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Growth Kinetics in Multicomponent Fluids

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The hydrodynamic effects on the late-stage kinetics in spinodal decomposition of multicomponent fluids are examined using a lattice Boltzmann scheme with stochastic fluctuations in the fluid and at the interface. In two dimensions, the three- and four-component immiscible fluid mixture (with a 1024^2 lattice) behaves like an off-critical binary fluid with an estimated domain growth of $t^{0.4 \pm 0.03}$ rather than $t^{1/3}$ as previously estimated, showing the significant influence of hydrodynamics. In three dimensions (with a 256^3 lattice), we estimate the growth as $t^{0.96 \pm 0.05}$ for both critical and off-critical quenches, in agreement with phenomenological theory.

KEY WORDS: Lattice Boltzmann method; growth kinetics; multicomponent fluids.

1. INTRODUCTION

It is well known that a binary fluid mixture undergoes phase separation if rapidly quenched from a high-temperature phase to a point in the coexistence region. Moreover, when the domain sizes are much larger than the interfacial thickness, there is only one dominant length scale in the system (for review see ref. 1). It is accepted that the late-time dynamics in a binary alloy or glass in which the order parameter is conserved, follows a growth law of $R(t) \sim t^{1/3}$, where $R(t)$ is the average size of the domains. This growth law is characteristic of the long-range diffusion of particles between domains and was first predicted for off-critical quenches by

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Lifshitz and Slyozov.(2) Methods to carry out simulations of phase segregating systems with hydrodynamic interactions include molecular dynamics (MD), direct numerical simulation of time-dependent Ginzburg–Landau equations, cell dynamical systems,(3) and more recently, lattice gas(4) and lattice Boltzmann (LB) models.(5, 6) Lattice Boltzmann simulations have recently been successfully used to study spinodal decomposition for critical quenches for a binary fluid in the presence(5) and absence of porous media.(6) The various simulation techniques have tried to address the question of the growth of single-phase domains and the scaling properties of the correlation or structure functions. The underlying aim has been to find certain "universality classes" for first-order phase transitions in which the growth law and scaling properties are independent of the details of the interactions, spatial dimension, and the number of components.

The aim of this work is to determine the growth law in two and three dimensions for three- and four-component fluids with equal volume fractions and compare the results to that for an off-critical binary quench with unequal volume fractions. We obtain results in which the hydrodynamic interactions are present as well as absent and show that hydrodynamics is important. In previous work on the LB method for studying phase segregation(5, 6) fluctuations due to correlations in the particles have been neglected. We include the effects of fluctuations on domain growth in multicomponent systems by incorporating stochastic fluctuations in the fluid stress tensor using a scheme recently proposed by Ladd(7) and fluctuations in the color gradient to perturb the interface.

The influence of the number of components on domain growth and scaling has been recently examined in two dimensions for a Potts model with two and three components using Monte Carlo(8) as well as for a three-component fluid using molecular dynamics (MD)(9) simulations. The Potts model does not include hydrodynamics and therefore the result of $t^{1/3}$ for the growth law for two and three components is not surprising. The conclusion of the MD work for a three-component fluid shows that the growth exponent is also 1/3. Since a thorough MD study for fluids requires a large number of particles and has to be run for very long times, we have undertaken a lattice Boltzmann approach while also incorporating fluctuations. Our results demonstrate that for off-critical binary quenches and for three- and four-component fluids the growth exponent for late times scales as $t^{0.4 \pm 0.03}$ in two dimensions. This is to be compared with $t^{1/3}$ if hydrodynamics was not relevant to phase segregation and domain growth in two dimensions. Our three-dimensional result is the first simulation for an off-critical quench to verify the result of Siggia(10) that is based on a phenomenological model of droplet coalescence. We also determine that dynamical scaling, which is a consequence of the existence of one dominant length scale, is valid in two and three dimensions. The multicomponent fluids show the same scaling behavior as an off-critical binary quench.

2. LATTICE BOLTZMANN MODEL FOR SPINODAL DECOMPOSITION

Lattice gas and lattice Boltzmann (LB) methods can be used to study hydrodynamic phase segregation using parallel computing techniques.(11) They simulate fluid properties, phase segregation, and the interface dynamics simultaneously and allow complex boundaries to be handled easily. The methods have been described as providing the most promising tools to study flow through porous media.(12) Unlike methods such as the use of the Langevin equation(13) that are based on a phenomenological model of fluid behavior and are computationally intensive, LB methods simulate hydrodynamic phase segregation in a natural way without the introduction of *ad hoc* relations between the order parameter fluctuations and the fluid dynamics. The lattice Boltzmann method is a discrete, in space and time, microscopic kinetic equation description for the evolution of the particle distribution function of a fluid. Point particles move along the links of a lattice (hexagonal in two dimensions), obey certain collision rules, and macroscopically mimic the Navier–Stokes equations in certain limits. The LB two-phase model we used is a modified version of the immiscible fluid model proposed by Grunau *et al.*(14) that is based on the original model introduced by Gunstensen *et al.*(15) The multicomponent LB model in this paper is an extension of the lattice gas(16) and lattice Boltzmann(17) models by Gunstensen and Rothman.

The local order parameter is defined as

$$\psi(\mathbf{x}, t) = \rho_1(\mathbf{x}, t) - \sum_{k=2}^{n} \rho_k(\mathbf{x}, t)$$

where $\rho_k = \sum_{i=0}^{N} f_i^k(\mathbf{x}, t)$ $(k = 1,..., n)$ is the particle density and $f_i^k(\mathbf{x}, t)$ is the distribution function for the kth component of the fluid mixture at site $\mathbf{x}$ and time t moving along the link in the direction i. Here n is the number of components and N is the number of velocity directions. Also,

$$f_i(\mathbf{x}, t) = \sum_{k=1}^{n} f_i^k(\mathbf{x}, t)$$

is the distribution function for the total fluid, where $i = 0, 1,..., 6$ represent the velocity directions at each site of a hexagonal lattice. The state $i = 0$

corresponds to a portion of the fluid at rest. The LB equation for $f_i(\mathbf{x}, t)$ can be written as

$$f_i^k(\mathbf{x}+\mathbf{e}_i, t+1) = f_i^k(\mathbf{x}, t) + \Omega_i^k \tag{1}$$

where k denotes the fluid component, and

$$(\Omega_i^k) = (\Omega_i^k)^{\mathrm{c}} + (\Omega_i^k)^{\mathrm{p}}$$

is the collision operator consisting of a term representing the rate of change of f_i^k due to collisions and a term representing the color perturbation. The vectors $\mathbf{e}_i$ are the velocity vectors along the links of a hexagonal lattice. The form of $(\Omega_i^k)^{\mathrm{c}}$ is chosen to have a single time relaxation with

$$(\Omega_i^k)^{\mathrm{c}} = -(f_i^k - f_i^{k(\mathrm{eq})})/\tau$$

where τ is the characteristic relaxation time and $f_i^{k(\mathrm{eq})}$ is the local equilibrium distribution.(18) The surface tension inducing perturbation $(\Omega_i^k)^{\mathrm{p}}$ and the recoloring procedure are chosen appropriately so that Laplaces's law holds for the model.(14)

The local color gradient for each component $\mathbf{G}^k(\mathbf{x})$ is defined by

$$\mathbf{G}^k(\mathbf{x}) = \sum_{i=1}^{N} \mathbf{e}_i f_i^k(\mathbf{x}+\mathbf{e}_i)$$

and the surface color perturbation that is added to segregate and stabilize the interface is

$$(\Omega_i^k)^{\mathrm{p}} = A\ |\mathbf{G}^k| \cos 2(\theta_i - \theta_{G^k})$$

where θ_i is the angle of lattice direction i and $\theta_{G^k} = \arctan(G_y^k/G_x^k)$ is the angle of the local color gradient for the kth component. It can be analytically calculated(15, 14) that the surface tension is proportional to $\sim A\tau\rho$. Here $\rho = \sum_{i,k} f_i^k$ is the total particle density. In the original models(15, 14) the recoloring step always makes the local color gradient to lie along the direction perpendicular to the interface.

To mimic the temperature effect on the interface, a noise is introduced to perturb the local color gradient direction θ_{G^k} by assuming that the angles are distributed according to a Gaussian distribution about θ_{G^k}. The variance of the angle distribution depends on some local temperature.

A cause for concern about the use of the LB method for studying spinodal decomposition has been the lack of statistical correlations in the particles. Previous simulations show that droplets form and collapse due to an initial random concentration field. It has been questioned if this initial fluctuation-driven system is equivalent in scaling dynamics to a system with intrinsic noise (such as lattice gas and molecular dynamics simulation) or a system with a noise drive due to the temperature of the system. In order to study the effects of fluctuations on domain growth, we have incorporated stochastic fluctuations in the fluid stress tensor according to a scheme proposed by Ladd.(7) The basic idea is that, on length scales and time scales intermediate between molecular and hydrodynamic, thermally induced fluctuations can be reduced to random fluctuations in the fluxes of conserved variables, for instance, the stress tensor. It is thus plausible that in an LB simulation, molecular fluctuations can be modeled realistically on intermediate scales, although the microscopic interactions are different from the real dynamics obtained from, for example, an MD simulation. To incorporate this effect, a stochastic term, $f_i'(r, t)$ representing the thermally induced fluctuations in the stress tensor is added to the time evolution of the density distribution. That is,

$$f_i^k(\mathbf{x}+\mathbf{e}_i, t+1) = f_i(\mathbf{x}, t) + (\Omega_i^k)^{\mathrm{c}} + (\Omega_i^k)^{\mathrm{p}} + f_i'(\mathbf{x}, t) \tag{2}$$

where f_i' is chosen so that its stress moment is nonzero, while conserving mass and momentum. The random stress fluctuations are uncorrelated in space and time and are sampled from a Gaussian distribution. The intensity of the random stress represents the magnitude of the local temperature (see ref. 7 for details).

3. SIMULATION RESULTS AND DISCUSSION

We performed critical quenches with $\langle \psi(x) \rangle = 0$, while $\langle \psi(x) \rangle \neq 0$ for off-critical quenches. The largest systems simulated were 1024^2 in two dimensions and 256^3 in three dimensions. Although we have investigated the domain growth and scaling properties for a variety of lattice sizes and parameters, we report on the domain growth for only one set of parameters. The results obtained with smaller lattices and different parameters are consistent with the data presented here.

The lattice was initialized with a random distribution of the different-colored fluids. The growth kinetics is characterized through the order parameter correlation function $G(r, t) = \langle \psi(r)\, \psi(0) \rangle - \langle \psi \rangle^2$ averaged over shells of radius r. The domain size $R(t)$ is then defined as the first zero of $G(r, t)$ and the Fourier transform of $G(r, t)$, the structure factor $S(k, t)$, is expected to have the scaling form $R(t)^d\, F(kR(t))$. The function $F(x)$ $[x = kR(t)]$ is the scaling function, which is expected to behave as x^{δ}, where for large x, $\delta = -(d+1)$, which is Porod's law.

We first discuss the two-dimensional results. The effects of fluctuations

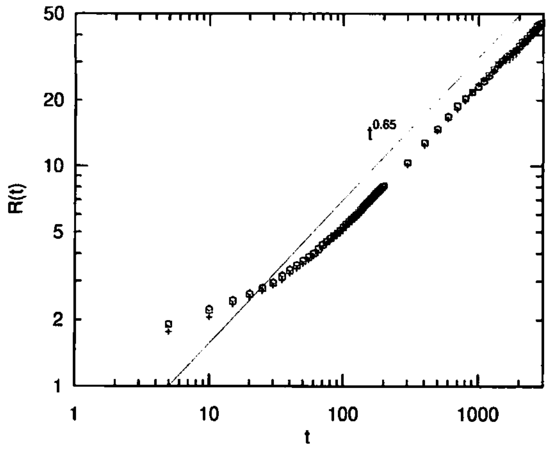

Fig. 1. The effects of fluctuations on domain growth for a binary fluid on a 512^2 lattice after a critical quench. The three cases represent growth due to (i) thermally induced fluctuations via the stress tensor (+), (ii) fluctuations due to initial velocity perturbation only (□), and (iii) both thermal and initial velocity fluctuations (◇).

on domain growth is shown in Fig. 1 for a binary fluid after a critical quench. The three cases shown are the growth due to (i) thermally induced fluctuations as described above (+), (ii) fluctuations due to initial velocity only with average velocity $\langle u \rangle = 0$ and $\langle u^2 \rangle \neq 0$ (□), and (iii) both thermal and initial velocity fluctuations included (◇). If neither (i) nor (ii) is present, no patterns are obtained. Since the energy will decay due to dissipation in the system, for very long times the systems with initial fluctuations or thermal fluctuations should behave differently. However, in the current study, up to the times simulated, the results are consistent with each other and show that the origin of the fluctuations has little bearing on domain growth. Thus, the previous use(5, 6) of an initial fluctuation in velocity only as the driving mechanism for droplet formation would seem justified. In all subsequent simulations in this work we have used case (iii).

Figure 2 shows the domain growth for a binary fluid after an off-critical quench for $\langle \psi(x) \rangle = 1/3$(◇) and $\langle \psi(x) \rangle = 1/2$(+). The early-time domain growth of $t^{1/3}$ and the later-stage growth of $t^{0.4 \pm 0.02}$ are clearly evident regardless of the average order parameter. We can interpret this as long-range diffusion of fluid particles across growing domains at early time giving way to domain growth where hydrodynamic or inertial effects become important. Our value appears to be consistent with the prediction $t^{2/5}$ made by Furukawa(19); however, it is not in accord with the $t^{1/3}$ prediction by Miguel *et al.*(19) for two-dimensional off-critical mixtures. In order

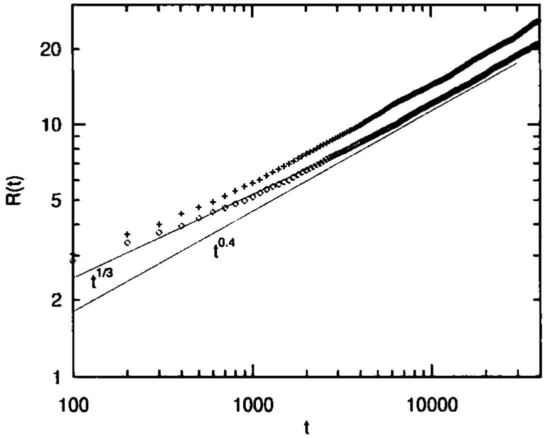

Fig. 2. Domain growth for a binary fluid after an off-critical quench for $\langle\psi(x)\rangle = 1/3$(◇) and $\langle\psi(x)\rangle = 1/2$(+). The straight lines represent fits to $\langle\psi(x)\rangle = 1/3$ of $\sim t^{1/3}$, showing the early-time diffusive growth, and $\sim t^{0.4}$ for the later-stage growth where inertial effects are important.

to understand the effects of hydrodynamics in the system, the velocity **u** of the particles is set to zero before the collision step. For such a system, the convective effect and dissipative mechanisms will disappear and the only dynamics left is the diffusive process. Figure 3 shows the domain growth for this diffusive motion. The minority phase forms small droplets at first,

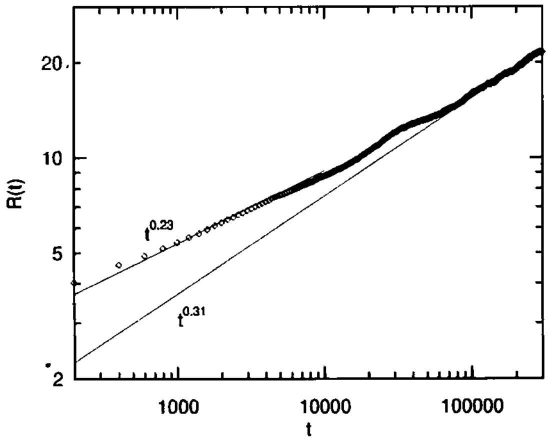

Fig. 3. Growth for a binary fluid after an off-critical quench for $\langle\psi(x)\rangle = 1/3$ without hydrodynamics. The transient behavior due to interfacial diffusion ($\sim t^{0.23}$) eventually leads to the expected long-range behavior.

which then grow very slowly into larger domains. The growth is very slow and is hampered by transient effects which slow down the ordering process. A transient regime where the growth scales as $t^{0.23}$, close to $t^{0.25}$, can be identified and is likely caused by short-range diffusion along the boundary of the domains, as explained by Mullin.(20) This short-range diffusion crosses over at very late times to a $t^{1/3}$ behavior characteristic of long-range diffusion. Though this has been previously predicted theoretically,(2) and has been detected using extrapolation methods, we are not aware of any off-critical simulations in which it has been directly observed.

We expect topological factors to play a significant role in the ordering process for three- and four-component fluid mixtures. We observe that the domains are rather compact and can eventually be hindered from growing, similar to the effects of confinement of a fluid mixture in a pore geometry.(5) Figure 4 shows the domain growth for three- and four-component mixtures in the presence of hydrodynamics. For the symmetric three- (◇) and four- (×) component mixtures an early $t^{1/3}$ regime can be identified which crosses over to a clear $t^{0.40 \pm 0.02}$ growth for late times where the domain morphology is compact. The growth for a three-component system with concentrations (0.2, 0.2, 0.6) (not shown here) has similar behavior. Comparison of Figs. 2 and 4 shows that, as expected, a ternary and four-component symmetric fluid mixture behaves like an off-critical binary mixture with an estimated growth law of $t^{0.4 \pm 0.03}$ for late times. The early-time behavior for (◇) and (×) shows the $t^{1/3}$ growth characteristic of long-range diffusion. Our results clearly indicate that hydrodynamics plays a significant role in the late-time behavior, contrary to an earlier MD work(9) which we suspect was not run long enough. A Langevin model simulation of Farrell and Valls(21) on a 100 by 100 grid for an off-critical binary quench predicts $t^{0.35 \pm 0.03}$. This result has been interpreted as showing $t^{1/3}$ behavior. However, the work by Wu *et al.*(22) with this method indicates an exponent very close to results reported here. It appears that large lattice sizes and long-time simulations are necessary to obtain correct growth kinetics by this method. The reason for the similar behavior of three- and four-component fluids to that of the off-critical binary fluid may be understood in terms of the number of contact points of different phases. The physics appears to be dominated by the two-phase line contacts rather than the finite number of three- or four-phase point contacts.

The behavior of the scaling function $F(x)$ during the late stages of growth is shown in Figs. 5–7 for an off-critical binary mixture, a three-component system, and a four-component system, respectively. The results show that dynamical scaling holds for all three cases and the scaling for large x $(x > 2)$ is consistent with an exponent around -3, showing that Porod's law is approximately satisfied. For $x < 1$, our results indicate an

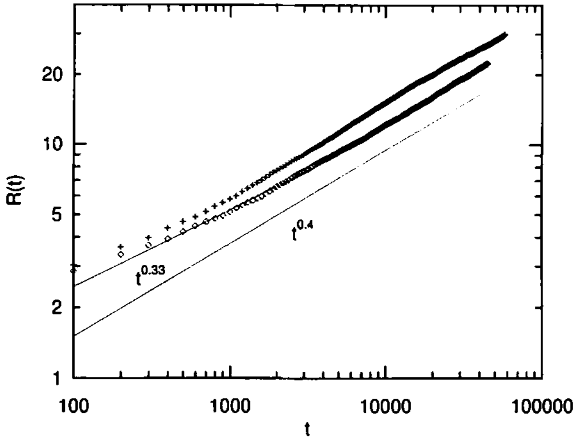

Fig. 4. Growth for a symmetric three-component system (◇) (1/3, 1/3, 1/3) and a symmetric four-component mixture (×) (1/4, 1/4, 1/4, 1/4).

exponent just over 2 for all three cases. This is in contrast with the value of 4 predicted by Furukawa(23) for a fluid mixture. However, it is consistent with estimates from other two dimensional simulation studies.(24, 25, 22) In particular, the recent results of Wu *et al.*(22) from a Langevin dynamics study suggest that the small x scaling δ depends on the "strength" of the hydrodynamic interaction.

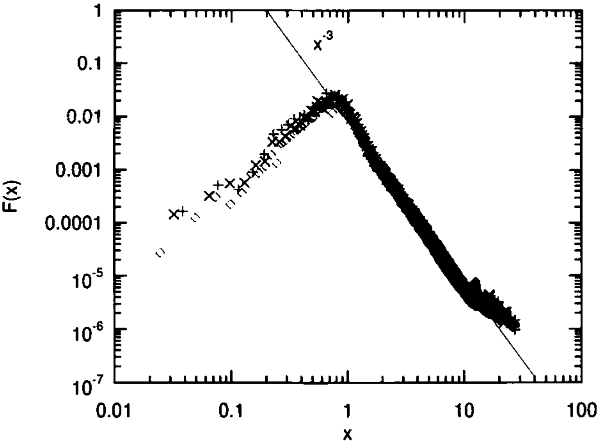

Fig. 5. The scaling function $F(x)$ as a function of x $[x = kR(t)]$ for an off-critical binary fluid with $<\psi(x)> = 0.5$ in two dimensions (1024^2). The data are for times t = 10,000 (◇), 15,000 (×), 20,000 (+) (lattice time units).

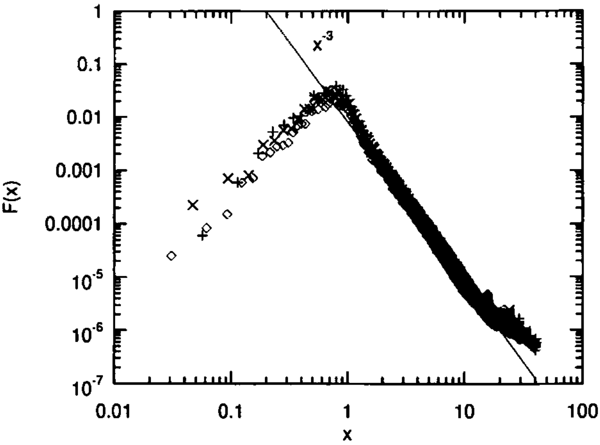

Fig. 6. The scaling function $F(x)$ as a function of x $[x = kR(t)]$ for a three-component mixture (1/3, 1/3, 1/3) in two dimensions (1024^2). The data are for times $t = 20{,}000$ (◇), 30,000 (×), 40,000 (+) (lattice time units).

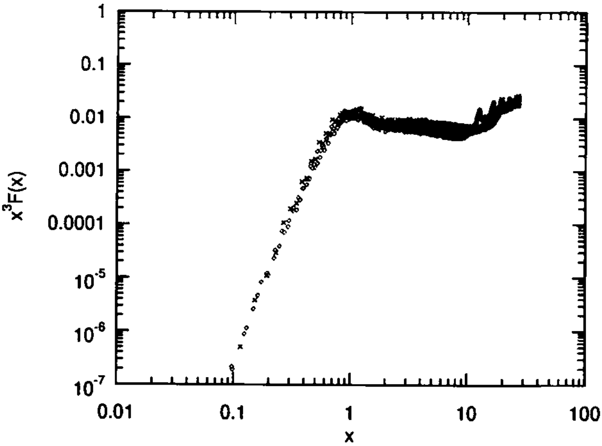

Fig. 7. Plot of $x^3F(x)$ as a function of x $[x = kR(t)]$ for a four component mixture (1/4, 1/4, 1/4, 1/4) in two dimensions (1024^2). The data are for times $t = 20{,}000$ (◇), 30,000 (×), 40,000 (+) (lattice time units). The scaling in $2 < x < 10$ approximately satisfies Porod's law.

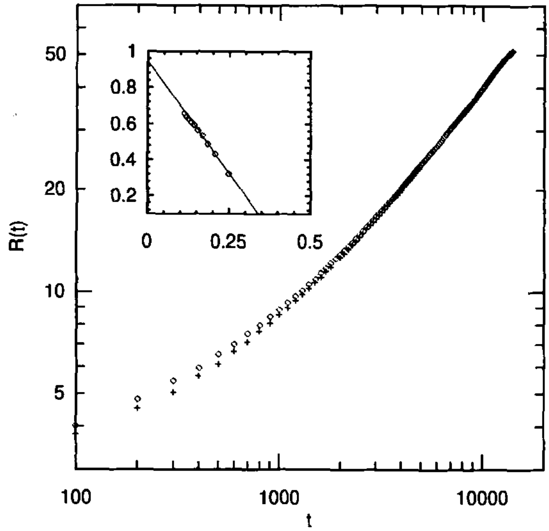

Fig. 8. Three-dimensional domain growth $R(t)$ on a 256^3 lattice for a critically quenched (◇) and off-critically quenched $[\langle\psi(x)\rangle = 0.5(+)]$ binary fluid, showing similar growth dynamics. The extrapolated growth exponent n_{eff} versus $1/R(t)$, as seen in the inset, gives $\sim t^{0.96}$, close to the expected growth law t^1.

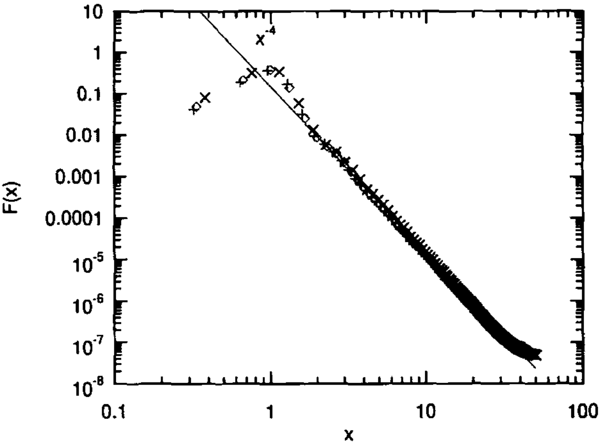

Fig. 9. The scaling function $F(x)$ as a function of x $[x = kR(t)]$ at times $t = 10,000$ (◇), 15,000 (×), 20,000 (+) (lattice time units) for a critical binary mixture in three dimensions on a 256^3 lattice.

The three-dimensional growth is shown in Fig. 8 for 256^3 for a critical (◇) and off-critical (+) quenched binary fluid with $\langle \psi(x) \rangle = 0.5$. Both cases show similar behavior. Finite-size effects have not allowed us to run the simulation long enough to estimate the exponents for later stage growth. However, if we calculate an effective exponent(26) n_{eff}, by considering the domain sizes at time Mt and t, where M is an integer, then the extrapolated behavior for large domain is $R(t) \sim t^{0.96 \pm 0.05}$ (see the insert of Fig. 8). The results are in agreement with Siggia's prediction.(10) We note, however, that the extrapolation procedure used may not be firmly established and could be a source of controversy.

The behavior of the scaling function $F(x)$ for accessible late times for a three-dimensional critical mixture is shown in Fig. 9. The results appear consistent with dynamical scaling and the behavior for $x > 2$ is in agreement with Porod's law ($\delta = -4$).

Experimental studies have been inconclusive in the estimates of growth exponents. The measurement of an off-critical quench of a simple acid–water mixture gave a growth exponent between 0.32 and 0.35,(27) whereas a growth rate of ~ 0.5 was observed in a very careful study of an off-critical quenched block copolymer.(28) Transient effects and the lack of error analysis in the previous studies make direct comparison with experiments difficult. A recent experiment in microgravity indicates a growth of 1/3 over several decades.(29) Thus we can only stress the need for more experiments on off-critical binary quenches and multicomponent fluids.

In conclusion, we have used a lattice Boltzmann model with fluctuations in the fluid and interface to study the kinetics of domain growth and scaling behavior in two and three dimensions for an off-critical quenched binary fluid and three- and four-component fluid mixtures. We find that the domain growth scales as $t^{0.4 \pm 0.03}$ in two dimensions, indicating that hydrodynamics is relevant to the kinetics of phase separation as seen in critical quench. Our three-dimensional results are in agreement with phenomenological theory.(10)

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