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Onset of ordering in driven bilayer lattice gases

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Abstract

We investigate the phase diagram of a system with two layers of an Ising lattice gas at half filling, as a function of the inter-layer coupling J, keeping the usual intra-layer nearest-neighbor attraction constant. In equilibrium, the phase diagram is symmetric under $J \to -J$, but exhibits different ground states. The effects of imposing a uniform external drive are studied by simulation and analytic techniques. In particular, we attempt to develop a simple intuitive picture which allows us to predict the approximate location of T_c in driven lattice gases.

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One of the major problems in the study of non-equilibrium steady states is the absence of a free energy, with its associated store of simple intuitive arguments, which often allow us to predict the qualitative phase diagram of equilibrium systems rather successfully. For example, the structure of ground states can be predicted based on simple energetics, while the existence of phase transitions, especially in systems with discrete symmetry, as well as the *qualitative* shape of the phase diagram can be obtained easily from Landau–Peierls-type arguments [1], balancing the competing effects of entropy and energy. More sophisticated tools, such as Monte Carlo simulations, mean-field theory or renormalization group methods, can then be invoked to obtain more quantitative details. In contrast, there are as yet no intuitive arguments that would enable us to predict, *qualitatively*, the basic features of phase diagrams even in *simple* non-equilibrium systems. A prime example for this situation is the driven lattice gas, introduced over a decade ago by Katz et al. [2]. In this model, particles hop to nearest-neighbor empty sites, subject to the usual Ising [3] Hamiltonian and a thermal bath at temperature T. The system is driven into a non-equilibrium steady state by a driving force which favors particle hops along a specific lattice axis. While many

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surprising features of this basic model are by now well understood [4], a few results remain puzzling. In particular, it is not yet clear why the critical temperature, $T_c(E)$, *increases* with E, saturating at about 40% above the Onsager temperature as $E \to \infty$ [5]. Since nearest-neighbor bonds are easily broken by sufficiently strong fields, one might in fact have expected a *lower* T_c. Additional data, demonstrating the presence of long-range correlations in the system[6], give a first indication as to why T_c might increase.

In the following, we will explore a related system, consisting of a *bilayer* driven lattice gas. The purpose of our work is twofold. First, this system is closely related to models for intercalated structures [7,8], and we will investigate its phase diagram and critical properties. Second, we wish to test which of these competing arguments successfully predicts the qualitative behavior of the phase diagram. The goal is to develop a simple intuitive picture which can be applied to other systems.

Earlier simulation work [9] investigated driven bilayer lattice gases, with *zero* cross-layer coupling. Surprisingly, *two* transitions were found: As T is lowered, the usual disordered (D) phase orders into a strip phase (S), displaying a single strip in each one of the layers. The two strips lie right on top of one another. At even lower temperatures, a first-order transition occurs into a second ordered state: here, one of the planes is mainly full, while the other is nearly empty, corresponding to homogeneous, opposite magnetizations on the two planes. This state will be referred to as FE. The lack of understanding, as to why there should be *two* transitions, as well as possible applications to the physics of intercalated compounds, motivates our study of a bilayer driven system with *inter-layer* interactions. The goal is to map out the phase diagram in the space spanned by temperature, drive and inter-layer coupling strength. In the following, we will briefly describe our model, report the main results from the simulations, and outline the first steps towards a renormalization group analysis of the continuous transitions in the model. Finally, we will briefly summarize a few intuitive arguments concerning the structure of the phase diagram. A more detailed description of the simulations and of our intuitive picture can be found in [10]. A thorough discussion of the field theory analysis and additional Monte Carlo data will be published elsewhere [11].

We consider two fully periodic $L \times L$ square lattices, arranged in a bilayer structure. Each site (j_1, j_2, j_3), with $j_1, j_2 = 1, \ldots, L$ and $j_3 = 1, 2$, can be either occupied by a particle or empty, denoted by $n(j_1, j_2, j_3) = 0$ or 1. Occasionally, we will use Ising spin language, $s(j_1, j_2, j_3) \equiv 2n(j_1, j_2, j_3) - 1$. To access the usual critical point of the Ising model, we study only half-filled systems, i.e., $\sum n = L^2$. This constraint imposes a conservation law on the dynamics. The particles interact with their nearest neighbors, both *within* and *across* layers, with coupling strengths J_0 and J, respectively. Thus, the Hamiltonian is given by $\mathcal{H} \equiv -J_0 \sum nn' - J \sum nn''$, where n and n' are nearest neighbors within a given layer, and n and n'' differ only by the layer index. Our study is restricted to positive J_0 , with several values of J/J_0 in the range $[-10, 10]$. Negative J's are motivated by the physics of intercalated materials [7,8], where elastic deformations of the host lattice may lead to repulsive effective interactions between

intercalating atoms in adjacent layers. To simulate equilibrium systems coupled to a thermal bath at temperature T, we use mostly standard spin-exchange (Kawasaki) [12] dynamics with Metropolis rates [13]. A few datapoints for large $|J|$ have been obtained using Glauber dynamics, in order to reduce the relaxation times. Since the steady-state properties of an *equilibrium* system are independent of the dynamics (as long as detailed balance is satisfied), this is a valid procedure. Finally, to drive the system into non-equilibrium steady states, we model the bias as an "electric" field (aligned with the 1-axis), acting on "charged" particles, by adding $\pm E$ to the change in configurational energy for hops against/along the field [2,4]. Of course, spin exchange dynamics is used for all simulations at non-zero E.

To distinguish different ordered phases, we choose the appropriate structure factors as order parameters [2,4]. Introducing the Fourier transform of the occupation variables of a given configuration,

$$\tilde{n}(l_1,l_2,l_3)\equiv\frac{1}{2L^2}\sum n(j_1,j_2,j_3)\,e^{2\pi i\left(\frac{j_1l_1+j_2l_2}{L}+\frac{j_3l_3}{2}\right)}, \tag{1}$$

the structure factor is defined as $S(l_1,l_2,l_3)\equiv\langle|\tilde{n}(l_1,l_2,l_3)|^2\rangle$. As usual, the $\langle\ \rangle$'s are time-averages, taken over the run. Clearly, $S(0,1,0)$ and $S(0,0,1)$ measure, respectively, the degree of ordering in the S- and FE-phases. Structure factors associated with a range of other wave vectors have also been monitored, in order to exclude the emergence of additional phases. First-order transitions are identified by the appearance of hysteresis loops in the order parameters as either T or J are varied. As a first approximation, we place the phase boundary at the mid-point between the values where the order parameter jumps in a hysteresis loop. Second-order transitions are associated with a peak in the fluctuations of $|\tilde{n}|^2$, measured as a function of T at fixed J and E. More precise estimates of the phase boundaries are clearly possible, by obtaining better statistics accompanied by a finite size scaling analysis. However, the accuracy of our data is sufficient to reach some conclusions about the nature of the phase transitions in our systems.

We now turn to a summary of our findings. In the equilibrium case, the low-temperature configurations can be determined from energetic arguments. Since the intra-layer coupling J_0 is always attractive, the system attempts to maximize the number of satisfied intra-layer bonds. For large negative J, broken *inter-layer* bonds are favored, and the FE phase results. In contrast, for positive J, the inter-layer bonds must also be satisfied. In the thermodynamic limit, the associated energetic gain offsets the extra interfacial cost for any $J>0$, so that the S-phase prevails in that regime. As the temperature is increased, the system disorders via a continuous transition at a critical temperature $T_c(J)$. Since a gauge transformation relates the $J>0$ system to the $J<0$ one, $T_c(J)$ is even in J. It takes its minimum at $J=0$, where the two layers decouple, so that $T_c(0)$ is given by the Onsager value. Then, as $|J|$ increases, $T_c(\pm J)$ also increases, reaching a value of $2T_c(0)$ for $J=\pm 10$, within the error bars. This bears out the expectation that, in the limit of $J\to\infty$, the system is equivalent to a single $d=2$ Ising model with coupling $2J_0$. Finally, the $J=0$ axis, between $T=0$ and

$T_c(0)$, is a line of first-order transitions, with the point $(T_c(0), J=0)$ being a bicritical point.

Next, we summarize the effects of turning on the drive. Since the drive violates the Ising symmetry, the phase diagram is no longer symmetric (Fig. 1). At high temperatures, the system is disordered, ordering via a continuous transition as the temperature decreases. Intriguingly, the critical temperature $T_c(J,E)$ is *lowered* for large $|J|$, compared to its equilibrium counterpart $T_c(J,E=0)$. This is quite surprising, given that $T_c(E)$ is greater than $T_c(0)$ in the single-layer case. The other remarkable feature is the shift of the bicritical point to *higher* values of T and negative J. Thus, the S-phase prevails, not only for positive J, but also in a finite, triangular region (inset, Fig. 1), characterized by small *negative* J. We conclude that neither energy nor entropy determine the steady state here. For larger values of the inter-layer repulsion, the FE phase is observed to be stable. Its presence is also somewhat surprising since one might have expected a phase with strips in each layer, but *staggered*. The phase boundary between the S- and FE-phases is a line of first order transitions. A few test runs at selected points in the phase diagram for larger systems indicate that none of these characteristics appear to be due to finite size effects.

In the following, we outline the first steps towards a full field-theoretic analysis [11] of critical behavior in this system, beginning with the equilibrium case. Our first task is to coarse-grain the microscopic Hamiltonian $\mathscr{H}$ in order to arrive at a Landau–Ginzburg Wilson Hamiltonian, $\mathscr{H}_c$, for the bilayer structure. A convenient approach is to perform a Hubbard Stratonovich transformation [11]. $\mathscr{H}_c$ then emerges as a functional of two fields which can be identified, in the usual way, with the local magnetizations $\varphi_1(j_1,j_2)$ and $\varphi_2(j_1,j_2)$ of the first and second layer, respectively. After a naive continuum limit, the discrete in-layer coordinate (j_1,j_2) is replaced by the continuous variable $\boldsymbol{x}$. For generality, we take $\boldsymbol{x}$ to be d-dimensional. A particularly simple form of $\mathscr{H}_c$ results if the single-layer magnetizations $\varphi_1(\boldsymbol{x})$ and $\varphi_2(\boldsymbol{x})$ are expressed in terms of their sum $\Sigma(\boldsymbol{x}) \equiv \frac{1}{\sqrt{2}}(\varphi_1+\varphi_2)$ and difference $\Delta(\boldsymbol{x}) \equiv \frac{1}{\sqrt{2}}(\varphi_1-\varphi_2)$:

$$\mathscr{H}_c[\Sigma,\Delta] = \int d^d x \left\{ \frac{1}{2}(\nabla\Sigma)^2 + \frac{1}{2}(\nabla\Delta)^2 + \frac{1}{2}\Sigma^2\left[\frac{1}{2d\beta J_0} - 2a\right] + \frac{1}{2}\Delta^2\left[\frac{1}{2d\beta J_0} - 2b\right] + \frac{a}{3!}(3a-1)\Sigma^4 + \frac{b}{3!}(3b-1)\Delta^4 + ab\Sigma^2\Delta^2 \right\}. \tag{2}$$

Here, ∇ denotes the gradient in d-dimensional space, β is the inverse temperature, and

$$a(J) \equiv \frac{\exp(\beta J)}{2\cosh(\beta J)}, \qquad b(J) \equiv \frac{\exp(-\beta J)}{2\cosh(\beta J)} = a(-J)$$

are coefficients originating from the Hubbard Stratonovich transformation. For simplicity, Boltzmann's constant is set to 1. The gauge invariance of the microscopic model translates into the symmetry of $\mathscr{H}_c$ under exchanging Σ and Δ, and replacing J by

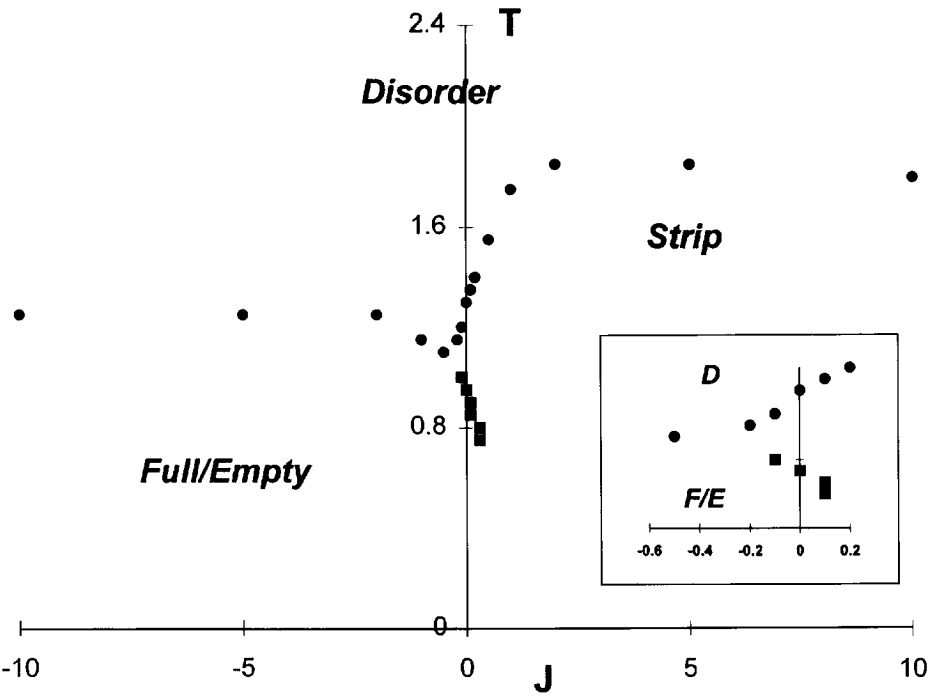

Fig. 1. Phase diagram for a bi-layer lattice gas at half filling, driven with $E = 25J_0$ [10]. The system size here is $L = 30$. T is given in units of the single-layer Onsager temperature: $0.5673J_0/k_B$; both J and E are measured in units of J_0. The disordered (D), strip (S), and full-empty (FE) phases are labeled. The D–S and D–FE transitions, denoted by •, are second order; while the S-FE transitions, shown by ■, are first order. The three lines join at a bicritical point.
Inset: Magnified view of the region near the bicritical point showing the presence of the S phase in $J < 0$ half-plane.

$-J$. It is easy to check that setting $J = 0$ reduces $\mathscr{H}_c$ to the sum of two completely decoupled ϕ^4-theories for φ_1 and φ_2, as expected.

Next, we show that the basic features of the zero-field phase diagram, namely, the nature of ordered states, the type of transitions, and the shape of the line of continuous transitions are correctly captured by a mean-field analysis of $\mathscr{H}_c$. Anticipating that spatial inhomogeneities are energetically costly, we will focus on homogeneous Σ, Δ. Seeking stable equilibrium phases, we determine the global minima of $\mathscr{H}_c$. Four types of extrema are present, at (a) $\Sigma = \Delta = 0$, (b) $\Sigma = 0$, $\Delta = \pm\Delta_0 \neq 0$, (c) $\Sigma = \pm\Sigma_0 \neq 0$, $\Delta = 0$, and finally (d) $\Sigma = \pm\Sigma_1 \neq 0$, $\Delta = \pm\Delta_1 \neq 0$. The explicit expressions for Σ_0, Δ_0, Σ_1 and Δ_1 are easily found from Eq. (2), but their precise values are not important here. Considering the matrix of second derivatives of $\mathscr{H}_c$, we find that (d) never corresponds to a minimum. In contrast, solution (a) is the only minimum at high temperatures, representing the disordered phase. Just below a critical temperature T_c, given implicitly by

$$T_c^{MF}(J) = 2dJ_0 \exp(\beta_c^{MF}|J|)/\cosh(\beta_c^{MF}|J|), \tag{3}$$

(a) becomes a saddle point, and the global minimum shifts to (b) for J negative, and to (c) if J is positive. Clearly, the former of these corresponds to the FE phase, while the latter represents the S phase. By virtue of Eq. (3), the phase boundary $T_c^{MF}(J)$ is symmetric in the cross-layer coupling, and monotonically increasing with its magnitude. The bicritical point is located at $J=0$. There, the two layers decouple, and $T_c^{MF}(0)=2dJ_0$ reduces to the mean-field critical temperature of a single-layer Ising model on the hypercubic lattice, as expected. Finally, it is particularly gratifying that the *ratio* $T_c^{MF}(J)/T_c^{MF}(0)$ approaches the observed limit for large $|J|$, namely, $T_c^{MF}(\pm\infty)/T_c^{MF}(0)=2$.

We now turn to the universal critical behavior described by $\mathscr{H}_c$. Focusing first on the D–S transition, i.e., $J>0$, we note that the ordering field here is Σ while Δ does not order. As a result, its coupling to Σ is irrelevant in the renormalization group sense, and the D–S transition is Ising-like, along the whole $J>0$ line. Similarly, for $J<0$, Δ and Σ simply exchange roles, so that the D–FE transitions also belong into the Ising universality class. However, the *dynamic* universality classes of the two lines differ, due to the conservation law: For the D–S transition, the order parameter Σ is conserved, so that the dynamic universal behavior is that of Model B, in the Halperin–Hohenberg scheme [14]. In contrast, the order parameter for the D–FE transition is Δ, which is not conserved. Since its coupling to the conserved, non-ordering Σ is quadratic in Σ, rather than linear, the dynamics here falls into the universality class of Model A.

In the presence of the drive, the analysis is complicated by several features. First, strong spatial anisotropies are generated since a specific lattice axis has been singled out. Second, the drive explicitly breaks the fluctuation–dissipation theorem so that the underlying Langevin equations are no longer Hamiltonian. Third, the transformation from (φ_1,φ_2) to (Σ,Δ) leads to novel, drive-dependent couplings in the field theory. Nevertheless, preliminary results suggest that the D–S transition belongs into the universality class of the single-layer driven lattice gas. Work is in progress to confirm this hypothesis, and to identify the universal properties of the D–FE transition [11].

Finally, we briefly review a first attempt towards developing a simple intuitive picture for the behavior of $T_c(J,E)$ [10]. It relies on the behavior of $G(\boldsymbol{x})$, the two-point correlation function, in the disordered phase. In the driven *single-layer* case, both its short- and long-range properties are affected by the drive: First, the *nearest-neighbor* correlations of the driven system are suppressed [15], compared to the equilibrium case, consistent with the picture that the drive acts as an extra noise in breaking bonds. This effect alone would lead to a *decrease* in $T_c(E)$, compared to $T_c(0)$. However, the long-range correlations in the driven system are vastly enhanced over those in the equilibrium system: while the latter decay exponentially, the former follow a power law $\propto 1/|\boldsymbol{x}|^d$, with an angle-dependent amplitude [6]. Specifically, correlations parallel (transverse) to E are positive (negative). Thus, the *long-range* correlations *promote* ordering into a single strip aligned with the drive, leading us to expect $T_c(E)$ to be *greater* than $T_c(0)$. Clearly, the effects of the short- and long-range parts of the transverse correlation function compete with one another, giving some insight as to why it is quite difficult

to predict the correct E dependence of $T_c(E)$. However, an extension of the above reasoning does "predict" the observed behavior in $d=3$ [16,10].

For the bi-layer system, the ratio $T_c(J,E)/T_c(J,0)$ is again determined by the competition between the short- and long-range properties of the transverse correlations. Now, an additional contribution due to the cross-layer correlations will be present. Since it is purely short-range in nature, we anticipate that it will tend to *lower* $T_c(J,E)$, due to the fact that E effectively reduces J. Of course, this effect may still be offset by the long-range in-plane correlations. However, if the effective cross-layer coupling is determined predominantly by E, while being only weakly dependent on J, then we expect to see a lowering of the *ratio* $T_c(J,E)/T_c(J,0)$ with increasing J. This hypothesis is indeed borne out by the simulation data. For small $J \geqslant 0$, the long-range part still dominates, leading to $T_c(J,E)/T_c(J,0) > 1$. Specifically, for $J=0$ we find $T_c(J,E)/T_c(J,0) \simeq 1.3$, comparable to the single-layer case, since the cross-layer correlations vanish here. In fact, the long-range part continues to dominate in a small region of *negative* J, so that the S-phase prevails there also. Thus, the bicritical point, together with its trailing first-order line, is "driven" into the $J<0$ half-plane (inset, Fig. 1). Returning to positive J, the ratio $T_c(J,E)/T_c(J,0)$ is indeed observed to decrease with increasing J. For $J \geqslant 5$, it even drops *below unity*, signalling that the short-range correlations dominate the behavior of $T_c(J,E)$ in this regime!

For large but *negative* J, we expect strong negative correlations across the layers, so that the system orders into the FE phase in equilibrium. Our data indicate that this process still dominates under the drive, so that the low-temperature phase of the driven system is also FE. However, the FE phase tends to be suppressed by *both* the short- and long-range parts of the correlations, the former effectively lowering $|J|$, the latter favoring an S phase. Thus, the critical temperature should be lower than its equilibrium counterpart. Further, in contrast to the $J>0$ case, the two effects cooperate rather than compete, so that the $J<0$ branch of $T_c(J,E)$ is significantly lower than the $J>0$ branch.

In summary, our study of an interacting bi-layer driven lattice gas provides new insight into the nature of ordering in driven systems. In an extended T–E–J phase space, we have shown that there are three phases, one disordered (D) and two ordered ones, namely, a strip (S) and a full-empty (FE) phase. The equilibrium phase diagram consists of two second-order lines, symmetric in J, joining at a bicritical point $(T_c(0), J=0)$ from which a first-order line stretches to $(T=0, J=0)$. The emergence of the ordered phases, the shape of the transition lines, and the universal behavior of the system near the continuous transitions can be easily understood. When driven, the phase diagram is modified, in a nontrivial way, by the competition between short- and long-range properties of the correlations. Specifically, the presence of large negative transverse correlations favors the strip phase, so that, for small $|J|$, the region associated with this phase is larger than in equilibrium. The associated shift of bicritical point and first-order line places the two $J=0$ transitions [9] into a wider, more comprehensible context. The second-order transition temperature, as a function of J and E, appears to be determined by a subtle interplay of the competing short- and long-range

components of the transverse correlation function. This hypothesis is currently being tested on a variety of other driven systems, with the hope of developing better intuitive insight into the qualitative structure of non-equilibrium phase diagrams.

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