Active matter: quantifying the departure from equilibrium

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Active matter systems are driven out of equilibrium at the level of individual constituents. One widely studied class are systems of athermal particles that move under the combined influence of interparticle interactions and self-propulsions, with the latter evolving according to the Ornstein-Uhlenbeck stochastic process. Intuitively, these so-called active Ornstein-Uhlenbeck particles (AOUPs) systems are farther from equilibrium for longer self-propulsion persistence times. Quantitatively, this is confirmed by the increasing equal-time velocity correlations (which are trivial in equilibrium) and by the increasing violation of the Einstein relation between the self-diffusion and mobility coefficients. In contrast, the entropy production rate, calculated from the ratio of the probabilities of the position space trajectory and its time-reversed counterpart, has a non-monotonic dependence on the persistence time. Thus, it does not properly quantify the departure of AOUPs systems from equilibrium.

Introduction. – The focus of our study are model active matter systems consisting of athermal self-propelled particles, which move due to interparticle interactions and self-propulsions, with the self-propulsions evolving independently of the positions of the particles [1, 2]. To fully define these systems one needs to specify the properties of the self-propulsions. Two popular choices are active Brownian particles (ABPs) [3, 4] which are endowed with self-propulsions of a constant magnitude and evolving via rotational diffusion, and active Ornstein-Uhlenbeck particles (AOUPs) [5–7] for which the self-propulsions evolve according to the Ornstein-Uhlenbeck stochastic process. Since our fundamental understanding of non-equilibrium systems is not well developed, one often used approach to study systems of self-propelled particles is to approximate them by appropriately chosen equilibrium systems [6, 8, 9]. Intuitively, whether such an approach is sensible depends on how non-equilibrium the former systems are [10]. Thus, the recurring question in the recent literature is how to quantify the departure of the systems of self-propelled particles from equilibrium. To paraphrase a recent article [12], we want to replace a binary in-out of equilibrium classification by a more quantitative one.

One possibility is to generalize the stochastic thermodynamics approach and to evaluate the entropy production defined through a ratio of the probability of the forward trajectory and that of its time-reversed version. For systems of AOUPs this approach was first proposed by Fodor *et al.* [7] and then elaborated on by Puglisi, Marconi, Maggi and collaborators [13, 14]. They defined the entropy production in terms of the ratio of the probabilities of the forward and reversed trajectories in the position space and derived a compact expression which makes a numerical evaluation of the entropy production straightforward.

There have been other attempts to define the entropy production. Mandal *et al.* [15] defined the entropy production in terms of the ratio of the probabilities of the forward trajectory and a trajectory following time-reversed evolution. Dabelow *et al.* [16] argued that the relation between the entropy production and the trajectory probability ratio involves an additional quantity originating from the "mutual information" between the trajectory and the environment. Shankar and Marchetti [17] proposed calculating the entropy production from the ratio of the probabilities of the forward and reversed trajectories in the enlarged phase space consisting of the particle's position and self-propulsion. While they considered only a single free self-propelled particle, their approach was generalized by one of us to a single AOUP in an external potential [18]. Additionally, Pietzonka and Seifert [19] argued that the most fundamental consideration of entropy production should also include the contribution from physico-chemical processes that give rise to the self-propulsion.

Here we present results of a simulational investigation of the entropy production according to Fodor *et al.* for systems of interacting AOUPs. The most intuitive control parameter tuning the departure of these systems from equilibrium is the persistence time of the self-propulsion. We present quantitative numerical results supporting this expectation. Then, we show that the expression for the entropy production derived in Ref. [7] has a non-monotonic dependence on the self-propulsion persistence time. Thus, it is not a good measure of the departure of AOUP systems from equilibrium. Numerical evaluation of the alternative proposals to define the entropy production [15–18] is left for a future study.

Simulations. – We simulated interacting, athermal AOUPs [5–7], moving in a viscous medium, without inertia, under the combined influence of the interparticle forces and self-propulsions, with the latter evolving according to the Ornstein-Uhlenbeck stochastic process. The equations of motions read

$$\dot{\mathbf{r}}_i = \xi_0^{-1}[\mathbf{F}_i + \mathbf{f}_i], \tag{1}$$

$$\tau_p \dot{\mathbf{f}}_i = \mathbf{f}_i + \boldsymbol{\eta}_i. \tag{2}$$

In Eq. (1) $\mathbf{r}_i$ is the position of particle i, ξ_0 is the fric-

tion coefficient of an isolated particle, $\mathbf{F}_i$ is the interparticle force, and $\mathbf{f}_i$ is the self-propulsion. In Eq. (2) τ_p is the persistence time of the self-propulsion and $\boldsymbol{\eta}_i$ is the internal Gaussian noise with zero mean and variance $\langle \boldsymbol{\eta}_i(t)\boldsymbol{\eta}_j(t')\rangle_{\rm noise} = 2\xi_0 k_B T_a \mathbf{I}\delta_{ij}\delta(t-t')$, where $\langle\ldots\rangle_{\rm noise}$ denotes averaging over the noise distribution, T_a is the "active" temperature, and $\mathbf{I}$ is the unit tensor. We choose a system of units such that $\xi_0 = 1$ and $k_B = 1$. We emphasize that T_a characterizes the strength of the self-propulsion; it is called the active temperature because it determines the long-time diffusion coefficient of a single free AOUP, $D_0 = k_B T_a/\xi_0 \equiv T_a$.

We studied a 50:50 binary mixture of $N = 1000$ particles interacting via the smoothed Weeks-Chandler-Andersen truncation of the Lennard-Jones potential, $V_{\alpha\beta}(r) = 4\epsilon\left[\left(\frac{\sigma_{\alpha\beta}}{r}\right)^{12} - \left(\frac{\sigma_{\alpha\beta}}{r}\right)^{6}\right] + V^{\rm cut}_{\alpha\beta}(r)$, where α,β denote the particle species A or B, $\epsilon = 1$, $\sigma_{AA} = 1.4$, $\sigma_{AB} = 1.2$, $\sigma_{BB} = 1.0$, and $V^{\rm cut}_{\alpha\beta}(r) = c_0 + c_2\left(r/\sigma_{\alpha\beta}\right)^2 + +c_4\left(r/\sigma_{\alpha\beta}\right)^4 + c_6\left(r/\sigma_{\alpha\beta}\right)^6$. The potential is truncated and shifted at $\varsigma_{\alpha\beta} = 2^{1/6}\sigma_{\alpha\beta}$ and the parameters c_0, c_2, c_4, and c_6 are chosen such that the potential and its first three derivatives are continuous at the cutoff. The resulting inter-particle force $\mathbf{F}_i = -\sum_{j\neq i}\partial_{\mathbf{r}_i} V_{\alpha\beta}(r_{ij})$ is purely repulsive. All the quantities presented in this work except for the velocity correlations, Eq. (3), pertain to all, *i.e.* large and small, particles. The velocity correlations were calculated for the large particles only; the correlations for the small particles are qualitatively the same.

Our control parameters were the active temperature T_a, the packing fraction $\phi = \pi N[\varsigma^3_{AA} + \varsigma^3_{BB}]/(12V)$ and the persistence time τ_p. We performed simulations along two lines in this three-dimensional space, specified by $[T_a = 1.0,\ \phi = 0.64]$ and $[T_a = 0.01,\ \phi = 0.58]$. As a shortcut, we refer to these two lines as two "state points", in spite of the fact that the full specification of the state point requires also τ_p. When the persistence time goes to zero at a fixed active temperature our system becomes equivalent to a Brownian system at temperature $T = T_a$. We chose the two state points in such a way that we could observe qualitatively different changes of the single-particle dynamics with increasing persistence time [20].

Equal time velocity correlations. – Intuitively, by increasing persistence time we displace an AOUP system farther from equilibrium. To give some quantitative support to this statement we investigated equal time correlations of the velocities of the active particles. These correlations are trivial for equilibrium thermal systems. We note that non-trivial equal time velocity correlations were observed in an experimental study of active cellular motion [21] and in a simulational investigation of the dense phase in an active system undergoing mobility-induced phase separation [11].

In our earlier investigations of glassy dynamics in interacting AOUP systems [20, 22, 23] we found that the equal time velocity correlation function defined below determines the short-time dynamics of the active particles and also appears in an approximate mode-coupling-like theory of the long time dynamics,

$$\omega_{\parallel}(q) = \frac{1}{N\xi_0^2}\left\langle\left|\sum_i \hat{\mathbf{q}}\cdot\left(\mathbf{f}_i + \mathbf{F}_i\right)e^{-i\mathbf{q}\cdot\mathbf{r}_i}\right|^2\right\rangle. \qquad (3)$$

Here $\hat{\mathbf{q}} = \mathbf{q}/|\mathbf{q}|$ and $\xi_0^{-1}\left(\mathbf{f}_i + \mathbf{F}_i\right)$ is the instantaneous velocity of particle i, see Eq. (1).

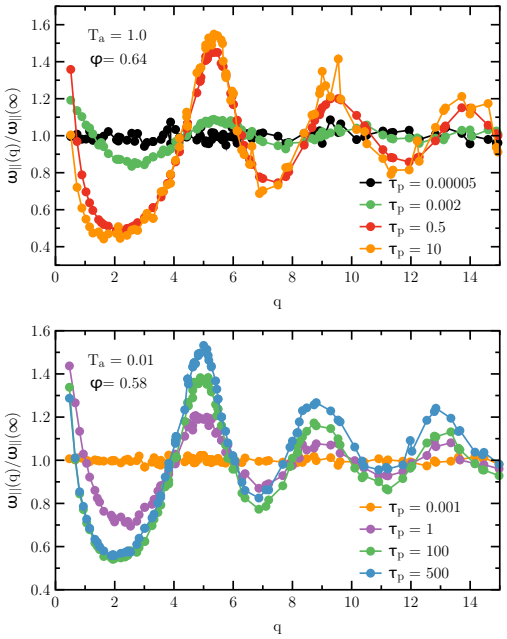

FIG. 1: The wavevector dependence of the equal-time nonequilibrium velocity correlation function $\omega_{\parallel}(q)$ normalized by its large wavevector limit, $\omega_{\parallel}(\infty)$, for (a) $T_a = 1.0$ and $\phi = 0.64$ and (b) $T_a = 0.01$ and $\phi = 0.76$, and different self-propulsion persistence times. Non-trivial character of the velocity correlations monotonically increases with increasing persistence time.

In Fig. 1 we show that, while velocity correlations as characterized by $\omega_{\parallel}(q)$ become trivial (featureless) in the limit of vanishing persistence time of the self-propulsions, they monotonically increase with the persistence time.

Effective temperature based on the Einstein relation. – The validity/violation of fluctuation-dissipation relations is a sensitive signature for a system to be in/out of equilibrium [24]. To further verify that with increasing persistence time a system of interacting AOPUs is progressively displaced away from equilibrium we test the validity of the simplest fluctuation-dissipation relation between the self-diffusion and mobility coefficients and we compare the effective temperature defined as the ratio of these coefficients to the active temperature.

In Fig. 2 we show the persistence time dependence of the mean-square displacement, $\langle\delta r^2(t)\rangle = N^{-1}\left\langle\sum_i\left(\mathbf{r}_i(t) - \mathbf{r}_i(0)\right)^2\right\rangle$, and of the self-diffusion co-

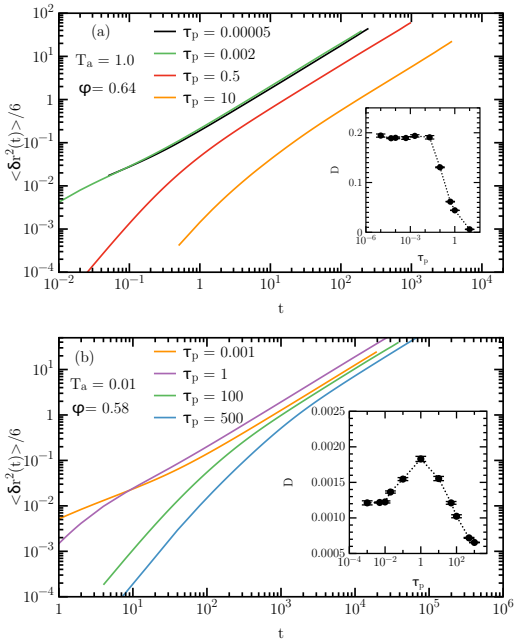

FIG. 2: The persistence time dependence of the mean-square displacement $\langle \delta r^2(t) \rangle$ for (a) $T_a = 1.0$ and $\phi = 0.64$ and (b) $T_a = 0.01$ and $\phi = 0.76$. The insets show the persistence time dependence of the self-diffusion coefficient at each state point. With increasing persistence time, the steady-state dynamics of the system studied at $T_a = 1.0$ monotonically slow down. However, with increasing persistence time the system studied at $T_a = 0.01$ the steady-state dynamics initially speed up and then begin to slow down for the largest persistence times studied.

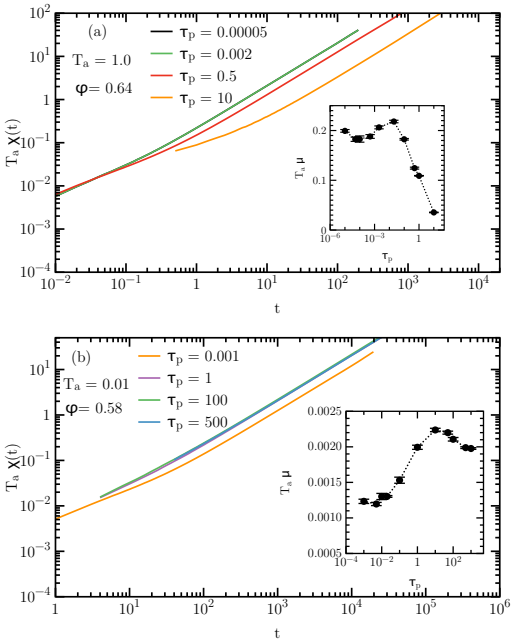

FIG. 3: The persistence time dependence of the time-dependent mobility function, $\chi(t)$, for (a) $T_a = 1.0$ and $\phi = 0.64$ and (b) $T_a = 0.01$ and $\phi = 0.76$. The insets show the persistence time dependence of the mobility coefficient at each state point. With increasing persistence time, the mobility of the system studied at $T_a = 1.0$ monotonically slows down. In contrast, with increasing persistence time the system studied at $T_a = 0.01$ the mobility initially speeds up and then begins to slow down for the largest persistence times studied.

efficient, $D = \lim_{t\to\infty} \langle \delta r^2(t) \rangle / (6t)$. We observe that at both state points D is initially almost persistence time-independent (there is some slight non-monotonic dependence of D on τ_p, analogous to that reported earlier [22]). Then, D starts to decrease rapidly with τ_p for the system at $T_a = 1.0$. In contrast, for the system at $T_a = 0.01$ D starts to increase with increasing τ_p. This behavior was previously observed in a similar system [20]. However, with increasing persistence time further than in this earlier study, the self-diffusion coefficient of the system at $T_a = 0.01$ goes through a maximum and starts decreasing.

We define the time-dependent mobility function as follows: at $t = 0$ a weak constant force $\lambda \mathbf{e}$ is applied to one (tagged, t) particle. Here λ measures the magnitude of the force and $\mathbf{e}$ is a unit vector. Under the influence of this force the average position of the tagged particle will change systematically,

$$\langle \mathbf{r}_t(t) - \mathbf{r}_t(0) \rangle = \lambda \chi(t) \mathbf{e} + o(\lambda), \tag{4}$$

where $\chi(t)$ is the time dependent mobility. We define the mobility coefficient through the long-time limit of the time-dependent mobility function, $\mu = \lim_{t\to\infty} \chi(t)/t$. We note that for an equilibrium system the Einstein relation holds and $\langle \delta r^2(t) \rangle /6 = T\chi(t)$, and $D = T\mu$, where T is the system's temperature.

To calculate the time-dependent mobility of our active matter system we used the procedure proposed earlier [25], which allows one to evaluate a linear response function of an AOUP system using trajectories generated without any external force. We note that, in general, longer trajectories are needed to accurately evaluate mobility function $\chi(t)$ than mean-square displacement $\langle \delta r^2(t) \rangle$.

In Fig. 3 we show the persistence time dependence of the time-dependent mobility function and of the mobility coefficient at both state points. Comparing the main panels of Figs. 2 and 3 we see that the time-dependence of $\langle \delta r^2(t) \rangle$ and $\chi(t)$ at short times is qualitatively different. At long times, however, both functions grow linearly with time. Comparing the insets in Figs. 2 and 3 we see that, although the persistence time dependence of D and μ is qualitatively similar, there are significant quantitative differences.

We recall that in the limit of the vanishing persistence time our system becomes equivalent to a Brownian system at temperature $T = T_a$. Thus, we expect that in the $\tau_p \to 0$ limit the ratio D/μ should be approaching T_a. For longer persistence times we *define* an effective

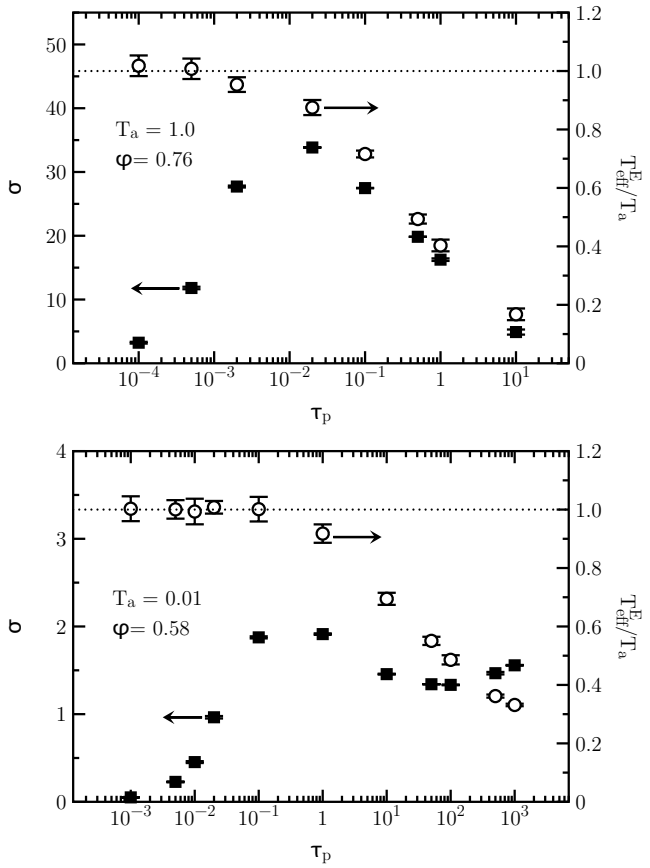

FIG. 4: The persistence time dependence of the ratio of the effective and active temperatures, $T^{\rm E}_{\rm eff}/T_a$ (open circles) and of the entropy production σ as defined by Fodor *et al.* [7] (filled squares). The ratio $T^{\rm E}_{\rm eff}/T_a$ monotonically decreases with increasing persistence time whereas σ exhibits a non-monotonic dependence on the persistence time.

temperature based on the Einstein relation as

$$T^{\rm E}_{\rm eff} = D/\mu. \tag{5}$$

The difference between the active temperature T_a and the effective temperature $T^{\rm E}_{\rm eff}$ based on the Einstein relation quantifies the departure of our active matter system from equilibrium [26].

In Fig. 4 we show the persistence time dependence of the ratio of the effective temperature $T^{\rm E}_{\rm eff}$ based on the Einstein relation and the active temperature T_a. We observe that for sufficiently short persistence times $T^{\rm E}_{\rm eff}/T_a$ is constant and equal to 1 (within error bars). With increasing persistence time this ratio starts decreasing monotonically [27]. This behavior agrees with physical expectations and a small τ_p expansion [7]. For short persistence times the system is in an effective equilibrium state that can be described in terms of the active temperature, but with increasing persistence time the system is progressively displaced away from equilibrium [29].

We note that the dependence of the ratio $T^{\rm E}_{\rm eff}/T_a$ on the persistence time is the opposite from the dependence on the shear rate of the ratio of the same effective temperature and the temperature T for a sheared Brownian system. In the latter case, the ratio $T^{\rm E}_{\rm eff}/T$ increases monotonically [30, 31] with increasing shear rate, *i.e.* with increasing departure from equilibrium.

Entropy production rate. – Fodor *et al.* [7] started from a definition of the entropy production in terms of a ratio of the probabilities of a position space trajectory and its time-reversed version and derived the following expression for the entropy production rate,

$$\sigma = \frac{\tau_p^2}{2T_a}\left\langle \left(\sum_i \dot{\mathbf{r}}_i \cdot \partial_{\mathbf{r}_i}\right)^3 \sum_{k>l} V(r_{kl}) \right\rangle. \tag{6}$$

We note that this expression involves third derivatives of the potential energy, and in order to avoid any problems associated with singular contributions we used an interparticle potential that has continuous first three derivatives.

We evaluated the persistence time dependence of the entropy production rate given by expression (6) for both state points. The results shown in Fig. 4 do not follow our physical expectation. The quantity given by expression (6) has a non-monotonic dependence on the persistence time, and, moreover, this dependence is qualitatively different for the two active temperatures T_a.

For both T_a, at small persistence times σ increases with increasing persistence time, as predicted in Ref. [7]. At $T_a = 1.0$, at the persistence time at which the ratio $T^{\rm E}_{\rm eff}/T_a$ is about 0.8, σ exhibits a maximum and then starts to decrease for longer persistence times. At $T_a = 0.01$, we observe initially a very similar persistence time dependence, with the maximum occurring for the persistence time at which the ratio $T^{\rm E}_{\rm eff}/T_a$ is about 0.9. However, in this case for longer persistence times σ exhibits a local minimum and then it starts to increase again. We note that the range of persistence times investigated at $T_a = 1.0$ is smaller than that investigated at $T_a = 0.01$. The reason is that at $T_a = 1.0$ at persistence times longer than $\tau_p = 10$ very slow dynamics prevented us from reaching a stationary state.

We note that the relation between the location of the maximum of the entropy production as a function of the persistence time and the value of the ratio $T^{\rm E}_{\rm eff}/T_a$ is not necessarily universal and deserves further study.

Discussion. – Our physical expectation is that increasing persistence time displaces an AOUP system progressively away from equilibrium. This is supported by the monotonically increasing non-trivial equal-time correlations between AOUPs velocities and the monotonically decreasing ratio of the effective temperature based on the Einstein relation and the active temperature. Surprisingly, the dependence of the entropy production rate calculated according to Fodor *et al.* on the persistence time does not agree with this expectation. This suggests

that expression (6) is not a good quantitative measure of the departure from equilibrium.

One may observe that the entropy production rate should be normalized by a quantity characterizing system's dynamics, *i.e.* that instead of σ one should examine $\sigma\tau_{\rm rel}$, where $\tau_{\rm rel}$ is a characteristic relaxation time of the system. $\sigma\tau_{\rm rel}$ quantifies the entropy produced while the system decorrelates from its given configuration. We used the characteristic diffusion time σ_{BB}^2/D, where σ_{BB} is the B particle size, as a measure of the relaxation time and we examined $\sigma\sigma_{\rm BB}^2/D$ as a function of the persistence time. We found that $\sigma\sigma_{\rm BB}^2/D$ monotonically increases with τ_p at $T_a = 1.0$ but has a non-monotonic dependence on τ_p at $T_a = 0.01$.

We recall that expression (6) also predicts vanishing entropy production for a single freely moving AOUP and for a single AOUP in a harmonic potential. Both findings are often claimed to be counter-intuitive. Some of the alternative approaches to the entropy production [15–18] find non-vanishing entropy production for a freely moving AOUP and/or for an AOUP in a harmonic potential. It would be interesting to check what do these approaches predict for systems of interacting AOUPs.

Finally, we note that approaches of Refs. [16–18] either implicitly or explicitly consider trajectories in the space of positions and self-propulsions. Thus, at least in spirit, they are similar to the approach of Pietzonka and Seifert [19] who argue that, in order to properly evaluate the entropy production, one has to consider the physicochemical processes giving rise to the self-propulsion. It would be very interesting to investigate whether the minimal approach adopted in Refs. [16–18] is sufficient to define and evaluate the entropy production or whether one has to follow the full treatment of Ref. [19].

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[26] We note that in the small density (single-particle) limit $T_{\rm eff}^{\rm E} = T_a$ for all persistence times. Thus, the ratio $T_{\rm eff}^{\rm E}/T_a$ is not a good indicator of the departure from equilibrium of a free AOUP.

[27] We note that an earlier study [28] reported that the effective temperature based on the Einstein relation is increasing with increasing persistence time of the persistent Monte Carlo dynamics. However, to compare with our result one would have to investigate for the model considered in Ref. [28] the persistence time dependence of the ratio of the effective temperature at finite density to that in the small density (single-particle) limit. Fig. 1c of Ref. [28] suggests that this ratio decreases with incresing persistence time.

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