Micromotion-induced Limit to Atom-Ion Sympathetic Cooling in Paul Traps

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We present and derive analytic expressions for a fundamental limit to the sympathetic cooling of ions in radio-frequency traps using cold atoms. The limit arises from the work done by the trap electric field during a long-range ion-atom collision and applies even to cooling by a zero-temperature atomic gas in a perfectly compensated trap. We conclude that in current experimental implementations this collisional heating prevents access to the regimes of single-partial-wave atom-ion interaction or quantized ion motion. We determine conditions on the atom-ion mass ratio and on the trap parameters for reaching the s-wave collision regime and the trap ground state.

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The combination of cold trapped ions and atoms [1–8] constitutes an emerging field that offers hitherto unexplored possibilities for the study of quantum gases. New proposed phenomena and tools include sympathetic cooling to ultracold temperatures [3–10], charge transport in a cold atomic gas [11, 12], dressed ion-atom states [13–16], local high-resolution probes [17–18], and ion-atom quantum gates [19, 20].

In contrast to atom traps based on conservative forces, Paul traps employ radiofrequency (RF) electric fields to create a time-averaged secular trapping potential for the ion [21]. The time-varying field can pump energy into the system if the ion’s driven motion is disturbed, e.g., by a collision with an atom [22]. The kinetic behavior of an ion in a neutral buffer gas has been observed in numerous experiments [23–30]. The ion’s equilibrium energy distribution was predicted analytically [31], as well as using Monte-Carlo techniques [26, 27, 32–33], and recently a quantum mechanical analysis has been performed [34]. For atom-ion mass ratios below a critical value, the ion is predicted to acquire a stationary non-thermal energy distribution with a characteristic width set by the coolant temperature [33].

The RF field drives micromotion of the ion at the RF frequency. At any position and time, the ion’s velocity can be decomposed into the micromotion velocity and the remaining velocity of the secular motion. Consider the simple case of a sudden collision with an atom that brings the ion to rest, a process which in a conservative trap would remove all kinetic energy. Immediately after such a collision the ion’s secular velocity is equal and opposite to the micromotion velocity at the time of the collision. This implies that the ion’s secular motion can be increased even in a collision that brings it momentarily to rest. Hence for cooling by an ultracold atomic gas [3, 4], the energy scale of the problem is no longer set by the atoms’ temperature but by the residual RF motion of the ion, caused, e.g., by phase errors of the rf drive or by dc electric fields which displace the trap minimum from the rf node [35]. Such technical imperfections have limited sympathetic cooling so far, with the lowest inferred ion temperature on the order of 0.5 mK [7]. Full quantum control in these systems [19, 20], on the other hand, likely requires access to the smaller temperature scales $\hbar \omega/k_B \sim 50\mu K$ for the trap zero-point motion and $E_s/k_B \sim 50$ nK for the s-wave collision threshold [11].

In this Letter, we show that even with the atomic gas at zero temperature and in a perfectly dc- and rf-compensated Paul trap, a fundamental limit to sympathetic atom-ion cooling arises from the electric field of the atom when polarized by the ion, or equivalently, the long-range ion-atom interaction. As the atom approaches, it displaces the ion from the RF node leading to micromotion, whose interruption causes heating. A second non-conservative process arises from the non-adiabatic motion of the ion relative to the RF field due to the long-range atom-ion potential; during this time, the trap can do work on the ion and increase its total energy. We find that, in realistic traps, the work done by the RF field dominates the effect of the sudden interruption of the ion’s micromotion and leads to an equilibrium energy scale that, for all but the lightest atoms and heaviest ions, substantially exceeds both the s-wave threshold $E_s$ and the trap vibration energy $\hbar \omega$. Our analysis shows that current atom-ion experiments [1–8] will be confined to the regimes of multiple partial waves and vibration quanta, and indicates how to choose particle masses and trap parameters in order to achieve full quantum control in future experiments. Our analytical results are supported by numerical calculations that furthermore reveal that in those collisions where the RF field removes energy from the system, the atom becomes loosely bound to the ion, leading to multiple subsequent close-range collisions until enough energy is absorbed from the RF field to eject the atom and heat the ion.

We consider a classical model and later confirm that this assumption is self-consistent, i.e. that the energies obtained from the model are consistent with a classical description. An atom of mass $m_a$ approaches from infinity to an ion of mass $m_i$ held stationary in the center of

\[ \frac{m_a}{m_i} \]
an RF quadrupole trap. At sufficiently low collision energies, the angular-momentum barrier will be located far from the collision point, and, once it is passed, the collision trajectory will be nearly a straight line. We initially assume this trajectory to be along an eigenaxis of the RF trap, resulting in a true 1D collision in a total potential given by $V(r_i, r_a, t) = e^2 C_4 |r_i - r_a|^2 + U(r)$, where $e$ is the ion’s charge, $r_i$ and $r_a$ are the ion and atom locations, respectively, $r = r_i - r_a$ is the ion-atom distance, and $\mathcal{E}(r_i, t) = gr_i \cos (\Omega t + \phi)$ is the RF electric field of the ion trap at frequency $\Omega$, parameterized by its quadrupole strength $q$. The ion-atom interaction potential at large distances is given by $U(r) = -C_4 / (2r^4)$ with $C_4$ the atom’s polarizability, and modeled as a hard-core repulsion at some small distance. Since in a three-dimensional collision the atom can approach the ion along directions that are perpendicular to the RF field or where the RF field vanishes, we expect our analysis to overestimate the heating by a factor of order unity, which we address below.

As the ion is pulled from the trap origin by the long-range interaction $U$ with the approaching atom, the oscillating electric field $\mathcal{E}$ causes it to execute sinusoidal micromotion with amplitude $gr_i / 2$, where $q = 2eg / (m_i \Omega^2)$ is the unitless trap Mathieu parameter. As long as the motion of the ion relative to the atom remains slow relative to the average RF micromotion velocity $v_{jm}$, the ion’s equations of motion during one RF cycle will remain linear, and the secular motion of the ion during each RF cycle can be described in terms of an effective conservative secular potential $V_s = \frac{1}{2} m_i \omega_i^2 r_i^2 + U(r)$, where $\omega = q \Omega / 2^{3/2}$ is the secular frequency of the ion within the trapping potential. Associated with $V_s$ are the characteristic length scale $R = C_4^{1/6} / (m_i \omega_i^2)^{1/6}$ at which the interaction potential $U$ is equal in magnitude to the trap harmonic potential, time scale $T = 2\pi / \omega$, and energy scale $E_{R} = \frac{1}{2} m_i \omega_i^2 R^2 = \frac{1}{2} (m_i^2 \omega_i^4 C_4)^{1/3}$.

In collisions with light atoms, $m_a < m_i$, we expect the ion to stay confined close to the trap origin. In this case, the ion-atom distance $r(t)$ is governed solely by the motion of the atom in the ion-atom potential $U$,

$$ r(t) \approx (9C_4 / \mu)^{1/6} |t|^{1/3}, $$

where the collision occurs at time $t = 0$ and $\mu = m_m m_a / (m_i + m_a)$ is the reduced mass of the system. The displacement $r_c$ of the ion from the center of the trap at the time of the hard-core collision with the atom can then be estimated by integrating the effect of the force $2C_4 / r^5$ exerted by the atom on the ion trapped in its secular potential, yielding $r_c \approx 1.11 \left( m_a / m_i \right)^{5/6} R$. In collisions with heavy atoms ($m_i < m_a$) on the other hand, the ion responds quickly to minimize the total secular potential energy until, at $(r_i, r_a) = (0.29, 1.76) R$, the deformed ion’s equilibrium position becomes unstable and the light ion quickly falls towards the atom with the collision occurring at the ion displacement $r_c \approx 1.76 R$. In general, we may express the collision point as $r_c / R = r_c (q, m_a / m_i) / (1 + m_i / m_a)^{5/6}$ where $1 < r_c < 2$.

Around the collision point $r_c$, we may expect the energy of the system to change as long as the ion moves non-adiabatically relative to the RF field, including the interval $-t_3 < 0 < t_3$ around the collision at $t = 0$, during which the ion’s velocity $\dot{r}_i$ is greater than its average micromotion velocity $v_{jm} \approx \omega r_c$. In this regime, the trap RF field can be thought of as a time-dependent perturbation to the ion-atom potential, doing work on the ion.
equal to

\[ W = e \int_{r_1}^{r_2} \mathcal{U}(r_i(t), t) \cdot \dot{r}_i(t) \, dt. \]  

(2)

The change in the system’s energy will depend on RF phase \( \phi \) at the time of the close-range ion-atom collision, taking on the maximal value \( W_{\text{max}} \) for \( \phi = \phi_{\text{max}} \). For \( \dot{r}_i \gg v_{\text{ium}} \), we may neglect the effect of the electric field on the ion’s trajectory and approximate the ion’s position in terms of the free collision trajectory \( r(t) \). Eq. (1) as \( r_i \approx r_c - m_i \tau/(m_i + m_a) \). In this case, the work done by the RF field can be written as \( W \approx K \sin \phi \), where

\[ K = W_0 \int_{-\Omega t_1}^{\Omega t_1} \left[ \frac{\sqrt{2r_c}}{(3|\tau|)^{2/3}} - \frac{q^{1/3}}{(3|\tau|)^{1/3}} \right] \sin |\tau| \, d\tau \]  

(3)

and

\[ W_0 = 2 \left( \frac{m_a}{m_i + m_a} \right)^{5/3} \left( \frac{m_i^2 \omega^4 C_4}{q^2} \right)^{1/3} \]  

(4)

is the characteristic scale for the work done on the ion by the RF field. The maximal energy gain \( W_{\text{max}} \approx K \) occurs for \( \phi = \phi_{\text{max}} \approx \pi/2 \), corresponding to the RF field changing sign at the time of the collision. The non-adiabatic condition \( |\dot{r}_i| > |v_{\text{ium}}| \) is equivalent to \( 3q\Omega t < (2/r_c)^{3/2} \), which, for the practically relevant values of \( q < 0.5 \), will always include the region \( |\Omega t| < 0.8 \) where the dominant contribution to the integral in (3) occurs. Consequently, we may extend the limits of integration to \( \pm \infty \) to obtain \( K/W_0 \approx 1.82r_c - 1.63q^{1/3} \), implying \( 0.7 < K/W_0 < 2.8 \) for \( q < 0.5 \) and at all mass ratios. Under the same conditions, \( K \) is at least three times larger than the ion’s average micromotion energy at the collision point \( E_{\text{ium}} \approx m_i v_{\text{ium}}^2/2 \) and the gradual energy change (2) dominates the effect of the sudden interruption of the ion’s micromotion. Intuitively, at low collision energies, the ion-atom potential dominates for a longer time during which the RF field does more work on the system. Since the trap electric field must be increased for higher RF frequencies to preserve the ion secular potential, the heating increases with a decrease in the Mathieu parameter \( (K > 15 \times E_{\text{ium}} \text{ for } q < 0.1) \).

Since \( W \) corresponds to the difference in the work done by the RF field during the incoming and outgoing parts of the collision, the energy change will depend on the phase \( \phi \) of the RF field at the time of the hard-core collision. For \( 0 < \phi < \pi \), the RF field accelerates the collision partners towards each other, increasing the system energy; for \( \pi < \phi < 2\pi \), the RF field opposes the collision, doing negative work and causing the atom to be bound to the ion with binding energy on the order of \( -W_0 \) (Figure 1). Since the \( r^{-4} \) potential does not possess stable orbits, bound ion-atom trajectories will include further close-range collisions. Depending on the RF phase during each subsequent collision, the system will gain or lose energy on the order of \( W_0 \), leading to a random walk in energy until the atom finally unbinds. Then \( \Delta E \) depends sensitively on the RF phases at each hard-core collision spaced in time by many RF cycles, leading to a sharp dependence of \( \Delta E \) on the RF phase \( \phi \) during the initial collision for \( \pi < \phi < 2\pi \) (Figure 2). Using the approach below, we calculate the net average energy gain for \( 0 < \phi < 2\pi \) and \( m_a/m_i = 1/2 \), \( q = 0.1 \) below as \( (\Delta E) = 1.0W_0 \).

To quantitatively verify the above heating model, we numerically calculated classical trajectories of low-energy 1D collisions as a function of \( \phi \), \( m_a/m_i \), and \( m_a/m_i \) (Figures 2 and 3). The ion was initially held at the trap center while the atom followed the analytic trajectory (1). At
a critical ion-atom distance \( r_0 \), the ion was displaced so that the trap’s secular force would balance the atom’s attraction, while keeping its velocity zero. Choosing \( r_0 = 3.8 (1 + m_i/m_a)^{1/3} R \) ensured that for all the trap parameters in this paper, the ion’s initial micromotion energy was smaller than \( 10^{-3} W_0 \) and both the atom’s kinetic energy and \( U (r_0) \) were smaller than \( 10^{-2} W_0 \). The equations of motion were integrated using the Dormand-Prince explicit Runge-Kutta method: away from collision points, the motion was integrated as a function of time while near the collisions, the ion-atom distance \( r \) was used with a hard-core radius \( \epsilon = 10^{-3} (1 + m_i/m_a)^{1/3} R \). The integration was stopped when the atom reached a distance \( r_a = 2.1 r_0 \) much larger than the ion motion, at which point the total secular energy of the system was evaluated. We confirmed the accuracy of our integration by replacing the RF potential with a time-independent secular potential and confirming energy conservation at the level of \( 10^{-3} W_0 \).

Figures 3a and 3b show the numerically calculated maximal energy gain \( W_{\text{max}} \) in the initial collision as a function of \( m_a/m_i \) and \( q \). For \( q \leq 0.1 \), the calculated \( W_{\text{max}} \) is within 30% of the analytic prediction \( W_{\text{max}} = K \); for \( q = 0.5 \), \( m_a/m_i = 1/2 \), \( W_{\text{max}} \) increases to 1.7K, partly because the analytic result does not include the micromotion interruption. The heating is insensitive to the the hard-core radius: a tenfold increase or decrease in \( \epsilon \) changes \( W_{\text{max}} \) by less than 1%. Our results are also very robust with respect to the initial conditions: the average energy gain in Figure 2 changes by less than 5% if the ion is started at rest in the center of the trap and the atom at rest a distance \( R \) away.

In three-dimensional linear quadrupole RF traps, the electric field \( \mathbf{E} \) and the ion position \( r_i \) in (2) are vectorial quantities. Assuming that the collision trajectory is still nearly one-dimensional, averaging over the atom’s approach direction rescales the work done by the RF field, Eq. (2), by \( 4/(3\pi) \), leading to a natural 3D heating scale \( W_0^{3D} = 4W_0/(3\pi) \). To check this, numerical simulations were done for the three-dimensional quadrupole RF trap from Ref. 2. We considered cold \(^{87}\text{Rb} \) atoms that have passed the angular momentum barrier at a large distance and are colliding head-on with a \(^{174}\text{Yb}^+ \) ion from a random direction and at a random time. The initial conditions for numeric integration were chosen by analogy to the 1D case. Figure 4 shows a sample ion-atom trajectory including multiple collisions. Due to a difference in the axial and radial frequencies of the ion trap, the collision trajectory precesses about the \( y \) axis, while remaining nearly one-dimensional close to the collision points. A histogram of the final system energies \( E \) after the atom is ejected to infinity is shown on Figure 4h, with an average energy gain of \( 0.9W_0^{3D} \), confirming that the heating is similar in one and three dimensions.

Table I shows \( W_0 \) together with the \( s \)-wave threshold energy \( E_s = \hbar^2/(2\mu^2 C_4) \) and the energy \( \hbar \omega \) of a trap vibrational quantum in various experimental systems. In the current systems \( 1 \) - \( 5 \), \( 7 \), the atom number per characteristic volume \( R^3 \) is much smaller than one, while the atom kinetic energy is much smaller than \( W_0 \), and we expect binary ion-atom collisions to be well described by our model. Given the typical duration of a collision of about \( T/2 \) and typical ion-atom collision rate coefficients \( \Gamma \), corrections due to three-body collisions are expected at critical atom densities on the order of \( n_c \sim 10^{14} \text{ cm}^{-3} \). At these densities, three-body inelastic processes become significant, leading to strong ion heating and loss as was recently observed \( 7 \). Thus best sympathetic cooling is expected in the density regime \( n < n_c \), where it is limited by the two-body energy scale \( W_0 \) derived above (Eq. 4) to temperatures \( T_{\text{min}} \sim W_0/k_B \). In currently realized systems, \( W_0 \) is more than one order of magnitude larger than the trap vibrational quantum and almost three orders of magnitude larger than the \( s \)-wave scattering limit.

Since, for light atoms, the ratio of heating to the \( s \)-wave collision threshold scales as \( W_0/\hbar \omega \propto (\omega C_4)^{1/3} m_a^{1/3}/m_i \) and the ratio of heating to the trap vibration quantum scales as \( W_0/\hbar \omega \propto (\omega C_4)^{1/3} m_a^{5/3}/m_i \), our model predicts that micromotion heating could be mitigated using light atoms and heavy ions trapped in weak RF traps, limited by the control of DC fields (since \( E_{\text{DC}}^3 \propto \omega \)). In particular, with control over the DC electric fields on the order of 10 mV/m - an order of magnitude better than current state-of-the-art - the \(^{87}\text{Yb}^+ / \text{Li} \) system may enter the \( s \)-wave regime. Heating could also be decreased by employing Raman transitions to produce molecular ions that are sufficiently

| ion / atom | \( \omega/(2\pi) \) kHz | \( q \) | \( E_s/k_B \) nK | \( E_{\text{DC}}^3 \) mV/m | \( \hbar \omega/k_B \) \( \mu \)K | \( W_0/k_B \) \( \mu \)K |
|-----------|----------------|-----|----------------|----------------|----------------|----------------|
| \(^{174}\text{Yb}^+ \text{ } ^{87}\text{Rb} \) | 200 | 0.013 | 44 | 4.6 | 9.6 | 540 |
| \(^{87}\text{Rb}^+ \text{ } ^{87}\text{Rb} \) | 350 | 0.24 | 79 | 7.7 | 17 | 210 |
| \(^{138}\text{Ba}^+ \text{ } ^{87}\text{Rb} \) | 200 | 0.11 | 52 | 4.5 | 9.6 | 150 |
| \(^{40}\text{Ca}^+ \text{ } ^{87}\text{Rb} \) | 110 | 0.20 | 200 | 2.6 | 5.3 | 50 |
| \(^{174}\text{Yb}^+ \text{ } ^{174}\text{Yb} \) | 67 | 0.14 | 44 | 1.6 | 3.2 | 41 |
| \(^{174}\text{Yb}^+ \text{ } ^{40}\text{Ca} \) | 250 | 0.25 | 270 | 14 | 12 | 32 |
| \(^{174}\text{Yb}^+ \text{ } ^{23}\text{Na} \) | 50 | 0.30 | 710 | 4.7 | 2.4 | 1.5 |
| \(^{174}\text{Yb}^+ \text{ } ^{7}\text{Li} \) | 50 | 0.30 | 6400 | 14 | 2.4 | 0.24 |
bound so as not to be affected by micromotion [34]. Another option may be the use of an optical trap for the ion, as was recently demonstrated [36].

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[1] A.T. Grier, M. Cetina, F. Oručević, and V. Vuletić, Physical Review Letters 102, 223201 (2009).
[2] C. Zipkes, S. Palzer, C. Sias, and M. Köhl, Nature 464, 388 (2010).
[3] C. Zipkes, S. Palzer, L. Ratschbacher, C. Sias, and M. Köhl, Physical Review Letters 105, 133201 (2010).
[4] S. Schmid, A. Härter, and J.H. Denschlag, Physical Review Letters 105, 133202 (2010).
[5] W.G. Rellergert, S.T. Sullivan, S. Kotochigova, A. Petrov, K. Chen, S.J. Schowalter, and E.R. Hudson, Physical Review Letters 107, 243201 (2011).
[6] F.H.J. Hall, M. Aymar, N. Bouloufa-Maafa, O. Dulieu, and S. Willitsch, Physical Review Letters 107, 243202 (2011).
[7] A. Härtner, A. Krükow, A. Brunner, W. Schnitzler, S. Schmid, J.H. Denschlag, Physical Review Letters 109, 123201 (2012).
[8] L. Ratschbacher, C. Zipkes, C. Sias, and M. Köhl, Nature Physics 8, 649-652 (2012).
[9] W. W. Smith, O. P. Makarov, and J. Lin, Journal of Modern Optics 52, 2253 (2005).
[10] E.R. Hudson, Physical Review A 79, 032716 (2009).
[11] R. Côté and A. Dalgarno, Physical Review A 62, 012709 (2000).
[12] R. Côté, Physical Review Letters 85, 5316 (2000).
[13] R. Côté, V. Kharchenko, and M.D. Lukin, Physical Review Letters 89, 093001 (2002).
[14] P. Massignan and C.J. Pethick, H. Smith Physical Review A 71, 023606 (2005).
[15] J. Goold, H. Doerk, Z. Idziaszek, T. Calarco, and T. Busch, Physical Review A 81, 041601(R) (2010).
[16] B. Gao, Physical Review Letters 104, 213201 (2010).
[17] C. Kollath, M. Köhl, and T. Giamarchi, Physical Review A 76, 063602 (2007).
[18] Y. Sherkunov, B. Muzykantskii, N. d’Ambrumenil, and B.D. Simons, Physical Review A 79, 023604 (2009).
[19] Z. Idziaszek, T. Calarco, and P. Zoller, Physical Review A 76, 033409 (2007).
[20] H. Doerk, Z. Idziaszek, and T. Calarco, Physical Review A 81, 012708 (2010).
[21] D. Leibfried, R. Blatt, C. Monroe, and D. Wineland, Reviews of Modern Physics 75, 281 (2003).
[22] F. G. Major and H. G. Dehmelt, Physical Review 170, 91 (1968).
[23] H. Schaal, U. Schmeling, and G. Werth, Appl. Phys. 251, 249 (1981).
[24] L. S. Cutler, R. P. Giffard, and M. D. Megoire, Appl. Phys. B 142, 137 (1985).
[25] M. D. N. Lunney, F. Buchinger, and R. B. Moore, Journal of Modern Optics 39, 349 (1992).
[26] F. Herfurth, J. Dilling, A. Kellerbauer, G. Bollen, S. Henry, and H. Kluge, Nucl. Instr. Meth. A 469, 254 (2001).
[27] A. Kellerbauer, T. Kim, R. B. Moore, and P. Varfalvy, Nucl. Instr. Meth. A 469, 276 (2001).
[28] M. Green, et al., Physical Review A 76, 1 (2007).
[29] J. Mikosch, et al., Physical Review Letters 98, 4 (2007).
[30] B. Flatt, et al., Nucl. Instr. Meth. A 578, 399 (2007).
[31] Y. Moriwaki, M. Tachikawa, Y. Maeno, and T. Shimizu, Jpn. J. Appl. Phys. 31, 1640 (1992).
[32] S. Schwarz, Nucl. Instr. Meth. A 566, 233 (2006).
[33] R. DeVoe, Physical Review Letters 102, 063001 (2009).
[34] L. H. Nguyen, A. Kalev, M. D. Barrett, and B.-G. Englert, Physical Review A 85, 052718 (2012).
[35] C. Zipkes, L. Ratschbacher, C. Sias, and M. Köhl, New Journal of Physics 13, 053020 (2011).
[36] C. Schneider, M. Endlerlein, T. Huber, and T. Schäetz, Nature Photonics 4, 772 (2010).