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Phase-ordering dynamics of binary mixtures with field-dependent mobility in shear flow

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Abstract. The effect of shear flow on the phase-ordering dynamics of a binary mixture with field-dependent mobility is investigated. The problem is addressed in the context of the time-dependent Ginzburg-Landau equation with an external velocity term, studied in self-consistent approximation. Assuming a scaling ansatz for the structure factor, the asymptotic behavior of the observables in the scaling regime can be analytically calculated. All the observables show log-time periodic oscillations which we interpret as due to a cyclical mechanism of stretching and break-up of domains. These oscillations are damped as consequence of the vanishing of the mobility in the bulk phase.

PACS. 47.20.Hw Morphological instability; phase changes – 05.70.Ln Nonequilibrium and irreversible thermodynamics – 83.50.Ax Steady shear flows

1 Introduction

A well-studied problem in non-equilibrium statistical mechanics is the growth of domains in quenching processes [1]. Namely, when a system is suddenly quenched from a disordered initial state into a thermodynamic region where different phases coexist, macroscopic domains can be observed, usually characterized by a single time-dependent length scale, which grows as a power law $L(t) \sim t^z$. The spatial patterns of the domains at two different times are related by a global change of this length scale. A signature of this dynamical scaling is the fact that the structure factor $C(\mathbf{k}, t)$ can be cast in the form

$$C(\mathbf{k}, t) = L^d(t) C_o(\mathbf{k} L(t)) \qquad (1)$$

where C_o is the scaling function.

In the case of a binary mixture, the evolution of the system is described by a scalar order parameter ϕ representing the difference of concentrations of the two liquids in the mixture. When hydrodynamic effects are not considered, the quenching process can be described by an equation of the form $\partial_t \phi = -\nabla \cdot \mathbf{j}$, where $\mathbf{j} = -\Gamma \nabla (\delta H / \delta \phi)$, H is a free-energy functional describing the ordered phases and Γ is a transport coefficient called mobility, usually taken constant. It has been argued [2] that the dynamics is more accurately mimicked considering a field-dependent mobility of the form

$$\Gamma(\phi) = (1 - a(T)\phi^2) \qquad (2)$$

where $a(T) \to 1$ for temperature $T \to 0$, $a(T) \to 0$ for $T \to T_c$. In this way it is possible to take into account the different nature of the two mechanisms which operate during the phase separation: surface diffusion and bulk diffusion. The first has a growth exponent $z = 1/4$ [3] and is due to the diffusion of molecules of the two species along the interfaces. The second, which is also called Lifshitz-Slyozov mechanism, is due to the diffusion of molecules of one species from more curved interfaces, where they evaporate, to less curved ones through the bulk of the other phase. The corresponding growth exponent is $z = 1/3$ [4]. At high temperatures (but still less than the critical temperature) the bulk diffusion is the only observed because it is faster than the other. Since bulk diffusion is a thermal activated process [5], one expects that, lowering the temperature, a regime with $z = 1/4$ can be observed. The proposed form (2) for the mobility is able to catch these features. For shallow quenches ($a \ll 1$) the mobility remains constant in the whole system, while for very deep quenches ($a = 1$) $\Gamma(\phi)$ vanishes in the bulk phases where $\phi^2 = 1$, suppressing the bulk diffusion. The diffusion along the interfaces is unaffected because $\Gamma(\phi) \simeq 1$ on domain boundaries.

The effect of an order parameter-dependent mobility (2) on systems with scalar order parameter has been studied by simulations [6,7]. It has been found that for $a = 1$ the length scale $L(t)$ grows as $t^{1/4}$ and for $0 < a < 1$ there is a crossover between $L(t) \sim t^{1/4}$ and $L(t) \sim t^{1/3}$. Recently, a non constant mobility has been also used to study the phase-ordering dynamics in systems with a vectorial order parameter [5,8]. In [5] the expression (2) for

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the mobility has been adopted. The limit $N \to \infty$, where N is the number of vectorial components, is analytically solvable and corresponds to the self-consistent approximation of the scalar case. It is characterized by a growth exponent 1/6 for the case $a = 1$. For $0 < a < 1$ the usual value 1/4 of the growth exponent for vectorial systems is recovered in the asymptotic regime. A more general form of the mobility has been introduced in [8], being given by

$$\Gamma(\phi) = (1 - \phi^2)^\alpha \tag{3}$$

where α is a positive real number.

In this paper we are concerned with the phase separation of a binary mixture subject to a uniform shear flow when the mobility is given by equation (3). When a shear flow is applied to a quenched binary mixture, the pattern of the phase-separating domains as well as the time evolution are strongly modified by the flow [9]. Domains greatly elongated in the flow direction have been observed in simulations [10,11] and in experiments [12]. A difference $\Delta z = 0.8 \div 1$ between growth exponents has been measured in some experiments, with the larger exponent in the flow direction [13,14]. The deformation of domains affects the rheological properties of the system, giving rise to an increase of the stress tensor, which is proportional to $\frac{1}{V}\int \mathrm{d}\mathbf{r}(\partial_\alpha\phi)(\partial_\beta\phi)$, where V is the system volume and α and β denote the spatial directions [15]. Since $(\nabla\phi)(\nabla\phi)$ behaves like a δ-function near the interface, during the phase separation larger stress contributions arise from interface deformations. This produces an increase $\Delta\eta$ of the viscosity, which is proportional to the diagonal term of the stress tensor [15,16]. This behavior has its explanation in the fact that mechanical energy is expended to deform the domains against the interfacial tension. When the domains are stretched to such an extent that they start to burst, the stored mechanical energy is dissipated and the excess viscosity decreases. The phase separation in steady shear for binary mixtures with constant mobility has been recently studied in [17]. There, using a self-consistent approximation, an equation for the time evolution of the structure factor has been derived and solved numerically. The existence of an anisotropic dynamical scaling theory with different growth exponents in flow and in other directions has been shown: the growth exponent in the shear direction is not affected by the shear and the difference between them is 1. It is found that the excess viscosity $\Delta\eta$, after reaching a maximum, relaxes to zero, exhibiting log-time periodic oscillations. Also other physical observables are modulated by such oscillations, which can be related to a cyclical mechanism of storing and dissipation of elastic energy. Here we want to see the effects of a non constant mobility on this *scenario*.

The outline of the paper is as follows. In Section 2 we present the model and derive the equation of time evolution for the structure factor in a self-consistent approximation. In Section 3 we report the asymptotic behavior of the model. We found two different growth exponents for the flow and the shear directions, given in the leading scaling regime by $z_x = (5+2\alpha)/2(2+\alpha)$ and $z_y = 1/2(2+\alpha)$, respectively. The asymptotic behavior of the rheological quantities is also calculated. Finally, in Section 4 we integrate numerically the time evolution equation of the structure factor and calculate the whole evolution of the physical observables as moments of the structure factor. In the asymptotic regime they are modulated by damped log-time oscillations. Our conclusions complete the article.

2 The model

We consider a binary mixture whose evolution is described by the diffusion-convection equation

$$\frac{\partial\phi(\mathbf{r},t)}{\partial t} + \nabla\cdot(\phi(\mathbf{r},t)\mathbf{v}) = \nabla\cdot\left[\Gamma(\phi)\nabla\left(\frac{\delta H[\phi(\mathbf{r},t)]}{\delta\phi}\right)\right] \tag{4}$$

where the field $\phi(\mathbf{r},t)$ describes the concentration difference between the two components of the mixture, $\Gamma(\phi)$ is the mobility, which depends on the order parameter as in equation (3). The order parameter ϕ is convected by an external velocity field [9]. We choose a planar shear flow with

$$\mathbf{v} = \gamma y \mathbf{e}_x, \tag{5}$$

where γ is the shear rate, assumed constant, and $\mathbf{e}_x$ is a unit vector in the flow direction. The free energy functional is chosen to be of the standard ϕ^4 form:

$$H[\phi] = \int \mathrm{d}\mathbf{r}\left[-\frac{1}{2}\phi^2(\mathbf{r},t) + \frac{1}{4}\phi^4(\mathbf{r},t) + \frac{1}{2}|\nabla\phi(\mathbf{r},t)|^2\right] \tag{6}$$

where we assume that all parameters have been rescaled into dimensionless units [18] and the system is below the critical temperature. The two minima of the polynomial part of $H[\phi]$ describe the pure states of the mixture.

In this article we deal with the non-linear term of equation (4) in a self-consistent approximation; hence the term ϕ^3 appearing in the functional derivative $\delta H/\delta\phi$ is linearized as $< \phi^2 > \phi$, where $< ... >$ stands for the average over the system. In the same way the term ϕ^2 in the mobility is substituted by $< \phi^2 >$. In the Fourier space equation (4) becomes:

$$\frac{\partial\hat{\phi}(\mathbf{k},t)}{\partial t} = \gamma k_x \frac{\partial}{\partial k_y}\hat{\phi}(\mathbf{k},t) - (1 - S(t))^\alpha k^2\left[S(t) - 1 + k^2\right]\hat{\phi}(\mathbf{k},t) \tag{7}$$

where $S(t) =< \phi^2(\mathbf{r},t) >$ and $\hat{\phi}(\mathbf{k},t)$ is the Fourier transform of $\phi(\mathbf{r},t)$.

The statistical quantity of experimental interest is the time-dependent structure factor $C(\mathbf{k},t)$ which is defined as $< \hat{\phi}(\mathbf{k},t)\hat{\phi}(-\mathbf{k},t) >$. It obeys to the evolution equation:

$$\frac{\partial C(\mathbf{k},t)}{\partial t} = \gamma k_x \frac{\partial}{\partial k_y}C(\mathbf{k},t) - 2(1 - S(t))^\alpha k^2\left[S(t) - 1 + k^2\right]C(\mathbf{k},t) \tag{8}$$

which is closed by the self-consistency condition

$$S(t) = \int_{|\mathbf{k}|<q} \frac{\mathrm{d}\mathbf{k}}{(2\pi)^d}\, C(\mathbf{k},t) \tag{9}$$

where q is a phenomenological cutoff. Rheological quantities of interest can be calculated as momentum integrals of the structure factor. Since we restrict the solution of the model to the two-dimensional case, we consider the excess viscosity $\Delta\eta$ and the first normal stress ΔN_1 defined by [19]

$$\Delta\eta = -\gamma^{-1} \int_{|\mathbf{k}|<q} \frac{\mathrm{d}\mathbf{k}}{(2\pi)^d}\, k_x\, k_y\, C(\mathbf{k},t) \tag{10}$$

$$\Delta N_1 = \int_{|\mathbf{k}|<q} \frac{\mathrm{d}\mathbf{k}}{(2\pi)^d}\, \left[k_y^2 - k_x^2\right] C(\mathbf{k},t). \tag{11}$$

3 The scaling behavior

Assuming simple scaling for the structure factor, we write, for arbitrary space dimensionality d,

$$C(\mathbf{k},t) = \prod_{i=1}^{d} R_i(t) F(\mathbf{X}, \tau(\gamma t)) \tag{12}$$

where the subscript i labels the space directions with $i = 1$ along the flow, R_i is the average size of domains in the i-th direction, $\mathbf{X}$ is a vector of components $X_i = k_i R_i(t)$ and F is a scaling function. Some comments are in order here about the scaling hypothesis. Our self-consistent approximation is equivalent to that made in the large-N limit for vectorial systems [20]. In that case, at $\gamma = 0$, simple scaling is not verified for $\alpha = 0$ [21] and $\alpha \neq 0$ [8] so that $C(\mathbf{k},t)$ has not the form (1) but can be written as $L^{d\xi(k/k_m)}$, where k_m is the position of the maximum in the structure factor and ξ is a function which depends continuously on k (multiscaling). After this article was completed we became aware of a recent work [22], in which the exact solution for the case $\gamma \neq 0$ with constant mobility is found in the large-N limit. In [22] it is shown that the structure factor exhibits multiscaling. At the moment a similar analysis is not available for the case with field-dependent mobility. However, since simple scaling is the leading approximation in the regions of the maxima of the structure factor, we can use it to obtain the correct value of the growth exponents (apart from logarithmic corrections) because the momentum integrals which define the observables are dominated by the maxima of $C(\mathbf{k},t)$. We also allow in (12) an explicit time dependence of the structure factor through $\tau(\gamma t)$; notice that since $C(\mathbf{k},t)$ scales as the domains volume below the critical temperature, τ must not introduce any further algebraic time dependence in $C(\mathbf{k},t)$. From the numerical results of the next section, we will see that F is a damped periodic function of τ.

Inserting the form (12) of $C(\mathbf{k},t)$ into equation (8) we obtain:

$$\gamma X_1 F_2 = R_1 R_2^{-1} \Bigg\{ \dot\tau\, \frac{\partial F}{\partial \tau} + \sum_{i=1}^{d} \Bigg[R_i^{-1} \dot R_i (F + X_i F_i) + 2\Big[1 - S(t)\Big]^{\alpha} R_i^{-2} X_i^2 \Bigg(\sum_{k=1}^{d} R_k^{-2} X_k^2 - 1 + S(t) \Bigg) F \Bigg] \Bigg\} \tag{13}$$

where $F_i = \partial F / \partial X_i$ and a dot means a time derivative. Under the assumptions that $R_1 \gg R_i \quad (i = 2, d)$ and $R_i \simeq \widetilde{R} \quad (i = 2, d)$ we can write

$$\gamma X_1 F_2 = \widetilde{R}^{-1} R_1 \Bigg\{ \dot\tau\, \frac{\partial F}{\partial \tau} + R_1^{-1} \dot R_1 (F + X_1 F_1) + \sum_{i=2}^{d} \Bigg[\widetilde{R}^{-1} \dot{\widetilde{R}} (F + X_i F_i) + 2\Big[1 - S(t)\Big]^{\alpha} \times \widetilde{R}^{-4} X_i^2 \Bigg(\sum_{k=2}^{d} X_k^2 - (1 - S(t)) \widetilde{R}^2 \Bigg) F \Bigg] \Bigg\}. \tag{14}$$

Since the l.h.s. of equation (14) has no explicit algebraic time dependence, one has the asymptotic solutions

$$\begin{aligned} R_1(t) &\sim \gamma\, t^{(5+2\alpha)/2(2+\alpha)} \\ \widetilde{R}(t) &\sim t^{1/2(2+\alpha)} \\ \big(1 - S(t)\big) &\sim t^{-1/(2+\alpha)} \\ \tau(\gamma t) &\sim \log \gamma t. \end{aligned} \tag{15}$$

The growth exponents in the flow and in the shear directions are $z_x = (5 + 2\alpha)/2(2 + \alpha)$ and $z_y = 1/2(2 + \alpha)$. We observe that $z_x - z_y = \dfrac{4 + 2\alpha}{2(2 + \alpha)} = 1$. The value z_y is the same found in [8], in the leading approximation, in a model having a field-dependent mobility with vectorial conserved order parameter without shear. Increasing the values of α, one obtains values smaller with respect to the case with constant mobility, when $z_x = 5/4$ and $z_y = 1/4$. The shear affects only the growth exponent z_x which remains greater than 1 for every real and positive value of α.

The previous arguments can be used to establish the scaling properties of the rheological coefficients. Inserting the form (12) into equation (10) we obtain

$$\Delta\eta(t) \sim (\gamma t)^{-(3+\alpha)/(2+\alpha)} \gamma^{-(1+\alpha)/(2+\alpha)} \times \int X_1 X_2 F\left[\mathbf{X}, \tau(t)\right] \mathrm{d}\mathbf{X}. \tag{16}$$

Therefore, in the scaling regime, for each value of γt, the functions $\Delta\eta$ corresponding to different values of γ collapse each on the others if rescaled as $\Delta\eta \to \gamma^{(1+\alpha)/(2+\alpha)} \Delta\eta$. A similar analysis can be done for the normal stress. It is straightforward to show that

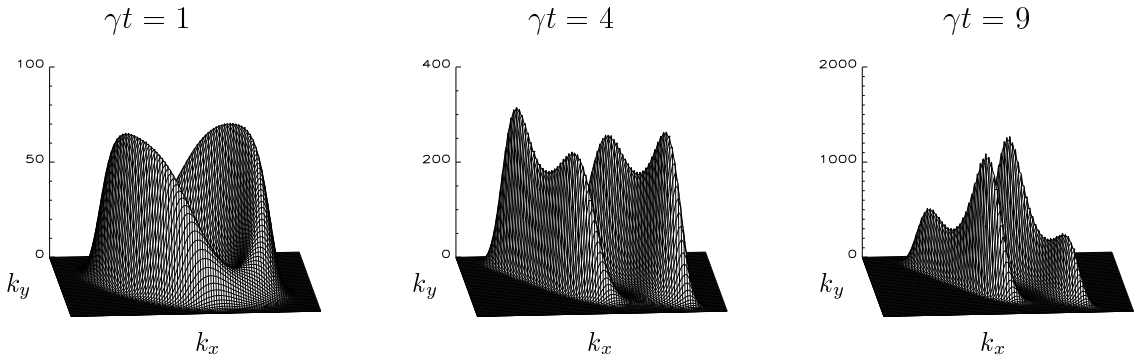

Fig. 1. The structure factor at consecutive times for $\gamma = 0.01$ and $\alpha = 1$. The k_x coordinate assumes positive values on the right of the picture, while the k_y is positive towards the upper part of the plane. The maximum value of $|k_y|$ is 0.8. In the other direction, k_x varies in the range $[-0.8, 0.8]$, $[-0.4, 0.4]$ and $[-0.2, 0.2]$, respectively. The positions of the peaks on the left foil of the structure factor is $(-0.22, 0.37)$ for the highest and $(-0.08, -0.24)$ for the other at $\gamma t = 4$. At $\gamma t = 9$ the highest peak is located at $(-0.05, -0.23)$ and the other, at $(-0.10, 0.36)$.

in the asymptotic regime

$$\Delta N_1 \sim t^{-1/(2+\alpha)} \int X_2^2 F\left[\mathbf{X}, \tau(t)\right] \mathrm{d}\mathbf{X}. \qquad (17)$$

Setting $\alpha = 0$, we recover the previous results for the case with constant mobility [17].

4 Results and discussion

In this section we consider the numerical solution of equation (7). We will present the results for the calculation of the average size of domains and of the rheological indicators $\Delta\eta$ and ΔN_1, stressing the effects of a non-costant mobility. We solved the equation of time evolution of the structure factor numerically in two dimensions, implementing a first-order Euler scheme with an adaptive mesh. The starting configuration for the structure factor is a constant value, which corresponds to a disordered state at very high temperature. Equation (7) has been solved for different values of γ. In the following, results will be shown for the case $\gamma = 0.01$. Similar results have been obtained in the other cases. The two values $\alpha = 1$ and $\alpha = 2$ for the parameter α appearing in (3) have been considered. At the beginning the function $C(\mathbf{k}, t)$ develops a circular volcano shaped structure. This is then deformed, as consequence of shear, into an elliptic structure. The sizes of the axes of the ellipse decrease in time at a different rate, this being larger in the k_x-direction. During this evolution two dips start to develop in the volcano edge until $C(\mathbf{k}, t)$ is made of two foils. In Figure 1 at $\gamma t = 1$, the shape of $C(\mathbf{k}, t)$ representative of this stage of the evolution is plotted. Later, on each foil, two well-formed peaks can be seen. At $\gamma t \simeq 4$ the peaks characterized by the larger values of $|k_y|$ prevail. Observe that the structure factor is symmetric with respect to the change $\mathbf{k} \to -\mathbf{k}$. The peaks with the smaller $|k_y|$ corresponding to a more isotropic configuration of domains, grow faster than the others until they prevail as it can be seen in Figure 1 at $\gamma t = 9$. These peaks continue to prevail along all the time evolution. What we observed is that the peaks continue always to grow and that the difference of their heights as a function of the shear strain γt is an increasing function being modulated by damped oscillations.

In order to get information about the growth of domains, we computed the typical domain size as

$$R_x(t) = \left(\frac{\int \mathrm{d}\mathbf{k} C(\mathbf{k}, t)}{\int \mathrm{d}\mathbf{k} k_x^2 C(\mathbf{k}, t)} \right)^{1/2} \qquad (18)$$

and the same for the other direction. The values of R_x and R_y are plotted in Figure 2 as function of the shear strain γt for $\alpha = 1$ and $\alpha = 2$. We plotted also the values for the case with constant mobility ($\alpha = 0$). Some comments are in order here. The asymptotic behavior is the one expected through the previous scaling analysis: for $\alpha = 1$ one has $R_x \sim t^{7/6}$ and $R_y \sim t^{1/6}$; for $\alpha = 2$, $R_x \sim t^{9/8}$ and $R_y \sim t^{1/8}$. The growth exponents in both the directions are decreasing functions of the mobility exponent α. This is reasonable since the mobility (3) becomes smaller and smaller when α increases, if $(1 - \phi^2) < 1$. Another consideration is about the superimposed log-time periodic oscillations. In the case with constant mobility these oscillations have an apparently constant amplitude. For nonzero values of α these oscillations are damped. We will see that this feature is common to all the observables and will be discussed later.

We turn now to the study of the rheological behavior of the system. The external velocity field causes additional stresses on the mixture. Important indicators are the excess viscosity and the first normal stress. We calculated numerically $\Delta\eta$ and ΔN_1 using their definitions (10) and (11), respectively. The results are shown in Figure 3 and Figure 4. The excess viscosity reaches its maximum at the onset of the scaling when the domains are expected to be maximally stretched in the flow direction and the structure of $C(\mathbf{k}, t)$ is the one shown at $\gamma t = 4$ in Figure 1. According to our analysis of the scaling behavior, we expect the excess viscosity to scale with γ as $\Delta\eta \sim \gamma^{-(1+\alpha)/(2+\alpha)}$ for fixed value of γt. In the inset of Figure 3 we report for the case $\alpha = 1$ the dependance of $\Delta\eta_M$ on γ. We find an

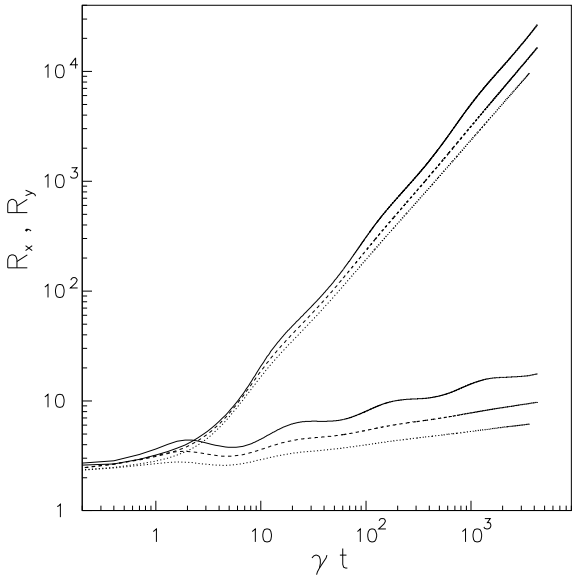

Fig. 2. The typical size of domains as function of the strain γt in the flow and in the shear directions for different values of α: $\alpha = 0$ (full line), $\alpha = 1$ (dashed line), $\alpha = 2$ (dotted line).

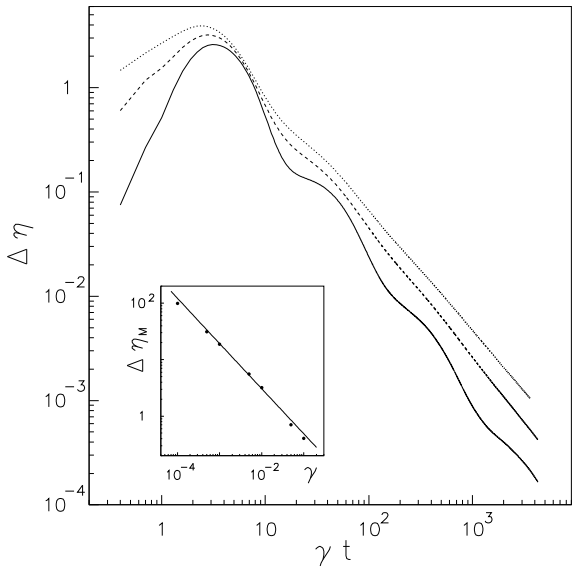

Fig. 3. Plots of the excess viscosity *vs.* the strain γt for different values of α: $\alpha = 0$ (full line), $\alpha = 1$ (dashed line), $\alpha = 2$ (dotted line). The inset shows the maxima of $\Delta\eta$ as function of γ for the case $\alpha = 1$. The slope of the straight line is 0.8.

exponent 0.8 slightly larger than the expected 2/3. The reason can be related to the fact that the excess viscosity reaches the maximum before the asymptotic regime is fully realized. Then $\Delta\eta$ decreases as a consequence of the dissipation of the elastic energy stored by domains which start to burst when they are stretched furtherly. Therefore more isotropic patterns form and the typical structure of the function $C(\mathbf{k}, t)$ is the one at $\gamma t = 9$ in Figure 1. The excess viscosity decreases with a power law behavior which is consistent with the predicted exponent $-(3+\alpha)/(2+\alpha)$. The first normal stress reported in Figure 4 decreases in

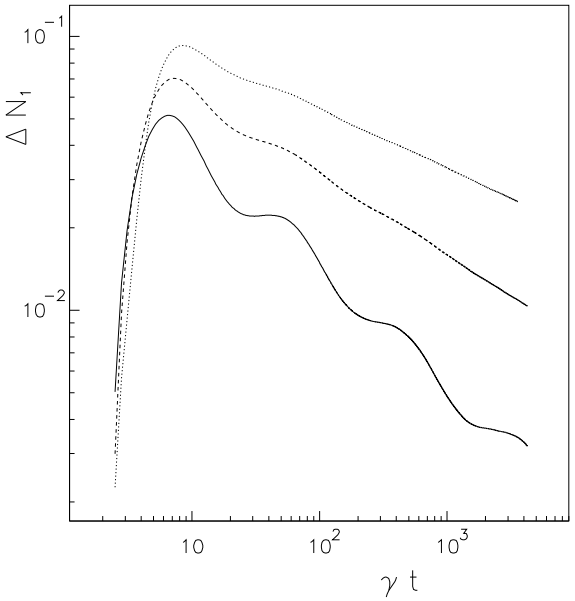

Fig. 4. The first normal stress as function of the strain γt for different values of α: $\alpha = 0$ (full line), $\alpha = 1$ (dashed line), $\alpha = 2$ (dotted line).

time according to the exponent $-1/(2+\alpha)$ after reaching a maximum. The amplitudes of all quantites plotted in Figures 2, 3 and 4 are modulated by damped log-time oscillations [23]. We believe that the physical explanation for the damping of oscillations may be found in the vanishing value of the mobility at equilibrium. In the case with constant mobility the origin of the oscillations is related to a cyclical mechanism of elongation and bursting of domains, which allows to store and dissipate elastic energy in the system [17]. In the present case, during the time evolution, the decreasing values of $\Gamma(\phi)$ suppress diffusion in the bulk phase and inhibit the growth of small bubbles coming from bursting. Therefore, they cannot be stretched too much by the flow. In this way it is more difficult to store elastic energy in the system and the excess viscosity can increase only by a small amount. This mechanism of growth inihibition becomes stronger and stronger in the course of evolution causing the observed damping of oscillations.

In conclusion, we have studied the phase separation of a binary mixture with field-dependent mobility in shear flow. We proved that dynamical scaling holds for this system. There are different growth exponents in the flow and in the shear directions which depend on the mobility exponent α. The difference in growth exponents is always 1. All the physical observables have amplitudes decorated by damped oscillations which are periodic in logarithmic time. We made a guess about the origin of this behavior. It would be an important endeavour to study this system in direct simulation of equation (4) to deeply understand this phenomenon.

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