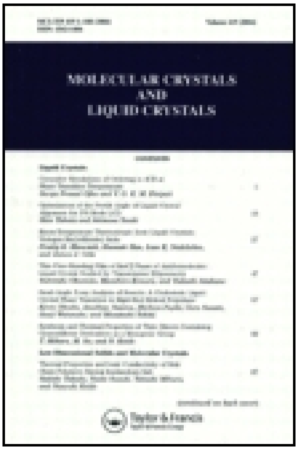

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EFFECT OF SHEAR FLOWS ON CRITICAL FLUCTUATIONS IN FLUIDS

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A shear flow distorts the equal time correlation functions $\langle|m_k|^2\rangle$ for a fluid system with order parameter m, just above a critical point. We analyse this effect here for the case where m is <u>not</u> a conserved quantity : physical examples may be found with the transition smectic C↔ smectic A or possibly smectic A↔ nematic. We find an important, anisotropic, reduction of fluctuations of wave vector k where $k\xi \gg 1$ provided that $s\tau > 1$ (where ξ is the correlation length and τ the relaxation time of the uniform mode).

Critical fluctuations have low relaxation rates. Thus, in liquid systems it may be possible to achieve velocity gradients s which are comparable to these rates. Then, as first suggested by Bergé[1], the fluctuations may be significantly reduced by shear. For instance near a demixtion point the fluctuations of wave length $2\pi/k$ comparable to the correlation length ξ show relaxation frequencies of order[2]

$$1/\theta_r = k_B T/6\pi\eta\xi^3$$

where T is the temperature, η the viscosity, and k_B is Boltzman's constant. For $\eta = 0.1$ poise and $\xi = 1000$ Å $\frac{1}{\theta_r} \sim 25 \text{sec}^{-1}$ at room temperature : shear rates of this magnitude are easily achieved in Couette flow.

We have attempted a simple analysis of these effects : as usual in dynamical critical phenomena[3], the essential feature is the existence (or absence) of a conservation law for the order parameter $m(\underset{\sim}{r},t)$. For the most usual critical points in a fluid phase (e.g. demixtion, or liquid gas, transitions), $\int m \, d\underset{\sim}{r}$ is a constant of the motion. But some more exotic transitions, such as smectic C to smectic A or smectic A to nematic[5], there is no conservation law. It turns out that the analysis is considerably simpler for the latter case : this is the only one to be discussed in this note.

Consider for instance an A↔C transition with smectic layers fixed in orientation and parallel to the xy plane. The order parameter has two components ψ_x and ψ_y describing two modes of tilt for the molecules. Above the critical point we assume that ψ is ruled by a linear time dependent, Landau Ginzburg equation. Consider for instance a simple shear flow parallel to the layers

$$v_x = sz \quad v_y = v_z = 0$$

and write the equation motion for $\psi_y \equiv m(\underset{\sim}{r},t)$ in the form[6]

$$\frac{\partial m}{\partial t} + \frac{m}{\tau} - D\nabla^2 m + \underset{\sim}{v}\cdot\nabla m = \phi \qquad (1)$$

Here τ is the relaxation time of the uniform mode, and $D = \xi^2/\tau$. The third term describes convection, and ϕ is a random source, with correlations fixed by the usual rules[7]

$$\langle Q(\underset{\sim}{r},t_1)\phi(r_2 t_2)\rangle = 2k_B T_c \chi/_\tau \ \delta(\underset{\sim}{r}_{12})\delta(t_{12}) \qquad (2)$$

where χ is the susceptibility associated with m; both χ and τ diverge near T_c. In our Van Hove approximation[8] χ and τ have the same divergence. More generally, eqs(1,2) may be retained for small wave vectors ($k\xi < 1$), even if the critical exponents of χ and τ are non classical. In the following we shall use eq(1) for all wave vectors: this is essentially equivalent to a neglect of the small exponent in both static and dynamic properties[9]. Note that eq(2) remains valid in shear flow: because of the factor $\delta(t_1-t_2)$ all convections of ϕ have no effect.

Introducing Green's function $G(r_1 t_1\ r_2,t_2)$ for the operator acting on m in eq(1) we may write

$$m(r_1 t) = \int_{-\infty}^{t} dt_3 \int dr_3\ G(31)\phi(3)$$

and for the equal time correlation function, after insertion of eq(2), we get

$$\langle m(r_1 t)\ m(r_2 t)\rangle = 2k_B T_c \chi/_\tau \int_{-\infty}^{t} dt_3 \int dr_3\ G(31)G(32) \quad (3)$$

where (3) represents $(\underset{\sim}{r}_3,t_3)$ and $t_1 = t_2 = t$. The function $G(\underset{\sim}{r}_3\ 0\ |r_1\ \theta)$ represents the spreading of a point source from point $\underset{\sim}{r}_3$ during the time θ, under the effects of diffusion, capture (through the rate $1/\tau$) and convection. Translational invariance imposes the following structure for G:

$$G(\underset{\sim}{r}_3 0\ r_1\theta) = \kappa_\theta(\underset{\sim}{r}_1-\underset{\sim}{r}_3-\theta\underset{\sim}{v}(\underset{\sim}{r}_1))\varepsilon^{-\theta/\tau} = \kappa_\theta(\underset{\sim}{\rho})\varepsilon^{-\theta/\tau} \qquad (4)$$

$K_\theta(\rho)$ is the spreading function for a particle starting for the origin and moving through the distance ρ, with the specification that the velocity field vanishes at the origin. It can be shown that $K_\theta(\rho)$ is gaussian, and that it's moments obey the equations

$$\frac{d}{d\theta}\langle \rho_z^2 \rangle = \frac{d}{d\theta}\langle \rho_y^2 \rangle = 2D$$

$$\frac{d}{d\theta}\langle \rho_x \rho_z \rangle = s\langle \rho_z^2 \rangle$$

$$\frac{d}{d\theta}\langle \rho_x^2 \rangle = 2D + s\langle \rho_x \rho_z \rangle$$

Solving for the moments, and going to the Fourier transform $K_\theta(k)$ we get

$$f_\theta(k) \equiv -\ln K_\theta(k) = Dk^2\theta + Dk_x k_z s\theta^2 + \frac{1}{3} Dk_x^2 s^2 \theta^3 \qquad (5)$$

Returning now to eq(3) we find (after some manipulation) that all phase factors due to the terms like $\theta v(r_3)$ in eq (4) drop out, and we get simply

$$\langle |m_k|^2 \rangle \equiv \int dr_2 \langle m(r_1,t)\, m(r_2 t) \rangle$$
$$= 2k_B T_c \,\chi/\tau \int_0^\infty d\theta \exp -2\left[f_\theta(k) + \frac{\theta}{\tau} \right] \qquad (6)$$

Consider first a wave vector k normal to the flow lines ($k_x = 0$) : then $f_\theta \to Dk^2\theta$ and we recover the Ornstein-Zernike form

$$\langle |m_k|^2 \rangle = \frac{\chi k_B T_c}{1+\xi^2 k^2} \qquad (7)$$

On the other hand, when k is parallel to the flow lines

$$\langle |m_k|^2 \rangle - \chi k_B T_c \int_0^\infty 2du \exp -2\left[(1+\xi^2 k^2)u + \frac{k^2 \xi^2 u^3 s^2 \tau^2}{3} \right] \qquad (8)$$

When $s\tau$ is small we again recover the Ornstein-Zernike form. But when

$$s\tau > \frac{(1+\xi^2 k^2)^{3/2}}{\xi k} \qquad (9)$$

the last term in the bracket of eq(8) dominates and the correlation function is strongly reduced

$$\langle |m_k|^2 \rangle \to \text{const.}\, \chi k_B T_c \,(k\xi)^{-2/3} (s\tau)^{-2/3} \tag{10}$$

This unusual law could in principle be tested by light scattering experiments.

We must emphasize again that these results depend on a "Van Hove approximation" for the critical dynamics. This approximation should be not too bad for systems where m is not conserved and not coupled strongly to other conserved quantities[9]: this is the situation found in the C→A transition (where backflow effects do not alter the general structure of the modes in the ordered phase). On the other hand for the λ point of helium (and probably also for the A→N transition) the order parameter is strongly coupled to flow variables and our discussion is useless.

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3. B. Halperin, P. Hohenberg, Phys. Rev. 177, 952 (1969).

4. See for instance P.G. de Gennes, "The Physics of Liquid Crystals", Oxford (2nd printing) 1976.

5. The A → N transition is complicated by the coupling between direction fluctuations and the smectic order parameter: see B. Helperin, T. Lubensky, S. Ma, Phys. Rev. Lett. 32, 292 (1974).

6. The equation for the other component ψ_x contains one extra term, linear in s, expressing that a uniform shear along x induces a tilt $\overline{\psi}_x$ = const.s. Putting $m = \psi_x - \overline{\psi}_x$ we return to the form (1).

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