

## Universal Relation between Entropy and Kinetics

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Relating thermodynamic and kinetic properties is a conceptual challenge with many practical benefits. Here, based on first principles, we derive a rigorous inequality relating the entropy and the dynamic propagator of particle configurations. It is universal and applicable to steady states arbitrarily far from thermodynamic equilibrium. Applying the general relation to diffusive dynamics yields a relation between the entropy and the (normal or anomalous) diffusion coefficient. The relation can be used to obtain useful bounds for the late-time diffusion coefficient from the calculated steady-state entropy or, conversely, to estimate the entropy based on measured diffusion coefficients. We demonstrate the validity and usefulness of the relation through several examples and discuss its broad range of applications, in particular, for systems far from equilibrium.

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Materials are characterized, on the one hand, by static thermodynamic properties (heat capacity, compressibility) and, on the other hand, by kinetic properties (viscosity, conductivity, diffusion coefficient). Clearly, the dynamics of a system dictates its steady state; however, the same steady state may arise from many different dynamics. Indeed, one of the remarkable feats of statistical mechanics is the ability to relate thermodynamic properties to kinetic ones in certain limits. Well-known examples of such relations arise from linear response theory close to equilibrium, for example, the Onsager relations, fluctuation-dissipation theorems (including the Einstein relation), and Green-Kubo relations [1]. Far from equilibrium, generalizations include fluctuation theorems [2–4] and generalized Green-Kubo relations [5]. Particularly remarkable are relations between kinetic coefficients and quantities such as temperature and entropy which are obtainable from one-time independent configurations (e.g., the Einstein relation). Other relations, such as the Green-Kubo ones, require two-time statistics.

In the present Letter, we derive, based on first principles, a rigorous general relation between a one-time steady-state variable, the entropy, and the dynamic propagator of particle configurations. This relation holds arbitrarily far from equilibrium. When applied to single-particle diffusion, it connects the entropy with the late-time diffusion coefficient. The relation, in its most generality, is an inequality. It becomes an equality under specified assumptions. Such a relation would be very useful, because the abilities to calculate or measure thermodynamic and kinetic properties may differ substantially.

Several general relations between entropy and kinetic properties were proposed over the years based on the equilibrium theory of fluids. The Adam-Gibbs relation between the entropy of a glass-forming liquid and its

relaxation time [6] has inspired later theories of the glass transition [7,8]. Rosenfeld [9,10] and later, independently, Dzugutov [11], proposed a phenomenological relation between the entropy per particle of a fluid  $s$  and the single-particle diffusion coefficient  $D$ . It reads  $D/(vl) = A \exp[b(s - s^{\text{id}})]$ , where  $l \sim \rho^{-1/3}$  is the mean interparticle distance ( $\rho$  being the mean density),  $v \sim T^{1/2}$  is the thermal velocity ( $T$  being the temperature),  $s^{\text{id}}$  is the entropy per particle of the ideal gas, and  $A$  and  $b$  are system-dependent phenomenological parameters to be found *ad hoc* by experiment or simulation. Similar entropy-diffusion relations were obtained rigorously for the specific case of a single particle at equilibrium, diffusing in a random external potential [12,13]. The relation derived below is not restricted to equilibrium and applies also to the correlated motion of many particles.

The phenomenological entropy-diffusion relation has been tested extensively in the last two decades against experiments and simulations. It has been used also in industrial applications to indirectly infer transport properties. For a recent review and literature survey, see Ref. [14]. The myriad of systems to which the relation has been applied ranges from simple liquids (e.g., Refs. [15–17]), through active particles [18], to planetary cores [19]. The success of the phenomenological entropy-diffusion relation has been inconsistent. Its physical origin, and thus the ability to account for its successes and failures, has not been resolved. A theory based on hidden scale invariance originating in the microscopic interactions has been proposed to account for the scaling with density and temperature [14,20,21]. Overall, the widespread use of the entropy-diffusion relation, even if it is empirical and inaccurate, attests to the far-reaching importance of relating thermodynamic and transport properties.

*Derivation outline.*—We consider a material at steady state, consisting of  $N$  identical particles whose microscopic configurations are given by  $\mathbf{X} = \{\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N\}$ . The state  $\mathbf{x}$  of each particle may include its position, orientation, velocity, etc. The many-particle configuration  $\mathbf{X}$  changes with time  $t$  according to the case-specific microscopic laws of motion, defining a trajectory  $\mathbf{X}(t)$  between  $t = 0$  and  $t = t_{\text{obs}}$ , the observation time. Our main assumption, valid for the vast majority of materials, is that the material has a finite relaxation time  $\tau$ . For times  $t > \tau$ , the configurations of the particles, which are regarded as indistinguishable, are mixed, and the material reaches steady state.

At the core of the theory is the distinction between two equivalent perspectives of the dynamics, regarding the particles as either indistinguishable or distinguishable (identifiable); see Fig. 1. We utilize the two perspectives to relate the steady-state behavior with the kinetic one. We discretize the trajectory  $\mathbf{X}(t)$  into  $K + 1$  consecutive “snapshots” separated by time intervals  $\tau$ ,  $\mathbf{X}^k = \mathbf{X}(k\tau)$ ,  $k = 0, 1, \dots, K = t_{\text{obs}}/\tau$ . We denote the probability to obtain a certain sequence of configurations by  $\tilde{\text{Pr}}[\{\mathbf{X}^k\}]$  for indistinguishable particles and by  $\text{Pr}[\{\mathbf{X}^k\}]$  for identifiable ones.

From the first perspective (top row of Fig. 1), we consider configurations of the  $N$  indistinguishable particles. Over time intervals larger than  $\tau$ , sufficiently many randomizing events (e.g., collisions) occur to mix the particles. Hence, the configurations at different instances along the discretized trajectory may be taken as independent,

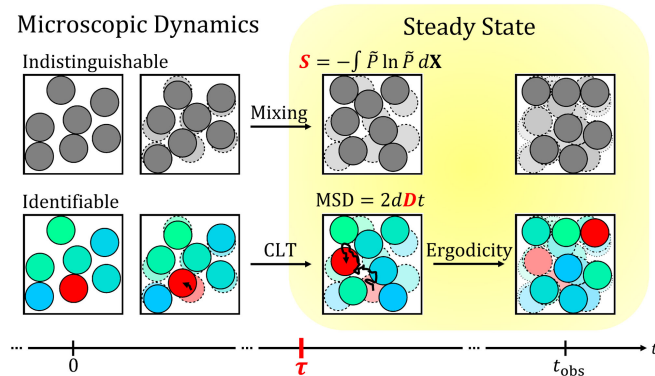


FIG. 1. Derivation outline. We assume the existence of a finite relaxation time  $\tau$ . For  $t \ll \tau$  (left) the particles follow microscopic dynamics (e.g., moving ballistically). For  $t > \tau$  (right) the system reaches a steady state. Two equivalent descriptions are considered. In the first (top row), particles are treated as indistinguishable. Their  $\tau$ -separated configurations  $\mathbf{X}$  are independent and drawn from the steady-state distribution  $\tilde{P}(\mathbf{X})$ , with entropy  $S$ . In the second description (bottom row), particles are treated as distinguishable (identifiable). Over time  $t > \tau$  each particle experiences many collisions, which (by the central limit theorem, CLT) leads to an effective diffusion with mean-squared displacement (MSD)  $2dDt$ . Comparing the two statistics leads to a relation between the steady-state entropy (top right) and diffusion (bottom right).

drawn each from the same steady-state probability distribution for indistinguishable particles  $\tilde{P}(\mathbf{X})$ . This implies the decomposition of  $\tilde{\text{Pr}}[\{\mathbf{X}^k\}]$  into a product of independent probabilities,  $\tilde{\text{Pr}}[\{\mathbf{X}^k\}] = \tilde{P}(\mathbf{X}^0)\tilde{P}(\mathbf{X}^1)\dots\tilde{P}(\mathbf{X}^K)$ . The steady-state entropy is the information content of  $\tilde{P}(\mathbf{X})$  [22],

$$S = - \int d\mathbf{X} \tilde{P}(\mathbf{X}) \ln \tilde{P}(\mathbf{X}), \quad (1)$$

up to a constant that fixes units (see Supplemental Material [23]).

From the second perspective (bottom row of Fig. 1), we treat particles as identifiable and follow their individual dynamics. Their configurations, separated by time intervals  $\tau$ , are typically correlated, as each particle has traversed a negligible part of the total available space. We assume that the identifiable configurations follow a stationary Markov process and denote the Markovian probability to change from configuration  $\mathbf{X}^{k-1}$  to configuration  $\mathbf{X}^k$  during time  $\tau$  (the propagator) as  $W_\tau(\mathbf{X}^k|\mathbf{X}^{k-1})$ . This enables another decomposition of the trajectory probability, this time for identifiable particles,  $\text{Pr}[\{\mathbf{X}^k\}] = P(\mathbf{X}^0)W_\tau(\mathbf{X}^1|\mathbf{X}^0)\dots W_\tau(\mathbf{X}^K|\mathbf{X}^{K-1})$ , where  $P(\mathbf{X}) = \tilde{P}(\mathbf{X})/N!$  is the steady-state distribution of configurations for identifiable particles [28].

Thus, we have decomposed the indistinguishable trajectory distribution  $\tilde{\text{Pr}}[\{\mathbf{X}^k\}]$  into steady-state distributions  $\tilde{P}(\mathbf{X}^k)$  and the identifiable trajectory distribution  $\text{Pr}[\{\mathbf{X}^k\}]$  into kinetic distributions  $W_\tau(\mathbf{X}^k|\mathbf{X}^{k-1})$ . To relate these two results to entropy, we define the following integral over all possible discretized trajectories [29]:

$$H = -\frac{1}{K} \int d\{\mathbf{X}^k\} \text{Pr}[\{\mathbf{X}^k\}] \ln(\text{Pr}[\{\mathbf{X}^k\}]). \quad (2)$$

When the decomposition of  $\text{Pr}$  is substituted in Eq. (2), the integral becomes  $H = (S + \ln N!)/K - \int d\mathbf{X}^0 P(\mathbf{X}^0) \int d\mathbf{X} W_\tau(\mathbf{X}|\mathbf{X}^0) \ln W_\tau(\mathbf{X}|\mathbf{X}^0)$  [23].

The main complication lies in the relation between the two trajectory distributions  $\text{Pr}$  and  $\tilde{\text{Pr}}$ . Indistinguishable trajectories correspond to many possible permutations of identifiable ones; yet, not all permutations are equally likely. For example, the probability that particles that start far apart will switch positions during time  $\tau$  is negligibly small, whereas colliding particles may permute with high probability.

First, we consider an extreme case, which sets a strict lower bound on  $H$ , ignoring all the possible particle-identity exchanges (except in the initial condition). With this “undercounting,” we get  $\tilde{\text{Pr}} \geq N! \text{Pr}$ . When the decomposition of  $\tilde{\text{Pr}}$  is substituted in Eq. (2), this inequality gives  $H \geq (S + \ln N!)/K + S$ . Comparing the two expressions for  $H$  arising from the two decompositions, we get

$$S \leq - \int d\mathbf{X}^0 P(\mathbf{X}^0) \int d\mathbf{X} W_\tau(\mathbf{X}|\mathbf{X}^0) \ln W_\tau(\mathbf{X}|\mathbf{X}^0). \quad (3)$$

Equation (3) is our first universal relation between the steady-state entropy and a kinetic property, the many-particle propagator. It relies only on the existence of a finite relaxation time and the Markov property. It can be rationalized as follows. During the finite relaxation time  $\tau$ , the identifiable particles cover a small (intensive) region of their phase space (right-hand side). Combining the regions covered by all particles, without distinguishing among them, is related to the available microstates in the indistinguishable picture (left-hand side). Loosely speaking, the right-hand side overcounts the overlap between regions explored by different particles, which leads to the inequality.

One is typically interested in the transport properties of a subset of particles much smaller than  $N$ , in particular, the effective diffusion coefficient of a single particle. If the particles are independent and the system is translation invariant, the propagator can be decomposed into single-particle ones,  $W_\tau(\mathbf{X}|\mathbf{X}^0) = w_\tau(\Delta\mathbf{x}_1)w_\tau(\Delta\mathbf{x}_2)\cdots w_\tau(\Delta\mathbf{x}_N)$ , where  $\Delta\mathbf{x}_j = \mathbf{x}_j - \mathbf{x}_j^0$ , leading to  $H = (S + \ln N!)/K - N \int d\Delta\mathbf{x} w_\tau(\Delta\mathbf{x}) \ln w_\tau(\Delta\mathbf{x})$ . If the particles are dependent, this relation sets another bound on  $H$  [23,33]. Overall, owing to the subextensive property of the entropy [23], Eq. (3) becomes a bound on the entropy per particle,  $s = S/N$ ,

$$s \leq - \int d\Delta\mathbf{x} w_\tau(\Delta\mathbf{x}) \ln w_\tau(\Delta\mathbf{x}). \quad (4)$$

Compared to Eq. (3), the inequality of Eq. (4) is less tight but more practical, as the single-particle propagator is more accessible in experiments, simulations, and coarse-grained theories.

In most materials, due to the central limit theorem, the single-particle propagator converges to a Gaussian over intervals  $\Delta t > \tau$ ,  $w_{\Delta t}(\Delta\mathbf{x}) = (4\pi D\Delta t)^{-d/2} e^{-|\Delta\mathbf{x}|^2/(4D\Delta t)}$ , where  $D$  is the diffusion coefficient and  $d$  is the dimensionality. Substitution into Eq. (4) gives

$$D\tau \geq (D\tau)^\circ \exp[(2/d)(s - s^\circ)], \quad (5a)$$

where the superscript  $^\circ$  denotes values for a reference system with a tighter bound. This is an exact and practically useful relation.

To improve the bound, we need to evaluate the contribution of the neglected permutations when relating indistinguishable and identifiable trajectories. Taking a mean-field approach, we assume that during each time step  $\tau$  every particle can be exchanged with  $z$  nearby particles. Hence,  $\tilde{\text{Pr}} \simeq N! z^{NK} \text{Pr}$ . As a result, Eq. (3) is replaced by  $S + N \ln z \leq - \int d\mathbf{X} W_\tau(\mathbf{X}|\mathbf{X}_0) \ln W_\tau(\mathbf{X}|\mathbf{X}_0)$ . Estimating  $z$  requires kinetic considerations. Each particle has a ‘‘sphere of influence’’ of radius  $\sim (D_s\tau)^{1/2}$ , where  $D_s$  is a short-time diffusion coefficient [34]. Therefore,  $z \sim \rho (D_s\tau)^{d/2}$ . This leads to [23]

$$D \geq D_s \exp[(2/d)(s - s^{\text{id}})], \quad (5b)$$

where  $s^{\text{id}} = -\ln \rho$  is the ideal-gas entropy. Equation (5b) is not exact, but it gives an improved bound under the mean-field assumption.

Equation (4) could be applied to any single-particle propagator  $w_\tau(\Delta\mathbf{x})$ . For example, consider anomalous diffusion where the particle’s mean-squared displacement (MSD) is equal to  $dF\Delta t^\alpha$ , with a generalized diffusion coefficient  $F$  [35]. A modified derivation [23] leads to the generalized relation [36]

$$F\tau^\alpha \geq (F\tau^\alpha)^\circ \exp[(2/d)(s - s^\circ)]. \quad (6)$$

Both inequalities (5) and (6) become equalities when trajectories do not mix and the dynamics is accurately represented by single-particle (generalized) diffusion.

Unlike the earlier phenomenological relations [9,11], (a) Eqs. (5) are *inequalities* in general; (b) they are not restricted to thermodynamic equilibrium; (c) the coefficient  $b$  has an explicit value,  $b = 2/d$ ; and (d) there is an explicit dependence on the relaxation time  $\tau$ . These differences explain the discrepancies between the phenomenological relations and observations. Departure from equality may be caused by strong correlations among particles [37,38] or extra dependencies of  $\tau$  and  $D_s$  on  $\rho$  and  $T$ , which deviate from  $D_s \sim v l \sim \rho^{-1/d} T^{1/2}$  as arising from simple kinetic theory. The latter effect is seen in Fig. 2(b) and also in Fig. S1, Supplemental Material [23].

We now demonstrate the validity and usefulness of Eqs. (5) and (6) in several examples.

*Previously tested examples.*—Quite a few empirical studies found values of  $b$  close to or bounded by  $2/d$ , as predicted by Eqs. (5). Simulations gave  $b = 0.65$  for a 3D hard-sphere gas [9],  $b = 0.67$ – $0.70$  for liquid metals [19,39], and  $b = 0.70$ – $0.73$  and  $0.65$  for water and methanol, respectively [40]. Experiments on colloidal monolayers ( $d = 2$ ) gave values bounded from above by  $b = 1$  [41–44] and at least twice as high  $b$  for the rotational diffusion coefficient ( $d = 1$ ) [43]. Still, recent extensive molecular dynamics simulations of Lennard-Jones fluids in 3D gave  $b = 0.751$  [45]. This deviation from  $b = 2/3$  can be attributed to the above-mentioned dependencies on density and temperature.

*Example: Homogenized diffusion.*—Under a periodic or random potential, the motion of a diffusing particle, after traversing sufficiently long distances, is accurately described by an effective (‘‘homogenized’’) diffusion [46–48]. Calculations of the homogenized  $D$  even for the simplest scenarios require elaborate mathematical analyses [12,13,47,48]. In certain cases, Eqs. (5) offer a far simpler alternative. Indeed, for a particle in a random potential, the analysis in Ref. [12] arrives at a relation that coincides with Eq. (5b) in the limit of a single particle at equilibrium. For a particle in a 1D periodic potential, the known analytical result for the homogenized  $D$  [46–48] agrees with Eq. (5a),

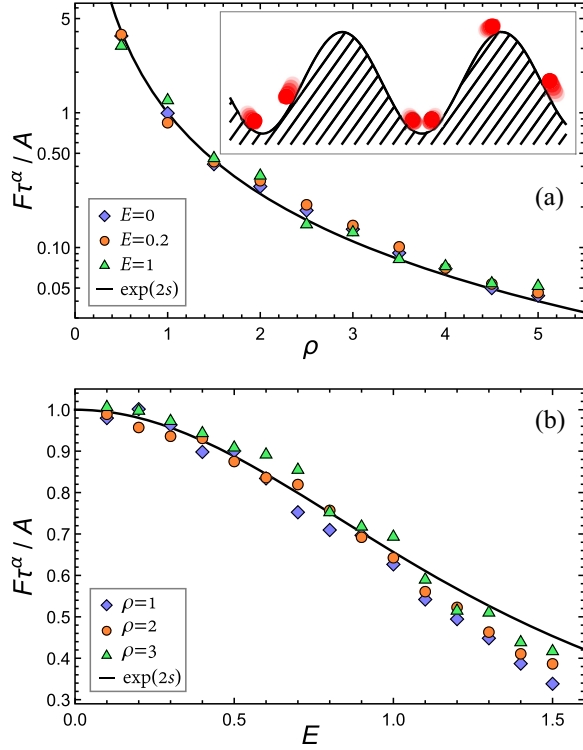


FIG. 2. Single-file diffusion in a periodic potential. (a) Anomalous MSD as a function of density for fixed potential strengths as indicated. The MSD is scaled by the predicted energy-dependent factor to make the data collapse. (b) MSD versus potential strength for fixed densities as indicated. The MSD is scaled by the predicted density-dependent factor to make the data collapse. Symbols show results of Langevin dynamics simulations and solid lines show the theoretical predictions obtained from entropy. Inset: Illustration of the simulated system.

where the inequality tends to an equality for weak to moderate potentials [23].

*Example: Anomalous diffusion in a single file.*—Single-file diffusion is relevant to a large variety of transport processes [49]. The model consists of  $N$  rigid particles of diameter  $a$ , restricted to move along a line of length  $L$  without bypassing each other. An isolated particle would perform normal Brownian motion with diffusion coefficient  $D_s$ . However, when put in a single file with others, its motion becomes subdiffusive, with the exact result [50]  $\text{MSD}(t) = Ft^{1/2}$ ,  $F = 2a(D_s/\pi)^{1/2}(1-\phi)/\phi$ , where  $\phi = Na/L$  is the line fraction occupied by the particles. This  $\phi$  dependence can be easily reproduced using Eq. (6). The single-file constraint prevents mixing of particle trajectories (i.e.,  $z = 1$  exactly), suggesting that the bound in Eq. (6) should be tight. The total entropy per particle is  $s = \ln[(1-\phi)/\phi]$ , and  $\tau$  is proportional to the time between encounters of neighboring particles,  $\tau \sim l^2/D_s$ , where  $l = a(\phi^{-1} - 1)$  is the mean distance between neighbors. Substituting these into Eq. (6) (with  $d = 1$ ), we obtain  $F \sim aD_s^{1/2}(1-\phi)/\phi$ . Equation (6) turns out to be an equality here, although the particles' motions are correlated.

*Example: Single-file diffusion in a periodic potential.*—To our best knowledge, there is no analytical result for the homogenized anomalous diffusion coefficient in this case [51,52]. Here, pointlike particles ( $a \rightarrow 0$ ) perform random walks without bypassing each other, under a potential with amplitude  $E$  (in units of  $k_B T$ ) and periodicity  $\lambda$ ,  $U(x) = E[1 - \cos(2\pi x/\lambda)]$ . This describes, for example, single-file diffusion occurring over a periodic substrate. The entropy per particle is  $s = -\ln \rho + f(E)$ , where  $f(E)$  is given in the Supplemental Material [23]. Using Eq. (6), we predict that  $F\tau^{1/2} = Ae^{2f(E)}/\rho^2$ , where  $A$  is a constant. To verify this prediction, we performed Langevin dynamics simulations with periodic boundary conditions for various particle densities  $\rho$  (number of particles per  $\lambda$ ) and potential strengths  $E$ . We measured the MSD and extracted the generalized diffusion coefficient  $F$ . Obtaining  $\tau$  is intricate in this example because of its dependence on both  $\rho$  and  $E$ . We assess it numerically from the crossover time between normal and anomalous diffusion [23]. Figure 2(a) shows  $F\tau^\alpha$  as a function of  $\rho$  for fixed  $E = 0, 0.2, 1$ . The prediction  $F\tau^\alpha \sim \rho^{-2}$  with  $\alpha = 1/2$  is verified. In Fig. 2(b), we plot  $F\tau^\alpha$  as a function of  $E$  for fixed  $\rho = 1, 2, 3$ . The prediction works well for small  $E$  but gets further from the numerical results as the potential strength is increased. This is due to an underestimation of  $\tau$  for large  $E$  [23]. Alternatively, one can use these results together with the analytical expression to infer  $\tau(\rho, E)$ .

*Example: Momentum entropy.*—For momenta, the decomposition into single-particle dynamics is exact. When the momentum propagator is used in Eq. (4),  $D\tau$  appearing in Eq. (5) is replaced by the mean-squared momentum. The latter is proportional to the mean kinetic energy, which in turn is proportional to  $T$ . Equation (5a) for  $d = 3$  then gives  $e^{2s/3} \sim T$ , in line with the classical result,  $s = \ln T^{3/2} + \text{const}$ .

*Summary.*—Equation (3) shows a rigorous generic mark left by the steady-state entropy of materials on their dynamics. It offers a theoretical framework for relating static and kinetic properties arbitrarily far from equilibrium, e.g., for fluids in steady flow or active matter. The only assumptions are the existence of a finite relaxation time and Markovian dynamics at late times. Thus, remarkably, quantitative predictions concerning kinetic coefficients could be obtained without a kinetic theory.

Equation (4) makes the general framework more applicable as it relates the entropy with the accessible dynamics of individual particles. We have used it to obtain relations between the entropy and the (generalized) late-time diffusion, Eqs. (5) and (6). With additional knowledge of the material's relaxation time [Eqs. (5a) and (6)] or short-time diffusion coefficient [Eq. (5b)], the relations can be used to obtain useful bounds for the late-time diffusion coefficient from entropy, and vice versa, as demonstrated in the examples above [53]. In essence, these relations imply that  $D\tau$ ,  $F\tau^\alpha$ , or  $D/D_s$  are actually static configurational properties. This is reminiscent of the Einstein relation

$D\eta \sim T$ ,  $\eta$  being a friction coefficient. The Einstein relation can be reproduced by a similar argument to the one used here, comparing the equilibrium entropy of the particle's fast-relaxing velocity,  $s = \ln T^{d/2} + \text{const}$ , with the information content of the resulting diffusive trajectories. The inequalities derived here, however, are much more general and hold arbitrarily far from equilibrium.

The examples given above highlight the conditions under which the inequality approaches an equality, namely, weak mixing of particle trajectories and weakly correlated dynamics. We expect the bound not to be as tight in more strongly correlated or slowly relaxing systems. The violation of the rigorous bound derived here might serve as a useful indicator of anomalous behaviors such as ergodicity breaking and aging [43].

The range of possible applications of Eqs. (5) and (6) is vast. The case of diffusion under an external potential analyzed above clearly shows how the relations could be used to infer binding parameters from measured diffusion. From the opposite direction, entropy is often fairly easy to find from equilibrium thermodynamics or estimate in nonequilibrium steady states [54–59]. This can be used to infer diffusion coefficients in complex scenarios, in particular, in active matter.

We conclude by highlighting the more general inequality relating the entropy with the dynamic propagator, Eq. (3), which enables various extensions of the theory beyond single-particle diffusion. It can be used to treat the correlated diffusion of two or more particles. Introducing weak interactions into the propagator  $W_\tau$  will improve the bound in Eq. (4). One can substitute nondiffusive propagators in Eq. (4) to study, for example, memory effects. It will be particularly important to derive relations for kinetic coefficients other than the diffusion coefficient, such as the heat conductivity and viscosity. The theoretical framework constructed here, utilizing the distinction between the fast relaxation of indistinguishable particle configurations and the coarse-grained dynamics of identifiable particles, will hopefully be instrumental in the progress toward a consistent general theory of nonequilibrium thermodynamics.

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