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## Phase coexistence far from equilibrium

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## Abstract

Investigation of simple far-from-equilibrium systems exhibiting phase separation leads to the conclusion that phase coexistence is not well defined in this context. This is because the properties of the coexisting nonequilibrium systems depend on how they are placed in contact, as verified in the driven lattice gas with attractive interactions, and in the two-temperature lattice gas, under (a) weak global exchange between uniform systems, and (b) phase-separated (nonuniform) systems. Thus, far from equilibrium, the notions of universality of phase coexistence (i.e., independence of how systems exchange particles and/or energy), and of phases with intrinsic properties (independent of their environment) are lost.

## 1. Introduction

Consider a liquid mixture, for example methanol and *n*-hexane, cooled below the unmixing temperature, so that two distinct phases emerge, separated by an interface of microscopic thickness. The properties of the coexisting phases can be predicted by equating the chemical potentials of the two components, in homogeneous samples of each phase. Similarly, knowing the chemical potential of the homogeneous liquid and vapor phases as functions of temperature and pressure permits one to predict the liquid–vapor coexistence curve. Is the same thermodynamic analysis possible far from equilibrium?

A central issue in nonequilibrium physics is whether thermodynamics can be extended to systems far from equilibrium, in particular, to steady states [1–12]. A key thermodynamic concept is phase coexistence; indeed, one of the principal applications of equilibrium thermodynamics is the prediction of phase coexistence based upon knowledge of the isolated phases.

While there has been much study of nonequilibrium statistical systems and their associated phase transitions [13–17], the issue of coexistence between far-from-equilibrium phases remains largely unexplored. An important contribution in this regard is the study of intensive thermodynamic parameters (ITPs) in nonequilibrium steady states (NESSs) by Bertin *et al*, who showed that such parameters can be defined in a consistent manner provided a certain asymptotic additivity property holds. These authors further show that ITPs of coexisting nonequilibrium phases are equal for a specific kind of exchange dynamics. Pradhan *et al* [8] tested, via numerical simulation of driven lattice gases in contact, the validity of the zeroth law for temperatures in NESS. They found that the zeroth law holds to good approximation, and suggest that observed deviations may be attributed to the nonuniformities induced by the contact itself. In [18] it is found that such inconsistencies, though small, exist even if nonuniformities are eliminated by using global rather than local exchange of particles, *unless* a particular class of exchange rates (Sasa–Tasaki (ST) rates, see below) are employed. Despite the inconsistencies noted in [18], Pradhan and Seifert [9] were able to construct a mean-field theory of phase coexistence in the driven lattice gas that compares well with simulation. Chatterjee and coworkers [10, 11] have argued that ITPs can be defined consistently for interacting systems in NESS provided spatial correlations are short-ranged, and that the systems interact weakly.

Instances of phase separation in NESSs have been known for some time, for example in the driven lattice gas with attractive interactions [13–15]. It appears to have been assumed, implicitly, that nonequilibrium phase coexistence is well defined.

Equilibrium phase coexistence enjoys a high degree of universality. That is, if phases A and B with different densities, for example, are known to coexist when present in a single nonuniform system, (e.g., as a result of phase separation), then they must also coexist if a pair of uniform systems, prepared in phases A and B, are permitted to exchange particles and/or energy across a fixed boundary. Here I show, via explicit examples, that which nonequilibrium phases actually coexist depends upon how they make contact, that is, on how they are permitted to exchange particles. Thus, far from equilibrium, the universality associated with equilibrium phase coexistence is lost.

Equilibrium statistical mechanics has established a very general notion of ‘phase’ in terms of Gibbs measures [19]; since such a formalism is not available far from equilibrium, I revert to simple operational definitions. I consider systems maintained out of equilibrium by a steady drive acting on the particles, or via contact with two reservoirs having different temperatures. The condition maintaining the system away from equilibrium is called a *drive*; the drive provokes a flux of energy and/or matter through the system.

A nonequilibrium phase corresponds to a macroscopic state of a system under a drive, having time-independent, reproducible properties that vary smoothly with the drive intensity and the external parameters (such as temperature and chemical potential) associated with the reservoir or reservoirs in contact with the system. If the macroscopic properties depend in a singular manner on the drive or other external parameters, the system is said to suffer a (nonequilibrium) phase transition.

Now consider two systems in NESSs, subject to the same drive and external parameters, but with distinct, spatially uniform macroscopic properties. The systems represent coexisting phases if, when allowed to exchange energy or matter, the net flux of the quantity or quantities they may exchange is zero. Nonequilibrium phase coexistence emerges spontaneously at *phase separation*, in which, varying some external parameter, a homogeneous phase becomes unstable, yielding a new stable steady state containing distinct macroscopic phases separated by a sharp interface. (The coexisting phases are clearly free to exchange particles and energy in this situation.)

The stochastic particle system known as the driven lattice gas with attractive interactions (or Katz–Lebowitz–Spohn (KLS) model [13]) provides a simple realization of phase separation far from equilibrium. The drive, which favors particle motion along a certain axis, tends to increase the potential energy, because it increases the likelihood of transitions with  $\Delta E > 0$  more than it does those with  $\Delta E < 0$ . Under steady conditions, energy increase due to the drive is balanced by energy transfer to a reservoir at temperature  $T_R$ , the temperature appearing in the transition rates, as specified in the next section.

At high temperatures, the state with uniform density is stable, but (in two or more dimensions) below a certain value of  $T_R$ , the system segregates into high- and low-density phases separated by a narrow interface, much as its equilibrium counterpart (equivalent to the Ising model with fixed total magnetization) undergoes phase separation. The high- and low-density regions relax to a state in which their macroscopic properties are time-independent, despite the presence of the drive. These ‘empirical’ facts about the model (known from extensive numerical simulations) provide an unambiguous example of coexisting nonequilibrium phases. I note that phase separation occurs in the KLS model for both Metropolis and Kawasaki transition rates in the direction perpendicular to the drive, as well as for the ST rates used here. The critical temperature depends upon which rate is chosen [15].

Suppose a pair of coexisting phases, A and B, are found in a driven, phase-separated system, and that we prepare uniform systems with the same macroscopic properties (such as density), as observed in phases A and B, respectively, subject to the same drive and external parameters as in the phase-separated system. We may now ask:

- (1) Are the isolated uniform phases A and B stable?
- (2) If they are stable, and are allowed to exchange particles and/or energy, will they coexist?

In equilibrium, the stability and coexistence of the uniform phases (given their coexistence under phase separation), is so ‘obvious’ that the corresponding questions have hardly been explored, far from equilibrium. I investigate these questions in the context of two far-from-equilibrium models: the KLS model mentioned above, and a two-temperature lattice gas (TTLG). Precise definitions of the models are given below.

In the present study, the transition rates for particle exchange between (potentially) coexisting phases are taken as ST rates<sup>1</sup>, which depend on the interaction energy before, but not after, the transition. I use ST rates for two reasons. First, as argued in [12], these rates correspond to thermally induced transitions over an energy

<sup>1</sup> The ST rate for transfer of a particle from system A to system B depends exclusively on parameters associated with A, and vice-versa [12]. Imposing detailed balance, the ST rate for this transition takes the form  $w_{ST} = \epsilon \exp[\beta(\mu_A - E_A)]$ , where  $E_A$  is the energy of interaction between the particle and its neighbors, and  $\epsilon$  is an arbitrary rate factor. The ST expression can be seen as resulting from a high energy barrier between A and B; the particle first makes a transition from A to the barrier, and from there to B.

barrier: as in the classic Kramers escape problem, the particle cannot ‘know’ the energy landscape on the other side of the barrier. Second, as shown in [18], consistent definitions of intensive properties (temperature, chemical potential) for the driven system are in general only possible using ST rates. (Here ‘consistent’ is used in the sense of the zeroth law of thermodynamics.)

The balance of this paper is organized as follows. In section 2 I report on studies of phase coexistence in the KLS model, followed, in section 3, by studies of the TTLG. Section 4 presents our conclusions.

## 2. KLS model

The KLS model [13–15], is a stochastic lattice gas in which each site  $i$  of a lattice is either vacant (occupation variable  $\sigma_i = 0$ ) or occupied ( $\sigma_i = 1$ ). The interaction energy is:

$$E = -\sum_{\langle i,j \rangle} \sigma_i \sigma_j, \quad (1)$$

where the sum is over nearest-neighbor (NN) pairs of sites; each NN particle pair lowers the energy by one unit. In equilibrium, this system is equivalent to the NN ferromagnetic Ising model; on the square lattice, it exhibits a continuous phase transition at temperature  $T_c = T_{c,i}/4 \simeq 0.5673$ , where  $T_{c,i}$  denotes the Onsager temperature of the Ising model [20]. The KLS model is equipped with a particle-conserving NN hopping dynamics, and, crucially, a nonequilibrium drive  $\mathbf{D} = D\mathbf{i}$  imposed via hopping rates favoring displacements along the  $+x$  direction (which must be periodic), and inhibiting those in the opposite sense. The acceptance probability for a particle displacement  $\Delta x$ , along the  $x$  direction is

$$p_{a,x} = \min\{1, \exp[-\beta(\Delta E - D\Delta x)]\}, \quad (2)$$

where  $\beta = 1/T_R$ . In the present work I study the infinite- $D$  limit: all attempts to hop along the  $+x$  direction are accepted (provided the target site is unoccupied), while hopping in the opposite direction is prohibited. The acceptance probabilities for hopping by particle  $j$  in the transverse directions ( $\pm y$ ) follow the ST prescription:

$$p_{a,y} = \exp[-\beta n_j], \quad (3)$$

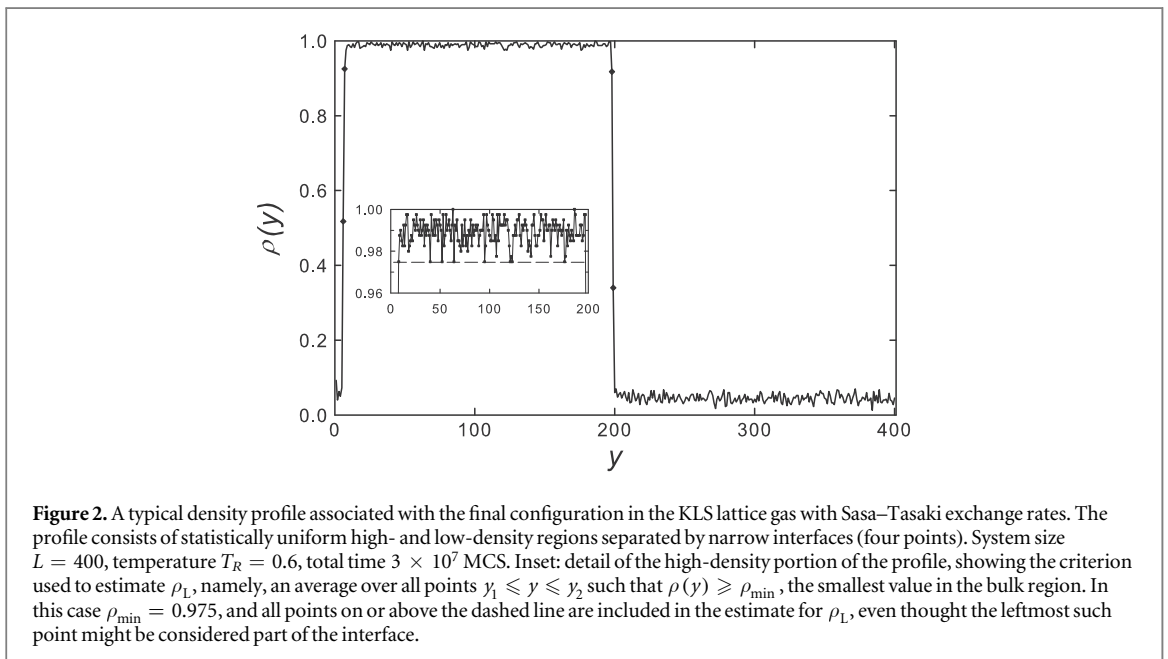
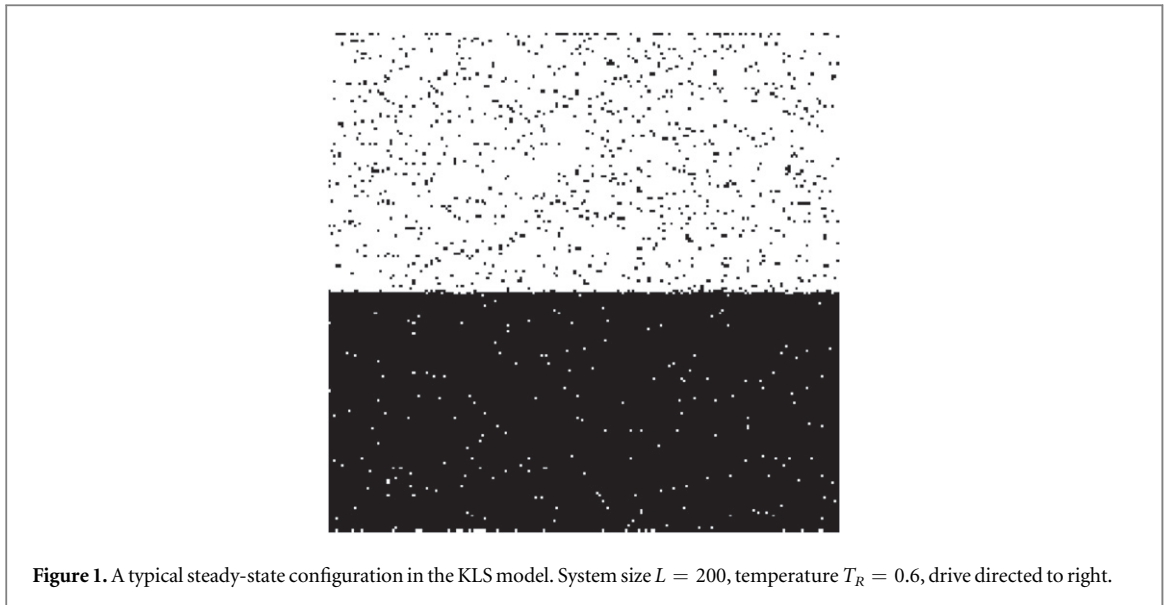
where  $n_j$  is the number of occupied NNs of particle  $j$  prior to hopping. I use ST rates for hopping perpendicular to the drive because, under phase separation, the interface is along the drive. (An interface perpendicular to the drive is unstable [13–15].) Thus exchange between coexisting phases is governed by ST rates.

I simulate the KLS model on square lattices of  $L_x \times L_y$  sites, with periodic boundaries, using  $L = 100, 200$  and  $400$ . (In most of the studies,  $L_x = L_y$ .) In the continuous-time stochastic evolution, each particle is equally likely to be the next to attempt to hop; hopping is always to a NN site. If the latter is unoccupied, the particle displacement is accepted with the probabilities defined above. A Monte Carlo step (MCS) corresponds to one attempted move per lattice site; simulations are run for a total of  $1-4 \times 10^7$  MCS, with an initial period of  $1-2 \times 10^7$  MCS for relaxation to the steady state. Averages are performed over 5–10 independent realizations.

A preliminary study revealed that phase separation occurs for temperatures  $T_r < T_c \simeq 0.90$ . (This is somewhat higher than the value,  $T_c = 0.769(2)$ , for the square lattice under infinite drive, using Metropolis rates [21].) For present purposes a precise result for  $T_c$  is not needed: the only need for an estimate of  $T_c$  is to study values well below it, to avoid finite-size effects and large fluctuations associated with the critical region.

Starting from a random, statistically uniform distribution of particles, the system evolves to a configuration with a number of dense stripes separated by rarefied regions: the hallmark of phase separation in the KLS model. To minimize interfacial effects and facilitate determination of the coexisting densities, I employ a *single-stripe* initial configuration in a half-filled system, which relaxes to a stationary state consisting of a single dense and a single rarefied region (see figure 1). The coexisting densities  $\rho_L$  and  $\rho_V$  are determined through analysis of the final density profile  $\rho(y)$  (see figure 2). To obtain reliable estimates for the coexisting densities and their uncertainties, six such profiles, taken after a total of  $3 \times 10^7$  MCS, are analyzed. Each profile yields estimates for  $\rho_L$  and  $\rho_V$ ; the results for the six independent studies are averaged to yield (for  $L = 400$  and  $T_R = 0.6$ ),  $\rho_L = 0.9890(2)$  and  $\rho_V = 0.0451(4)$ , where the figures in parentheses denote statistical uncertainties (standard deviation of the mean). Profiles taken after only  $2 \times 10^7$  MCS yield densities of  $0.9889(2)$  and  $0.0450(5)$ , showing that the densities have indeed relaxed to their stationary values. Studies of a smaller system ( $L = 200$ ) yield coexisting densities of  $0.9893(2)$  and  $0.0459(3)$ , showing that finite-size effects are minimal.

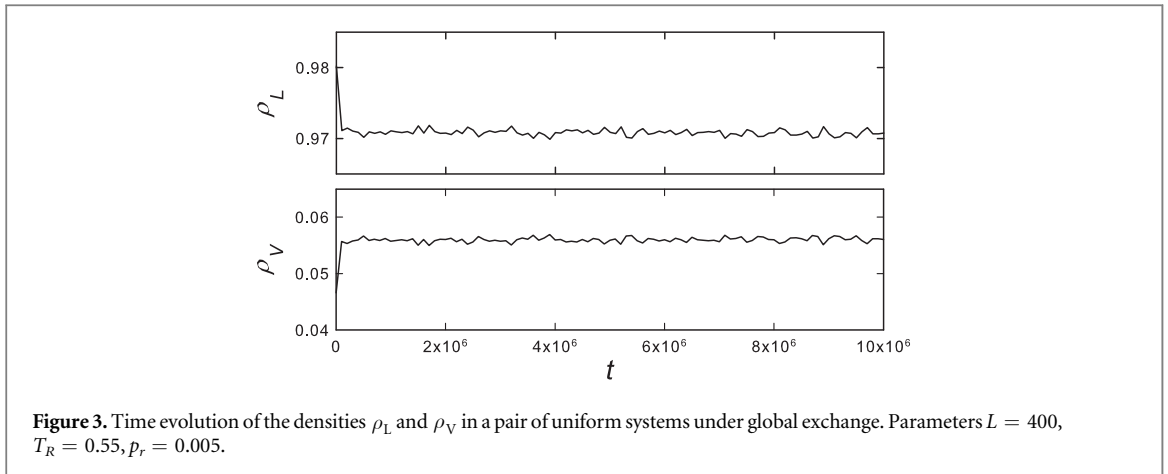
All profiles exhibit well defined bulk regions, in which the density  $\rho(y)$  is free of any significant linear trend or curvature; the bulk regions are separated by interfaces. To decide if a given point  $y$  (near the interface) should be taken as part of the bulk, I use the criterion illustrated in the inset of figure 2: the minimum value  $\rho_{\min}$ , within the bulk of the high-density region is identified, and the maximal set  $\{y_1, \dots, y_2\}$  such that  $\rho(y) \geq \rho_{\min}$  is taken as the bulk, and used to calculate  $\rho_L$ . An analogous criterion (using the maximum density in the low-density region) is employed in calculating  $\rho_V$ . Note that this procedure might be expected to yield a slight underestimate



of  $\rho_L$ , and a slight overestimate of  $\rho_V$ , as relatively rare density values, that might arguably be assigned to the interface, are included in the bulk. The absence of significant finite-size effects nevertheless suggests that such overestimates (or underestimates) are minimal.

In this manner, I determine the coexisting densities for temperatures ranging from 0.5 to 0.7, well below the critical temperature. In contrast to the equilibrium lattice gas (or the KLS model using Metropolis rates), here the coexisting densities do not obey  $\rho_L + \rho_V = 1$ . This is because ST rates do not respect particle-hole symmetry. I verify that the coexisting densities are insensitive to modest changes in the aspect ratio, that is, using  $L_x = 2L_y$  or  $L_x = L_y/2$ .

I turn now to studies of *composite* systems, consisting of a pair of *uniform* systems of the same size (with periodic boundaries), at the same reservoir temperature and subject, as before, to an infinite drive. The two systems, L and V, are prepared with the densities  $\rho_L$  and  $\rho_V$  found to coexist in the phase-separated systems. These systems are allowed to exchange particles with an overall attempt rate  $p_r$ , again using ST rates, via weak global exchange. (It is important to note that there are *no interactions* between particles in different systems.) Global exchange means that any particle in one system may attempt to jump to any site in the other. Weak exchange corresponds to the limit  $p_r \rightarrow 0$ , and is similar to the weak interaction condition defined in [11]. Important consequences of weak exchange are: (1) the systems in contact are statistically independent; (2) particle exchange does not provoke spatial inhomogeneities within these systems; (3) under weak global



**Figure 3.** Time evolution of the densities  $\rho_L$  and  $\rho_V$  in a pair of uniform systems under global exchange. Parameters  $L = 400$ ,  $T_R = 0.55$ ,  $p_r = 0.005$ .

**Table 1.** KLS model: coexisting densities in a single phase-separated system ( $\rho_{L,1}$ ,  $\rho_{V,1}$ ), and in a pair of uniform systems under weak global exchange ( $\rho_{L,2}$ ,  $\rho_{V,2}$ ).

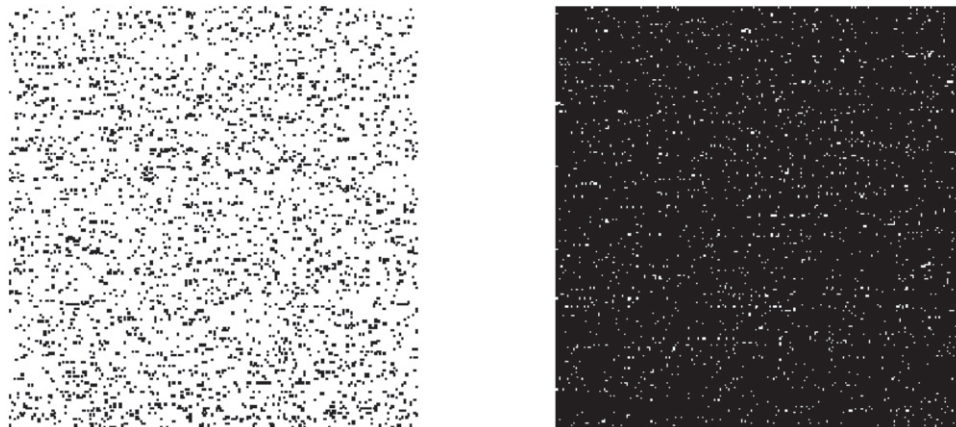
$T_R$	$\rho_{L,1}$	$\rho_{V,1}$	$\rho_{L,2}$	$\rho_{V,2}$
0.50	0.9965(1)	0.0216(1)	0.9826(1)	0.0360(1)
0.55	0.9939(1)	0.0330(2)	0.97088(2)	0.05603(2)
0.60	0.9890(2)	0.0451(4)	0.9521(1)	0.0818(1)
0.625	0.9859(1)	0.0531(1)	0.9411(1)	0.1008(1)
0.65	0.9821(2)	0.0621(1)	0.9297(1)	0.1266(1)

exchange using ST rates, it is possible to associate a temperature and a chemical potential with *spatially uniform* NESSs in a thermodynamically consistent manner [18].

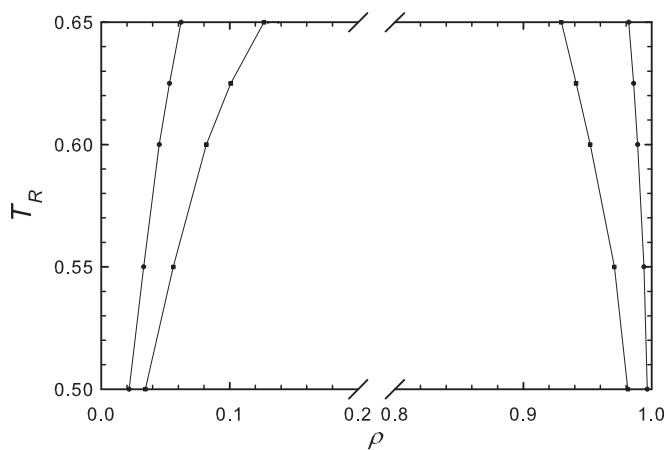
Using the same simulation algorithm as before, I monitor the particle densities in systems L and V over periods of order  $10^7$  MCS; relaxation to stationary values typically requires fewer than  $10^6$  MCS. The final configuration is checked to assure that both systems remain spatially uniform, i.e., that phase separation has not occurred within either system. Figure 3 shows a typical evolution of the densities in the uniform systems under global exchange. Typical steady-state configurations are shown in figure 4. Varying the exchange attempt rate  $p_r$  between 0.01 and 0.001, no significant change in the stationary densities  $\rho_L$  and  $\rho_V$  is found.

In the stationary state, systems L and V coexist, but *not* at the densities observed under phase separation in a single system. Compared to the single, phase separated system, the density  $\rho_L$  in the uniform system is consistently smaller, while  $\rho_V$  is consistently larger. (Recall that the procedure for estimating coexisting densities in the single phase-separated system is likely to *underestimate* not overestimate, these differences.) A quantitative comparison of the coexisting densities is given in table 1, for system size  $L = 400$  (no significant differences are observed between studies using  $L = 200$  and  $L = 400$ ); figure 5 illustrates the general trends. The coexisting densities under phase separation and under weak global exchange between uniform systems are clearly incompatible. While the differences between the coexisting liquid densities  $\rho_{L,1}$  and  $\rho_{L,2}$  amount to but a few percent, those for the vapor are considerably larger, with  $\rho_{V,2} \simeq 2\rho_{V,1}$  at the highest temperatures studied.

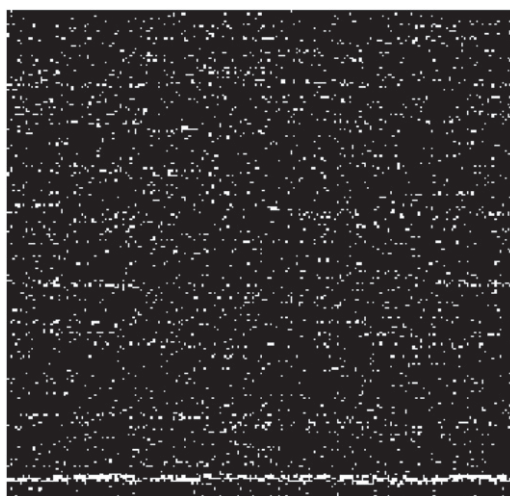
In the two-system studies, the total particle number  $N$  is determined using the bulk densities observed in the single, phase-separated system:  $N = L^2(\rho_{L,1} + \rho_{V,1})$ . Initially, one system is fully occupied ( $N_L(t=0) = L^2$ ), while the other contains the remaining  $N_V = N - L^2$  particles, inserted at randomly chosen sites. During the stochastic evolution under exchange, the particle numbers  $N_L$  and  $N_V$  quickly attain stationary values, subject, of course, to the constraint of fixed  $N = N_L + N_V$ . For temperatures  $T_R \leq 0.6$ , the final configurations are spatially uniform (see figure 4). At higher temperatures, however, one observes the formation of stripes: one or both systems have undergone phase separation, showing that the phase that is stable in the single system is *unstable* in the composite system (see figure 6). To determine the coexisting densities in this case, I search for values of  $N$  such that both systems remain uniform. In table 1, the values for  $T_R \geq 0.625$  were obtained in this manner; the densities listed are the smallest for which the composite system is stable. Stable coexistence in the composite system is not observed for *any* density, for  $T_R = 0.7$ . (The situation is reminiscent of a somewhat different system, studied by Achahbar and coauthors, [22, 23], who employed exchange between pairs of corresponding sites, rather than global exchange, in a pair of KLS models. These authors observed phase separation within each system (in corresponding regions), for temperatures between the critical values for the



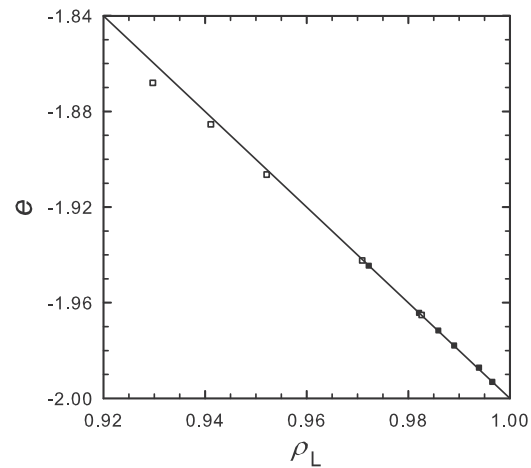
**Figure 4.** A typical steady-state configuration in the KLS model: two-system coexistence under weak global exchange (V on left, L on right). System size  $L = 200$ , temperature  $T_R = 0.6$ , exchange rate  $p_r = 0.003$ , drive directed to right.



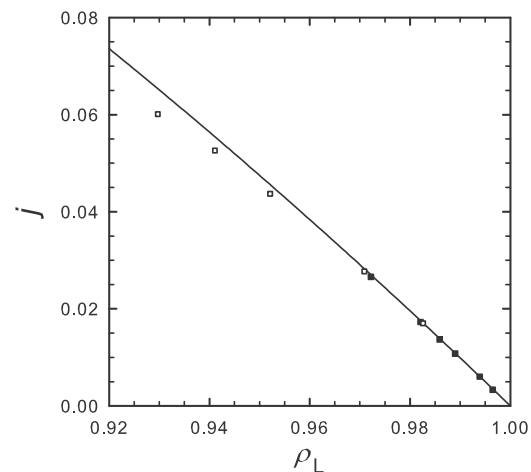
**Figure 5.** KLS model: coexisting densities  $\rho_L$  (right side) and  $\rho_V$  (left side) versus reservoir temperature  $T_R$ . The points nearer the vertical axes correspond to the single phase-separated system; those further from the axes correspond to uniform systems under weak global exchange. System size  $L = 400$ . Error bars smaller than symbols.



**Figure 6.** A typical steady-state configuration of the dense system (L) in the KLS model under weak global exchange. (The other system (V) is uniform and is not shown.) Although systems L and V were given initial densities equal to those observed in the single phase-separated system with the same parameters, an empty strip has formed. System size  $L = 200$ , temperature  $T_R = 0.65$ , drive directed to right.



**Figure 7.** KLS model: mean interaction energy per particle  $e$  versus density  $\rho$  at coexistence in the high-density phase, in the single system (filled symbols) and the composite system (open symbols). Solid line: random-mixing prediction,  $e = 2\rho$ .



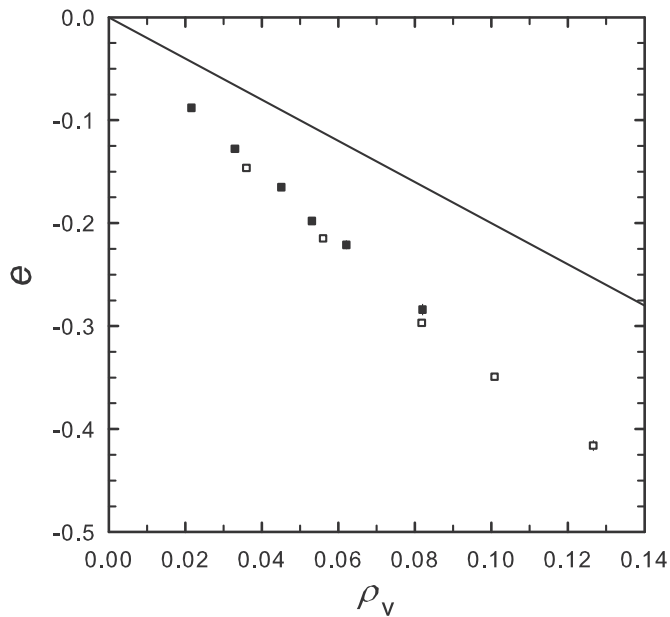
**Figure 8.** KLS model: mean current  $j$  versus density  $\rho$  at coexistence in the high-density phase, in the single system (filled symbols) and the composite system (open symbols). Solid line: random-mixing prediction,  $j = \rho(1 - \rho)$ .

driven and undriven systems, and phase coexistence between uniform systems at temperatures below the critical value for the undriven model.)

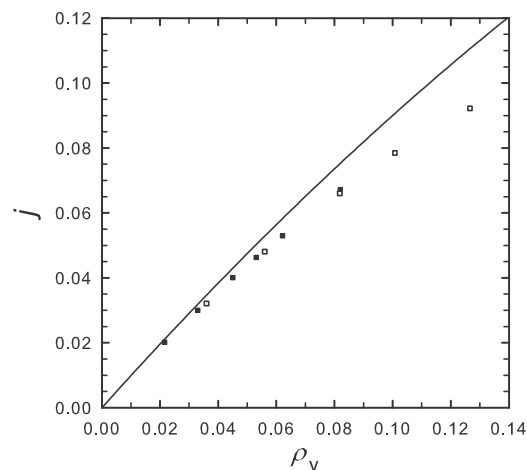
Discrepancies between coexisting densities in the single and composite systems naturally lead to differences in other stationary macroscopic properties, such as the mean interaction energy per particle  $e$ , and mean current density  $j$ . Plotting these functions versus particle density  $\rho$  affords some insight into the nature of the coexisting phases. In the high-density (L) phase, both  $e$  and  $j$  appear to be functions of  $\rho$  alone, and are well-approximated by the random-mixing expressions  $e = 2\rho$  and  $j = \rho(1 - \rho)$ , as shown in figures 7 and 8. The corresponding plots for the low-density (V) phase (see figures 9 and 10) show substantial differences between the single- and composite-system properties, and strong deviations from the random-mixing predictions. This is rather natural, since we expect  $e$  and  $j$  to depend on density *and* temperature, and when the densities are equal, the temperatures are different. The low sensitivity to temperature evident in the L phase is consistent with its approximation to a random mixture, which would appear to be a feature of a density close to unity and a correspondingly short correlation length.

Summarizing results for the KLS model, at all temperatures studied, there is a clear discrepancy between the densities characterizing coexisting phase in a single, phase-separated system, and those associated with a pair of uniform systems that coexist under global exchange.





**Figure 9.** KLS model: mean interaction energy per particle  $e$  versus density  $\rho$  at coexistence in the low-density phase, in the single system (filled symbols) and the composite system (open symbols). Solid line: random-mixing prediction,  $e = 2\rho$ .

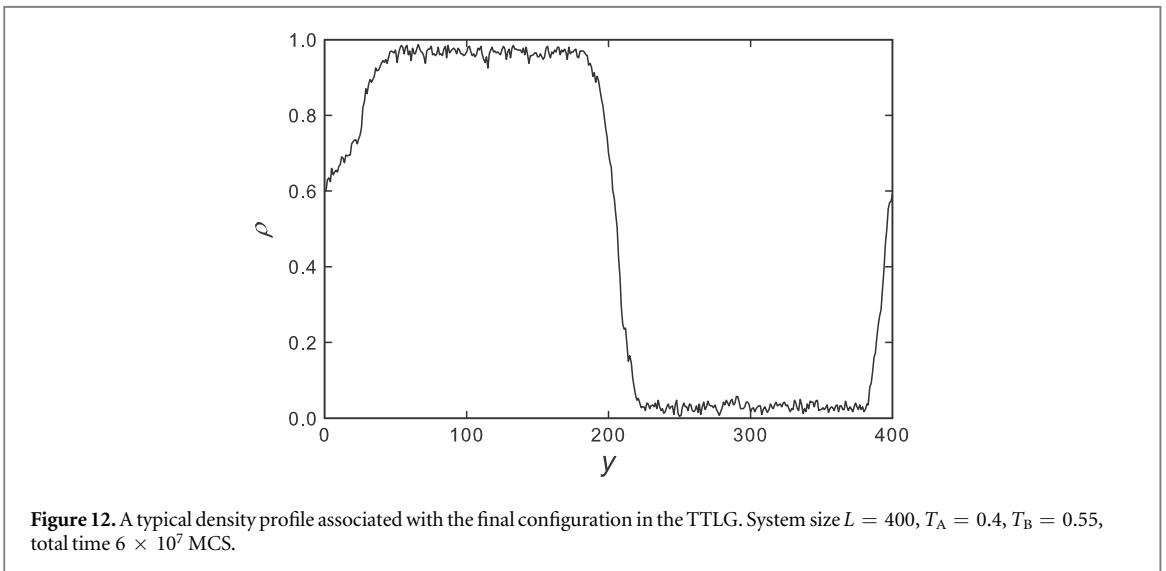
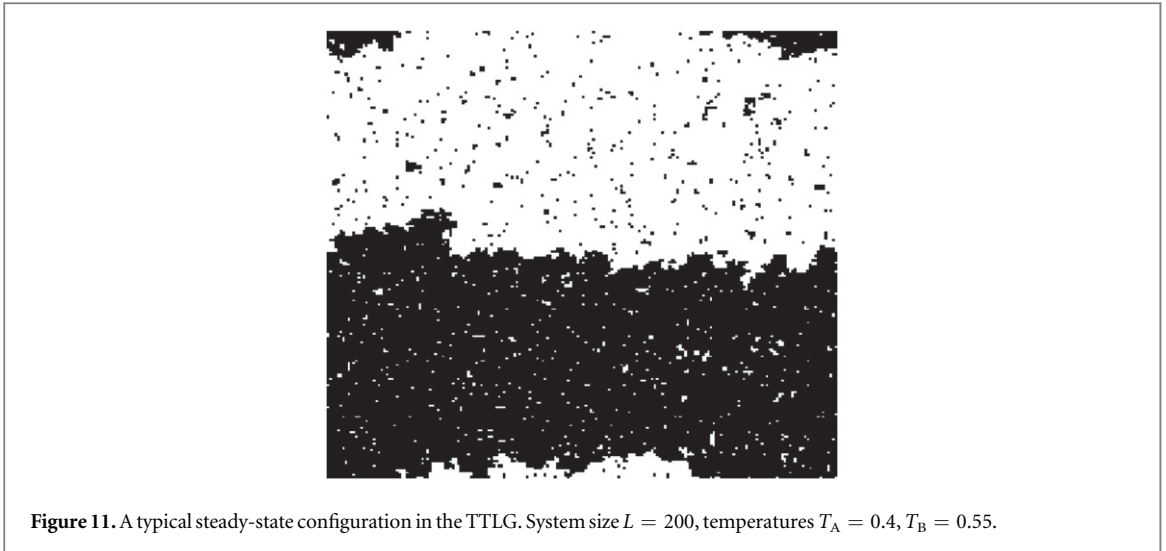


**Figure 10.** KLS model: mean current  $j$  versus density  $\rho$  at coexistence in the low-density phase, in the single system (filled symbols) and the composite system (open symbols). Solid line: random-mixing prediction,  $j = \rho(1 - \rho)$ .

### 3. Two-temperature lattice gas

The KLS model is strongly anisotropic, and features a particle current along one of the lattice axes. It is natural to ask whether these features are somehow responsible for the nonuniversality of phase coexistence documented above. This question motivates study of a second far-from-equilibrium system, a TTLG. The interaction energy is again given by equation (1). The stochastic evolution is via NN particle hopping, with acceptance probabilities as in equation (3), in *both* the  $x$  and  $y$  directions. The nonequilibrium drive in this case takes the form of two reservoir temperatures,  $T_A$  and  $T_B$ , associated with sublattices A and B. (Sublattices A and B comprise the sets of sites  $(i, j)$  with  $i + j$  even and odd, respectively.) The case  $T_A = T_B$  corresponds to the equilibrium lattice gas, equivalent to the NN Ising model. For  $T_A \neq T_B$  the system cannot reach equilibrium. Note however that the system is isotropic and that there is no net particle current. (The drive  $T_A - T_B$  induces an energy flux between the sublattices.)

The TTLG exhibits a line of Isinglike phase transitions in the  $T_A - T_B$  plane [24–26]. To observe phase separation in the form of coexisting strips, I initialize the system with all sites in half of the system ( $j \leq L/2$ ) occupied, and the other half vacant. Coexisting densities are determined from the final density profile, as in the



KLS studies. I verify that the estimates for system sizes of  $L = 400$  and  $L = 200$  agree to within uncertainty. Next, a pair of uniform systems, with particle densities corresponding to the coexisting densities found under phase separation, are prepared, and studied in simulations of  $1-3 \times 10^7$  MCS. As before, the particle densities in the coexisting systems are monitored; final configurations are checked for uniformity.

As a test, I set  $T_A = T_B = 0.5$ , and observe  $\rho_{L,1} = 0.9552(4)$  and  $\rho_{V,1} = 0.04454(2)$  in the single, phase-separated system, while coexistence in the composite system yields  $\rho_{L,2} = 0.9556(3)$  and  $\rho_{V,2} = 0.0444(3)$ . Thus, as expected, at equilibrium the two modes of coexistence yield phases with densities that agree to within uncertainty, and, moreover,  $\rho_L + \rho_V = 1$ . For unequal sublattice temperatures there are discrepancies. Studies using  $T_A = 0.4$  and  $T_B = 0.55$ , for example, yield  $\rho_{L,1} = 0.9644(3)$  and  $\rho_{V,1} = 0.03001(3)$ , in a single phase-separated system. A typical configuration is shown in figure 11. Although the interface is now rough (since there is no particle current), well defined bulk phases are present. The coexisting densities are again obtained via analysis of the final density profiles; a typical profile is shown in figure 12. As before, coexisting densities between two uniform systems under weak global exchange are determined by monitoring the particle numbers in the respective systems, once they have attained a steady state. For these temperatures, the composite-system studies furnish  $\rho_{L,2} = 0.9511(15)$  and  $\rho_{V,2} = 0.0333(4)$ , clearly incompatible with the values of  $\rho_{L,1}$  and  $\rho_{V,1}$  cited above. Similar inconsistencies are found at other values of  $T_A$  and  $T_B > T_A$ , as shown in table 2.

In the TTLG with sublattice temperatures  $T_A = 0.4$  and  $T_B = 0.6$ , separation into well defined coexisting phases in a single system is observed, but it appears to be impossible to stabilize coexistence of two uniform phases in a composite system, regardless of how the total density is varied. This may be related to the fact that  $T_B$  is above the critical temperature of the equilibrium lattice gas.

**Table 2.** TTLG: coexisting densities in a single phase-separated system ( $\rho_{L,1}, \rho_{V,1}$ ), and in a pair of uniform systems under weak global exchange ( $\rho_{L,2}, \rho_{V,2}$ ).

$T_A$	$T_B$	$\rho_{L,1}$	$\rho_{V,1}$	$\rho_{L,2}$	$\rho_{V,2}$
0.30	0.50	0.9828(2)	0.0098(3)	0.9709(1)	0.01202(1)
0.35	0.55	0.9706(1)	0.02053(2)	0.951(3)	0.0240(16)
0.40	0.55	0.9644(3)	0.03001(3)	0.9511(15)	0.0333(4)

In both the KLS and TTLG systems, the difference (at a given temperature) between coexisting densities is smaller in the composite system than in the single system. The reason for this tendency, while not obvious, is presumably connected with the presence of an interface in the single system and its absence in the composite system. One might speculate that the one-step transitions taking a particle directly from one bulk phase to the other facilitate exchange in the composite system, as compared with the series of ‘uphill’ displacements required to transport particles across the interface in the single system. This line of argument appears to fail, however, when we note that for very small exchange rates  $p_r$ , the rate of transfers between phases in the composite system is much smaller than that in the single system. The coexisting densities in the composite system are nevertheless essentially independent of  $p_r$ , as it tends to zero.

## 4. Conclusions

Studies of two of the simplest nonequilibrium models exhibiting phase separation are found to have coexisting bulk properties that depend on how the phases coexist. Violations of universality in coexistence are observed in two systems, one anisotropic and bearing a particle current, the other isotropic and free of such a current, suggesting that such violations are generic to phase coexistence far from equilibrium. Put another way, the results suggest that the notion of *phase* as a state of matter with bulk properties depending only on a small set of intensive parameters does not apply far from equilibrium. The properties of the coexisting phases in the KLS and TTLG models depend not only upon the reservoir temperature(s) and the drive strength, but on the spatial relation between the phases. Of course, many further possibilities exist, each presumably leading to a different set of coexisting densities. For example, allowing long-range hopping in the single phase-separated system destabilizes the coexisting regions, leading to multiple-stripe configurations.

As shown in previous work on the KLS model and the lattice gas with nearest-neighbor exclusion (NNE) [18, 27], it is possible to implement steady-state thermodynamics (SST) in a consistent manner in spatially uniform systems, if (and in general only if) ST exchange rates are used, and provided these rates tend to zero. By contrast, SST is inconsistent and without predictive value in spatially nonuniform NNE models. Here, rather than imposing nonuniformities via a nonuniform drive, or walls, they arise spontaneously due to phase separation. Our results imply that in this case as well, intensive properties (whose equality would predict phase coexistence) cannot be assigned to the coexisting phases consistently. While one could in principle assign a temperature and chemical potential to spatially uniform systems via coexistence with thermal and particle reservoirs [18], these parameters would be of no use in predicting phase coexistence in a single, phase-separated system.

Although ST rates guarantee the zeroth law of thermodynamics, it remains possible that other rates might also be consistent with the zeroth law, if a free energy function can be constructed [11]. In such a case it might be possible to devise particle-exchange rates for which the notion of phase coexistence of NESS is well defined.

Our results suggest a new viewpoint regarding nonequilibrium phases. From the equilibrium context, we are used to thinking of a phase as having intrinsic properties, immutable under coexistence with another phase. Far from equilibrium, the properties of coexisting phases depend not only on the control parameters (drive, reservoir temperature), but also on precisely how exchanges of matter and energy between the phases are realized.

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