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Dynamical scaling and crossover from algebraic to logarithmic growth in dilute systems

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The ordering dynamics of the two-dimensional Ising antiferromagnet with mobile vacancies and nonconserved order parameter is studied by Monte Carlo temperature-quenching experiments. The domain-size distribution function is shown to obey dynamical scaling. A crossover is found from an algebraic growth law for the pure system to effectively logarithmic growth behavior in the dilute system, in accordance with recent experiments on ordering kinetics in impure chemisorbed overlayers and off-stoichiometric alloys.

Impurities,[1-3] vacancies,[4-7] random fields,[8-10] and second-phase particles[11] are known to have a dramatic effect on condensed-matter systems undergoing ordering processes. The case of quenched (immobile) impurities or vacancies[1,2,4,7] and random fields[8-10] has been studied in quite some detail, and theory,[1,8,10] computer simulations,[2,7,9] and some experiments[12] have suggested that the dominant growth mode at late times is logarithmic. The The case of annealed (mobile) vacancies and impurities has been considered much less.[3-6] However, a recent time-resolved x-ray experiment[13] on the effect of extra Cu on the ordering dynamics in thin films of Cu_3Au has indicated that also in the annealed case do impurities (vacancies or excess atoms) lead to an effective logarithmic growth law. Similarly, we point out that the recent finding by high-resolution low-energy electron diffraction (LEED) experiments[14] of a crossover to slow growth in nitrogen-doped oxygen overlayers on W(112) surfaces gives further testimony to the general nature of this crossover.

In this paper, we have performed a computer-simulation study, in the simplest possible setting, of the ordering dynamics in two-dimensional systems with mobile vacancies (inert impurities). We have been able to study the ordering process in three different time regimes: (i) an early-time regime of nucleation and growth of ordered domains, (ii) an intermediate-time regime of vacancy precipitation and trapping of the vacancies at the domain boundaries, and (iii) a late-time regime of diffusion of vacancies along the domain boundaries towards the surface leading to complete phase separation. The migration of the vacancies to the domain boundaries leads to a lowering of the interfacial tension and a screening of the domain-domain interactions. The resulting slowing down of the growth constitutes the main result of the present work in terms of a crossover from an algebraic growth law to an effectively logarithmic growth law as the vacancy concentration is increased. This result appears not to depend on the symmetry of the ordering.

The model we study is the site-diluted Ising antiferromagnet on a square lattice with free surfaces described by the Hamiltonian

$$\mathcal{H} = J\left[\sum_{i>j}^{\mathrm{NN}} \sigma_i\sigma_j + \alpha \sum_{i>j}^{\mathrm{NNN}} \sigma_i\sigma_j\right], \tag{1}$$

with coupling constant $J>0$, and $\sigma_i = 0, \pm 1$. The value $\sigma_i = 0$ is associated with vacant sites. The global vacancy concentration is c. We have in Eq. (1) included the possibility of next-nearest-neighbor interactions in order to allow for different types of ordered structures. Most of our results are obtained for $\alpha = 0$ and $\alpha = 1$ corresponding to, respectively, twofold and fourfold degenerate (2×1) ordering. These two types of symmetries, whose ordering kinetics previously[15-19] has been extensively studied for $c = 0$, are realized experimentally in the ordering of atomic oxygen overlayers on the surfaces W(112) and W(110). In those systems atomic nitrogen may act as a diluting impurity.[14] Since the computer-simulation results obtained for the two ordering symmetries are very similar, we have restricted ourselves in this paper to reporting the results for $\alpha = 0$.

The microscopic dynamics governing the ordering processes of the model is chosen to be a combination of single-site Glauber spin-flip dynamics and Kawasaki nearest-neighbor spin-vacancy exchange. For convenience, the time scales of these two types of local equilibration mechanisms are chosen to be the same. By these mechanisms the order parameter is nonconserved and the site dilution is annealed. The global vacancy concentration is a conserved quantity. For the present Hamiltonian, Eq. (1), this dynamics will, at low temperatures, make the system relax into an equilibrium characterized by phase separation where all of the spins are gathered in a single domain of uniform order. The dynamics is implemented using a vectorized algorithm: The lattice is subdivided into 16 equivalent, interpenetrating, and noninteracting sublattices. The sublattices are visited randomly and for each sublattice, a certain fraction of the sites of that sublattice is updated simultaneously. We have verified that our particular implementation does not influence the results by varying this fraction from no vectorization to

maximum vectorization. The quenches are performed globally by initiating the system in a disordered state characteristic of infinite temperature and by suddenly changing the temperature to a value $k_B T_f/J > 0$ below the equilibrium transition temperature. The results are derived as averages over 10–20 independent quenches. The lattice sizes used correspond to 100^2 and 200^2 sites.

Figure 1 shows a log-log plot of the results for the time dependence of the average domain size[20] $\bar{R}(t)$ in the case of quenches to $k_B T_f/J = 0.25$ for different vacancy concentrations ranging from $c = 0$–50%. The time is measured in units of Monte Carlo steps per site (MCS/S). It is noted that the algebraic growth law

$$\bar{R}(t) \sim t^n \tag{2}$$

holds for $c = 0$ with the expected classical Lifshitz-Allen-Cahn value $n = \frac{1}{2}$. As the system gets diluted, the growth slows down and there is a crossover from an algebraic growth law at early times to a much slower growth mode at late times. At no stage does the growth become pinned, not even at zero temperature.[21] The larger c is, the earlier the crossover point occurs. In Fig. 2 the same data are reanalyzed in a semilogarithmic plot which shows that the dilute systems at late times have an ordering dynamics consistent with an effectively logarithmic growth law

$$\bar{R}(t) \sim \ln t . \tag{3}$$

The same statement holds for other quench temperatures. The higher the temperature is, the earlier the crossover to the slow growth mode occurs.

The spatial organization of the dilute system during the ordering process suggests that the following scenario occurs: At early times there is a fast nucleation of ordered domains and a concomitant precipitation of vacancies. As the domains grow, the vacancies become localized in the domain-boundary network. The further growth is then slowed down. At late times the vacancies migrate towards the surface of the system. This process, which proceeds predominantly along the domain-boundary network and

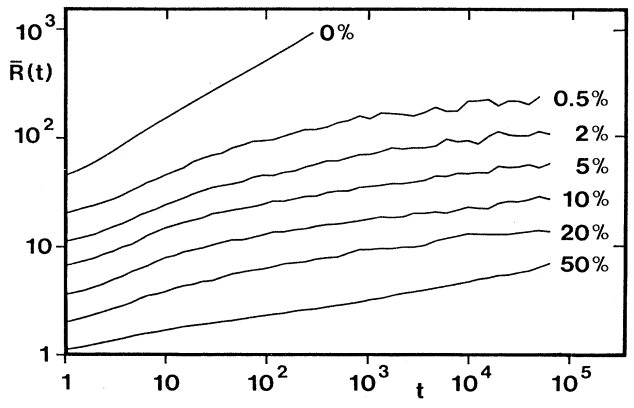

FIG. 1. Double-logarithmic plots of the average domain size $\bar{R}(t)$ vs time t (in units of MCS/S) for quenches to a temperature $k_B T_f/J = 0.25$ for different vacancy concentrations. For the sake of clarity, the various data sets have been appropriately translated along the vertical axis.

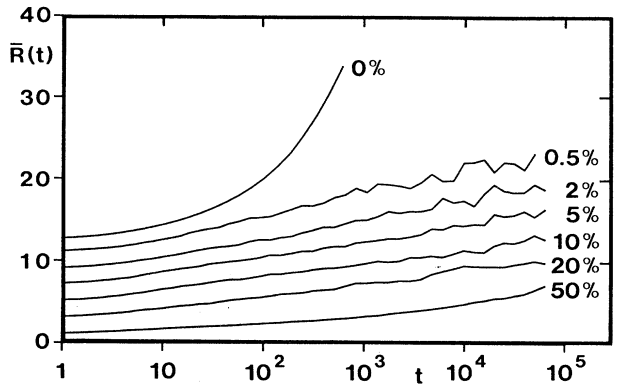

FIG. 2. Semilogarithmic plot of the data in Fig. 1.

hence is very slow, eventually leads to complete phase separation. For high dilution, the vacancies expelled to the domain boundaries effectively decouple the direct domain-domain interactions, leading to an archipelago of isolated islands. These islands coarsen and coalesce by a slow evaporation-condensation process, which is not the Lifshitz-Slyozov mechanism, since the order parameter is not conserved. This provides a possible explanation of the logarithmic growth behavior, since this evaporation-condensation process is an activated process and the time it takes for two islands to merge depends on the size of the islands. Hence the annealed randomness of the present model leads to a similar type of growth mode as the quenched randomness of the random-field Ising model.[10] The crossover to the effectively logarithmic growth mode depends on the diffusivity of the vacancies, i.e., the ratio of time scales between the spin-flip and spin-vacancy exchange mechanisms, which in this work is chosen to be unity. For slower diffusivity, the crossover will occur at a later time.[3]

As illustrative examples of this scenario, time series of microconfigurations are shown in Fig. 3 for quenches to temperatures $k_B T_f/J = 0.50$ and 0.25 for very dilute systems, $c = 20\%$ and 50%. The corresponding domain-size distribution function, $P(R,t)$ [subject to the normalization $\int_0^\infty P(R,t)dR = 1$], is given in Fig. 4 for one of the cases. $P(R,t)$ is a measure of the probability of occurrence of a domain of linear extension R at time t. It displays, as expected, an asymmetric peak at a finite, temperature-dependent value of the domain size. It has a long tail towards the large domain sizes corresponding to an approximate log-normal distribution. By introducing the time-dependent scaling variable $x = R(t)/\bar{R}(t)$ into the distribution function $P(R,t)$ and by renormalizing in order to define a dynamical scaling function $\tilde{P}(x)$ [i.e., $\int_0^\infty \tilde{P}(x)dx = 1$] we discover, as demonstrated in the inset of Fig. 4, that the entire data set in Fig. 4, from $t \simeq 10^2$ to 2×10^5, can be collapsed into a single function, $\tilde{P}(x)$. Hence, the ordering process exhibits dynamical scaling over more than three decades of time. A similarly remarkable result holds for other vacancy concentrations and temperatures.

In the case of fourfold degenerate (2×1) ordering [$\alpha = 1$ in Eq. (1)] we have found results similar to those presented in Figs. 1, 2, and 4. In addition, the time evolu-

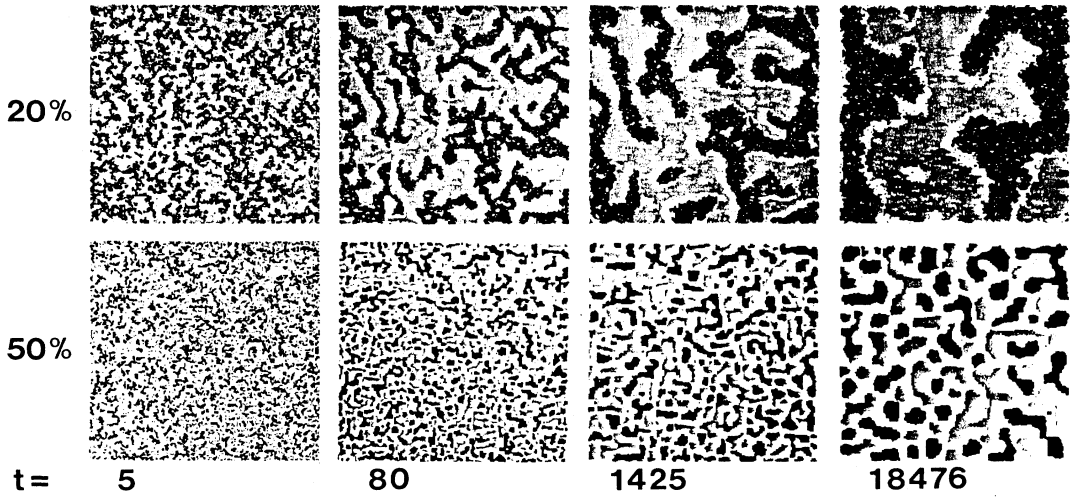

FIG. 3. Snapshots of microconfigurations as they evolve in time t (in units of MCS/S) after quenches to temperatures $k_BT_f/J=0.50$ and 0.25 for vacancy concentrations $c=20\%$ and 50%. The two types of (2×1) antiferromagnetically ordered domains are indicated by grey and black regions.

tion of the vacancy distribution proceeds in a manner qualitatively similar to that seen in, e.g., Fig. 3. Hence we conjecture that the results of a crossover from power-law growth to effectively logarithmic growth in annealed dilutions is a general result that does not depend on the symmetry and the degeneracy of the order parameter.

Annealed randomness in systems undergoing ordering processes is realized in a number of different experimental systems. Here, we shall refer to two recent experimental studies on ordering dynamics in mixtures. The first one is the high-resolution LEED study of oxygen ordering in a monolayer chemisorbed on W(112) of Zuo, Wang, and Lu.[14] In this case the oxygen atoms have an ordering with the same symmetry as the (2×1) structures of the present model study. In the presence of nitrogen impurities a crossover from Lifshitz-Allen-Cahn algebraic growth to logarithmic growth is observed.[14] This result was interpreted[14] as a case of the random-field Ising model, i.e., in terms of quenched randomness. However, surface diffusion data[22] for O and N on tungsten surfaces in the pertinent temperature range indicate that the diffusion of nitrogen impurities should not be appreciably slower than that of oxygen. The impurities therefore do not couple directly to the local order parameter[1] as in the random-field Ising model. Hence, we reinterpret the LEED data as a manifestation of logarithmic growth due to annealed randomness. The second experimental study is that of Shannon, Harkless, and Nagler,[13] who studied the effect of extra Cu on the ordering kinetics in thin films of Cu_3Au. These authors found that the extra Cu, in an off-stoichiometric quench, has the same effect on the growth behavior as a diffusing impurity leading to a logarithmic growth law. This finding is consistent with the general interpretation of the results of the present work. Moreover, the picture of the ordering process suggested by Shannon *et al.*[13] by which the extra Cu precipitates into the domain boundaries is supported by our computer-simulation data. Finally, we wish to remark that the model studied in this work may be a useful simple model for studying certain general aspects of sintering processes[23] and the annealing of radiation-damaged materials.

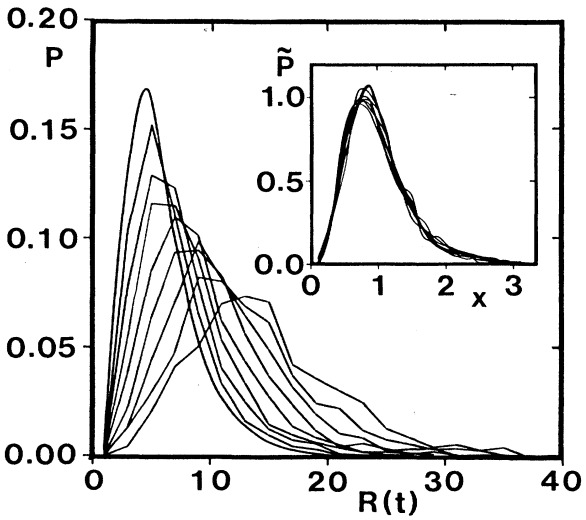

FIG. 4. Domain-size distribution functions $P(R,t)$ for quenches to a temperature $k_BT_f/J=0.25$ for a vacancy concentration $c=50\%$; cf. Fig. 1. The maximum of $P(R,t)$ moves to the right as times lapses. Results are given for the following times, $t=79$, 164, 340, 708, 1424, 2853, 5765, 11 764, and 18 475 MCS/S. The inset shows the corresponding dynamical scaling function $\tilde{P}(x)$ where $x=R(t)/\bar{R}(t)$ is the scaling variable.

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[20]The average domain size $\bar{R}(t)$ is for $c>0$ calculated from the linear-size distribution function $P(R,t)$. The linear size of a given domain is defined as the square root of the number of spins in that domain. For the pure system $c=0$, $R(t)$ by this definition is an unreliable late-time measure of linear domain size of twofold degenerate ordering due to percolation; cf. G. S. Grest and D. J. Srolovitz, Phys. Rev. B **30**, 5150 (1984). Hence for $c=0$ we have used the excess energy $\overline{\Delta E}(t)=\bar{E}(t)-E(t\rightarrow\infty)$ via the scaling relation $\overline{\Delta E}^{-1}(t)\sim\bar{R}(t)$ as a more accurate measure of linear domain size.

[21]The lack of pinning in the present calculations may be contrasted to the finding by Srolovitz and Hassold (Ref. 3; see also Ref. 5) of pinning in $T\simeq 0$ quenches on Ising ferromagnets with mobile vacancies, in which the growth stops at late times. The reason for this difference is that, whereas we allow for simultaneous Kawasaki and Glauber excitations, Srolovitz and Hassold treat these excitations independently. This implies that in their implementation the motion of the vacancies trapped at domain-boundary kinks on the square lattice becomes an activated process. We have verified that the diluted ferromagnet indeed becomes unpinned as the temperature is raised.

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