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Critical temperatures in driven binary mixtures with conserved and non-conserved dynamics

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Abstract. We consider the critical behaviour of binary mixtures described by the φ^4 Landau–Ginzburg free energy and subject to an external shear flow. The critical temperature is calculated as a function of the shear rate γ in the limit of an infinite number of components of the field. The two cases with conserved and non-conserved order parameter are analysed. The values of the critical temperature depend on the dynamics considered. In particular, at small γ, the critical temperature grows as $\gamma^{1/4}$ in the conserved case and as $\gamma^{1/2}$ in the non-conserved case.

1. Introduction

There are many physically relevant cases of statistical systems where an external driving field is also the source of a flux of energy through the system in stationary conditions. In systems where an order–disorder transition can occur, a natural question to ask is how the critical temperature depends on the strength of the driving field [1]. This question assumes a particular experimental relevance for the case of a binary mixture subject to an external shear flow [2]. The knowledge of generalized phase diagrams in the two-parameter space of temperature and shear rate is important for the study of the rheological properties of fluid mixtures [3].

Many experiments and theories have shown that the critical properties of a binary mixture are strongly affected by a shear flow [4]. If hydrodynamic fluctuations are neglected, it is expected that the critical fluctuations are suppressed by the shear flow. This implies the raising of the critical temperature T_c together with a mean-field behaviour of the critical exponents. This problem has been studied by Monte Carlo simulations [5], but it was not possible to extract from simulations the quantitative behaviour of T_c due to numerical uncertainties and the relevance of finite-size effects [6].

In this paper we analyse the effects of the shear on the critical temperature of a binary mixture described in thermodynamic equilibrium by the usual φ^4 Landau–Ginzburg free energy. We study the critical behaviour of this system in the limit of an infinite number of components of φ. This approximation, which allows us to perform explicit calculations is often used in equilibrium statistical mechanics to calculate critical properties [7], and has also been proved to be very useful in dynamical problems such as phase separation with applied flows [8].

In the case of a binary fluid mixture the order parameter, which is the total difference of concentrations between the two components of the mixture, is conserved during the evolution of the system. In this paper we consider both the cases with conserved and not conserved order parameters. The second case is relevant for the behaviour of liquid crystals in a shear flow [9]. We will show that the conservation law affects the behaviour of the critical temperature which depends on the particular dynamics considered. We calculate this behaviour explicitly at small values of the shear rate.

The paper is organized as follows. In the next section we describe our model and write the equations for the critical temperature in the conserved and non-conserved cases. In section 3 we solve these equations for the binary fluid and in section 4 for the non-conserved case. A discussion with conclusions will follow.

2. The model and the equations for the critical temperature

We consider a system in $d = 3$ spatial dimensions described at equilibrium by the Ginzburg–Landau free-energy functional

$$\mathcal{F}\{\varphi\} = \int \mathrm{d}^d r \left\{ \frac{a}{2}\varphi^2 + \frac{b}{4}\varphi^4 + \frac{1}{2}(\vec{\nabla}\varphi)^2 \right\} \tag{1}$$

where φ is the order parameter field which represents the concentration difference between the two components of the mixture. The parameter b is always positive, while the value of a distinguishes between the ordered and the high-temperature disordered state.

The dynamics of the model is governed by the convection–diffusion equation [4]

$$\frac{\partial\varphi}{\partial t} + \vec{\nabla}\cdot(\varphi\vec{v}) = -\Gamma\frac{\delta\mathcal{F}}{\delta\varphi} + \eta \tag{2}$$

where Γ is the kinetic coefficient and η is a Gaussian white noise with expectations

$$\begin{aligned} &\langle\eta(\vec{r},t)\rangle = 0 \\ &\langle\eta(\vec{r},t)\eta(\vec{r}',t')\rangle = 2T\Gamma(\vec{r})\delta(\vec{r}-\vec{r}')\delta(t-t'). \end{aligned} \tag{3}$$

We study both the cases with conservation of the order parameter,

$$\Gamma(\vec{r}) = -\Gamma\nabla^2 \qquad \text{(COP)} \tag{4}$$

and without conservation of the order parameter,

$$\Gamma(\vec{r}) = \Gamma \qquad \text{(NCOP)}. \tag{5}$$

The imposed velocity field is of the form

$$\vec{v} = \gamma y \vec{e}_x \tag{6}$$

where γ is the spatially homogeneous shear rate and $\vec{e}_x$ is a unit vector in the flow direction.

The presence of the cubic term in the derivative $\delta\mathcal{F}/\delta\varphi$ prevents an exact solution of equation (2), as in the case without shear [10]. However, a solvable model is recovered in the so-called $N \to \infty$ limit, which amounts to the factorization of the cubic term of equation (2) as

$$\varphi^3 \to \langle\varphi^2\rangle\varphi. \tag{7}$$

It is possible to show [7] that the substitution (7) becomes exact in models with a vectorial order parameter when the number N of the components of the field becomes infinite. Since $\langle\varphi^2\rangle = S(t)$ does not depend on space, due to translational invariance, the substitution (7) formally linearizes the theory.

A quantity of interest for the critical properties of the model, related to the inverse susceptibility, is the structure factor

$$C(\vec{k}, t) = \langle \varphi(\vec{k}, t)\varphi(-\vec{k}, t)\rangle \tag{8}$$

where $\varphi(\vec{k}, t)$ is the Fourier transform of $\varphi(\vec{r}, t)$. It is straightforward to show that in the large-N approximation $C(\vec{k}, t)$ satisfies the equation [8]

$$\frac{\partial C(\vec{k}, t)}{\partial t} - \gamma k_x \frac{\partial C(\vec{k}, t)}{\partial k_y} = -2\Gamma(\vec{k})[k^2 + S(t) - 1]C(\vec{k}, t) + 2\Gamma(\vec{k})T \tag{9}$$

where

$$\Gamma(\vec{k}) = \Gamma \qquad \text{(NCOP)} \tag{10}$$

$$\Gamma(\vec{k}) = \Gamma k^2 \qquad \text{(COP)} \tag{11}$$

and the function $S(t)$ is given self-consistently by

$$S(t) = \int_{|\vec{k}|<q} \frac{\mathrm{d}\vec{k}}{(2\pi)^d} C(\vec{k}, t) \tag{12}$$

with q being a high-momentum phenomenological cut-off. For simplicity we have put in equation (9) $b = 1, a = -1$ and for the following we set $\Gamma = \frac{1}{2}$. This corresponds to a redefinition of the time, space and field scales.

In order to study the critical behaviour we consider the stationary limit where the structure factor is time independent. The solution of equation (9), obtained by applying the methods of characteristics, reads as

$$\begin{aligned}\mathcal{C}(\vec{k}) = \lim_{t\to\infty}\bigg[&C_0 \exp\bigg(-\int_0^t \mathcal{K}^p(z)[\mathcal{K}^2(z) + \mathcal{S} - 1]\,\mathrm{d}z\bigg) \\ &+T\int_0^t \mathcal{K}^p(z)\exp\bigg(-\int_0^z \mathcal{K}^p(s)[\mathcal{K}^2(s) + \mathcal{S} - 1]\,\mathrm{d}s\bigg)\mathrm{d}z\bigg]\end{aligned} \tag{13}$$

where C_0 is a constant value for the structure factor at the initial time, $p = 0, 2$ for the NCOP and the COP case, respectively,

$$\mathcal{S} = \int_{|\vec{k}|<q} \frac{\mathrm{d}\vec{k}}{(2\pi)^d}\mathcal{C}(\vec{k}) \tag{14}$$

and

$$\mathcal{K}(s) = \vec{k} + \gamma k_x \vec{e}_y s. \tag{15}$$

When $\mathcal{S} > 1$ only the second integral of the above expression survives in the limit $t \to \infty$. The self-consistent relation

$$\mathcal{S} = \int_{|\vec{k}|<q} \frac{\mathrm{d}\vec{k}}{(2\pi)^d} T \int_0^\infty \mathcal{K}^p(z)\exp\left[-\int_0^z \mathcal{K}^p(s)[\mathcal{K}^2(s) + \mathcal{S} - 1]\,\mathrm{d}s\right]\mathrm{d}z \tag{16}$$

is analogous to the state equation of the ϕ^4-model in the disordered phase in the $N \to \infty$ limit [10]. Also here, in the limit $\mathcal{S} \to 1^+$ the integral becomes infrared divergent for $d \leqslant d_c^{inf} = 2$, while it tends to a finite value for $d \geqslant 3$. For $d \geqslant 3$, the value $\mathcal{S} = 1$ defines the critical point of the model with the critical temperature fixed by the relation

$$\frac{1}{T_c(\gamma)} = \int_{|\vec{k}|<q} \frac{\mathrm{d}\vec{k}}{(2\pi)^d} \int_0^\infty \mathcal{K}^p(z)\exp\left[-\int_0^z \mathcal{K}^p(s)\mathcal{K}^2(s)\,\mathrm{d}s\right]\mathrm{d}z. \tag{17}$$

As in the ϵ-expansion calculations of [4], the critical point of equation (17) is where the inverse susceptibility $1/C(\vec{k})$ with $k_x = 0$ vanishes in the limit $k \to 0$ (see equation (9)).

We conclude this section with some considerations on how to fix the ultraviolet cut-off q. These considerations are pertinent because one expects that the critical temperature, as it occurs in the $N \to \infty$ limit in the case without shear [10], depends on q. Indeed, if we put $\gamma = 0$ in equation (17), then $\vec{\mathcal{K}}(s) = \vec{k}$ and the integral gives

$$T_c(0) = \frac{2\pi^2}{q}. \tag{18}$$

We impose that the shear is not effective at the molecular scales fixed by the cut-off. This assumption can be made quantitative by the following observations. From many experiments and phenomenological considerations it results that the effects of shear become relevant when $\gamma t > 1$ [2]. A shear time-scale is fixed as $\tau_s = 1/\gamma$. In stationary problems, in the disordered phase, only the fluctuations which decay in a time larger than τ_s will be affected by the shear. The relaxation time τ of a fluctuation depends on its wavevector k and a critical value of k can be defined by the relation $\tau(k_c) = \tau_s$ [4]. Only the fluctuations with $k < k_c$ will be distorted by the shear flow. For the model defined by equation (2) it is possible to show that $k_c \sim \gamma^{1/(2+p)}$ [4], so that, in fixing the cut-off, the shear will be properly taken into account if

$$q > k_c \qquad \text{or} \qquad q\gamma^{-\frac{1}{2+p}} > 1. \tag{19}$$

3. The case with conserved dynamics

In this section we calculate the critical temperature of a binary mixture in the COP case with $p = 2$. We define the function

$$g(\gamma, q) = \int_{|\vec{k}|<q} \frac{d\vec{k}}{(2\pi)^d} \int_0^\infty \mathcal{K}^2(z) \exp\left[-\int_0^z \mathcal{K}^4(s)\, ds\right] dz \tag{20}$$

with $\vec{\mathcal{K}}(z)$ given by equation (15) and $\mathcal{K}^4 \equiv (\mathcal{K}^2)^2$. The integral in the argument of the exponential can be performed and the function $g(\gamma, q)$ can be written using spherical coordinates as

$$\begin{aligned} g(\gamma, q) &= \int_0^q \frac{k^4}{(2\pi)^d}\, dk \int d\Omega \int_0^\infty dz\, [1 + \alpha\gamma z + \beta\gamma^2 z^2] e^{-zk^4 f(\gamma z)} \\ &= \gamma^{1/4} \int_0^{\bar{q}} \frac{k^4}{(2\pi)^d}\, dk \int d\Omega \int_0^\infty dy\, [1 + \alpha y + \beta y^2] e^{-k^4 h(y)} \end{aligned} \tag{21}$$

with $d\Omega = d\phi \sin\theta\, d\theta$, $\bar{q} = q/\gamma^{1/4}$, $h(y) = yf(y)$,

$$f(y) = 1 + \alpha y + (\alpha^2 + 2\beta)\frac{y^2}{3} + \alpha\beta\frac{y^3}{2} + \beta^2\frac{y^4}{5} \tag{22}$$

and

$$\alpha = \sin^2\theta \sin 2\phi \qquad \beta = \sin^2\theta \sin^2\phi. \tag{23}$$

By changing the order of integrations and using equations (17) and (18) we obtain the result

$$\frac{T_c(0)}{T_c(\gamma)} = \frac{g(\gamma, q)}{g(0, q)} \equiv \tilde{g}(\bar{q}) = \frac{1}{16\pi\bar{q}} \int d\Omega \int_0^\infty dy\, [1 + \alpha y + \beta y^2] \frac{\gamma\left(\frac{5}{4}, \bar{q}^4 h(y)\right)}{h^{5/4}(y)} \tag{24}$$

where $\gamma(\alpha, x) = \int_0^x \mathrm{d}t\, \mathrm{e}^{-t} t^{\alpha-1}$ is the incomplete γ-function. It is also useful to introduce the analytical function $\gamma^*(\alpha, x) = x^{-\alpha}\gamma(\alpha, x)/\Gamma(\alpha)$ in terms of which $\tilde{g}(\bar{q})$ can be rewritten as

$$\tilde{g}(\bar{q}) = \frac{\Gamma\left(\frac{5}{4}\right)}{16\pi}\bar{q}^4 \int \mathrm{d}\Omega \int_0^\infty \mathrm{d}z\, (1 + \alpha z + \beta z^2)\gamma^*\left(\frac{5}{4}, \bar{q}^4 z f(z)\right). \tag{25}$$

The limit $\bar{q} \to \infty$. In order to evaluate this limit we write

$$\tilde{g}(\bar{q}) = I_1(\bar{q}) + I_2(\bar{q}) \tag{26}$$

where

$$I_1(\bar{q}) = \frac{\Gamma\left(\frac{5}{4}\right)}{16\pi} \int \mathrm{d}\Omega \int_0^{\bar{q}^4} \mathrm{d}z \left(1 + \alpha\frac{z}{\bar{q}^4} + \beta\left(\frac{z}{\bar{q}^4}^2\right)\gamma^*\left(\frac{5}{4}, zf\left(\frac{z}{\bar{q}^4}\right)\right)\right) \tag{27}$$

and

$$I_2(\bar{q}) = \frac{\Gamma\left(\frac{5}{4}\right)}{16\pi}\bar{q}^4 \int \mathrm{d}\Omega \int_1^\infty \mathrm{d}z\, (1 + \alpha z + \beta z^2)\gamma^*\left(\frac{5}{4}, \bar{q}^4 z f(z)\right). \tag{28}$$

We first consider the behaviour of the integral $I_2(\bar{q})$. In the limit $x \to \infty$ $\gamma^*(\alpha, x) \sim x^{-\alpha} + \mathrm{O}\left(\frac{\mathrm{e}^{-x}}{x}\right)$ [11] so that

$$I_2(\bar{q}) = \frac{\Gamma\left(\frac{5}{4}\right)}{16\pi\bar{q}} \int \mathrm{d}\Omega \int_1^\infty \mathrm{d}z\, \frac{1 + \alpha z + \beta z^2}{z^{5/4} f(z)^{5/4}} + \mathrm{O}\left(\frac{\mathrm{e}^{-q^4}}{q^4}\right). \tag{29}$$

The above integral can be calculated numerically and the result is

$$I_2(\bar{q}) \sim \frac{0.347\,46}{\bar{q}}. \tag{30}$$

The evaluation of I_1 is more elaborate. The function $\gamma^*\left(\frac{5}{4}, zf(z/\bar{q}^4)\right)$ can be written at large $\bar{q}$ as a Taylor expansion. Each term of this expansion contributes with terms proportional to $\gamma^*\left(\frac{5}{4}, z\right)(z/\bar{q})^n$ and terms like $\mathrm{e}^{-z} b^{2n-1}(z)/\bar{q}^n$, where $b^s(z)$ is a polynomial of degree s in z. Therefore, for large $\bar{q}$ we can write

$$I_1(\bar{q}) = \int \mathrm{d}\Omega \int_0^{\bar{q}^4} \sum_{k=0}^{\infty} \left(\left(\frac{z}{\bar{q}^4}\right)^{2k} \mathcal{P}_{2k-1}(z, \theta, \phi)\, \mathrm{e}^{-z} + \mathcal{C}_k(\theta, \phi)\gamma^*\left(\frac{5}{4}, z\right)\left(\frac{z}{\bar{q}^4}\right)^{2k}\right) \mathrm{d}z \tag{31}$$

where $\mathcal{P}_{2k-1}(z, \theta, \phi)$ is a polynomial of degree $2k-1$ in z and both $\mathcal{P}_{2k-1}(z, \theta, \phi)$ and $\mathcal{C}_k(\theta, \phi)$ contain polynomials of α and β. In equation (31) only even powers appear since the other terms give a zero contribution after the integration over the solid angle. The integration in $\mathrm{d}\Omega$ gives

$$I_1(\bar{q}) = \sum_{k=0}^{\infty} \int_0^{\bar{q}^4} \sum_{k=0}^{\infty} \left(\left(\frac{z}{\bar{q}^4}\right)^{2k} p_{2k-1}(z)\, \mathrm{e}^{-z} + c_k\gamma^*\left(\frac{5}{4}, z\right)\left(\frac{z}{\bar{q}^4}\right)^{2k}\right) \mathrm{d}z \tag{32}$$

where now the p_{2k-1} are polynomials depending only on z and the c_k are real numbers. Then, using the results

$$\int_0^{\bar{q}^4} \mathrm{e}^{-z} z^n\, \mathrm{d}z = n! + \mathrm{O}(\bar{q}^{4n}\mathrm{e}^{-\bar{q}}) \tag{33}$$

and

$$\int_0^{\bar{q}^4} \left(\frac{z}{\bar{q}^4}\right)^{2k} \gamma^*\left(\frac{5}{4}, z\right) \mathrm{d}z = \frac{1}{\bar{q}}\frac{4}{8k-1} - \frac{1}{\bar{q}^{8k}}\frac{4(2k)!}{\Gamma\left(\frac{5}{4}\right)(8k-1)} + \mathrm{O}(\bar{q}^{8k}\mathrm{e}^{-\bar{q}^4}) \tag{34}$$

it turns out that

$$I_1(\bar{q}) = \frac{A}{\bar{q}} + \sum_{k=0}^{\infty} \frac{B_k}{\bar{q}^{8k}} + \mathrm{O}(\bar{q}^{8k}\mathrm{e}^{-\bar{q}^4}). \tag{35}$$

The coefficients B_k take contributions from the second term of the right-hand side of equation (34) and from the terms proportional to p_{2k-1} in equation (32). We have calculated the first three coefficients with the result

$$B_0 = 1 \qquad B_1 = -0.057\,143 \qquad B_2 = -0.391\,11. \tag{36}$$

The constant A is given by

$$A = \sum_{k=0}^{\infty} \frac{4c_k}{8k-1} \sim -0.908\,35 \tag{37}$$

where contributions up to $k = 5$ have been considered and the remainder of the series has been evaluated as less than 10^{-6}.

Putting together equations (24), (26), (35)–(37) we obtain

$$\tilde{g}(\bar{q}) \to 1 - \frac{0.560\,89}{\bar{q}} - \frac{0.057\,14}{\bar{q}^8} - \frac{0.391\,11}{\bar{q}^{16}} + \mathrm{O}(1/\bar{q}^{24}). \tag{38}$$

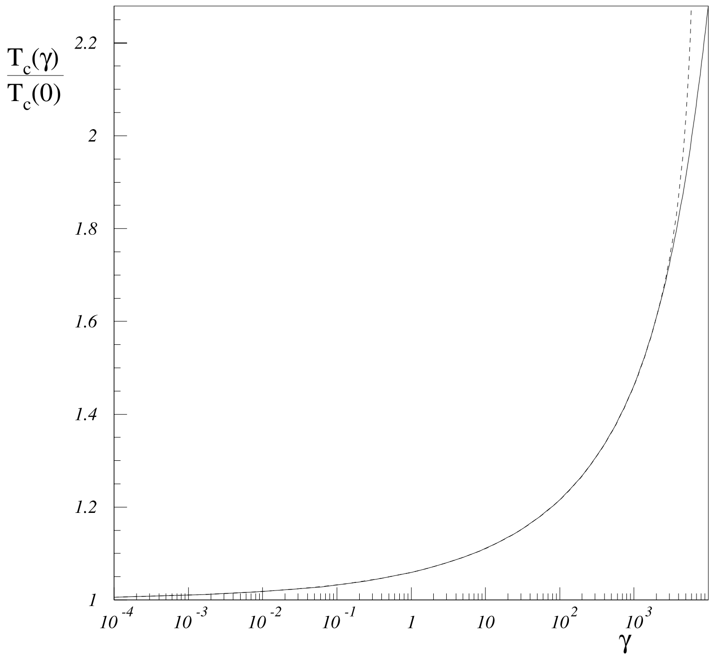

Figure 1. The critical temperature as a function of the shear rate γ in the COP case. The broken curve corresponds to the result of equation (38), while the full curve is a numerical evaluation of the integral of equation (17).

The critical temperature. From equation (38) it results that the critical temperature behaves at small γ as $(T_c(\gamma) - T_c(0))/T_c(0) \sim \gamma^{1/4}$. When the other terms of equation (38) are considered, one obtains the critical curve plotted in figure 1. In figure 1 we also show the critical temperature (full curve) obtained by the numerical evaluation of the integral (17). There is a good agreement between the analytical and the numerical results in the range considered with $\bar{q} > 1$. The situation would be different for $\bar{q} < 1$ where we have checked that our analytical results are very poor in describing the behaviour of the integral (17).

4. The NCOP case

When the order parameter is not conserved the critical temperature is given by equation (17) with $p = 0$. Repeating the same steps of the above section we arrive at the equation

$$\frac{T_c(0)}{T_c(\gamma)} = \tilde{c}(\tilde{q}) = \frac{\Gamma\left(\frac{3}{2}\right)}{8\pi}\tilde{q}^2 \int \mathrm{d}\Omega \int_0^\infty \mathrm{d}y\, \gamma^*\left(\frac{3}{2}, \tilde{q}^2 y d(y)\right) \tag{39}$$

where

$$d(y) = 1 + \tfrac{1}{2}\alpha y + \tfrac{1}{3}\beta y^2 \tag{40}$$

and $\tilde{q} = q\gamma^{-1/2}$. Also in this case we can evaluate the expression $\tilde{c}(\tilde{q})$ in the limit $\tilde{q} \to \infty$. As before the function $\tilde{c}(\tilde{q})$ can be written as

$$\tilde{c}(\tilde{q}) = J_1(\tilde{q}) + J_2(\tilde{q}) \tag{41}$$

where

$$J_1(\tilde{q}) = \frac{\Gamma\left(\frac{3}{2}\right)}{8\pi}\tilde{q}^2 \int \mathrm{d}\Omega \int_0^1 \mathrm{d}y\, \gamma^*\left(\frac{3}{2}, \tilde{q}^2 y d(y)\right) \tag{42}$$

and

$$J_2(\tilde{q}) = \frac{\Gamma\left(\frac{3}{2}\right)}{8\pi}\tilde{q}^2 \int \mathrm{d}\Omega \int_1^\infty \mathrm{d}y\, \gamma^*\left(\frac{3}{2}, \tilde{q}^2 y d(y)\right). \tag{43}$$

The integral J_1 and J_2 can be calculated with a procedure similar to that applied in the previous section. The result is

$$J_2(\tilde{q}) = \frac{0.494\,81}{\tilde{q}} + \mathrm{O}\left(\frac{\mathrm{e}^{-\tilde{q}^2}}{\tilde{q}^2}\right) \tag{44}$$

$$J_1(\tilde{q}) = \frac{D}{\tilde{q}} + \sum_{k=0}^{\infty} \frac{E_k}{\tilde{q}^{4k}} + \mathrm{O}(\tilde{q}^{4k}\mathrm{e}^{-\tilde{q}^2}). \tag{45}$$

with $D = -0.898\,22$, $E_0 = 1$, $E_1 = -0.044\,44$, $E_2 = 0$ hence

$$\tilde{c}(\tilde{q}) \to 1 - \frac{0.403\,41}{\tilde{q}} - \frac{0.044\,44}{\tilde{q}^4} + \mathrm{O}(1/\tilde{q}^{12}). \tag{46}$$

The above equation implies that at small γ the critical temperature behaves as $(T_c(\gamma) - T_c(0))/T_c(0) \sim \sqrt{\gamma}$. The full behaviour described by equation (46) is plotted in figure 2 together with the numerical evaluation of the integral.

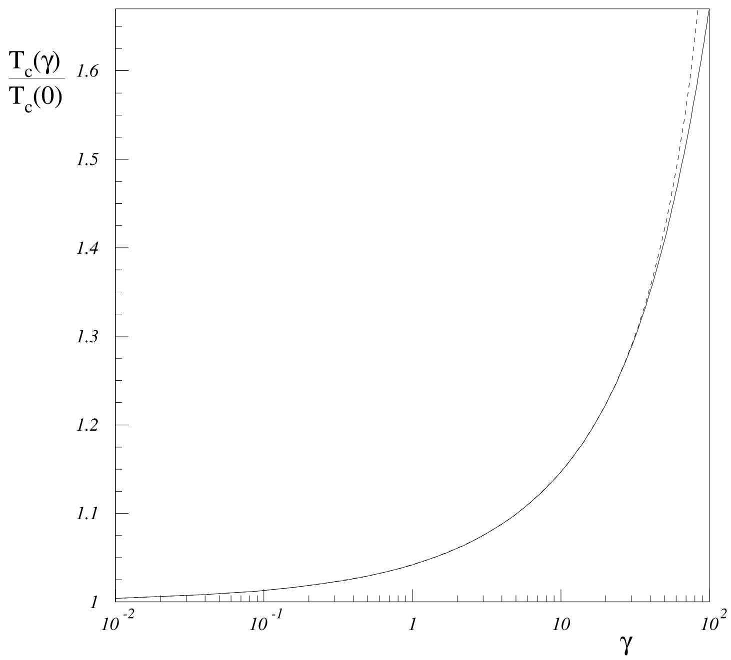

Figure 2. The critical temperature in the NCOP case. The broken and full curve describe analytical and numerical results as in figure 1.

5. Discussion and conclusions

In this paper we have studied the behaviour of the critical temperature of a binary mixture subject to a shear flow. In both cases with conserved and non-conserved order parameter the critical temperature is found to grow with the shear rate. A simple analysis, not reported here, shows that the critical exponents remain the same as in the ϕ^4-model without flow in the $N \to \infty$ limit. The case with the conserved order parameter describes the behaviour of a fluid mixture. Equation (38) shows that $(T_c(\gamma) - T_c(0))/T_c(0) \sim \gamma^{1/4}$ at small γ. ϵ-expansion results of [4] show that $(T_c(\gamma) - T_c(0))/T_c(0) \sim \gamma^{0.54}$. The different exponent is due to the use of different approximations. In fluid mixtures one should also consider the effects of velocity fluctuations not taken into account in this paper. The concentration fluctuations are enhanced by the coupling with the velocity fluctuations and this produces a lowering of T_c which competes with the effect described in this paper. The net change of T_c will depend on the balance between the two effects and varies with the particular system considered.

When the order parameter is not conserved we find that $(T_c(\gamma) - T_c(0))/T_c(0) \sim \gamma^{1/2}$. We do not know of previous calculations for this case. Moreover, our results show with an explicit example that the critical temperature of a driven system depends on the dynamics considered. In particular, the suppression of thermal fluctuations at small γ is more enhanced in the case with conserved dynamics. It would be interesting to check these observations outside the framework of the $N \to \infty$ approximation. Finally, we mention that an increase of the critical temperature is also observed in Ising driven models studied in other contexts [1]. The fact that in our model we do not observe a saturation of the value of the critical temperature at large values of the imposed field is probably due to the particular approximation used.

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