wave model. As an unmodulated beam current increases, the spontaneous emission becomes strong, but its bandwidth is still narrow ($\Delta\omega/\omega \sim 1.5\times10^{-2}$ at $I_b = 2.3$ mA). The amplitude of the emission increases exponentially and becomes saturated, where the corresponding $f_b(U)$ are similar with those in Fig. 2, though the energy spread is rather broad. Therefore, trapping of beam electrons is still dominant in this case.

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Nonlocal Shear Viscosity and Order-Parameter Dynamics near the Critical Point of Fluids*

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We derive and solve numerically the self-consistent set of equations for the nonlocal shear viscosity and the order-parameter decay rate near the critical point of fluids. This removes the ambiguity associated with the so-called "high frequency" shear viscosity entering the theoretical expressions for the Rayleigh linewidth, and improves the agreement with the measured linewidth of Chang *et al.*

Decay rates of order-parameter fluctuations near the critical point of fluids have been extensively investigated lately by the techniques of inelastic scattering of laser light,[1] and have been compared with the following mode-mode-coupling theoretical expression[2] for the decay rate Γ_{qs} with the wave vector $\vec{q}$:

$$\Gamma_{qs} = [k_B T/6\pi\eta^*\xi^3] K_0(q\xi), \tag{1}$$

where ξ is the range of correlation of order-parameter fluctuations and

$$K_0(x) = \tfrac{3}{4}[1 + x^2 + (x^3 - x^1)\tan^{-1}x]. \tag{2}$$

Here η^* is customarily referred to as "high frequency" shear viscosity and is temporarily identified with the shear viscosity $\eta(T)$ obtained by macroscopic measurements since theoretically $\eta(T)$ must remain finite at the critical point.[2] However, there are theoretical[3] and experimental[4] indications that the shear viscosity should

have at least a cusplike sharp peak at the critical point, and hence the value of η^* to be used in (1) remains somewhat ambiguous. The situation was critically examined recently by Chang *et al.*,[5] and the problem remains quite unsettled.

It is the purpose of the present communication to remove this ambiguity by considering the pair of equations which determine self-consistently both Γ_{qs} and the wave-number–dependent nonlocal shear viscosity η_q. Such equations can be obtained from the self-consistent set of equations for propagators $G_{qu}(t)$ and $G_{qs}(t)$ for transverse velocity u and order parameter s, respectively, which are normalized to reduce to unity for $t=0$. In the approximation where vertex corrections are ignored, they are given, for instance, by Eqs. (30) and (31) of Ref. 3, where the concentration c should be replaced by a more general order parameter s. In view of the fact that the transverse local velocity decays to zero infinitely faster than the order parameter at the critical point,[2,3] Eq. (31) can be approximated by the "Markovian" equation for $G_{qs}(t)$ with the result that $G_{qs}(t)=\exp(-t\Gamma_{qs})$, where the decay rate is given by

$$\Gamma_{qs}=\frac{k_BT}{(2\pi)^3}q^2\int d^3k\,\sin^2\theta\,\frac{\chi_{\vec{q}}\,\vec{q}\cdot\vec{k}}{\chi_{\vec{q}}k^2\eta_k}. \tag{3}$$

Here

$$\eta_q=(\rho/q^2)\Big[\int_0^\infty G_{qu}(t)\,dt\Big]^{-1} \tag{4}$$

defines the nonlocal (or q-dependent) shear viscosity, $\chi_{\vec{k}}=\langle|s_{\vec{k}}|^2\rangle$, and θ is the angle between $\vec{q}$ and $\vec{k}$. Here the "background" term, denoted as $q^2D_q{}^0$ in Ref. 3, has been ignored. This form of $G_{qs}(t)$ is substituted back into Eq. (30) of Ref. 3 to give the following expression for η_q:

$$\eta_q=\eta^0+\frac{k_BT}{2q^2}\frac{1}{(2\pi)^3}\int d^3k\,k^2\sin^2\theta\,\chi_{\vec{k}}\,\chi_{\vec{q}\cdot\vec{k}}\left(\frac{1}{\chi_{\vec{k}}}-\frac{1}{\chi_{\vec{q}\cdot\vec{k}}}\right)^2\left[\Gamma_{\vec{k}s}+\Gamma_{\vec{q}\cdot\vec{k}s}\right]^{-1}. \tag{5}$$

Equations (3) and (5) thus constitute the self-consistent set of equations for Γ_{qs} and η_q. Unfortunately, however, the integral in (5) diverges at large k, as noted in Ref. 3 and these equations cannot be immediately handled. However, if we choose to give up predicting the critical anomaly in the macroscopic shear viscosity

$$\eta(T)=\lim_{q\to 0}\eta_q,$$

but rather to focus our attention only on the nonlocality (or q dependence) of η_q, then this divergence difficulty disappears as one can readily verify by subtracting $\eta(T)$ from both sides of (5). Assuming the Ornstein-Zernike form for $\chi_k\sim(k^2+\xi^{-2})^{-1}$, we find by inspection of the resulting self-consistent equations that η_q and Γ_{qs} take the following forms:

$$\eta_q=\eta(T)[1-F(q\xi)], \tag{6}$$

$$\Gamma_{qs}=[k_BT/6\pi\eta(T)\xi^3]K(q\xi). \tag{7}$$

The functions F and K are then determined by the following integral equations:

$$K(x)=\frac{3}{4\pi^2}x^2(1+x^2)\int d^3y\,\sin^2\theta\,\frac{1}{y^2}\,\frac{1}{1+(\vec{x}-\vec{y})^2}\,\frac{1}{1-F(y)}, \tag{8}$$

$$F(x)=\frac{3}{8\pi^2x^2}\int d^3y\,\sin^2\theta\,\frac{y^2}{1+y^2}\left\{\frac{(2\vec{x}\cdot\vec{y})^2}{2K(y)}\,\frac{1}{1+y^2}-\frac{[x^2-2\vec{x}\cdot\vec{y}]^2}{K(y)+K(|\vec{x}-\vec{y}|)}\,\frac{1}{1+(\vec{x}-\vec{y})^2}\right\}, \tag{9}$$

where θ is the angle between the vectors $\vec{x}$ and $\vec{y}$. Note that the so-called high-frequency viscosity η^* is related to the macroscopic shear viscosity $\eta(T)$ by

$$\eta^*=[K_0(q\xi)/K(q\xi)]\eta(T) \tag{10}$$

The equations (8) and (9) have been solved numerically by iteration starting with $K=K_0$ and the results are shown in Figs. 1, 2, and 3. Note that the correction on Γ_{qs} arising from the nonlocality of η_q is fairly important, amounting to roughly 30% for $q\xi=20$, and remains finite ($\sim 5.5\%$) even in the hydrodynamic regime. The new linewidth function K is compared with the observed linewidth for the binary critical mixture 3-methylpentane-nitroethane along with the old linewidth function K_0 in Fig. 4 where very precise measurements of $\eta(T)$ are available.[6] Note that no adjustable parameter enters the theoretical curve and there is a clear improvement in the fit with data points. In the cases where $\eta(T)$ is not avail-

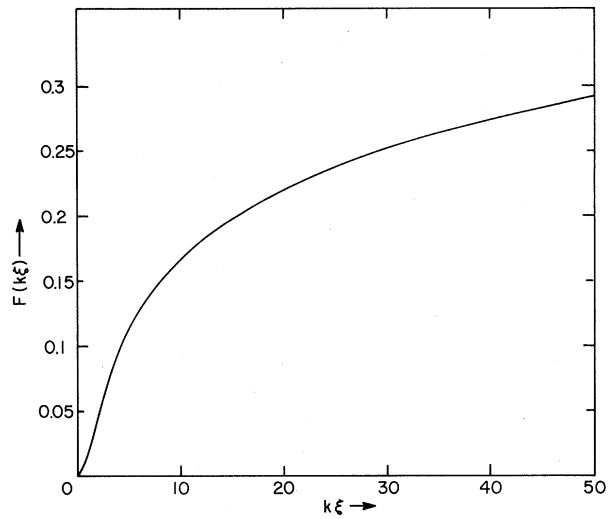

FIG. 1. The function F defined by Eq. (6).

able, the theoretical curve K can still be tested by measuring linewidths at different scattering angles but at the same temperature. It would be also quite interesting to test the form of η_q, Eq. (6), by inelastic light scattering by interfaces,[7] as well as by measuring the diffusion constant of fine particles dispersed in fluids.[8]

We conclude this communication with the following additional remarks: (i) The background contribution to Γ_{qs}, which has been ignored so far, on the one hand tends to enhance the theoretical linewidth; on the other hand it also acts to reduce the difference between $K(q\xi)$ and $K_0(q\xi)$. The net result presumably will further improve the agreement with experimental results. (ii) The mode-mode coupling that enters (5) is less well-founded than that entering (3) in the sense that three-, four-, ··· mode intermediate states cannot be ignored *a priori*. (iii) In the present work we have not included a possible frequency dependence of the shear viscosity and the order-parameter decay rate since a complete treatment of this effect would enormously complicate the problem. As indicated by the recent work of Perl and Ferrell on the same problem,[9] inclusion of this effect would tend to reduce further the difference between K and K_0, especially in the critical regime.

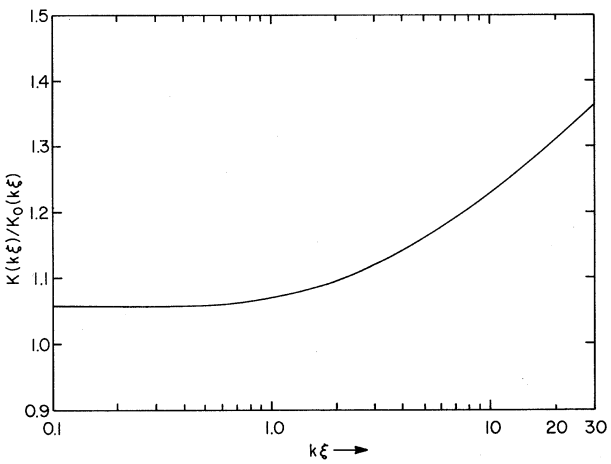

FIG. 3. Ratio of the new and old linewidth functions.

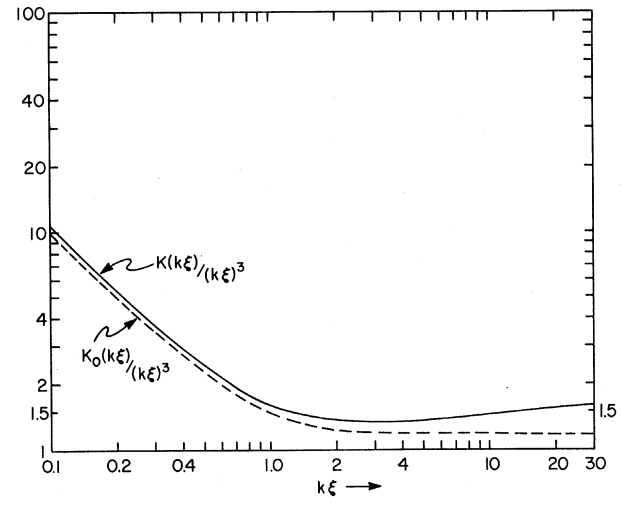

FIG. 2. New and old linewidth functions $K(k\xi)/(k\xi)^3$ and $K_0(k\xi)/(k\xi)^3$ defined by Eqs. (2) and (8), respectively.

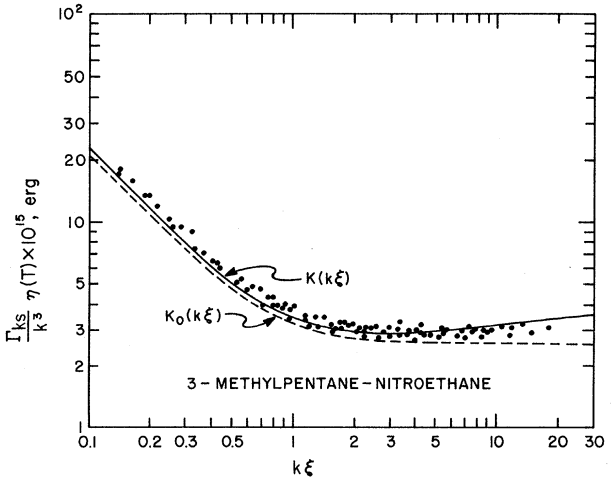

FIG. 4. Comparison of the linewidth functions with the data of Chang *et al.* (Ref. 5). The data points are taken from Fig. 11 of the second reference of Ref. 5, and the solid and dashed lines represent the new and old theoretical linewidth functions, respectively.

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Critical Viscosity and Diffusion in the Binary-Liquid Phase Transition*

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The fluctuation-dissipation formula for the viscosity in terms of the stress-tensor fluctuations reproduces the experimental logarithmic temperature dependence of the hydrodynamic viscosity. Using the theoretical wave-number– and frequency-dependent viscosity in the problem of critical diffusion, we find that the effects of nonlocality and retardation practically cancel, resulting in satisfactory agreement with a recent light-scattering determination of the effective viscosity.

Kawasaki[1] has shown that the critical variation of the diffusion coefficient in a binary liquid near its critical point can be represented by the Einstein relation $D = T\mu_\xi$, where T is the temperature (we use natural units such that Boltzmann's constant is unity), ξ is the correlation length for the concentration fluctuations, and $\mu_\xi = (6\pi\eta\xi)^{-1}$ is Stokes's formula for the mobility of a sphere of radius ξ moving through a liquid of viscosity η. This result has also been established by one of the present authors[2] by a different method, and gives the rate of relaxation of a concentration fluctuation of wave number q as

$$\Gamma_q = Dq^2 = Tq^2/6\pi\eta\xi, \tag{1}$$

provided $\xi \ll q^{-1}$; i.e., the wavelength should be much greater than the correlation length. But, as the critical point is approached, $T \to T_c$ and $\xi \to \infty$. The above inequality is then no longer satisfied and D becomes a function of q, corresponding to "nonlocal" diffusion. This change is carried out in Eq. (1) by substitution of an effective value for ξ^{-1} according to

$$(\xi^{-1})_{\text{eff}} = a_{\text{eff}} q, \tag{2}$$

where a_{eff} is some numerical constant of order of magnitude unity. Equation (2) expresses in a quantitative form the qualitative rule of dynamical scaling[3,4] that all temperature dependence is to be expressed in terms of ξ, and that as $T \to T_c$