Comment on "Phase Separation in Two-Dimensional Fluid Mixtures"

In a recent Letter [1], the authors report results of molecular dynamic simulations of phase segregation in two-dimensional one-component and binary fluids quenched into the coexistence region. They find that the growth exponent n for the domain sizes, $R(t) \propto t^n$, is $n \leq \frac{1}{2}$. This is consistent with the results of [2] on similar systems but differs from many simulations of binary mixtures involving lattice Boltzmann [3,4], fluctuating hydrodynamic [5,6], Ginzburg-Landau [7], and most recently microscopic lattice models [8]. All these simulations find $R(t) \propto t^n$ with $n \simeq \frac{2}{3}$, in accord with the predictions of Furukawa for such systems [7,9]. No discussion of this discrepancy is given in [1].

Here we want to point out that, as discussed in [8] in connection with the results of [2] for binary mixtures, the source of this difference is most likely the relative size of the domains $R(t)$ probed in the different simulations compared to the relevant characteristic hydrodynamic length scale of the system $R_h = \nu^2\rho/\sigma$ [7]; ν is the kinematic viscosity, ρ is the density, and σ is the surface tension coefficient. As indicated in [8], we expect in two dimensions a crossover from $n = \frac{1}{2}$, for $R(t) < R_h$, suggested by [10] for growth dominated by droplet coalescence and $n = \frac{2}{3}$ for $R(t) > R_h$ when the dominant mechanism is surface tension driven by hydrodynamic flow balanced by inertial effects. As we know of no estimates of the viscosity and surface tension coefficient for the binary fluid model used in [1,2], we base our remarks on the one-component Lennard-Jones fluid and very rough estimates for the mixture using a related model [11]. For the Lennard-Jones liquid, the simulations described in [1] at $T = 0.76T_c$ and $\rho = 0.325$ correspond to $\eta \simeq 3$ and $\sigma \simeq 0.02$ [12], giving $R_h \simeq 1400$ which is much larger than the largest $R(t)$ observed there. For the binary fluid our rough estimate gives $R_h \simeq 50$ at $\rho = 0.74$ and $k_BT/\epsilon = 2$ (the lowest temperature probed), again bigger than the $R(t)$ probed in [1,2]. (In [2] the density is smaller than in [1] so R_h is even bigger. R_h should also increase with the temperature.) The simulations [3,7,8], on the other hand, are clearly in the $R(t) \geq R_h$ regime [8]. We plan to carry out simulations in which one can see the actual crossover.

We also note here that in three dimensions, near critical concentrations, we expect two crossovers starting with $n = \frac{1}{3}$, when $R(t)$ is very small compared to R_h and Brownian coalescence dominates [13], to $n = 1$ at later stages, predicted by Siggia [13], when hydrodynamic flows are controlled by dissipation to an $n = \frac{2}{3}$ regime when $R(t)$ is large compared to R_h and inertial effects are important [7]. The intermediate regime is absent in two dimensions due to the instability of the corresponding flow [10]. The first two regimes have so far been probed in 3D by experiments [14] and simulations [3,4,15], and there is a good indication that the third regime was also observed in computer simulations using a microscopic lattice model [16]. One would like to observe also the crossover between $n = 1$ and $\frac{2}{3}$, and that should be possible through lattice Boltzmann simulations [8].

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