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EXPERIMENTAL EVIDENCE OF THE "CRITICAL REGION" OF A BINARY MIXTURE BY MEANS OF INELASTIC SCATTERING OF LIGHT

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The "critical region" of a binary mixture is found to be characterized by the appearance of an asymptotic dependence such as $\Gamma=AK^3$.

As is well known, the formula giving the spectral linewidth Γ of the light scattered by a binary mixture in the hydrodynamic region is

$$\Gamma=DK^2(1+\xi^2K^2)^{1-\eta/2},$$

with D = mass diffusion coefficient, K = scattering vector, and ξ = correlation length.

This formula, however, is no longer valid when $\xi K \gtrsim 1$, as we have shown in a previous paper.[1] In order to obtain the relationship between Γ and K in this region, we have performed systematic measurements in the immediate vicinity of the critical temperature of a cyclohexane-aniline binary mixture. To avoid the difficulties arising from eventual thermal fluctuations of the thermostatic bath, the photocurrent due to the scattered light was recorded on an Ampex tape recorder, which demanded a relatively short time during which the temperature was stable to better than 10^{-3}°C. The rather long spectral analysis was performed at a later time.

Figure 1 shows the observed angular dependence of the spectral linewidth Γ, for different values of $T-T_c$. This angular dependence (for $T-T_c=6\times10^{-3}$°K, 5.5×10^{-3}°K, 4×10^{-3}°K, 3.5×10^{-3}°K, and 1.5×10^{-3}°K) is consistent with a law such as $\Gamma=AK^3$ (with $A=1.3\times10^{-13}$ cm^3 sec^{-1} and independent of $T-T_c$). This result is not surprising, because for all these values of $T-T_c$, $\xi K\gtrsim 1$, even for the smallest recorded value of the scattering angle $\theta=30°$.

At temperatures this close to T_c one might expect spurious effects due to multiple scattering and concentration gradients with height. However a separate experiment conducted on three cylindrical cells of different diameters (14, 10, and 5 mm) has shown that, in spite of variable amounts of multiple scattering (as shown by the observed beating signal-to-shot-noise ratios) the linewidth of the scattered spectrum does not depend on the cell diameter. This experiment shows that while multiple scattering strongly affects the intensity of the scattered light, it does not affect its linewidth. Thus the observed $\Gamma\sim K^3$ behavior cannot be ascribed to a spurious multiple-scattering effect.

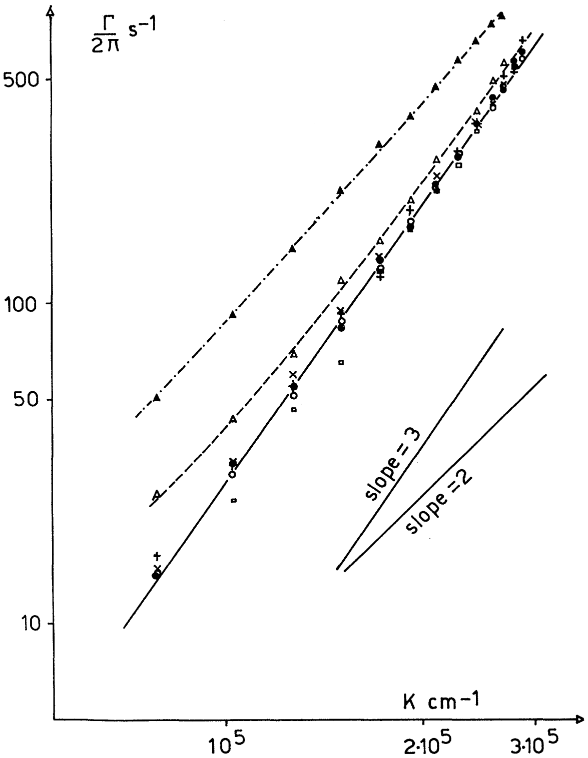

FIG. 1. Angular dependence of the spectral width Γ vs $T-T_c$. Closed triangles, 121×10^{-3}°K; open triangles, 12×10^{-3}°K; closed circles, 6×10^{-3}°K; crosses, 5.5×10^{-3}°K; plusses, 4×10^{-3}°K; open squares 3.5×10^{-3}°K; open circles, 1.5×10^{-3}°K.

One can also rule out a spurious effect due to a vertical concentration gradient in the cell. First of all this concentration gradient has been shown to be small.[2] Moreover, even if such a gradient were present, it would have as an effect the transformation of the stoichiometric mixture into a mixture locally nonstoichiometric. Separate experiments conducted on mixtures voluntarily nonstoichiometric have shown that the linewidth varies as $\sim K^2$ near T_c, with a very small Fixman correction,[3] and such behavior is absolutely different from the observed $\Gamma\sim K^3$ dependence.

Figure 1 also shows the data of an experiment performed at $T-T_c = 12\times10^{-3}$ °K; at this temperature $\xi K \gtrsim 1$ for $\theta \gtrsim 70°$. Thus this curve illustrates the behavior of Γ at the transition between "critical" and "hydrodynamic" regions.

Finally, in the experiment performed at $T-T_c = 121\times10^{-3}$ °K, the asymptotic behavior is never attained. This was expected because at this temperature one has $\xi K = 0.25$ for the highest recorded value of $\theta = 130°$ and thus one is still appreciably far from the "critical region" $\xi K \gtrsim 1$.

A theoretical justification of the observed $\Gamma \sim K^3$ behavior can be summarized as follows, according to scaling theory.[4] In a general way the spectral linewidth can be expressed by

$$\Gamma = K^x f(\xi K),$$

where $f(\xi K)$ is a homogeneous function of ξK. It is known that far from the critical temperature one has

$$\Gamma \sim DK^2 \text{ with } D \sim 1/\xi,$$

so that

$$\Gamma \sim K^2/\xi \text{ or } \Gamma \sim K^3/\xi K.$$

One thus obtains $x = 3$ and $f(\xi K) = (\xi K)^{-1}$. Moreover scaling theory shows that when $\xi K \to 1$, $f(\xi K)$ tends toward a constant value independent of T. One then expects

$$\Gamma = AK^3 \text{ with } A = \text{const},$$

which is our experimental result.

As a conclusion, it seems that the "critical region" of a binary mixture is characterized by the appearance of an asymptotic behavior $\Gamma = AK^3$.

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