Microcanonical analysis of a finite-size nonequilibrium system

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Microcanonical analysis is a powerful method that can be used to generalize the concept of phase transitions to finite-size systems. However, microcanonical analysis has only been applied to equilibrium systems. I show that it is possible to conduct the microcanonical analysis of a finite-size nonequilibrium system by generalizing the concept of microcanonical entropy. A one-dimensional asymmetric diffusion process is studied as an example for which such a generalized entropy can be explicitly found, and the microcanonical method is used to define a generalized phase transition for the finite-size nonequilibrium system.

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I. INTRODUCTION

Microcanonical analysis is a powerful method that can be used to generalize the concept of phase transitions to finite-size systems [1,2]. In this approach, the form of microcanonical entropy is examined to determine whether there is a convex region. The existence of such a region signals the onset of inhomogeneity, and the system is considered to undergo a first-order-like transition in this region. Such a generalized notion of phase transition stands in contrast to the phase transition in traditional canonical ensemble approaches in that the latter is defined only in terms of singularities of physical quantities in the infinite-size limit. Microcanonical analysis has been applied to the study of generalized phase transitions of various finite-size systems, such as spin models [3–6], atomic clusters and nuclei [2,7], polymers [8–16], peptides [17], and proteins [18–22].

Because the probability distribution of energy is proportional to the exponential of microcanonical entropy only in an equilibrium system, the microcanonical method has thus far been exclusively used to analyze equilibrium systems. In this work I show that it is possible to apply this method to analyze generalized phase transitions in a finite-size nonequilibrium system [23–30] through the proper generalization of the concept of microcanonical entropy.

Let us first briefly review the connection between the convex region of microcanonical entropy and generalized phase transition [1,2]. We consider a finite closed system with a conserved quantity, say energy E, and denote the number of corresponding microstates as $\Omega_L(E)$, where the subscript denotes the dependence on the system size L. Microcanonical entropy is then defined as

$$S_L(E) = \ln \Omega_L(E), \tag{1}$$

where we use the unit with $k_B = 1$. Now, suppose that we construct a larger system by assembling two identical subsystems of energy E and size L. We let the two subsystems make thermal contact, but we also let the coupling between the two subsystems be weak enough so that the total energy is $E_{\rm tot} = 2E = E_A + E_B$, where E_A and E_B are the energy values of the two subsystems. We then determine the most probable distribution of the energy values of the subsystems, (E_A, E_B), under the constraint $E_A + E_B = 2E$. Because $P(E_A, E_B) \propto \exp[S_L(E_A) + S_L(E_B)]$, it is sufficient to find the value of E_A that maximizes $S_L(E_A) + S_L(2E - E_A)$ with respect to E_A.

If $S_L(E)$ is a concave function, then by definition

$$S_L[pE_A + (1-p)E_B] > pS_L(E_A) + (1-p)S_L(E_B) \tag{2}$$

for any values of E_A, E_B, and p such that $E_A \neq E_B$ and $0 < p < 1$. Substituting $p = 1/2$ and $E_B = 2E - E_A$, we find that $2S_L(E) > S_L(E_A) + S_L(2E - E_A)$, leading to $P(E,E) > P(E_A, 2E - E_A)$ for any value of E satisfying $E_A \neq E$. Therefore, the homogeneous distribution of energy among the subsystems is preferred.

On the other hand, if there is a convex region in $S_L(E)$ and E is in that region, then one can find $E_A \neq E$ such that $2S_L(E) < S_L(E_A) + S_L(E - E_A)$. In this case, an inhomogeneous distribution is favored over the homogeneous distribution, and we say that the system is in the phase coexistence region. In fact, if the total system is assembled from a large number of subsystems with convex microcanonical entropy, the sizes of the droplets of high-energy and low-energy phases increase and decrease, respectively, with increasing energy, and the system can be considered to undergo first-order-like transition. The argument can be easily generalized to the case of subsystems of different sizes and multiple conserved quantities [1,2].

We note that the only relevant property of $S_L(E)$ exploited in the argument is that when a conserved quantity $\mathbf{Q} = \mathbf{Q_A} + \mathbf{Q_B}$ of the total system is distributed over two subsystems A and B, the probability distribution of $\mathbf{Q_A}$ and $\mathbf{Q_B}$ is proportional to $\exp[S_L(\mathbf{Q_A}) + S_{L'}(\mathbf{Q_B})]$, where L and L' denote the sizes of the subsystems A and B. In terms of Ω, the relation can be written as

$$P(\mathbf{Q_A}, \mathbf{Q_B}) \propto \Omega_L(\mathbf{Q_A})\Omega_{L'}(\mathbf{Q_B}). \tag{3}$$

We can also impose a certain boundary condition at the interface between the subsystems. For example, there can be a free-energy cost for creating a phase boundary. In this case, $P(\mathbf{Q_A}, \mathbf{Q_B})$ becomes a *conditional* probability which can be used to analyze whether the system prefers inhomogeneous or homogeneous distribution of the conserved quantity $\mathbf{Q}$ with respect to the boundary, under the given condition. Again, $S_L(E)$ in Eq. (3) can be used to define the generalized phase transition of the finite-size system.

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Therefore, it is clear that even for a nonequilibrium system, if the probability of the distribution of a conserved quantity $\mathbf{Q}$ among subsystems under appropriate boundary conditions can be expressed in the form Eq. (3), then we can consider $\Omega_L(\mathbf{Q})$ as the generalized density and

$$S_L(\mathbf{Q}) \equiv \log \Omega_L(\mathbf{Q}) \tag{4}$$

as the generalized entropy, which can then be used as the target of the microcanonical analysis. Below, I will consider an asymmetric diffusion model on a periodic lattice as an example of a nonequilibrium model for which the microcanonical analysis can be performed, and show that the concept of phase transition can be generalized to a *finite-size* nonequilibrium system.

II. THE GENERALIZED MICROCANONICAL ENTROPY FOR AN ASYMMETRIC DIFFUSION MODEL

In this work, I consider a diffusion model where two types of particles, labeled 1 and 2, move asymmetrically on a periodic lattice of length L [24,28–32]. Treating the vacancy as a particle with label 0, the transition rates $g_{\alpha\beta}$ for the particle exchanges of the type $(\alpha,\beta) \to (\beta,\alpha)$ at neighboring sites are given as [28–32]

$$g_{10} = g_{02} = 1,\ g_{12} = q,\ g_{21} = 1, \tag{5}$$

with all other components of gs being zero. We note that the numbers of both types of particles are separately conserved, which we will denote as n_1 and n_2. The matrix representation of the stationary state for this process has already been found and is given as [28–32]

$$P_{st}(\beta_1,\dots\beta_L) \propto \mathrm{tr}\mathbf{G}_{\beta_1}\cdots\mathbf{G}_{\beta_L}, \tag{6}$$

where β_k denotes the particle type at the kth site, and the components of the three infinite-dimensional matrices $\mathbf{G}_\beta$ $(\beta = 0,1,2)$ are given as

$$(G_0)_{ij} = \delta_{1i}\delta_{1j}, \quad (G_1)_{ij} = a_i\delta_{ij} + t_i\delta_{ij-1},$$
$$(G_2)_{ij} = a_i\delta_{ij} + s_j\delta_{i-1j}, \tag{7}$$

where

$$a_k = \frac{1+q^{2-k}-2q^{1-k}}{q-1},$$
$$s_k t_k = \frac{1-q^{-k}}{(q-1)^2}(1-q^{3-k}+4q^{2-k}-4q^{1-k}). \tag{8}$$

Now let us suppose that there are vacancies at sites a and b. The entire periodic lattice can be divided into two regions bounded by these two sites, and we would like to obtain the conditional probability for the particles in these two regions being $\mathbf{n}_A = (n_1^{(A)},n_2^{(A)})$ and $\mathbf{n}_B = (n_1^{(B)},n_2^{(B)})$. Obviously, we see from Eq. (6) that it is proportional to

$$P(\mathbf{n}_A,\mathbf{n}_B) \propto \sum_{\gamma_1,\cdots,\gamma_L} \mathrm{tr}(\mathbf{G}_{\gamma_1}\cdots\mathbf{G}_{\gamma_{a-1}}\mathbf{G}_0\mathbf{G}_{\gamma_{a+1}}\cdots\mathbf{G}_{\gamma_{b-1}}$$
$$\times\mathbf{G}_0\mathbf{G}_{\gamma_{b+1}}\cdots\mathbf{G}_{\gamma_L})\delta\left(\sum_{i\in A}\delta_{\gamma_i,1},n_1^{(A)}\right)$$
$$\times\delta\left(\sum_{j\in A}\delta_{\gamma_j,2},n_2^{(A)}\right)\delta\left(\sum_{k\in B}\delta_{\gamma_k,1},n_1\right)^{(B)}$$
$$\times\delta\left(\sum_{l\in B}\delta_{\gamma_l,2},n_2^{(B)}\right), \tag{9}$$

where $\delta(a,b) = \delta_{a,b}$ denotes the Kronecker δ function that vanishes when the indices are not equal. Note that

$$\begin{aligned}
&\mathrm{tr}(\mathbf{G}_{\gamma_1}\cdots\mathbf{G}_{\gamma_{a-1}}\mathbf{G}_0\mathbf{G}_{\gamma_{a+1}}\cdots\mathbf{G}_{\gamma_{b-1}}\mathbf{G}_0\mathbf{G}_{\gamma_{b+1}}\cdots\mathbf{G}_{\gamma_L})\\
&= \mathrm{tr}(\mathbf{G}_0\mathbf{G}_{\gamma_{a+1}}\cdots\mathbf{G}_{\gamma_{b-1}}\mathbf{G}_0\mathbf{G}_{\gamma_{b+1}}\cdots\mathbf{G}_{\gamma_{a-1}})\\
&= [\mathbf{G}_{\gamma_{a+1}}\cdots\mathbf{G}_{\gamma_{b-1}}]_{11}[\mathbf{G}_{\gamma_{b+1}}\cdots\mathbf{G}_{\gamma_{a-1}}]_{11}\\
&= \mathrm{tr}(\mathbf{G}_0\mathbf{G}_{\gamma_{a+1}}\cdots\mathbf{G}_{\gamma_{b-1}})\mathrm{tr}(\mathbf{G}_0\mathbf{G}_{\gamma_{b+1}}\cdots\mathbf{G}_{\gamma_{a-1}}).
\end{aligned} \tag{10}$$

Therefore, the conditional probability for the steady state is expressed in the form of Eq. (3), where the generalized density for a system of size L is now defined as

$$\begin{aligned}
\Omega_L(\mathbf{n}) &= \sum_{\gamma_1,\cdots,\gamma_{L-1}} \mathrm{tr}\left(\mathbf{G}_0\prod_{k=1}^{L-1}\mathbf{G}_{\gamma_k}\right)\delta\left(\sum_{i=1}^{L-1}\delta_{\gamma_i,1},n_1\right)\delta\left(\sum_{j=1}^{L-1}\delta_{\gamma_j,2},n_2\right)\\
&= \sum_{\gamma_1,\cdots,\gamma_{L-1}} \left[\prod_{k=1}^{L-1}\mathbf{G}_{\gamma_k}\right]_{11}\delta\left(\sum_{i=1}^{L-1}\delta_{\gamma_i,1},n_1\right)\delta\left(\sum_{j=1}^{L-1}\delta_{\gamma_j,2},n_2\right),
\end{aligned} \tag{11}$$

where the system size L includes one vacancy. We analyze the generalized phase transition of the current model by performing the microcanonical analysis on the generalized entropy $S_L(\mathbf{n}) = \log \Omega_L(\mathbf{n})$. It is expressed in terms of $(L/2) \times (L/2)$ submatrices of $\mathbf{G}_\beta$, which can be computed exactly for given values of q and L [28–32].

Analytic computations, Monte Carlo simulations, mean field calculations [28,29], and a partition function zero analysis [30] have been carried out to argue that this system undergoes a nonequilibrium phase transition in the limit of $L \to \infty$. There is a $q_c > 1$ such that the system remains homogeneous for $q \geqslant q_c$, but inhomogeneities of the particle densities appear for a certain range of particle numbers when $q < q_c$. In fact, the latter can be considered as a region of the first-order-like transition between fluid and condensed phases, as will be elaborated below.

III. THE GENERALIZED PHASE TRANSITION FOR THE FINITE-SIZE NONEQUILIBRIUM SYSTEM

From the viewpoint of microcanonical analysis, the criterion for a first-order-like transition is the existence of a nonconcave region in the microcanonical entropy, a set of points where one can find a direction with a positive second derivative [1,2]. For the current model where the conserved quantity $\mathbf{n}$ is discrete, I examined the discretized second derivatives

$$\begin{aligned}
\Delta_a\Delta_b S_L \equiv\ & S_L(n_1+a,n_2+b)\\
&+S_L(n_1-a,n_2-b) - 2S_L(n_1,n_2)
\end{aligned} \tag{12}$$

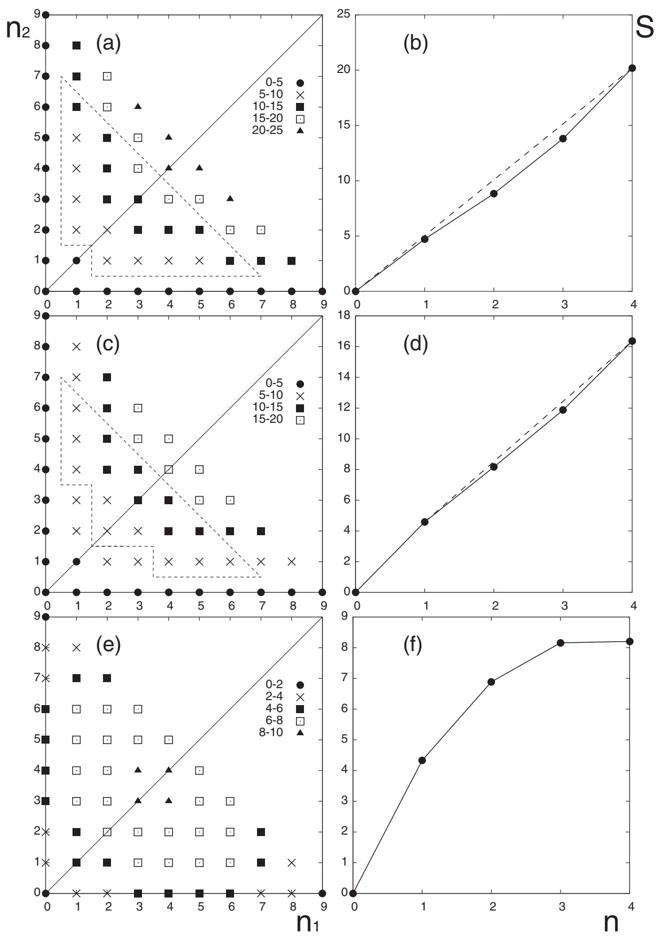

FIG. 1. The generalized entropy function $S_L(n_1,n_2)$ for $L = 10$ is displayed with distinct symbols for points belonging to different ranges of function values, for (a) $q = 0.5$, (c) $q = 0.7$, and (e) $q = 2.0$. The nonconcave regions are enclosed by dashed lines. The cross sections along the diagonal lines $n_1 = n_2$ in (a), (c), and (e) are displayed in (b), (d), and (f). The concave envelopes are drawn in figures (b) and (d) with dashed lines as visual guides.

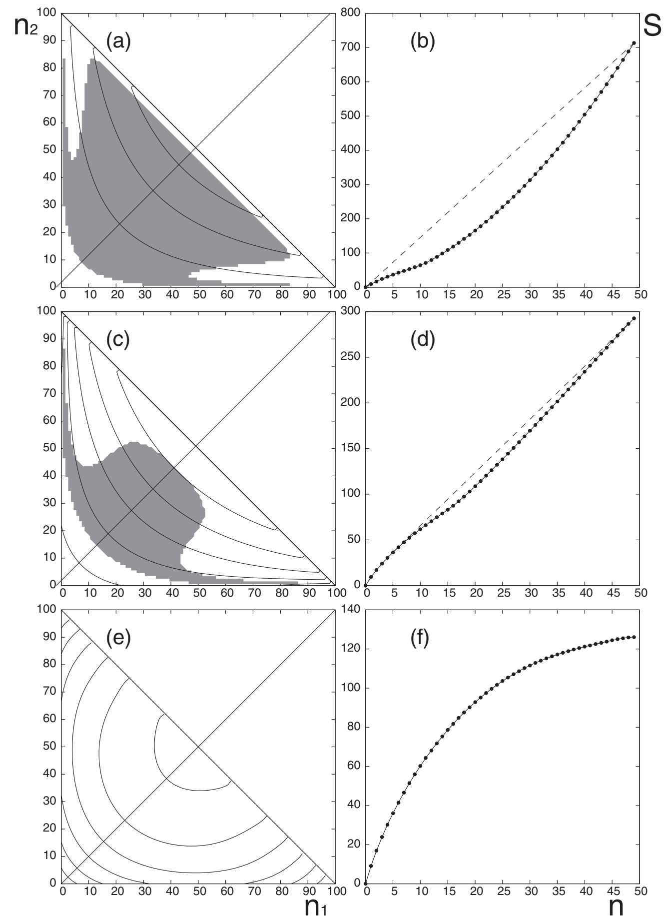

FIG. 2. The contours of the generalized entropy function $S_L(n_1,n_2)$ for $L = 100$ are drawn for (a) $q = 0.8$, (c) $q = 1.1$, and (e) $q = 2.0$ at intervals of 200, 50, 20, respectively. The nonconcave regions are colored in gray. The cross sections along the diagonal lines $n_1 = n_2$ in (a), (c), and (e) are displayed in (b), (d), and (f). The concave envelopes are drawn in figures (b) and (d) with dashed lines as visual guides.

along the horizontal $[(a,b) = (1,0)]$, vertical $[(a,b) = (0,1)]$, and two diagonal $[(a,b) = (1,\pm 1)]$ directions. A point in the interior is nonconcave if any one of these four quantities has a positive value.

The generalized entropy function $S_L(n_1,n_2)$ is shown in the left panels of the Figs. 1 and 2 for $L = 10$ and $L = 100$, respectively, for various values of q. Note that the entropy has the symmetry with respect to the line $n_1 = n_2$ due to the invariance under the simultaneous application of particle-type exchange $1 \leftrightarrow 2$ and the parity inversion $k \leftrightarrow -k$. We see that for small enough values of q, a nonconcave region appears in the generalized entropy, enclosed by dashed lines in Fig. 1 and denoted as gray regions in Fig. 2. As q increases, the nonconcave region shrinks and eventually disappears for large enough values of q. We find that the nonconcave region always includes a part of the line $n_1 = n_2$. The second derivative at such a point is also the largest along the $(1,1)$ direction, which tells us that for a sufficiently small q, when the system is divided into subsystems with respect to a pair of vacancies, it is most probable that there is an inhomogeneity for the total particle numbers, but that there are the same numbers of both species at both sides. Note that this is an *exact* statement for a *finite* value of L, in contrast to the results of previous works where the limit $L \to \infty$ was considered [28–30,32].

The cross sections of $S_L(n_1,n_2)$ along the line $n_1 = n_2 = n$, $S_L(n,n)$, are also displayed in the right panels of Figs. 1 and 2. When there is a convex intruder, the region of the first-order-like transition is defined in terms of the concave envelope constructed by drawing a straight line that is tangent to the curve at two points. The segment of the line bounded by the two contact points are shown with dashed lines in the right panels whenever they exist. The two contact points then define upper and lower boundaries $\rho_\pm$ of the transition region for $n_1 = n_2$ in the space of particle density $\rho \equiv n/L$ $(0 \leqslant \rho < 0.5)$. From here on, we will restrict ourselves to the subspace of $n_1 = n_2$ that was extensively studied in the literature [29,30,32].

The boundaries $\rho_\pm$ are drawn as functions of q to produce a phase diagram in Fig. 3 for $L = 10$ and $L = 100$. The mean field result $\tilde{q}(\rho) = (1 + 6\rho)/(1 + 2\rho)$ in the limit of $L \to \infty$ is shown in Fig. 3 with a dashed line for comparison [29], where $\tilde{q}(\rho)$ is the inverse function of $\rho_\pm(q)$.

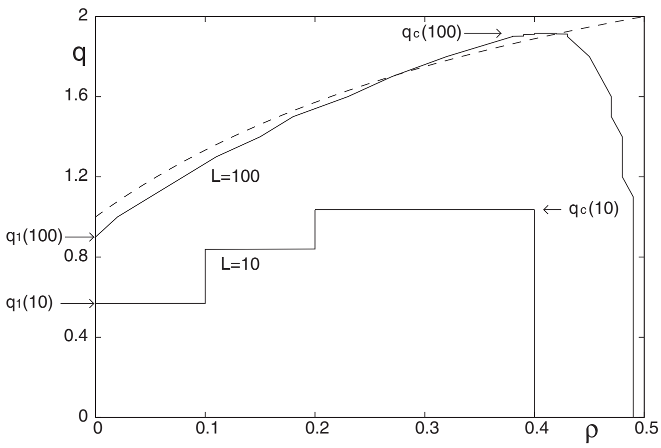

FIG. 3. The phase boundary $\rho_\pm(q)$ for $n_1 = n_2$, for $L = 10$ and $L = 100$. The mean field result in the limit of $L \to \infty$ is shown with a dashed line for comparison.

The high-density side of the phase boundary, $\rho \geqslant \rho_+$, corresponds to the condensed phase. Note that there is a $q_1(L)$ such that $\rho_- = 0$ for $q \leqslant q_1(L)$, in which case the low-density phase $\rho = \rho_- = 0$ is just the vacuum without any particles present. For $q > q_1(L)$, the low-density phase $\rho \leqslant \rho_-$ is the fluid phase. As q increases, $\rho_\pm$ approach each other and eventually merge at the critical point $q = q_c(L)$, after which the system is in a homogeneous phase. The values of q_1 and q_c for $L = 10$ and $L = 100$ are indicated with arrows in Fig. 3. The mean field prediction for these parameters are $q_1(\infty) = 1$ and $q_c(\infty) = 2$, as can be easily read from the analytic expression for $\tilde{q}(\rho)$.

The regions $q \leqslant q_1$, $q_1 < q < q_c$, and $q \geqslant q_c$ have been called pure, mixed, and disordered phases [29]. However, the microcanonical analysis shows that in ρ space, each of the regions $q \leqslant q_c$ and $q_1 < q < q_c$ is divided into the vacuum (or fluid) phase ($\rho \leqslant \rho_-$), condensed phase ($\rho \geqslant \rho_+$), and the phase coexistence region ($\rho_- < \rho < \rho_+$). The situation is analogous to the two-dimensional Ising model with the conserved magnetization M and the temperature T. When one simply considers the T dependence, then there is a critical temperature T_c such that the system is in a disordered phase for $T \geqslant T_c$ and an ordered phase for $T < T_c$. However, by examining the M-dependent behavior of the system, one realizes that the ordered phase is divided into an up-spin phase, down-spin phase, and the region of the first-order transition between the up and down phases.

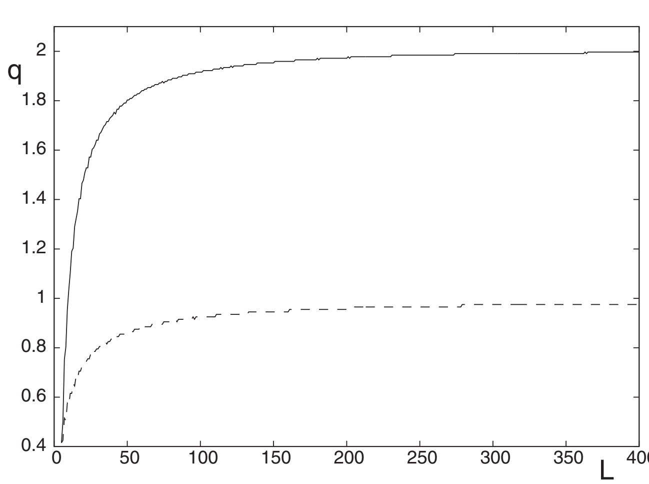

FIG. 4. The critical values q_c (solid line) and q_1 (dashed line) for $n_1 = n_2$ as functions of the system size L. A first-order-like transition exists for $q < q_c$. The low-density phase is a vacuum phase for $q \leqslant q_1$ and a fluid phase for $q_1 < q < q_c$.

I also plot $q_c(L)$ and $q_1(L)$ as functions of L in Fig. 4. Both $q_c(L)$ and $q_1(L)$ approach their mean field values $q_c(\infty) = 2$ and $q_1(\infty) = 1$.

IV. DISCUSSION

In this work, I applied microcanonical analysis to nonequilibrium steady states of an asymmetric diffusion model on a periodic lattice to study a generalized phase transition. This was possible by generalizing the concept of microcanonical entropy to a nonequilibrium steady state via Eqs. (3) and (4), and by defining the generalized phase transition in terms of inhomogeneous distribution of particles among the subsystems defined with respect to a pair of vacancies.

The generalized entropy can be used analyze properties of a nonequilibrium steady state even when there is no convex intruder. In equilibrium statistical physic, derivatives of the microcanonical entropy are taken with respect to energy, volume, or particle number to define temperature, pressure, and chemical potential. Analogously, in the current model we can define the generalized chemical potentials in terms of the discretized first derivatives

$$\tilde{\mu}_i \equiv -S_L(\mathbf{n} + \mathbf{e_i}) + S_L(\mathbf{n} - \mathbf{e_i}), \tag{13}$$

where $\mathbf{e}_1 \equiv (1,0)$ and $\mathbf{e}_2 \equiv (0,1)$.[1] Now suppose a lattice is divided into two regions A and B by a pair of vacancies and $\tilde{\mu}_i(A) > \tilde{\mu}_i(B)$ at some instant. If the system can still be divided into the subsystems A and B with respect to the same pair of vacancies after the steady state is reached, then most probably the particle of species i has flowed from A to B in the process. This is the analog of the statement that energy flows from a high-temperature region to a low-temperature region, or particles flow from a high-chemical-potential region to a low-chemical-potential region, when an infinite-sized system reaches the equilibrium.

The nonequilibrium phase transition of the current model has also been analyzed using the partition function zeros (PFZs) [30]. There, a partition function of the form $\sum_{n_1,n_2} \Omega_L(n_1,n_2) x^{n_1+n_2}$ was constructed where x was called the fugacity. Then the PFZs in the complex plane of x were analyzed to claim that there is a first-order transition as $L \to \infty$, for sufficiently small values of q. It is obvious that $\Omega_L(\mathbf{n})$ was implicitly used as the generalized density of states, but it was not explained why $\Omega_L(\mathbf{n})$ should have such a special status. Also, the physical meaning of the fugacity was unclear, because $\Omega_L(\mathbf{n})$ was regarded as describing particles on a periodic

[1] $\tilde{\mu}$ in fact corresponds to μ/T in equilibrium statistical physics, where μ is the chemical potential. Since T is undefined in the current model, we use the dimensionless quantity $\tilde{\mu}$ instead.

lattice of size L, which is an isolated system. The current work not only justifies the use of $\Omega_L(\mathbf{n})$ as a generalized density, via the factorization Eq. (3), but also shows that $\Omega_L(\mathbf{n})$ in the PFZs approach describes a subsystem of size $L-1$ bounded by a pair of vacancies, rather than the entire system. Then $x = e^{\tilde{\mu}_{\text{bath}}}$, where $\tilde{\mu}_{\text{bath}}$ is the generalized chemical potential of the rest of the system, whose size is much larger than L, which acts as an infinite-size particle reservoir.[2] The microcanonical analysis presented in the current work is more general because a closed system with a finite size can be analyzed.

It has been claimed that the current model does not exhibit a phase transition in the infinite-size limit in the sense that no singularities appear in the physical quantities [32]. This result is in disagreement with those of other works that claim a phase transition for the same system [28–30], but even if this scenario is true, the conclusion of the current work does not change, because we are considering a generalized phase transition defined in terms of the convex intruder in the generalized entropy.

In fact, the generalized phase transition for a finite-size nonequilibrium system is introduced for the first time in the current work through the microcanonical analysis, which would be a subject of much interest for future studies.

[2]This is a special case where $\tilde{\mu}_1 = \tilde{\mu}_2 = \tilde{\mu}_{\text{bath}}$ for the particle reservoir. It is straightforward to consider a case with $\tilde{\mu}_1 \neq \tilde{\mu}_2$.

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