Dimensionality Dependence of Aging in Kinetics of Diffusive Phase Separation: Behavior of order-parameter autocorrelation

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Behavior of two-time autocorrelation during the phase separation in solid binary mixtures are studied via numerical solutions of the Cahn-Hilliard equation as well as Monte Carlo simulations of the Ising model. Results are analyzed via state-of-the-art methods, including the finite-size scaling technique. Full forms of the autocorrelation in space dimensions 2 and 3 are obtained empirically. The long time behavior are found to be power-law type, with exponents unexpectedly higher than the ones for the ferromagnetic ordering. Both Chan-Hilliard and Ising models provide results consistent with each other.

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I. INTRODUCTION

Properties of a nonequilibrium system change with growing age \[1\]. Understanding of such aging phenomena is of fundamental importance in all branches of science and technology. There have been serious activities on this issue concerning living \[2, 3\] as well as nonliving matters, especially in problems related to domain growth \[1, 4–14\] and glassy dynamics \[15–19\]. Among other quantities, aging phenomena is studied via the two-time autocorrelation function \[4\]

\[
C(t, t_w) = \langle \psi(\vec{r}, t)\psi(\vec{r}, t_w) \rangle - \langle \psi(\vec{r}, t)\rangle \langle \psi(\vec{r}, t_w)\rangle.
\]

In Eq. (1), \(\psi\) is a space \((\vec{r})\) and time dependent order parameter, \(t_w\) is the waiting time or age of the system and \(t (> t_w)\) is the observation time.

In phase ordering systems \[20\], though time translation invariance is broken, \(C(t, t_w)\) is expected to exhibit scaling with respect to \(t/t_w\). Important examples are ordering of spins in a ferromagnet, kinetics of phase separation in a binary \((A + B)\) mixture, etc., having been quenched to a temperature \((T)\) below the critical value \((T_c)\), from a homogeneous configuration. Though full forms are unknown even for very simple models, asymptotically \(C(t, t_w)\) is expected to obey power-law scaling behavior as \[4, 6\]

\[
C(t, t_w) \sim x^{-\lambda}; \; x = \ell/\ell_w.
\]

In Eq. (2), \(\ell\) and \(\ell_w\) are the average sizes of domains, formed by spins or particles of similar type, at times \(t\) and \(t_w\), respectively. Typically \(\ell\) and \(t\) are related to each other via power-laws.

For nonconserved order-parameter dynamics, e.g., ordering in a ferromagnet, such scaling has been observed and the values of the exponent \(\lambda\) have been accurately estimated \[6, 14\] in different space dimensions \(d\). There the exponents follow the bounds

\[
\frac{d}{2} \leq \lambda \leq d,
\]

(3)
predicted by Fisher and Huse (FH) \[4\]. In kinetics of phase separation in solid mixtures, for which the order parameter is a conserved quantity, the state of understanding is far from satisfactory, due to theoretical as well as computational difficulties. There exist reports \[11\] of violation of scaling with respect to $t/t_w$. The latter observation, our results indicate, is due to the fact that scaling is achieved for $t_w \gg 1$. Access to such long time is constrained by inadequate computational resources. This difficulty, in some studies \[9, 13\], might have led to the conclusion about incorrect values of $\lambda$.

In an important work, Young et al. \[7\] put a more general lower bound on $\lambda$, valid irrespective of the conservation of the total order parameter, as

$$\lambda \geq \frac{\beta + d}{2}, \quad (4)$$

where $\beta$ is the exponent for small wave-vector power-law enhancement of equal time structure factor which, depending upon the dynamics, becomes important for $t_w \gg 1$, as stated below. In nonconserved dynamics, $\beta = 0$ and so the lower bound in Eq. (3) is recovered. For conserved order parameter dynamics, on the other hand, $\beta = 4$ in both $d = 2$ and 3 at late time. Thus, the upper bound in Eq. (3) is violated. Simulations of the Cahn-Hilliard (CH) equation \[20\]

$$\frac{\partial \psi(\vec{r}, t)}{\partial t} = -\nabla^2 \left[ \psi(\vec{r}, t) + \nabla^2 \psi(\vec{r}, t) - \psi^3(\vec{r}, t) \right], \quad (5)$$

by Young et al. \[7\], observed $\lambda > 3$ in $d = 2$, consistent with Eq. (4). From these simulations, the authors, however, did not accurately quantify $\lambda$; scaling of $C(t, t_w)$ with respect to $t/t_w$ was not demonstrated; focus was rather on the sensitivity of the aging dynamics to the correlations in the initial configurations. Situation is far worse in $d = 3$, with respect to the CH equation as well as the Ising model \[1, 20\]

$$H = -J \sum_{<ij>} S_i S_j; \quad S_i = \pm 1; \quad J > 0. \quad (6)$$
In this paper, we study both CH equation and the Ising model, used for understanding diffusive phase separation as in solid mixtures, in \( d = 2 \) (on regular square lattice) and \( d = 3 \) (on simple cubic lattice), via extensive simulations, to quantify the decay of \( C(t, t_w) \). We observe scaling of \( C(t, t_w) \) with respect to \( x \) in which the power-law of Eq. (2) is realized for large \( x \). Via computations of the instantaneous exponent \[ \lambda_i = -\frac{d \ln[C(t, t_w)]}{d \ln x}, \] and application of the finite-size scaling technique \[24, 25\], we find that \( \lambda \approx 3.6 \) in \( d = 2 \) and \( \approx 7.5 \) in \( d = 3 \). Though these numbers respect the bounds in Eq. (4), the high value in \( d = 3 \) is surprising. But this comes from both CH and Ising models, from various reliable analyses. Furthermore, a general analytical form for the full scaling functions has been obtained empirically.

II. METHODS

One numerically solves the CH equations on a regular lattice, usually via Euler discretization method. With the Ising model, phase separation kinetics in a solid binary mixture is studied via the Kawasaki exchange Monte Carlo (MC) \[25\] simulations, to be referred to as KIM. An up spin \((S_i = +1)\), for this problem, may correspond to an \( A \) particle and a down spin \((-1)\) to a \( B \) particle. In this MC scheme, one randomly chooses a pair of nearest neighbor spins and tries their position exchange. The moves are accepted according to standard Metropolis algorithm \[25\]. Due to the coarse-grained nature of the CH equation, as opposed to the atomistic Ising model, one can explore large effective length in simulations. The order parameter in Eq. (5) corresponds to a coarse-graining \[26\] of the Ising spins, typically over the equilibrium correlation length \( \xi \). Then, a positive value of \( \psi \) means an \( A \)-rich region and for a \( B \)-rich region, \( \psi \) will have a negative number. For the calculation of \( C(t, t_w) \), we have used binary numbers +1 and
The average domain length, \( \ell \), in our simulations was measured from the first moment of domain size distribution, \( P(\ell_d, t) \), as

\[
\ell = \int \ell_d P(\ell_d, t) \, d\ell_d,
\]

where \( \ell_d \) is the distance between two successive domain boundaries in any direction. Throughout the paper, all lengths are presented in units of the lattice constant \( a \). In MC simulations, time is counted in units of Monte Carlo steps (MCS), each MCS consisting of \( L^d \) trial moves, where \( L \) is the linear dimension of a periodic square or cubic system. In CH equation, \( t \) is expressed dimensionless units [27]. All results are presented after averaging over at least 50 initial configurations, for quenches from random initial configurations to \( T = 0.6T_c \).

## III. RESULTS

In Fig. 1(a), we present the plots of \( C(t, t_w) \), vs \( x \), for different values of \( t_w \), from the solutions of CH model in \( d = 2 \). As seen, one needs large enough value of \( t_w \) to observe appropriate scaling behavior, compared to ordering in ferromagnets [14]. Between the two data sets with largest values of \( t_w \), the deviation from each other, for large \( x \), is due to the finite-size effects. Similar plots for the \( d = 3 \) CH model are presented in Fig. 1(b). Here all \( t_w \)’s are large enough, providing good scaling. Again, deviation from the master curve, starting at different values of \( x \) for different \( t_w \), are primarily related to the finite-size effects. In both these figures, 1(a) and 1(b), the system sizes are kept fixed, only the values of \( t_w \) are varied. A similar observation, with respect to the above mentioned deviation for different choices of \( t_w \), can be made, when, for same value of \( t_w \), data are presented for different system sizes.

In the scaling parts, both in Fig. 1(a) and Fig. 1(b), continuous bending is observable, in these log-log plots. Thus, power-laws, if exist, carry corrections. The solid lines in these figures
FIG. 1. (a) Autocorrelation function, $C(t, t_w)$, from the $d = 2$ Cahn-Hilliard model, are plotted vs $x = \ell/\ell_w$, for different values of $t_w$. The solid line there corresponds to a power-law decay with exponent 3. (b) Same as (a) but for the $d = 3$ CH model. The solid line there has a power-law decay exponent 3.5. The system sizes used are $L = 256$ ($d = 2$) and 200 ($d = 3$).

are power-law decays with exponents 3 and 3.5, respectively, corresponding to the bounds in Eq. (4). For large $x$, simulation data in $d = 2$ appear reasonably consistent with the bound. The asymptotic exponent, in $d = 3$, on the other hand, appear much higher than 3.5.

With the expectation that power laws indeed exist, in Fig. 2 we present plots of instantaneous exponents $\lambda_i$ for both $d = 2$ and 3, vs $1/x$. In addition to providing $\lambda$, from the extrapolations to $x = \infty$, such exercise may be useful for obtaining crucial information on the full forms of $C(t, t_w)$. For $d = 2$, the data are obtained for $t_w = 5 \times 10^3$, and for $d = 3$, the data correspond to $t_w = 10^3$. In both the cases, the results appear reasonably linear.
FIG. 2. Instantaneous exponents $\lambda_i$ are plotted vs $1/x$. Results are shown only from the solutions of the CH equations, in both $d = 2$ and 3. The solid lines are guides to the eye. The $d = 2$ data are for $t_w = 5 \times 10^3$ with $L = 400$. In $d = 3$ the numbers are $10^3$ and 200.

The solid lines there are extrapolations to $x = \infty$, accepting the linear trends. These indicate $\lambda \simeq 3.60$ in $d = 2$ and $\simeq 7.80$ in $d = 3$. Again, while the value in $d = 2$ is consistent and close to the bound of Yeung et al., the observation of surprisingly high number in $d = 3$ is certainly interesting. We intend to obtain more accurate values via appropriate finite-size scaling analyses [24, 25]. This is considering the fact that the choice of the regions in Fig. 2 for performing least-square fitting, is not unambiguous due to finite-size effects and strong statistical fluctuations at large $x$. Also, for very small $x$ (data excluded), there is rapid decay of $C(t, t_w)$ related to the fast equilibration of domain magnetization.

Since the corrections to the asymptotic decay laws are seen to be strong for finite $x$, a reasonable idea about the full forms of the decays is essential for accurate finite-size scaling analyses. Those, however, are nonexistent in the literature. Here we obtain the forms empirically. Assuming power-law behavior of the data sets in Fig. 2 we write

$$\lambda_i = \lambda - \frac{A_c}{x^\gamma},$$

(9)
FIG. 3. Finite-size scaling plot of $C(t, t_w)$ from $d = 2$ CH model. The scaling function $Y$ is plotted vs $y$, using data from different system sizes. The optimum collapse of data, the presented one, was obtained for $\lambda = 3.47$. The inset presents the same exercise for the CH model in $d = 3$. Here the value of $\lambda$ is 7.30. See text for values of $t_w$.

where $A_c$ and $\gamma$ are constants. Combining Eq. (9) with Eq. (7), we obtain

$$C(t, t_w) = C_0 \exp \left( - \frac{A_c}{\gamma x^\gamma} \right) x^{-\lambda},$$

$C_0$ being a constant. For finite-size scaling analysis, one needs to introduce a scaling function

$$Y(y) = C(t, t_w) \exp \left( \frac{A_c}{\gamma x^\gamma} \right) x^\lambda; \quad y = L/\ell.$$  

For appropriate choices of $A_c$, $\gamma$ and $\lambda$, one should obtain a master curve for $Y$, when data from different system sizes are used. The behavior of $Y$ should be flat in the finite-size unaffected region and a deviation from it will mark the onset of finite-size effects.

By examining the data in Fig. 2 (also see Fig. 4(b) for KIM), we fix $\gamma$ to 1. In the main frame of Fig. 3 we show a finite-size scaling plot for data from the $d = 2$ CH model. The presented results correspond to best collapse, obtained for $A_c = 2.25$ and $\lambda = 3.47$. The value of $t_w$ used for all the data sets is $10^4$. A similar exercise for the $d = 3$ CH data is presented in the inset of Fig. 3. Again the data collapse looks quite reasonable and was obtained for $A_c = 5.1$ and $\lambda = 7.30$. The value of $t_w$, in this case, was set to $10^3$. The reason behind
choosing smaller value of $t_w$ in $d = 3$, than in $d = 2$, is computational difficulty. It is extremely difficult to accumulate data for further decades in time, starting from very high value of $t_w$, particularly in $d = 3$. Nevertheless, this chosen value of $t_w$ falls within the scaling regime. Note that for similar temperatures, amplitude of growth is larger in $d = 3$ and the scaling of $C(t, t_w)$ is related more closely to the value of $\ell_w$. In this connection we mention that the longest run lengths (associated with largest systems) for the CH model are $t = 2 \times 10^6$ and $2 \times 10^5$ in $d = 2$ and 3, respectively; for the Ising model these numbers are $5 \times 10^7$ and $4 \times 10^6$.

![Graphs showing autocorrelations](image)

**FIG. 4.** (a) Same as Fig. 1(a) but for the $d = 2$ Ising model. (b) Same as Fig. 2 but for Ising model.

The oscillatory behavior of large $x$ data in $d = 2$ is statistical fluctuation. In $d = 2$ the results are from $L = 512$ and in $d = 3$, we have used $L = 100$.

We now move to present results from KIM. In Fig. 4(a) we show the autocorrelations from
different values of $t_w$ in $d = 2$, for $L = 512$. Scaling is poor for $t_w$ below $10^4$ MCS and so those results are excluded. Despite strong statistical fluctuations, it is recognizable that the decay of $C(t, t_w)$ in the latter part is on the higher side of the bound in Eq. (1), represented by the solid line.

![Graph showing $C(t, t_w)$ vs $x$ for the CH model in $d = 2$ and $d = 3$. The inset shows corresponding results for the KIM. The solid lines are fits to the form in Eq. (10), with $\gamma = 1$. The $t_w$ values are mentioned on the figure. We have discarded data suffering from finite-size effects. The system sizes are $L = 400$ and $512$ for CH and Ising models in $d = 2$, whereas, $L = 200$ and $100$ in $d = 3$.]

FIG. 5. $C(t, t_w)$ are plotted vs $x$, for the CH model in $d = 2$ and $3$. Inset shows corresponding results for the KIM. The solid lines are fits to the form in Eq. (10), with $\gamma = 1$. The $t_w$ values are mentioned on the figure. We have discarded data suffering from finite-size effects. The system sizes are $L = 400$ and $512$ for CH and Ising models in $d = 2$, whereas, $L = 200$ and $100$ in $d = 3$.

In Fig. 4(b) we show the instantaneous exponents for the Ising model in $d = 2$ and $3$, vs $1/x$. In each dimension, we have included two values of $t_w$ from the scaling regime. While results for different $t_w$s, in a particular dimension, are consistent with each other, finite-size effects appear earlier for larger value of $t_w$, as expected. Thus, for extrapolations to $x = \infty$, data sets with smaller $t_w$ are used. This exercise provides $\lambda \simeq 3.60$ and $\simeq 7.30$ in $d = 2$ and 3, respectively. These values are in agreement with the ones obtained for the CH model via various methods of analysis.

Finally, in Fig. 5 we show the fits of the simulation data to the form in Eq. (10). The main frame is for the CH model in $d = 2$ and $3$, whereas the inset contains similar results from the
KIM. The fits look quite satisfactory. This exercise provides $\lambda = 3.55$ and $3.76$ in $d = 2$ for CH and Ising models, respectively. The numbers in $d = 3$ are $7.64$ and $7.37$.

IV. CONCLUSIONS

In conclusion, we have studied aging dynamics for the phase separation in solid binary mixtures via Cahn-Hilliard and Ising models. Results for the two-time autocorrelation, $C(t, t_w)$, are presented from simulations in both $d = 2$ and $3$. Decays of $C(t, t_w)$ appear power law in large $x$ limit. The exponents for these power laws were obtained via various different analyses, including finite-size scaling, which is new for this purpose. For the finite-size scaling analysis, full forms of the autocorrelations were essential which we obtained empirically. All these methods provide consistent values of the decay exponent $\lambda$ for different models. These are $\lambda \simeq 3.6$ in $d = 2$ and $\lambda \simeq 7.5$ in $d = 3$, within 5% error.

Very high value of $\lambda$ in $d = 3$, far above the lower bound in Eq. (1), can be due to the fact that domains are more mobile in this dimension than in $d = 2$. From the numbers obtained in $d = 2$ and $3$, it may be tempting to predict a dimensionality dependence as $\lambda = f(d - 1)$ with $f \simeq 3.75$. However, we caution the reader not to jump into such conclusion. Even though the influence of the dimension $d$ appear more important than in the bound of Eq. (1), the contribution of $\beta$ is significant, particularly at lower dimension. Results from other dimensions are necessary to make such a conclusion. In $d = 1$ one should exercise the caution that $\beta (= 2)$ has a different value [28].
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