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LIGHT SCATTERING IN AN IMPURE BINARY LIQUID MIXTURE NEAR THE CRITICAL POINT

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Light-scattering measurements were made in a ternary critical mixture of phenol and water with hypophosphorous acid as an impurity. The exponents which characterize the divergence of the intensity and inverse linewidth near the critical point are found to have nearly the same value in this system as in pure phenol-water. In addition it was observed that the light-scattering cross section and the inverse linewidth were appreciably larger in the impure system than in the pure one. Some of the experimental results are compared with theory.

We report here a measurement of the intensity I and spectral width Γ (more precisely called the inverse correlation time) of light scattered by concentration fluctuations in a critical mixture of phenol and water with 5.7 wt% hypophosphorous acid (H_3PO_2) as an impurity. The critical exponents γ and γ^* are obtained from the equations $I^{-1} \propto |\Delta T|^{\gamma}$ and $\Gamma \propto |\Delta T|^{\gamma^*}$, where $\Delta T \equiv T - T_c$, with T_c being the critical temperature.[1] The exponents are found to have approximately the same value in this ternary mixture as in the pure phenol-water system in which γ^* was previously measured.[2]

For a binary system in which the photon momentum transfer K is much less than the reciprocal of the correlation length, the following relations hold[3,4]:

$$I^{-1} \propto (\partial \mu / \partial c)_{T,p} \tag{1}$$

$$\Gamma = DK^2. \tag{2}$$

Here μ is an appropriately defined chemical potential, c is the concentration of one of the components, p is pressure, and D is the mutual diffusion coefficient. The coefficient D may be written as the product[5]

$$D = \alpha^* (\partial \mu / \partial c)_{T,p}, \tag{3}$$

where α^* is a purely dynamic factor called the concentration conductivity. The observed weak dependence of γ on impurity concentration is predicted by Fisher[6] on the basis of thermodynamic arguments. It also follows from the analysis of certain lattice gas models containing impurities.[7,8] Since the correlation time reflects a nonequilibrium property of a critical system, the above theories can have nothing to say about the effect of impurities on the value of γ^*. Nor are they concerned with the following intensity and spectral-width ratios:

$$R_I = I(\Delta T)/I(-\Delta T)$$

and

$$R_\Gamma = \Gamma(-\Delta T)/\Gamma(\Delta T),$$

where the functional dependence of I and Γ on ΔT has been explicitly designated. Here the argument ΔT ($-\Delta T$) refers to measurements made above (below) T_c. These ratios were also found to have approximately the same values in the pure and impure systems.

Before proceeding to a more detailed discussion of the results, some relevant experimental details will be presented.

The apparatus used to determine $\Gamma(\Delta T)$ was a photon correlator whose method of operation has been previously described.[2,9] The intensity measurements were corrected to take into account background, extinction, and density fluctuations, all of which were small.

In the impure system, both I and Γ were measured at two scattering angles, $\theta = 30°$ and $\theta = 81°$. The temperature interval covered was $0.6°\text{C} \lesssim |\Delta T| \lesssim 8°\text{C}$. In this range γ and γ^* could be determined from the observed linear dependence of $\ln I$ and $\ln\Gamma$ on $\ln|\Delta T|$, both above and below T_c. Data obtained at the two scattering angles gave the same exponents within the experimental error.

Measurements in both the pure and impure systems were carried out at the critical concentration. Sample preparation of the pure binary system is described in Ref. 2. Its critical parameters were $T_c = 66.05°\text{C}$, $c(\text{phenol}) = 35$ wt%, and its impurity content was less than 0.02 wt%. The H_3PO_2 in the impure system depressed the critical temperature to 36.04°C and shifted c(phenol) to 36.3 wt%. The critical solution was prepared as follows: A number of samples, all containing 5.7 wt% H_3PO_2, but with various phenol/water ratios, were elevated in temperature and then slowly cooled until phase separation occurred. That particular sample which developed a phase boundary dividing the fluid into equal volumes was selected as being of critical concentration. With temperature plotted on the vertical axis, the various samples referred to here fall in a plane P_S which is parallel to the phenol-water base plane of the triangular ternary phase diagram. The line of plait points intersects P_S if one of the samples has the plait-point composition. This point of intersection selects out the desired sample, i.e., the one with critical composition. It also locates its critical temperature. The critical temperature was more precisely located, in the sample thus selected, as that for which the light-scattering cross section was a maximum.

The results of this study are presented in Table I. There it may be seen that within experimental error γ, γ^*, and R_I/R_Γ have approximately the same value in both systems. From Eqs. (1)-(3) and the definitions of R_I and R_Γ, it may be seen that $R_I/R_\Gamma = \alpha^*(\Delta T)/\alpha^*(-\Delta T)$. It is especially interesting that this purely dynamic quantity appears even less impurity dependent than either R_I or R_Γ. Attention is also drawn to the fact that in the two-phase region, γ and γ^* are measurably larger in the low-density (water-rich) phase than in the high-density (phenol-rich) phase. It is, however, possible that these discrepancies would vanish if the measurements were carried to sufficiently small values of $T_c - T$.

Not shown in Table I is our interesting observation that the light-scattering cross section at a given angle and temperature is approximately twice as large in the impure system as in the pure one, with Γ being smaller by roughly a factor of 3.

As already noted, γ is the only parameter in Table I whose change with impurity level may be estimated theoretically. Recent calculations[6-8] indicate that both above and below T_c, γ will at most be changed to a value

$$\gamma_X = \gamma/(1-\alpha), \qquad (4)$$

where α is the heat-capacity exponent.[10] Exponents other than γ_X refer to the pure binary system. Equation (4) is actually valid only in the limit $|\Delta T| \to 0$ or at finite $|\Delta T|$ if the impurity is in a sense infinitely effective in altering the critical behavior of the pure system. When these conditions are not satisfied, a reasonable estimate of γ_X is provided by the equation[11] $\gamma_X - \gamma \lesssim f\gamma\alpha/(1-\alpha)$, where f is the fractional change in T_c

Table I. Measured critical parameters for the pure and impure phenol-water systems. The symbols are defined in the text.

	$T > T_c$		$T < T_c$ Low Density Phase					$T < T_c$ High Density Phase				
	γ	γ^*	γ	γ^*	R_Γ	R_I	$\frac{R_I}{R_\Gamma}$	γ	γ^*	R_Γ	R_I	$\frac{R_I}{R_\Gamma}$
Phenol-Water	1.32 (0.03)	0.68 (0.03)	1.36 (0.03)	0.68 (0.03)	2.29 (0.13)	5.6 (0.6)	2.4 (0.4)	1.28 (0.04)	0.60 (0.02)	1.84 (0.09)	5.0 (0.3)	2.7 (0.3)
Impure Phenol-Water	1.27 (0.04)	0.70 (0.03)	1.45 (0.06)	0.69 (0.04)	1.96 (0.10)	4.5 (0.6)	2.3 (0.5)	1.22 (0.05)	0.59 (0.03)	1.50 (0.10)	3.8 (0.4)	2.5 (0.5)

produced by the addition of the impurity X. From a measurement of the heat capacity of phenol-water near the critical temperature,[12] we roughly estimate α to be less than 0.1. Using the fact that $f \simeq 0.1$ in our system, one estimates $\gamma_X - \gamma \lesssim 0.02$. This result is consistent with the data in Table I when experimental uncertainties are taken into account.

To summarize, it is found that impurities added to a critical system have a small effect on at least some of its thermodynamic and transport properties. Aside from the theoretical implications of this work, it suggests that one need not use extremely high-purity samples to obtain meaningful values of γ and γ^* in fluid systems.[13]

We are grateful to Professor M. E. Fisher and Mr. P. E. Scesney for permission to quote their work prior to its publication. Also we have profited by correspondence and conversations with Professor B. Widom and Professor Fisher. One of us (C.S.B.) would like to thank Dr. P. N. Pusey for assistance in sample preparation and linewidth measurements.

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OBSERVATION OF SUPERFLUID-HELIUM PERSISTENT CURRENT BY DOPPLER-SHIFTED SPLITTING OF FOURTH-SOUND RESONANCE*

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Persistent currents in superfluid helium contained in the pores of a packed powder have been observed by means of the Doppler-shifted splitting of an azimuthal resonant fourth-sound mode of the cylindrical resonator containing the helium.

A direct method to determine the velocity of a fluid is to measure the Doppler shift of a sound wave. The purpose of this Letter is to describe the first such determination of the velocity of a circulating persistent current in superfluid helium. Previous measurements of persistent currents depend on gyroscopic effects associated with their angular momentum.[1] In an imaginative series of experiments Reppy and his coworkers have exploited such measurements to study a variety of phenomena.[1] The momentum measurements yield the product $\rho_s(T)\omega_s(T)$ and the relative temperature dependence of ρ_s (ρ_s and ω_s are the superfluid component density and angular velocity, respectively). If ρ_s is known at one temperature then $\rho_s(T)$ and $\omega_s(T)$ are determined. The only circumstance in which there is an uncertainty in ρ_s occurs where size effects are important[2] and this is possible since in such experiments the normal fluid is locked by the small pores of a porous medium which fills the test chamber. We determine the velocity of persistent currents by the measurement of the Doppler shift experienced by fourth sound, the sound wave which propagates in the helium-filled pores of such systems.[2] Since the fundamental condition on the quantized vortices which comprise the velocity field establishes their velocity rather than their momentum, it would appear that such a measurement is preferable to a momentum de-