Power-Law Distributions for a Trapped Ion Interacting with a Classical Buffer Gas

Ralph G. DeVoe

Physics Department, Stanford University, Stanford, California 94305, USA

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Classical collisions with an ideal gas generate non-Maxwellian distribution functions for a single ion in a radio frequency ion trap. The distributions have power-law tails whose exponent depends on the ratio of buffer gas to ion mass. This provides a statistical explanation for the previously observed transition from cooling to heating. Monte Carlo results approximate a Tsallis distribution over a wide range of parameters and have *ab initio* agreement with experiment.

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The behavior of ions in the collision-free regime of a radio frequency ion trap is well understood. Laser cooling and the properties of the quantum-mechanical ground state [1] have been examined in great detail. It is therefore surprising that the more accessible regime of ions cooled by buffer gas collisions has never been thoroughly analyzed. Major and Dehmelt first showed in 1968 that collisions with neutral buffer gases could either cool or heat the ion [2], depending on their relative masses. His theory, still widely accepted today, hypothesized that the recoil from a collision with a heavy neutral atom heated the ion by disrupting its response to the rf field (i.e., micromotion). The theory relied on the ''pseudopotential'' or time-averaging approximation and did not address the statistics of the ion's distribution function, which were assumed to be noncritical. Subsequent workers have introduced Maxwell-Boltzmann (MB) statistics in several ways, for example, by assuming a Gaussian velocity distribution function [3] or by hypothesizing Gaussian random noise in a Langevin equation [4]. Numerical work has shown apparent instability in individual trajectories without addressing the statistics [5].

In this Letter we compute the ion's distribution function using a combination of Monte Carlo and analytic methods. Our results show that the distribution is not in general Gaussian and does not follow MB statistics. Collisions with heavy neutrals give the distribution function power-law tails $E^{-\alpha}$ in place of the Gaussian equivalent $\exp(-E/kT)$. Here E is a time-averaged ''pseudoenergy'' which is not conserved during collisions [2]. In previous work the instability was thought to arise from a positive heating rate $dE/dt > 0$, leading to exponential runaway. Our results lead to a different picture in which stationary power-law tails lead to a small but constant rate of ion loss. This leads to orders-of-magnitude differences in predicted ion lifetimes.

In the past decade non-Gaussian statistics have been studied in many different contexts [6]. In atomic physics such distributions have been observed primarily in laser-cooled atoms operating near the quantum-mechanical ground state. Subrecoil laser cooling has been shown to obey Lévy statistics [7] with a nonstationary distribution and nonergodicity [8,9]. Lévy walks, anomalous diffusion [10], and a tunable Tsallis distribution [11] have been observed in optical lattices. The present case is remarkable in that it shows non-Gaussian statistics in a classical system not far removed from the ideal gases originally studied by Maxwell and Boltzmann.

An additional motivation for understanding collisional heating is that ion traps have recently come into use as probes of collision physics. Hybrid traps comprising both ion traps and magneto-optical traps have been constructed yielding results for charge exchange cross sections [12] and radiative lifetimes [13]. The theory of ultracold atom-ion collisions has been developed [14–16], and novel effects of an ion in a Bose-Einstein condensate have been predicted [17]. Room temperature buffer gases have been used to study molecular ions [18] and the properties of multipole traps [19]. This is in addition to the more traditional use of buffer gas cooling in trace element detection [20,21], the trapping of radioactive ion beams [5], and several other applications. It is necessary to understand collisional heating to disentangle the effect of the trap fields from the collision physics.

Previous Monte Carlo work has used numerical integration to compute the ion's motion between collisions. This is neither fast nor accurate enough for the total of $\approx 10^{10}$ collisions needed to compute the distribution function. Instead we use the classical time-evolution matrix M of the ion

$$\begin{pmatrix} x(t_2) \\ v(t_2) \end{pmatrix} = M(t_2, t_1) \begin{pmatrix} x(t_1) \\ v(t_1) \end{pmatrix} \tag{1}$$

to propagate the position and velocity of the ion from one collision to the next. Consider first the case of a simple harmonic oscillator, which obeys $\ddot{x}(t) + \beta^2 x(t) = 0$. The time-evolution matrix S of this system is

$$S(t_2, t_1) = \begin{pmatrix} \cos\beta(t_2 - t_1) & \frac{1}{\beta}\sin\beta(t_2 - t_1) \\ -\beta\sin\beta(t_2 - t_1) & \cos\beta(t_2 - t_1) \end{pmatrix}. \tag{2}$$

This is a special case of the general solution [22]

$$M(t_2, t_1) = \frac{1}{D}\begin{pmatrix} S_1P_2 - Q_2R_1 & P_1Q_2 - P_2Q_1 \\ S_1R_2 - S_2R_1 & S_2P_1 - Q_1R_2 \end{pmatrix}, \quad (3)$$

where P and Q are the two linearly independent solutions of a second order linear differential equation, R and S are the respective time derivatives, and the Wronskian $D = S_1P_1 - Q_1R_1$. The rf ion trap obeys a Mathieu equation

$$\frac{d^2x}{dt^2} + (a - 2q\cos 2t)x = 0, \quad (4)$$

for which P and Q are given by the Fourier solutions [1,2]

$$P_i = \sum_{m=-\infty}^{m=\infty} \cos[(\beta + 2m)t_i)]C_{2m}, \quad (5)$$

where Q_i has $\sin[(\beta + 2m)t_i)]$ in place of the cosine. Here $q = 2eV_0/m_i\Omega^2 r_0^2$, $a = 4eU_0/m_i\Omega^2 r_0^2$, E_0 and U_0 are the rf and dc applied potentials, r_0 is the trap radius, and the unit of time is $2/\Omega$, where Ω is the angular frequency of the applied rf. The coefficients up to and including $C_{\pm 8}$ are evaluated to 24 bit accuracy (5×10^{-8} error) with a recursive routine. Error propagation has been tested with the identity $M(t_N, t_1) = \prod_{i=1}^{N-1} M(t_{i+1}, t_i)$. For N up to 10^6 the discrepancy $< 1 \times 10^{-5}$ for randomly chosen times t_i.

The distribution function is computed by Monte Carlo averaging over six random variables for each collision: the time t_i, the center-of-mass angles θ_i and φ_i, and the three random velocities v_i^x, v_i^y, and v_i^z of the buffer gas, which obey a Maxell-Boltzmann distribution at temperature T, where $T = 100$, 300, or 1000 K. We assume a linear ion trap with transverse rf confinement and a dc potential along the z axis, represented by the Mathieu matrices M_x and M_y, where $q_y = -q_x$, and static harmonic oscillator matrix S_z as in Eq. (2) above. We combine the three matrices for the x, y, and z axes into a single 6×6 matrix equation

$$\begin{pmatrix} r_x(t_N) \\ r_y(t_N) \\ r_z(t_N) \end{pmatrix} = \prod_{i=1}^{N} C(\theta_i, \varphi_i, \vec{v}_i)\vec{M}(t_i, t_{i-1})\begin{pmatrix} r_x(t_0) \\ r_y(t_0) \\ r_z(t_0) \end{pmatrix}, \quad (6)$$

where $\vec{M}$ has M_x, M_y, and S_z along the diagonal, and where, for example, r_x is the column vector $(x, \dot{x})$. Collisions are represented by a matrix C which leaves the coordinates unchanged but transforms the velocities according to a hard sphere (isotropic in the center of mass) collision model. The ion-neutral atom collisions are modeled by Langevin scattering in which the cross section $\sigma \propto 1/v$ so the time between collisions is a random variable independent of the relative velocity v. An ensemble typically consists of 10^6 trials each containing 500–50 000 collisions. All distributions for >500 collisions agree within statistics.

Distribution functions for a single $^{136}Ba^+$ ion at $q = 0.1$ are shown in Fig. 1. Six different buffer gases with masses $m_B = 4$, 40, 84, 136, 170, and 200 amu have been assumed, corresponding to the noble gases He, Ar, Kr, Xe, as in a recent experiment [20,21], and to two hypothetical

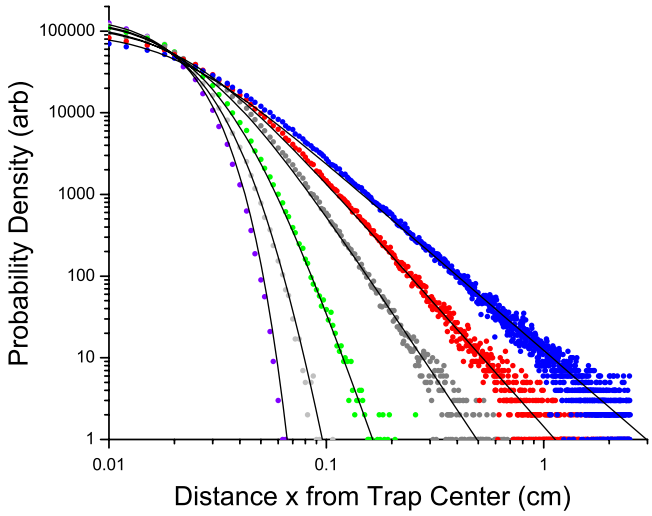

FIG. 1 (color online). Monte Carlo distributions for a single $^{136}Ba^+$ ion cooled by six different buffer gases at 300 K ranging from $m_B = 4$ (left) to $m_B = 200$ (right). Note the evolution from Gaussian to power law (straight line) as the mass increases. The solid lines are Tsallis functions [Eq. (7)] with fixed $\sigma = 0.0185$ cm and the exponents of Table I.

heavier gases. The distribution for $^{136}Ba^+$ in He is a good fit to a MB distribution with a classical $\sigma \approx \sqrt{2kT/m_I\omega^2}$, where ω is the secular frequency $\beta\Omega/2$. All of the other gases show non-Gaussian distributions which develop broad power-law tails as the mass increases. The four heaviest gases fit a power law $x^{-2\alpha}$ with good χ^2 over at least 3 orders of magnitude. For $m_B = 84$, 136, 170, and 200 the best fits are $\alpha = 3.2$, 1.98, 1.5, and 1.17, respectively. A typical fit will have $\chi^2/n \leq 1.1$ for $n > 100$ degrees of freedom.

In the absence of an analytic theory, the data have been fit to a Tsallis function $T(x/\sigma, n)$

$$T(x/\sigma, n) = \frac{T_0}{[1 + (x/\sigma)^2/n]^n}, \quad (7)$$

which is a generalization of the Gaussian. For $n \to \infty$, T reduces to a Gaussian, while for small n it has power-law tails of the form $(x/\sigma)^{-2n}$. The exponent n is related to the more familiar "entropic" Tsallis parameter q_T by $q_T = 1 + 1/n$. The Tsallis function arises in the theory of non-

TABLE I. Tsallis parameters n and q_T fit from Fig. 1.

Buffer gas	m_I/m_B	n	q_T
He	34.5	>60	1.03
Ar	3.40	8.2	1.12
Kr	1.70	3.8	1.26
Xe	1.0	1.98	1.51
170	0.80	1.50	1.80
200	0.68	1.15	1.87

extensive entropy [6], but at present we treat it empirically. Table I shows the value of n extracted from fitting the distributions to Eq. (7), where σ was held constant for all m_B and T_0 normalizes the distribution to unity. The fit is qualitative since the χ^2 is poor due to systematic deviation near the origin, where the standard deviation $<0.3\%$. Nevertheless, Fig. 1 shows good agreement over a factor of 10^5 in probability density and a factor of 50 in buffer gas mass. The value of the Tsallis exponent n is close to the value of α extracted from the power-law fit above. Similar data at 100 and 1000 K give comparable fits with the same σ scaled by $\sqrt{T}$.

The usefulness of the Tsallis function is that it shows that n and σ act independently of each other, to first order. In the light gas limit $m_B \rightarrow 0$, the ion has a Gaussian distribution with a temperature T equal to that of the buffer gas, so that $\sigma \approx \sqrt{2kT/m_I\omega^2}$. As m_B increases, σ changes very slowly, so that the distribution retains a Gaussian-like core of constant width as the power-law tails get stronger. This indicates that the increase in the mean energy of the ion is not the cause of ion loss. A three-parameter fit, in which σ, n, and the normalization are optimized for each value of m_B, shows a weak dependence of σ on m_I/m_B. For example, the best value of $\sigma = 0.0175$ cm at $m_B = 4$ rises to $\sigma = 0.022$ cm at $m_B = 200$, an increase of only 26% for a 50-fold decrease of m_I/m_B. Similarly, changing the temperature of the buffer gas does not alter the power-law exponent. To generalize further, the Tsallis exponent n is approximated by the simple relation $n \approx 2m_I/m_B$, which is accurate in the exponent to about $\pm 20\%$.

The Monte Carlo simulation also computes the ion lifetime. The ion is started at the origin with zero energy and is propagated through i collisions, until $r_i = \sqrt{x_i^2 + y_i^2} \geq r_0$, the trap radius. In general the ion lifetime $\tau \propto r_0^{2n}$, where n is the Tsallis exponent of Table I. However, since the trap depth $U \propto r_0^2$ (for constant q), it is more general to plot τ versus U, which yields $\tau \propto U^n$ as shown in Fig. 2. Interestingly, τ is not sensitive to initial conditions, and an ion starting with an energy ≈ 1 eV has τ only slightly shorter than with zero energy. This is because most of the hot ions equilibrate to 300 K in a few dozen collisions. It is only in a very large ensemble (10^6 trials in Fig. 1) or a very large number of collisions ($N = 5 \times 10^5$ in Fig. 2) that extreme values of r_i are reached. In contrast to the exponential runaway model, Fig. 2 suggests that ion traps may be designed to achieve a specific ion lifetime.

Power-law tails dominate the trap stability whenever a stationary distribution function exists. However, when the Tsallis exponent falls below 1, which occurs for $m_B > 1.55m_I$, the distribution becomes time dependent, as in a Lévy distribution [7], the mean values diverge, and the ion's energy increases with each collision. In this case exponential runaway occurs as originally suggested in [2].

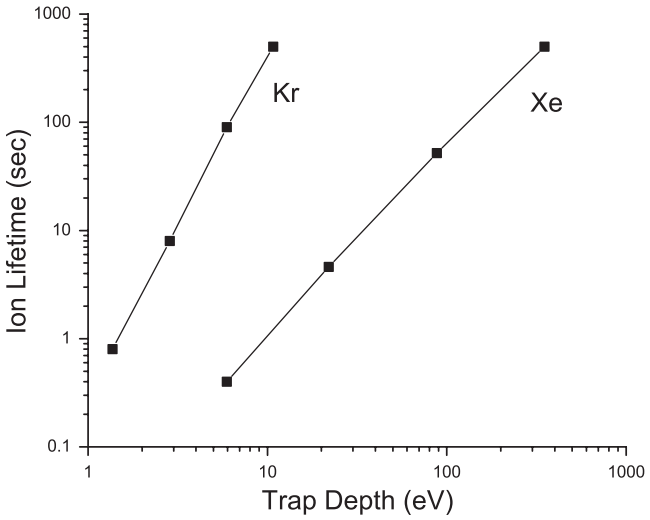

FIG. 2. Predicted power-law ion lifetime $\tau \propto U^n$ where U is the trap depth, n is the Tsallis exponent of Table I, and the parameters correspond to Fig. 1.

The Monte Carlo results have no free parameters and the predicted ion lifetimes agree with the results of a recent experiment [20,21] in which a single Ba^+ was confined in a trap of radius $R_T = 0.26$ cm and $q = 0.52$. Stable trapping was observed for He gas, Ar gas was measured to give an ion lifetime of 50–100 sec, while Kr and Xe had lifetimes too short to measure (<5 sec). When the above R_T and q

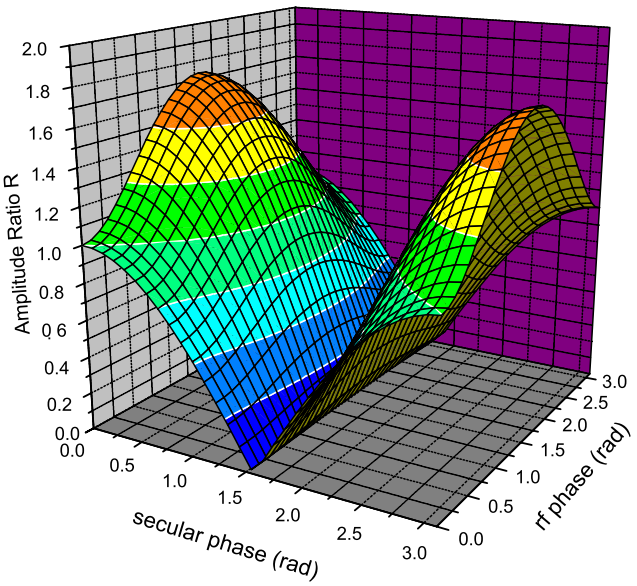

FIG. 3 (color online). A heating and cooling diagram for a single collision in the case $m_b = m_i$ or $\alpha = 0$ in Eq. (8). Heating occurs for $R > 1$ and cooling for $R < 1$. The explanation given in the text does not depend on the exact form of this result and requires only that there be a small phase space volume $\beta < 1$ for $R > 1$.

are input into the Monte Carlo program it yields lifetimes of 45 sec for Ar and <0.1 sec for Kr and Xe.

It remains to provide a physical explanation of these results. An analytic expansion of the Mathieu matrix Eqs. (3) and (6) shows that the power-law tails are a result of a multiplicative random process [23], i.e., products of random variables. These may be contrasted with the better-known additive random processes (sums of random variables), which obey the central limit theorem and produce Gaussian statistics. Multiplicative random products are not well understood, but they do not in general lead to Gaussian distributions. Multiplicative fluctuations have recently been studied in a Langevin equation [24] and have been shown to lead to a tunable Tsallis distribution.

A model for multiplicative fluctuations can be derived by expanding the Mathieu matrix Eq. (3) to first order in q and substituting the result in Eq. (6). In the same limit used by Major and Dehmelt [2] one can show [25] that the matrix product Eq. (6) can be approximated by a product of numbers $\prod_{i=1}^{N} R(\varphi_s^i, \varphi_m^i)$, where

$$R(\varphi_s, \varphi_m) = \sqrt{\cos^2\varphi_s + \alpha^2\sin^2\varphi_s + 2(\alpha-1)^2\cos^2\varphi_s\sin^2\varphi_m - \sqrt{2}\alpha(\alpha-1)\sin 2\varphi_s \sin\varphi_m}. \quad (8)$$

Here φ_s^i and φ_m^i are the phases of the secular motion (''macromotion'') and driven rf oscillations (''micromotion'') at the time of the ith collision, and $\alpha = (m_I - m_B)/(m_I + m_B)$ is a recoil parameter. Multiplicative random products tend to be dominated by rare events of large amplitude [23]. In the present case these events can be identified as N consecutive heating collisions without an intervening cooling collision. Consider a volume of phase space $\beta < 1$ around the heating maximum $R \approx \sqrt{3}$ in Fig. 3. N consecutive collisions will give an amplitude of $3^{N/2}$ with a probability of β^N. This provides a mechanism for the power-law tails leading to ion loss. If the trap were of infinite size, cooling collisions, which occupy most of the phase space, would eventually return the ion to the origin. In this sense, the ion loss is due to a non-Gaussian *fluctuation* rather than to heating. In the light mass limit $\alpha \to 1$ Gaussian statistics return since each term in the product is near unity, $R = 1 + \epsilon(\varphi_s, \varphi_m)$, where $\epsilon \ll 1$. The product reduces to a sum $1 + \sum \epsilon(\varphi_s, \varphi_m)$, the fluctuations become additive, and the central limit theorem applies.

This work has several implications. For statistical mechanics it provides a simple, classical system which shows tunable non-Gaussian statistics. For trapping and cooling experiments it shows how traps may be engineered for a specific ion lifetime, as in Fig. 2. This should be useful in trace atom detection [20,21] and in trapping radioactive ion beams [5]. It is also necessary for understanding recent ion trap collision experiments (Refs. [11–17]), since the non-Gaussian distribution function can alter their interpretation.

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