Two-scale competition in phase separation with shear

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The behavior of a phase separating binary mixture in uniform shear flow is investigated by numerical simulations and in a renormalization group (RG) approach. Results show the simultaneous existence of domains of two characteristic scales. Stretching and cooperative ruptures of the network produce a rich interplay where the recurrent prevalence of thick and thin domains determines log-time periodic oscillations. A power law growth of the average domain size, with \( \alpha = 4/3 \) and \( \alpha = 1/3 \) in the flow and shear direction respectively, is shown to be obeyed.

The kinetic behaviour of the binary mixture is described by the Langevin equation

\[
\frac{\partial \varphi}{\partial t} + \nabla (\varphi \vec{v}) = \Gamma \nabla^2 \frac{\delta \mathcal{F}}{\delta \varphi} + \eta
\]

where the scalar field \( \varphi \) represents the concentration difference between the two components of the mixture \([1]\). The equilibrium free-energy can be chosen as usual to be

\[
\mathcal{F}\{\varphi\} = \int d^d x \left\{ \frac{a}{2} \varphi^2 + \frac{b}{4} \varphi^4 + \frac{\kappa}{2} \right\} \nabla \varphi \right| \right|^2
\]

where \( b, \kappa > 0 \) and \( a < 0 \) in the ordered phase. \( \vec{v} \) is an external velocity field describing plane shear flow with average profile given by

\[
\vec{v} = \gamma y \vec{e}_x
\]

where \( \gamma \) is the shear rate and \( \vec{e}_x \) is the unit vector in the flow direction. \( \eta \) is a gaussian white noise, representing thermal fluctuations, with mean zero and correlation \( \langle \eta(x, t) \eta(x', t') \rangle = -2 \gamma T \nabla^2 \delta(x - x') \delta(t - t') \), where \( \Gamma \) is a mobility coefficient, \( T \) is the temperature of the fluid, and the symbol \( \langle \ldots \rangle \) denotes the ensemble average.

The Langevin equation \([1]\) has been simulated in \( d = 2 \) by first-order Euler discretization scheme. Periodic boundary conditions have been used in the flow direction while in the \( y \) direction the point at \( (x, y) \) is identified with the point at \( (x + \gamma L, y + L) \), where \( L \) is the size of the lattice \([1]\). Lattices with \( L = 1024, 2048, 4096 \) and space discretization intervals \( \Delta x = 0.5, 1 \) were used. In the phase separation process the initial configuration of \( \varphi \) is a high temperature disordered state and the evolution of the system is studied in model \([1]\) with \( a < 0 \). Parameterization invariance of \([1]\) allows one to set \( \Gamma = |\alpha| = b = \kappa = 1 \). The structure factor is defined as \( C(k,t) = \langle \varphi(k,t) \varphi(-k,t) \rangle \) where \( \varphi(k,t) \) are the Fourier components of \( \varphi \). Results will be shown for the case \( \gamma = 0.0488 \), \( T = 0 \), \( L = 4096 \), \( \Delta x = 1 \), \( \langle \varphi \rangle = 0 \). Similar results have been obtained for other values of the parameters.

A sequence of configurations at different values of the strain \( \gamma t \) is shown in Fig. 1. After an early time, when well defined domains are forming, the usual bicontinuous structure of phase separating domains starts to be distorted for \( \gamma t \gtrsim 1 \).
The growth is faster in the flow direction and domains assume the typical striplike shape aligned at an angle \( \theta(t) \) with the direction of the shear which decreases with time. As the elongation of the domains increases, nonuniformities appear in the system: Regions with domains of different thickness can be clearly observed at \( \gamma t = 11 \). The evolution at still larger values of the strain \( \gamma t = 20 \). The domains with the smallest thickness eventually break up and burst with the formation of small bubbles.

A systematic existence of two scales in the size distribution of domains is suggested by the behavior of the structure factor, shown in Fig. 2. At the beginning (see the picture at \( \gamma t = 0.2 \)) \( C(\vec{k}, t) \) exhibits an almost circular shape, corresponding to the early-time regime without sharp interfaces. Then shear-induced anisotropy becomes evident, \( C(\vec{k}, t) \) is deformed into an ellipse, changing also its profile and, for \( \gamma t \geq 1 \), four peaks can be clearly observed. The position of each peak identifies a couple of typical lengths, one in the flow and the other in the shear direction. The peaks are related by the \( \vec{k} \rightarrow -\vec{k} \) symmetry so that, for each direction, there are two physical lengths. This corresponds to the observation of domains with two characteristic thicknesses, made in Fig. 1.

The dips in the profile of \( C(\vec{k}, t) \) develop with time until \( C(\vec{k}, t) \) results to be separated in two distinct foils at \( \gamma t \approx 4 \). The evolution of the system until this stage is well described by the solution of the linear part of (4) (letting \( b = 0 \)):

\[
C(\vec{k}, t) = C_0 \exp \left(-\int_0^t k^2(s)(k^2(s) - a) \, ds \right)
\]

where \( \vec{k}(s) = (k_x, k_y + \gamma s k_x) \) and \( C_0 \) is the structure factor at the initial time [9]. Then non-linear effects become essential in producing the patterns shown in Fig. 2 at \( \gamma t = 11, 20 \). At \( \gamma t = 11 \) we evaluate the positions of the peaks at \( (k_x, k_y) = (0.015, 0.107) \) and \( (k_x, k_y) = (0.038, 0.23) \). This gives a value around two for the ratio between the characteristic sizes of domains; the same value is found for the ratio between the positions of the two peaks in the histogram of the domain size distribution. The relative height of the peaks in one of the foils of \( C(\vec{k}, t) \) can be more clearly seen in Fig. 3 where the two maxima are observed to dominate alternatively at the times \( \gamma t = 11 \) and \( \gamma t = 20 \).

The competition between two kinds of domains is a cooperative phenomenon. In a situation like that at \( \gamma t = 11 \), the peak with the larger \( k_y \) dominates, describing a prevalence of stretched thin domains. When the strain becomes larger, a cascade of ruptures occurs in those regions of the network where the stress is higher and elastic energy is released. At this point the thick domains, which have not yet been broken, prevail and the other peak of \( C(\vec{k}, t) \) dominates, as at \( \gamma t = 20 \). In the large-\( N \) limit the prevalence of one or the other peak
has been shown to continue periodically in time \[\eta\]. Here we have an indication of a similar behavior, although the observation of the recurrent dominance of the peaks on longer timescales is hardly accessible numerically.

The hallmarks of this dynamics are found in the behavior of the average size of domains, \(R_x(t)\) and \(R_y(t)\), in the flow and shear direction. In our simulations these quantities have been calculated through the following expressions:

\[
\langle C(\vec{k}, t) \rangle = \frac{1}{\sqrt{V}} \int d\vec{r} \phi(\vec{r}, t) e^{i\vec{k}\cdot\vec{r}}
\]

\[
\delta \phi(\vec{k}, t) = \left( \gamma k_x \nabla_x C(\vec{k}, t) \right) - \nabla_y C(\vec{k}, t)
\]

\[
\frac{\partial \phi(\vec{k}, t)}{\partial t} = -\Gamma k_x^2 \frac{\delta}{\delta \phi(\vec{k}, t)} + \eta_k(t)
\]

and the field transformation

\[
\phi_x(t) = b^{\alpha_x} k_x \phi_x(t')
\]

\[
k_x \rightarrow k'_x = k_x b^{\alpha_x}, \quad k_y \rightarrow k'_y = k_y b^{\alpha_y}, \quad t \rightarrow t' = tb^{-1}
\]

The RG scheme of \[13\] by considering the change of scale \[10\] and the field transformation

\[
\phi_x(t) = b^{\alpha} \phi_x(t')
\]

where \(b\) is the rescaling factor and the meaning of the exponents \(\alpha_x, \alpha_y, \zeta\) will be clarified in the following. Dimensional analysis implies that the structure factor can be written as

\[
C(\vec{k}, t) = R_x(t)R_y(t)f(x, y)
\]

with \(x = k_x R_x(t), y = k_y R_y(t)\). Its invariance in form with respect to the transformations \[14\] gives \(\zeta = (\alpha_x + \alpha_y)\).
The invariance of the scaling variables \( x, y \) under the transformations \( (5) \) implies that \( \alpha_x, \alpha_y \) correspond to the growth exponents.

The dynamical scaling regime is described by a fixed point of the above recursions with \( \Gamma' \), \( \gamma' \) and flow (upper curve) directions. The straight line has slope 4/3.

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**FIG. 4.** Evolution of the average domain size in the shear (lower curve) and flow (upper curve) directions. The straight line has slope 4/3.

A fixed point of the above recursions with \( \Gamma', \gamma' \neq 0 \) is obtained when \( \alpha_y = 1/3, \alpha_x = \alpha_y + 1 \), with the temperature being not relevant for the process of phase separation. We observe that a difference between the growth exponents \( \Delta \alpha = \alpha_x - \alpha_y \) in the range 0.8 - 1 has been measured in [3,4]. Moreover, the above analysis suggests that, for a constant value of the strain \( \gamma t \), the excess viscosity scales as \( \Delta \eta \sim \gamma^{-\beta} \) with \( \beta = 1/3 \).

In conclusion, we have studied the phase-separation kinetics of a binary fluid in an uniform shear flow by direct numerical simulation of the constitutive equations and in a RG approach. Results show the simultaneous existence of domains of two characteristic sizes in each direction. The two kind of domains alternatively prevail, because the thicker are thinned by the strain and the thinner are thickened after cascades of ruptures in the network. This mechanism produces an oscillation which decorates the expected power-law growth \( R(t) \sim t^\alpha \) of the average size of the domains, with \( \alpha = 4/3 \) and 1/3 in the flow and shear direction, respectively. The oscillations occur on logarithmic time-scale as in models describing propagation of fractures in materials where the releasing of elastic energy is measured [18]. Finally, in a recent paper [19], a first treatment of the effects of hydrodynamics on the phase separation of a binary mixture in uniform shear has been given. It would be interesting to know if and how hydrodynamics affects the global picture described in this letter.

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