Phase Transitions in Driven Bilayer Systems: A Monte Carlo Study

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We investigate the phase diagram of a system with two layers of an Ising lattice gas at half filling. In addition to the usual intralayer nearest neighbor attractive interaction, there is an interlayer potential J. Under equilibrium conditions, the phase diagram is symmetric under $J \to -J$, though the ground states are different. The effects of imposing a uniform external drive, studied by simulation techniques, are dramatic. The mechanisms responsible for such behavior are discussed. [S0031-9007(96)00614-X]

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Over a decade ago, motivated by the physics of fast ionic conductors, Katz *et al.* [1] introduced a simple modification to the well known Ising lattice gas [2], so that *nonequilibrium* steady states may be studied. The dynamics of this model consists of particle hopping, or Kawasaki exchange [3], controlled by the usual Ising Hamiltonian and a thermal bath at temperature T, as well as a bias in one direction so as to describe the effect of a uniform, dc "electric" field E, acting on the "charged" particles. In the ensuing years, many unexpected properties have been discovered in the prototype model and numerous of its variants, and, by now, some are well understood [4]. On the other hand, a few of the surprising results observed in Ref. [1] remain unexplained. An example is the basic question: Why should the critical temperature, $T_c(E)$, increase with E, saturating at about 40% above the Onsager temperature as $E \to \infty$ [5]? Indeed, one might have predicted a *lowering* of T_c, since large fields should overwhelm the nearest neighbor coupling whenever hops along the field are attempted, so that the system is effectively subjected to an extra noise. Given simulation data and a better understanding of other phenomena displayed by this system, simple arguments in favor of an *increased* T_c emerged. However, to date, there is still no intuitive picture which guides us to the correct behavior. One motivation of our study is to explore similar systems, in order to test which type of argument is successful in "predicting" the qualitative behavior of the novel phase diagram.

At an entirely different level, this work is motivated by interesting properties in driven multilayered structures, observed in both physical systems [6] and Monte Carlo simulations [7,8]. In the former, the process of intercalation, where foreign atoms or molecules diffuse into a layered host material, is well suited for modeling by driven, layered lattice gases [8]. On the simulation front, the effects of particle transfer between *two decoupled* Ising systems, subject to a global conservation law, turn out to be quite intriguing: *two* transitions were found [7]. As T is lowered, the disordered (D) phase transforms into a state with strips in both layers, reminiscent of two entirely unrelated, yet aligned, single-layer driven systems. We will refer to this state as the strip phase (S). As T is lowered further, a first order transition occurs. The ordered state now resembles the equilibrium case, displaying homogeneous, opposite magnetization on the two planes. In the lattice gas picture, the planes are mainly full or empty, so that this state will be labeled by FE. Why there should be *two* transitions was not well understood. This impasse, as well as the generic microscopics of intercalated compounds, motivates our study of a bilayer driven system with *interlayer* interactions. The remainder of this Letter is devoted to a brief description of our model and the simulation results. A consistent, intuitive picture emerges. We conclude with suggesting possible analytic approaches and other tests of this picture.

Our system consists of two fully periodic $L \times L$ square lattices, arranged in a bilayer structure. The sites are labeled by (j_1, j_2, j_3), with $j_1, j_2 = 1, \ldots, L$ and $j_3 = 1, 2$. Each may be empty or occupied by a particle, so that a configuration of the system is specified by the set of occupation numbers $\{n(j_1, j_2, j_3)\}$, where $n = 0$ or 1. Alternatively, we may use the spin language: $s \equiv 2n - 1 = \pm 1$. For simplicity, we study only half-filled systems, i.e., $\sum n = L^2$ or $\sum s = 0$. Next, we endow the spins with nearest neighbor interactions, so that 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, while n and n'' differ only by the layer index. Thus, the first sum represents the usual two-dimensional Ising model with coupling J_0, and the second sum takes into account the interactions across the layers. Our study is restricted to positive J_0, with several values of J/J_0 in the range $[-10, 10]$. The choice of negative J's is motivated by the physics of intercalated materials [6,8]. To simulate equilibrium systems coupled to a thermal bath at temperature T, we use spin-exchange (Kawasaki) [3] dynamics with the usual Metropolis rate [9]. So, particles are allowed to hop to nearest neighbor holes with probability $\min\{1, \exp(-\Delta\mathcal{H}/k_B T)\}$, where $\Delta\mathcal{H}$ is the change in energy due to the hop. Since this dynamics becomes very slow for large $|J|$, we exploit spin flip (Glauber) dynamics to explore the transitions in

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these regions, rather than a more complex algorithm [10]. Finally, to drive this system into nonequilibrium steady states, we incorporate the "electric" field (aligned with the 1 axis) in the standard way, i.e., by adding $\pm E$ to $\Delta\mathcal{H}$ for hops against or along the field [1,4]. The goal of this study is to map out the phase diagram in the T-J-E space. In the following, the main results are reported, while details will be published elsewhere [11]. Here, T is given in units of the single layer Onsager temperature, $0.5673 J_0/k_B$; both J and E are given in units of J_0.

The systematic part of our runs involves lattices with $L = 12$ and 30 and 100×10^3 Monte Carlo steps (MCS) per site. Typically, these runs are set at fixed J, E, and T, starting with two types of initial conditions, ordered and random. Discarding the first 10×10^3 MCS, measurements are then taken every 200 MCS. To explore first order transitions, we look for hysteresis by sweeping in both T and J, where T (J) is raised or lowered in steps of 0.05 (0.02) after 100×10^3 MCS. Apart from these systematic studies, we have performed very long runs (up to 5×10^6 MCS) at a few points in parameter space, mainly to explore metastability. We have also investigated a few systems with various sizes up to 100×100, in order to be more confident about the existence of certain steady states in the thermodynamic limit. As order parameters, we have chosen the appropriate structure factors [1,4]. In the S and FE phases, they are $S(0,1,0)$ and $S(0,0,1)$, respectively, where $S(l_1, l_2, l_3) \equiv \langle |\tilde{n}(l_1, l_2, l_3)|^2 \rangle$ and

$$\tilde{n}(l_1, l_2, l_3) \equiv \frac{1}{2L^2} \sum n(j_1, j_2, j_3) e^{2\pi i [(j_1 l_1 + j_2 l_2)/L + j_3 l_3/2]}.$$

Of course, in practice, the $\langle\,\rangle$'s are time averages, taken over the run. Occasionally, when simple averaging produces highly irregular results, typically near first order transitions, we resort to time traces of $|\tilde{n}|^2$. These reveal the system being "caught" for an extended period of time (say, 30×10^3 MCS) in a metastable state, before settling into the stable, steady state for the rest of the run. In these instances, we reperform the average using only the last part of the data. In all cases, the resultant values fall within the expected statistical variations.

To identify the second order transitions, we consider the fluctuations of $|\tilde{n}|^2$ as a function of T, with fixed J and E. The critical temperature is then associated with the peak of this function. We estimate that this method is accurate to about 5%, the error coming from both finite size effects and statistics. On the other hand, for first order transitions, $T_c(J, E)$ or $J_c(T, E)$ is identified with the midpoint between the values where the order parameter jumps in a hysteresis loop. We simply use these jump values to assign the error, which is presumably overestimated. More accurate estimates of the phase boundaries are clearly possible, using longer runs on larger systems and more sophisticated techniques, such as finite size scaling. However, high precision data are not necessary for us to reach some conclusions about the nature of the phase transitions in our systems. In this spirit, we discuss our findings and their implications.

In the equilibrium case, a gauge transformation relates the $J > 0$ system to the $J < 0$ one. Thus, the phase diagram in the T-J plane is symmetric (Fig. 1). Note that, in the limit of $J \to \infty$, the bilayer structure is irrelevant and the system reduces to a single $d = 2$ Ising model with coupling $2J_0$. Our simulations certainly confirm that $T_c(J = \pm 10)$ is $2T_c(0)$, within the errors. Given the conservation law, the two ground states are different, however. For $J > 0$ ($J < 0$), the system orders into an S (FE) state, to minimize interface free energy. Thus, there is actually a line of first order transitions on the $J = 0$ axis, between $T = 0$ and $T_c(0)$. In this sense, the junction of the three lines is a bicritical point. Since $L < \infty$ systems are unavoidable in a Monte Carlo study, we must be aware of finite size effects and expect, e.g., the first order transitions to occur at small, $O(1/L)$, positive J values.

When the drive is turned on, the most prominent new features are (i) the *lowering* of the critical temperature for large $|J|$ and (ii) the shift of the bicritical point to *higher* values of T and negative J. The loss of symmetry in the phase diagram (Fig. 2) is not surprising, since the drive violates Ising symmetry. On the other hand, given that $T_c(E)$ is greater than $T_c(0)$ in the single-layer case, it is quite unexpected that $T_c(|J| \gg 1, E \gg 1)$ is *smaller* than its equilibrium counterpart.

Perhaps most unexpected is the presence of a finite, triangular region (inset, Fig. 2) in the phase diagram, in which an S phase is stable even for *negative*, albeit small, J. Since similar spins lie on top of each other here, such a phase could not exist if either energy or entropy were to play dominant roles in determining the steady state. It

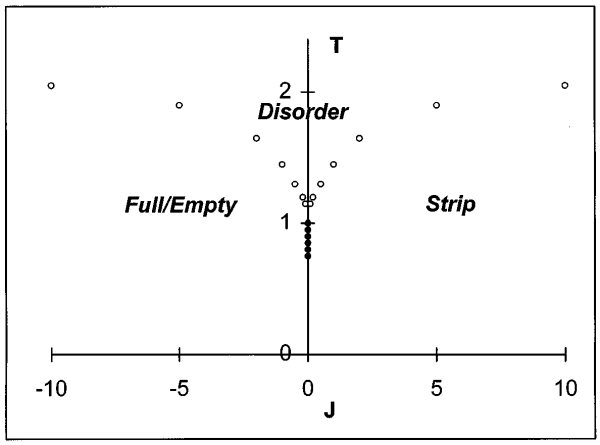

FIG. 1. Phase diagram for an equilibrium bilayer lattice gas at half filling. The disordered (D), strip (S), and full-empty (FE) phases are labeled. The D-S and D-FE transitions, denoted by ○, are second order; while the S-FE transitions, shown by ●, are first order. The three lines join at a bicritical point.

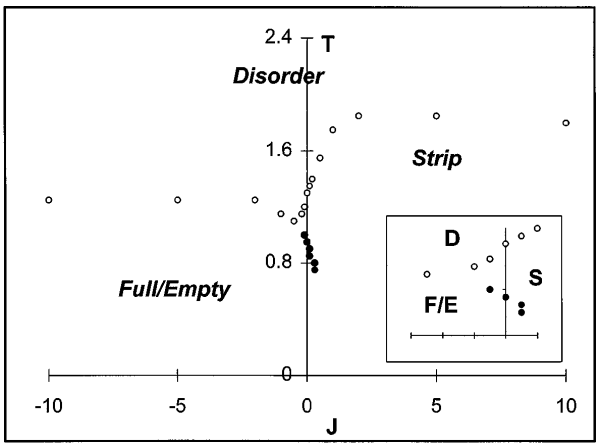

FIG. 2. Phase diagram for a bilayer lattice gas at half filling, driven with $E = 25J_0$. The symbols and the scale used are the same as in Fig. 1. Inset: Magnified view of the region near the bicritical point showing the presence of the S phase in $J < 0$ half plane.

is thus natural to ask whether its presence might simply be a finite size effect. To provide a partial answer, we carried out simulations using $L = 12, 16, 24, 30$, and 100, with $J = -0.1$, $E = 25$, and $T \in [1.00, 1.20]$. In all cases, the S phase prevailed. Deferring details to a later publication [11], we conjecture that this region exists even in the thermodynamic limit.

In order to develop a simple intuitive picture for these phenomena, let us review the arguments which attempt to "predict" the field dependence of T_c in the driven single-layer case. To provide a wider context, we begin with highlighting the differences between the driven system and an equilibrium Ising model, exhibited in $G(\vec{x})$, the two-point correlations. Even in the disordered phase, both the small and large $|\vec{x}|$ behaviors are affected by the drive. First, the *nearest neighbor* correlations of the driven system are somewhat suppressed [12], consistent with the picture that the drive acts as an extra noise in breaking bonds, so that the effective strength of the nearest neighbor coupling is reduced. This observation alone would lead to a *decreasing* $T_c(E)$. However, these properties stand in stark contrast to the large $|\vec{x}|$ behavior, where G falls off as $1/|\vec{x}|^d$, with an amplitude that depends on the angle θ between $\vec{x}$ and the direction of the drive [13]. Specifically, the amplitude changes sign as θ increases from 0 to $\pi/2$, such that correlations parallel (transverse) to E are positive (negative). In either case, the *magnitude* of G is greatly enhanced over its exponentially decaying counterpart in equilibrium. The enhanced negative correlations at large transverse $\vec{x}$ are expected to help "push" the particles together into a strip aligned with the drive. The positive longitudinal correlations should also promote this ordering process, so that one would 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, shedding some light on the origin of the contradictory expectations concerning the E dependence of $T_c(E)$. To predict which is the dominant effect will be difficult, of course. In the prototype model, $T_c(E)$ is observed to increase with E in both $d = 2$ and 3, indicating that the long-range effects "win." However, the two cases differ in a subtle way. In $d = 3$, there is an *additional* competition among the negative long-range correlations, since there are *two* transverse directions here. Long-range order (i.e., positive correlations) must develop in *one* of these directions as T_c is approached. Thus, we might expect a less significant rise in $T_c(E)$ in $d = 3$. Intriguingly, this is indeed the case: $T_c(E)$ saturates at only 7% higher than the equilibrium $T_c(0)$ [14].

Turning to the bilayer case, we need to take into account the effect of cross-layer correlations on $T_c(J, E)$. Focusing on $J = 0$, where there are no additional short-range effects, we are led to $T_c(0, E)/T_c(0, 0) > 1$. Indeed, this ratio is comparable to that in the single-layer case. As for the lower transition, which is first order in nature, we refrain from using these arguments, since the role played by long-range correlations in a first order transition is unclear. Instead, we will return to examine this transition in the context of a larger perspective.

Next, we consider the effect of having a positive J. Without the drive, $T_c(J, 0)$ is, of course, enhanced over $T_c(0, 0)$. With $E \neq 0$, $T_c(J, E)/T_c(J, 0)$ is again determined by the competition of the short- and long-range properties of the transverse correlations. Evidently, for small J, the long-range part still dominates, so that this ratio is greater than unity. At the other extreme, if $J \gg J_0$, the presence of E effectively lowers J, according to our argument that E acts as an extra noise which breaks even very strong nearest neighbor bonds. A lower effective J naturally leads to a lower T_c. Thus, we would "predict" that $T_c(J, E)/T_c(J, 0)$ could decrease considerably as J increases. In fact, the simulations show that this ratio drops *below unity* for $J \geq 5$. The interplay of the competing effects is so subtle that either can dominate, in different regions of the phase diagram.

Next, for large $J < 0$, we expect strong negative correlations across the layers, so that the system orders into the FE phase in equilibrium. That the low temperature phase of the driven system is also FE indicates an ordering process dominated by J. However, under the drive, *both* the short- and long-range parts of the correlations tend to suppress the FE phase: 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 (see Fig. 2). We should add that a reasonable expectation for two driven layers with $J < 0$ would be a phase with strips in each layer, but *staggered.* The absence of such a state

may be due to energetics. The transverse long-range anticorrelations presumably also act across layers, so that the price of interfaces is not compensated for by the system having staggered strips instead of being in an FE state.

Finally, the presence of the S phase, in the triangular region $J \leq 0$ (inset, Fig. 2), can also be understood qualitatively. Since long-range negative correlations transverse to E supposedly dominate the ordering process for $J \geq 0$, it is not too surprising that this mechanism continues to be effective for a small region of negative J. As a consequence, the bicritical point together with its trailing first order line are "driven" to the $J < 0$ half plane. Evidently, however, the relative importance of E and J is easily reversed, so that the system orders into the FE pattern if J is sufficiently negative. Similarly, for low temperatures at $J = 0$, energetics appear to outweigh the nonequilibrium-induced effects so that the FE phase prevails here also. At present, it is unclear if, like the equilibrium case, finite size effects account for the presence of the FE phase in a small $J > 0$ region.

To conclude, our study of an *interacting* bilayer driven lattice gas provides new insight into the nature of ordering in driven systems. It is a nontrivial extension of a previous investigation [7] which used only decoupled layers and simply reported the existence of two transitions into two different ordered states. We have shown that, in an extended T-E-J phase space, these two states are generically present and, in equilibrium cases, can be easily understood. When driven, the negative long-range correlation favors the strip phase so that, for small $|J|$, the region associated with this phase is larger than in equilibrium. On the other hand, for $|J| \gg J_0$, the decorrelating effects of the drive on the nearest neighbors are so disruptive that, compared to the equilibrium model, T_c is lower. The overall result is a shift of the bicritical point, and a portion of its associated first order line, to the $J < 0$ region. Thus, the two phase transitions found in the $J = 0$ case [7] are placed into a comprehensible context. The behavior of the second order transition temperature, as a function of J and E, is determined by a subtle interplay of the competing short- and long-range components of the transverse correlation function. Work is in progress to test this hypothesis further, by studying a single-layer driven lattice gas with anisotropic interactions. Our prediction is that $T_c(E)$ will be higher (lower) if the drive is aligned with the stronger (weaker) bonds.

On the analytic front, we have formulated continuum field theoretic approaches, for both the equilibrium and the driven cases [4]. In the former case, we obtained [11] the (mean-field) critical temperature $T_c^{\mathrm{MF}}(J)$ and $T_c^{\mathrm{MF}}(\infty)/T_c^{\mathrm{MF}}(0)$ is found to be 2. The actual $T_c(J)$ is modified by fluctuations, of course. With this starting point, we hope to find the *additional* effect due to the drive, at least qualitatively, as another way to confirm the intuitive picture presented above. Since these approaches are best suited for the study of universal properties, our goal is to compute various quantities at, e.g., the bicritical point, and to make quantitative comparisons with extensive finite size scaling Monte Carlo studies.

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