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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 particle (AOUP) 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 nonmonotonic dependence on the persistence time. Thus, it does not properly quantify the departure of AOUP systems from equilibrium.

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I. 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 nonequilibrium systems is not well developed, one oft-used approach to study systems of self-propelled particles is to approximate them by appropriately chosen thermal equilibrium systems [6,8,9]. Intuitively, whether such an approach is sensible depends on the degree of nonequilibrium in the active systems[10]. Thus, a recurring question in the recent literature is how to quantify the departure of the systems of self-propelled particles from thermal equilibrium. To paraphrase a recent article [12], we want to replace a binary in-out of equilibrium classification with 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 Nardini *et al.* [13] and by Puglisi, Marconi, Maggi, and collaborators [14,15]. 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.* [16] 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.* [17] 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 [18] 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 [19]. Additionally, Pietzonka and Seifert [20] argued that the most fundamental consideration of entropy production should also include the contribution from physicochemical processes that give rise to the self-propulsion.

Here we present the 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 nonmonotonic dependence on the self-propulsion persistence time. Thus, it is not a good measure of the departure of AOUP systems from thermal equilibrium. Numerical evaluation of the alternative proposals to define the entropy production [16–19] is left for a future study.

II. 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 friction 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, $\langle \boldsymbol{\eta}_i(t)\boldsymbol{\eta}_j(t')\rangle_{\text{noise}} = 2\xi_0 k_B T_a \mathbf{I}\delta_{ij}\delta(t-t')$, where $\langle \cdots \rangle_{\text{noise}}$ 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 in $d = 3$, interacting via the smoothed Weeks-Chandler-Andersen truncation of the Lennard-Jones potential, $V_{\alpha\beta}(r) = 4\epsilon[(\frac{\sigma_{\alpha\beta}}{r})^{12} - (\frac{\sigma_{\alpha\beta}}{r})^{6}] + V_{\alpha\beta}^{\text{cut}}(r)$, 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 $V_{\alpha\beta}^{\text{cut}}(r) = c_0 + c_2(r/\sigma_{\alpha\beta})^2 + +c_4(r/\sigma_{\alpha\beta})^4 + c_6(r/\sigma_{\alpha\beta})^6$. 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 interparticle 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 $\phi = \pi N[\varsigma_{AA}^3 + \varsigma_{BB}^3]/(12L^3)$, and the persistence time τ_p, where L is the simulation box length. 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 [21].

III. EQUAL-TIME VELOCITY CORRELATIONS

Intuitively, by increasing persistence time we displace an AOUP system farther from thermal 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 nontrivial equal-time velocity correlations were observed in an experimental study of active cellular motion [22] and in a simulational investigation of the dense phase in an active system undergoing mobility-induced phase separation [11].

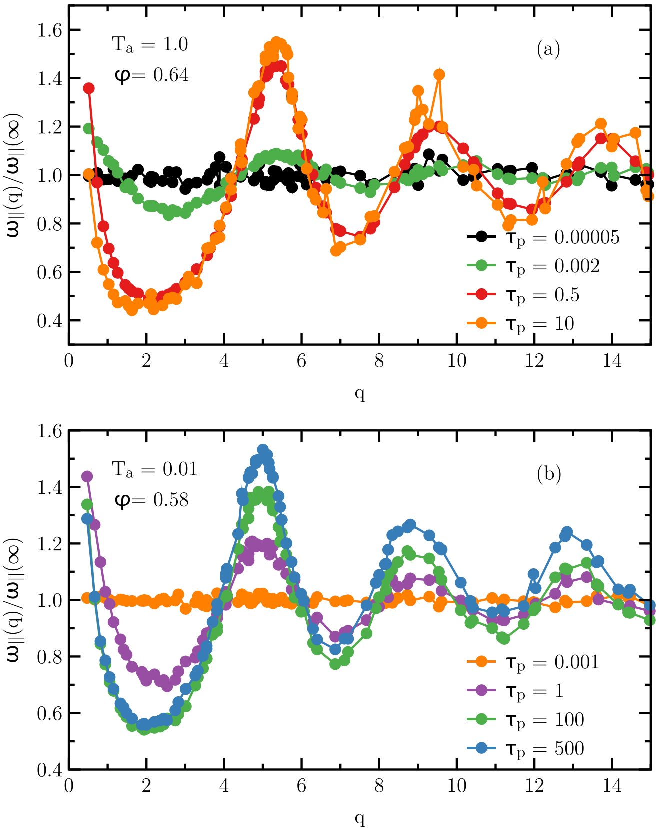

FIG. 1. The wave-vector dependence of the equal-time nonequilibrium velocity correlation function $\omega_{\parallel}(q)$ normalized by its large wave-vector limit, $\omega_{\parallel}(\infty)$, for (a) $T_a = 1.0$ and $\phi = 0.64$ and (b) $T_a = 0.01$ and $\phi = 0.76$, and different self-propulsion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

In our earlier investigations of glassy dynamics in interacting AOUP systems [21,23,24], 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,

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

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

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 increase monotonically with the persistence time. We note that we have already reported on this increase before [21]. The results shown in Fig. 1 pertain to the present system with interaction potential vanishing smoothly at the cutoff, and they have been obtained for a significantly larger range of persistence times.

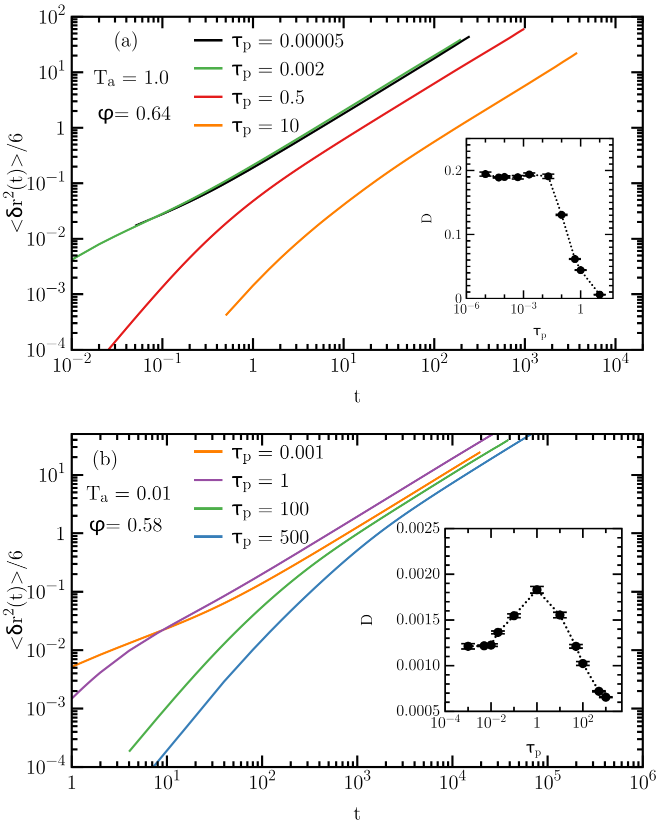

FIG. 2. The persistence time dependence of the mean-square displacement $\langle \delta r^2(t) \rangle$ for (a) $T_a = 1.0$ and $\phi = 0.64$ and (b) $T_a = 0.01$ and $\phi = 0.76$. The insets show the persistence time dependence of the self-diffusion coefficient at each state point. With increasing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

IV. 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 [25]. To further verify that a system of interacting AOUPs is progressively displaced away from equilibrium with increasing persistence time, we test the validity of the simplest fluctuation-dissipation relation between the self-diffusion and mobility coefficients. 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, $\langle \delta r^2(t) \rangle = N^{-1} \langle \sum_i [\mathbf{r}_i(t) - \mathbf{r}_i(0)]^2 \rangle$, and of the self-diffusion coefficient, $D = \lim_{t\to\infty} \langle \delta r^2(t) \rangle / (6t)$. At both state points, we observe that D is initially almost persistence time-independent (there is some slight nonmonotonic dependence of D on τ_p, analogous to that reported earlier [23]). 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 [21]. 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 in Ref. [26]: 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 in Ref. [26], 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 the mobility function $\chi(t)$ than the 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 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 temperature based on the Einstein relation as

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

The difference between the active temperature T_a and the effective temperature $T_{\rm eff}^{\rm E}$ based on the Einstein relation quantifies the departure of our active matter system from thermal equilibrium. We note, however, 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.

In Fig. 4 we show the persistence time dependence of the ratio of the effective temperature $T_{\rm eff}^{\rm E}$ based on the Einstein relation and the active temperature T_a. We observe that for sufficiently short persistence times $T_{\rm eff}^{\rm E}/T_a$ 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].

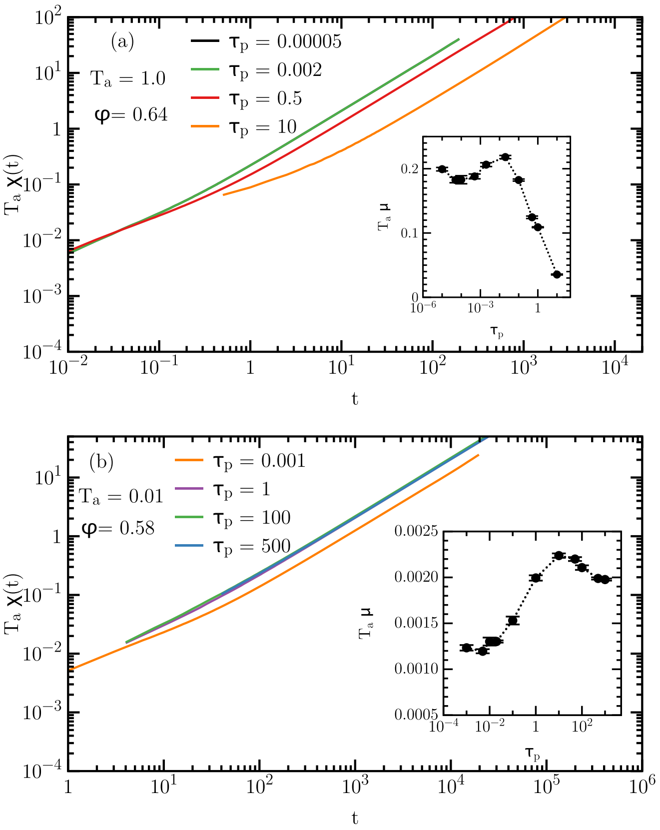

FIG. 3. The persistence time dependence of the time-dependent mobility function, $\chi(t)$, for (a) $T_a = 1.0$ and $\phi = 0.64$ and (b) $T_a = 0.01$ and $\phi = 0.76$. The insets show the persistence time dependence of the mobility coefficient at each state point. With increasing persistence time,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

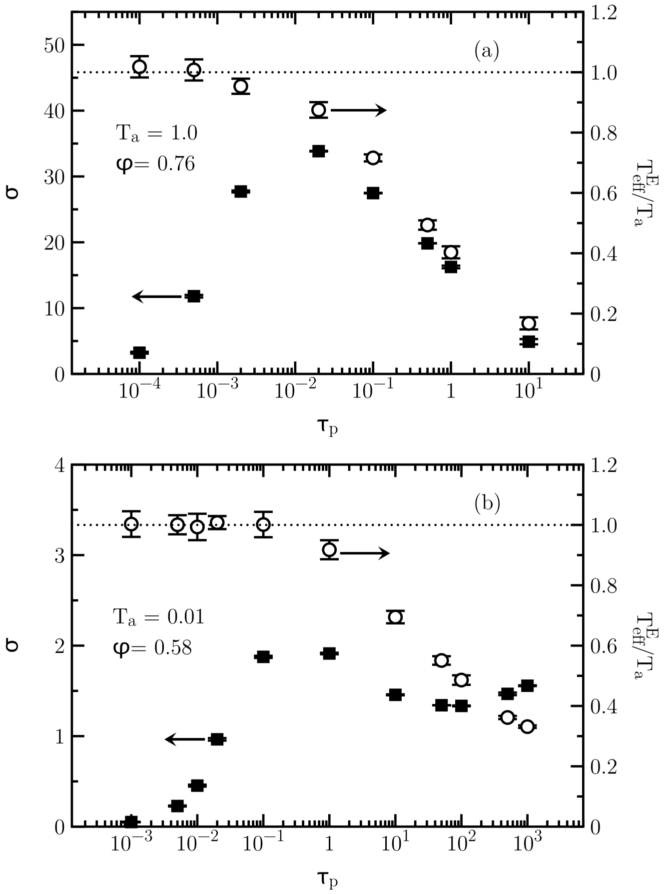

FIG. 4. The persistence time dependence of the ratio of the effective and active temperatures, $T_{\rm eff}^{\rm E}/T_a$ (open circles), and of the entropy production σ as defined by Fodor *et al.* [7] (filled squares). The ratio $T_{\rm eff}^{\rm E}/T_a$ decreases monotonically with increasing persistence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

The results shown in Fig. 4 significantly extend the ones reported in Ref. [26]: Here we show that the monotonic dependence of $T_{\rm eff}^{\rm E}/T_a$ on the persistence time is observed even though the self-diffusion and mobility coefficients exhibit a nonmonotonic dependence on τ_p. In contrast, in Ref. [26] we only examined a limited range of persistence times for which the self-diffusion and mobility coefficients were varying monotonically with τ_p (for a slightly different system from the one investigated there).

We note that the dependence of the ratio $T_{\rm eff}^{\rm E}/T_a$ on the persistence time is opposite 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 $T_{\rm eff}^{\rm E}/T$ increases monotonically [30,31] with increasing shear rate, i.e., with increasing departure from equilibrium.

V. 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 they 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 whose first three derivatives are continuous.

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 nonmonotonic 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 $T_{\rm eff}^{\rm E}/T_a$ 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 $T_{\rm eff}^{\rm E}/T_a$ is

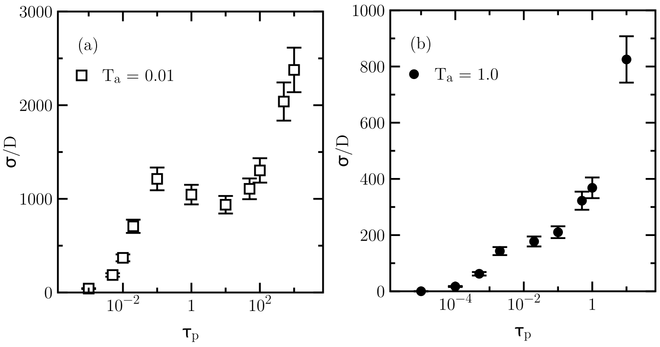

FIG. 5. The persistence time dependence of the entropy production rate normalized by the self-diffusion coefficient, σ/D.

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 $T_{\text{eff}}^{\text{E}}/T_a$ is not necessarily universal and deserves further study.

Finally, one may observe that the physically important quantity is the amount of entropy produced during the time it takes the system to forget its given state. In other words, the entropy production rate should perhaps be normalized by a characteristic relaxation time of the system τ_{rel}, i.e., instead of σ one should examine $\sigma\tau_{\text{rel}}$. 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 $\sigma\sigma_{BB}^2/D$ as a function of the persistence time. In Fig. 5 we show that σ/D (recall that $\sigma_{BB} = 1$) exhibits a nonmonotonic dependence on τ_p at $T_a = 0.01$ but increases monotonically with τ_p at $T_a = 1.0$.

VI. DISCUSSION

Our physical expectation is that increasing persistence time displaces an AOUP system progressively away from equilibrium. This is supported by the monotonically increasing nontrivial equal-time correlations between AOUP 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 thermal equilibrium.

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 counterintuitive. Some of the alternative approaches to the entropy production [16–19] find nonvanishing entropy production for a freely moving AOUP and/or for an AOUP in a harmonic potential. It would be interesting to check what these approaches predict for systems of interacting AOUPs.

We note that approaches of Refs. [17–19] 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 [20], 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. [17–19] is sufficient to define and evaluate the entropy production or whether one has to follow the full treatment of Ref. [20].

Finally, we admit that one can adopt an alternative view of the results presented here by acknowledging that the expression derived by Fodor *et al.* really measures the breakdown of the time-reversal symmetry. Then, our results imply that increasing departure from thermal equilibrium, quantified by the nontrivial character of the equal-time velocity correlations or of the effective temperature, is not necessarily monotonically related to the degree of the breakdown of the time-reversal symmetry. A deeper investigation of this disconnect is left for future work.

ACKNOWLEDGMENTS

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