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Krypton on graphite and the striped helical Potts model

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A generalization of the helical Potts model, with two species of domain wall due to explicit triaxial helical symmetry breaking, is studied via position-space renormalization-group methods and is discovered to exhibit striped, as well as hexagonal, phases. The disordering transition of the commensurate ferromagnetic phase belongs to the symmetric Potts universality class. No evidence is found for a chiral melting transition. Commensurate-incommensurate phase diagrams for oversaturated krypton on graphite are constructed.

There are many two-dimensional systems in which adsorbed atoms form a solid in registry with the underlying substrate. The melting of this commensurate solid, via a change in temperature or ambient vapor pressure, has been extensively studied. Of immediate concern here is the commensurate melting transition of physisorbed krypton on graphite. At densities less than a monolayer, the krypton atoms select one of the three equivalent triangular sublattices of adsorption sites to form an ordered solid. The permutation symmetry of the three sublattices leads to a disordering transition in the three-state Potts universality class, as pointed out by Alexander[1] and others,[2,3] and confirmed by experiments.[4] At densities exceeding a monolayer, extra atoms can be accommodated into the first layer via dense domain walls[5,6] separating the commensurate regions. Kadar and Berker[7] pointed out that there are two types of domain walls (heavy and superheavy) with different energies and hence at these densities the system must be described by a "helical" Potts model.[8,9] An explicit calculation[7] revealed a transition to a dense domain-wall liquid[6,10] that was still in the three-state Potts universality class.

Motivated by the experiments of Moncton *et al.*,[6] Huse and Fisher[11] suggested that although helicity is irrelevant at the Potts transition in triaxial systems, sufficiently strong triaxial symmetry breaking terms could incur a crossover to "triaxial chiral" melting. This new transition would be governed by an entirely different fixed point and belong to an altogether new universality class of asymmetric chiral transitions. Huse and Fisher[12] suggest that this transition was not observed in the calculation of Kadar and Berker[7] because in the latter treatment all helicity, which results from the difference between heavy and superheavy wall energies, ends up directly associated with the wall crossings. In this Rapid Communication, we present and analyze the striped helical Potts model on the triangular lattice, which overcomes this problem and incorporates all the basic physics of two species of domain walls. Hence, within the limitations of a lattice approximation, the energetics of different microdomain walls, their crossings and dislocations[10] are faithfully represented. The Hamiltonian allows for both hexagonal and striped phases. Our position-space renormalization-group calculation, in addition to recovering the fixed points that govern the disordering transitions of the hexagonal phases, also uncovers for the first time fixed points that control the disordering transitions of striped phases. We find no evidence of the speculated new multicritical point of Huse and Fisher[11] (separating Potts and chiral melting transitions). At the Potts fixed point, triaxial helicity is irrelevant as expected, and the disordering of the commensurate phase, despite the presence of triaxial symmetry breaking, is *always* characterized by three-state Potts exponents. Phase diagrams for krypton on graphite in the temperature-chemical potential plane are obtained using a suitably modified version of previous parametrizations. Connection with the recent work of Caflisch, Berker, and Kardar[13] permits conversion to temperature-pressure space and a more direct comparison with experimental results.

As in a previous calculation[7] we subdivide the graphite adsorption sites into hexagonal patches of size l (in units of commensurate adatom separation). Each patch represents a microdomain of krypton atoms adsorbed in one of the three possible sublattices a, b, or c. Neighboring patches either occupy the same sublattice (thus forming part of a larger domain), or different sublattices in which case they are separated by heavy or superheavy domain walls. A vertical heavy domain wall, for example, separates a and b domains and contains an extra one-third adsorbate column, whereas a superheavy domain wall, while separating a and c domains, accommodates twice as many adsorbate atoms (see Fig. 1 of Ref. 7). We assign an energy per unit length of $(\mu/3-J)$ for a heavy wall, and $2(\mu/3-J)$ for a superheavy wall, where μ is the adsorbate chemical potential and $J>0$ is an effective nearest-neighbor repulsion. The centers of the hexagonal patches form a triangular lattice, subdivided into three sublattices A, B, and C. The Hamiltonian of the striped helical Potts model is given by

$$-\beta\mathcal{H}=\sum_t \epsilon(S_t^A,S_t^B,S_t^C) \ ,$$

where the summation runs over all triplets t of the triangular lattice, and $S_t^i=a$, b, or c (depending on the patch occupation) is the triplet spin on sublattice i. The possible triplet energies are[7]

$$\epsilon(a,a,a)=0\equiv F \ ,$$

$$\epsilon(a,b,c)=\frac{3l}{2}\left(\frac{\mu}{3}-J\right)-\tfrac{3}{2}J\equiv P \ ,$$

$$\epsilon(a,c,b)=\frac{3l}{2}\left(\frac{2\mu}{3}-2J\right)-\tfrac{3}{2}J\equiv N \ ,$$

$$\epsilon(a,a,b)=\frac{\sqrt{3}l}{2}\left(\frac{2\mu}{3}-2J\right)\equiv U \ ,$$

$$\epsilon(b,b,a)=\frac{\sqrt{3}l}{2}\left(\frac{\mu}{3}-J\right)\equiv D \ .$$

A repulsive energy of $-(\frac{3}{2})J$ is included for both heavy and superheavy wall crossings, and a patch size $l=5$ (related to wall width[5]) is selected. Note that the above triplet energies are for up-pointing triplets—the last two expressions being interchanged for down-pointing triplets. By associating different triplet energies with the microdomain walls in the (a,a,b) and (b,b,a) configurations, we have remedied the objection of Huse and Fisher[12] to the earlier triplet helical Potts model[7] and thereby permitted the possibility of striped commensurate phases.

To determine the general topology of the phase diagram and calculate critical exponents for the above striped helical Potts model, we employ position-space renormalization-group methods in the finite cluster approximation. The three-cell cluster originally employed by Schick, Walker, and Wortis[14] is used. However, the simple majority rule projection operator[15] is no longer sufficient and a more complex projection operator is used. Our projection scheme maps a configuration of nine spins to a triplet representing the ground state requiring the least number of spin flips, and in case of a tie, gives equal weight to the equidistant states. In the four-dimensional parameter space we discovered 19 fixed points (sinks included), 15 of which were already discussed in the $U=D$ subspace,[7] and the new fixed points governing the heavy and superheavy wall striped phases for $U\neq D$. Each striped phase is ninefold degenerate: there is a trivial three-state Potts degeneracy, and a threefold degeneracy of the three equivalent directions (at 120° to each other) for the stripes. In our renormalization-group approximation the disordering of the striped phase is continuous with a thermal exponent $y_t=0.864$ (the true transition could well be first order). We found no evidence for a new fixed point governing a chiral melting transition as speculated by Huse and Fisher.[11,12] The ferromagnetic Potts fixed point appears in the $P=N$, $U=D$ subspace. Breaking the $P-N$ and $U-D$ equalities corresponds to introducing irrelevant triaxial symmetry-breaking terms with crossover exponents $\phi_{P\neq N}=-1.07$ and $\phi_{U\neq D}=-3.50$, respectively. The former agrees well with the estimate $-\frac{19}{18}$ obtained by scaling arguments.[12]

Translation of our results into the variables μ and J of the microscopic model yields the phase diagrams given in Fig. 1. The energetics of microdomain walls manifests itself most distinctly in the region $\mu\approx 3J$ where both heavy and superheavy wall energies vanish. As before, the commensurate phase appears as the ferromagnetic phase of the striped helical Potts model, while the incommensurate phase at large chemical potentials ($\mu >> 3J$) corresponds to the Potts negative helicity phase (a hexagonal net of superheavy walls). The phase diagram with both heavy and superheavy wall crossings repulsive is indicated in Fig. 1(a). There is a small region of the phase diagram occupied by a striped phase of *superheavy domain walls* (possibly representing a different incommensurate phase of the krypton on graphite system). The phase diagram with only the superheavy wall crossing made attractive is given in Fig. 1(b). There is now a direct first-order transition between the commensurate and incommensurate phases, and this situation, as discussed in Ref. 7, is relevant to coadsorption of krypton and deuterium on graphite.[16] We have also included phase diagrams with only the heavy wall crossing attractive [Fig. 1(c)], and with both heavy and superheavy wall crossings attractive [Fig. 1(d)]. There can now be an additional phase composed of a hexagonal net of *heavy* domain walls. The striped phase of superheavy domain walls appears only when both types of wall crossing are repulsive. Figures 1(a)

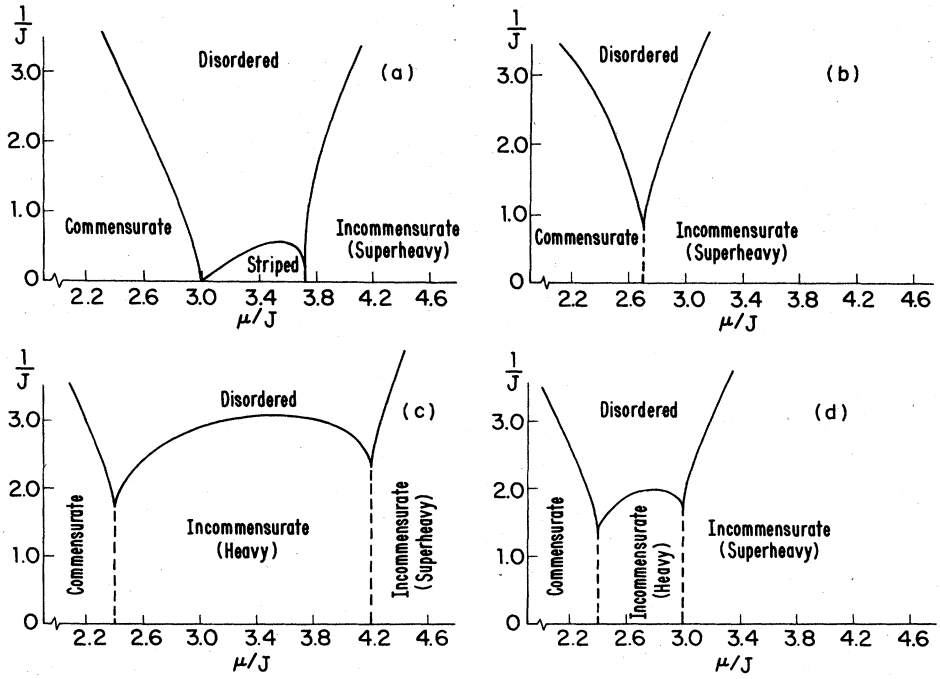

FIG. 1. Overlayer phase diagrams with (a) repulsive heavy and superheavy wall crossings, (b) repulsive heavy and attractive superheavy wall crossings, (c) attractive heavy and repulsive superheavy wall crossings, (d) attractive heavy and superheavy wall crossings.

and 1(b) are in agreement with predictions by Bak, Mukamel, Villain, and Wentowska[5] concerning the nature of phases and order of phase diagrams when only one type of domain wall is included. Our results, therefore, generalize their conclusions to two species of domain wall. It would be most interesting for experiments to overcome the challenge of probing the low-pressure-temperature regions where the new phases may appear.

To make better contact with experiment we must relate the lattice-gas chemical potential μ of the microscopic model to the actual ambient pressure p of the krypton on graphite system. The recent work of Caflisch *et al.*[13] indicates that μ can be related to pressure p and temperature T through

$$\mu = \ln\left[\frac{p\lambda^3}{kT}\right] + \sum_{\alpha=x,y,z} \ln\left[\tfrac{1}{2}\,\mathrm{csch}\left[\frac{\hbar\omega_\alpha}{kT}\right]\right] + \frac{E}{kT} \ ,$$

where $\lambda = h/\sqrt{2\pi mkT}$ is the thermal wavelength, ω_α are characteristic frequencies [$(\hbar\omega_x)/k = (\hbar\omega_y)/k = 19.8$ K, $(\hbar\omega_z)/k = 65.1$ K], and $E/k \simeq 1461$ K is the energy gain by krypton atoms upon adsorption on graphite. Also, the coupling J is estimated[13] to be 83 K/T. Using these results the phase diagrams of Figs. 1(a) and 1(b) are translated into the pressure-temperature diagrams given in Figs. 2(a) and 2(b), respectively. As is apparent from the former figure, the striped phase appears at very low pressures making it experimentally inaccessible. At higher temperatures ($T \sim 130$ K) the commensurate phase melts into a dilute fluid. It has been shown both experimentally[17] and theoretically[13] that the dilute fluid phase and the dense domain-wall liquid phase are continuously connected.

To elucidate the chiral melting hypothesis, Huse and Fisher[12] have proposed a particularly simple model to study the effects of triaxial helicity breaking[7] on the three-state Potts transition. With spins at each site j of a triangular lattice taking on three possible complex values $\psi_j = 1$, $\exp(\pm 2\pi i/3)$, their Hamiltonian is [Ref. 12, Eq. (2.3)]

$$-\beta\mathscr{H} = J\sum_{\langle ij\rangle} \mathrm{Re}(\psi_i^*\psi_j) - \sum_{\langle i,j,k\rangle} [J_3\,\mathrm{Re}(\psi_i\psi_j\psi_k) + \Delta\epsilon_{ijk}\,\mathrm{Im}(\psi_i\psi_j\psi_k)] \ ,$$

where $\epsilon_{ijk} = +1(-1)$ for up(down) pointing triplets, and the sums run over all nearest-neighbor pairs and elementary triplets, respectively. This model Hamiltonian can be mapped onto the $P = N$ subspace of our striped helical Potts model. Without loss of generality we choose $J_3 = -3J/4$, which corresponds to setting the zero of the energy scale for P,N triplets. The phase diagram in the remaining parameters J and Δ is exhibited in Fig. 3. For all values of Δ, the triaxial helicity term, the ferromagnetic disordering transition is in the Potts universality class and no chiral melting transition is observed. This result, together with Baxter's exact solution of the hard hexagon problem,[18] provides further evidence for the absence of a chiral melting transition in triangular systems. Indeed, Huse[19] has made the observation that simple lattice-gas models may possess a momentum-space symmetry not shared by the real krypton on graphite system, and thus may not exhibit the chiral melting transition. (A possibly asymmetic experimental structure factor is so far the only evidence for such a transition.) On the other hand, lattice-gas models have so far proved most successful in describing adsorbed gas systems such as krypton on graphite.[3,7,13]

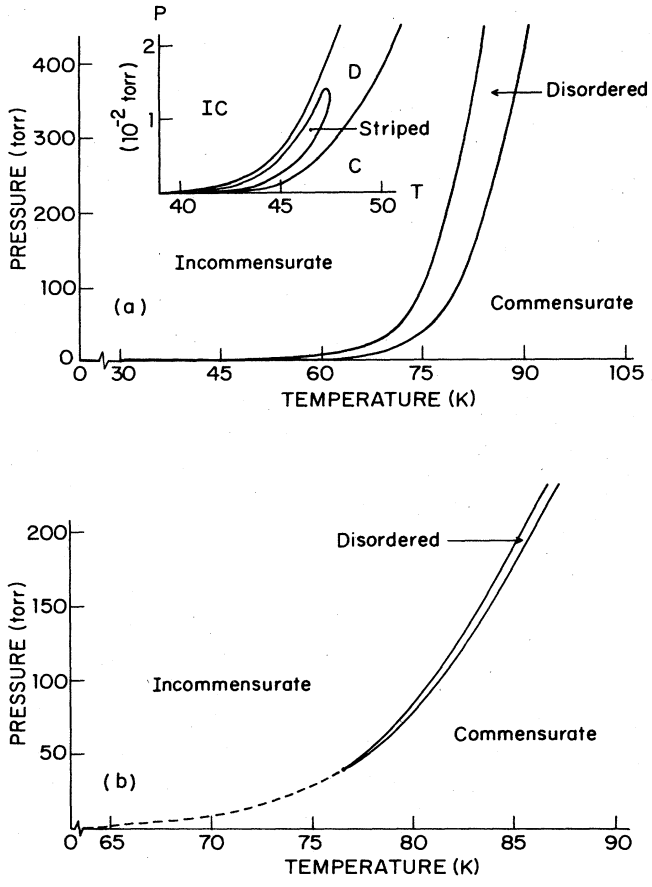

FIG. 2. Pressure-temperature overlayer phase diagrams with (a) repulsive heavy and superheavy wall crossings—inset shows the striped phase, (b) repulsive heavy and attractive superheavy wall crossings.

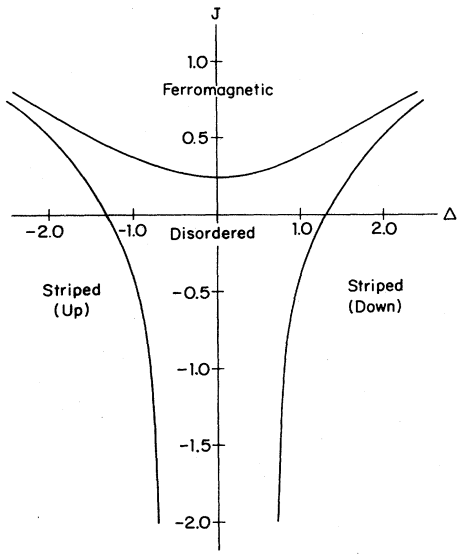

FIG. 3. Phase diagram for the model proposed by Fisher and Huse. The disordering transition of the ferromagnetic phase belongs to the symmetric Potts universality class.

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