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Nonequilibrium statistical mechanics of mixtures of particles in contact with different thermostats

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We introduce a novel type of locally driven systems made of two types of particles (or a polymer with two types of monomers) subject to a chaotic drive with approximately white noise spectrum, but different intensity; in other words, particles of different types are in contact with thermostats at different temperatures. We present complete systematic statistical mechanics treatment starting from first principles. Although we consider only corrections to the dilute limit due to pairwise collisions between particles, meaning we study a nonequilibrium analog of the second virial approximation, we find that the system exhibits a surprisingly rich behavior. In particular, pair correlation function of particles has an unusual quasi-Boltzmann structure governed by an effective temperature distinct from that of any of the two thermostats. We also show that at sufficiently strong drive the uniformly mixed system becomes unstable with respect to steady states consisting of phases enriched with different types of particles. In the second virial approximation, we define nonequilibrium "chemical potentials" whose gradients govern diffusion fluxes and a nonequilibrium "osmotic pressure," which governs the mechanical stability of the interface.

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I. INTRODUCTION

Paraphrasing Tolstoy [1], all equilibrium systems are alike, each nonequilibrium system is out of equilibrium in its own way. Of particular current interest is a variety of nonequilibrium systems which are driven locally, such that energy transduction occurs at the scale of the individual components. Examples include bacterial swimmers [2,3], their artificial models [4,5], molecular motors [6,7], and other enzymatic systems performing local mechanical work [8] at the expense of the free energy excess of supplied reagents (e.g., ATP hydolysis). An instructive comparison can be done with three-dimensional turbulence. In turbulence, the system is driven globally, on the largest scale, and then energy cascades down and dissipates locally. By contrast, active systems are driven locally, and the question is how the disorganized local drive can produce observable macroscopic consequences on the largest scale, such as spontaneous flows [9] or phase segregation [10–17]. The interest in such questions is motivated by biological applications, but these studies shed also light on fundamental statistical mechanics.

A useful classification of locally driven systems may be based on symmetry, as an active drive may have a scalar or vectorial character [18]. Bacterial swimmers, for instance, have vectorial symmetry, as their activity takes the form of force generation with a certain direction, even though the direction randomizes after some reorientation time (by either separate tumbling events [run-and-tumble particles (RTPs)] or rotational diffusion [active Brownian particles (ABPs)]. The classification as an "active system" is generally reserved for vectorial activity. Purely scalar activity may be exemplified by local chemical transformations, such as chromatin methylation or acetylation, which amounts to local change of solvent quality for the chromatin fiber viewed as a polymer.

Theoretical efforts in the field are mostly about vectorial active particles at high concentrations, where all effects are most pronounced, and the works are dominated by simulations, including comprehensive ones [11,12,15,17,19]. Building on numerical results, successful phenomenological theories were developed, notably Ref. [20]. However, the field suffers from a lack of analytically solvable microscopic models and approaches enabling to develop physical insight into how disorganized, incoherent local drive generates coherent macroscopic behavior. The goal of the present paper is to offer a class of locally driven systems allowing for such a systematic treatment.

Specifically, we consider a suspension of two types of particles, $\mathcal{A}$ and $\mathcal{B}$, exposed to different heat baths with temperatures $T_{\mathcal{A}} \geqslant T_{\mathcal{B}}$. Our main technical idea is to look at systems of sufficiently low concentration, where we build up the nonequilibrium analog of a second virial approximation, considering only pair collisions between particles. This system, even relatively dilute, is not only far from equilibrium, but also *locally driven* in the sense that energy transduction occurs at a local scale, with energy flowing from the hotter to the colder reservoir via the local interactions between particles. Although this system is rather simple, and does allow for systematic theoretical treatment, our main result is that the properties of this system are highly nontrivial. In particular, we show that some statistical features of this nonequilibrium system, such as pair correlation functions, are described by an unusual quasi-Boltzmann distribution, governed by some effective temperature not imposed by any thermostat, which is a linear combination of $T_{\mathcal{A}}$ and $T_{\mathcal{B}}$. Furthermore, this leads directly to the phenomenon of activity-induced phase segregation, in this purely scalar system having no directional force generation or quorum sensing feedback mechanism. Since all these results arise from a simple model and a completely systematic treatment, which does not employ any *a priori* analogies with the equilibrium case, our model could be an instructive playground for physical intuition.

Our treatment (with slight modification) may also apply to a polymer chain, a copolymer with $\mathcal{A}$- and $\mathcal{B}$-type monomers connected in a certain sequence. Such polymers were considered in Ref. [21], with the idea to model spatial segregation between eu- and hetero-chromatin, i.e., between actively processed and almost silent parts of the genome in a eukaryotic cell nucleus. It is argued in this paper that genes which are being expressed and, therefore, subject to RNA polymerization and other active processes, should be viewed as active monomers, and silent genes as passive monomers. The observed compartmentalization between the two kinds of chromatin is then understood as an actively driven microphase segregation [22]. Since active processes in this case are of scalar character, if we neglect time-dependent correlations between them, i.e., approximate them as a white noise, we can view $\mathcal{A}$ and $\mathcal{B}$ monomers as exposed to two different thermostats (which is what was done in the simulation of Ref. [21]). While our work was already completed, we became aware of Ref. [23] where the same model at high concentration was examined by molecular dynamics simulations, and the activity-driven phase segregation was indeed observed.

There are many examples of systems whose description involves two distinct temperatures, ranging from plasmas (see basics in, e.g., Ref. [24]), to spin glasses [25] and heteropolymers [26]; other examples are given in Refs. [27–30]. In all these works (with the notable exception of Ref. [30]), the two temperatures are used to describe motions on vastly different time scales. In our system, there is no such separation of time scales, and our goal is to explore how the access to two different heat baths enhances the tendency toward phase segregation.

Our starting point is the overdamped Langevin equation

$$\zeta_i \dot{x}_i = -\partial_i U + (2T_i\zeta_i)^{1/2}\xi_i(t), \tag{1}$$

for every particle i in the system. Here x_i indicates the position of particle i (for brevity, we make no distinction between particles in one or three dimensions). We assume that all forces acting on a particle derive from a potential energy U, while ∂_i is the derivative with respect to x_i. The friction coefficient for particle i is ζ_i, and $\xi_i(t)$ is a standard zero mean and unit variance Gaussian white noise, independent for all particles i. Finally, T_i is the temperature (in energy units) of the heat bath interacting with particle i; it is either $T_{\mathcal{A}}$ or $T_{\mathcal{B}}$. A major assumption of our model is that we ignore hydrodynamic interactions (Rouse model), each particle experiencing a local friction against an immobile solvent.

II. TWO PARTICLES

We first examine a system with only two particles, one $\mathcal{A}$ and one $\mathcal{B}$. Equation (1) consists then of two coupled Langevin equations. Dotsenko *et al.* [30] studied this problem when the potential energy $U(x_{\mathcal{A}},x_{\mathcal{B}})$ is a positive-definite quadratic form of its two variables. We only assume here that U depends on the distance between particles, $r = x_{\mathcal{A}} - x_{\mathcal{B}}$, i.e., $U = u^{\mathcal{AB}}(r)$, and consider mostly cases where $u^{\mathcal{AB}}(r)$ vanishes at large r. We derive a Langevin equation for the variable r, by combining the two equations (1):

$$\zeta_r \dot{r} = F(r) + (2\zeta_r\overline{T})^{1/2}\xi_r, \tag{2}$$

where the relevant friction is $\zeta_r = \zeta_{\mathcal{A}}\zeta_{\mathcal{B}}/(\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}})$ and the relevant temperature is the mobility-weighted average $\overline{T} = (\zeta_{\mathcal{B}}T_{\mathcal{A}} + \zeta_{\mathcal{A}}T_{\mathcal{B}})/(\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}})$. The definition of the effective temperature $\overline{T}$ is dictated by the condition that the noise $\xi_r(t)$ is a zero mean, unit variance Gaussian white noise. It is always between $T_{\mathcal{A}}$ and $T_{\mathcal{B}}$. The Langevin equation (2) imposes that the relative distance between particles, r, in a steady state is Boltzmann distributed with temperature $\overline{T}$, despite the fact that the system is out of equilibrium:

$$P(r) = \exp[-U(r)/\overline{T}]z^{-1}; \tag{3}$$

z is here the "partition sum" ensuring normalization. Equation (3) is the central technical result of this work. See also Appendix A regarding the enhancement of the joint diffusion of two particles when $T_{\mathcal{A}} \neq T_{\mathcal{B}}$.

III. FOKKER-PLANCK EQUATION, CURRENTS, AND VIOLATION OF DETAILED BALANCE

The Langevin equations (1) can be recast as a Fokker-Planck equation for the joint probability distribution of the coordinates of all particles $P(\{x\})$ and the corresponding currents J_i:

$$\dot{P} = -\sum_i \partial_i J_i, \quad J_i = -\partial_i U\ P/\zeta_i - T_i\partial_i P/\zeta_i. \tag{4}$$

At steady state for two particles, the probability $P(r)$ depends only on the distance $r = x_{\mathcal{A}} - x_{\mathcal{B}}$. Then $\partial_{\mathcal{A}}P = -\partial_{\mathcal{B}}P$, as well as $\partial_{\mathcal{A}}U = -\partial_{\mathcal{B}}U$. Furthermore, under these conditions, there is no current in the direction of $r = x_{\mathcal{A}} - x_{\mathcal{B}}$, which means $J_{\mathcal{A}} - J_{\mathcal{B}} = 0$, or

$$(-\partial_{\mathcal{A}}U/\zeta_{\mathcal{A}} + \partial_{\mathcal{B}}U/\zeta_{\mathcal{B}}) - (T_{\mathcal{A}}\partial_{\mathcal{A}}P/\zeta_{\mathcal{A}} - T_{\mathcal{B}}\partial_{\mathcal{B}}P/\zeta_{\mathcal{B}}) = 0.$$

This leads back to the Boltzmann distribution with the average temperature (3). Now compute the current $J_{\mathcal{A}}$:

$$J_{\mathcal{A}} = J_{\mathcal{B}} = \frac{T_{\mathcal{A}} - T_{\mathcal{B}}}{T_{\mathcal{A}}\zeta_{\mathcal{B}} + T_{\mathcal{B}}\zeta_{\mathcal{A}}} P(r)\partial_{\mathcal{A}}U. \tag{5}$$

As expected, it vanishes for an equilibrium system ($T_{\mathcal{A}} = T_{\mathcal{B}}$), where detailed balance is obeyed. However, if $T_{\mathcal{A}} \neq T_{\mathcal{B}}$, detailed balance is violated. For instance, if the system is one-dimensional, its configuration space $(x_{\mathcal{A}},x_{\mathcal{B}})$ is a two-dimensional plane, and there are current loops in this plane as shown in Fig. 1. Physically, Eq. (5) suggests that these current loops are such that the passive particle moves, on average, in the direction of the force acting on them, while the active particle moves in the direction opposite to the force acting on them.

IV. POWER TRANSFER

As pointed out above, energy is transferred everywhere from the "hot heat bath" to the "cold" bath, or from the energy source of active motion to the surrounding passive medium. The transferred power from $\mathcal{A}$ to $\mathcal{B}$ is the average $w = \langle -\dot{x}_{\mathcal{B}}\partial_{\mathcal{B}}U\rangle$. The velocity is $\dot{x}_{\mathcal{B}} = J_{\mathcal{B}}/P$, while the average involves an integration with a weight P. Therefore, $w = -\int J_{\mathcal{B}}\partial_{\mathcal{B}}U\, dx_{\mathcal{A}}\, dx_{\mathcal{B}}$, leading to

$$w_{\mathcal{A}\to\mathcal{B}} = \frac{T_{\mathcal{A}} - T_{\mathcal{B}}}{T_{\mathcal{A}}\zeta_{\mathcal{B}} + T_{\mathcal{B}}\zeta_{\mathcal{A}}} \int [\partial_r u^{\mathcal{AB}}(r)]^2 \frac{e^{-u^{\mathcal{AB}}(r)/\overline{T}}}{z}\, dr. \tag{6}$$

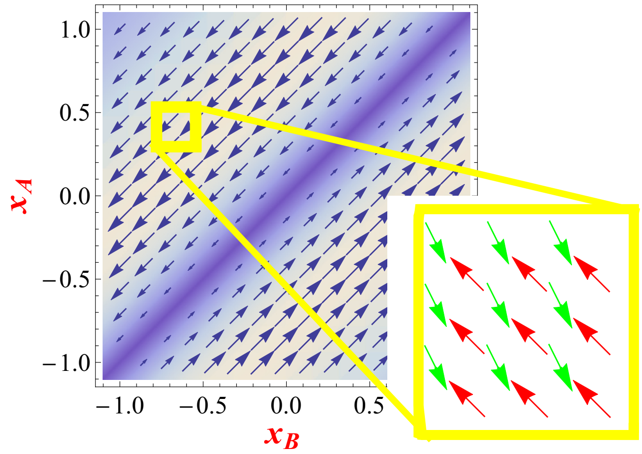

FIG. 1. (Color online) Field of currents. For illustration purposes, the interaction potential is chosen in the form $u(r) = 1/(r^2 + 1)$, and the temperatures are $T_{\mathcal{A}} = 1$ and $T_{\mathcal{B}} = 0.5$. In the main figure, the arrows are the current vectors with components $J_{\mathcal{A}} = (-1 + \frac{T_{\mathcal{A}}}{\overline{T}})e^{-u(r)/\overline{T}}\partial_r u/\zeta$ and $J_{\mathcal{B}} = (1 - \frac{T_{\mathcal{B}}}{\overline{T}})e^{-u(r)/\overline{T}}\partial_r u/\zeta$ [assuming for illustration $\zeta_{\mathcal{A}} = \zeta_{\mathcal{B}}$; see Eq. (4)]. In the enlarged window, the drift current (upward and to the left arrows, corresponding to the first terms in the expressions for $J_{\mathcal{A}}$ and $J_{\mathcal{B}}$) and the diffusion current (downward and to the right arrows, corresponding to the second terms in the expressions of the currents) are shown separately; unlike for equilibrium systems, they are not collinear, indicating a detailed balance violation. Their vector sum gives rise to the nonpotential current field.

As expected, this power transfer from $\mathcal{A}$ to $\mathcal{B}$ vanishes at equilibrium ($T_{\mathcal{B}} = T_{\mathcal{A}}$), but it is strictly positive if $T_{\mathcal{A}} > T_{\mathcal{B}}$.

To understand the meaning of the result (6), consider a three-dimensional system and a repulsive potential U with amplitude U_0 and spatial scale b. The integral in Eq. (6) is then estimated as $(U_0/b)^2 b^3/L^3$, with L^3 the box volume (which enters in z); assuming for simplicity $\zeta_{\mathcal{A}} = \zeta_{\mathcal{B}} = \zeta$, the result can be rearranged as

$$w \sim [b(\overline{T}/\zeta)/L^3]\big[(T_{\mathcal{A}} - T_{\mathcal{B}})U_0^2/\overline{T}^2\big]. \tag{7}$$

The first factor in the square brackets is the inverse Smoluchowski collision time between two particles, and the second factor gives an estimate of the energy transferred during one "collision."

V. MANY PARTICLES

Consider now a system of N_i particles i ($i,j = \mathcal{A},\mathcal{B}$). The Fokker-Planck equation (4) is generalized for any number of particles. Integrating the Fokker-Planck equation over all coordinates except for one, we derive a diffusion equation for the single-particle probability for every particle species:

$$\begin{aligned}\frac{\partial p_1^{\mathcal{A}}(\mathbf{r})}{\partial t} &= \frac{N_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\int \frac{\partial u^{\mathcal{A}\mathcal{A}}}{\partial \mathbf{r}} p_2^{\mathcal{A}\mathcal{A}}(\mathbf{r},\mathbf{r}')d\mathbf{r}'\right]\\ &\quad + \frac{N_{\mathcal{B}}}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}}\left[\int \frac{\partial u^{\mathcal{A}\mathcal{B}}}{\partial \mathbf{r}} p_2^{\mathcal{A}\mathcal{B}}(\mathbf{r},\mathbf{r}')d\mathbf{r}'\right] + \frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\nabla_{\mathbf{r}}^2 p_1^{\mathcal{A}}(\mathbf{r}).\end{aligned} \tag{8}$$

A similar equation is obtained for $p_1^{\mathcal{B}}(\mathbf{r})$. These equations for single-particle probabilities include the pair probabilities p_2. By integrating the multiparticle Fokker-Planck equation over all coordinates except for two, we derive equations for p_2, which include the three-body correlations p_3, and then a hierarchy of equations (see details about this hierarchy in Appendix B). However, if the density is small enough, we can ignore all terms involving p_3, thus obtaining a closed equation for p_2. Consistent with the two-particle system, we obtain in this approximation

$$p_2^{ij}(\mathbf{r},\mathbf{r}') = p_1^i(\mathbf{r})p_1^j(\mathbf{r}')\exp[-u^{ij}(\mathbf{r}-\mathbf{r}')/T_{ij}], \tag{9}$$

where $i,j = \mathcal{A},\mathcal{B}$. The effective temperatures entering these expressions are different for the three types of interactions: $T_{\mathcal{A}\mathcal{A}} = T_{\mathcal{A}}$, $T_{\mathcal{B}\mathcal{B}} = T_{\mathcal{B}}$, and $T_{\mathcal{A}\mathcal{B}} = \overline{T}$.

It is important to note that these distributions form only as a result of an averaging over many collisions happening in the system under steady state conditions (similar in this respect to an equilibrium system).

Inserting the approximation (9) into Eqs. (8) and introducing the concentrations $c^i(\mathbf{r}) = N_i p_1^i(\mathbf{r})$, we obtain closed equations for the concentrations:

$$\frac{\partial c^i(\mathbf{r})}{\partial t} = \frac{1}{\zeta_i}\frac{\partial}{\partial \mathbf{r}}\left(c^i\frac{\partial \mu_i}{\partial \mathbf{r}}\right). \tag{10}$$

These equations look like regular diffusion equations, but they are governed by *nonequilibrium* analogs of chemical potentials (see detailed derivation of nonequilibrium chemical potentials in the Appendix C):

$$\mu_{\mathcal{A}} = T_{\mathcal{A}}\ln c^{\mathcal{A}} + T_{\mathcal{A}}B_{\mathcal{A}}c^{\mathcal{A}} + \overline{T}B_{\mathcal{A}\mathcal{B}}c^{\mathcal{B}} \tag{11}$$

and a similar equation for $\mu_{\mathcal{B}}$. The virial coefficients are defined each with its own temperature, as $B_{ij} = \int[1 - e^{-u^{ij}(\mathbf{r})/T_{ij}}]d^3\mathbf{r}$.

Nonequilibrium chemical potentials, as quantities whose gradients determine fluxes, were discussed in Ref. [20]. It was shown that, unlike its equilibrium counterpart, a nonequilibrium chemical potential, cannot in general be obtained as a derivative of a free energy. This is what makes our case interesting, because our nonequilibrium chemical potentials $\mu_{\mathcal{A}}$ and $\mu_{\mathcal{B}}$ are in fact the partial derivatives $\mu_i = \frac{\partial f}{\partial c^i}$ of a function, which looks like a two-temperature free energy (per unit volume):

$$\begin{aligned} f &= T_{\mathcal{A}}c^{\mathcal{A}}\ln(c^{\mathcal{A}}/e) + T_{\mathcal{B}}c^{\mathcal{B}}\ln(c^{\mathcal{B}}/e)\\ &\quad + (1/2)T_{\mathcal{A}}B_{\mathcal{A}}c_{\mathcal{A}}^2 + (1/2)T_{\mathcal{B}}B_{\mathcal{B}}c_{\mathcal{B}}^2 + \overline{T}B_{\mathcal{A}\mathcal{B}}c^{\mathcal{A}}c^{\mathcal{B}}.\end{aligned} \tag{12}$$

Furthermore, in a very similar manner, we could also consider the case of finite range interaction potentials, when spatial gradient terms are present in the effective free energy, as in Ref. [20], but chemical potentials are still the derivatives of this free energy.

VI. INSTABILITY OF THE UNIFORM STATE AND "SPINODAL"

Suppose that $c_0^{\mathcal{A}}$ and $c_0^{\mathcal{B}}$ are the averaged spatially uniform concentrations of the two components. By introducing space-dependent perturbations $c^i(\mathbf{r}) = c_0^i + \delta c^i(\mathbf{r})$, we perform a

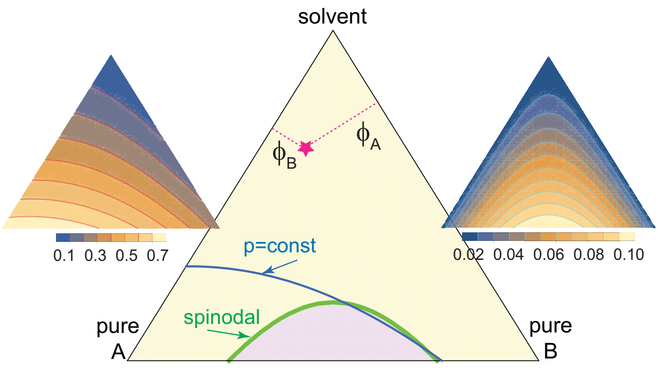

FIG. 2. (Color online) *Main figure:* Triangular phase diagram, for a three-component system of $\mathcal{A}+\mathcal{B}+$ solvent; for every point in the triangle (exemplified by a star), the volume fractions of $\mathcal{A}$ and $\mathcal{B}$ particles are given by the distances to the triangle sides, while the volume fraction of the solvent $1-\phi_{\mathcal{A}}-\phi_{\mathcal{B}}$ is the distance to the bottom of the triangle. The symmetric line is the "spinodal" Eq. (13); below this line, the uniformly mixed state is unstable. For this figure, we used the parameters $\beta = B_{\mathcal{AB}}/\sqrt{B_{\mathcal{A}}B_{\mathcal{B}}} = 2.95$ (an instability in an equilibrium system requires $\beta > 3$), $T_{\mathcal{A}}/T_{\mathcal{B}} = 3$ (which is sufficient to open an instability region). The asymmetric curve is a constant osmotic pressure line (14) $pB_{\mathcal{A}}B_{\mathcal{B}}/B_{\mathcal{AB}}\overline{T} = 0.493$, assuming $B_{\mathcal{A}} = B_{\mathcal{B}}$; this line crosses the instability region thus indicating the possibility of two coexisting phases. *Left inset:* Lines of constant pressure (14). *Right inset:* Level lines of the left-hand side of formula (13).

linear stability analysis. It shows that an instability occurs macroscopically under the condition

$$\frac{\phi_{\mathcal{A}}}{1+\phi_{\mathcal{A}}}\frac{\phi_{\mathcal{B}}}{1+\phi_{\mathcal{B}}} > \frac{T_{\mathcal{A}}T_{\mathcal{B}}}{\overline{T}}\frac{B_{\mathcal{A}}B_{\mathcal{B}}}{B_{\mathcal{AB}}^2}, \tag{13}$$

where we have defined the volume fractions $c^i B_i = \phi_i$. In general, the virial coefficients depend on temperature in a complex way. A simple limit that we study in the following corresponds to purely excluded volume interaction potentials such that the B do not depend on temperature.

In the plane $\phi_{\mathcal{A}}$ and $\phi_{\mathcal{B}}$, the nonequilibrium "spinodal line" (13) is a hyperbola (see Fig. 2). The contrast between temperatures favors instability; it works in the same direction as contrast between interactions. But this instability, to have a physical meaning, must occur at $\phi_{\mathcal{A}}+\phi_{\mathcal{B}} < 1$. For instance, consider the most symmetric case of identical particles in all respects except driven by different temperatures: $\zeta_{\mathcal{A}} = \zeta_{\mathcal{B}}$, $B_{\mathcal{A}} = B_{\mathcal{B}} = B_{\mathcal{AB}}$. In this case the spinodal line is in the physical range $\phi_{\mathcal{A}}+\phi_{\mathcal{B}} < 1$ if the ratio of the two temperatures is outside the range $17-12\sqrt{2} \approx 0.029 < \frac{T_{\mathcal{A}}}{T_{\mathcal{B}}} < 17+12\sqrt{2} \approx 34$ (A more general discussion of the instability condition driven by a contrast of temperatures, frictions, and interactions is given in Appendix D). Thus, a numerically large temperature contrast is required to achieve an instability by temperature difference alone. This is roughly consistent with numerical observation of Ganai *et al.* [21], who used $T_{\mathcal{A}}/T_{\mathcal{B}} = 20$.

VII. PRESSURE AND "BINODAL"

To address not only the loss of stability of the uniformly mixed state, but also the steady state phase segregation, in addition to nonequilibrium chemical potentials we also need a nonequilibrium equivalent of the osmotic pressure. Given that nonequilibrium chemical potentials are the derivatives of a "quasi-free energy" (12), we can expect the analog of osmotic pressure to be given by the Gibbs-Duhem formula $p = c^{\mathcal{A}}\mu_{\mathcal{A}} + c^{\mathcal{B}}\mu_{\mathcal{B}} - f$, yielding

$$p = T_{\mathcal{A}}c^{\mathcal{A}} + T_{\mathcal{B}}c^{\mathcal{B}} + (1/2)T_{\mathcal{A}}B_{\mathcal{A}}c_{\mathcal{A}}^2 + (1/2)T_{\mathcal{B}}B_{\mathcal{B}}c_{\mathcal{B}}^2 + \overline{T}B_{\mathcal{AB}}c^{\mathcal{A}}c^{\mathcal{B}}. \tag{14}$$

We have directly derived this result in two independent ways, first, by computing the force exerted on the wall by replacing the wall by a potential ramp (not necessarily the same for both particle species), and second by relating the pressure to the pair correlation function in the bulk [31,32]. Since we do not consider nonspherical particles, which can experience a torque upon interactions with the wall, the complications studied in recent works [33–35] do not play any role, and, indeed, the two derivations (details of both derivations are presented in Appendix E) yield identical results (14). This result means that the densities of two coexisting phases at steady state are found by the Maxwell common tangent construction based upon the quasi-free energy function (12). Although we derive here this result to the second virial approximation and for spatially uniform states only, it is possible to derive, in a similar way, the nonlocal corrections to the effective free energy. Based on that, we argue that our results remain correct more generally as long as the range of interaction potential between particles and/or their sizes remain microscopic, while all the complications discussed in Refs. [20,34,35] and having to do with nonlocal dependence of fluxes on densities and their derivatives are governed by finite particle sizes.

The calculation of the dissipation (6) can be generalized to the dissipation per unit volume of the solution:

$$w = c^{\mathcal{A}}c^{\mathcal{B}}\frac{T_{\mathcal{A}}-T_{\mathcal{B}}}{T_{\mathcal{A}}\zeta_{\mathcal{B}}+T_{\mathcal{B}}\zeta_{\mathcal{A}}}\int\left(\frac{\partial u^{AB}}{\partial \mathbf{r}}\right)^2 e^{-\frac{u^{AB}(\mathbf{r})}{T}}\, d^3\mathbf{r}. \tag{15}$$

It shows that in a phase-separated system the dissipation mostly happens around the phase boundary. Interestingly, a more detailed derivation of the dissipated power shows that, in a system with pairwise interactions only, the dissipated power can be expressed in terms of the two- and three-body correlation functions only, as shown in Appendix F.

VIII. CONCLUSION AND DISCUSSION

Although this is not our main thrust, it is interesting to discuss the implications of our approach for mixtures of passive particles and more "traditional" polar active particles, such as RTPs or ABPs [10]. One of our important results is that the virial corrections to the dilute limit might be interesting. In dilute systems, the reorientation time of one swimmer is much smaller than the time between collisions: $\tau_r \ll \tau_c$. Then the particle trajectory between collisions is that of a random walk, characterized by an effective diffusivity. We can then identify $T_{\mathcal{B}} = T$ with the ambient temperature while $T_{\mathcal{A}} - T_{\mathcal{B}} \simeq v_0^2\tau_r\zeta/6$ is related to the level of activity measured by the swimming speed v_0. The collision time is estimated by the Smoluchowski formula, leading to the condition $\tau_r \ll \tau_c$ conveniently formulated in terms of the Péclet number $v_0\tau_r/b \equiv \mathrm{Pe} \ll 1/\sqrt{\phi}$, b being the particle size. In this limit, the behavior of swimmers between collisions is correctly described by the two-temperature model.

Whether multiple collisions for swimmers average to build up an effective Boltzmann distribution (3) remains an open question. It is certainly true for repulsive interparticle potentials in three dimensions, but it might require further scrutiny for other cases.

Returning to our other results, one may want to have a qualitative feel for the existence of the phase segregation. It is best given by considering a large temperature contrast, $T_\mathcal{A} \gg T_\mathcal{B}$; for the sake of the argument, we consider the case where $T_\mathcal{B} = 0$, when $\mathcal{B}$ particles do not move at all unless pushed by $\mathcal{A}$ particles. Assuming also for simplicity purely repulsive, excluded volume interactions, if two (or several) $\mathcal{B}$ particles come close enough to one another, then they cannot part, leading to aggregation and assembly of $\mathcal{B}$ particles in a separate $\mathcal{B}$-rich phase under these nonequilibrium conditions.

The idea to replace local active drive with some "active temperature" was discussed many times in the biophysical context; see, for example, Ref. [36]. With all the known drawbacks [37], it remains attractive because of its simplicity. Nevertheless, previous works treated specific and rather complex models. By contrast, our simple model is reduced to bare essentials. In this sense, a mixture of particles exposed to different thermostats which we examined represents a peculiar class of active systems: peculiar in the sense that each component would not be active on its own, while the mixture is active. Despite all the limitations, we believe that the theory developed here is useful because it is physically transparent and may be instructive as a source of physical intuition for these highly unusual driven systems.

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APPENDIX A: TWO PARTICLES: "CENTER OF FRICTION" DIFFUSION

In the main text, we analyzed the relative motion of two particles $\mathcal{A}$ and $\mathcal{B}$ by looking at the variable $r = x_\mathcal{A} - x_\mathcal{B}$. It is interesting to find also how the presence of two distinct temperatures affects their joint diffusion in space. To do so, it is convenient to define their joint coordinate R in such a way that the noises in r and R are statistically independent. This is achieved by choosing

$$R = \frac{\zeta_\mathcal{A} T_\mathcal{B}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}} x_\mathcal{A} + \frac{\zeta_\mathcal{B} T_\mathcal{A}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}} x_\mathcal{B}. \tag{A1}$$

Then the Langevin equation for R reads

$$(\zeta_\mathcal{A} + \zeta_\mathcal{B})\dot{R} = \frac{T_\mathcal{B} - T_\mathcal{A}}{\overline{T}} F(r) + \sqrt{2(\zeta_\mathcal{A} + \zeta_\mathcal{B})\frac{T_\mathcal{A} T_\mathcal{B}}{\overline{T}}}\,\xi_R(t), \tag{A2}$$

where $\xi_R(t)$ is a zero mean unit variance Gaussian white noise (independent of ξ_r, as stated), and $\overline{T} = (\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A})/(\zeta_\mathcal{A} + \zeta_\mathcal{B})$ was defined in the main text. In an equilibrium system, at $T_\mathcal{A} = T_\mathcal{B}$, the interparticle force $F(r)$ does not couple to the joint motion. Not so out of equilibrium: since on average the force $F(r)$ vanishes (by symmetry), $\langle F(r) \rangle = 0$, the $F(r)$ term provides an additional noise driving the diffusion of the variable R. Of course, it is not a white noise, so that the dynamics of R is not a simple diffusion on time scales shorter or comparable to the correlation time of r. But on longer time scales R undergoes simple diffusion, with a diffusion coefficient which can be directly read out of the Langevin equation (A2), because $F[r(t)]$ is statistically independent from $\xi_R(t)$:

$$D_R = \frac{T_\mathcal{A} T_\mathcal{B}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}} + \frac{1}{2}\left(\frac{T_\mathcal{A} - T_\mathcal{B}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}\right)^2 (F^2)_{\omega=0}; \tag{A3}$$

here the power the spectrum of the force $F(r)$ at zero frequency is

$$(F^2)_{\omega=0} = \int_{-\infty}^{\infty} \langle F[r(t)] F[r(t+\tau)] \rangle d\tau. \tag{A4}$$

In order to find the power spectrum of the force, we Fourier transform the Langevin equation for r [Eq. (2) of the main text]:

$$F_\omega = \imath\omega\zeta_r r_\omega - (2\zeta_r \overline{T})^{1/2}\xi_\omega. \tag{A5}$$

Then multiplying it by the complex conjugate and assuming $[\omega F_\omega]_{\omega=0} = 0$, we obtain $(F^2)_{\omega=0} = 2\zeta_r \overline{T}$. This yields

$$D_R = \frac{T_\mathcal{A} T_\mathcal{B}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}} + \frac{1}{2}\frac{(T_\mathcal{A} - T_\mathcal{B})^2}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}\frac{\zeta_\mathcal{A}\zeta_\mathcal{B}}{(\zeta_\mathcal{A} + \zeta_\mathcal{B})^2}. \tag{A6}$$

We see that the difference in temperatures, independently of the sign, enhances the joint diffusion.

It is also instructive to write the Fokker-Planck equation in terms of the variables r and R:

$$\begin{aligned}\partial_t P = &-\left[\frac{\zeta_\mathcal{A} + \zeta_\mathcal{B}}{\zeta_\mathcal{A}\zeta_\mathcal{B}}\right]\partial_r(F(r)P) - \left[\frac{T_\mathcal{B} - T_\mathcal{A}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}\right]F(r)\partial_R P \\ &+ \left[\frac{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}{\zeta_\mathcal{A}\zeta_\mathcal{B}}\right]\partial_r^2 P + \left[\frac{T_\mathcal{A} T_\mathcal{B}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}\right]\partial_R^2 P.\end{aligned} \tag{A7}$$

Here the right-hand side has explicitly the form of a divergence and can be written as

$$\partial_t P = -\partial_r J_r - \partial_R J_R, \tag{A8}$$

where the components of the flux are

$$J_r = \left[\frac{\zeta_\mathcal{A} + \zeta_\mathcal{B}}{\zeta_\mathcal{A}\zeta_\mathcal{B}}\right]F(r)P - \left[\frac{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}{\zeta_\mathcal{A}\zeta_\mathcal{B}}\right]\partial_r P, \tag{A9a}$$

$$J_R = \left[\frac{T_\mathcal{B} - T_\mathcal{A}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}\right]F(r)P - \left[\frac{T_\mathcal{A} T_\mathcal{B}}{\zeta_\mathcal{A} T_\mathcal{B} + \zeta_\mathcal{B} T_\mathcal{A}}\right]\partial_R P. \tag{A9b}$$

APPENDIX B: HIERARCHY OF EQUATIONS FOR THE CORRELATION FUNCTIONS

1. Notations, definitions, and symmetries

As stated in the main text, we operate with a multidimensional Fokker-Planck equation for the probability density P as a function of the positions of all the particles in the system. More specifically, consider a system of $N_{\mathcal{A}}$ particles $\mathcal{A}$ and $N_{\mathcal{B}}$ particles $\mathcal{B}$; their coordinates are $\mathbf{r}_1^{\mathcal{A}},\mathbf{r}_2^{\mathcal{A}},\dots,\mathbf{r}_{N_{\mathcal{A}}}^{\mathcal{A}}$ and $\mathbf{r}_1^{\mathcal{B}},\mathbf{r}_2^{\mathcal{B}},\dots,\mathbf{r}_{N_{\mathcal{B}}}^{\mathcal{B}}$. The potential energy includes single-particle potentials and pairwise additive interactions:

$$\begin{aligned} U&\big(\mathbf{r}_1^{\mathcal{A}},\mathbf{r}_2^{\mathcal{A}},\dots,\mathbf{r}_{N_{\mathcal{A}}}^{\mathcal{A}};\mathbf{r}_1^{\mathcal{B}},\mathbf{r}_2^{\mathcal{B}},\dots,\mathbf{r}_{N_{\mathcal{B}}}^{\mathcal{B}}\big)\\ &=\sum_i^{N_{\mathcal{A}}} u_1^{\mathcal{A}}\big(\mathbf{r}_i^{\mathcal{A}}\big)+\sum_j^{N_{\mathcal{B}}} u_1^{\mathcal{B}}\big(\mathbf{r}_j^{\mathcal{A}}\big)+\frac{1}{2}\sum_{i\neq j}^{N_{\mathcal{A}}} u^{\mathcal{AA}}\big(\mathbf{r}_i^{\mathcal{A}}-\mathbf{r}_j^{\mathcal{A}}\big)\\ &\quad+\sum_i^{N_{\mathcal{A}}}\sum_j^{N_{\mathcal{B}}} u^{\mathcal{AB}}\big(\mathbf{r}_i^{\mathcal{A}}-\mathbf{r}_j^{\mathcal{B}}\big)+\frac{1}{2}\sum_{i\neq j}^{N_{\mathcal{B}}} u^{\mathcal{BB}}\big(\mathbf{r}_i^{\mathcal{B}}-\mathbf{r}_j^{\mathcal{B}}\big). \end{aligned} \tag{B1}$$

The probability density P is also a function of all the coordinates: $P=P(\mathbf{r}_1^{\mathcal{A}},\mathbf{r}_2^{\mathcal{A}},\dots,\mathbf{r}_{N_{\mathcal{A}}}^{\mathcal{A}};\mathbf{r}_1^{\mathcal{B}},\mathbf{r}_2^{\mathcal{B}},\dots,\mathbf{r}_{N_{\mathcal{B}}}^{\mathcal{B}})$. It is normalized:

$$\int P\big(\mathbf{r}_1^{\mathcal{A}},\mathbf{r}_2^{\mathcal{A}},\dots,\mathbf{r}_{N_{\mathcal{A}}}^{\mathcal{A}};\mathbf{r}_1^{\mathcal{B}},\mathbf{r}_2^{\mathcal{B}},\dots,\mathbf{r}_{N_{\mathcal{B}}}^{\mathcal{B}}\big)d\{\mathbf{r}\}=1. \tag{B2}$$

Define single-particle probability densities, two-particle probability densities, etc., as

$$p_1^{\mathcal{A}}(\mathbf{r})=\int\delta\big(\mathbf{r}_i^{\mathcal{A}}-\mathbf{r}\big)P(\{\mathbf{r}\})d\{\mathbf{r}\}, \tag{B3a}$$

$$p_1^{\mathcal{B}}(\mathbf{r})=\int\delta\big(\mathbf{r}_i^{\mathcal{B}}-\mathbf{r}\big)P(\{\mathbf{r}\})d\{\mathbf{r}\}, \tag{B3b}$$

$$p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')=\int\delta\big(\mathbf{r}_i^{\mathcal{A}}-\mathbf{r}\big)\delta\big(\mathbf{r}_j^{\mathcal{A}}-\mathbf{r}'\big)P(\{\mathbf{r}\})d\{\mathbf{r}\}, \tag{B3c}$$

$$p_2^{\mathcal{BB}}(\mathbf{r},\mathbf{r}')=\int\delta\big(\mathbf{r}_i^{\mathcal{B}}-\mathbf{r}\big)\delta\big(\mathbf{r}_j^{\mathcal{B}}-\mathbf{r}'\big)P(\{\mathbf{r}\})d\{\mathbf{r}\}, \tag{B3d}$$

$$p_2^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')=\int\delta\big(\mathbf{r}_i^{\mathcal{A}}-\mathbf{r}\big)\delta\big(\mathbf{r}_j^{\mathcal{B}}-\mathbf{r}'\big)P(\{\mathbf{r}\})d\{\mathbf{r}\}. \tag{B3e}$$

There are four types of three-particle densities: $p_3^{\mathcal{AAA}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')$, $p_3^{\mathcal{AAB}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')$, $p_3^{\mathcal{ABB}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')$, $p_3^{\mathcal{BBB}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')$. All these densities are independent of i and j etc.; i.e., the probability density is the same for every particle of a given species.

There are several normalization and symmetry properties ($\mathcal{X},\mathcal{Y}=\mathcal{A},\mathcal{B}$):

$$\int p_1^{\mathcal{X}}(\mathbf{r})\,d\mathbf{r}=1, \tag{B4a}$$

$$p_2^{\mathcal{XX}}(\mathbf{r},\mathbf{r}')=p_2^{\mathcal{XX}}(\mathbf{r}',\mathbf{r}), \tag{B4b}$$

$$p_2^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}')=p_2^{\mathcal{YX}}(\mathbf{r}',\mathbf{r}), \tag{B4c}$$

$$\int p_2^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}')\,d\mathbf{r}\,d\mathbf{r}'=1, \tag{B4d}$$

$$\int p_2^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}')\,d\mathbf{r}'=p_1^{\mathcal{X}}(\mathbf{r}), \tag{B4e}$$

$$\int p_2^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}')\,d\mathbf{r}=p_1^{\mathcal{Y}}(\mathbf{r}'). \tag{B4f}$$

2. Fokker-Planck equations for the densities

Integrating out all variables except for one, or except for two, etc., we obtain the following dynamic equations for the densities:

$$\begin{aligned} \frac{\partial p_1^{\mathcal{A}}(\mathbf{r})}{\partial t}&=\frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\nabla_{\mathbf{r}}^2 p_1^{\mathcal{A}}(\mathbf{r})+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u_1^{\mathcal{A}}(\mathbf{r})}{\partial\mathbf{r}}p_1^{\mathcal{A}}(\mathbf{r})\right]\\ &\quad+\frac{N_{\mathcal{A}}-1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\int\frac{\partial u^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')}{\partial\mathbf{r}}p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')d\mathbf{r}'\right]\\ &\quad+\frac{N_{\mathcal{B}}}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}}\left[\int\frac{\partial u^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')}{\partial\mathbf{r}}p_2^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')d\mathbf{r}'\right]. \end{aligned} \tag{B5}$$

A similar equation is obtained for $p_1^{\mathcal{B}}(\mathbf{r})$, which is not given for brevity. For large numbers of particles, we can replace $N_{\mathcal{A}}-1\simeq N_{\mathcal{A}}$:

$$\begin{aligned} \frac{\partial p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')}{\partial t}&=\frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\big(\nabla_{\mathbf{r}}^2+\nabla_{\mathbf{r}'}^2\big)p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')\\ &\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u_1^{\mathcal{A}}(\mathbf{r})}{\partial\mathbf{r}}p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')\right]\\ &\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')}{\partial\mathbf{r}}p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')\right]\\ &\quad+\frac{N_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\int\frac{\partial u^{\mathcal{AA}}(\mathbf{r},\mathbf{r}'')}{\partial\mathbf{r}}p_3^{\mathcal{AAA}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right]\\ &\quad+\frac{N_{\mathcal{B}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\int\frac{\partial u^{\mathcal{AB}}(\mathbf{r},\mathbf{r}'')}{\partial\mathbf{r}}p_3^{\mathcal{AAB}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right]\\ &\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}'}\left[\frac{\partial u_1^{\mathcal{A}}(\mathbf{r}')}{\partial\mathbf{r}'}p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')\right]\\ &\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}'}\left[\frac{\partial u^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')}{\partial\mathbf{r}'}p_2^{\mathcal{AA}}(\mathbf{r},\mathbf{r}')\right]\\ &\quad+\frac{N_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}'}\left[\int\frac{\partial u^{\mathcal{AA}}(\mathbf{r}',\mathbf{r}'')}{\partial\mathbf{r}'}p_3^{\mathcal{AAA}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right]\\ &\quad+\frac{N_{\mathcal{B}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}'}\left[\int\frac{\partial u^{\mathcal{AB}}(\mathbf{r}',\mathbf{r}'')}{\partial\mathbf{r}'}p_3^{\mathcal{AAB}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right]. \end{aligned} \tag{B6}$$

A similar equation (not given here) is obtained for $p_2^{\mathcal{BB}}$. But the equation for the mixed probability deserves to be written:

$$\begin{aligned} \frac{\partial p_2^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')}{\partial t}&=\left(\frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\nabla_{\mathbf{r}}^2+\frac{T_{\mathcal{B}}}{\zeta_{\mathcal{B}}}\nabla_{\mathbf{r}'}^2\right)p_2^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')\\ &\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u_1^{\mathcal{A}}(\mathbf{r})}{\partial\mathbf{r}}p_2^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')\right]\\ &\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')}{\partial\mathbf{r}}p_2^{\mathcal{AB}}(\mathbf{r},\mathbf{r}')\right]\\ &\quad+\frac{N_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\int\frac{\partial u^{\mathcal{AA}}(\mathbf{r},\mathbf{r}'')}{\partial\mathbf{r}}p_3^{\mathcal{ABA}}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right] \end{aligned}$$

$$
\begin{aligned}
&+\frac{N_{\mathcal{B}}}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\int \frac{\partial u^{\mathcal{A}B}(\mathbf{r},\mathbf{r}'')}{\partial \mathbf{r}} p_3^{ABB}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right]\\
&+\frac{1}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}'}\left[\frac{\partial u_1^{\mathcal{B}}(\mathbf{r}')}{\partial \mathbf{r}'} p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')\right]\\
&+\frac{1}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}'}\left[\frac{\partial u^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')}{\partial \mathbf{r}'} p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')\right]\\
&+\frac{N_{\mathcal{A}}}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}'}\left[\int \frac{\partial u^{BA}(\mathbf{r}',\mathbf{r}'')}{\partial \mathbf{r}'} p_3^{ABA}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right]\\
&+\frac{N_{\mathcal{B}}}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}'}\left[\int \frac{\partial u^{\mathcal{B}B}(\mathbf{r}',\mathbf{r}'')}{\partial \mathbf{r}'} p_3^{ABB}(\mathbf{r},\mathbf{r}',\mathbf{r}'')\,d\mathbf{r}''\right].
\end{aligned}
\tag{B7}
$$

The equations for p_3 involve p_4, and so on, *ad infinitum*.

If the density is small enough, we can neglect all triple collisions, i.e., directly discard all terms involving p_3. Indeed, given the normalization, any term containing p_2 in Eq. (B6) or (B6) is of order $1/V^2$, while every term containing p_3 is of order N/V^3. This, of course, simplifies the equations quite dramatically and reduces them essentially to what we obtained for the case of two particles. Omitting the single-particle potential terms, we obtain

$$
\begin{aligned}
\frac{\partial p_2^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')}{\partial t} &= \frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\left(\nabla_{\mathbf{r}}^2+\nabla_{\mathbf{r}'}^2\right)p_2^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')\\
&\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')}{\partial \mathbf{r}} p_2^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')\right]\\
&\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}'}\left[\frac{\partial u^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')}{\partial \mathbf{r}'} p_2^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')\right]
\end{aligned}
\tag{B8}
$$

(and a similar equation for $p_2^{\mathcal{B}B}$),

$$
\begin{aligned}
\frac{\partial p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')}{\partial t} &= \left(\frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}}\nabla_{\mathbf{r}}^2+\frac{T_{\mathcal{B}}}{\zeta_{\mathcal{B}}}\nabla_{\mathbf{r}'}^2\right)p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')\\
&\quad+\frac{1}{\zeta_{\mathcal{A}}}\partial_{\mathbf{r}}\left[\frac{\partial u^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')}{\partial \mathbf{r}} p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')\right]\\
&\quad+\frac{1}{\zeta_{\mathcal{B}}}\partial_{\mathbf{r}'}\left[\frac{\partial u^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')}{\partial \mathbf{r}'} p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}')\right].
\end{aligned}
\tag{B9}
$$

These equations are simple enough to guess the solution based on our knowledge of the two-particle case:

$$
p_2^{\mathcal{A}A}(\mathbf{r},\mathbf{r}') = p_1^{\mathcal{A}}(\mathbf{r})p_1^{\mathcal{A}}(\mathbf{r}')\exp\left[-\frac{u^{\mathcal{A}A}(\mathbf{r}-\mathbf{r}')}{T_{\mathcal{A}}}\right], \tag{B10a}
$$

$$
p_2^{\mathcal{B}B}(\mathbf{r},\mathbf{r}') = p_1^{\mathcal{B}}(\mathbf{r})p_1^{\mathcal{B}}(\mathbf{r}')\exp\left[-\frac{u^{\mathcal{B}B}(\mathbf{r}-\mathbf{r}')}{T_{\mathcal{B}}}\right], \tag{B10b}
$$

$$
p_2^{\mathcal{A}B}(\mathbf{r},\mathbf{r}') = p_1^{\mathcal{A}}(\mathbf{r})p_1^{\mathcal{B}}(\mathbf{r}')\exp\left[-\frac{u^{\mathcal{A}B}(\mathbf{r}-\mathbf{r}')}{\overline{T}}\right]. \tag{B10c}
$$

Of course, the central feature of this result is the appearance of the average temperature, as defined in the main text

$$
\overline{T} = \frac{\zeta_{\mathcal{A}}T_{\mathcal{B}}+\zeta_{\mathcal{B}}T_{\mathcal{A}}}{\zeta_{\mathcal{A}}+\zeta_{\mathcal{B}}}. \tag{B11}
$$

In order to obtain the result (9), we look for a solution of the form $p_2^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}') = q^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}')\exp[-\beta u^{\mathcal{X}Y}(\mathbf{r},\mathbf{r}')]$, plug it into Eq. (B8) or (B9), and discover, that in the remaining equation for q, the variables separate, meaning that q factorizes into a factor that depends only on $\mathbf{r}$ and a factor that depends only on $\mathbf{r}'$.

APPENDIX C: DIFFUSION EQUATIONS AND NONEQUILIBRIUM CHEMICAL POTENTIALS

Plugging the *ansatz* (9) [(B10)] into Eqs. (8) [(B5)], we obtain closed results for the densities. This involves the integral

$$
\int \frac{\partial u^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')}{\partial \mathbf{r}} p_1^{\mathcal{A}}(\mathbf{r}')e^{-\frac{u^{\mathcal{A}A}(\mathbf{r},\mathbf{r}')}{T_{\mathcal{A}}}}\,d\mathbf{r}', \tag{C1}
$$

which can be integrated by parts. Finally, we obtain equations that look like diffusion equations for a regular system in contact with a thermostat,

$$
\frac{\partial c^{\mathcal{A}}(\mathbf{r})}{\partial t} = \frac{1}{\zeta_{\mathcal{A}}}\frac{\partial}{\partial \mathbf{r}}\left(c^{\mathcal{A}}\frac{\partial \mu_{\mathcal{A}}}{\partial \mathbf{r}}\right), \tag{C2a}
$$

$$
\frac{\partial c^{\mathcal{B}}(\mathbf{r})}{\partial t} = \frac{1}{\zeta_{\mathcal{B}}}\frac{\partial}{\partial \mathbf{r}}\left(c^{\mathcal{B}}\frac{\partial \mu_{\mathcal{B}}}{\partial \mathbf{r}}\right), \tag{C2b}
$$

but these equations contain *nonequilibrium chemical potentials*, as stated in the main text [Eq. (11)].

1. Linear stability analysis

Suppose that $c_0^{\mathcal{A}}$ and $c_0^{\mathcal{B}}$ are the averaged spatially uniform concentrations of both components. By introducing small space-dependent perturbations $c^{\mathcal{A}}(\mathbf{r}) = c_0^{\mathcal{A}}+\delta c^{\mathcal{A}}(\mathbf{r})$ and $c^{\mathcal{B}}(\mathbf{r}) = c_0^{\mathcal{B}}+\delta c^{\mathcal{B}}(\mathbf{r})$, we perform a linear stability analysis in the standard way:

$$
\begin{aligned}
\frac{\partial \delta c^{\mathcal{A}}}{\partial t} &= \frac{1}{\zeta_{\mathcal{A}}}\nabla^2\left[\left(T_{\mathcal{A}}+T_{\mathcal{A}}c_0^{\mathcal{A}}B_{\mathcal{A}}\right)\delta c^{\mathcal{A}}+\left(\overline{T}c_0^{\mathcal{A}}B_{\mathcal{A}B}\right)\delta c^{\mathcal{B}}\right]\\
\frac{\partial \delta c^{\mathcal{B}}}{\partial t} &= \frac{1}{\zeta_{\mathcal{B}}}\nabla^2\left[\left(\overline{T}c_0^{\mathcal{B}}B_{\mathcal{A}B}\right)\delta c^{\mathcal{A}}+\left(T_{\mathcal{B}}+T_{\mathcal{B}}c_0^{\mathcal{B}}B_{\mathcal{B}}\right)\delta c^{\mathcal{B}}\right].
\end{aligned}
\tag{C3}
$$

This shows that an instability occurs (at $q=0$, i.e., macroscopically) under the condition that the determinant of this matrix vanishes; i.e., the system is unstable if

$$
c_0^{\mathcal{A}}c_0^{\mathcal{B}}\overline{T}^2B_{\mathcal{A}B}^2 > T_AT_B\left(1+c_0^{\mathcal{A}}B_{\mathcal{A}}\right)\left(1+c_0^{\mathcal{B}}B_{\mathcal{B}}\right). \tag{C4}
$$

At the instability, the unstable combination (eigenvector whose eigenvalue flips sign) is

$$
\frac{\delta c^{\mathcal{A}}(\mathbf{r})\zeta_{\mathcal{A}}}{\sqrt{T_{\mathcal{A}}B_{\mathcal{A}}c_0^{\mathcal{A}}\left(1+B_{\mathcal{A}}c_0^{\mathcal{A}}\right)}} - \frac{\delta c^{\mathcal{B}}(\mathbf{r})\zeta_{\mathcal{B}}}{\sqrt{T_{\mathcal{B}}B_{\mathcal{B}}c_0^{\mathcal{B}}\left(1+B_{\mathcal{B}}c_0^{\mathcal{B}}\right)}}. \tag{C5}
$$

In the plane ($c_0^{\mathcal{A}}$, $c_0^{\mathcal{B}}$), the spinodal line (C4) is a hyperbola (Fig. 3). A better way to represent it is to use a triangular phase diagram as given in Fig. 2.

APPENDIX D: NONEQUILIBRIUM "SPINODAL" LINE FOR ATHERMAL PARTICLES

The dimensionless parameters of the system are as follows:

(1) Contrast of excluded volumes,

$$
\beta = \frac{B_{\mathcal{A}B}}{\sqrt{B_{\mathcal{A}}B_{\mathcal{B}}}}. \tag{D1}
$$

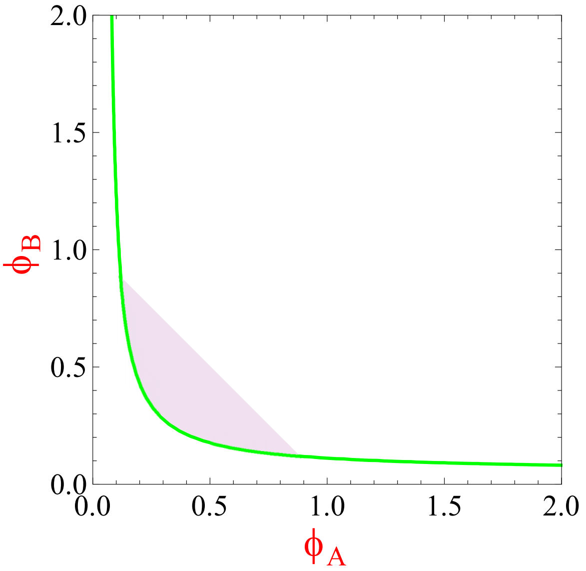

FIG. 3. (Color online) Spinodal line (C4), shown for one particular choice of parameters ($T_{\mathcal{A}}/T_{\mathcal{B}}, \zeta_{\mathcal{A}}/\zeta_{\mathcal{B}}, B_{\mathcal{A}}B_{\mathcal{B}}/B_{\mathcal{AB}}^2$). Physical meaning has only region $\phi_{\mathcal{A}} + \phi_{\mathcal{B}} < 1$, the instability region within this region is shaded.

(2) Contrast of temperatures,

$$\tau = \frac{T_{\mathcal{A}} - T_{\mathcal{B}}}{T_{\mathcal{A}} + T_{\mathcal{B}}}\ ; \quad -1 < \tau < 1. \tag{D2}$$

(3) Contrast of frictions,

$$\kappa = \frac{\zeta_{\mathcal{A}} - \zeta_{\mathcal{B}}}{\zeta_{\mathcal{A}} + \zeta_{\mathcal{B}}}\ ; \quad -1 < \kappa < 1. \tag{D3}$$

The condition that the spinodal exists within the physical range $\phi_{\mathcal{A}} + \phi_{\mathcal{B}} < 1$ reads

$$\frac{4\kappa^2\tau^2 - 4\kappa\tau + 1}{1 - \tau^2}\beta^2 > 9. \tag{D4}$$

This condition is presented graphically in two different ways, in Fig. 4 in the form of a three-dimensional surface and in Fig. 5 as an array of two-dimensional plots. Beautifully, the contrast of frictions κ becomes irrelevant for the equilibrium system, when $\tau = 0$.

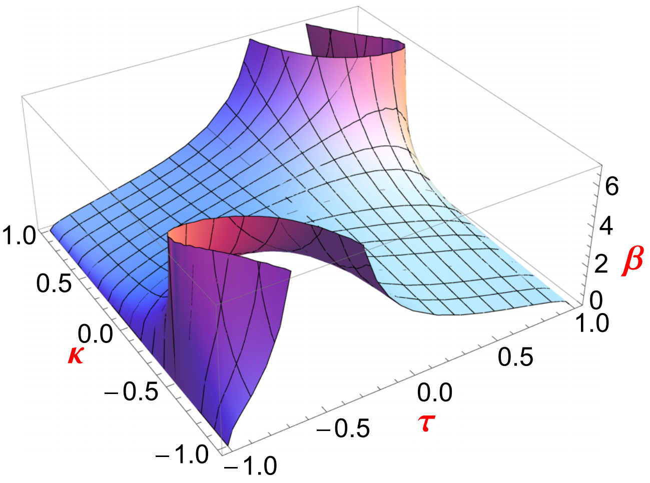

FIG. 4. (Color online) Instability exists above this surface; see condition (D4).

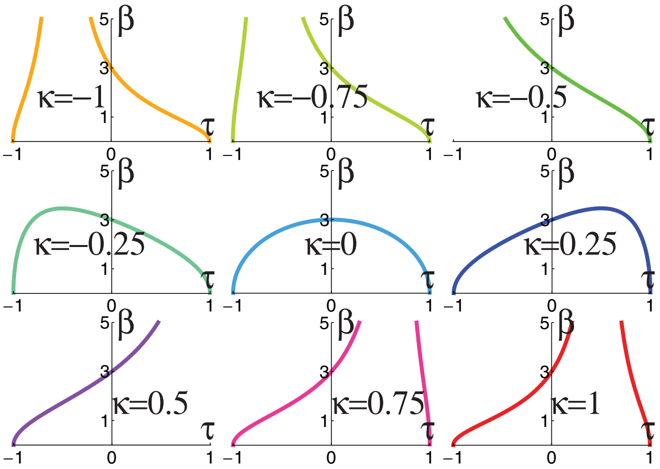

FIG. 5. (Color online) For every κ, instability exists above the line, according to Eqs. (D4).

APPENDIX E: NONEQUILIBRIUM OSMOTIC PRESSURE AND “BINODAL”

To address the steady state phase segregation, in addition to the nonequilibrium chemical potentials we also need to define a nonequilibrium osmotic pressure. We derive it in two different ways.

1. Derivation 1

To find the osmotic pressure, imagine that the system “feels” single-particle potentials $u_1^{\mathcal{A}}(\mathbf{r})$ and $u_1^{\mathcal{B}}(\mathbf{r})$ such that they are both like a box, except that one wall of this box has a (not necessarily very) sharp potential “ramp” in the direction, perpendicular to the wall: $u_1^{\mathcal{A},B}(\mathbf{r}) = f_{\mathcal{A},B}x$, as shown in Fig. 6. In this case, the pressure is found according to

$$p = f_{\mathcal{A}} \int_0^\infty c^{\mathcal{A}}(x)\,dx + f_{\mathcal{B}} \int_0^\infty c^{\mathcal{B}}(x)\,dx, \tag{E1}$$

because every particle $\mathcal{A}$ present in the ramp area exerts on the wall the force $f_{\mathcal{A}}$, and similarly for $\mathcal{B}$. We emphasize that this is actually an *osmotic* pressure, in the sense that the ramp potentials $u_1^{\mathcal{A},B}(\mathbf{r}) = f_{\mathcal{A},B}x$ act only on the $\mathcal{A}$ and $\mathcal{B}$ particles while the solvent penetrates everywhere completely freely. This means that our ramp potentials represent an osmotic piston.

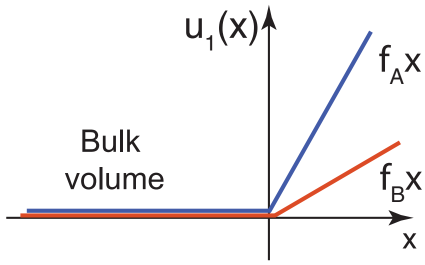

FIG. 6. (Color online) Ramp potential used to calculate pressure.

A similar expression for the osmotic pressure was also used in Ref. [34], where it is derived from the expression of the Helmhotz partition sum, i.e., from equilibrium statistical mechanics. We feel necessary to emphasize that Eq. (E1) is derived on purely mechanical grounds, and it has nothing to do with thermodynamic equilibrium. As such, it is perfectly applicable to our present problem.

To find the steady state concentration profile in the presence of ramp potentials, we slightly generalize the diffusion Eqs. (10) and (C2) by including the external potentials u_1:

$$\mu_{\mathcal{A}} \rightarrow \mu_{\mathcal{A}} + u_1^{\mathcal{A}} \quad \text{and} \quad \mu_{\mathcal{B}} \rightarrow \mu_{\mathcal{B}} + u_1^{\mathcal{B}}. \tag{E2}$$

At steady state, the concentration profile must be such that $\mu + u_1 = \text{const}$ for both the $\mathcal{A}$ and $\mathcal{B}$ components. This can be written as

$$c^{\mathcal{A}}(\mathbf{r}) = C^{\mathcal{A}} e^{-\frac{u_1^{\mathcal{A}}}{T_{\mathcal{A}}}} \left[1 + c^{\mathcal{A}}(\mathbf{r}) B_{\mathcal{A}} + c^{\mathcal{B}}(\mathbf{r}) B_{\mathcal{A}B} \frac{\overline{T}}{T_{\mathcal{A}}} \right], \tag{E3a}$$

$$c^{\mathcal{B}}(\mathbf{r}) = C^{\mathcal{B}} e^{-\frac{u_1^{\mathcal{B}}}{T_{\mathcal{B}}}} \left[1 + c^{\mathcal{B}}(\mathbf{r}) B_{\mathcal{B}} + c^{\mathcal{A}}(\mathbf{r}) B_{\mathcal{A}B} \frac{\overline{T}}{T_{\mathcal{B}}} \right]. \tag{E3b}$$

Here $C^{\mathcal{A}}$ and $C^{\mathcal{B}}$ are normalization factors. To make things simple, we assume that the "ramps" are not too shallow, such that the normalization integral is dominated by the bulk volume V where both ramp potentials vanish. Given that the virial terms in the chemical potentials are the corrections to the ideal gas, we solve iteratively and get

$$\begin{aligned} c^{\mathcal{A}}(\mathbf{r}) = \frac{N_{\mathcal{A}}}{V} e^{-\frac{u_1^{\mathcal{A}}}{T_{\mathcal{A}}}} \bigg[1 &+ \frac{N_{\mathcal{A}}}{V} B_{\mathcal{A}} \big(1 - e^{-\frac{u_1^{\mathcal{A}}}{T_{\mathcal{A}}}}\big) \\ &+ \frac{N_{\mathcal{B}}}{V} B_{\mathcal{A}B} \frac{\overline{T}}{T_{\mathcal{A}}} \big(1 - e^{-\frac{u_1^{\mathcal{B}}}{T_{\mathcal{B}}}}\big) \bigg] \end{aligned} \tag{E4}$$

and similarly for $c^{\mathcal{B}}(\mathbf{r})$. Note that the result does not depend on the ramp forces $f_{\mathcal{A}}$ and $f_{\mathcal{B}}$, which do not have to be identical.

2. Derivation 2

Our starting point of the second derivation is the kinetic expression of the pressure

$$p = p_{\text{ideal}} - \frac{N}{6V} \left\langle \sum_{i \neq j} \mathbf{r}_{ij} \cdot \partial_{\mathbf{r}_{ij}} u(\mathbf{r}_{ij}) \right\rangle. \tag{E5}$$

Sometimes it is called Irving-Kirkwood formula [31,32]. As in the first derivation, the important point is that this equation follows from pure mechanics and does not make any assumption related to equilibrium statistical mechanics. In terms of pair distributions p_2^{ij} the Irving-Kirkwood formula reads

$$\begin{aligned} p = p_{\text{ideal}} &- \frac{N_{\mathcal{A}}^2}{6} \int \mathbf{r}_{12} \cdot \partial_{\mathbf{r}_{12}} u^{\mathcal{AA}}(\mathbf{r}_{12}) p_2^{\mathcal{AA}}(\mathbf{r}_1, \mathbf{r}_2) \\ &- \frac{N_{\mathcal{A}} N_{\mathcal{B}}}{3} \int \mathbf{r}_{12} \cdot \partial_{\mathbf{r}_{12}} u^{\mathcal{AB}}(\mathbf{r}_{12}) p_2^{\mathcal{AB}}(\mathbf{r}_1, \mathbf{r}_2) \\ &- \frac{N_{\mathcal{B}}^2}{6} \int \mathbf{r}_{12} \cdot \partial_{\mathbf{r}_{12}} u^{\mathcal{BB}}(\mathbf{r}_{12}) p_2^{\mathcal{BB}}(\mathbf{r}_1, \mathbf{r}_2). \end{aligned} \tag{E6}$$

Using the ansatz (9, 9) for p_2, and integrating by parts (and remembering that $\nabla \cdot \mathbf{r} = 3$), we obtain the same result as before for the osmotic pressure.

APPENDIX F: POWER TRANSFER

In the main text, we outlined the derivation of the power transfer in the cases of either two particles or many particles with only pairwise collisions. Here we establish a more general result which suggests that the power transfer is expressed in terms of only pair and triple correlation functions (but not higher order ones). Consider the work performed by all forces per unit time on all $\mathcal{A}$ particles, which is also the power received by $\mathcal{A}$ particles:

$$W = \sum_i^{N_{\mathcal{A}}} \int \frac{\partial U}{\partial \mathbf{r}_i^{\mathcal{A}}} \left[\frac{T_{\mathcal{A}}}{\zeta_{\mathcal{A}}} \frac{\partial P}{\partial \mathbf{r}_i^{\mathcal{A}}} + \frac{\partial U}{\partial \mathbf{r}_i^{\mathcal{A}}} \frac{P}{\zeta_{\mathcal{A}}} \right] d\{\mathbf{r}\}, \tag{F1}$$

with $U = U(\{\mathbf{r}_j^{\mathcal{A}}\}, \{\mathbf{r}_k^{\mathcal{B}}\})$ the total potential energy of the system. In the integral (F1), the first factor is the force which acts on particle $\mathcal{A}_i$ due to all other particles, while the second factor (in square brackets) is the current, i.e., the velocity of the particle $\mathcal{A}_i$ multiplied by the probability density P. Thus, the integral (F1) is the average power transfer to one particle $\mathcal{A}_i$. By symmetry, it is independent of i, so that the summation over i reduces to a factor $N_{\mathcal{A}}$. As long as the interaction potentials are pairwise additive (B1), the force is also a sum:

$$\frac{\partial U}{\partial \mathbf{r}_i^{\mathcal{A}}} = \sum_{j \neq i}^{N_{\mathcal{A}}} \frac{\partial u^{\mathcal{AA}}\big(\mathbf{r}_i^{\mathcal{A}} - \mathbf{r}_j^{\mathcal{A}}\big)}{\partial \mathbf{r}_i^{\mathcal{A}}} + \sum_k^{N_{\mathcal{B}}} \frac{\partial u^{\mathcal{AB}}\big(\mathbf{r}_i^{\mathcal{A}} - \mathbf{r}_k^{\mathcal{B}}\big)}{\partial \mathbf{r}_i^{\mathcal{A}}}. \tag{F2}$$

The first term is the force acting on particle $\mathcal{A}_i$ due to other $\mathcal{A}$ particles, by symmetry this term vanishes on average in the sum over $\mathcal{A}$ particles. And the second term, which is the due to $\mathcal{B}$ particles on $\mathcal{A}$ particles, yields

$$\begin{aligned} W = {} & \frac{N_{\mathcal{A}} T_{\mathcal{A}}}{\zeta_{\mathcal{A}}} \int \frac{\partial u^{\mathcal{A}B}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \times \frac{\partial p_2^{\mathcal{A}B}(\mathbf{r}, \mathbf{r}')}{\partial \mathbf{r}} d^3\mathbf{r} d^3\mathbf{r}' \\ & + \frac{N_{\mathcal{A}}}{\zeta_{\mathcal{A}}} \int \left[\frac{\partial u^{\mathcal{A}B}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \right]^2 p_2^{\mathcal{A}B}(\mathbf{r}, \mathbf{r}') d^3\mathbf{r} d^3\mathbf{r}' \\ & + \frac{N_{\mathcal{A}}^2}{\zeta_{\mathcal{A}}} \int \frac{\partial u^{\mathcal{A}B}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \times \frac{\partial u^{\mathcal{AA}}(\mathbf{r} - \mathbf{r}'')}{\partial \mathbf{r}} p_3^{ABA}(\mathbf{r}, \mathbf{r}', \mathbf{r}'') \\ & \times d^3\mathbf{r} d^3\mathbf{r}' d^3\mathbf{r}'' \\ & + \frac{N_{\mathcal{A}} N_{\mathcal{B}}}{\zeta_{\mathcal{A}}} \int \frac{\partial u^{\mathcal{A}B}(\mathbf{r} - \mathbf{r}')}{\partial \mathbf{r}} \times \frac{\partial u^{\mathcal{A}B}(\mathbf{r} - \mathbf{r}'')}{\partial \mathbf{r}} p_3^{ABB} \\ & \times (\mathbf{r}, \mathbf{r}', \mathbf{r}'') d^3\mathbf{r} d^3\mathbf{r}' d^3\mathbf{r}''. \end{aligned} \tag{F3}$$

Neglecting the three-body collisions (terms with p_3), and using the known expression (9) for p_2, we return to the result given in the main text.

Here, we emphasize once again that, as long as interaction potentials are pairwise additive, as in Eq. (B1), Eq. (F3) is exact.

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