

## Vacancy-driven ordering in a two-dimensional binary alloy

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Domain growth in a system with nonconserved order parameter is studied. We simulate the usual Ising model for binary alloys with concentration 0.5 on a two-dimensional square lattice by Monte Carlo techniques. Measurements of the energy, jump-acceptance ratio, and order parameters are performed. Dynamics based on the diffusion of a single vacancy in the system gives a growth law faster than the usual Allen-Cahn law. Allowing vacancy jumps to next-nearest-neighbor sites is essential to prevent vacancy trapping in the ordered regions. By measuring local order parameters we show that the vacancy prefers to be in the disordered regions (domain boundaries). This naturally concentrates the atomic jumps in the domain boundaries, accelerating the growth compared with the usual exchange mechanism that causes jumps to be homogeneously distributed on the lattice.

### I. INTRODUCTION

Binary alloys are known to exhibit a rich variety of phases as a function of temperature and concentration. Some of the phase-transition lines can be understood as order-disorder phase transitions between a high-temperature phase in which the species are randomly distributed on the lattice sites and a low-temperature phase in which the symmetry is broken and two or more sublattices have different concentration of the species. Although their equilibrium phase diagrams begin to be well understood from first-principles theories,<sup>1</sup> the dynamics of the relaxation to equilibrium is still under discussion. A typical experiment consists in quenching a binary alloy from the high-temperature disordered state to a temperature  $T$  well below the order-disorder phase transition. The long-range order develops by the nucleation and growth (or spinodal ordering) of ordered domains. This represents an example of a domain growth process, that have been studied intensively for the last two decades. Such processes are very common in nature and appear in a wide variety of systems. Other typical examples are crystal growth, phase separation, magnetic domain growth, and so on. These kind of problems are not only of interest in the field of materials science but represent as well a fundamental problem in nonequilibrium statistical mechanics. Here, the main interest has been to classify in few classes the growth processes depending on very general features like the existence of conservation laws. This possibility seems appropriate in the late stages of growth when the domain sizes are larger than the microscopic lengths in the system. Hence, by analogy with critical phenomena, where the order parameter correlation length is much larger than the microscopic lengths, one expects as a result, power laws, scaling and universality.<sup>2</sup> In particular, the domain size should increase as  $L(t) \sim t^x$  and the structure factor scale as

$$S(\mathbf{k}, t) = [L(t)]^{d\hat{S}} [kL(t)] \quad (1)$$

$d$  being the dimensionality and  $\hat{S}$  the dynamical scaling function. For systems with nonconserved order parameter, like in ordering binary alloys, a dynamical exponent  $x = 1/2$  is proposed (Allen-Cahn law<sup>3</sup>). This result has been confirmed by Monte Carlo (MC) simulations for pure systems.<sup>4</sup> However the exponent seems to be sensitive to the existence of diffusing impurities,<sup>5</sup> quenched (immobile)<sup>6</sup> and annealed (mobile)<sup>7</sup> disorder or self-pinned effects.<sup>8</sup> Experimentally, measurements are difficult to perform and interpret. A number of experiments are in agreement with  $x = 1/2$ .<sup>9</sup> Nevertheless, impurities<sup>10</sup> or nonstoichiometry<sup>11</sup> may reduce the values of  $x$ . The existence of large universality classes is nowadays controversial.

Monte Carlo simulation studies of ordering kinetics in AB binary systems with conserved density frequently use the unrealistic Kawasaki dynamics, i.e., the elementary movements consist in neighbor atom exchanges. Actually, it is known that in metallic alloys atomic diffusion mainly proceeds via vacancies.<sup>12</sup> This fact was already taken into account by Flynn and McManus<sup>13</sup> who implemented the vacancy mechanism in the first MC simulations of bcc binary alloys. Details of vacancy motion in alloys were studied using MC simulations by Beeler and Delaney<sup>14</sup> who showed that for low vacancy concentrations, the vacancy random walk concentrates in the ordered regions. Recently Fultz<sup>15</sup> indicate that vacancy trapping increases when ordering develops, and that low lattice coordination numbers favors trapping at the anti-phase domain boundaries.

The problem of domain growth in systems with vacancies has been considered by Mouritsen and Shah.<sup>7</sup> They have performed simulation studies of ordering with mixed spin-flip and vacancy mechanism showing a crossover from a  $x = 1/2$  to a slower logarithmic law due to annealed vacancies. Actually this behavior should be associated with the fact that vacancies are only allowed to jump to nearest-neighbor (NN) positions. Yaldrum and Binder<sup>16</sup> have considered the problem of phase separation of a 2D binary alloy (conserved order parameter) with a

fixed small vacancy concentration. The vacancies are allowed to jump to the different nn sites with different rates for  $A$  and  $B$  exchanges. They obtain a behavior similar to the behavior corresponding to the direct  $AB$  exchange mechanism model.

In a recent paper<sup>17</sup> we have reexamined the problem of ordering kinetics in a two-dimensional (2D) binary alloy with conserved density. The dynamics is introduced by a single vacancy that performs jumps to the 4 NN and the 4 next-nearest-neighbors (NNN) with equal probability. Although the nn jumps are responsible for the ordering, the NNN jumps are very important because they enable the movement of the vacancy in the ordered regions with no energy cost, preventing trapping phenomena. The main results are (i) after an initial period of domain formation, the growth follows a power law with  $x \simeq 0.77$ , clearly faster than  $x = 1/2$ , (ii) the fast growth is also deduced from finite-size scaling analysis, and (iii) scaling relations for the structure factor [Eq. (1)] are well satisfied during the domain growth period.

In this paper we analyze carefully the behavior of the vacancy path in the system during the domain growth for different probability rates between NN and NNN jumps. We discuss the possibility that non-uniform excitations, arising from the interaction with a heat bath and driving the system to equilibrium, may strongly modify the universal laws commented above. We also discuss the relation of these results with experiments.

The paper is organized as follows. In Sec. II we present the details of the model and the simulation. In Sec. III, we present the MC results. In Sec. IV we discuss the results and analyze the reasons for the observed time evolution of the system in detail. Finally, in Sec. V we conclude.

## II. MODEL AND MONTE CARLO SIMULATION DETAILS

We have used the standard lattice binary mixture model for the description of a  $A_x B_{1-x}$  binary alloy. Let us consider a 2D square lattice of dimensions  $L \times L$  with periodic boundary conditions. On each site  $\{i = 1, \dots, N = L^2\}$  we define a variable  $S_i$  taking values 1 or  $-1$  if that site is occupied by an atom  $A$  or  $B$ , respectively. The number  $N_A (N_B)$  of  $A (B)$  particles is kept constant. We include a single vacancy ( $S_i = 0$ ) in the system, substituting an  $A$  atom. The concentration of  $A$  atoms is then  $\rho = N_A / N = 0.5 - L^{-2}$ . For the sizes we have considered ( $L = 30, 50, 100, 200, 500$ ) the existence of a single vacancy does not affect the equilibrium properties compared to the perfect system. The case of concentrations different from  $\rho = 0.5$  might be quite different from the present one, and will be studied in the future. The interaction between the  $A$  and  $B$  atoms, responsible for the ordering phenomena, is modeled by the nn antiferromagnetic Ising model Hamiltonian:

$$H^* = H/J = \sum_{\langle NN \rangle} S_i S_j, \quad (2)$$

where  $\langle NN \rangle$  means sum over all the NN pairs and  $J$  is a parameter for the interaction which is taken as unit of en-

ergy. We also define a dimensionless scale of temperature in the usual way:  $T^* = k_B T / J$ . The ground state of the system can be described as two alternating sublattices filled by  $A$  and  $B$  atoms in a checkerboard way. We call the two sublattices  $+$  and  $-$  and we consistently define the sign of a site  $i$  [ $\text{sgn}(i)$ ] as  $+1$  or  $-1$  depending on the sublattice to which it belongs. This ground state structure is  $N$  times degenerated since  $+$  and  $-$  sublattices can be exchanged, and the vacancy is also free to stay in any of the  $N/2$  sites of the sublattice occupied by the  $A$  atoms. At finite temperatures this model exhibits an Ising-like order-disorder phase transition at practically the same temperature as the standard 2D Ising model.

Standard MC simulations have been performed. Instead of the usual Kawasaki dynamics exchanging  $A$  and  $B$  atoms we have considered the following dynamics (vacancy mechanism): Only exchanges between the vacancy and the 8 NN and NNN atoms are allowed. During each MC step we propose  $N$  vacancy jumps to the NN or NNN positions. The probability of proposing a jump to NN is  $W$  and to NNN is  $1 - W$ . The four NN (NNN) have equal probability to be proposed as new positions for the vacancy. The proposed jump is accepted or not according to the usual Metropolis probability:

$$p = e^{-\Delta H^* / T^*}, \quad (3)$$

where  $\Delta H^*$  is the energy change corresponding to the proposed jump. Since the proposed movements are reversible and the acceptance probability is the standard Metropolis, detailed balance is satisfied for each individual movement. We have not considered the existence of additional energy barriers that could modify the dynamic behavior of the system.

We have performed quenches from initially disordered states to a temperature  $T^* = 1.0$ , well below the order-disorder phase transition ( $T_c^* \simeq 2.27$ ). Usually runs of 3000 MCS steps are enough to equilibrate the system in the  $L = 500$  case. For each lattice size ( $L = 30, 50, 100, 200, 500$ ) averages over 50, 30, 20, 15, and 8 different runs have been, respectively, taken. We have excluded the cases that lead to final ‘‘slab’’ configurations, corresponding to two competing domains separated by a flat interface, and which are characterized by a very small value of the long-range order parameter defined below in Eq. (4).

During the simulation, at the end of each MC step, we have monitored the following quantities: the energy [ $E(t)$ ], the acceptance ratio of the proposed jumps to NN [ $\phi_1(t)$ ] and to NNN [ $\phi_2(t)$ ], the long-range order parameter  $m(t)$  defined as

$$m = \frac{1}{N} \left| \sum_{i=1}^N S_i (-1)^{\text{sgn}(i)} \right| \quad (4)$$

and the following two local order parameters:

$$m_s = \left\langle \frac{1}{25} \left| \sum_{5 \times 5} S_i (-1)^{\text{sgn}(i)} \right| \right\rangle, \quad (5)$$

$$m_v = \frac{1}{24} \left| \sum_{5 \times 5}^v S_i (-1)^{\text{sgn}(i)} \right|, \quad (6)$$

where  $\sum_{5 \times 5}$  means a sum over all the sites inside a generic square subblock of size  $5 \times 5$ ,  $\sum_{5 \times 5}^v$  means the sum over all the sites inside a  $5 \times 5$  subblock centered on the vacancy position, and the angular brackets mean an average over all the possible nonoverlapping subblocks in the  $L \times L$  lattice. The first local-order parameter is a measure of the short-range order in the lattice (it reaches a value  $\simeq 1$  when the size of the correlations exceeds a  $5 \times 5$  subblock), and the second one is the same measure but restricted to a local environment around the vacancy.

### III. MONTE CARLO RESULTS

The first surprising result, completely different from the simulations using standard Kawasaki dynamics, is the fast growth behavior. A log-log plot of the energy evolution versus MC steps, for different lattice sizes and  $W=0.5$ , is presented in Fig. 1. The final equilibrium energy  $E_F$  has been evaluated using long enough simulations ( $10^4$  MCS), and does not differ from the one expected for a system with no vacancy. In a previous paper<sup>17</sup> we have also analyzed the growth of the domain sizes measuring the structure factor  $S(\mathbf{k}, t)$ . For all lattice sizes the growth is faster than the Allen-Cahn law. An estimation of the exponent gives  $x \simeq 0.77$ . For the small systems the exponent is even bigger.

In Fig. 2 we present the same log-log figure for the case of  $L=500$  and different values of the probability  $W$  ( $W=0.3, 0.5, 0.7, 0.9$ , and  $1.0$ ). As can be seen, the fast growth behavior is independent of the value of  $W$ , except for the case of  $W=1.0$  (NN jumps only) for which the known logarithmic behavior  $E(t) \sim \log(t)$  is found.<sup>7</sup> This is demonstrated in the inset plot where the curve corresponding to the case  $W=1.0$  is shown in semilogarithmic scales. For this case the change in the dynamics is very dramatic. In the ordered regions a jump of the vacancy to a NN position is accepted with a probability  $p = e^{-6/T^*} \simeq 2 \times 10^{-3}$  while a jump to a NNN position is

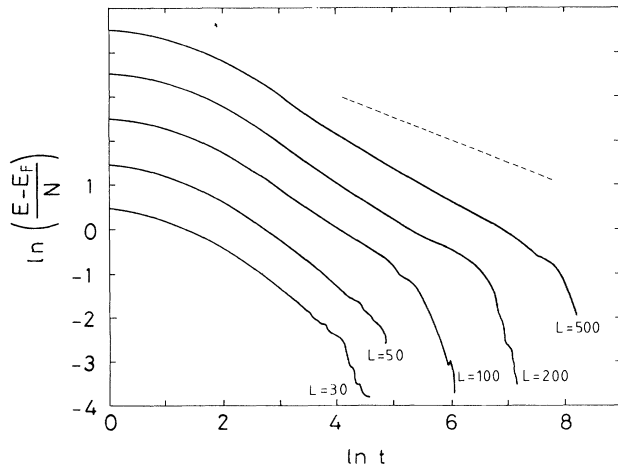


FIG. 1. Log-log plot of the excess energy per particle vs time for different system sizes. The curves are sequentially shifted one vertical unit in order to clarify the picture. The dashed line represents the  $t^{-0.5}$  behavior.

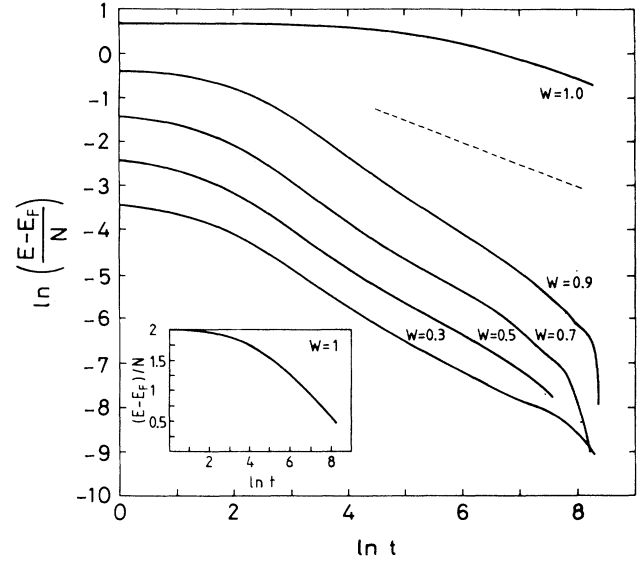


FIG. 2. Log-log plot of the excess energy per particle vs time for different values of the probability  $W$  of proposing nn jumps in front of nnn. The curves are sequentially shifted one vertical unit in order to clarify the picture. The dashed line represents the  $t^{-0.5}$  behavior. The inset shows the curve with  $W=1$  (nn jumps only) in semilog scales.

accepted with  $p=1$ . Therefore it is very important to allow jumps to NNN (very few are enough) in order to prevent the logarithmic growth.

Figure 3 shows the evolution of the acceptance ratios of the proposed jumps to NN and NNN ( $\phi_1$  and  $\phi_2$ ) defined in the previous section. As can be seen the behavior at long times is the same independent of  $W$ . As the lattice orders, more NNN jumps are accepted and less NN jumps are possible.

It should be noticed that the NNN jumps in the or-

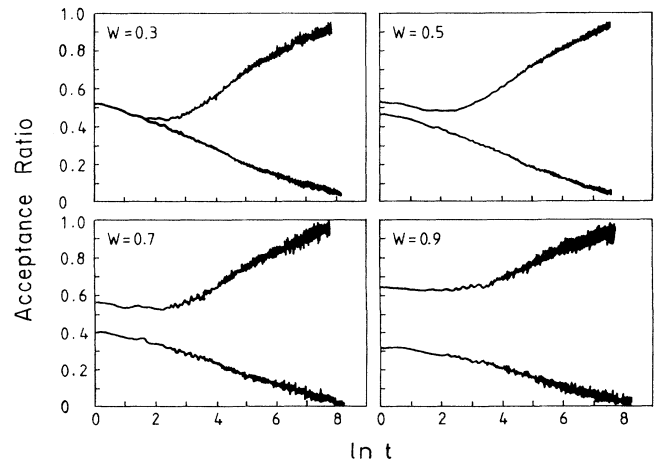


FIG. 3. Time evolution of the acceptance ratio of the vacancy jumps to NNN  $\phi_2$  (curves above) and NN  $\phi_1$  (curves below), for different values of the parameter  $W$ .

dered domains cost no energy at all, but only the NN jumps are really contributing to increase the order in the system since they exchange atoms from one sublattice to the other. Therefore the fast ordering dynamics is only due to nn jumps, and the role played by the NNN is to improve the mobility of the vacancy inside the ordered regions, preventing the trapping phenomena (logarithmic growth) and helping the vacancy to find the disordered regions (domain boundaries) where it operates increasing the degree of order in the system.

The difference in the initial time behavior can also be understood from the above considerations. In the initial fully disordered configuration one has  $\phi_1 = \phi_2$ , but since only nn jumps increase order, the higher  $W$  the faster the lattice orders in the first MC step. This explains the difference between  $\phi_1$  and  $\phi_2$  after the first MC step observed for high  $W$  values. Also the faster the system orders in the first MC step, the smaller is the value of the acceptance ratio  $\phi_1$  for the NN jumps, and correspondingly the highest is the acceptance ratio  $\phi_2$  for the NNN jumps. A direct observation of the configuration evolution in the case of  $W = 0.9$  shows that after the first MC steps, the system has formed large ordered domains and the vacancy travels fast through them, looking for the antiphase boundaries. The above picture is also demonstrated in Fig. 4 where we plot the local order parameter averaged over the whole system ( $m_s$ ) against the local order parameter around the vacancy ( $m_v$ ), as defined in the previous section, for the case  $W = 0.5$  and  $L = 500$ . For a Kawasaki exchange mechanism the averaged local order parameter and the local order parameter around the position where exchanges are performed are equal, since jumps are proposed in a homogeneous way. In our case, we can clearly distinguish two regions. In the initial stages (less than 1 MC step) the order around the vacancy

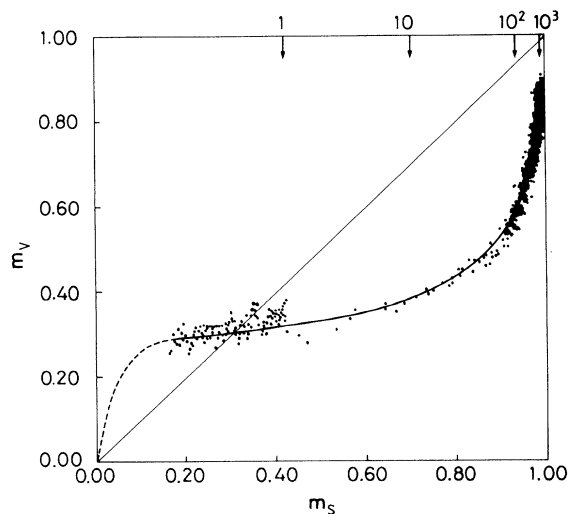


FIG. 4. Local order parameter around the vacancy position  $m_v$  vs the average local order parameter  $m_s$ . The straight line corresponds to the expected behavior if the vacancy homogeneously sweeps the lattice. The arrows on top indicate the corresponding time evolution in MC steps.

is higher than in the system since only the vacancy can develop order. The data corresponding to this region shows a higher dispersion since it corresponds to averages over short intervals inside the first MC step. As soon as the vacancy has performed 3 or 4 MC steps, the figure shows that the order around the vacancy is smaller than the average order in the system. This is due to the fact that the vacancy spends more time in the disordered domain boundaries than in the ordered domains. By this method the excitations in the system (proposed jumps) are naturally concentrated in the disordered regions, which accelerates the growth law compared to the case in which the excitations are performed homogeneously on the lattice.

#### IV. COMPARISON WITH THE EXCHANGE MECHANISM

The results presented above, suggest that the nonhomogeneity of the excitations in the system is responsible for the observed fast behavior. In a general way, when the system exhibits dynamical scaling, it has been shown<sup>18</sup> that the exponent  $x$  is related to the way the interface velocity changes under uniform magnification of the linear dimensions of the system. In particular if the velocity scales as

$$v(\lambda L) = \lambda^\beta v(L), \quad (7)$$

where  $\lambda$  is the magnification factor, then it is obtained that  $x = 1/(1-\beta)$ . This argument can be used in our case, because previous studies of the structure factor  $S(\mathbf{k}, t)$  (Ref. 17) have clearly shown that scaling [Eq. (1)] is present after an initial transient period.

Let us now consider a system of linear dimension  $L$  with an interface of length  $P$  and study how the velocity of the interface changes when the system is magnified a factor  $\lambda$ . If the excitations in the system are homogeneous, like in the case of usual exchange dynamics (no matter if the exchanges are proposed randomly or sequentially on the lattice), in each unit of time (1 MC step) every site in the interface is, in average, visited once, so the interfaces advance the same distance independent of  $\lambda$ . Thus, in the system of dimension  $\lambda L$  the velocity  $v(\lambda L)$  changes as:

$$v(\lambda L) = \lambda^{-1} v(L) \quad (8)$$

giving  $x = 1/2$ , as deduced from a deterministic Langevin equation assuming a Ginzburg-Landau free energy functional.<sup>3</sup> Consider now the extreme case in which all excitations are concentrated on the  $P$  interface sites. During a unit of time (1 MC step) the sites of the interface are visited  $L^2/P$  times. Then the interface advance per unit of time is proportional to  $L$ , giving:

$$v(\lambda L) = v(L) \quad (9)$$

which leads to a maximum dynamical exponent  $x = 1$ . In our case the vacancy spends most of the time on the interface but sometimes it travels across ordered domains, slowing the growth. This effect might be more important the larger the system size is, justifying the possible de-

crease of the exponent  $x$  when  $L$  increases and  $\rho$  decreases (see Fig. 1).

With the above considerations in mind, we compare the results obtained with the exchange mechanism with those corresponding to the vacancy mechanism. In both cases, during the scaling regime, only the NN jumps (or exchanges) are responsible for ordering.

For the case of exchanges, the NN jumps are practically only accepted when the exchanges are proposed on the interface, so the acceptance ratio  $\phi_1$  is proportional to the length of the interfaces, and consequently it goes like  $\phi_1^h(t) \sim t^{-0.5}$ . For the extreme case in which the excitations are only proposed on the interface,  $\phi_1$  would be constant until the interface disappears. In our case of vacancy mechanism (nonhomogeneous) an intermediate behavior is expected. Figure 5 shows the evolution of the nn acceptance ratio  $\phi_1$  for the case of exchanges ( $\phi_1^h$ ) and vacancy ( $\phi_1^{nh}$ ) mechanism for a system with  $L = 100$ .  $\phi_1^{nh}$  is compared with a phenomenological linear fit. The inset shows  $\phi_1^h$  in logarithmic scales compared with a  $t^{-0.5}$  law.

The change in the growth law from  $x = 1/2$  to  $x = 0.77$  can be understood as a change of time scale between the homogeneous and nonhomogeneous case. The corresponding time scales  $\tau^h$  and  $\tau^{nh}$  are proportional to the inverse of the acceptance ratios ( $\phi_1^h, \phi_1^{nh}$ ). The ratio between the two time scales is then given by

$$C(t) = \frac{\tau^h(t)}{\tau^{nh}(t)} = \frac{\phi_1^{nh}(t)}{\phi_1^h(t)}. \quad (10)$$

Figure 6(a) shows  $C(t)$  calculated from the MC results, together with the theoretical behavior obtained from the

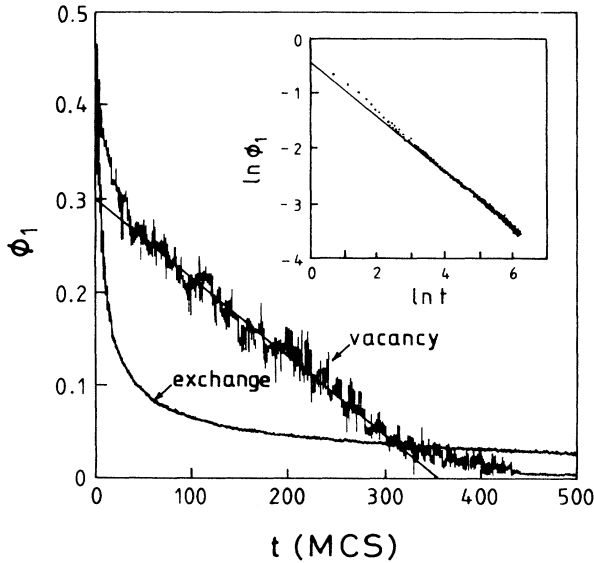


FIG. 5. Comparison of the acceptance ratio to nn positions  $\phi_1$  vs time, obtained with vacancy mechanism and with the usual exchange mechanism. The straight line corresponds to a phenomenological linear fit. The inset shows the case corresponding to exchange mechanism in log-log plot, together with the  $t^{-0.5}$  behavior. These results correspond to a system with  $L = 100$ .

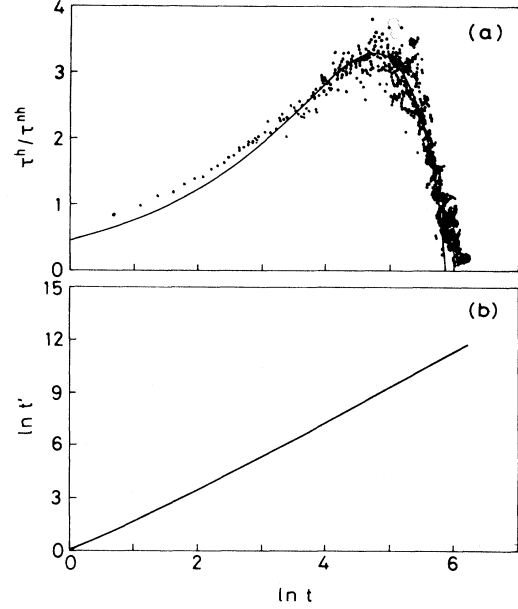


FIG. 6. (a) Ratio between the time scales corresponding to the homogeneous case (exchange mechanism) and the nonhomogeneous case (vacancy mechanism) obtained from data in Fig. 5. The continuous line is the ratio between the two fits also in Fig. 5. (b) Corrected time scale vs noncorrected time scale in log-log scales.

ratio between the two fitted behaviors (linear and  $t^{-0.5}$ ) in Fig. 5. The time correction that should be applied to the nonhomogeneous case in order to recover the homogeneous behavior is given by

$$t'(t) \simeq \int_0^t C(s) ds \quad (11)$$

which is shown in Fig. 6(b). Figure 7 shows the behavior of  $E(t)$  (in a log-log scale) for the vacancy mechanism before (curve 1) and after (curve 3) the time correction, together with the results corresponding to the exchange mechanism (curve 2). An exponent  $x = 0.46 \pm 0.03$  is obtained close to the  $x = 1/2$  when the nonhomogeneity of the excitations is corrected. Small differences could be associated with the existence of a small fraction of NNN

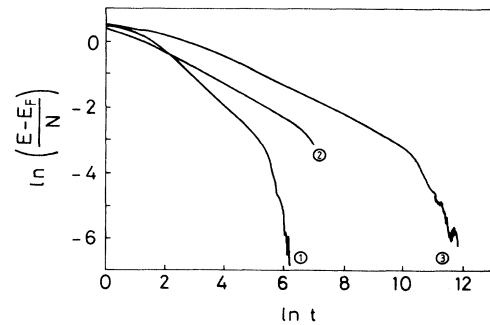


FIG. 7. Log-log plot of the excess energy per particle vs time computed using the vacancy mechanism (1) and the exchange mechanism (2). Curve (3) corresponds to curve (1) after applying the time correction plotted in Fig. 6(b).

jumps on the interface contributing to local ordering. This result justifies the assumption that the nn jumps in disordered regions (interfaces) are the main ordering mechanism and are responsible for the fast growth.

Most of the simulation results existing in the literature are performed with the standard homogeneous exchange mechanism. In fact, nonhomogeneous methods (like the  $n$ -fold way<sup>19</sup>) have been used as artifacts to accelerate MC simulations but the growth results have always been "corrected" for the nonhomogeneity. Here we propose that such nonhomogeneities could naturally appear in systems where ordering is controlled by vacancies and where no other slowing mechanism appear. Binder has recently used the vacancy mechanism to simulate a case of phase separation (conserved order parameter). In that case no difference is found between the vacancy mechanism and the usual exchange mechanism, since the growth is controlled by the particle diffusion giving  $x = 1/3$ . Also very recently, simulations of a model for order-disorder phase transitions with strain induced coupling<sup>20</sup> leads to a growth law faster than the usual Allen-Cahn law. It is difficult to see if in that case the excitations are homogeneous or not, but could be that the existence of strains produces a nonhomogeneous behavior of the system in agreement with our results.

To our knowledge no experimental measurements reporting such fast growth law in the case of ordering binary alloys exist. Different reasons for that can be argued. Firstly most experiments are performed on 3D systems. The behavior of the vacancy walk in 3D is different from the 2D case and this could modify the 3D growth law. Future simulation studies for the 3D case will clarify this point. Secondly, the presented results could change due to vacancy-vacancy correlations for larger vacancy concentration. Also, in real systems, the effective vacancy concentration could change during growth which will also influence the time scale. Finally, the ex-

istence of additional energy barriers other than those coming from the local order, could slow down the movement of the vacancy and give a smaller exponent  $x$ .

Nevertheless, in the light of the present results, we strongly encourage more experimental work specially designed to reveal this vacancy driven mechanism. Specially, stoichiometric binary alloy films with a well controlled vacancy concentration could be good candidates to gain understanding in the present problem.

## V. CONCLUSIONS

We have simulated the domain growth process in a binary alloy quenched below its order-disorder phase transition. Instead of the usual pair exchange dynamics we have used a single vacancy dynamics. Trapping phenomena has been prevented by allowing vacancy jumps not only to next but also to next-nearest neighbors. This dynamics models an interaction between a heat bath and the system which is established only through the vacancy. For energy reasons the vacancy prefers to stay in the disordered regions. Then, the excitations in the system (vacancy jumps) are no longer homogeneous, but naturally concentrate in the antiphase boundaries. This accelerates the ordering process giving rise to a growth exponent larger than  $1/2$ . The process is curvature driven but it cannot be simply described by the usual (Allen-Cahn model) formalism for a system with nonconserved ordered parameter and conserved density.<sup>3</sup> A third equation describing the dependence of the vacancy diffusion with the local order should be incorporated.

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