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OBSERVATION OF CRITICAL PARAMETER RENORMALIZATION IN A THREE-COMPONENT LIQUID MIXTURE (*)

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Résumé. — Nous avons étudié, en fonction de la température et de l’angle de diffusion, l’intensité et la dépendance temporelle de la lumière diffusée par un mélange liquide de trois composants, bromobenzène-eau-acétone, à la fois au-dessus et au-dessous du point critique de démixtion $T_c$. On peut décrire ce système comme un système binaire très impur, l’acétone jouant le rôle d’une impureté inerte. La concentration de l’acétone dans le mélange était environ de 57 %, en poids. À la fois au-dessus et au-dessous de $T_c$, la théorie de Kawasaki décrit bien la constante d’atténuation des fluctuations d’intensité de la lumière diffusée. Les exposants critiques et leurs rapports semblent présenter une renormalisation due à la présence de l’impureté, comme l’ont prévu Fisher et Scesney.

Abstract. — We have studied, as a function of temperature and scattering angle, the intensity and time dependence of light scattered by the three-component liquid mixture, bromobenzene-water-acetone, both above and below its critical mixing temperature $T_c$. This system can be pictured as a highly impure binary system, with acetone playing the role of an inert impurity. The concentration of acetone in the mixture was about 57 %, in weight. Above and below $T_c$, the decay rate of intensity fluctuations in the scattered light is well described by the Kawasaki theory. The critical exponents and ratios appear to show renormalization due to the presence of the impurity, as predicted by Fisher and Scesney.

I. Introduction. — We report here further light scattering measurements on a three-component liquid mixture near its critical point. The system chosen for study was a critical mixture of bromobenzene (19 wt %), water (24 wt %), and acetone (57 wt %). This system (henceforth designated as BWA) may be regarded as a highly impure mixture of water and bromobenzene, with the acetone playing an active role in permitting the mutual solution of these otherwise immiscible components, water and bromobenzene.

The earlier light scattering measurements on BWA by Bak et al. [1] were confined to the one-phase region. We have repeated this work using a different sample and a more efficient 20-channel correlation spectrometer which has been described elsewhere [2]. In addition, measurements were made below $T_c$ on both sides of the coexistence curve.

The results provide a partial verification of the spectral-width calculation of Kawasaki [3] and the renormalization theory of Fisher [4]. In addition, some of the measurements can be compared with recent calculations of Fisher and Scesney [5]. These authors have carried out a detailed numerical analysis of a decorated Ising lattice model and have thereby shown that only very close to the critical temperature and at very high impurity concentrations (near 100 %) can the experimenter expect to see full critical exponent renormalization. They have also calculated the effect of renormalization on the ratio of magnetic susceptibility of their system at equal, but small, intervals above and below the critical temperature, $T_c$.

We report precise measurements of the spectral width $\Gamma$ in both the one- and two-phase regions and will also discuss the results of less precise measurements of the average scattered light intensity, $I$. Physically, $\Gamma$ is a measure of the inverse lifetime of the concentration fluctuations.

The new measurements reported here are in satisfactory agreement with those presented in reference [1] (to be referred to as [1]). The observations in the two-phase region enable further comparison with the predictions of Fisher and Scesney.

II. Theory. — In this section we summarize the main theoretical results with which the measurements in section III will be compared.

a) THE NEAR-HYDRODYNAMIC REGIME. — In this domain, the intensity is expected to conform to the Ornstein-Zernike (OZ) prediction

$$I \propto k_B T \left( \frac{\partial \mu}{\partial c} \right)^{-1} \left( 1 + K^2 \xi^2 \right)^{-1} ,$$

(1)
where \( \mu \) is an appropriately defined chemical potential difference [6] and \( k_B \) is Boltzmann's constant. The derivative \( (\partial \mu / \partial c)_T \) is analogous to the isothermal susceptibility \( \chi_T \) in a ferromagnet and the isothermal compressibility \( \kappa_T \) in a simple fluid. Here \( \zeta \) is the OZ correlation length, and the photon momentum transfer \( K = (4 \pi n / \lambda) \sin (\theta / 2) \), where \( \lambda \) is the wavelength of the incident light and \( n \) is the refractive index of the solution. In these experiments the light source was a He-Ne laser with \( \lambda = 6328 \text{ Å} \). The divergence of \( (\partial \mu / \partial c)_T \) is characterized by the exponent \( y \), whence

\[
I \propto k_B T e^{-\varepsilon(1 + K^2 \zeta)^{-1}}
\]

(2)

where

\[
\varepsilon \equiv \frac{T - T_c}{T_c}.
\]

(3)

The temperature dependence of \( \zeta \) is given by the equation

\[
\zeta = \zeta_0 \varepsilon^{-y}.
\]

(4)

From eq. (2),

\[
\lim_{\varepsilon \to 0} I \propto e^{-\varepsilon^y}.
\]

(5)

We consider next the dynamic behavior of a critical mixture. Kawasaki [3] has predicted that when \( K \zeta \leq 1 \),

\[
\Gamma = DK^2 (1 + \frac{3}{5} K^2 \zeta^3).
\]

(6)

An equation similar to (6) was first derived by Fixman (1).

We also note that the diffusivity, \( D \), in (6) may be written as

\[
D = \alpha^* (\partial \mu / \partial c)_T.
\]

(7)

where \( \alpha^* \) is called the concentration conductivity [8].

Kawasaki [3] and Ferrell [9] have recently shown that in simple fluids and binary mixtures, \( D \) is related to the shear viscosity of the medium through a Stokes-Einstein type of equation:

\[
D = \frac{k_B T}{6 \pi \eta \zeta},
\]

(8)

where \( \eta \) is the viscosity and \( k_B \) is Boltzmann's constant. The theory utilized in the derivation (8) predicts that \( \eta \) does not exhibit a divergence near the critical point. It then follows that

\[
D \propto \zeta^{-1} \propto \varepsilon^y.
\]

(9)

The validity of eq. (8) and (9) remains somewhat open to question, since a number of binary mixtures are known to exhibit a peak in the shear viscosity near \( T_c \) (2). When analyzing the experimental results of section III, we will assume in the absence of contrary evidence that \( \eta \) was constant over the temperature interval studied.

Theoretically the concentration conductivity \( \alpha^* \) is expected to diverge as \( \varepsilon^{-y} \), and this type of divergence is typically seen [8], [9], [11], [12]. Yet it is now known that in several pure fluids the thermal conductivity \( \Lambda \) appears to be the sum of two terms, one of which diverges and the other which does not [10], [13]. The background contribution to \( \Lambda \) in SF, for example, seems to be in large measure responsible for the observed anomalous exponent describing the temperature dependence of the thermal diffusivity above \( T_c \) [13]. Since \( \alpha^* \) is the binary-mixture analog of \( \alpha \), one wonders whether a similar term may not be present in these systems. Again, in the absence of relevant data, it will be assumed that there is no appreciable background contribution to this transport coefficient in the system under study here.

b) Critical regime \( (K \zeta > 1) \). — In the extreme limit \( K \zeta \gg 1 \), the dynamic scaling ansatz of Halperin and Hohenberg [14] requires that \( \Gamma \propto K^3 \). A detailed mode-coupling calculation, valid at all values of \( K \zeta \), has been made by Kawasaki [3]. He finds that

\[
\Gamma = \frac{2 A}{K^3} \left[ y^3 + y^2 + (1 - y^4) \tan^{-1} (y^{-1}) \right],
\]

(10)

where \( y = (K \zeta)^{-1} = (K \zeta_0)^{-1} \varepsilon^y \). This equation reduces to eq. (6) when \( K \zeta \) is small and also has the limiting form \( \lim_{\varepsilon \to 0} \Gamma = AK^3 \), where \( A = k_B T / 16 \eta \).

While all of the equations in this section were derived for pure systems, it will be assumed that the presence of an impurity changes the parameters, but not the form of the algebraic expressions. This assumption is in conformity with the spirit and the results of renormalization theory and is also supported by the experimental results reported in section III.

c) Ratios. — In addition to the exponents \( y \) and \( \nu \) defined above, two other quantities of interest are the intensity ratio \( R_I \) and the correlation length ratio \( R_\xi \). These quantities are defined by the equations:

\[
R_I = \lim_{\varepsilon \to 0} \frac{I(\Delta T)}{I(-\Delta T)},
\]

(11)

and

\[
R_\xi = \frac{\xi(\Delta T)}{\xi(-\Delta T)}.
\]

(12)

In eq. (11) and (12), the functional dependences of \( I \) and \( \xi \) on \( \Delta T = T - T_c \) are explicitly designated. Eq. (1) implies

\[
R_I = \frac{(\partial \mu / \partial c)_{\Delta T}}{(\partial \mu / \partial c)_{\Delta T}}.
\]

(13)

The prime superscript will denote the value of the parameter in the two-phase region.

d) Renormalization. — The presence of an impurity in a system has two important effects. For one, it

(1) See, for example, reference [7].

(2) A recent review article by Sengers [10] presents the results of many viscosity measurements in binary mixtures and simple fluids.
provides an additional degree of freedom, \( z \) \(^{(3)} \) to the system. For compelling reasons, Fisher [4] chooses \( z \) to be the thermodynamic potential conjugate to the fractional impurity concentration \( x \). Secondly, the fact that \( x \) is held at a fixed value, assures that the system will be constrained to a particular path in the space of its thermodynamic variables. Since the constraints imposed on even a pure system determine the degree of the divergence which is observed \(^{(4)} \) it is perhaps not surprising that the imposition of this added constraint results in a change of the critical exponents.

With only a minimum number of thermodynamic assumptions, Fisher was able to relate the value of exponents such as \( y \) in the impure system to their counterparts in the pure one. The result is that in the limit of very large \( x(x \rightarrow 1) \), and at temperatures sufficiently close to the critical point, \( y(x) \) in the impure system take on its "fully renormalized" value as \( y(x) \), and shows that

\[
\gamma_x = \frac{(y - 1)}{a}, \quad \gamma_x' = \frac{(y' - 1)}{a'}.
\]

In (14), \( a \) characterizes the divergence of the specific heat. The exponents on the right side of this equation are those associated with the pure system. Away from the asymptotic limits, \( x \rightarrow 1, |\Delta T| \rightarrow 0 \), exponents, prefactors, and ratios such as \( R \) are expected to have values which fall between their fully renormalized and pure-system values. By curve-fitting the expressions derived for their decorated Ising model, Fisher and Scesney demonstrate that the apparent exponents \( y(x) \), \( a(x) \), etc. exhibited by the model, depend not only on \( x \) but also on the temperature interval over which the fit is made. For example, with \( x = 0.15 \), they find that \( y(x) \) differs from its pure-system value by less than one percent, even if the curve fitting is carried out in the interval \( 10^{-7} < \epsilon < 10^{-5} \). Full renormalization would correspond to a 15% change in \( y(x) \) if \( y \) and \( x \) were to have their Ising model values of 1.25 and 1/8 respectively. It thus is apparent that renormalization effects can be studied only at very large impurity levels. A previously reported study [16] of a slightly impure \( (x = 0.057) \) mixture of phenol and water supports this conclusion.

III. Experimental results. — We begin by reviewing the results of the previous study of BWA above the critical point [1]. By measuring \( I \) and \( \Gamma \) in the near-hydrodynamic regime and fitting the data to eq. (1) and (6), one can obtain values of the parameters \( \gamma \) and \( \nu \). This procedure led to the following numerical results reported in [1]: From intensity measurements,

\[
\gamma = 1.50 \pm 0.08 \quad 2 \nu = 1.50 \pm 0.08 \quad (15)
\]

and from linewidth measurements,

\[
2 \nu = 1.58 \pm 0.12. \quad (16)
\]

The data of (15) and (16) are in conformity with the scaling prediction, \((2 - \eta) \nu = \gamma \), assuming \( \eta = 0 \) [15]. In addition, the parameter \( R_I \) was measured to be

\[
R_I = 1.41 \pm 0.1. \quad (17)
\]

The sample used in that study had a critical temperature of 28.39°C.

The present experiments were performed on a sample with a slightly different composition, as evidenced by the fact that the critical temperature was 32.36 ± 0.01°C. Attention was centered on measurements of the fluctuation lifetime. The results are shown in figure 1 \((T > T_c)\) and figure 2 \((T < T_c)\). The circles and crosses in figure 2 refer to the low-density and high-density phases respectively. In analyzing the measurements on both sides of the coexistence curve, \( \Gamma'' \) (lower phase) and \( \Gamma' \) (upper phase) were averaged together to effect a partial correction for the (small) index of refraction change with temperature. The average index of refraction of the mixture was measured to be \( n = 1.381 \). The observed photocount correlation functions were well described by single exponentials, indicating a single mode of decay [2].

The data of figures 1 and 2 were analyzed by making a three-parameter least-squares fit to the Kawasaki expression, eq. (10). The resulting values for the para-

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The data of figures 1 and 2 were analyzed by making a three-parameter least-squares fit to the Kawasaki expression, eq. (10). The resulting values for the para-
meters $A$, the exponents $v$, and the magnitudes of the correlation lengths were

$$A = 3.05 \pm 0.1 \text{ cm}^3/\text{s} \quad (18)$$

$$\xi = (242 \pm 10) (T - T_c)^{-\nu} \text{Å}, \quad v = 0.81 \pm 0.03 \quad (19)$$

and

$$A' = 2.96 \pm 0.05 \text{ cm}^3/\text{s} \quad (20)$$

$$\xi' = (212 \pm 5) (T_c - T)^{-\nu'} \text{Å}, \quad v = 0.72 \pm 0.02. \quad (21)$$

Plots of $\Gamma/k^3$ vs $(K\xi)^{-1}$ are given in figures 3 and 4. The solid lines are the results of putting the above values of $A$ and $\xi$ into eq. (10). Agreement with the theory is good over the whole range of $K\xi$, though there is some evidence of systematic deviation from Kawasaki behavior for $T > T_c$. It can be seen that the asymptotic values, $\lim_{K\xi \to \infty} \Gamma/k^3$ are the same above and below $T_c$. This result is reasonable since it is to be expected that any quantity which becomes independent of $T$ as $T$ tends to $T_c$ will be continuous through the phase transition. The shear viscosities obtained from the values of $A$ given in (18) and (20) are

$$\eta = (8.6 \pm 0.3) \times 10^{-2} \text{ poise}$$

and

$$\eta' = (8.8 \pm 0.2) \times 10^{-2} \text{ poise}.$$}

There exists no direct measurement of this ratio. It is seen to be weakly temperature dependent, varying from 1.14 at $|\Delta T| = 1 \text{ °C}$ to 1.0 at $|\Delta T| = 5 \text{ °C}$. The fact that $v$ is greater than $v'$ may be due to failure to take into account background contributions to the diffusivity, as in SF$_6$ [13]. On the other hand, it may be a real effect in this system and in others, such as phenol-water [17], where a similar discrepancy was observed.

In the course of performing the linewidth measurements, record was kept of the average scattered intensity $I(\Delta T)$. While considerably higher accuracy could be obtained in an experiment designed specifically for measuring $I(\Delta T)$, these data were sufficiently precise to provide information concerning $\xi$, $\gamma$ and $R_t$. The exponents $\gamma$ and $\gamma'$ were found to be roughly equal to each other and $\gamma$ was in agreement with the results quoted in [1] (see eq. (15)). The intensity ratio (eq. (11)) was found to be

$$R_t = 1.39 \pm 0.2, \quad (23)$$

again in agreement with the previously reported measurement [1]. Here, as before, the result quoted in (23) is an average over the two phases.

Finally we report our observation that the correlation length determined from intensity measurements (eq. (1)) exceeds the value $\xi$ obtained from the Kawasaki equation (eq. (10)) by a factor of about 1.25. The same factor was observed both above and below $T_c$ and was also seen in the measurements reported in [1]. We do not understand the origin of this discrepancy.

IV. Conclusions. — In this section the experimental results will be compared with the theoretical predictions in section II and with Ising model calculations. First we take up the question of critical exponent renormalization. To establish unambiguously that BWA exhibits renormalization, it would be necessary to measure critical exponents in a pure mixture of bromobenzene and water. Since these two fluids are almost completely immiscible at ordinary temperatures and pressures, there is really no possibility of determining the unrenormalized exponents $\alpha$ and $\gamma$. The only alternative, then, is to insert «reasonable» values of these parameters into eq. (14) and into Fisher and Sceensney's calculation of $\gamma(x)$ and $R_t(x)$. A choice which conforms satisfactorily to both experimental results and three-dimensional Ising model calculations is $\alpha = 0.125$, $\gamma = 1.3$ [8], [15]. When these values are inserted into the right-hand side of (14), one finds

$$\gamma_s = \left[ \frac{1.3}{1 - 0.125} \right] \simeq 1.5. \quad (24)$$

Assuming $\gamma = 2v$, this should be compared with $2v = 1.62 \pm 0.06$ and $2v = 1.44 \pm 0.04$ and with the apparent exponent $\gamma(x = 0.6)$ calculated by Fisher and Sceensney (see section IIId and eq. (19) and (21)). They find that in the temperature interval

$$10^{-3} \leq \epsilon \leq 10^{-5}, \quad \gamma(0.6) \simeq 1.35.$$ 

It appears then either that the calculations of Fisher and Sceensney underestimate the degree of renormalization or that the unrenormalized exponents $\alpha$ and $\gamma$ are
considerably different in this system than in most fluid systems [8].

We turn next to a comparison of measured and calculated values of $R_T$ and $R_k$. Fisher and Scesney have shown that when $\gamma = \gamma'$, the fully renormalized ratio,

$$\left( \frac{\partial \mu}{\partial \phi} \right)_{\Delta T} = \left( \frac{\partial \mu}{\partial \phi} \right)_{\Delta T}^\ast,$$

is related to the corresponding specific heat ratio through the equation

$$R_{fx} = \left[ \left( \frac{\partial \mu}{\partial \phi} \right)_{\Delta T}^\ast \right]_x = \left[ \left( \frac{\partial \mu}{\partial \phi} \right)_{\Delta T}^\ast \right]_{x=0} \left[ \frac{B(\Delta T)}{B'(\Delta T)} \right]_{x=0},$$

(25)

where $B(\Delta T)$ and $B'(\Delta T)$ are the specific heat amplitudes of the pure system. The Ising model yields numerical values for both ratios on the right side of (25) viz. [15]

$$\left[ \left( \frac{\partial \mu}{\partial \phi} \right)_{\Delta T}^\ast \right]_{x=0} = 5.2 \pm 0.05$$

and

$$\left[ \frac{B(\Delta T)}{B'(\Delta T)} \right]_{x=0} = 2.1 \pm 0.1.$$  

(26)

Assuming $\gamma = 1.3$ and invoking eq. (13), the fully renormalized intensity ratio is

$$R_{fx} = 5.2(2.1)^{-1.2} = 2.0.$$  

(27)

Fisher and Scesney have also evaluated $R_k(x)$ and found that renormalization of this quantity should be relatively easy to observe even appreciably far from the asymptotic limit, $x \rightarrow 1, |e| \rightarrow 0$. According to their calculations, $R_k(x \approx 0.6, |e| \approx 10^{-2}) \approx 4$, whereas the experimental value given in (23) is $R_k = 1.39 \pm 0.2$. Again it appears that either the theory underestimates the degree of renormalization or that one or another of the unrenormalized coefficients was chosen incorrectly.

Since $R_f$ and $R_k$ are expected to be related, a measurement of the latter ratio provides an additional check on Fisher and Scesney’s calculations of $R_{fx}$ and $R_k(x)$. Assume, for example, that OZ theory is valid in the hydrodynamic regime. A direct consequence of the theory is the relation $\xi^2 = F(T) (\partial \mu/\partial \phi)^{-1}$, where $F(T)$ is a slowly varying function of temperature through the critical point. This equation, together with (1), gives

$$\frac{I(\Delta T)}{I(- \Delta T)} \propto \frac{\xi^2(\Delta T)}{\xi^2(- \Delta T)}.$$  

(28)

Thus, from (12)

$$R_T = R_k^2.$$  

(29)

Substituting into (29) the experimental value of $R_k$ from (22) gives $R_k = 1.30 \pm 0.12$, whereas the directly measured $R_T$ is $1.39 \pm 0.2$ from eq. (23). Agreement between these two ratios is therefore good.

Despite this agreement it should be realized that there is reason to doubt the validity of eq. (28). Recent light scattering measurements in xenon [13], [19] and in CO$_2$ [20] imply the correctness of the OZ theory in the near-hydrodynamic regime. In the language of simple fluids, one of the theory’s predictions is [18]

$$\xi^2 \propto \kappa_r$$  

(30)

and this relation in particular is verified in the xenon experiment. On the other hand, eq. (30) seems to yield predictions which are strikingly at odds with the Ising model result, $\kappa_r(\Delta T)/\kappa_r(- \Delta T) = 5.2 \pm 0.05$. To see this, consider the ratio, $(\partial \mu/\partial \phi)|_{\Delta T}$, which is equal to $\xi^2/\xi^2_0$ if $\gamma = \gamma'$. While $\xi^2/\xi^2_0$ has not yet been evaluated for a three-dimensional system, it is reasonable to assume that it falls between the two-dimensional Ising and mean-field values. These are respectively 2 and $\sqrt{2}$ [15]. Since three-dimensional exponents are usually closer to the mean-field values, we guess $\xi^2/\xi^2_0 = 1.6$ or $\xi^2/\xi^2_0 \approx 2.6$. But if (30) were correct, this ratio would also be $\kappa_r(\Delta T)/\kappa_r(- \Delta T) = 5.2$, rather than 1.6. It appears thus that there is little theoretical justification for assuming that

$$\frac{I(\Delta T)}{I(- \Delta T)} \equiv R_f = \left( \frac{\xi^2}{\xi^2_0} \right)^2.$$  

(5) (7)

In spite of uncertainties in the above arguments, it seems safe to assert that the BWA system exhibits renormalization and that the effect may be readily seen from measurements of $R_k$ as well as $R_T$.

V. Summary. — A three-component mixture such as BWA is characterized by a line of critical points, each point having associated with it a critical temperature and a critical composition [21]. The particular mixture studied here had a critical temperature of 32.36°C and contained ~ 60% acetone with the remaining fraction being roughly equally divided between the two other components. The acetone, being soluble in all proportions in each of the other components, may be regarded as an impurity. Our light scattering measurements, which were carried out in both the one and two phase regions, lend support to Kawasaki’s mode coupling analysis and to the renormalization calculations of Fisher and Scesney. An interesting result of this study is that renormalization is more readily visible through its effect on $R_T$ and $R_k$ than through its influence on critical exponents such as $\gamma$ and $v$. The results of the present study are summarized and compared with theory in table I.

Finally we wish to call attention to an observation made in [1] but not previously mentioned in this paper, viz. the experimental accessibility of the critical regime ($K_2 \gg 1$) in BWA. The following argument, due to Widom (7), suggests that this property should be

(5) A better estimate might be $R_f = (\xi^2/\xi^2_0)^{2-\eta}$. [15], but for typical value of $\eta < 0.05$, this modification to the OZ theory cannot alone account for the large discrepancy discussed here.

(7) We are indebted to Prof. M. E. Fisher for an illuminating discussion concerning the validity of the OZ theory.
Parameter | Experiment | Theory (*) | Calculations of Fisher and Scesney (*)
--- | --- | --- | ---
--- | --- | --- | ---
$v$ | 0.81 ± 0.03 | 0.65 | 0.67
$v'$ | 0.72 ± 0.02 | 0.65 | 0.75
$R_f$ | 1.39 ± 0.2 | 5.2 | 4.0
$R_g$ | 1.14 ± 0.06 | See section IV for discussion | |

(*) In the theoretical calculations, the exponents $\alpha$ and $\alpha'$ have been taken as 1/8.

(†) These are approximate 3-D Ising model values.

(‡) Using eq. [14] and taking $\gamma = 2v$.

(§) Reference [5].

One of us (WG) is grateful to many colleagues in the departments of Physics, Applied Physics, and Chemistry for hospitality extended during a Sabbatical Leave at Cornell University 1970-1971.

Note Added in Proof. — The comments in this paper concerning the anomalous exponent of $SF_6$ no longer appear to be applicable, since recent experiments did not show anomalous behavior. See: Lim (T. K.), Swinney (H. L.), Langley (K. H.) and Kachnowski (T. A.), Phys. Rev. Letters, 1971, 27, 1776, and Feke (G. T.), Hawkins (G. A.), Lastovka (J. B.), and Benedek (G. B.), Phys. Rev. Letters, 1971, 27, 1780.

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