Monte Carlo Studies of Nonequilibrium Vacancy Mediated Dynamics in Binary Alloys

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While the statistical mechanics of system in thermal equilibrium is a well established discipline, nonequilibrium systems are fundamentally much less well understood, even though most natural phenomena fall into the latter category. In particular, there is as yet no nonequilibrium analog for the systematic formalism of Gibbs ensemble. Rather than using the anlytic approach as a tool to study the nonequilibrium problem, this study is restricted to the utilizing Monte Carlo computer simulations.

We have performed the simulations of noneqilibrium vacancy-driven disordering process of an initially phase segregated binary alloys system that is rapidly heated to a temperature above the critical temperature, T_c. To quantify the evolution of the system, we measure the number of broken bonds, $\mathcal{A}(L,t)$, as a function of time and system size. This disorder parameter allows us to identify three temporal regimes, distinguished by the distribution of the vacancies through the system. The temporal growth of $\mathcal{A}$ during the latter regime is captured by the exponent ~ 0.5. If the final temperature is infinite, $T=\infty$, the motion of the vacancies is a simple Brownian random walk. For *finite* temperature $T \gtrsim 1.5T_c$, we observe that these exponents still hold with some deviation. Even though interparticle interactions now play a role, correlations in the system are still short-range. The vacancy is still a random walk by nature if viewed on a length scale that exceeds the correlation length.

I. INTRODUCTION

The nonequilibrium dynamics of binary alloys, undergoing mixing or segregation processes, has been of interest for several years especially in the physics and materials science communities [1]. Much interest has focused specifically on the dynamics of phase ordering and domain growth, following a rapid quench below the coexistence curve. Starting from an initial nonequilibrium configuration, the system evolves toward its final equilibrium state. This gives rise to the fundamental questions in both disciplines. Crucial to metallurgy, the domain morphology is intimately linked to structural stability. Statistical physics, however, focuses on the universal, self-similar aspects of its temporal evolution that is manifested in characteristic growth exponents and dynamic scaling.

A key ingredient in the study of these processes is the mechanism by which two particles exchange positions. From the viewpoint of modelling and simulation purposes, *direct* particle-particle exchange obviously lead to the simplest codes and coarse-grained equations. Moreover, invoking universality, it is expected that the details of the microscopic mechanisms do not affect large-wavelength, long-time properties, and universal scaling functions. In this spirit, Kawasaki dynamics and the Cahn-Hilliard equation have been extensively used to describe phase ordering in binary alloys. However, in most real solids, microscopic atom-atom exchange can be mediated by a variety of processes [2], direct exchange plays a rather small role since steric hindrance tends to create large energy barrier. The most common mechanisms involve exchange of defects, such as vacancies or interstitial sites. For these reasons, alloys have often been modelled by three-state models [3] whose dynamics is controlled by atom-vacancy exchange. Direct exchange is completely forbidden. Two problems in particular have attracted considerable attention: first, the effect of vacancies on phase separation and domain growth [4,5], and second, their role in atomic interdiffusion [6,7]. A number of studies has also addressed vacancy-mediated ordering in a variety of *antiferromagnetic* alloys [8], as well as surface modes of unstable droplets in a stable vapor phase [9].

In this paper, we will describe a third aspect of defect-mediated dynamics, namely, the inverse of the phase ordering problem. Instead of studying the growth of order in response to a sudden temperature decrease (quenching), we focus on the *disordering* of a finite system, following a rapid *increase* in temperature. Starting at zero-temperature ferromagnetic configuration, i.e., a perfectly phase-segregated system with sharp interfaces, we monitor how the interfaces "roughen" and how particles of one species are transported into regions dominated by the other species. Clearly, if the final temperature is sufficiently high, the interfaces will eventually disappear, resulting in a homogeneous final state. Several questions emerge quite naturally: Are there characteristic time scales on which the disordering takes place, and how do they depend on the system size, temperature, and other controlling parameters? How do local density profiles and correlation functions evolve with time? Are there any scaling regimes, and what are the appropriate scaling variables? How do these features respond to changes in the relative concentrations of vacancies and alloy components?

In this paper, we address some of these questions with a simple model for defect-mediated interface destruction and bulk disordering. We consider a symmetric (Ising-like) binary alloy of A and B atoms which is diluted by a very small number of vacancies (10^{-5}, found in most real systems). Following an up-quench from *zero* to a *finite* temperature, T, the vacancies act as a "catalyst" for the disordering process, exchanging with neighboring particles according to the usual energetics of the (dilute) Ising model. The particles themselves form a passive background whose dynamics are slaved to the defect motion. Thus, this system corresponds to a real material in which the characteristic time scale for vacancy diffusion is much faster than the ordinary bulk diffusion time. While vacancies are typically distributed uniformly in the bulk, certain defects may prefer to accumulate at the interfaces. Thus, the number of defects is not necessarily extensive in system size.

While we allow for some variation in the vacancy number, we consider equal concentrations of A and B atoms. Thus, our work forms a natural complement to the only other study [10] of vacancy-mediated disordering in the literature. There, the alloy composition is chosen highly asymmetric: 95% of A atoms versus only 5% for the B species, with a single vacancy. Thus, the A atoms form a *matrix* for a B-*precipitate*. The alloy is first equilibrated at a very low temperature, so that small clusters of B atoms are present. It is then rapidly heated to a higher temperature, and the number and size of B clusters are monitored. Three different scenarios are observed, depending on whether the final temperature is below the miscibility gap, above the miscibility gap but below T_c, or above T_c. In the first case, the precipitates remain compact. They dissolve partially at first, but then equilibrate again by coarsening. In the second case, the precipitates also remain compact but eventually dissolve completely, mostly through "evaporation" from their surfaces. In the third case, the clusters decompose rapidly ("exploded") into a large number of small fragments which then disappear diffusively.

The complete or partial mixing of two materials at an interface plays a key role in many physical processes, such as corrosion or erosion phenomena [11]. We mention just two applications with huge technological potential for device fabrication. The first concerns nanowire etching by electron beam lithography [12]: If a thin film of platinum is deposited on a silicon wafer, interdiffusion of Pt and Si produces a mixing layer. If this layer is heated locally by, e.g., exposure to a conventional electron beam, silicides, such as Pt_2Si and PtSi, form. The unexposed platinum can subsequently be etched away, leaving conducting nanoscale structures behind. The second example concerns mesoscopic superlattice structures, consisting of alternating magnetic and nonmagnetic metallic layers. If adjacent magnetic layers couple antiferromagnetically, the application of a large uniform magnetic field to these layered structures results in giant magnetoresistance [13]. However, the performance of these devices requires precisely engineered layer thicknesses and interfaces, and can be significantly affected by disorder [14], including interdiffusion or interfacial fluctuations.

Motivated by these applications, our study can only form a baseline here, for further work on more realistic models. However, it also has some rather fundamental implications. First, it serves as a testing ground for a basic problem in statistical physics, namely, how a system approaches its final steady state, starting from an initial *non-stationary* configuration. A second view of our study addresses the effect of a random walker on its background medium. Each move of a vacancy slightly rearranges the background atoms, leaving a trail behind like a child running across a sandy beach. In the simplest case, the walker is purely Brownian. In our language, this corresponds to an up-quench to infinite temperature, $T = \infty$, where energy barriers are irrelevant. In this case, there is no feedback from the background to the local motion of the vacancy. Nevertheless, each displaced atom displays its own intriguing dynamics (especially in $d = 2$) [15,16]. To study the collective behavior of the atoms, [17] explored a lattice filled with just two species of (indistinguishable) particles. Beyond this simple case is a system heated to *finite* temperatures. Since the background affects the vacancy through the local energetics associated with the next move, no exact solutions are known. Instead, progress relies mainly on simulations.

The key result of our study is the observation of *three* distinct temporal regimes, separated by two crossover times, provided the final temperature is not too close to T_c. The *intermediate and late* stages of the disordering process exhibit dynamic scaling, with characteristic exponents and scaling functions that are computed analytically. For the system size considered here, a clear breakdown of these scaling forms is observed for temperatures within about 10% of T_c. We argue that this occurs when the correlation length becomes comparable to the system size. In contrast, the early stage of the disordering process consists of interfacial destruction in a highly anisotropic manner [16].

This article is organized as follows. We first introduce our model and define an appropriate disorder parameter. We then give the two-dimensiononal system results starting from the the purely diffusive case, corresponding to $T = \infty$

and less diffusive at finite tempearture $T_c < T < \infty$ where particle-particle interactions come into play in Section III. We summarize and conclude with some comments on the effect up-quenches to temperatures, $T < T_c$, and details concerning interfacial destruction in the Section IV.

II. THE MODEL

In this section, we describe the dilute Ising model underlying our Monte Carlo simulations. It is defined as a two-dimensional ($d = 2$) square lattice of dimension $L \times L$, with sites denoted by a pair of integers, $\mathbf{r} \equiv (x, y)$. The boundary conditions are fully periodic in all directions. To model the two species of particles, black ("spin up") and white ("spin down", displayed as gray in the figures), and the vacancies, we introduce a spin variable $\sigma_{\mathbf{r}}$ at each site which can take three values: $+1$ (-1) if the site is occupied by a black (white) particle, and $\sigma_{\mathbf{r}} = 0$ if it is empty. Multiple occupancy is forbidden. The numbers of black (N^+) and white (N^-) particles are conserved and differ by at most 1. The number of vacancies, M, is much smaller: $M \ll N^+$. In fact, most of our simulations will be restricted to $M = 1$, to model the minute vacancy concentrations in real systems.

The particles and vacancies interact with one another according to a dilute Ising model:

$$\mathcal{H}[\sigma_{\mathbf{r}}] = -J \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} \sigma_{\mathbf{r}} \sigma_{\mathbf{r}'} \tag{1}$$

with a ferromagnetic, nearest-neighbor coupling $J > 0$. Since the dilution is so small, the behavior of the system is that of the ordinary (non-dilute) two-dimensional Ising model. Thus, it has a phase transition, from a disordered to a phase-segregated phase, at the Onsager critical temperature, $T_c = 2.267...J/k_B$ [18]. The ground state is doubly degenerate. It consists of a strip of black and a strip of white particles, each filling half the system, separated by two planar interfaces running parallel to a lattice axis. Since the particle-particle interactions are ferromagnetic, only few vacancies will accumulate at the interfaces.

Next, we turn to the dynamics of the model. Only particle-hole exchange is allowed and performed with the usual Metropolis [19] rate: $\min\{1, \exp(-\beta \Delta \mathcal{H})\}$, where $\Delta \mathcal{H}$ is the energy difference of the system before and after the jump, and $\beta = 1/(k_B T)$ is the inverse temperature. The initial configuration of the system is perfectly phase-segregated, with two interfaces chosen to lie along the x-axis. The vacancies are located at random positions along one of the interfaces. Since this is technically a zero-temperature configuration, while the dynamics occurs at temperature $T > 0$, the vacancies will move around, disordering the interfaces and, if $T > T_c$, dissolving them eventually. The final steady state is an equilibrium state of the usual two-dimensional Ising model. Therefore, many of its properties are exactly known [20].

In our simulations, the system size is $L = 60$. The final temperature T, measured in units of the Onsager temperature T_c, varied between $1.1T_c$ and infinity. Our data are averages over 10^2 to 10^4 realizations (or runs) depending on the desired quality of the data. The time unit is one Monte Carlo step (MCS) which corresponds to M attempted particle-hole exchange. All systems investigated equilibrate after about 10^8 MCS.

To monitor the evolution of the system, we measure a "disorder parameter", defined as the average number of black and white nearest-neighbor pairs, $\mathcal{A}(L, t)$, as a function of (Monte Carlo) time, t. This quantity is easily related to the Ising energy,

$$\mathcal{A}(L, t) = \frac{d}{2} L^d + \frac{1}{2J} \langle \mathcal{H} \rangle + O(L^\gamma) \tag{2}$$

where $\langle \cdot \rangle$ denotes the configurational average over runs. The correction is due to the vacancies that remains much smaller than the two leading terms.

III. MONTE CARLO SIMULATION RESULTS

In this Section, we show the computer simulation results of various cases: starting from the simplest case, up-quenches to infinite temperature, and to the more complicate finite temperature. We shall describe the results for these two cases as follows

A. The infinite temperature results: $T = \infty$.

For the infinite temperature, consequently the nearest-neighbor coupling J plays no role, and all attempted particle-hole exchange are executed with unit rate. Thus, the vacancies perform a Brownian random walk, regardless of their local environment. Resulting in a rougher interface as time progresses. To illustrate the gradual destruction of the interfaces and the disordering of the bulk, Fig. 1 shows the evolution of a typical configuration, in a 60×60 system with a single vacancy (represented by a white square in the figure). At $t = 0$, the vacancy is located at the interface in the center which begins to break up slowly. Eventually, the second interface also becomes affected. As more particles are transported into regions of opposite color, the system finally becomes completely disordered. For later reference, we note that the *last two* configurations, at 10^7 and 10^8 MCS, are both already fully random. The disordering process is clearly reflected in the number of broken bonds: $\mathcal{A}(60, t)$, shown in Fig. 2, increases from its minimum of $O(L)$ for the initial configuration at $t = 0$, to $O(L^2)$ for the fully equilibrated system at $t = 10^7$. One clearly distinguishes three regimes, shown schematically in the inset: the *early* regime (I), the *intermediate* or *scaling* regime (II), and finally the *late* or *saturation* regime (III) in which the system has effectively reached the steady state. Tracking the motion of the vacancy, the physical origin of these three regimes is easily identified. For *early* regime (I), the vacancy is still localized in the vicinity of its starting point, far from the boundaries of the system. After $O(L^2)$, however, the vacancy has explored the whole system and is effectively equilibrated. This marks the onset of the *intermediate* regime (II). The particle distribution is still strongly inhomogeneous and does not equilibrate until the system enters the *saturation* regime (III). We emphasize that the second and third regimes emerge only in a *finite* system. In an infinite system, regime (I) persists for all times.

B. The finite temperature results: $T_c < T < \infty$.

When the final temperature of the system is *finite*, $T < \infty$, the full Hamiltonian, Eqn (1), comes into play when a vacancy attempts to move. In particular, the defects no longer perform a simple random walk, since the jump rates now carry information about the local environment, i.e., the distribution of black and white particles around the originating and receiving site. Thus, a feedback loop between vacancy and background is established, in stark contrast to the case $T = \infty$. One might expect that this has significant consequences for profiles and disorder parameter, and that dynamic scaling exponents might be modified. It is immediately obvious that we should distinguish final temperatures above T_c from those below. In the former case, the final steady-state configurations are still homogeneous, even if correlations become more noticeable. In the latter case, however, the equilibrium state itself is phase-separated, so that the only effect of the vacancies is to "soften" the initial interfaces. This aspect of vacancy-induced disordering should be particulary interesting in two dimensions, where Ising interfaces are known to be rough [21]. There, the vacancies must mediate both the development of an "intrinsic interfacial width" and the emergence of large-scale wanderings of the interface [22]. Certainly, we expect these two phenomena to take place on drastically different time scales, since they are associated with local *vs.* global disorder.

In this Section, we focus on final temperatures *above* criticality: $T_c < T < \infty$, and a single vacancy, $M = 1$. Starting with a set of typical evolution pictures, we investigate the dynamic scaling properties of the disorder parameter. Since the equations of motion now become highly nonlinear, we have not been able to solve them exactly. Thus, we offer only a few comments in conclusion.

Fig. 3 shows the evolution of a typical configuration, in a 60×60 system with a single vacancy at $T = 1.5T_c$. Note that the MC times are the same as in Fig. 1. Again, we observe the gradual, yet eventually complete, destruction of the interfaces. In comparison with Fig. 1, however, two obvious differences emerge: First, the system takes longer time to reach the final steady state. Second, the final configuration shows clear evidence of a $finite$ correlation length. Both features are induced by the interactions. In particular, the disordering process is slowed down since the breaking of bonds is energetically costly. At a more quantitative level, this is documented in Fig. 4 which shows the disorder parameter, for a 60×60 system at several temperatures. One observes that the late crossover time shifts to later times as the temperature decreases. In contrast, the early crossover time appears to be less affected. Also, the saturation value of the disorder parameter decreases. However, being essentially the Ising energy, it remains extensive, with a T-dependent amplitude.

We comment briefly on some analytic results. First, we can conclude that the saturation values, $\mathcal{A}_{sat}(L)$, are consistent with exact results for the two-dimensional Ising value. Table 1 shows this comparison for a 60×60 system with a single vacancy at several temperatures. $\mathcal{A}_{sat}^{MC}$ is the *measured* saturation value of the disorder parameter. $\mathcal{A}_{sat}^{TH}$ is calculated on the basis of Eqn (2), where $\langle \mathcal{H} \rangle$ is the *exact* bulk value for the average energy of the usual Ising model [20]. The effect of the vacancy has been neglected. The agreement is within the statistical errors of our data. The discrepancies are of course largest for the last column, with T closest to T_c.

IV. SUMMARY AND CONCLUSION

To summarize, we have analyzed the vacancy-driven disordering process of an initially phase segregated binary system that is rapidly heated to a temperature T above criticality. To quantify the evolution of the system, we measure the number of broken bonds, $\mathcal{A}(L,t)$, as a function of time. This disorder parameter allows us to identify three temporal regimes, distinguished by the distribution of the vacancies through the system. The temporal growth of $\mathcal{A}$ during the latter regime is captured by the exponent ~ 0.5. If the final temperature is infinite, $T=\infty$, the motion of the vacancies is a simple Brownian random walk. For *finite* temperature $T \gtrsim 1.5T_c$, we observe that these exponents still hold. Even though interparticle interactions now play an important role, correlations in the system are still short-range. And the vacancy is still a random walk by nature when the length scale exceeds the correlation length. As T approaches T_c, however, the correlation length reaches $O(L)$, the simple random walk scenario breaks down.

All the investigations above are focused on an up-quenches to the disordered state. A natural extension of this project is to set the final temperature below criticality. Needless to say, new questions and interesting phenomena can be expected. In particular, the final equilibrium state is still phase separated and all the interfacial properties come into play. With a single vacancy in a finite system, there may be a further separation of times scales. It is expected that soon after the early stage, the intrinsic density profile of the interface can be built. At this point, the interfacial width is most likely controlled by the correlation length . In the $d=2$ case, the interface is always rough, so that we can always expect the latter crossover to occur. We could study the equilibrium probability profile of the *vacancy*, i.e., where the vacancy spends most of its time. In case more than one vacancy are present, this profile should map into a density profile for the vacancies. In either case, it is expected that the vacancies will be trapped at the interface. Now, if more vacancies are added to system, the interface profile should be altered, since a preponderance of vacancies may significantly modify the interfacial energy. For $d=3$ Ising models, there are further interesting phenomena, associated with roughening transitions. Given that interfacial energies should depend on vacancy concentrations, how the locations (if not the nature) of such transitions are affected is a natural question. Finally, it would be extremely interesting to study the dynamic content of these systems, such as scaling properties when the up-quench is set at a roughening temperature.

We conclude that even though our model is very simple, it forms the basis for the description of a large variety of related systems. Moreover, it is truly expected that considerable analytic progress is possible for a problem that is both *nonlinear* and *time-dependent*.

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T/T_c	∞	3.5	1.5	1.1
$\mathcal{A}_{sat}^{TH}$	3600.0	3134.6	2367.0	1602.9
$\mathcal{A}_{sat}^{MC}$	3598.6	3134.0	2368.9	1615.7

TABLE I. Comparison of exact and measured values of the saturated disorder parameter for several temperatures.

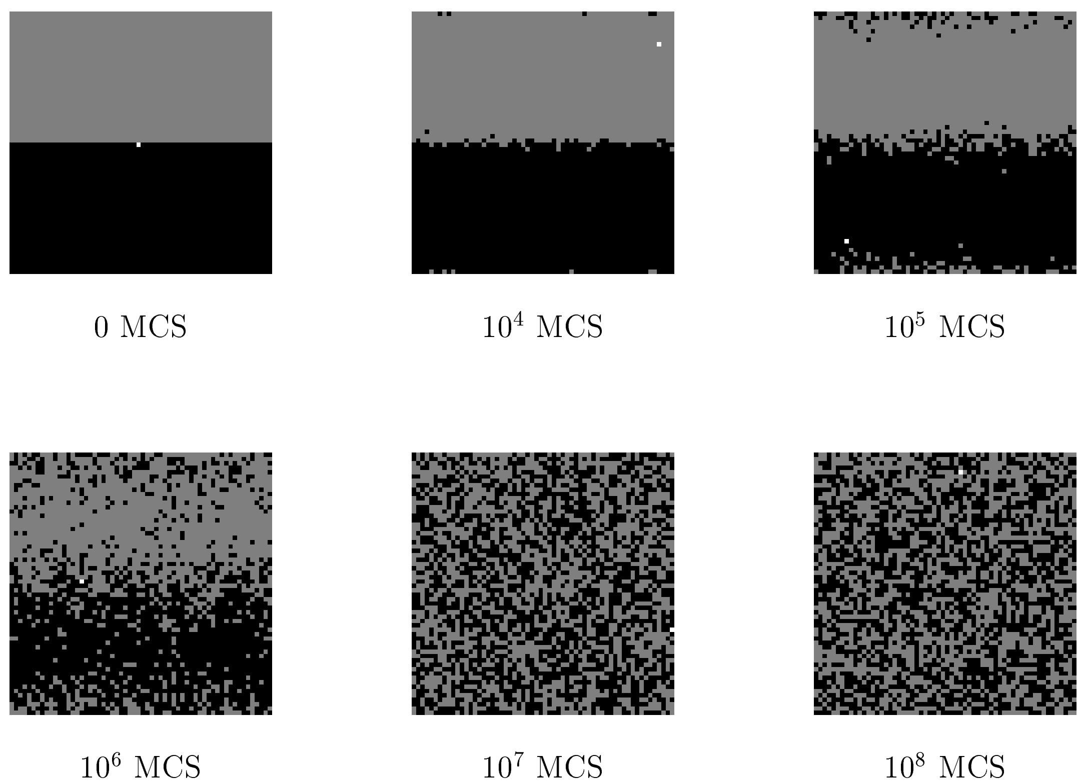

FIG. 1. Sequence of snapshots showing the disordering process of a 60×60 system with $T = \infty$. The black and gray squares represent the two types of particles ($\sigma = \pm 1$) and the white square denotes the vacancy ($\sigma = 0$). The configurations were recorded after $0, 10^4, 10^5, 10^6, 10^7$ and 10^8 MCS.

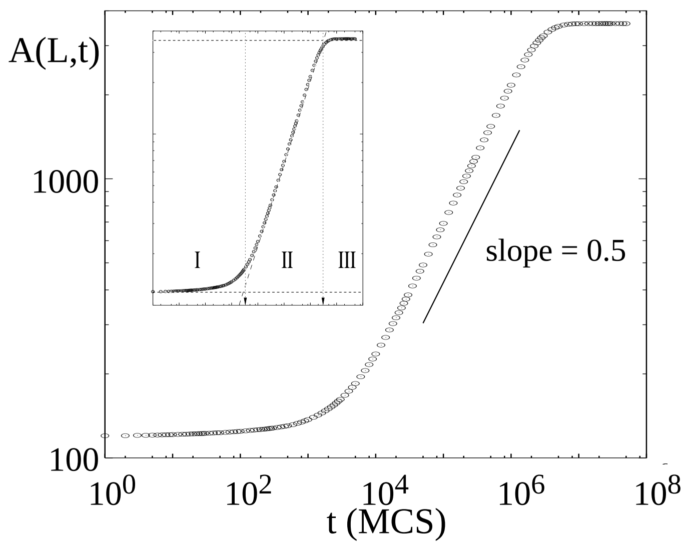

FIG. 2. Plot of the total number of broken bonds, $A(L,t)$ vs t, for $L = 60$ and $T = \infty$. It shows the emergence of an *early* regime (I), an *intermediate* or *scaling* regime (II), and a *late* or *saturation* regime (III). The straight reference line has slope 0.5.

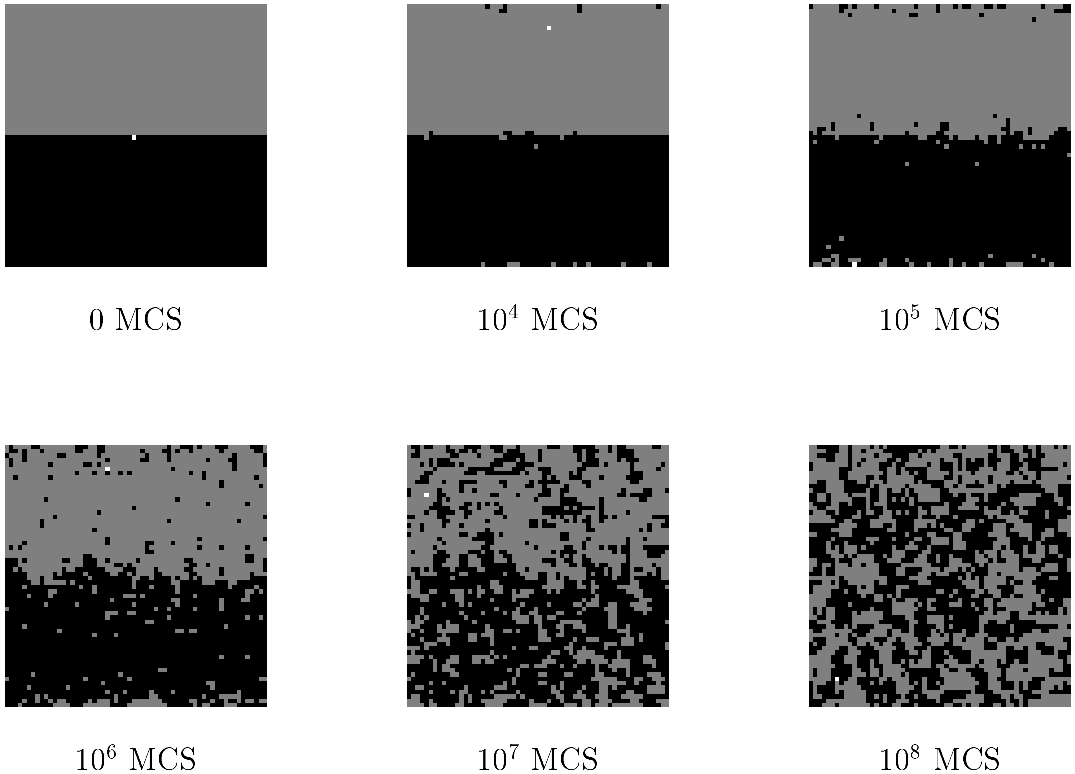

FIG. 3. Sequence of snapshots showing the disordering process of a 60×60 system with $T = 1.5T_c$. The black and gray squares represent the two types of particles ($\sigma = \pm 1$) and the white square denotes the vacancy ($\sigma = 0$). The configurations were recorded after $0, 10^4, 10^5, 10^6, 10^7$ and 10^8 MCS.

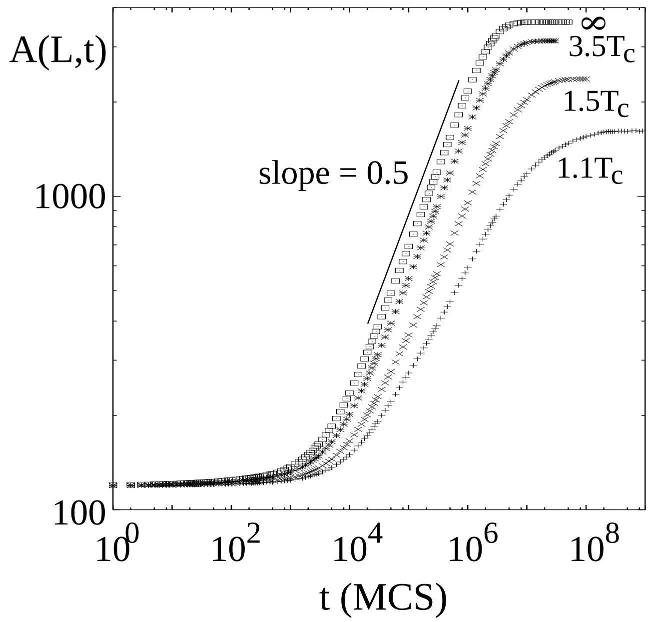

FIG. 4. Plot of $A(L,t)$ vs t for $L = 60 \times 60$ at $T = \infty$, $3.5T_c$, $1.5T_c$ and $1.1T_c$. This shows the increasing in the disordering parameter at the equilibrium state as the final temperature increases.