Superstatistical Energy Distributions of an Ion in an Ultracold Buffer Gas

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An ion in a radio frequency ion trap interacting with a buffer gas of ultracold neutral atoms is a driven dynamical system which has been found to develop a nonthermal energy distribution with a power law tail. The exact analytical form of this distribution is unknown, but has often been represented empirically by q-exponential (Tsallis) functions. Based on the concepts of superstatistics, we introduce a framework for the statistical mechanics of an ion trapped in an rf field subject to collisions with a buffer gas. We derive analytic ion secular energy distributions from first principles both neglecting and including the effects of the thermal energy of the buffer gas. For a buffer gas with a finite temperature, we prove that Tsallis statistics emerges from the combination of a constant heating term and multiplicative energy fluctuations. We show that the resulting distributions essentially depend on experimentally controllable parameters paving the way for an accurate control of the statistical properties of ion-atom hybrid systems.

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The advent of hybrid systems of cold ions immersed in ultracold neutral atoms has opened up new perspectives for exploring two- and many-body effects in a regime intermediate between strong ion-ion and weak neutral-neutral couplings [1–3]. A range of applications in atomic, molecular, and chemical physics has recently emerged including studies of ion-neutral collisions and chemical reactions at very low energies [4–8], of many-body physics in dense systems [9,10], and of the quantum dynamics of an ion under the influence of an ultracold buffer gas [11,12].

Ion-atom hybrid systems are realized by superimposing cold ions in a radio frequency (rf) trap with trapped ultracold atoms [1–3]. rf traps use rapidly oscillating electric fields to dynamically confine the ions. In an adiabatic regime [13], the resulting motion of an ion can be represented as a thermal component ("secular motion") superimposed by small-amplitude oscillations at the rf frequency ("micromotion") [13]. In a hybrid trap, the ion undergoes frequent collisions with neutral atoms which disrupt its motion and lead to energy exchange between the secular motion and the rf field [14–19].

These processes lead to a distortion of the ion's secular-energy distribution from thermal (Boltzmann) to one better described by a power law at high energy [16–18,20]. The precise knowledge of the ion energetics is crucial for understanding the properties and dynamics of hybrid systems and their derived applications. Consequently, this problem has been the subject of intense recent research [16–19,21]. In the high-energy limit, expressions for the mean energy and the power-law exponent have been derived [18]. The complete ion-energy distribution has often been modeled [12,16,22] by Tsallis (q-exponential) functions [23,24],

$$e_q(x) = [1 + (1 - q_T)x]^{1/(1-q_T)} \tag{1}$$

for $q_T > 1$, where C_q is a normalization factor. q_T is a parameter which characterizes the deviation from a standard exponential function which is recovered in the limit $q_T \to 1$. However, the application of q exponentials has remained empirical [12,21,22] since their first introduction for fitting numerical energy distributions [16].

Using the formalism of superstatistics [25,26], we introduce a framework for the statistical mechanics of ion-atom hybrid systems. We derive analytic ion secular-energy distributions both neglecting and including the thermal energy of the ultracold buffer gas and confirm their validity by comparison with numerical simulations. For a buffer gas at zero kelvin, we obtain an energy distribution with no steady state and an exponential decay at high energies. For a buffer gas at finite temperature, we prove from first principles the emergence of Tsallis statistics, thus vindicating its application in the present context. The energy distributions derived here depend on experimentally adjustable parameters, which opens the door for a rational experimental control of the statistical properties of ion-atom hybrid systems.

The motion in each direction r_j, $j \in (x, y, z)$, of an ion in a quadrupole rf trap is given by the Mathieu differential equations,

$$\ddot{r}_j(\tau) + [a_j - 2q_j \cos(2\tau)]r_j = 0, \tag{2}$$

where $\tau = \Omega t/2$ and q_j, a_j are the Mathieu stability parameters [27]. In a hybrid system, the ion interacts with neutral atoms through a polarization potential. We treat the dynamics as a series of elastic collisions in the Langevin approximation with an energy-independent rate [18,28]. The velocity $\mathbf{v}$ of the ion after a collision is [17,18],

$$\mathbf{v}' = \frac{1}{1+\tilde{m}}\mathbf{v} + \frac{\tilde{m}}{1+\tilde{m}}\mathbf{v}_n + \frac{\tilde{m}}{1+\tilde{m}}R \cdot (\mathbf{v} - \mathbf{v}_n), \tag{3}$$

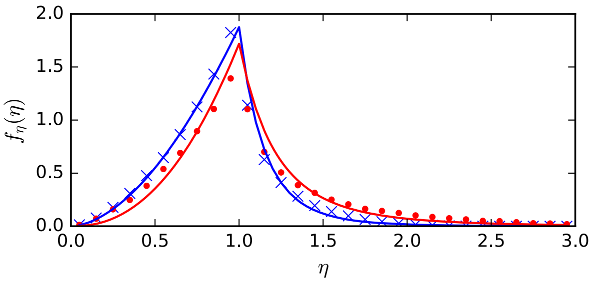

FIG. 1. Distributions of the energy-transfer factor η in ion-atom collisions for $q = 0.1$, $\tilde{m} = 0.75$ (blue crosses) and $q = 0.5$, $\tilde{m} = 1.25$ (red points) starting from a thermal state with ion temperature $T_0 = 1$ mK. The points are binned normalized data from 100 000 numerical simulations of a collision. The lines represent an empirical asymmetric log-Lapace distribution, see text.

where $\tilde{m} = m_n/m_i$ is the ratio of the atom's to the ion's mass, R is a rotation matrix [29], and $\mathbf{v}_n$ is the velocity of the neutral atom. Primes refer to postcollision quantities. As we have no control over the instantaneous velocities of the particles at the time of collisions, $\mathbf{v}$ and $\mathbf{v}_n$ are random variables. Generally, we denote the distribution f of a random variable x as $f_x(y)$, where the argument y indicates the variable in which this function is expressed.

From Eq. (3), the ion's secular energy after a collision can be derived to be [30],

$$E' = \eta E + c_1\sqrt{E\epsilon} + c_2\epsilon, \tag{4}$$

where $\epsilon = (m_n/2)|\mathbf{v_n}|^2$ is the kinetic energy of the neutral atom and η, c_1, c_2 are coefficients [30]. Assuming that the buffer gas density is uniform, these coefficients are independent of the values of E and ϵ. For an ion much hotter than the buffer gas ($E \gg \epsilon$), we approximate $E' \approx \eta E$. The stability of the ion motion in the buffer gas with respect to runaway heating is determined by the distribution of η. As a rule, the motion is stable for a mass ratio $\tilde{m} \lesssim 1.4$ for Mathieu parameters $q \ll 1$ [15–19].

Figure 1 shows numerical distributions $f_\eta(\eta)$ for the energy-transfer parameter η for $q = 0.1$, $\tilde{m} = 0.75$ and $q = 0.5$, $\tilde{m} = 1.25$ which correspond to stable and unstable ion motions, respectively. The numerical simulations were performed following DeVoe's approach [16]. The solid lines in Fig. 1 correspond to log-Laplace distributions of the form [35]

$$f_\eta(\eta) = \frac{1}{\delta}\frac{a_1 a_2}{a_1 + a_2}\begin{cases} \left(\frac{\delta}{\eta}\right)^{a_1+1} & \eta \geq \delta \\ \left(\frac{\eta}{\delta}\right)^{a_2-1} & 0 < \eta < \delta, \end{cases} \tag{5}$$

with $a_1, a_2 > 0$ which have previously been used to model processes involving multiplicative fluctuations [35]. The parameter δ representing the maximum of the distribution was found to be ≈ 1, reflecting the fact that most collisions result in minor changes to the ion's energy. The values of a_1 and a_2 may be estimated by calculating the first and second moment $\langle\eta\rangle$ and $\langle\eta^2\rangle$, respectively, of the distribution using Eq. (5) and matching them to the expressions found numerically from Eq. (4).

Let us now assume that the ion is initially prepared in a thermal state at temperature T_0, as may be the situation after Doppler laser cooling [36,37]. The resulting distribution for the ion's initial energy E_0 is

$$f_{E_0}(E_0) = \frac{E_0^k \beta_0^{k+1}}{\Gamma(k+1)} e^{-E_0\beta_0}, \tag{6}$$

where $\beta_0 = 1/(k_B T_0)$, Γ is the Gamma function, and the preexponential factor represents the density of states ($k = 2$ for a three-dimensional harmonic oscillator [38]).

We now consider the effects of collisions with the neutral atoms. Initially, we neglect their thermal energy and set $\epsilon = 0$ in Eq. (4) such that $E' = \eta E_0$. The resulting energy distribution can be written as [39],

$$\begin{aligned} f_{E'}(E') &= \int_{\eta=0}^{\eta=\infty} \frac{1}{\eta} f_{E_0}(E'/\eta) f_\eta(\eta) d\eta \\ &= \int_{\eta=0}^{\eta=\infty} \frac{1}{\eta}\frac{(E'/\eta)^k \beta_0^{k+1}}{\Gamma(k+1)} e^{-(E'/\eta)\beta_0} f_\eta(\eta) d\eta. \end{aligned} \tag{7}$$

We first consider the case in which every collision multiplies the energy by a fixed amount η_c. The distribution for η is then given by a Dirac δ function,

$$f_\eta(\eta) = \delta(\eta - \eta_c), \tag{8}$$

so that

$$f_{E'}(E') = \frac{E'^k \beta_0^{k+1}}{\eta_c^{k+1}\Gamma(k+1)} e^{-E'\beta_0/\eta_c}. \tag{9}$$

This is still a thermal distribution, except that it can now be written in terms of $\beta' = \beta_0/\eta_c$.

We now generalize this approach to an arbitrary $f_\eta(\eta)$ by making the change of variables $\beta' = \beta_0/\eta$ in Eq. (7),

$$f_{E'}(E') = \int_{\beta'=0}^{\beta'=\infty} \frac{E'^k \beta'^{k+1}}{\Gamma(k+1)} e^{-E'\beta'} \frac{\beta_0}{\beta'^2} f_\eta\left(\frac{\beta_0}{\beta'}\right) d\beta'. \tag{10}$$

The energy distribution after a collision can thus be represented by a superposition of thermal states. This problem can be treated within the formalism of superstatistics, i.e., the superpositions of several statistics as in our case the ones of η and E in Eq. (7) [25,26,40].

We can now define a distribution for β',

$$f_\beta(\beta') = \frac{\beta_0}{\beta'^2} f_\eta\left(\frac{\beta_0}{\beta'}\right), \tag{11}$$

which is used to recast Eq. (10) into the form

$$f_{E'}(E') = \int_{\beta'=0}^{\beta'=\infty} \frac{E'^k \beta'^{k+1}}{\Gamma(k+1)} e^{-E'\beta'} f_\beta(\beta') d\beta'. \quad (12)$$

Equation (12) has the form of a Laplace transform $\mathcal{L}$. For general distributions $f_\beta(\beta)$, $f_\eta(\eta)$ one gets

$$f_\beta(\beta') = \int_{\eta=0}^{\eta=\infty} \eta f_\beta(\eta\beta') f_\eta(\eta) d\eta. \quad (13)$$

A repeated application of Eq. (13) and substitution into Eq. (12) can then be performed to obtain the energy distribution of an ion after n collisions.

Thus, we formulate a recurrence relation for β after collision number i,

$$\beta_i = \beta_{i-1}/\eta_i. \quad (14)$$

Since the ion is initially in a thermal state, we take β_0 to be constant. After n collisions starting from β_0, we get

$$\beta_n = \beta_0 \prod_{i=1}^{n} 1/\eta_i. \quad (15)$$

Each value of η is assumed to be independently and identically distributed, and so by applying the central limit theorem the product $\prod_{i=1}^{n} 1/\eta_i$ is log-normally distributed for large n [39]. Hence, from Eq. (11) we write

$$f_{\beta_n}(\beta_n) = \frac{1}{\sqrt{2\pi n}\sigma\beta_n} \exp\left(-\frac{(\ln\beta_n - \ln\beta_0 + n\mu)^2}{2n\sigma^2}\right), \quad (16)$$

where $\mu = \langle \ln\eta \rangle$ and $\sigma^2 = \langle (\ln\eta)^2 \rangle - \langle \ln\eta \rangle^2$.

We now return to the energy distribution. By inserting Eq. (16) into Eq. (12), we obtain,

$$f_{E_n}(E_n) = \int_{\beta_n=0}^{\beta_n=\infty} \frac{E_n^k \beta_n^{k+1}}{\Gamma(k)} e^{-E_n\beta_n} \times \frac{1}{\sqrt{2\pi n}\sigma\beta_n} \exp\left(-\frac{(\ln\beta_n - \ln\beta_0 + n\mu)^2}{2n\sigma^2}\right) d\beta_n. \quad (17)$$

We use the Laplace integration method [41] to find an approximate analytical solution for $k = 2$. We obtain

$$f_{E_n}(E_n) = \frac{\hat{\beta}^3 E_n^2}{4\sqrt{\hat{\beta}E_n n\sigma^2 + 1}} \exp(-\hat{\beta}E_n) \times \left[\mathrm{erf}\left(\sqrt{\frac{\hat{\beta}E_n n\sigma^2 + 1}{2n\sigma^2}}\right) + 1\right] \times \exp\left(-\frac{n\sigma^2}{2}(\hat{\beta}E_n - 2)^2\right), \quad (18)$$

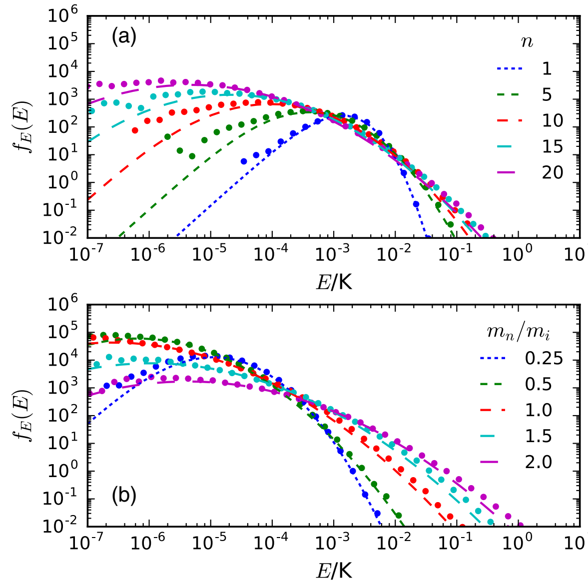

FIG. 2. (a) Energy distributions of an ion in a rf trap after n collisions with a neutral buffer gas at zero kelvin with a mass ratio $\tilde{m} = m_n/m_i = 1.5$. (b) The ion-energy distribution after 25 collisions at a range of mass ratios. The lines show corresponding energy distributions computed with Eq. (18). The points show numerical data sampled after 100 000 simulations.

where $\hat{\beta}$ is the point at which the integrand of Eq. (17) is maximal. In the high-energy limit for $k = 0$, Eq. (18) has been shown to exhibit an exponential decay [42,43]. From the general property of the Laplace transform,

$$\mathcal{L}[\beta^{k+1} f_\beta(\beta)] = (-1)^{k+1} \frac{d^{k+1}}{dE^{k+1}} \mathcal{L}[f_\beta(\beta)], \quad (19)$$

it follows that if the high-energy behavior for $k = 0$ is an exponential decay, then this holds true for any integer value of k. Thus, we conclude that a purely multiplicative model of the heating process does not lead to Tsallis statistics, which is characterized by a power-law tail for the distribution at high energies.

In order to test the validity of Eq. (18), a series of simulations were performed at a buffer gas temperature $T = 0$ K and varying the mass ratio or number of collisions. The results are plotted in Fig. 2 along with the distributions computed from Eq. (18). The μ and σ parameters were computed from numerical distributions $f_\eta(\eta)$ such as the ones shown in Fig. 1. At low collision numbers, the agreement is generally poor, which is expected due to the assumption in the derivation of Eq. (18) that the central limit theorem can be applied. Moreover, for all collision numbers, the agreement is not as good at low energies due to the Laplace integration method being valid only in the limit $E \to \infty$. However, for higher energies and numbers of collisions, Eq. (18) becomes an increasingly better representation of the simulated data.

For comparison, the numerical data for 25 collisions at a mass ratio of 1.0 are presented in Fig. 3 together with the

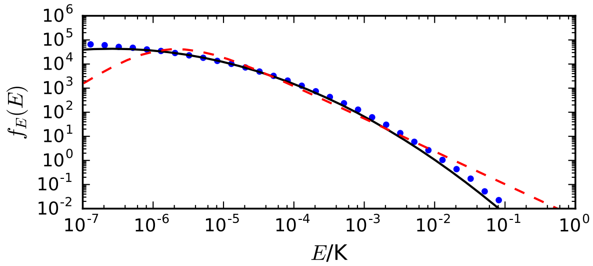

FIG. 3. Comparison between the ion-energy distribution Eq. (18) for a buffer gas at 0 K (black dashed line) and a Tsallis distribution (red dashed line) for an ion in a Paul trap after 25 collisions with a mass ratio 1.0. The points represent numerical data sampled from 100 000 simulations.

distribution predicted using Eq. (18). The red dashed line represents a Tsallis distribution obtained from a maximum-likelihood estimation (MLE) to the numerical data. It can be clearly seen that Tsallis statistics is a poor match for a buffer gas at zero kelvin, while Eq. (18) provides much better agreement.

Neither the energy nor the β distributions, Eqs. (18) and (16), respectively, converge to a steady state with increasing n. This is a known property of an unbounded multiplicative random walk, and in the present case results from the neglect of the temperature of the buffer gas allowing the ion to reach an arbitrarily low temperature [20,44].

For a buffer gas at a finite temperature, we have to adopt a different procedure as the change of the ion energy following a collision is no longer a purely multiplicative process, see Eq. (4). Because the buffer gas velocity distribution is isotropic, the c_1 coefficient averages to zero such that it can be neglected. We are thus left with $E' = \eta E + c_2\epsilon$. Assuming again that the ion's energy distribution can be represented as a superposition of thermal states as in Eq. (10), it follows that $\langle E\rangle = (1+k)k_B\langle T\rangle$. This suggests that we can rephrase the problem of finding an energy distribution to one of finding the underlying temperature distribution. We approximate that the contributions from ϵ in Eq. (4) can be treated as a constant source of heating proportional to the temperature of the buffer gas T_a which ensures that the ion's steady-state temperature is nonzero. This is a good approximation if the thermal fluctuations of the buffer gas are much smaller than the ones of the ion. The ion temperature after a collision is then

$$T_i = \eta_i T_{i-1} + \kappa T_a, \tag{20}$$

where κ is a heating coefficient [30].

To find the required temperature distribution, we solve the recurrence relation Eq. (20). The mathematical solution of this problem has been outlined in Refs. [44,45] and leads to a gamma distribution for β:

$$f_\beta(\beta) = \frac{1}{\beta\Gamma(n_T)} e^{-\beta n_T/\langle\beta\rangle}\left(\frac{\beta n_T}{\langle\beta\rangle}\right)^{n_T}. \tag{21}$$

Multiplying by the density of states and applying the Laplace transform we obtain the ion-energy distribution

$$f_{E,T}(E) = \left(\frac{\langle\beta\rangle}{n_T}\right)^{k+1}\frac{\Gamma(k+n_T+1)}{\Gamma(k+1)\Gamma(n_T)}\frac{E^k}{(\frac{\langle\beta\rangle E}{n_T}+1)^{k+n_T+1}}. \tag{22}$$

The parameter n_T can be obtained from the condition [44]

$$\int_{\eta=0}^{\eta=\infty} f_\eta(\eta)\eta^{n_T}d\eta = 1. \tag{23}$$

This integral may be solved numerically, or alternatively we make use of the empirical distribution equation (5). From substituting Eq. (5) into Eq. (23), we obtain,

$$n_T = a_1 - a_2 = \frac{\langle\eta\rangle - 4\langle\eta^2\rangle + 3\langle\eta\rangle\langle\eta^2\rangle}{\langle\eta\rangle - 2\langle\eta^2\rangle + \langle\eta\rangle\langle\eta^2\rangle}, \tag{24}$$

assuming $\delta = 1$ in Eq. (5). To fully characterize Eq. (22), we also require the value for $\langle\beta\rangle$. From Eq. (20), it follows that

$$\langle T\rangle = \langle\eta\rangle\langle T\rangle + \kappa T_a = \frac{\kappa T_a}{1-\langle\eta\rangle}. \tag{25}$$

Averaging $T = 1/(k_B\beta)$ over Eq. (21), we get

$$\langle T\rangle = \frac{1}{k_B\langle\beta\rangle}\frac{n_T}{n_T-1}. \tag{26}$$

Equating Eqs. (25) and (26), we find

$$\langle\beta\rangle = \frac{1}{k_B\kappa T_a}\frac{n_T}{n_T-1}(1-\langle\eta\rangle). \tag{27}$$

This derivation is valid only for $n_T > 1$ and $\langle\eta\rangle < 1$. If either of these conditions is not met, the mean temperature diverges because the ion motion becomes unstable.

The distribution Eq. (22) has the form of a q exponential Eq. (1) multiplied by a E^k term. For $k = 0$ (one dimensional), it reduces to the standard q exponential, and for $k = 2$ (three dimensional) it is equivalent to the form used in Ref. [12], if we set their exponent $n = n_T + 3$. We have therefore shown that Tsallis statistics are physically meaningful for the present problem under the condition that the variance of the thermal fluctuations are sufficiently small so that the additive noise due to the thermal energy of the atoms can be approximated as a constant.

Figure 4 shows a comparison of the MLE values of the parameters n_T and $1/\langle\beta\rangle$ extracted from numerical simulations with their predictions from Eqs. (24) and (27), respectively. Below the critical mass ratio given by the intersection of the curves with the gray horizontal line in Fig. 4(a), the ion motion is stable. Up to near this point, the predictions for both parameters are very close to the values

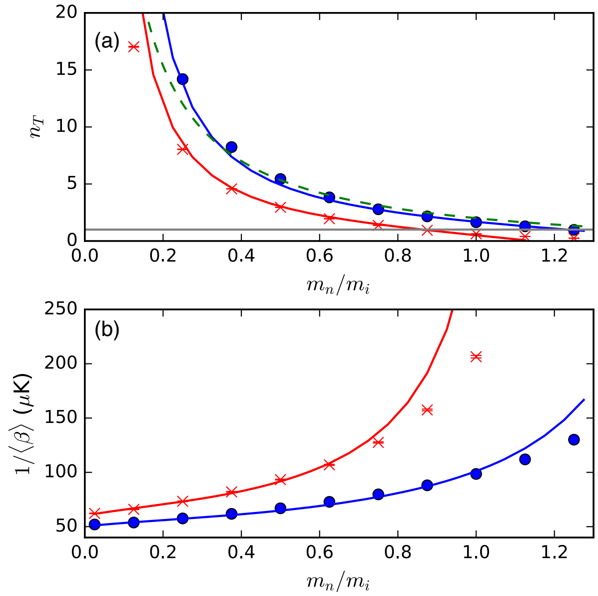

FIG. 4. (a) Tsallis parameter n_T at Mathieu parameter $q = 0.1$ (blue circles) and $q = 0.5$ (red crosses) calculated by a maximum likelihood estimation (MLE) of a Tsallis function to the steady-state ion-energy distribution obtained from numerical simulations (100 000 trials per point). The blue and red lines show the predictions using Eq. (24). The green dotted line indicates the approximate result for $q < 0.4$ from Ref. [18] and the gray horizontal line indicates the critical exponent $n_T = 1$ below which the mean energy is undefined. (b) As (a) for $1/\langle\beta\rangle$. Error bars correspond to the standard errors of the MLE values and are plotted when larger than the size of the symbols.

extracted from numerical data, vindicating the assumptions leading to the derivation of Eq. (21). Above the critical mass ratio, the predicted mean $\langle\beta\rangle$ becomes increasingly inaccurate as a result of energy correlations between different coordinate axes not accounted for in the present model (see [18,30]).

From Eqs. (22) and (23), it becomes clear that the energy distribution and therefore the statistical properties of the ion depend on the buffer gas temperature and the distribution $f_\eta(\eta)$. The latter depends on system parameters such as the atom-ion mass ratio and the Mathieu parameters of the trap which are defined in advance by the experimenter. By varying these parameters, $f_\eta(\eta)$ and therefore the Tsallis distribution Eq. (22) can be tuned in a deterministic manner, allowing for a control of the statistical properties of the system.

Beyond the current application, the formalism developed here represents a general framework for describing the statistical mechanics of an ion in a buffer gas which can be used to, e.g., compute thermodynamic functions [46]. The present treatment can also be extended to localized buffer gases. These developments will be reported elsewhere.

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