Anisotropic Spinodal Decomposition under Shear Flow

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When a critical fluid is brought into the unstable region in the presence of shear flow, growing fluctuations are greatly elongated in the flow direction, giving rise to strongly anisotropic light scattering. In the strong shear case the linear growth theory becomes applicable in a sizable time region $0 < t \lesssim t_{c'}$ in which the characteristic size along the flow direction grows as Dt/x, $1/x$ being the correlation length perpendicular to the flow in the strong shear case. For $t \gtrsim t_c$ the characteristic size in the perpendicular directions starts to increase. By applying a computational method of Langer, Bar-on and Miller, it is found to increase as $t^{a'}$ with $a' \cong 0.2$, whereas the characteristic size in the flow direction continues to increase roughly as t.

§ 1. Introduction

Recently an experiment of the spinodal decomposition has been performed in a critical binary mixture in the presence of a periodic shear flow.[1] One of marked results is that the spinodal ring in light scattering is strongly anisotropic, which means that the growing clusters are greatly elongated in the flow direction. In the experiment the spinodal decomposition has been realized periodically[2],[3] and the fluid appears to have been mostly in the strong shear regime $D\tau_{\varepsilon} \gtrsim 1$[4]~[7] except for narrow time intervals in which $D \cong 0$. Here D is the shear rate and τ_{ε} is the equilibrium characteristic relaxation time of the order parameter $(16\eta_0/k_B T_c)\xi^3, \eta_0$ and ξ being the shear viscosity and the (equilibrium) correlation length in the final state.*) For $D\tau_{\varepsilon} \ll 1$, however, only considerably developed clusters can be affected by the shear, so the spinodal ring starts out being isotropic and subsequently develops anisotropy.

In this paper we investigate the anisotropic growth of the fluctuations in the stationary, strong shear case. Here, the shear strongly suppresses the growth of the fluctuations. As a result, the role of the nonlinear interactions among the growing fluctuations is greatly reduced in an early stage, in which the liner growth theory holds. This time region is expressed as $0 < t < t_c$ with

$$t_c \sim 3(16\eta_0 k_c/x^4 k_B T)\log[\bar{\tau}_s(D)/(1 - T_f/T_c)], \qquad (1\cdot1)$$

where k_c and $\bar{\tau}_s(D)$ are a characteristic wave number and a characteristic reduced temperature defined in $(2\cdot9)$ and $(2\cdot16)$ below, respectively T_f is the final temperature, and T_c is the critical temperature slightly dependent on D. In this time region the characteristic wave numbers parallel and perpendicular to the flow direction are $k_{\parallel}(t) = x/Dt$ and $k_{\perp}(t) = x$, respectively, where $x \propto (1 - T_f/T_c)^{1/2}$ is independent of t.

For $t > t_c$ the nonlinear interactions become important and $k_{\parallel}(t)$ and $k_{\perp}(t)$ will decrease obeying some different power laws. It would be convenient to express the

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*) For $D\tau_{\varepsilon} \gg 1$ the characteristic size and the life time of the fluctuations are no longer given by ξ and τ_{ε}.

scattered light intensity in the form[8]

$$I_k(t) = k_\parallel(t)^{-1} k_\perp(t)^{-2} F(k_x/k_\parallel(t), \boldsymbol{k}_\perp/k_\perp(t)), \tag{1·2}$$

where the x-axis is taken to be along the flow and $\boldsymbol{k}_\perp = (k_y, k_z)$ is the perpendicular part of $\boldsymbol{k}$ to the flow. Recall that in the usual case without flow $I_k(t)$ is spherical and can be written as $k(t)^{-3} F(k/k(t))$, where $F(x)$ is nearly independent of time. On the other hand, our scaling function in (1·1) will change its functional form at $t \cong t_c$. In this paper we calculate $k_\parallel(t)$ and $k_\perp(t)$ for $t \gtrsim t_c$ using a computational method of Langer, Bar-on and Miller.

This paper is organized as follows. In §2, we introduce basic stochastic equations and characteristic parameters in the presence of shear. In §3, we investigate consequences of the linear approximation and find an anisotropic growth of the fluctuation spectrum. In §4, we present a nonlinear theory and the onset time $t_{c'}$ (1·1), is estimated.

§ 2.   Model

We consider a binary fluid mixture under a shear flow of the form

$$\boldsymbol{u}(\boldsymbol{r}) = Dy \boldsymbol{e}_x, \tag{2·1}$$

$\boldsymbol{e}_x$ being the unit vector along the x-axis. Near the criticality the relevant variables are the order parameter (=concentration deviation from the critical value) $s(\boldsymbol{r}, t)$ and the transverse part of the deviation of the local velocity from its average $\boldsymbol{u}(\boldsymbol{r})$ which is denoted by $\boldsymbol{v}(\mathbf{r}, t)$. They are governed by

$$\frac{\partial}{\partial t} s = L_0 \nabla^2 \frac{\delta}{\delta s} \varPhi - Dy \frac{\partial}{\partial x} s - \boldsymbol{v} \cdot \nabla s + \theta, \tag{2·2}$$

$$\frac{\partial}{\partial t} \boldsymbol{v} = \eta_0 \nabla^2 \boldsymbol{v} - (\nabla s \frac{\delta}{\delta s} \varPhi)_\perp + (\boldsymbol{\zeta})_\perp, \tag{2·3}$$

where $(\cdots)_\perp$ denotes taking the transverse part, L_0 and η_0 are (bare) kinetic coefficients, θ and ζ are Gaussian Markov noises characterized by

$$\langle \theta(\boldsymbol{r}, t) \theta(\boldsymbol{r}', t') \rangle = -2L_0 \nabla^2 \delta(\boldsymbol{r} - \boldsymbol{r}') \delta(t - t'), \tag{2·4}$$

$$\langle \zeta_\alpha(\boldsymbol{r}, t) \zeta_\beta(\mathbf{r}', t') \rangle = -2\eta_0 \nabla^2 \delta(\boldsymbol{r} - \boldsymbol{r}') \delta(t - t') \delta_{\alpha\beta} \tag{2·5}$$

with no correlation between θ and ζ. The thermodynamic potential $\varPhi$ is of the form

$$\varPhi = \int d\boldsymbol{r} [\frac{1}{2} (\nabla s)^2 + \frac{1}{2} r_0 s^2 + \frac{1}{24} g_0 s^4], \tag{2·6}$$

where r_0 is proportional to the temperature difference measured from the (mean field) critical value and g_0 is a positive constant. We are assuming that the fluid is at the critical composition.

In this paper we assume a sudden lowering of the temperature which brings the fluid from the one phase region to the coexistence region. The change of the parameter r_0 is described by

$$r_0 = \begin{cases} r_i > 0 & \text{for} \quad t < 0, \\ r_f = -x^2 < 0 & \text{for} \quad t > 0. \end{cases} \tag{2·7}$$

One of our main purposes is to follow the growth of the fluctuation spectrum

$$I_k(t) = \langle |s_k(t)|^2 \rangle, \tag{2·8}$$

where $s_k(t)$ is the Fourier transform of $s(r,t)$.

In the presence of shear we introduce a characteristic wave number k_c by[4],[5]

$$k_c = (16\eta_0 D/k_B T)^{1/3}. \tag{2·9}$$

In the one-phase region the effect of shear is strong in the case $k_c \xi \gtrsim 1$, ξ being the equilibrium correlation length. There, the fluctuations with wave numbers smaller than k_c are suppressed, giving rise to only small renormalization contributions, and mean field theory can be applied once the short wavelength fluctuations with $k > k_c$ are eliminated. In the case of quench we define a dimensionless number l_0 by

$$l_0 = x/k_c. \tag{2·10}$$

The depth of the quench is represented by $l_0^2 = x^2/k_c^2$. In this paper we are mainly interested in the case of strong shear or shallow quench with $l_0 < 1$ and defer the analysis of the case of weak shear or deep quench with $l_0 > 1$ to the future work.

The fluctuations with wave numbers greater than k_c are insensitive to the quench for $l_0 < 1$ (if the fluid was in the strong shear regime before the quench.) Therefore they may be assumed to be coarse-grained at the starting equations of motion. Among the thermal fluctuations with $k > x$, which can contribute to renormalizing L_0 and g_0, those with wave numbers k in the region $x < k < k_c$ are suppressed by the shear. Therefore we may set

$$L_0 \cong (k_B T/16\eta_0) k_c^{-1} = D k_c^{-4}, \tag{2·11}$$

$$g_0 \cong g^* k_c, \tag{2·12}$$

where $g^* = 16\pi^2 \varepsilon/3 + O(\varepsilon^2)$ to first order in $\varepsilon = 4 - d, d$ being the spatial dimensionality. The critical anomaly of η_0 is weak and negligible. Therefore the upper cutoff wave number in the renormalization scheme is k_c in the strong shear case. Since $x < k_{c'}$ the parameter r_0 is proportional to $T - T_c$ as

$$r_0 = \xi_0^{-2}(k_c \xi_0)^{(\gamma-1)/\nu}(T/T_c - 1), \tag{2·13}$$

ξ_0 being a microscopic length and γ and ν are the usual critical exponents near equilibrium. Thus,

$$x = \xi_0^{-1}(k_c \xi_0)^{(\gamma-1)/2\nu}(1 - T_f/T_c)^{1/2}, \tag{2·14}$$

T_f being the final temperature. From (2·10) we have

$$l_0 = (k_c \xi_0)^{-(1/2\nu + \eta/2)}(1 - T_f/T_c)^{1/2}, \tag{2·15}$$

where η is the Fisher critical exponent and is negligibly small.

The fluctuations with wave numbers greater than k_c gives rise to the multiplicative factors k_c^{-1} in (2·11) and $(k_c \xi_0)^{(\gamma-1)/\nu}$ in (2·13). The usual results near equilibrium can be obtained by replacing k_c in the above factors by ξ^{-1}. The length ξ_0 may be determined

by the equilibrium relation $\xi = \xi_0 (T/T_c - 1)^{-\nu}$. In the strong shear case a characteristic reduced temperature $\bar{\tau}_s(D)$ is defined by

$$\bar{\tau}_s(D) = (k_c \xi_0)^{1/\nu} \cong (16 \eta_0 \xi_0^3 / k_B T)^{0.53} D^{0.53}. \tag{2.16}$$

Then,

$$l_0 \cong (1 - T_f/T_c)^{1/2} / \bar{\tau}_s(D)^{1/2}. \tag{2.17}$$

The fluctuations with wave numbers smaller than k_c are suppressed in the one phase region, giving rise to a slight lowering of the critical temperature as [4),5)]

$$T_c(D) = T_c(O)[1 - 0.0832 \varepsilon \bar{\tau}_s(D)], \tag{2.18}$$

which is valid to first order in ε. In the limit of shallow quench $l_0 \ll 1$ most of the fluctuations with $k < k_c$ are unaffected by the quench and the critical temperature should be lowered as (2.18).

§ 3. Linear approximation

If we neglect the nonlinear interactions, we obtain a linear equation for $I_k(t)$:

$$\frac{\partial}{\partial t} I_k(t) = 2L_0 k^2 - 2L_0 k^2 (k^2 - x^2) I_k(t) + D k_x \frac{\partial}{\partial k_y} I_k(t). \tag{3.1}$$

This is integrated to give

$$I_k(t) = e^{f(k,t)} I_{k(t)}(0) + 2L_0 \int_0^t ds\, e^{f(k,t-s)} k(s)^2, \tag{3.2}$$

where

$$f(k,t) = 2L_0 \int_0^t ds [x^2 k(s)^2 - k(s)^4] \tag{3.3}$$

with

$$k(t) = k + Dt k_x e_y. \tag{3.4}$$

The time scale of the growing fluctuations $1/L_0 x^4$ is longer than the distortion time by the shear D^{-1} in the strong shear case $l_0 < 1$. Hence we are interested in the time region $Dt > 1$. Then we may set in (3.2)

$$k(t)^2 \simeq (k_y + Dt k_x)^2 + k_z^2. \tag{3.5}$$

This approximation is equivalent to replacing k^2 in (3.1) by $k_\perp^2 = k_y^2 + k_z^2$.

The calculations can be conveniently performed using the following scale changes

$$l_\perp = (l_y, l_z) = x^{-1} k_\perp, \tag{3.6}$$

$$l_x = k_c^4 k_x / x^5 = x^{-1} l_0^{-4} k_x, \tag{3.7}$$

$$\tau = L_0 x^4 t \cong (k_B T / 16 \eta_0 k_c) x^4 t, \tag{3.8}$$

$$\mathcal{I}(l, \tau) = x^2 I_k(t). \tag{3.9}$$

Then, (3·1) and (3·2) become

$$\frac{\partial}{\partial\tau}\mathscr{I}(\boldsymbol{l},\tau)=2l_\perp^2-2l_\perp^2(l_\perp^2-1)\mathscr{I}(\boldsymbol{l},\tau)+l_x\frac{\partial}{\partial l_y}\mathscr{I}(\boldsymbol{l},\tau), \tag{3·10}$$

$$\mathscr{I}(\boldsymbol{l},\tau)=e^{F(\boldsymbol{l},\tau)}\mathscr{I}(\boldsymbol{l}(\tau),0)+2\int_0^\tau du e^{F(\boldsymbol{l},u)}l_\perp^2(u), \tag{3·11}$$

where $\boldsymbol{l}(\tau)=\boldsymbol{l}+\tau l_x\boldsymbol{e}_{y'}$ and

$$F(\boldsymbol{l},\tau)=2\int_0^\tau du[l_\perp^2(u)-l_\perp^4(u)], \tag{3·12}$$

$$l_\perp^2(\tau)=(l_y+\tau l_x)^2+l_z^2=l_\perp^2+2\tau l_x l_y+\tau^2 l_x^2. \tag{3·13}$$

Since $F(\boldsymbol{l},\tau)$ is complicated, we neglect the cross term $2\tau l_x l_y$ in (3·13) because it does not affect the limiting behavior of $\mathscr{I}(\boldsymbol{l},\tau)$. Then,

$$F(\boldsymbol{l},\tau)\simeq 2(l_\perp^2-l_\perp^4)\tau+\frac{2}{3}l_x^2(1-2l_\perp^2)\tau^3-\frac{2}{5}l_x^4\tau^5. \tag{3·14}$$

Equation (3·14) indicates that the fluctuations with $l_\perp>1$ cannot be enhanced, so that $l_\perp$ may be assumed to be considerably smaller than 1 and $l_\perp^2-l_\perp^4\cong l_\perp^2$. Note also that $F(\boldsymbol{l},\tau)$ goes to $-\infty$ for $\tau\gg|l_x|^{-4/5}$ and hence $\mathscr{I}(\boldsymbol{l},\tau)$ tends to a limit

$$\chi_l=\mathscr{I}(\boldsymbol{l},\infty). \tag{3·15}$$

For $l_x=0,\mathscr{I}(\boldsymbol{l},\tau)$ grows indefinitely. For $l_x\neq 0$ some manupilations yield

$$\chi_l\cong 2|l_x|^{-4/5}\int_0^\infty dp(l_\perp^2+|l_x|^{2/5}p^2)\exp\Big(2c_1p+\frac{2}{3}c_2p^3-\frac{2}{5}p^5\Big), \tag{3·16}$$

where $p=|l_x|^{4/5}\tau$, use has been made of (3·14), and

$$c_1=(l_\perp^2-l_\perp^4)|l_x|^{-4/5}, \qquad c_2=(1-2l_\perp^2)|l_x|^{-2/5}. \tag{3·17}$$

For $|c_1|<1$ and $|c_2|<1$ or for $l_\perp<|l_x|^{2/5}$ and $|l_x|>1$ the integration region $p\lesssim 1$ is dominant in (3·16) and

$$\chi_l\cong(2/5)^{2/5}\Gamma(3/5)|l_x|^{-2/5}, \tag{3·18}$$

which is nearly equal to the variance in the strong shear case of the disordered phase[4],[5] $I_k\simeq 1/[r_0+\text{const }k_c^{8/5}|k_x|^{2/5}+k^2]$. Thus the fluctuations in that wave number region are insensitive to the quench. On the other hand, those in the region $l_\perp<1$ and $|l_x|<1$ are enhanced. The dominant contribution in the integral (3·16) comes from p that maximizes $2c_1p+\frac{2}{3}c_2p^3-\frac{2}{5}p^5$, namely, from the region $p^2\sim\frac{1}{2}c_2+\sqrt{c_2^2/4+c_1}$. For $l_\perp\ll 1/\sqrt{2}$ this region is given by $p\sim c_2^{1/2}\sim|l_x|^{-1/5}$ and χ_l becomes

$$\chi_l\cong(2\pi)^{1/2}|l_x|^{-1/2}\exp\Big(\frac{4}{15}|l_x|^{-1}\Big), \tag{3·19}$$

where the second term of (3·14) is responsible for the enhancement.

The time when $\mathscr{I}(\boldsymbol{l},\tau)$ begins to saturate is roughly determined by $(\partial/\partial\tau)F(\boldsymbol{l},\tau)=0$ or $l_\perp(\tau)^2=1$. Using (3·13) this yields $\tau\sim 1/|l_x|$ for $l_\perp<1$ and $|l_x|<1$, at which $F(\boldsymbol{l},\tau)\sim 1/|l_x|$. Thus the exponential factor of (3·19) can be reproduced. For $\tau\gg 1/|l_x|,\mathscr{I}(\boldsymbol{l},\tau)$

saturates to χ_l, (3·19).

In experiments the scattered light intensity is strongly anisotropic even at $\tau \sim 1$.

§ 4. Role of nonlinear interactions

In the usual phase separation without flow the nonlinear interactions among fluctuations are important almost immediately after the quench. However, in the presence of shear flow the onset of the nonlinear regime is delayed by the suppression of the fluctuations with $|l_x| = \chi^{-1} l_0^{-4} |k_x| \gtrsim 1$. This means that the growing fluctuations vary very slowly in the flow direction, leading to the reduction of the nonlinear effects.

In this paper we use a computational scheme of Langer, Bar-on and Miller,[9] which will be referred to as LBM. Then we obtain the following equation for $I_k(t)$:

$$\frac{\partial}{\partial t} I_k(t) = 2L_0 k^2 - 2L_0 k^2 (k^2 - \chi^2 \mu) I_k(t) + D k_x \frac{\partial}{\partial k_y} I_k(t)$$

$$+ \frac{1}{(2\pi)^3} \frac{k_B T}{\eta_0} \int d\boldsymbol{q} \frac{|\boldsymbol{k} \times \boldsymbol{q}|^2}{|\boldsymbol{k} - \boldsymbol{q}|^4} [(k^2 - q^2) I_k(t) I_q(t) + I_k(t) - I_q(t)]. \quad (4 \cdot 1)$$

The last term arises from the hydrodynamic interaction, which has been obtained by the decoupling approximation.[10] Following LBM, we define $\mu(\tau)$ as

$$\mu(\tau) = 1 - \frac{g_0}{6\chi^2} \langle s^4(\boldsymbol{r}, t) \rangle / \langle s^2(\boldsymbol{r}, t) \rangle. \quad (4 \cdot 2)$$

The quantity $-\chi^2 \mu(\tau)$ represents the effective temperature deviation. The integration in the last term of (4·1) may be restricted within the needle-like region $k_\perp \lesssim \chi$ and $|k_x| \lesssim \chi l_0^4$.

Using (2·11) and the scale changes (3·6)~(3·9) we have

$$\frac{\partial}{\partial \tau} \mathcal{I}(\boldsymbol{l}, \tau) = 2l_\perp^2 - 2l_\perp^2 (l_\perp^2 - \mu(\tau)) \mathcal{I}(\boldsymbol{l}, \tau) + l_x \frac{\partial}{\partial l_y} \mathcal{I}(\boldsymbol{l}, \tau)$$

$$+ \frac{2}{\pi^3} l_0^3 \int d\boldsymbol{m} \frac{|\boldsymbol{l}_\perp \times \boldsymbol{m}_\perp|^2}{|\boldsymbol{l}_\perp - \boldsymbol{m}_\perp|^4} [(l_\perp^2 - m_\perp^2) \mathcal{I}(\boldsymbol{l}, \tau) \mathcal{I}(\boldsymbol{m}, \tau) + \mathcal{I}(\boldsymbol{l}, \tau) - \mathcal{I}(\boldsymbol{m}, \tau)].$$

$$(4 \cdot 3)$$

The integral in (4·3) is in the region $|m_x| < 1$ and $m_\perp < 1$. The factor l_0^3 in (4·3) and (A·9) below greatly reduces the magnitude of the nonlinear interaction. This is one of new effects brought about by the shear flow. Furthermore, expecting that $\mathcal{I}(\boldsymbol{l}, \tau)$ is only weakly anisotropic in the l_y-l_z plane (if l_x is fixed),[5] the angular integration in the last term of (4·3) may be carried out to give

$$\text{the last term} = \frac{2}{\pi^2} l_0^3 \int_0^1 dm_\perp m_\perp \int_{-1}^1 dm_x Q(l_\perp / m_\perp)$$

$$\times [(l_\perp^2 - m_\perp^2) \mathcal{I}(\boldsymbol{m}, \tau) \mathcal{I}(\boldsymbol{l}, \tau) - \mathcal{I}(\boldsymbol{m}, \tau) + \mathcal{I}(\boldsymbol{l}, \tau)], \quad (4 \cdot 4)$$

where

$$Q(x) = Q(1/x) = (x^2 + 1)/|x^2 - 1| - 1. \quad (4 \cdot 5)$$

The quantity in the brackets of (4·4) vanishes at $l_\perp = m_\perp$ when integrated with respect to

m_x. Hence, although $Q(x)$ diverges at $x=1$, the integral in (4·4) is well-defined.

4.1. *Onset time of the nonlinear regime*

At early time we can assume the Gaussian distribution for $s(\boldsymbol{r},t)$ and we may approximate $\mu(\tau)$, (4·2), as

$$\mu(\tau)=1-(g^*l_0{}^3/16\pi^3)\int d\boldsymbol{m}\,\mathcal{I}(\boldsymbol{m},\tau). \qquad (4\cdot6)$$

The second term of (4·6) grows exponentially in time and the onset time τ_c is roughly estimated by setting $\mu(\tau_c)=1/2$ or

$$(g^*l_0{}^3/8\pi^3)\int d\boldsymbol{m}\,\mathcal{I}(\boldsymbol{m},\tau_c)=1. \qquad (4\cdot7)$$

Assuming (3·11) and (3·14) and setting $\mathcal{I}(\boldsymbol{l},0)=0$ for simplicity, we have

$$\tau_c\cong6\log(1/l_0). \qquad (4\cdot8)$$

On the other hand, the last term of (4·3) enhances the kinetic coefficient and make it time-dependent as $L(l,\tau)=L_0+\varDelta L(l,\tau)$. This interaction is not negligible for $\tau>\tau_H$ where τ_H is determined by $\varDelta L(\boldsymbol{l},\tau_H)\sim L_0$. Some perturbation calculations readily show that $\tau_H\sim\tau_c\sim6\log(1/l_0)$. Namely, the two nonlinear interactions become important roughly at the same time $\tau=\tau_c$.

4.2. *Intensities in the linear regime*

In the time region $0<\tau<\tau_c$ the linear theory is applicable and (3·11) is valid. Note that $\mathcal{I}(\boldsymbol{l},\tau)$ is nearly equal to (3·19) for $\tau>1/|l_x|$ in the region $l_\perp\ll1/\sqrt{2}$ and $|l_x|\ll1$. Thus, for $\tau\lesssim\tau_c$ we have

$$\mathcal{I}(\boldsymbol{l},\tau)\cong(2\pi)^{1/2}|l_x|^{-1/2}\exp\left(\frac{4}{15}|l_x|^{-1}\right) \quad \text{for} \quad \tau^{-1}<|l_x|<1 \text{ and } l_\perp\ll1/\sqrt{2}. \qquad (4\cdot9)$$

For $|l_x|<1/\tau$ and $l_\perp<1/\sqrt{2}$ we have

$$\mathcal{I}(\boldsymbol{l},\tau)\cong2e^{F(\boldsymbol{l},\tau)}\cong2\exp\left(2l_\perp{}^2\tau+\frac{1}{3}l_x{}^2\tau^3\right), \qquad (4\cdot10)$$

where the first term of (3·11) is neglected. For $l_\perp\ll1$ the maximum of $\mathcal{I}(\boldsymbol{l},\tau)$ is attained at $|l_x|\sim1/\tau$ and is of order $\exp(4\tau/15)$ which reaches $\exp(4\tau_c/15)\sim l_0{}^{-8/5}$ at $\tau=\tau_c$.

Thus the characteristic size in the flow direction, $\xi_\parallel(t)$, is given by

$$\xi_\parallel(t)=1/k_\parallel(t)=\varkappa^{-1}l_0{}^{-4}\tau=\varkappa^{-1}Dt, \qquad (4\cdot11)$$

whereas that perpendicular to the flow, $\xi_\perp(t)=1/k_\perp(t)$, is fixed at $\varkappa^{-1}$. In the early stage, $\tau<\tau_c$ or $t<\tau_c/L_0\varkappa^4$, the intensity $I_k(t)$ continues to be peaked along the k_x-axis. The scattered light intensity in strong only in the needle-like region $|k_x|\lesssim\varkappa/Dt$ and $|k_\perp|\lesssim\varkappa$.

4.3. *Nonlinear regime*

In the presence of the flow term $-l_x(\partial/\partial l_y)\mathcal{I}(\boldsymbol{l},\tau)$, (4·3) becomes extremely complicated when integrated. Therefore, further simplifications are needed at present to make the problem tractable. First, in determining $\mu(\tau)$ we neglect the effect of the hydrodynamic interaction which would affect $\mathcal{I}(\boldsymbol{m},\tau)$ appearing in the integral (A·10).

A similar approximation was used in Ref. 10). Second, the hydrodynamic interaction will be taken into account only as a first order perturbation. That is, first numerically solving (A·8) we have replaced $\mathscr{I}(\boldsymbol{m},\tau)$ in the integrand of (A·10) by

$$\mathscr{I}(\boldsymbol{m},\tau)\cong 2\int_0^\tau du e^{-G(\boldsymbol{m},\tau,u)}\boldsymbol{m}(u)^2, \tag{4·12}$$

where

$$G(\boldsymbol{m},\tau,u)=2\int_0^u dv m_\perp^2(v)(m_\perp^2(v)-\mu(\tau-v)). \tag{4·13}$$

Then (A·8), (A·10) and (4·12) constitute a closed set of equations and $\mu(\tau)$ can be obtained numerically. After that, $\mathscr{I}(\boldsymbol{l},\tau)$ is calculated from (4·3), where $\mathscr{I}(\boldsymbol{m},\tau)$ in the hydrodynamic interaction is replaced by (4·12), however.

In Fig. 1, $\mu(\tau)$ is plotted against τ for various l_0, where we have set $\mathscr{I}(\boldsymbol{l},0)=0$ for simplicity. For $\tau\gtrsim\tau_c,\mu(\tau)$ begins to decrease abruptly. The presence of the onset time τ_c is apparent. Subsequently, however, the decrease of $\mu(\tau)$ is decelerated by the slow variations of long wavelength fluctuations. $\mathscr{I}(\boldsymbol{l},\tau)$ is found to be strongly anisotropic. Its peak positions can be obtained for each fixed $\theta\equiv\tan^{-1}(l_y/l_x)$ as a closed curve in the l_x-l_y plane, where $l_z=0$, as displayed in Fig. 2. We write the values of the peak intensities at several points in Fig. 2. The peak intensity at $l_x=0$ is much larger than that of the usual case without flow, which is a consequence of the delayed onset of the nonlinear regime. For $l_\perp=0$, the peak intensity begins to decrease as $\mathscr{I}(\boldsymbol{l},\tau)$ approaches the final steady value.

The peak positions are plotted against τ at $\theta=0$ and $\pi/2$ in Fig.3. At $\theta=\pi/2$, the peak position $l_m(\tau)$ for $\tau>\tau_c$ will be represented by a power law

$$l_m(\tau)\sim\tau^{-a'}. \tag{4·14}$$

Here $a'\sim0.2$ for $\tau\gtrsim\tau_c$ which is independent of l_0 and is close to LBM's value $a'=0.21$. At $\theta=0$, we have found $a'\sim1$. Namely, the nonlinear interaction does not affect the exponent in the flow direction. However, we must note that it might be changed by a more satisfactory treatment of the hydrodynamic interaction.
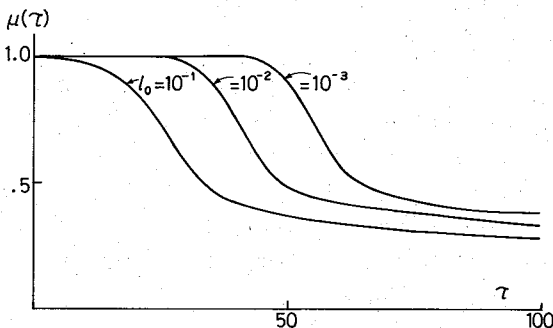

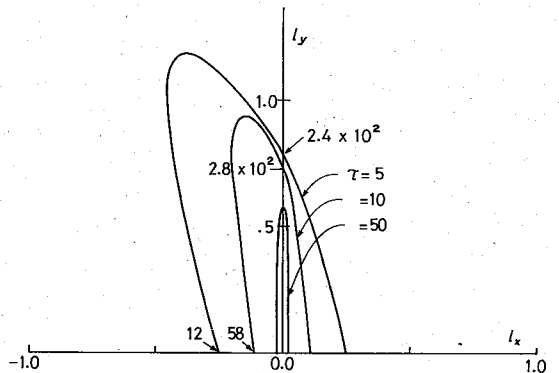Fig. 1. $\mu(\tau)$ vs τ for various l_0. The logarithmic dependence of τ_c on l_0 is clearly seen.

Fig. 2. Time-development of peak positions of $\mathscr{I}(l,\tau)$ in the l_x-l_y plane for $l_0=0.1$. The curves are the trajectories of the maxima obtained with the angle $\theta=\tan^{-1}(l_y/l_x)$ fixed.
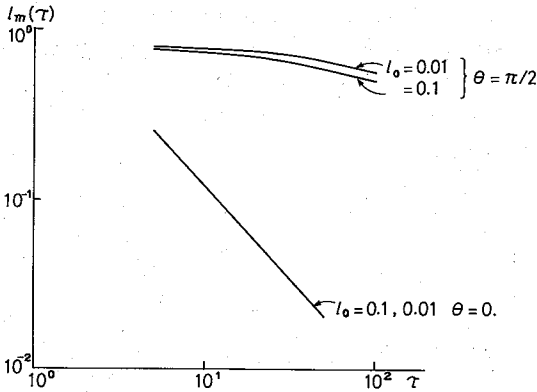
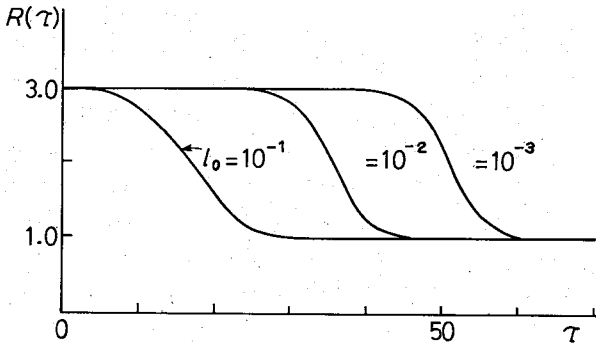Fig. 3. Peak positions $l_m(\tau)$ vs τ at $\theta = 0$ and $\pi/2$ for various l_0.

Fig. 4. $R(\tau) \equiv \langle x^4 \rangle / \langle x^2 \rangle^2$ vs τ.

In Fig. 4, we show $R(\tau) \equiv \langle x^4 \rangle / \langle x^2 \rangle$ versus τ where $x = (g^* k_c / 6\chi^2)^{1/2} s(r, t)$ (see the Appendix). Here $R(\tau) \simeq 3$ for $\tau \lesssim \tau_{c'}$ indicating that the distribution of s is nearly Gaussian. At long time periods $R(\tau)$ becomes very close to 1. Then the one-point distribution function $\rho_1(x, \tau)$ defined by (A·1) has very sharp two peaks at $\pm(\langle x^2 \rangle)^{1/2}$. The two peaks are much sharper than in the case of the original LBM. This is because the growing fluctuations must have wave numbers with very small x-components. Then the wave number integration becomes very small as compared to the case without flow and $\rho_1(x, t)$ attains a structure with δ-function-like peaks. However, the shift of the peak positions to greater absolute values is slow as in the case without flow.

§ 5. Summary and discussion

In this paper we have examined the phase separation process in critical fluids under shear flow. The new aspect of the problem is the competition of the growth of fluctuations due to the thermodynamic instability and its suppression by the shear flow. The suppression delays the onset of the nonlinear regime, giving rise to a possibility of detecting the linear regime experimentally in the case of strong shear. The fluctuation spectrum $I_k(t)$ is strongly anisotropic reflecting elongated spatial regions of the emerging new phase. In the flow direction, the position of the maximum of $I_k(t)$, $k_{\parallel}(t)$, changes as $k_{\parallel}(t) \sim t^{-1}$ after the quench. In the direction perpendicular to the flow, the position $k_{\perp}(t)$ remains stationary at $\varkappa/\sqrt{2}$ in the linear regime, and behaves as $k_{\perp}(t) \sim t^{-a'}$ in the nonlinear regime. The value of a' has been estimated to be about 0.2 by numerical analysis. This value coincides with that of a solid model.[9] It may still be altered by a more satisfactly treatment of the hydrodynamic interaction than the one in this paper.

We also remark that the fluctuations in the flow direction with size smaller than $1/k_{\parallel}(t)$ are suppressed and do not affect the long time behavior of $I_k(t)$. In this sense our problem is asymptotically a two-dimensional phase separation. The extremely large light scattering intensity in the flow direction (see (3·19), (4·9) and (4·10)) can be attributed to this effective reduction of the spatial dimensionality in the course of phase separation under flow.

Finally we must comment on the data of Beysens and Perrot.[1] They made the shear oscillate as $D(t) = D_0[\cos(\Omega t) + 1]/2$. There, the temperature T was fixed, while $T_c(D)$ was oscillated. Therefore when $T_c(D)$ is slightly greater than T, they should have observed the spinodal decomposition in the strong shear case. However, only when $D = 0$, they measured the characteristic wave numbers $k_\parallel$ and $k_\perp$ from the anisotropic spinodal ring and they found that $\Delta \equiv k_\perp/k_\parallel \sim 2$ and $k_\perp \propto \Omega^{1/3}$. In such cases the effect of shear is weakest; therefore, quantitative comparison of our results and their data is difficult unfortunately. The periodic case has its own new features and should be analyzed in the future.[2],[3]

Ackhowledgements

Appendix

In the LBM scheme a truncation approximation is made for the one-point distribution function

$$\rho_1(s_1, t) = \langle \delta(s(\boldsymbol{r}, t) - s_1) \rangle. \tag{A·1}$$

Similarly to the hydrodynamic interaction[7] the flow term $Dy(\partial/\partial x)s$ in (2·2) does not affect the equation for $\rho_1(s_1, t)$. The original equation of LBM remains unchanged as

$$\frac{\partial}{\partial t} \rho_1(s, t) = L_0 \frac{\partial}{\partial s} \Big[G + (\Delta/V_c) \frac{\partial}{\partial s} \Big] \rho_1(s, t), \tag{A·2}$$

where

$$G = \frac{1}{6} g_0 \Delta [s^3 - \langle s^3 \rangle - \langle s^4 \rangle s / \langle s^2 \rangle] + Ws / \langle s^2 \rangle, \tag{A·3}$$

$$W = (2\pi)^{-3} \int d\boldsymbol{k} k^2 (k^2 - k^2 \mu) I_k. \tag{A·4}$$

However, the wave number of s is restricted within the region $|k_x| < \varkappa l_0^4$ and $k < \varkappa$. Then the coarse-grained volume V_c becomes much greater than that of LBM as

$$V_c^{-1} = (2\pi)^{-3} \int d\boldsymbol{k} = 2(4\pi^2)^{-1} \int_0^{\varkappa l_0^4} dk_x \int_0^k dk_\perp k_\perp = (4\pi^2)^{-1} \varkappa^3 l_0^4. \tag{A·5}$$

The quantity Δ is defined by

$$\Delta = V_c (2\pi)^{-3} \int d\boldsymbol{k} k^2 = \frac{1}{2} \varkappa^2. \tag{A·6}$$

Next we make the equations dimensionless using the scale changes (3·6)~(3·9) and setting $x = s/M$, where

$$M = (6\varkappa^2/g_0)^{1/2} = (6\varkappa l_0/g^*)^{1/2}. \tag{A·7}$$

Then,

$$\frac{\partial}{\partial \tau}\rho_1 = \frac{1}{2}\varepsilon_0\frac{\partial^2}{\partial x^2}\rho_1 + \frac{\partial}{\partial x}\left[\frac{1}{2}(x^3 - \langle x^3\rangle - \langle x^4\rangle x/\langle x^2\rangle) + \varepsilon_0\bar{S}x/\langle x^2\rangle\right]\rho_1, \qquad (A\cdot8)$$

where

$$\varepsilon_0 = 2\varDelta/(V_c\chi^4M^2) = (g^*/24\pi^2)l_0^3 \ll 1, \qquad (A\cdot9)$$

$$\bar{S} = \int_{-1}^{1}dm_x\int_0^1 dm\ m^3(m^2-\mu)\mathcal{I}_m, \qquad (A\cdot10)$$

$$\mu = 1 - \langle x^4\rangle/\langle x^2\rangle, \qquad (A\cdot11)$$

We can make the hierarchy of the equations for the moments $\langle x^n\rangle$ and it can be truncated by assuming a displaced double Gaussian form for ρ_1 as in the original LBM.

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