SUPPLEMENTAL MATERIAL

The energy distribution of an ion in a radiofrequency trap interacting with a nonuniform neutral buffer gas

I. Rouse[a] and S. Willitsch[a]

[a] Department of Chemistry, University of Basel, Klingelbergstrasse 80, 4056 Basel, Switzerland

ARTICLE HISTORY
Compiled March 8, 2019

Appendix A. Energy change during collisions at the centre of the trap

We consider the extreme case in which collisions may only occur at the centre of the trap, under the assumption that there is no excess micromotion due to external forces (see, e.g., Ref. [1]). This may be achieved using the same procedure to calculate the change in energy as described in Section 2.2 of the main text, with the exception that there is now an additional constraint that the collision must occur at the centre of the trap, $r_j(\tau) = 0$ for each $j \in (x, y, z)$, where $r_j(\tau)$ is defined as in Eq. (5) of the main text. That is,

$$A_j(\cos\phi_j \ \mathrm{ce}_j(\tau) - \sin\phi_j \ \mathrm{se}_j(\tau)) = 0. \tag{A1}$$

For non-zero values of A_j, corresponding to an ion with a non-zero amplitude of motion passing through the trap centre, solutions to Eq. (A1) can be found by requiring that,

$$\tan\phi_j = \frac{\mathrm{ce}_j(\tau)}{\mathrm{se}_j(\tau)}. \tag{A2}$$

Since $\tan\phi_j$ is periodic, there are two possible solutions to this equation, which physically represent the fact that the velocity may correspond to motion in either the $+j$ or $-j$ direction. The velocity is given by the derivative of Eq. (5) of the main text with respect to τ, and substituting in the solutions given by Eq. (A2) we obtain,

$$v_j(\tau, r_j = 0) = \pm\frac{A_j W_j}{\sqrt{\mathrm{ce}_j(\tau)^2 + \mathrm{se}_j(\tau)^2}}. \tag{A3}$$

Here, $W_j = \mathrm{ce}_j(\tau)\dot{\mathrm{se}}_j(\tau) - \dot{\mathrm{ce}}_j(\tau)\mathrm{se}_j(\tau)$ is the Wronskian, which is a time-independent quantity [2]. For $q_j = 0$, $\mathrm{ce}_j(\tau), \mathrm{se}_j(\tau)$ reduce to $\cos\beta_j\tau$ and $\sin\beta_j\tau$ respectively, such that $\mathrm{ce}_j(\tau)^2 + \mathrm{se}_j(\tau)^2 = 1$. In this case, the velocity at the centre of the trap is independent of τ, as expected for a harmonic oscillator. However, for non-zero q_j this no longer holds. Indeed, by plotting the phase-space trajectory of the ion (Fig. A1), it can be seen that the velocity of the ion at $r_j = 0$ is not equal to the secular velocity and takes a range of values.

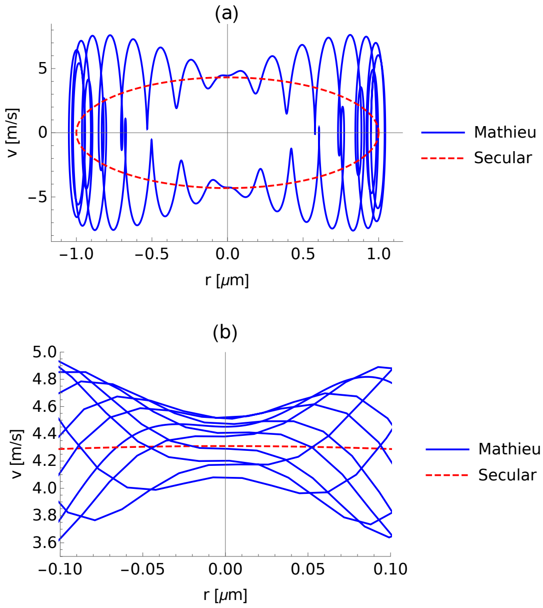

Figure A1. The phase-space trajectory of an ion in a radiofrequency trap with $q_j = 0.1, a_j = -0.000625/2, \Omega = 20 \times 2\pi$ MHz, an amplitude of $A_j = 1$ μm, and the phase set to $\phi_j = 0$. (a) The trajectory over one period of the secular motion comparing the exact solution to the Mathieu equation (solid line) to the secular motion (dashed line). (b) The velocity of the ion close to the centre of the trap, $r = 0$, shown for multiple periods of the secular motion to highlight the presence of micromotion at the centre of the trap.

We estimate the magnitude of this effect as follows. Using the Fourier series definitions for $\mathrm{ce}_j(\tau), \mathrm{se}_j(\tau)$ and trigonometric identities, it can be shown that,

$$\mathrm{ce}_j(\tau)^2 + \mathrm{se}_j(\tau)^2 = \sum_{m,n} c_{2m,j} c_{2n,j} \cos[2(m-n)\tau]. \tag{A4}$$

As a result of the terms in this sum with $m \neq n$, this function is time dependent and contains components with frequencies of integer multiples of Ω. Evaluating Eq. (A4) for $m, n \in (-1, 0, 1)$, and using the approximate values for the Mathieu coefficients $c_0 = 1, c_{\pm 2,j} = -q_j/4$ [3], we find,

$$\mathrm{ce}_j(\tau)^2 + \mathrm{se}_j(\tau)^2 \approx 1 - q\cos(2\tau). \tag{A5}$$

The velocity is therefore approximated by,

$$v_j(\tau, r_j = 0) \approx \pm \frac{A_j W_j}{\sqrt{1 - q_j \cos(2\tau)}}, \tag{A6}$$

and expanding this as a Taylor series to first order in q_j around $q_j = 0$ we obtain,

$$v_j(\tau, r_j = 0) \approx \pm A_j W_j [1 + \frac{q_j}{2}\cos(2\tau)]. \tag{A7}$$

An approximate value for the Wronskian W_j can be found using the $m = 0$ terms of the Fourier series expansions of $\mathrm{ce}_j(\tau), \mathrm{se}_j(\tau)$, which produces $W_j \approx c_0^2 \beta_j$. Using this approximation with $c_0 \approx 1$, and converting from τ to t, we find,

$$v_j(t, r_j = 0) \approx \pm A_j \omega_j [1 + \frac{q_j}{2} \cos(\Omega t)], \tag{A8}$$

where the definition of the secular frequency $\omega_j = \beta_j \Omega/2$ has been employed to simplify the result. This form of the result bears a resemblance to the adiabatic approximation for the motion of the ion [1],

$$r_j(t) = A_j \cos(\omega_j t + \phi_j)[1 - \frac{q_j}{2} \cos(\Omega t)], \tag{A9}$$

where the sign of the $\frac{q_j}{2}$ term used here differs from that of Ref. [1] as a result of the use of a different convention for the Mathieu equation. Taking the derivative of Eq. (A9) with respect to t produces,

$$v_j(t) = A_j \cos(\omega_j t + \phi_j)[\Omega \frac{q_j}{2} \sin(\Omega t)] - A_j \omega_j \sin(\omega_j t + \phi_j)[1 - \frac{q_j}{2} \cos(\Omega t)]. \tag{A10}$$

For the ion to be at the centre of the trap with $|q_j| < 2$ it is required that $\cos(\omega_j t + \phi_j) = 0$, and therefore $\sin(\omega_j t + \phi_j) = \pm 1$. Thus,

$$v_j(t, r_j = 0) = \mp A_j \omega_j [1 - \frac{q_j}{2} \cos(\Omega t)], \tag{A11}$$

which is equivalent to Eq. (A7) up to the sign of the $\frac{q_j}{2}$ term. This discrepancy in the sign is a consequence of the fact that $\omega_j q_j$ is approximately proportional to q_j^2 [3], and terms of this order are not included in the adiabatic approximation. Using an improved approximation for the motion of the ion including terms up to order q_j^2 as in Ref. [4] produces a result in agreement with Eq. (A7).

The velocity of the ion at the centre of the trap is therefore approximately given by the sum of the secular velocity and a term proportional to $q_j/2 \cos(\Omega t)$, i.e., a micromotion term. For the trajectory shown in Fig. A1 with $q_j = 0.1$, the secular velocity is given by ≈ 4.3 m/s, and the actual velocity spans $4.1 - 4.5$ m/s, in agreement with this result. Thus, we conclude that an ion which is passing through the centre of the trap exhibits a contribution to the velocity from the micromotion proportional to the secular velocity of the ion. If the ion is perfectly cooled to the centre of the trap, then $A_j = 0$ and this micromotion vanishes, as expected. However, for non-zero values of A_j, the magnitude of this micromotion increases proportionally to the secular velocity.

The post-collision energy is given as before by Eq. (11) of the main text, as this expression is valid for collisions at an arbitrary point in the trap. However, in this case the set of phases ϕ_j are determined by Eq. (A2), such that each $f_{\phi_j}(\phi_j)$ is sharply peaked at the two possible values which we assume to occur with equal probability. Assuming a thermal distribution for the components of the velocity of the buffer gas, an isotropic random rotation matrix, and taking τ to follow a uniform distribution, we find,

$$\langle E'_j \rangle = \frac{\langle E_j \rangle}{(1+\tilde{m})^2} + \kappa_j k_B T_b + \sum_{k \in (x,y,z)} \frac{\tilde{m}^2 c_{0,j}^2 W_k^2 \beta_j^2}{3(1+\tilde{m})^2 c_{0,k}^2 W_j^2 \beta_k^2} \mathcal{M}_j[(\mathrm{ce}_k(\tau)^2 + \mathrm{se}_k(\tau)^2)^{-1}] \langle E_k \rangle, \tag{A12}$$

where,

$$\kappa_j = \frac{\tilde{m}}{(1+\tilde{m})^2} \frac{c_{0,j}^2 \beta_j^2}{W_j^2}, \tag{A13}$$

and $\mathcal{M}_j$ is defined in Eq. (21) of the main text. As a further simplification, we assume that the temperature of the buffer gas is negligible, and that each of the three components of the mean energy before the collision are approximately equal in magnitude, i.e., $\langle E_x \rangle = \langle E_y \rangle = \langle E_z \rangle = \frac{1}{3}\langle E \rangle$. The ratio of the post-collision energy, $\langle E' \rangle = \sum_j \langle E'_j \rangle$, to the pre-collision energy $\langle E \rangle$ is then given by,

$$\frac{\langle E' \rangle}{\langle E \rangle} = \frac{1}{(1+\tilde{m})^2} + \frac{\tilde{m}^2}{9(1+\tilde{m})^2} \sum_{j,k\in(x,y,z)} \frac{c_{0,j}^2 W_k^2 \beta_j^2}{c_{0,k}^2 W_j^2 \beta_k^2} \mathcal{M}_j [(\mathrm{ce}_k(\tau)^2 + \mathrm{se}_k(\tau)^2)^{-1}]. \tag{A14}$$

For non-zero temperatures, we may solve Eq. (A12) for the steady-state values of $\langle E_j \rangle$ by setting $\langle E'_j \rangle = \langle E_j \rangle$, and solving the resulting set of linear equations. For sufficiently large values of $\tilde{m}$, no physically meaningful solution exists corresponding to values of $\tilde{m}$ greater than the critical mass ratio.

Appendix B. Secular phase distribution

The probability that a collision takes place at a given location $\mathbf{r}$ in a time interval Δt is proportional to the density $\rho(\mathbf{r})$ of the buffer gas,

$$p(c|\mathbf{r}) = k_c \Delta t \rho(\mathbf{r}). \tag{B1}$$

where the notation $c|\mathbf{r}$ indicates the probability of a collision (c) at a specific position $\mathbf{r}$ and where k_c is the collision rate constant. By employing Bayes' theorem, this may be converted to the probability for the ion to be at position $\mathbf{r}$ at the time of a collision [5],

$$p(\mathbf{r}|c) = \frac{p(c|\mathbf{r})p(\mathbf{r})}{\int p(c|\mathbf{r})p(\mathbf{r})d\mathbf{r}}. \tag{B2}$$

To proceed, we make the simplification that the density of the buffer gas changes sufficiently slowly such that it depends only on the secular position of the ion, and take the secular position to be given by $r_j = A_j \cos(\phi_j + \omega_j t) = A_j \cos\tilde{\phi}_j$. This approximation is appropriate for a buffer gas which is not strongly localised, i.e., the density of the buffer gas does not vary significantly over the length scale given by the amplitude of the micromotion, that is, the density is approximately constant over an interval of width $r_j q_j$ centred at r_j. For a given value of A_j, the probability for a component of the secular position to take a specific value in the interval $[-A_j, A_j]$ is [6],

$$p(r_j) = \left(\pi\sqrt{A_j^2 - r_j^2}\right)^{-1}, \tag{B3}$$

and so, assuming that the position for each axis is independent,

$$p(\mathbf{r}) = \prod_{j\in(x,y,z)} p(r_j) = \prod_{j\in(x,y,z)} \left(\pi\sqrt{A_j^2 - r_j^2}\right)^{-1}. \tag{B4}$$

Typically, the neutral buffer gas is confined in a potential which is approximately harmonic at the centre of the trap, such that the density of the buffer gas follows a Gaussian density distribution,

$$\rho(\mathbf{r}) = \rho_x(r_x)\rho_y(r_y)\rho_z(r_z), \tag{B5}$$

where,

$$\rho_j(r_j) = \frac{1}{\sqrt{2\pi}\sigma_j} e^{-\frac{r_j^2}{2\sigma_j^2}}. \tag{B6}$$

Substituting Eqs. (B1), (B4) and (22) (see main text) into Eq. (B2) and evaluating the integral produces,

$$p(\mathbf{r}|c) = \prod_{j\in(x,y,z)} \frac{\exp\left(\frac{1}{2}\left(\frac{A_j^2-2r_j^2}{2\sigma_j^2}\right)\right)}{\pi\sqrt{A_j^2 - r_j^2}\, I_0\left(\frac{A_j^2}{4\sigma_j^2}\right)}, \tag{B7}$$

where $I_n(x)$ is the modified Bessel function of the first kind [3]. Employing a change of variables $r_j = A_j \cos\tilde{\phi}_j$, we obtain the distribution for the instantaneous secular phase for the motion along each axis at the time of a collision,

$$f_{\tilde{\phi}_j}(\tilde{\phi}_j|c) = \frac{1}{2\pi} \frac{e^{-\frac{A_j^2\cos(2\tilde{\phi}_j)}{4\sigma_j^2}}}{I_0\left(\frac{A_j^2}{4\sigma_j^2}\right)}. \tag{B8}$$

Appendix C. Analytical expression for $\langle \eta_1 \rangle$

In the main text, it is demonstrated that in the presence of a buffer gas confined in a harmonic potential, the change in the secular energy of an ion as the result of a collision is approximated by,

$$E' \approx (\eta_0 - \eta_1 E)E + \epsilon. \tag{C1}$$

Here, we provide an expression for the expectation value of η_1, i.e., the value averaged over all the variables contributing to the outcome of a collision. This value is obtained by substituting $\phi_j = \tilde{\phi}_j - \beta_j\tau$ into the expression for η as given in the Supplementary Material of Ref. [7], then averaging the result over the distributions for $\tilde{\phi}_j$ derived in Appendix B. Applying Eq. (27) of the main text to this expression produces a set of terms independent of the secular energy, i.e., η_0, and a set of terms linearly proportional to the energy, i.e., η_1. Averaging the terms contributing to η_1 over the remaining variables produces,

$$\langle\eta_1\rangle = \int\int\int \frac{f_\tau(\tau) f_{\theta_\rho}(\theta_\rho) f_{\phi_\rho}(\phi_\rho)}{(\tilde{m}+1)^2 k_B T_b} \sum_{j\in(x,y,z)} \frac{F_j(\tau) P_j^2 \omega_{j,b}^2}{\mathrm{c}_{0,j}^2 W_j^2 \omega_j^2} d\tau d\theta_\rho d\phi_\rho, \tag{C2}$$

where the remaining average over τ is left unevaluated due to the complexity of integrals involving the Mathieu functions, and the averages over ϕ_ρ, θ_ρ are left unevaluated due to the lack of an accurate analytical form for the distributions of these two variables. In the above, P_j is defined as in the main text and is a function of θ_ρ, ϕ_ρ (see text following Eq. (12)) and the function $F_x(\tau)$ is defined by,

$$\begin{aligned}
&F_x(\tau) = \\
&\quad \frac{\tilde{m}^3 c_{0,y}^2 W_x^2 \beta_y^2}{24 c_{0,x}^2 W_y^2 \beta_x^2}\Big[\mathrm{cs}_y(\tau)\left[2\dot{\mathrm{ce}}_x(\tau)\dot{\mathrm{se}}_x(\tau)\sin(2\tau\beta_x) + \left(\dot{\mathrm{ce}}_x(\tau)^2 - \dot{\mathrm{se}}_x(\tau)^2\right)\cos(2\tau\beta_x)\right]\Big] \\
&\quad + \frac{\tilde{m}^3 c_{0,z}^2 W_x^2 \beta_z^2}{24 c_{0,x}^2 W_z^2 \beta_x^2}\Big[\mathrm{cs}_z(\tau)\left[2\dot{\mathrm{ce}}_x(\tau)\dot{\mathrm{se}}_x(\tau)\sin(2\tau\beta_x) + \left(\dot{\mathrm{ce}}_x(\tau)^2 - \dot{\mathrm{se}}_x(\tau)^2\right)\cos(2\tau\beta_x)\right]\Big] \\
&\quad + \frac{\tilde{m}^3}{12}\Big[\mathrm{cs}_x(\tau)\dot{\mathrm{ce}}_x(\tau)\dot{\mathrm{se}}_x(\tau)\sin(2\tau\beta_x)\Big] \\
&\quad + \frac{\tilde{m}^2}{4} W_x\Big[\left(\mathrm{se}_x(\tau)\dot{\mathrm{se}}_x(\tau) - \mathrm{ce}_x(\tau)\dot{\mathrm{ce}}_x(\tau)\right)\sin(2\tau\beta_x) \\
&\qquad\qquad + \left(\dot{\mathrm{ce}}_x(\tau)\mathrm{se}_x(\tau) + \mathrm{ce}_x(\tau)\dot{\mathrm{se}}_x(\tau)\right)\cos(2\tau\beta_x)\Big] \\
&\quad + \frac{\tilde{m}^3}{12}\Big[3\mathrm{ce}_x(\tau)\mathrm{se}_x(\tau)\left(\dot{\mathrm{ce}}_x(\tau)^2 + \dot{\mathrm{se}}_x(\tau)^2\right)\sin(2\tau\beta_x) \\
&\qquad + \left(\dot{\mathrm{ce}}_x(\tau)^2\left(2\mathrm{ce}_x(\tau)^2 - \mathrm{se}_x(\tau)^2\right) + \dot{\mathrm{se}}_x(\tau)^2\left(\mathrm{ce}_x(\tau)^2 - 2\mathrm{se}_x(\tau)^2\right)\right)\cos(2\tau\beta_x)\Big]
\end{aligned} \tag{C3}$$

where $\mathrm{cs}_j(\tau) = \mathrm{ce}_j(\tau)^2 + \mathrm{se}_j(\tau)^2$. The functions $F_y(\tau)$ and $F_z(\tau)$ have the same general form and are found by switching a pair of indices, e.g, $F_y(\tau)$ is found by replacing x with y and vice versa.

Appendix D. Moments of superstatistical distributions

For a general energy distribution which can be expressed as a superposition of thermal states, i.e.,

$$f_E(E) = \int \frac{E^k}{(k_B T)^{k+1}\Gamma(k+1)} f_T(T) e^{-\frac{E}{k_B T}} dT, \tag{D1}$$

the moments are given by,

$$\langle E^n\rangle = \int E^n f_E(E) dE = \int\int E^n \frac{E^k}{(k_B T)^{k+1}\Gamma(k+1)} f_T(T) e^{-\frac{E}{k_B T}} dT dE. \tag{D2}$$

Exchanging the order of integration to first integrate over E produces,

$$\langle E^n \rangle = k_B^n \frac{\Gamma(k+n+1)}{\Gamma(k+1)} \int f_T(T) T^n dT, \tag{D3}$$

for $k+n > -1$ and where the terms independent of T have been moved outside the integral. The integral itself is the definition of the expectation value of T^n, i.e., $\langle T^n \rangle$ [5]. Thus,

$$\langle E^n \rangle = k_B^n \frac{\Gamma(k+n+1)}{\Gamma(k+1)} \langle T^n \rangle. \tag{D4}$$

Appendix E. The Bessel-Tsallis distribution

When the buffer gas follows a Gaussian density distribution, the change in the mean energy with each collision can be approximated by,

$$\langle E' \rangle = \langle \eta_0 \rangle \langle E \rangle - \langle \eta_1 \rangle \langle E^2 \rangle + \langle \epsilon \rangle, \tag{E1}$$

and using Eq. (D4) we obtain,

$$\langle T' \rangle = \langle \eta_0 \rangle \langle T \rangle - 4k_B \langle \eta_1 \rangle \langle T^2 \rangle + \frac{\langle \epsilon \rangle}{3k_B}. \tag{E2}$$

We assume that the multiplicative term η is the most significant source of noise, and so in the recurrence relation of the random variable T the other variables can be treated as constants. We have shown previously that multiplication of the energy by a random value is equivalent to multiplying the temperature by the same random value, that is, $E' = \eta E$ is equivalent to $T' = \eta T$ [8]. Neglecting the fluctuations in both the additive term and η_1, a suitable recurrence relation for the random variable T is,

$$T' = \eta_0 T - 4k_B \langle \eta_1 \rangle T^2 + \kappa T_b, \tag{E3}$$

where $\kappa T_b = \langle \epsilon \rangle / (3k_B)$ and κ is defined as in Eq. (33) of the main text. We solve this recurrence relation using the method in Ref. [9] by converting it to a Langevin equation for the variable $x = \ln T$. We first consider the case where $\langle \eta_1 \rangle = 0, \kappa T_b = 0$ to establish a suitable representation for the noise term η_0. Since $T' = \eta_0 T$, it follows that,

$$\ln T' = \ln \eta_0 + \ln T. \tag{E4}$$

We approximate the finite difference by a differential, $\frac{dx}{dt} = \ln T' - \ln T$, and separate $\ln \eta_0$ into its mean value $\mu = \langle \ln \eta_0 \rangle$ and a fluctuating term, $\hat{\zeta}(t)$, such that,

$$\frac{dx}{dt} = \mu + \hat{\zeta}(t). \tag{E5}$$

This has converted the multiplicative stochastic process in terms of T and η into an additive stochastic process in terms of x and $\ln \eta$. By itself, Eq. (E5) does not produce a stable steady-state distribution for x [9], and it does not include the effects of the temperature of the buffer gas or the reduction in η due to localisation. To find a representation for these terms, we use a different approximation for the derivative, $\frac{dx}{dt} \approx \frac{T'-T}{T}$ following Ref. [9]. Using this approximation with Eq. (E3) produces,

$$\frac{dx}{dt} \approx \langle \eta_0 \rangle + \hat{\eta}(t) - 1 - 4k_B \langle \eta_1 \rangle e^{x} + \kappa T_b e^{-x}, \tag{E6}$$

where we have separated η_0 into its mean $\langle \eta_0 \rangle$ and a fluctuating term $\hat{\eta}(t)$ as before. Notice that since we have used a less accurate approximation for $\frac{dx}{dt}$, this equation is defined in terms of $\eta_0 - 1$ rather than $\ln \eta_0$. However, $\langle \eta_0 \rangle - 1 \approx \mu$ and the variances of $\hat{\eta}(t)$ and $\hat{\zeta}(t)$ are approximately equal [9], such that Eq. (E6) is approximately equivalent to Eq. (E5) in the limit where $T_b = 0$ and $\langle \eta_1 \rangle = 0$. Moreover, this approximation provides a representation for the effects of both a non-zero buffer gas temperature and the non-uniform density of the buffer gas. We therefore augment Eq. (E5) with the terms proportional to T_b and $\langle \eta_1 \rangle$ of Eq. (E6) to produce,

$$\frac{dx}{dt} = \mu + \hat{\zeta}(t) + \kappa T_b e^{-x} - 4\langle \eta_1 \rangle k_B e^{x}. \tag{E7}$$

We make the approximation that $\hat{\zeta}(t)$ can be modelled as following a Gaussian distribution, i.e., it represents the fluctuations in x averaged over multiple collisions [10]. This follows from the fact that the fluctuations in x are additive, and the sum of independent random variables approaches a Gaussian distribution by the central limit theorem [10]. This approximation enables the derivation of an analytically tractable Fokker-Planck equation for the probability distribution $f_x(x)$ [9],

$$\frac{\sigma^2}{2} \frac{d^2}{dx^2} f_x(x) - \frac{d}{dx} \left[(\mu + \kappa T_b e^{-x} - 4\langle \eta_1 \rangle k_B e^{x}) f_x(x) \right] = 0, \tag{E8}$$

where σ^2 is the variance of $\hat{\zeta}(t)$. If $\langle \eta_1 \rangle = 0$, then this equation reduces to the one obtained in Ref. [8], and a steady-state solution exists if $\mu < 0$ and $T_b \neq 0$. Conversely, if $T_b = 0$, then a solution exists only if $\langle \eta_1 \rangle$ is non-zero and $\mu > 0$. These conditions correspond to the overall drift of x towards a lower or an upper bound [9]. We proceed assuming that both $T_b, \langle \eta_1 \rangle$ are non-zero, such that both upper and lower bounds exist, and the existence of a steady-state does not depend on the sign of μ. Subject to the boundary conditions that $f_x(x) \to 0$ for $x \to \pm\infty$, the steady-state distribution for T is,

$$f_T^{(L)}(T) = \frac{2^{\nu-1} \left(\frac{b}{\nu}\right)^{-\frac{\nu}{2}} T^{-\nu-1} \left(\frac{k_B^2}{E_\ell}\right)^{-\frac{\nu}{2}} e^{-\frac{\nu}{b k_B T} - \frac{k_B T}{4E_\ell}}}{K_\nu\left(\sqrt{\frac{\nu}{bE_\ell}}\right)}, \tag{E9}$$

where $K_y(z)$ is the modified Bessel function of the second kind with order y and argument z, and the superscript (L) is used to indicate that this is the distribution obtained in the presence of a localised buffer gas. The parameters are defined in terms of the coefficients of Eq. (E8) as,

$$b = \frac{-\mu}{k_B \kappa T_b}, \tag{E10}$$

$$\nu = \frac{-2\mu}{\sigma^2}, \tag{E11}$$

$$E_\ell = \frac{\sigma^2}{32\langle\eta_1\rangle}, \tag{E12}$$

and the distribution is normalisable if $E_\ell > 0$ and $b/\nu > 0$. These definitions for the parameters are accurate in the limit in which $\hat{\zeta}(t)$ can be approximated as following a Gaussian distribution, but do not take into account corrections due to the exact form of the distribution of $\ln\eta_0$ [8, 9].

The energy distribution is defined as a superposition of thermal distributions,

$$f_E(E) = \int f_E(E|T) = \frac{E^k}{(k_B T)^{k+1}\Gamma(k+1)} e^{-\frac{E}{k_B T}} f_T(T) dT. \tag{E13}$$

Evaluating Eq. (E13) using Eq. (E9) produces,

$$f_E^{(BT)}(E) = \frac{E^k \left(\frac{bE}{\nu}+1\right)^{-\frac{1}{2}(k+\nu+1)} \left(\frac{b}{\nu E_\ell}\right)^{\frac{k+1}{2}} K_{k+\nu+1}\left(\sqrt{\frac{E}{E_\ell}+\frac{\nu}{bE_\ell}}\right)}{2^{k+1}\Gamma(k+1) K_\nu\left(\sqrt{\frac{\nu}{bE_\ell}}\right)}. \tag{E14}$$

The moments of this distribution are difficult to evaluate directly due to the complexity of integrals involving the Bessel function. However, the moments of Eq. (E9) may be easily calculated by evaluating $\int T^n f_T(T)dT$, and applying Eq. (D4) produces,

$$\langle E^n\rangle = \frac{2^n\Gamma(k+n+1)\left(\frac{b}{\nu E_\ell}\right)^{-\frac{n}{2}} K_{\nu-n}\left(\sqrt{\frac{\nu}{bE_\ell}}\right)}{\Gamma(k+1)K_\nu\left(\sqrt{\frac{\nu}{bE_\ell}}\right)}. \tag{E15}$$

The mean energy $\langle E\rangle$ evaluated using this expression is defined as long as $E_\ell > 0$ and $b/\nu > 0$.

Appendix F. Parameter estimation

Although the values of $\mu, \sigma, \langle\eta_1\rangle$ required to calculate ν, b, E_ℓ are in theory defined in terms of the mass ratio and trapping parameters, in practice these cannot be accurately evaluated a priori due to the fact that the distributions of $\ln\eta_0$, ϕ_ρ and θ_ρ are not known analytically. Instead, the required values can be obtained from numerical simulations by sampling the distribution of η. To do so, a series of collisions is simulated to produce a value of E under the same conditions as for the simulations used to obtain the energy distribution. For the final collision, the buffer gas temperature is set to $T_b = 0$ K, such that the change in energy is given by $E' = \eta E$. Dividing the post-collision energy by the pre-collision energy provides a value for η, and repeating this process (typically for 1'000'000 iterations) produces a set of values of E, η. From these, the coefficients for the linear expansion $\eta = \eta_0 - \eta_1 E$ are obtained by least-squares linear regression, and we take $\langle\eta_1\rangle$ to be equal to the value of η_1 extracted by this method. If the density of the buffer gas is set to a uniform distribution for the final collision in addition to setting the temperature equal to zero, the η_1 term is eliminated, and we have $\eta = \eta_0$. The values of $\mu = \langle \ln \eta_0 \rangle$ and $\sigma^2 = \langle (\ln \eta_0)^2 \rangle - \mu^2$ are then calculated from the values of η_0 obtained in this manner. This method is in general found to produce acceptable results except for the particular case of $\tilde{m} = 1, \omega_{r,b} = 100$ Hz, for which the ion's energy in the steady-state remains small enough that η_1 cannot be accurately estimated from collisions. Consequently, for that mass ratio η_1 is obtained by setting the initial temperature of the ion to $T = 1$ K and performing a single collision, rather than allowing the ion to reach the steady-state first. This leads to a slightly different value of η_1 than would be obtained in the steady-state, but in practice for this mass ratio the energy of the ion in the steady-state remains sufficiently low that the effect of localisation is unimportant, i.e., $E << E_\ell$, and so the error in η_1 does not significantly affect the shape of the distribution. Indeed, the steady-state energy distribution is found to essentially follow Tsallis statistics over the range of energies sampled in numerical simulations.

Alternatively, the distribution may be fit to numerical data through maximum-likelihood estimation. Analytical expressions for the maximum-likelihood estimates of the parameters b, ν, E_ℓ have not yet been obtained due to the complexity of derivatives of the Bessel function with respect to ν. Thus, the estimation is performed numerically with respect to the parameters $\tilde{b} = b/\nu, \nu, E_\ell$. The use of $\tilde{b}$ ensures that this parameter is strictly positive, reducing the range of values to optimise over and eliminating the constraint that b must have the same sign as ν. The parameters of the exponential-Tsallis distribution and the standard Tsallis distribution are also found through numerical maximum likelihood estimation.

References

[1] D.J. Berkeland, J.D. Miller, J.C. Bergquist, W.M. Itano and D.J. Wineland, J. Appl. Phys. **83**, 5025 (1998).

[2] W.E. Boyce, R.C. DiPrima and D.B. Meade, *Boyce's Elementary Differential Equations and Boundary Value Problems*, 11th ed. (Wiley, NJ, 2017).

[3] F.W.J. Olver, D.W. Lozier, R.F. Boisvert and C.W. Clark, editors, *NIST Handbook of Mathematical Functions* (Cambridge University Press, New York, NY, 2010).

[4] D.J. Wineland, C. Monroe, W.M. Itano, D. Leibfried, B.E. King and D.M. Meekhof, J. Res. Natl. Inst. Stan. **103**, 259 (1998).

[5] K. Riley, M. Hobson and S. Bence, *Mathematical Methods For Physics and Engineering*, 3rd ed. (Cambridge University Press, Cambridge, 2010).

[6] R. Robinett, Am. J. Phys. **63** (9), 823–832 (1995).

[7] I. Rouse and S. Willitsch, Phys. Rev. A **97** (4), 042712 (2018).

[8] I. Rouse and S. Willitsch, Phys. Rev. Lett. **118** (14), 143401 (2017).

[9] D. Sornette and R. Cont, J. Phys. I **7** (3), 431–444 (1997).

[10] J. Honerkamp, *Statistical Physics: An Advanced Approach with Applications*, 2nd ed. (Springer, Berlin, 2002).