PHYSICAL REVIEW A VOLUME 33, NUMBER 4 APRIL 1986

Turbidity in the binary fluid mixture methanol-cyclohexane

D. T. Jacobs
Department of Physics, The College of Wooster, Wooster, Ohio 44691
(Received 20 June 1985)

The turbidity of a near-critical binary fluid mixture, methanol-cyclohexane, has been carefully measured above its (upper) critical consolute point. These measurements covered the reduced temperature region $10^{-5} < t < 10^{-2}$ and allowed theoretical predictions for the critical exponents ν and γ to be verified. The amplitude ξ_0 of the correlation length was also determined to be 3.24 ± 0.2 Å while τ_0 was $(6.9\pm0.1)\times10^{-6}$ cm^{-1}. Our result for ξ_0 is compared with other experimental determinations, and when combined with τ_0 and coexistence curve data, is used to determine the ratio $R=\xi_0(B^2/k_BT_c\chi_0)^{1/3}=0.71_5\pm0.02_4$ which is consistent with the prediction 0.65—0.67 from two-scale-factor universality.

I. INTRODUCTION

The study of systems close to a second-order phase transition has progressed enormously since its inception over 100 years ago. The last 15 years have witnessed numerous examples of very diverse systems[1] (from biomembranes to isotopic yields in nuclear physics) which, at first glance, would seem far removed from a thermodynamic model. However, a system near a second-order phase transition can be viewed[2] as a scale transformation that has the properties of a group. The renormalization-group theory that resulted has been very successful in predicting the similarities seen in such diverse systems yet showing the quantitative distinctions that have been observed in different universality classes. Each universality class is described by a separate model. The model which is expected (and has experimentally been well confirmed) to describe binary liquid mixtures, binary alloys, ferromagnetism, and a liquid-gas transition is the three-dimensional Ising model.[3]

Renormalization-group theory predicts only two linearly independent critical exponents with all others being related with equalities replacing the strict thermodynamic inequalities. These relationships have been well verified experimentally.[3] The predictions for the values of the exponents are much harder to obtain since expansions, and hence approximations, must be made for the three-dimensional (3D) Ising model. Three exponents that are particularly relevant for this work are the correlation length exponent ν, the osmotic compressibility exponent γ, and the light scattering correlation function η. These three are related by $(2-\eta)\nu=\gamma$. The current predictions[4] for a 3D Ising model are $\nu=0.63$, $\gamma=1.241$, and hence $\eta=0.03$.

More recent predictions of renormalization-group theory suggest that the amplitudes of the anomalies described by critical exponents will also be related. In particular, only two scale factors are needed to relate the amplitudes. This two-scale-factor universality[5,6] predicts a relationship between the leading amplitude ξ_0 of the correlation length ξ,

$$\xi=\xi_0 t^{-\nu}, \tag{1}$$

and the leading amplitude A of the heat capacity anomaly C_p,

$$C_p=At^{-\alpha}/\alpha , \tag{2}$$

where α is another critical exponent, t is the reduced temperature from critical, $t\equiv|T-T_c|/T_c$, T is the absolute temperature, and T_c is the critical temperature. The two-scale-factor prediction is $A\xi_0^3/k_B=0.0165$ where k_B is Boltzmann's constant. This prediction has been verified[5,7] in liquid-gas systems, but not conclusively validated[8] in binary mixtures because of the difficulty of determining ξ_0 and measuring A.

Another prediction of two-scale-factor universality involves the amplitudes B of the coexistence curve, χ_0 of the osmotic compressibility, and ξ_0. These are defined by Eq. (1) and by

$$\phi-\phi_c=Bt^{\beta},\quad T<T_c \tag{3}$$

and

$$\chi=\chi_0 t^{-\gamma},\quad T>T_c \tag{4}$$

where ϕ is the volume fraction of one component in the upper or lower phase, ϕ_c is the critical composition, and β is a critical exponent predicted to be 0.325. Sengers and Moldover[7] showed these amplitudes were related by

$$R=\xi_0[B^2/(k_BT_c\chi_0)]^{1/3} \tag{5}$$

with a predicted value[5,6] of 0.65 from series expansions and 0.67 from ϵ expansions in a renormalization-group setting. Equation (5) is particularly relevant to this work since both ξ_0 and χ_0 can be determined from the turbidity. Verifying this relationship will depend upon an accurate determination of ξ_0 and, less critically, of χ_0.

Determining the amplitude ξ_0 is the major thrust of this investigation. Of the several techniques available to determine ξ_0, monitoring the transmitted light intensity was chosen. The reasons are several: the transmitted intensity provides a sensitive probe of several competing effects in the mixture so that χ_0 and ξ_0 can both be determined, the intensity is strong enough to be easily measured, and multiple scattering has a small influence. The transmitted light intensity I_t can be compared to the incident intensity I_0 to determine an effective extinction length called the turbidity τ. The turbidity is thus defined by $\tau=(-1/L)\ln(I_t/I_0)$, where L is the length of the scattering medium. The technique for measuring the turbidity of the fluids is completely described in the experimental section.

Using Ornstein-Zernike scattering, where η is assumed zero, the turbidity can be related[9] to the correlation length and osmotic compressibility. Although a development[10] where $\eta\neq 0$ has been used to determine τ, η is so small that its influence on τ is negligible at the level of the current experiments. The turbidity can thus be described[9] in terms of the critical parameters of the system by

$$\tau=\tau_0 t^{-\gamma}\{(2a^2+2a+1)[\ln(1+2a)]/a^3-2(1+a)/a^2\}\ , \tag{6}$$

where

$$\tau_0=\frac{\pi^3}{\lambda_0^4}\left[\frac{\partial n^2}{\partial\phi}\right]^2 k_B T\chi_0\ , \tag{7}$$

$a=2k_0^2\xi^2$, $k_0=2\pi n/\lambda_0$, n is the refractive index of the mixture, λ_0 is the vacuum wavelength of the light, and τ_0 is a quantity dependent on the system. The ξ_0 dependence enters in a complicated fashion through the ξ dependence of a. Equation (6) becomes[11] a simple power-law dependence far above T_c, $\tau=8\tau_0 t^{-\gamma}/3$ for $T\gg T_c$, but turns into a slowly varying function close to T_c, $\tau=(\tau_0/k_0^2)\ln(2a)$ for $T\gtrsim T_c$. As can be seen in these limits, τ_0 and ξ_0 will be coupled for data close to T_c, but only τ_0 becomes important at temperatures well above T_c.

This work describes precise measurements of the turbidity in the one-phase region close to the critical point of the binary fluid mixture methanol-cyclohexane. The data set is carefully analyzed to determine the values of ξ_0, τ_0, ν, and γ. These results are compared to other experimental measurements of the turbidity in this system and an analysis of the combined data test exponent predictions, parameter coupling, and two-scale-factor universality.

II. EXPERIMENTAL DETAILS

A. Fluids and cell

The binary fluid mixture methanol-cyclohexane was chosen for this experiment because the location of the critical point is well known,[12,13] the effect of some impurities has been investigated,[13] gravity effects should be negligible due to the density match of the components,[12] the effect of multiple scattering is known,[14] some heat-capacity data exist,[15] and a separate investigation[16] has measured the turbidity very close to the critical point.

The fluids were high-purity solvents: the methanol was Baker's Photrex $99.9^{+}\%$ pure with less than 0.05% water and the cyclohexane was Kodak's Spectro ACS $99.9^{+}\%$ pure with less than 0.02% water. These were used without further purification. The components were loaded into the cell in a dry box under a dry nitrogen atmosphere. All transfer syringes and the cell had already been carefully cleaned and dried.

The cell was a commercial (Spectrocell) cylindrical cell made of optical Pyrex with a 22-mm diameter, 10-mm path length, and a 50-mm fill tube which was heated and drawn closed to seal the fluids in the cylinder. The optical path length was measured with a Gaertner traveling microscope to be 0.999 ± 0.005 cm. The prepared composition of the fluids was $(29.47\pm0.05)\%$ by weight methanol. This is consistent with the critical composition found previously.[12,13] The location of the meniscus provides a sensitive test[17] of the composition—at the critical composition, the meniscus will remain at the center of the cell when very close to T_c. The meniscus of our sample appeared in the center whenever the system went two phase.

B. Temperature control and measurement

The cell was immersed in a cylindrical, 10-l, well-stirred water bath. The walls of the bath were made of polycarbonate and further insulated with Styrofoam below, bubble wrap on the sides, and polypropylene spheres on the top. A piece of acrylic plastic fit over the top of the bath to reduce evaporation; openings were cut to provide access. The bath fluid was distilled water with a poison (sodium azide) to eliminate algae growth. Stirring was optimized to maximize mixing yet minimize turbulence or air bubbles that would affect the optical measurements. The water in the bath was filtered continuously to remove particles larger than 10 μm. Temperature control was accomplished with a Tronac PTC-41 controller and a 100-W copper-sheathed heater. The temperature was controlled to ±0.7 mK excursions over an hour; equilibration time for the cell and fluids was a few minutes.

The temperature was monitored by a separate ac Wheatstone bridge. An ultrastable thermometrics thermistor was used to sense the temperature. The thermistor and a Vishay standard resistor formed one side of the Wheatstone bridge; the other side was a Gertsch RT-11R (six digit) ratiotransformer. The bridge was powered by a 0.1-V, 2.2-kHz sine wave and the error signal was monitored with a Princeton Applied Research 5101 lock-in amplifier. The thermistor was calibrated using the same bridge circuitry against calorimeter thermometers with a resolution of 0.01 °C and tested against National Bureau of Standards (NBS) standards to 0.002 °C at each thermometer's midpoint. The resulting accuracy in calibrating the monitor thermistor was ±10 mK with a resolution of ±0.2 mK.

C. Optics

The optical system, shown in Fig. 1, was designed and refined to optimize the precision and accuracy of the light intensity measurements which allowed the turbidity to be calculated. 2.5-cm-diam glass tubes ending in thin quartz windows were sealed in the walls of the water bath to minimize the light path in the water. The water bath,

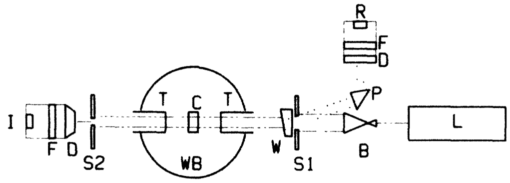

FIG. 1. Optical schematic of our turbidity experiment, as seen from above, with *L* a He-Ne laser, *B* a spatial filter and beam expander, *S*1 a 1-cm aperture, *W* a 2° wedge prism, *P* a right-angle prism, *D* a diffuser, *F* a 632.8-nm bandpass filter, *R* the reference detector, *T* the glass tubes sealed in the walls of the water bath WB and ending in thin quartz windows on the fluid side, *C* the 1-cm path length cell, *S*2 a 2-mm aperture, and *I* the transmitted beam detector. The cell can be removed from the light beam yet kept in the bath.

temperature probes, stirrer, and filter were mounted on a 2-cm-thick aluminum plate which was in turn bolted to a heavy wooden table. A 1-m optical breadboard was mounted below the plate and provided the support for all the optical components. This arrangement minimized vibration of the optical parts. A wooden frame rested on the table and surrounded all the optical components. This frame was then wrapped in black cloths and plastic to prevent any interference from stray light.

A Melles Griot 05-LLP-831 3-mW polarized He-Ne laser ($\lambda_0 = 632.8$ nm) beam passed through a spatial filter and beam expander to produce a 1-cm-wide columnated beam. Using a wide beam has several advantages—the intensity is reduced and spread over a large area to reduce local heating in the fluids, the beam was larger than the detector area so that an average light intensity could be measured, any local imperfections in an optical component would not influence the transmitted beam, and the detectors were much less sensitive to any "beam walk." The laser power varies slowly by ±2% so that the incident intensity must be monitored. A 2° wedge prism was placed in the beam to reflect a portion of the incident beam to a right-angle prism that reflected the beam into a reference detector. The wedge prism had an antireflection coating on its perpendicular face which prevented interference fringes in the reference beam.

The main beam continued into the water bath to the cell which could be removed from the beam. The cell was attached to a rod which had a vertically scribed mark on it. The rod slipped through an angular scale attached to the plastic top of the bath that allowed orientation of the cell to ±2°. After passing through the bath, the beam encountered a 2-mm aperture before striking the transmitted beam detector. The 2-mm aperture coupled with the 15-cm distance from the cell to the aperture resulted in a negligible contribution from multiple scattering[14] even at our closest point to critical. Both detector assemblies consisted of a cosine diffuser, a 632.8-nm transmission filter (2 nm width), and a 0.2-cm^2 active area photodiode. The photodiodes were optimized for dc light levels and were amplified by United Detector Technology's 101A amplifier for the reference detector and a model 350 for the transmitted beam detector. These produced an analog voltage which was measured by a $5\frac{1}{2}$-digit voltmeter under computer control to provide average and standard-deviation values of the ratio of the transmitted to reference voltages over 2–3 min. This averaged out any short-term fluctuations in the laser and corrected for laser power variations. Typical resolution was ±0.3% in the ratio.

D. Procedure

To obtain a data point, the temperature controller was set and the bath allowed to come to temperature equilibrium as monitored by the ac bridge. The cell was moved out of the light beam, yet remained in the bath while measurements of the ratio of the intensity transmitted through the bath to the reference were collected. Several ratio averages were obtained, and agreed within our 0.3% precision. The cell would then be oriented perpendicularly to the beam. The orientation was done using the angular scale affixed to the bath and by aligning the vertically scribed mark; the error in this alignment caused less than 0.06% uncertainty in τ. Several averages of the ratio of

TABLE I. Turbidity data in methanol-cyclohexane near its critical point. T is the temperature in degrees Celsius, τ is the turbidity of the fluid mixture, τ_b is a background turbidity that can be calculated, and $\delta\tau$ is the standard deviation of the turbidity at that temperature. The data were taken in the order listed during a three-week period. $T_c = 45.944\,^\circ\text{C}$.

T (°C)	$\tau+\tau_b$ (cm^{-1})	$\delta\tau$ (cm^{-1})
46.114	0.157	0.003
46.041	0.251	0.004
46.028	0.270	0.004
46.014	0.312	0.005
46.007	0.334	0.005
46.000	0.357	0.006
45.992	0.385	0.005
45.986	0.424	0.006
45.979	0.468	0.006
45.972	0.520	0.005
45.965	0.596	0.007
45.960	0.649	0.009
45.9564	0.719	0.007
45.9533	0.808	0.007
45.950	0.940	0.011
46.001	0.37	0.010
45.999	0.351	0.008
45.972	0.519	0.006
45.9505	0.909	0.014
45.9495	0.965	0.009
45.951	0.914	0.009
45.9481	1.100	0.008
46.067	0.204	0.007
46.065	0.208	0.007
46.108	0.168	0.007
46.175	0.131	0.006
46.247	0.092	0.003
46.386	0.067	0.003
46.597	0.049	0.003
46.811	0.042	0.003
48.812	0.021	0.003

intensities with the cell in place were obtained.

These averages had standard deviations of 0.3% and would agree with each other unless the fluids were within 10 mK of the critical temperature. When close to critical, the error signal from the lock-in amplifier in the temperature monitoring ac bridge was monitored by the computer acquisition system. The variation in turbidity of the binary fluid mixture could then be correlated to the temperature variation of the bath and allow a more precise determination of the turbidity and its corresponding temperature from critical. Due to the 20-min period in the bath's temperature oscillation, all of the data obtained close to T_c showed the fluids in quasistatic equilibrium with the bath.

At least a half hour was allowed between data points. Data were taken over several weeks while the temperature was both increased and decreased. No systematic variations due to time or temperature step was observed. The data are shown in Table I in the order they were taken. The error appearing next to the turbidity is the experimental standard deviation from the light intensity measurements. The determination of the critical temperature and the background turbidity τ_b will be discussed in the next section.

III. ANALYSIS AND DISCUSSION

By measuring the ratio of transmitted to reference beam intensities, both with and without the cell in place, the effects from the bath fluid and windows could be determined. The only unknowns were the reflections due to the cell's windows, the turbidity of the binary fluid mixture, and the scattering from the water displaced by the cell. Because the bath was filtered, it was felt this last effect could be neglected when compared to our experimental uncertainty. However, a parameter was still allowed to enter in our calculations of the fluid's turbidity that would include this effect and the effect of the reflection from the cell windows. Both these effects are small and can be safely approximated as an additional term τ_b appearing in

$$\tau = -\frac{1}{L}\ln(I_1/I_2) - \tau_b \ . \tag{8}$$

Here I_1 represents the ratio of transmitted to reference intensities with the cell in place, I_2 is the ratio of transmitted to reference intensities without the cell in the beam path, and τ is the turbidity of the binary liquid mixture. τ_b would be a constant determined by the length of the light path in the cell and the two effects discussed above. It should be noted that by using a wide beam, cosine diffusers on the detectors, and thin quartz windows on the cell and bath, any possible interference patterns in the transmitted beam were reduced to an unmeasurable level.

The contribution to τ_b from the reflection off the cell windows can be calculated[18] knowing the refractive indices of the cell walls, water, and the critical fluid mixture. The latter two have been measured [1.329 for water[19] and 1.379 for the near-critical binary fluid mixture methanol-cyclohexane[12] at this temperature (46 °C) and wavelength (632.8 nm)]. The refractive index of the glass walls was provided by the manufacturer as 1.52. These allow the contribution to τ_b due to reflection to be calculated as 0.013_6 cm^{-1}. As discussed below, this value agrees with the fits of the theory to the data and indicates that, indeed, the scattering due to the displaced water is negligible. We further tested this by placing a duplicate *empty* cell in the beam and found the enhanced loss due to reflection to be $\tau_b = 0.08_3 \pm 0.01$ which compared well with the calculated $\tau_b = 0.094 \pm 0.0003$. Thus, the background contribution to the turbidity can be well determined by this experimental procedure. Knowing τ_b allows other parameters to be better determined.

The temperature at which two phases formed and the location of the meniscus were both monitored. As stated previously, the meniscus appeared in the center of the cell which indicates our sample was very close to the critical composition. The transition temperature, where the fluid system goes from one to two phases, should then be close to the system's critical temperature T_c.[17] The transition temperature was observed to lie between 45.941 and 45.948 °C. The critical temperature was determined from fitting the theory to the data (as described below) to be $T_c = 45.944$ °C. This value is consistent with other determinations in this system considering the small amount of impurities present in the components.[12,13]

The data shown in Table I can be analyzed using a properly weighted least-squares fit where the parameters τ_b, T_c, τ_0, ξ_0, ν, and γ can all be adjusted in Eqs. (6) and (8). A properly[20] weighted fit utilizes both the experimental uncertainty in the turbidity and the propagated uncertainty in the temperature away from critical $(T - T_c)$. These two errors are combined in quadrature to provide the error in the turbidity used in the weighted fit. To allow ν and γ also to vary, a nonlinear fit described in Bevington[21] and used by us and others was performed on a microcomputer. This fit is very reliable when order-of-magnitude starting values for the parameters are provided; these starting values were easy to obtain.

Since the parameters ν and γ are well known theoretically and experimentally, those values were fixed at their predicted values (0.63 and 1.241, respectively) in the early fits. The parameters τ_0 and ξ_0 were allowed to vary in all the fits. τ_b and T_c were also well determined experimentally, yet allowed to vary within a small range to find the optimal value. These two parameters act independently at opposite reduced temperature extremes as can be seen in Fig. 2. The background turbidity τ_b was determined by performing successive fits and finding the minimum in the reduced chi-square while varying τ_b; the resulting value was 0.013 ± 0.001 which agreed with the value calculated from reflection losses in the cell described above. The critical temperature T_c was determined in a similar fashion and resulted in a value of $T_c = 45.944 \pm 0.001$ °C. The error in $(T - T_c)$ was thus taken to be 0.002 K when performing the remaining fits. These values of T_c and τ_b were used in subsequent fits when ν and γ were also varied and when combined data sets were fit as will be discussed shortly.

Various fits to the data in Table I are shown in Table II. Fit 1 had ν, γ, τ_b, and T_c fixed and provided a very good fit to the data with $\tau_0 = (6.90 \pm 0.11) \times 10^{-6}$ cm^{-1}

and $\xi_0=(3.24\pm0.06)$ Å where the error estimates are one standard deviation. These parameters were used to calculate the solid line in Fig. 2. As can be seen, the line not only describes the data reported in Table I, but also extrapolates to provide excellent agreement with the Kopelman *et al.*[16,22] data very close to T_c. The Kopelman *et al.* data are plotted using the τ_b and T_c values reported in their article.[16] This same line also agrees with Eden's[23] data as is shown in Fig. 2. The values we find for τ_0 and ξ_0 differ from Kopelman *et al.* ($\tau_0=9.62\times10^{-6}$ cm^{-1} and $\xi_0=3.87$) when ν and γ are fixed identically. As they suggest,[16] the parameters τ_0 and ξ_0 become decoupled only much further from T_c [see the limit of Eq. (6)] than their experiment could probe. By taking data further from T_c and determining τ_b, we feel our data better determine τ_0 and ξ_0. However, these parameters are also coupled to the

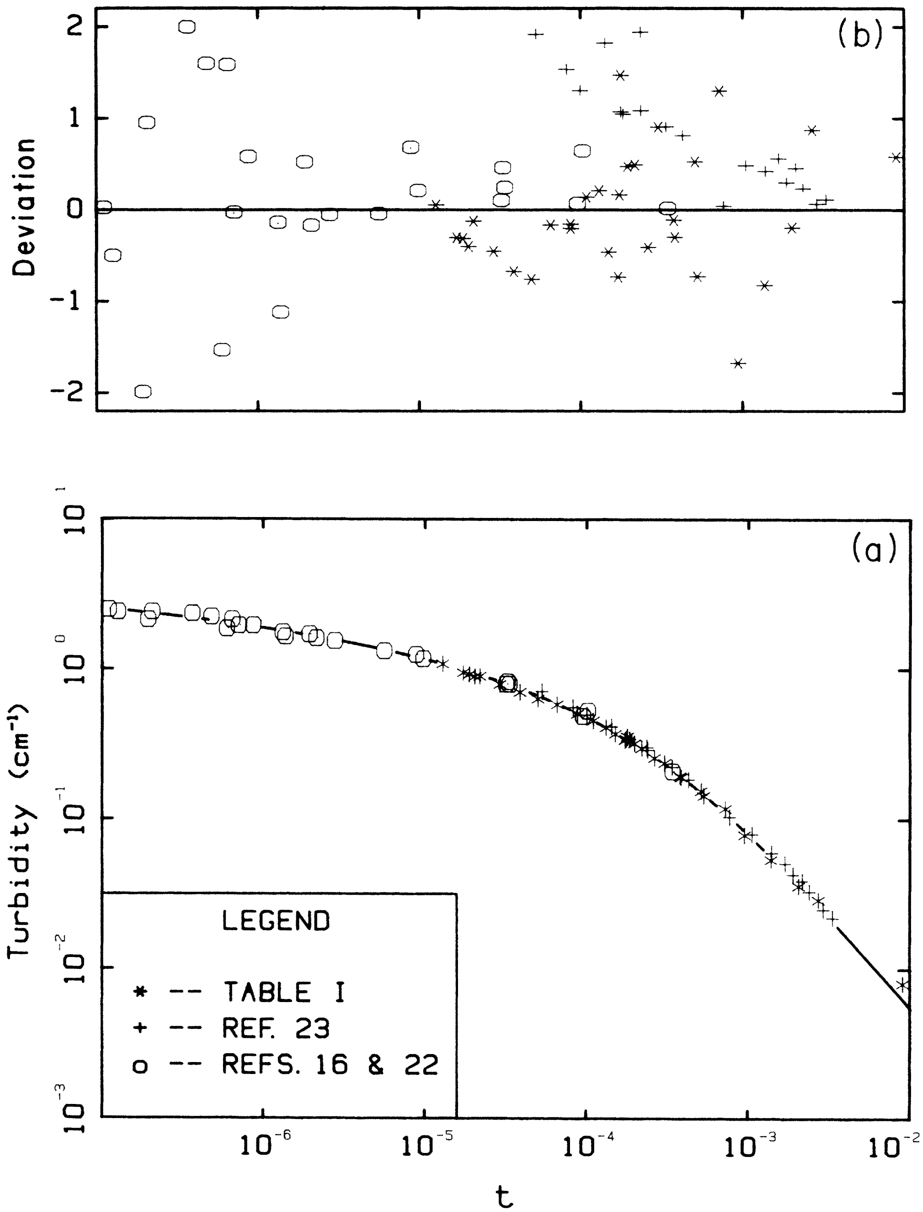

FIG. 2. (a) The lower plot shows the turbidity τ of the binary fluid mixture methanol-cyclohexane as a function of reduced temperature $t\equiv(T-T_c)/T_c$. Our data are from Table I and shown as $*$. The solid line is the best fit (fit 1 in Table II) to our data giving $\xi_0=3.24$ Å and $\tau_0=6.9\times10^{-6}$ cm^{-1} when ν and γ are fixed at 0.63 and 1.241, respectively. This line also extrapolates to fit the data from Refs. 16, 22, and 23 quite well. (b) The upper curve shows the deviation, in units of the error on each point, of this solid line from the data point. The systematic deviation of Eden's data (Ref. 23) from the line is most likely due to uncertainties in the critical temperature T_c and the background turbidity τ_b, neither of which were adjusted. Data from Refs. 16 and 22 and our data fall randomly about the line which was the best fit to our data.

TABLE II. The values of parameters used in fitting turbidity data in the binary fluid mixture methanol-cyclohexane. Data set A is our data reported in Table I with $T_c = 45.944$ and $\tau_b = 0.013$, set B is from Refs. 16 and 22, while C is from Ref. 23. Numbers in parentheses are fixed in a given fit. χ^2/N is the reduced chi-square as defined in Ref. 21. All fits were properly weighted as described in the text and determine the one standard deviation errors quoted.

Fit	Data set	ν	γ	$10^6\tau_0$ (cm^{-1})	ξ_0 (Å)	χ^2/N
1	A	(0.63)	(1.241)	6.90±0.11	3.24±0.06	0.46
2	A	(0.63118)	(1.23786)	7.01±0.11	3.17±0.06	0.45
3	A,B,C	(0.63)	(1.241)	6.98±0.09	3.24±0.04	0.69
4	A,B,C	0.64±0.02	1.25±0.05	6.4±1.9	3.0±0.2	0.69
5	A,C	0.64±0.02	1.26±0.05	6.1±1.9	3.1±0.3	0.64
6	A,C	(0.63)	(1.241)	6.88±0.11	3.22±0.04	0.62

choice of ν and γ and as the predicted values of these change, so would the values of τ_0 and ξ_0.

To test this, a second fit (fit 2 in Table II) at slightly different values of ν and γ (values recently calculated by George and Rehr[24]) cause a small shift in τ_0 and ξ_0 yet illustrate the coupling of the parameters. To better determine the values of ξ_0, τ_0, ν, and γ, several fits were attempted on combined sets of turbidity data. In using the Kopelman *et al.*[16,22] and Eden[23] data, no attempt was made to adjust T_c or τ_b in any of the sets of data; the experimental errors for τ and T_c were taken to be 0.1 cm^{-1} and 3×10^{-6} K in the Kopelman *et al.* data, and 0.01 cm^{-1} and 2 mK in Eden's data. Table II illustrates the parameter values when ν and γ were fixed (fit 3) when ν and γ were allowed to very (fits 4 and 5). Although fit 3 gives very precise values for τ_0 and ξ_0, fits 4 and 5 show the interdependence of the parameters and the resulting uncertainty in the parameter values.

Despite the coupling of the parameters, it is still possible to provide the best determination of ξ_0 for the binary mixture methanol-cyclohexane: $\xi_0 = 3.24 \pm 0.2$ Å. This value encompasses all the values found in the fits in Table II when using ours as well as other researchers' turbidity data.

Other determinations of ξ_0 for methanol-cyclohexane have been done in various ways. Sorensen *et al.*[14] found $\xi_0 = 2.38$ Å, $\nu = 0.654$, and $\gamma = 1.037$ when applying a dynamic-droplet-model to Rayleigh linewidth data; mode-mode coupling theory gave $\xi_0 = 2.85$ Å and $\nu = 0.627$ when applied to the same data. When fitting refractive index anomaly data, Hartley *et al.*[25] found $\xi_0 = 4.7$ Å when ν was fixed at 0.60. Yosida[26] investigated 90° light scattering data and found $\xi_0 = (4.1 \pm 0.5)$ Å, $\nu = 0.65 \pm 0.03$, and $\gamma = 1.27 \pm 0.06$. Finally, Kopelman *et al.*[16] used turbidity measurements in an extremely thin cell very close to T_c to find $\xi_0 = 3.87 \pm 0.97$ Å and $\tau_0 = 9.62\times10^{-6}$ cm^{-1} when ν and γ were fixed at 0.63 and 1.241, respectively.

Two-scale-factor universality can be tested in two ways. Kopelman *et al.*[16] calculated a value of $\xi_0 = (3.47 \pm 0.35)$ Å assuming two-scale-factor universality as well as estimating a value for the heat-capacity amplitude A corresponding to $\alpha = 0.110$ for data taken and analyzed by Anisimov *et al.*[15] This value of ξ_0 agrees with our determination.

A separate test of two-scale-factor universality can be done with the results of the analysis of this turbidity data combined with our previous measurements[12,13] of the coexistence curve in this system. The coexistence curve was measured by a refractive index technique and fit to $n_u - n_L = B' t^{\beta}$ where n_u (n_L) is the refractive index of the upper (lower) phase and B' is the measured amplitude which was found[13] to be $B' = 0.143 \pm 0.002$ (with $\beta = 0.325$). B' can be related to B in Eq. (5) by $B = (B'/2)(\partial\phi/\partial n)$ where $\partial\phi/\partial n$ can be calculated[12] from the Lorentz-Lorenz relation. The amplitude $k_B T_c \chi_0$ appearing in Eq. (5) can be determined from τ_0 using Eq. (7). Thus Eq. (5) can be rewritten in terms of directly measurable quantities:

$$R = \xi_0 \left[\frac{(B' n_c)^2 \pi^3}{\lambda_0^4 \tau_0} \right]^{1/3} , \tag{9}$$

where the term $\partial\phi/\partial n$ cancels out since $\partial n^2/\partial\phi = 2n_c \partial n/\partial\phi$. Since the theoretical predictions for R correspond to predicted values of the exponents,[7] a test of Eq. (9) should use amplitudes determined when the exponents were fixed at those values. Thus, Fits 1, 3, and 6 in Table II were used to determine τ_0 as 6.94 ± 0.11 and ξ_0 as 3.23 ± 0.06 (one standard deviation errors). Using these in Eq. (9) gives a value of $R = 0.71_5 \pm 0.02_4$. This value is higher than the predicted value of 0.65—0.67, but within three standard deviations. Other experimental values range from 0.68 to 0.69 in one-component systems,[7] and from 0.47 to 0.86 in binary mixtures.[8]

IV. SUMMARY

By taking transmitted light intensity data in the one-phase region of the binary liquid mixture methanol-cyclohexane, the turbidity could be well determined in the reduced temperature range $10^{-5} < t < 10^{-2}$ with the background turbidity being calculable. When the critical exponents ν and γ were fixed at 0.63 and 1.241, respectively, the best fit to our data gave $\xi_0 = 3.24$ Å and $\tau_0 = 6.9\times10^{-6}$ cm^{-1}. These parameters are consistent with all known turbidity data on this system. By combining our data with Kopelman *et al.*[16,22] the critical exponents ν and γ were found to be 0.64 ± 0.02 and 1.26 ± 0.05. These exponent values agree with predicted values of $\nu=0.630$ and $\gamma=1.241$. The value we determine for ξ_0 and τ_0 were combined with coexistence curve data to calculate the universal amplitude ratio $R=\xi_0(B^2/k_B T_c X_0)^{1/3}=0.71_5\pm0.02_4$ which is higher than the predicted range of 0.65—0.67, but does not rule out either value. More accurate data on the heat capacity in this mixture would be useful to further test two-scale-factor universality.

Note added in proof. A similar experiment on methanol-cyclohexane to the one described here has recently appeared.[27] They find $\xi_0=3.24\pm0.23$ and $\tau_0=(6.74\pm0.53)\times10^{-6}$ cm^{-1} which agrees with our results.

ACKNOWLEDGMENTS

I appreciate helpful discussions with R. B. Kopelman, R. W. Gammon, M. R. Moldover, J. V. Sengers, J. Shanks, and H. C. Burstyn. The effort by K. Gruner in developing the optical system was particularly helpful. This work was supported by the National Science Foundation under Grant No. CHE-83-18352.

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