s (see Supplemental Material [28] for an ion cooling model).

In addition to the ^{85}Rb MOT, there is the presence of the background gas (b.g.) of Rb, trace amounts of K, and other unidentified gases in our chamber. We observe that the K^+ ion cooling is suppressed [Fig. 2(b)], when performing the same experiment at an elevated b.g. pressure (obtained by operating the Rb dispenser at higher currents), while adjusting the MOT parameters such that the MOT numbers and density are approximately the same. The Rb background vapor contributes to the heating of the $^{39}K^+$ ions consistent with ion heating by uniform buffer gas atoms of higher mass as predicted initially by Major and Dehmelt [15] and recently by others [16–18].

We note that, in earlier experiments with $^{85}Rb^+$ and ^{85}Rb [12,24], the cooling of ions was faster, because the ions and atoms were of the same mass, for which collisional cooling is more efficient, and, in addition, resonant charge exchange (RCE) might also have been active [30,31]. Since K and Rb are different species, RCE is not possible, but nonresonant charge exchange (nRCE) is. However, careful experimental tests allow us to conclude that this channel is too weak [32] and is not detected in our experiment (see Supplemental Material [28]). Therefore, the dominant cooling channel is multiple elastic collisions, which reduces the cooling rate of the $^{39}K^+$ ions by the ^{85}Rb MOT as compared to the cooling of $^{85}Rb^+$ by ^{85}Rb [12,24]. The essence of the present experiment is that, in the ideal situation with no background gas, a spatially small MOT at the precise center of the ion trap would always cool a trapped ion via elastic collisions, irrespective of the ion-atom mass ratio but at different rates.

Rubidium ion cooling by cesium atoms.—To further validate our results, we perform another experiment in an entirely different experimental apparatus consisting of a linear Paul trap for $^{85}Rb^+$ ions [12,21] and a MOT for ultracold ^{133}Cs atoms (see Supplemental Material [28]). To demonstrate cooling of $^{85}Rb^+$ ions by ^{133}Cs atoms, we follow an experimental sequence similar to Fig. 1(d) except

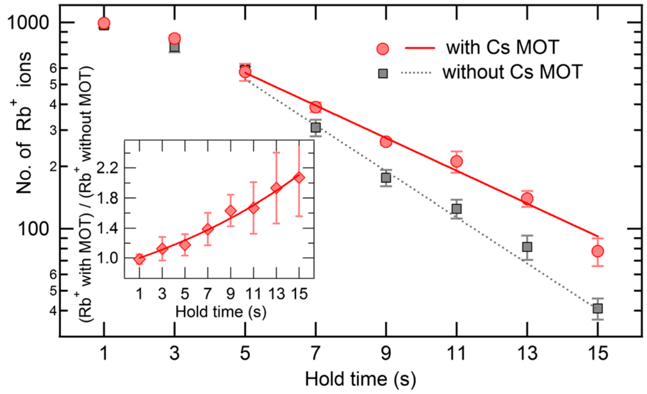

FIG. 3. The number of ^{85}Rb^{+} ions remaining in the linear Paul trap for different values of the hold time either in the absence (squares) or the presence (circles) of the ^{133}Cs MOT. The dotted and the solid lines are single exponential fits for the respective cases. The error bars represent the width (1 s.d.) of the underlying ion number distribution. Inset: The ratio between the number of surviving ^{85}Rb^{+} ions in the presence and absence of the ^{133}Cs MOT.

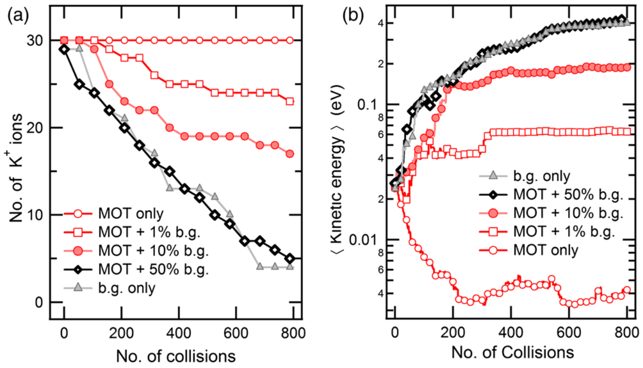

FIG. 4. (a) The number of surviving ^{39}K^{+} ions in the simulation as a function of the number of collisions each ion experiences. The ions collide with both the background gas of Rb atoms and the ^{85}Rb MOT atoms. At high background pressures, the ion losses with and without a MOT are the same, as seen experimentally in Fig. 2(b). (b) The mean kinetic energy of the ions for the above simulations. As the background gas density is reduced, cooling due to the MOT overcomes the heating due to background vapor and therefore lowers the ion kinetic energy.

that the first, fourth, and fifth rows of the figure now are a ^{85}Rb laser, ^{133}Cs laser, and ^{133}Cs MOT, respectively. The result of the experiment is shown in Fig. 3—a larger number of ^{85}Rb^{+} ions survive when held in the presence of the ^{133}Cs MOT. This can be explained only by the cooling of ^{85}Rb^{+} ions due to ultracold ^{133}Cs atoms. The inset in Fig. 3 shows the ratio between the number of surviving ^{85}Rb^{+} ions in the presence and the absence of the ^{133}Cs MOT, obtained by averaging over ten experimental runs. The ratio increases with increasing t, and a fit to an expression of the form $Ae^{k_{\mathrm{eff}}(t-1)}$ yields $A = 1.00(\pm 0.03)$ and $k_{\mathrm{eff}} = 0.053(\pm 0.003)\ \mathrm{s}^{-1}$. This value of k_{eff} is much greater than that for the cooling of ^{39}K^{+} ions by ^{85}Rb atoms shown earlier. This is partly because the atom-ion mass ratio (1.565) in this case is comparatively lower, resulting in faster cooling compared to the ^{39}K^{+} ion–^{85}Rb atom case. Notably, for long hold times, a significant reduction in the width of the ^{85}Rb^{+} ion arrival time distribution is observed when the ^{133}Cs MOT is present [e.g., at 13 s (width with a MOT)/(width without a MOT) $= 0.91 \pm 0.01$], providing independent evidence for the cooling of ^{85}Rb^{+} ions by the ^{133}Cs atoms. Furthermore, since this experiment is done in a different kind of ion trap, the ion cooling seems to be robust against changes in ion trap parameters. For brevity, in the rest of the Letter, we focus our discussion on the cooling of ^{39}K^{+} ions by ^{85}Rb atoms.

Simulations.—We use a MC algorithm and solve the equations of motion to track how the ions collisionally cool (see Supplemental Material [28]). In Fig. 4(a), we plot the lifetime of 30 noninteracting ^{39}K^{+} ions in the presence of a ^{85}Rb MOT of FWHM $d = 235\ \mu$m, whose atoms are at zero velocity, and a b.g. of Rb, whose velocity distribution is consistent with room temperature. As the density of the b.g. decreases with respect to the MOT peak density, the effective cooling rate increases, and the ion survives longer. When the b.g. density is zero, the ions are held in perpetuity. Figure 4(b) illustrates the competition between the cooling and the heating of the ion, for the cases in Fig. 4(a), and shows cooling to a steady state temperature when background vapor of Rb is absent. In the absence of a b.g., the steady state temperature T_s of the ions is proportional to d^2 for given trap parameters. In Supplemental Material [28], we discuss the simulations in greater details and show that ions are cooled with localized and precisely centered ensembles of atoms irrespective of the atom-ion mass ratio.

Conclusion.—The experiments demonstrate the collisional cooling of light ions by heavier atoms. The reported result is also a demonstration in support of no critical mass ratio for the cooling of ions with atoms, in hybrid traps. The net ion cooling rate can be increased by loading the MOT from a Zeeman slower, a pulsed atomic source, or by using a liquid nitrogen cold finger, all of which will reduce the background gas pressure. Judicious choice of ion-atom species, MOT spatial extent, and the secular trap depth of the ion trap could allow a single trapped ion to be cooled to very low temperatures, where possibly the low partial wave regime of ion-atom collisions can be attained at least momentarily [33,34]. Our demonstration also opens up the possibility of simultaneously trapping multiple ionic species with an ensemble of ultracold atomic gas, irrespective of the mass ratio with applications in cold ion chemistry and laboratory astrophysics [35] with trapped protons, H_2^{+}, or HD^{+} collisionally cooled with ^{6}Li.

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