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Tricritical Spinodal Decomposition in a Two-Dimensional Metamagnet

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Results are presented of a Monte Carlo study of the spinodal decomposition of a two-dimensional metamagnet, quenched from an initial high-temperature disordered state to below its tricritical point. It is found that the structure factors for both the order parameter and magnetization exhibit instability as manifested via growing peaks. The order-parameter structure factor $S(k, t)$ is shown to exhibit an early-time scaling of the form $t^{-2x}S(k, t) = F(kt^x)$, with $x \simeq 0.35$, where k is the wave number and t the time.

The dynamics of first-order phase transitions, which includes the evolution of metastable and unstable states, is currently a subject of considerable experimental and theoretical interest. Most theoretical attention has been given so far to kinetic Ising models of binary alloys quenched below a critical point, in which the order parameter is conserved (spin-exchange dynamics). Quite accurate descriptions of the time evolution for such models have been obtained via Monte Carlo (M. C.) studies[1] which have shown, among other things, that the distinction between the spinodal decomposition of unstable states and the decay of metastable states is not as sharp as was originally predicted. Quite recently these methods have also shown that the structure factor for a kinetic antiferromagnetic Ising model with spin-exchange dynamics exhibits a striking scaling behavior in the early-time, spinodal-decomposition region.[2] The scaling length is a time-dependent domain size which has been successfully predicted[3,4] to grow like $t^{1/2}$.

In this paper we report the first M. C. studies of tricritical spinodal decomposition for a two-dimensional metamagnet whose dynamics is assumed to be given by Kawasaki spin exchange.[1] The dynamics of this model is intrinsically more difficult than that of binary alloys or Ising antiferromagnets, because of the existence of a non-

conserved long-range–order parameter (the sublattice magnetization) and the conserved magnetization, whose time evolution is coupled. Our motivation for this study is threefold: Alloys such as Fe-Al are known to have tricritical points[3] and exhibit spinodal decomposition. Also, Hohenberg and Nelson[5] have recently predicted, on the basis of a linearized Cahn-Hilliard theory, that ^{3}He-^{4}He should exhibit an induced instability in a conserved variable (the ^{3}He concentration) in addition to the natural instability which the nonconserved (superfluid) order parameter should show. Finally, it is natural to examine how the qualitative features of spinodal decomposition differ between a critical and tricritical quench. Our model is sufficiently simple to analyze by M. C. methods, and realistic enough to provide some insight into tricritical spinodal decomposition. It should serve as the basis for further studies such as ^{3}He-^{4}He mixtures in which one also needs to consider the effects of hydrodynamics.

Before giving the details of our results, we summarize our major conclusions. First, the magnetization structure factor exhibits an induced instability (as qualitatively predicted by Hohenberg and Nelson for ^{3}He-^{4}He) through a peak which grows with time and whose peak position occurs at finite wave number k, which decreases

with time, much like that of a conserved binary alloy quenched below its critical point. Second, the structure factor for the sublattice magnetization peaks at the smallest allowed wave number, which in the bulk limit would be at $k = 0$. Third, we find that the sublattice magnetization structure factor exhibits an early-time scaling form quite similar to that found by Phani et al.,[2] but with a different characteristic exponent.

Our model for the metamagnet consists of a square lattice with a spin at each lattice point r, with a Hamiltonian

$$\mathcal{H} = J \sum_{n.n.} \sigma(\vec{r})\sigma(\vec{r}')$$
$$- \alpha J \sum_{n.n.n.} \sigma(\vec{r})\sigma(\vec{r}'') + H \sum_{r} \sigma(\vec{r}), \qquad (1)$$

where the sums are over the nearest and next-nearest neighbors on a square lattice. The tricritical point for this model is known[6] rather accurately for $\alpha = 0.5$ to be $T_t \simeq 1.30J$, $H_t \simeq 3.95J$, and $M_t \simeq 0.45$, where M_t is the tricritical magnetization. We have studied the dynamical properties of this system with $\alpha = 0.5$ for a $N = 60 \times 60$ lattice with periodic boundary conditions, assuming a Kawasaki spin-exchange model in which the transition probability for the ith and jth spins to flip is given by

$$W = \tau^{-1} e^{-\beta \Delta E}/(1 + e^{-\beta \Delta E}), \qquad (2)$$

where ΔE is the change in the energy of the configuration resulting from exchanging the randomly chosen pair of opposite spins. It is to be noted that both nearest- and next-nearest-neighbor spins are allowed to exchange. The time scales for these two processes are chosen for simplicity to be the same and equal to τ.

We quench a state of magnetization $M = 0.5$ prepared at a very high temperature to a temperature $T = 0.8J$ below the tricritical temperature and observe the system evolve for 2800 M.C. steps. We next obtain the circular-averaged structure factors discussed below. In order to ensure an appropriate ensemble average, we average over six runs. In addition to obtaining the structure functions we also monitored the sublattice magnetization M_s defined as

$$M_s = N^{-1} \sum_{r} (-1)^{\vec{r}} \sigma(\vec{r}). \qquad (3)$$

Similar to Phani et al.,[2] we observe that in three of our runs $M_s(t)$ develops a nonzero value which increases steadily towards the appropriate equilibrium value whereas in the other three runs $M_s(t)$ remains near zero. It should be noted that because of the existence of the conserved variable

M, our time scales are greater than those for the pure antiferromagnet.

In Fig. 1 we display our results for the magnetic structure factor which in our M.C. calculation is given by

$$S_{MM}(k,t) = N^{-1}|\sum_{\vec{r}} e^{i\vec{k}\cdot\vec{r}} \sigma(\vec{r})|^2, \qquad (4)$$

where $|\vec{k}| = (2\pi/\sqrt{N})j$, $j = 1, 2, \ldots, 13$. This function develops a peak at early times, indicating the existence of unstable modes. Just as for the binary alloy, we parametrize the peak position $k_m(t)$ by $k_m(t) \sim t^{-a}$ and the peak height $S_{MM}(k_m(t),t) \sim t^b$. Our results are consistent with $a \simeq 0.21$ and $b \simeq 0.5$. We have also analyzed the behavior of the first moment $\bar{k}(t) = \sum kS_{MM}(k,t)/\sum S_{MM}(k,t) \sim t^{-a'}$ and have found $a' \simeq 0.21$, in good agreement with $k_m(t)$. The behavior of the order-parameter structure function

$$S_{M_s M_s}(k,t) = N^{-1}|\sum_{\vec{r}} e^{i\vec{k}\cdot\vec{r}}(-1)^{\vec{r}} \sigma(\vec{r})|^2 \qquad (5)$$

is qualitatively different, since the peak height always occurs at the smallest allowed value of $k_1 = 2\pi/\sqrt{N} = \pi/30$. In the thermodynamic limit this peak would occur at $k = 0$, so that we have also included in Fig. 2 the behavior at these values. In addition to computing the magnetization and order-
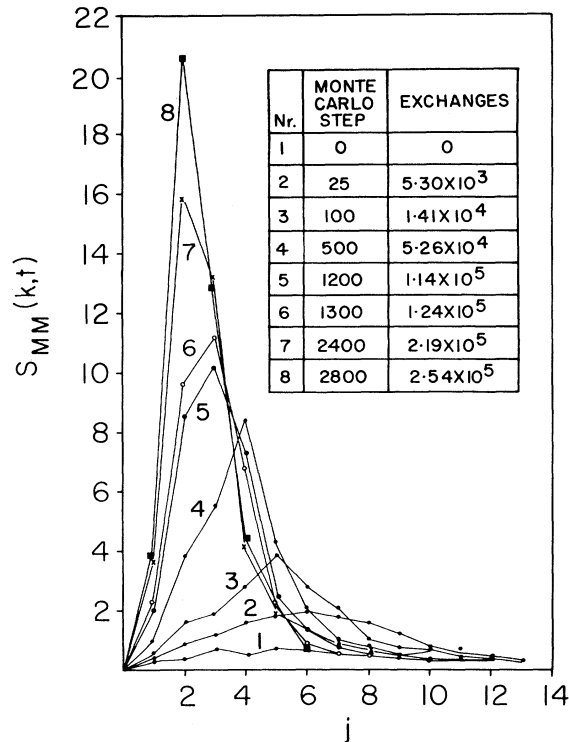

Nr.	MONTE CARLO STEP	EXCHANGES
1	0	0
2	25	5.30×10^3
3	100	1.41×10^4
4	500	5.26×10^4
5	1200	1.14×10^5
6	1300	1.24×10^5
7	2400	2.19×10^5
8	2800	2.54×10^5

FIG. 1. The magnetic structure factor $S_{MM}(k,t)$ vs k for different times.

Nr.	MONTE CARLO STEP	EXCHANGES
1	25	5.30×10^3
2	50	8.44×10^3
3	75	1.14×10^4
4	300	3.33×10^4
5	1400	1.32×10^5
6	1900	1.76×10^5
7	2400	1.19×10^5
8	2800	2.54×10^5
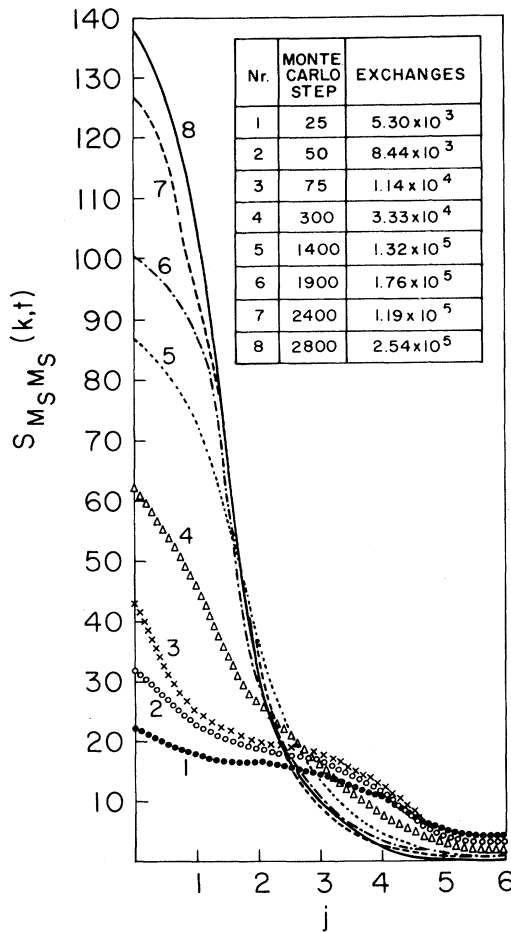
FIG. 2. The sublattice magnetization structure factor $S_{M_S M_S}(k,t)$ vs k for different times.

FIG. 3. The function $t^{-2x} S_{M_S M_S}(k,t)$ vs kt^x with x = 0.35, which exhibits the scaling form discussed in the text. The dots denote actual data points, while the crosses are points obtained from Fig. 2.

parameter structure factors, we have also calculated the cross structure factor, $S_{MM_S}(k,t)$. Its behavior is qualitatively similar to $S_{MM}(k,t)$.

We have observed from sampling various M.C. configurations that in the early stages spinodal decomposition develops by the formation of domains in a "paramagnetic" background of up spins whose average size $l(t)$ gradually increases, reminiscent of the study of Phani et al. Motivated by their work we have attempted to find a scaling region in which $t^{-2x} S_{M_S M_S}(k,t) = F(kt^x)$. As can be seen from Fig. 3 we find that in the early time domain ranging from $t = 1400$ to our maximum t = 2800 scaling does seem to be satisfied, with our best fit corresponding to $x = 0.35$. We should emphasize that this is only an approximate estimate of x, due to the large fluctuations encountered in M.C. experiments. It seems natural to identify the length scale t^{+x} with the domain size $l(t)$, but
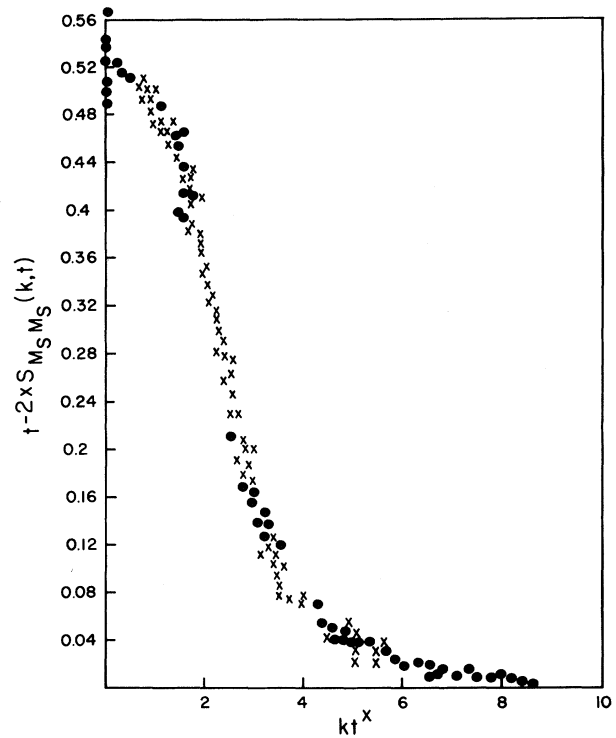
further investigation of this point is necessary. It should also be noted that the region in which we observe scaling is somewhat later than for the pure antiferromagnet, in which the scaling domain is apparently $0 \lesssim t \lesssim 100$, in contrast to our $1400 \lesssim t \lesssim 2800$. We believe scaling might hold for times greater than 2800 M.C. steps (but smaller than the "equilibrium" time) even though we have not studied this region yet. There are two points which need to be made concerning scaling. The first is that the deviations from our "universal function" $F(kt^x)$ are everywhere less than 13% in the time interval shown in Fig. 3. This is significantly less than in the pure antiferromagnet in which for very short times near zero the deviation is a factor of 4 larger than in our case. If we allow similar deviations, it would appear that we could extend the scaling region to a lower time of about $t \simeq 300$. Secondly, as a result the difference in time scales between the pure antiferromagnet and the metamagnet (due to the dynamical significance of the conservation law for M), the time for $M_s(t)$ to approach its equilibrium value is much longer than for the pure antiferro-

magnet, in which equilibrium is usually achieved in a time ranging between 175 and 800 M.C. steps. In our case on the basis of one long run we estimate this time to be approximately 5600 M.C. steps. Thus we are always in an "early-time" region in which domains are small compared to the sample size. As a consequence we do not encounter the late-time finite-size effects observed and analyzed in the antiferromagnetic model.

Finally, we note that we have also found a scaling behavior for $S_{MM}(k,t)$ of the type observed earlier by Marro, Lebowitz, and Kalos[7] for the binary alloys. Namely, $t^{-2a}S_{MM}(k,t) = G(kt^a)$ for $1000 \lesssim t \lesssim 2800$, where $a \simeq 0.21$.

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Three-Branch Polariton Dispersion Curve in CuCl

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A two-oscillator model, taking into account both the Z_3 and the Z_{12} exciton, is used to calculate the polariton dispersion curve in CuCl. The new curve has three branches. Values of the parameters characterizing the two oscillators are given, which yield a good agreement between theoretical predictions calculated from this model and several experimental results.

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It is well known that the coupling between the electromagnetic radiation field and the polarization field associated with transverse excitons in a crystal results in mixed eigenstates, called polaritons.[1] In CuCl, the theoretical dispersion curve of the excitonic polaritons has usually been calculated by considering a single-oscillator model,[2-5] taking into account only the Z_3 exciton. The corresponding dispersion relation $E(k)$ is then

$$\frac{\hbar^2 c^2 k^2}{E^2} = \epsilon_\infty + \frac{4\pi\beta}{1 - E^2/E_T^2(k)},\tag{1}$$

where ϵ_∞ is the high-frequency dielectric constant, expressing the effect of all the oscillators located at higher resonant energies, and considered here as a constant.

The parameter $4\pi\beta$, which characterizes the oscillator strength, is related to the energies E_T of the transverse exciton and E_L of the longitudinal exciton at $k = 0$ by

$$E_L^2/E_T^2 = 1 + 4\pi\beta/\epsilon_\infty = 1 + 4\pi\tilde{\beta}\tag{2}$$

with $\tilde{\beta} = \beta/\epsilon_\infty$. Finally, the spatial dispersion is also taken into account in Eq. (1) by letting

$$E_T(k) = E_T + \hbar^2 k^2/2m^*\tag{3}$$

as given by the effective-mass approximation, m^* being the effective mass of the Z_3 exciton.

This model has proved satisfactory in predicting the experimental positions of the luminescence line N_T due to the recombination of "cold" biexcitons leaving a transverse exciton (lower polariton) in the crystal under various geometrical configurations,[2,3] as well as that of the R_T^- and R_T^+ emission lines resulting from the process of two-photon resonant Raman scattering in the same spectral region,[6] provided ϵ_∞ was made