

Phase separation in two-dimensional binary fluids: A molecular dynamics study

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We present results of molecular dynamics computer simulation experiments of phase separation in a two-dimensional model binary fluid. For critical quenches the tubularlike domain structure grows at late times according to a power law with an exponent $1/2$ which later crosses over to $2/3$. This latter result has recently been questioned by Ossadnik *et al.* [Phys. Rev. Lett. **72**, 2498 (1994)], and we present further evidence that the fast exponent is indeed present in the dynamical growth. That these exponents are hydrodynamic in origin is demonstrated by performing runs where the velocity field is disturbed at regularly spaced intervals of time. The perturbation consists of resetting the velocities of the particles to those characteristic of a Boltzmann distribution, which destroys the hydrodynamic tail of the velocity autocorrelation function. The resulting exponent changes to that predicted by experiments using stochastic techniques (Monte Carlo), i.e., $1/3$. Also, we present results of experiments for off-critical quenches. The late-stage growth exponent for not too off-critical concentrations is close to $1/3$, but we observe a continuous transition from this exponent to that characteristic of a critical quench. [S1063-651X(96)02906-6]

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I. INTRODUCTION

Recently there has been much interest in the kinetic processes which occur when a binary fluid is quenched into the region of liquid-liquid immiscibility [1]. Binary fluids are especially interesting systems since hydrodynamic effects are expected to play a significant role in the dynamics of phase separation [2]. In particular, these effects have been observed to accelerate the formation of domains of the two separating phases [3].

The growth of domains is governed in the scaling regime by a power-law relation of the form $R(t) \propto t^n$, where t is time, $R(t)$ is the average domain size, and n is an exponent characterizing the growth. Numerical values for the exponents depend on the character and conservation law of the order parameter and the dimensionality. In previous work [4,5], we have applied the technique of molecular dynamics (MD) to study the late-stage separation of a symmetric binary fluid mixture in two dimensions (2D). In this model molecules of the same species interact through Lennard-Jones (LJ) potentials, whereas molecules of different species interact through a purely repulsive potential. We found quite different exponents for the cases where the concentration x_A of one of the species is critical ($x_A = 1/2$) and off critical ($x_A \neq 1/2$). The exponents obtained, $2/3$ for critical and $\sim 1/3$ for off-critical quenches, can be rationalized by the different morphology of the domain structures. However, recent work by Ossadnik *et al.* [6] using similar simulation techniques questioned our results for the critical quench experiments. These authors performed averages over 12 independent realizations of the separation dynamics and concluded, as found previously by us, that the growth exponent is $1/2$ but that our claim from our earlier simulations that there is a late-stage, fast-growth exponent was an artifact of the lack of statistical averaging in our study.

In an effort to verify the accuracy of our previous results and collect more statistically significant information, we un-

dertook further numerical experiments and used different probes to measure the size of the structure, namely, the structure factor and the internal energy per molecule. We believe the latter to be the more accurate and simple probe. Our new results clearly show that for critical quenches the late-time exponent is indeed $2/3$ and that hydrodynamic flow is responsible for the fast growth. This is demonstrated by suppressing the hydrodynamic modes in the deterministic MD simulation, which brings about a slowing down of the growth. The hydrodynamic modes can be removed from the system by perturbing the velocity distribution of the ensemble, which destroys the tail of the velocity autocorrelation function. These results reveal the validity of MD simulation as a valuable tool in kinetics studies of phenomena influenced by hydrodynamics and cast doubts on the validity of Monte Carlo, a purely stochastic technique, as a convenient methodology in investigations of phase separation processes in real fluids. Our MD simulation study has also been extended to off-critical quenches, which show a progressively slower dynamical growth as the solute fraction is decreased.

The paper is organized as follows. The next section sets up the interaction model and gives some details of the simulations. In Sec. III we present the results of our computer experiments and end up in Sec. IV with a discussion and a summary.

II. INTERACTION MODEL AND SIMULATION TECHNIQUE

We consider a symmetric binary fluid made up of two species of molecules, A and B . In order to induce phase separation it is usual practice to model the interactions with nonadditive repulsive cores and possibly attractive potentials with different depths. However, it is computationally convenient, though not necessarily realistic, to induce the phase separation by simply considering equal-sized molecules for

all the interactions but a less attractive AB interaction than the AA and BB interactions. A rather extreme case is when the interaction potentials are of the form

$$\phi_{AA}(r) = \phi_{BB}(r) = \phi_{LJ}^-(r), \quad \phi_{AB}(r) = \phi_{LJ}^+(r), \quad (1)$$

with

$$\phi_{LJ}^\pm(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} \pm \left(\frac{\sigma}{r} \right)^6 \right]. \quad (2)$$

In our previous work [4] we used this model but with a power 3 instead of 6 for the tails of the potentials. The above interactions, which will be henceforth called ‘‘LJ $^\pm$,’’ are used in the present work. In addition, we have considered the following modification of the AB interaction:

$$\phi_{AB}(r) = \phi_{WCA}(r), \quad (3)$$

with

$$\phi_{WCA}(r) = \begin{cases} 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] + \epsilon, & r < 2^{1/6}\sigma, \\ 0, & r \geq 2^{1/6}\sigma. \end{cases}$$

Here the components A and B correspond to pure Lennard-Jones (LJ) fluids whereas molecules of types A and B interact through a purely repulsive WCA potential. This model will be referred to as ‘‘LJ/WCA.’’ (In the following the parameters ϵ and σ set the energy and length scales, respectively, and quantities will be given in reduced units according to these scales.)

The above interaction models are capable of inducing phase separation at sufficiently low temperatures and high densities. The phase diagrams of such mixtures have been recently calculated by us [5,7] using an improved perturbation theory of mixtures, which provided quite accurate estimates of the coexistence concentrations and distribution functions. As an example, the temperature-concentration phase diagram of the LJ-WCA mixture is reproduced in Fig. 1. We also obtained the mean-field spinodals which limit the region where separation takes place by spinodal decomposition from the nucleation region. The latter lines give us a reference with respect to which we can assess the qualitative difference in the initial and final stages of phase separation following quenches at different concentrations.

In the phase separation experiments, systems of $N=80\,000$ and $40\,000$ molecules at temperature $T=1$ and with density $\rho=0.8$ were considered. The equations of motion were integrated using the Verlet algorithm and temperature was controlled by coupling the system to a Nosé-Hoover thermostat. The time interval was $h=0.005$ in reduced LJ time units. Our experiments were aimed at studying the effect on the late-time dynamics of (i) the size of the system, (ii) the length of the time interval in which the phase separation was observed, and (iii) the averaging over independent runs. In a first series of runs, which used the bigger system and will be referred to as ‘‘experiment 1,’’ molecules interacted via LJ $^\pm$ potentials and the separation of phases was monitored for 1600 reduced time units. Six independent samples were obtained and the domain size $R(t)$ was determined from the first zero of the total correlation function (see

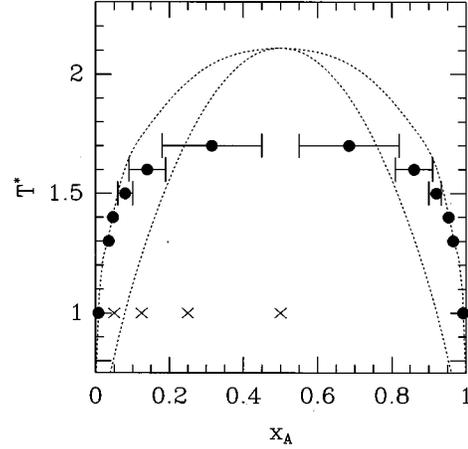


FIG. 1. The T - x_A phase diagram for a two-dimensional LJ-WCA mixture. The solid lines are the coexistence and spinodal lines as obtained from a simple mean-field theory, whereas the dashed lines correspond to a Guggenheim approximation. Dots indicate the location of the coexistence concentrations as derived from Gibbs-ensemble simulations. Crosses indicate state points at which quenches were performed.

below). The second experiment, which will be called ‘‘experiment 2,’’ used the smaller system and LJ-WCA potentials. The experiment was extended to 10^4 time units (an increase of one decade with respect to both our previous experiment and other studies [6,8]) and the dynamical behavior of a single run was characterized by the internal energy per molecule $u(t)$; this quantity is much easier to calculate than the radial distribution function, allowing the simulation to be extended to much longer times.

The critical quenches are carried out by first equilibrating a homogeneous liquid of Lennard-Jones molecules and randomly assigning each molecule to be of type A or B . This procedure ensures that no large force fields arise locally since the repulsive parts of the potentials are the same, regardless of the type of molecules in a pair. Some runs were also started from a triangular configuration. The molecule fraction was set to $x_A = 1/2$ which is the critical concentration of the liquid since the molecular interactions are symmetric with respect to A - B interchange. The temperature was chosen to be $T=1$ in reduced units; this is well below the temperature where evaporation of the coexisting liquid occurs.

In our previous work [4], we measured the average size of the domains from the first zero of the total correlation function of one of the molecular species, say A , $h_{AA}(r) = g_{AA}(r) - 1$, where $g_{AA}(r)$ is the radial distribution function of the A component. The calculation of this function entails heavy computer work. Therefore we used another probe, the excess internal energy, $u(t) - u_\infty$. This quantity is a measure of the interfacial energy, and hence also of the total length of the interface, which scales as $R(t)^{-1}$. We have monitored the excess internal energy at every time step and used the scaling relation

$$u(t) - u_\infty \propto t^{-n} \quad (4)$$

to obtain the exponent n . Here u_∞ is the energy of the coexisting phases at equilibrium, where the phases are separated

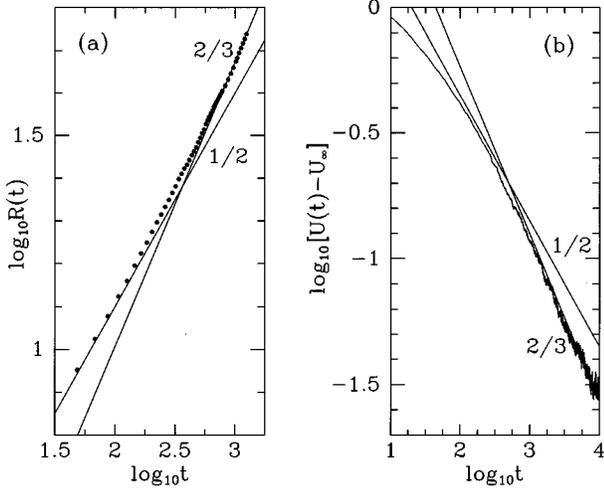


FIG. 2. Time dependence of the average domain size $R(t)$ of the growing structure with concentration $x_A=1/2$ from experiment 1 (see text), as determined from the radial distribution function of one of the species (a); excess energy $u(t) - u_\infty$ as a function of time from experiment 2 (see text) (b). In both cases straight lines of slopes $1/2$ and $2/3$ are shown for comparison.

by a flat interface. Our estimate for u_∞ was obtained by equilibrating a configuration initially consisting of two pure phases separated by a completely flat interface. This system was found to equilibrate very slowly but after 4×10^6 time steps the energy showed no drift. After equilibration averages were performed over 10^6 time steps and we obtained $u_\infty = -1.931 \pm 0.001$ [9].

III. RESULTS AND DISCUSSION

Let us first concentrate on the critical quenches, the results of which are shown in Figs. 2(a) and 2(b). In Fig. 2(a) the domain size $R(t)$ corresponds to an average over six independent runs using 80 000 molecules (experiment 1). The typical domain size shows a $1/2$ -type behavior followed by a faster growth, with an average slope of 0.67 ± 0.16 which is consistent with an exponent of $2/3$. The result of the second experiment (a critical quench using 40 000 molecules) is given in Fig. 2(b). The excess energy shows the same late-time exponent as obtained in the previous experiment from $R(t)$. Note that in Fig. 2(b) only a single run is shown, but the simulation was extended an additional decade with respect to experiment 1 and previous MD studies [6,4,8].

The exponent $2/3$ can be understood in terms of a simple dimensional analysis of the equation of motion for the order parameter, in the so-called inertial hydrodynamic regime. This exponent was first derived by Furukawa [10] for a system in 3D. Here we extend his argument to the 2D case [11] (the derivation closely follows that made in 3D [2]). The starting point is the time-dependent Ginzburg-Landau (Langevin) equation for the order parameter $\phi(\mathbf{r}, t)$,

$$\frac{\partial \phi}{\partial t} + \mathbf{v} \cdot \nabla \phi = \lambda \nabla^2 \mu. \quad (5)$$

In this equation \mathbf{v} is the local velocity field, λ is a transport coefficient, and μ is the chemical potential. At late times Eq. (5) has to be supplemented by the Navier-Stokes equation for \mathbf{v} ,

$$\rho \left(\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right) = \eta \nabla^2 \mathbf{v} - \nabla p - \phi \nabla \mu, \quad (6)$$

where p is the pressure, η the shear viscosity, and ρ the (constant) density. In the viscous regime the left-hand side terms in Eq. (6) are neglected and dimensional analysis leads to the well-known $t^{1/2}$ law (possibly with logarithmic corrections). On the other hand, neglecting the viscous term we are left with two dominant terms, the inertial term which in the scaling regime (when a typical domain size R is established) scales as $\rho R/t^2$, and the driving term due to the spatial gradient of the chemical potential, which scales as γ/R^2 , γ being the surface tension. At later times, when the inertial term starts being comparable to the driving force, we have $\rho R/t^2 \sim \gamma/R^2$, leading to $R \sim t^{2/3}$. When the condition $R \sim \eta^2/\rho\gamma$ is fulfilled the phase separating system is in the inertial hydrodynamic regime. We can check whether the latter condition holds for our fluid by assigning approximate values for the viscosity and surface tension. From independent simulations, we estimate $\gamma = 0.9 \pm 0.1$ and $\eta \approx 3.0$ for $\rho = 0.8$ and $T = 1$, [12] which gives $\eta^2/\rho\gamma \sim 12.5\sigma$. Thus the scaling arguments [2] suggest a crossover to a growth regime with an exponent of $2/3$ when the domains reach a size significantly bigger than about 12 molecular diameters, i.e., clusters of about a 100 molecules. At late times, for the growth with $n = 2/3$, our domains do certainly verify the condition $R \geq 12.5$.

The $t^{2/3}$ growth, although not observed experimentally, has been obtained by Farrell and Valls in their Langevin simulations [13] of a model closely related to model H of critical dynamics, which is the relevant continuum model for fluids. Also, Alexander *et al.* [14] and later Osborn *et al.* [15] examined a lattice Boltzmann model of hydrodynamic phase segregation and their results indicate an exponent consistent with ours. Recently, Bastea and Lebowitz [16] have also obtained phase separation in a binary mixture in the inertial regime with a power-law exponent of $2/3$, using a lattice-gas model with hydrodynamics. Our finding is to be contrasted with the results of a recent MD simulation by Ossadnik *et al.* [6], who found a $t^{1/2}$ growth law all the way up to the latest times in their runs. Interestingly, the simulations of Ossadnik *et al.* are identical to ours, except that the system size was smaller ($N = 17\,000$) and the algorithm used to integrate the equations of motion was a Gear algorithm. We have checked independently that this algorithm, although not time reversible and symplectic, unlike Verlet's, is capable of reproducing our results. At present we are unable to advance a possible explanation for the different outcomes of these (seemingly) identical experiments. Also, in another recent MD work, Leptoukh *et al.* [8] failed to confirm our predictions for the fast growth; the latter results are to be taken with circumspection, however, since the simulation times are clearly too short. In additional work we have investigated the growth of a mixture with a WCA potential for the AB interaction, and found that when the temperature is $0.79T_c$ the dynamics does not reach the fast growth within

our simulation time; a calculation of the surface tension and viscosity indicates that whereas the latter changes very little on increasing the temperature from $T=1$ to $0.59T_c$, the surface tension goes down quite significantly and the inertial hydrodynamic condition is much more severe (larger domain size). This means that the simulation time necessary to reach this regime is much longer and also the system size has to be increased. This fact seems to have been overlooked in [8].

Another point of interest in our previous work [4] was the reported presence of transient $t^{1/4}$ behavior before the late-time regime sets up. As correctly stated by the authors of Ref. [6], this behavior is a property of independent realizations, and does not appear in an average over independent runs. The $1/4$ has been associated with transport of particles along interfaces and was probably favored by our early model potential which contained a very soft repulsive and long-ranged AB interaction. In fact, with the present, much weaker interaction potentials, we have in no case obtained such transients.

With a view to assessing the role of hydrodynamic interactions in the growth behavior, we have performed some further experiments at critical concentration, where the velocity flow was disturbed at regularly spaced intervals of time. This was done by resetting the velocities of all the molecules according to a (random) Boltzmann distribution, with a variance given by the (constant) temperature of the system. The motivation for these experiments was the following. The hydrodynamic modes show up in the velocity autocorrelation function $\Psi(\tau)$ between the velocities at time t and $t + \tau$, [17]

$$\Psi(\tau) = \frac{\langle \mathbf{v}_i(t+\tau)\mathbf{v}_i(t) \rangle}{\langle \mathbf{v}_i(t)^2 \rangle} \quad (7)$$

where $\langle \rangle$ stands for an average over particles in the system. In real fluids (and also in model fluids simulated by MD) the hydrodynamic modes cause a long-time correlation of the velocities which results in a slowly (algebraically) decaying autocorrelation function $\Psi(\tau)$ [18]. On the other hand, stochastic dynamics, like Langevin dynamics and MC, do not contain these modes and the resulting autocorrelation functions decay rapidly (exponentially) to zero. When the particle velocities are perturbed no large force gradients are introduced (at least not larger than those already present); however, as the velocities are random, we cannot ensure that the forces have a certain direction. In fact they are randomized and the ‘‘memory’’ of the correlated motion is lost: we are actually destroying the autocorrelation in the velocities, Eq. (7), for times longer than the time interval between two consecutive perturbations, and it follows that the hydrodynamic modes, which are responsible for the correlated motion, are decoupled from the order parameter and may disappear.

The results of these experiments are shown in Fig. 3(a) where again the excess internal energy is represented as a function of time. In the figure different curves correspond to different time intervals τ between consecutive disturbances. When the perturbation is applied a short transient follows where the collective modes are reinstated locally. This is visible at short times by the small amplitude wiggles in the curves. At sufficiently late times these modes cannot be reinstated and they effectively disappear. When the perturba-

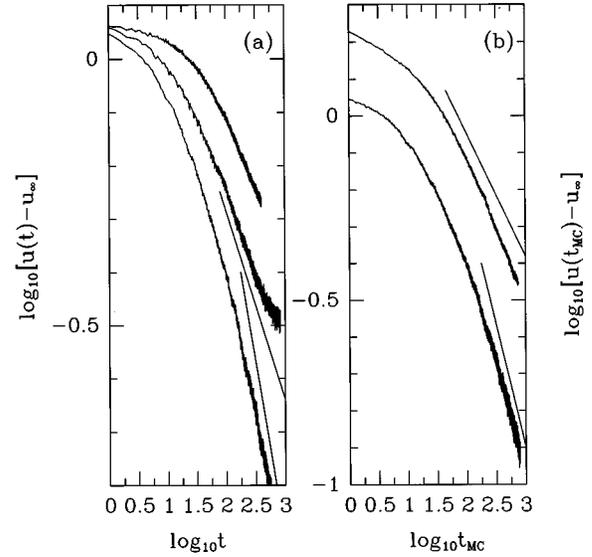


FIG. 3. Behavior of $u(t) - u_\infty$ with time for different values of the parameter τ ; from the top, $\tau=20$, 100, and ∞ , in MD steps (a). Comparison between the excess energy from the MD experiment and that obtained from the MC run (b). The MC data have been shifted upwards for the sake of clarity. t_{MC} stands for ‘‘Monte Carlo time,’’ which is proportional to the number of MC moves.

tion is applied moderately often ($\tau \geq 0.1$ time reduced units = 20 MD time steps, top curve in the figure), the structure succeeds in growing initially but only slowly and by single-particle diffusion, which is reflected in the low exponents attained. For larger τ the saturation in the growth occurs at increasingly long times and the system can set a growth law close to $t^{1/3}$. This law persists even for $\tau=100$ MD steps, which is a time much longer than the typical time needed for hydrodynamics to get established locally. For even larger τ the growth becomes faster and the full hydrodynamic exponent is recovered [19]. The procedure just outlined allows us to tune the hydrodynamics by changing τ ; this parameter plays a role similar to the hydrodynamic coupling constant λ introduced by Wu *et al.* [20] in their Langevin study of phase separation. These authors observe a smooth transition from a low ($n \sim 1/3$) to a high ($n \sim 0.7$) exponent as λ is increased. We believe our results are qualitatively similar to theirs, but have a microscopic basis.

Another simulation technique to explore thermodynamic and structural properties of equilibrium many-particle systems is Monte Carlo (MC). Despite minor technical differences of implementation, both techniques, MD and MC, provide essentially identical equilibrium results since they both sample the system’s configurational space correctly. There is, however, no connection between the MC trajectories and the real trajectories followed by a system. Thus when it comes to exploring nonequilibrium phenomena, where collective interactions play a role, the use of MC will necessarily lead to different results since this procedure consists of single-particle moves and hydrodynamic modes are suppressed. This can be shown easily by conducting MC experiments of the same system simulated above. In Fig. 3(b) we reproduce the results of a run generated by continuum stochastic MC dynamics. As expected [21], we find $R(t) \sim t^{1/3}$ (note that here the variable t stands not for real time but for

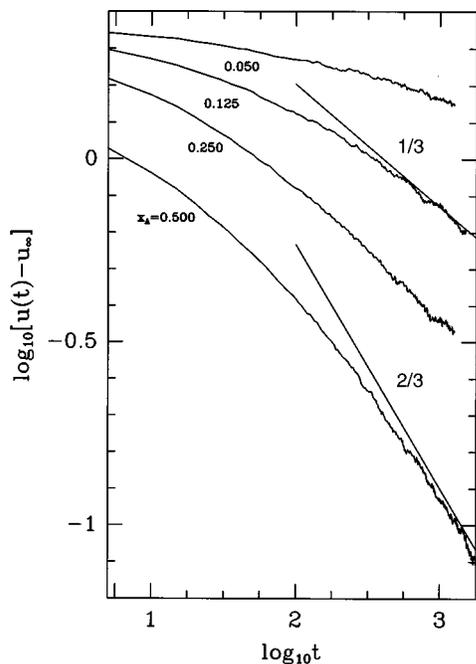


FIG. 4. Time dependence of the excess energy for different solute concentrations x_A .

“MC time,” which is proportional to the number of MC particle moves).

Finally, we have performed MD experiments at several off-critical concentrations, using the LJ-WCA interaction model and a system of $N=40\,000$ molecules. The setup of the experiments was identical with that of the critical quenches except that at the quench type A was assigned to 10 000, 5 000, and 2 000 molecules, giving $x_A=0.250$, 0.125, and 0.050, respectively. The corresponding state points are marked by crosses in Fig. 1. Note that the first state point is well inside the “spinodal region,” the second one is close to the mean-field spinodal line, and the last state point is in the region between the coexistence line and the spinodal (the “nucleation region”). Figure 4 shows the results of the quenches. The run at $x_A=0.500$ has also been included for comparison. The feature which first comes to one’s attention is the high exponent of the $x_A=0.250$ quench. Even though it is likely that the final state has not been attained, the measured exponent, $n\sim 0.45$, is still quite close to $1/2$ and much larger than $1/3$. This may indicate the activation of more than one growth mechanism and a smooth transition, within the length of our simulations, from one type of growth (motion of interfaces) to the other (cluster coalescence and evaporation-condensation of single particles). The run of the $x_A=0.125$ quench shows a $1/3$ exponent, consistent with theoretical expectations for off-critical quenches. Finally, the quench at the nucleation region exhibits a very slow growth and the measured exponent is probably not statistically significant in view of the small fraction of A (solute) molecules involved and the short simulation times. Figure 5 shows snapshots of molecular configurations taken after 250 000 MD time steps, where the different morphology of the structure with the solute fraction is apparent.

In summary, we have investigated the dynamics of phase separation in a two-dimensional binary fluid at both critical

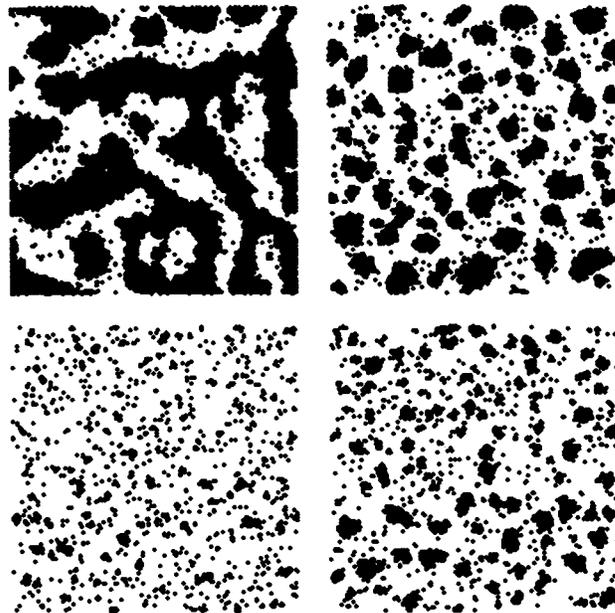


FIG. 5. Snapshots of molecular configurations taken after 250 000 MD time steps. Starting from the lower-left picture and proceeding counterclockwise, $x_A=0.050, 0.125, 0.250$, and 0.500.

and off-critical concentrations using the molecular dynamics technique. We observe a smooth variation in the late-time growth exponent with respect to concentration x_A from a value of $2/3$ for a critical quench to a value close to $1/3$ for noncritical quenches. For critical quenches the growth starts as $t^{1/2}$, and later crosses over to the faster growth $t^{2/3}$. These results are in agreement with theoretical predictions. MD simulations represent the dynamics of real fluids in an accurate way and explain the formation and evolution of macroscopic single-phase domains in phase segregation kinetic processes exclusively from microscopic molecular interactions. However, they are computationally expensive investigations, and this fact limits the size of the systems and the times which may be necessary to access the late-stage regimes of spinodal decomposition. In spite of our long runs, it could be that even longer times are required to attain the asymptotic region. Nevertheless, we have seen a definite departure of the growth law from the purely diffusive prediction $t^{1/3}$ to $t^{1/2}$ at critical concentration, found evidence of scaling in the correlation functions [4], and obtained data strongly in favor of a faster growth at longer times. Finally, by designing perturbed MD dynamics experiments we have been able to switch off collective modes in a controlled manner and recover the behavior expected in a system not governed by hydrodynamic interactions.

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- [21] Our continuum stochastic Monte Carlo simulation is equivalent to a Monte Carlo simulation of the kinetic Ising model with conserved order parameter. There has been some controversy in the past over the numerical value of the late-time exponent of these models. In the case of the latter model, the expected 1/3 was not confirmed and instead lower values were found. By using the effective exponent $n_{eff}(t)$ introduced by Huse [*Phys. Rev. B* **34**, 7845 (1986)] and long simulation runs, J. G. Amar *et al.* [*Phys. Rev. B* **37**, 196 (1988)] managed to extrapolate to infinite times to obtain the 1/3 exponent, attributing the previously reported 1/4 to long-persistent transients. Much the same story can be told for the continuum model *B*. Nowadays there is wide consensus that both these models are in the same universality class and that their late-time exponent is 1/3. Note that the 1/3 exponent we obtain disagrees with that reported in [4]; as advanced in the latter reference, we believe that the difference can be attributed to the model potential used in [4] for the *AB* interaction, which might penalize evaporation-condensation events, thus suppressing the 1/3 exponent and favoring diffusion along interfaces, which leads to 1/4. The model for the *AB* interaction used here might more easily allow for such events.