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Critical Behavior in Quaternary Microemulsion Mixtures: Experimental Determination of Universal Scaling Laws

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We present light scattering, density, and interfacial tension results obtained for a four-component system in two distinct points of a critical line. The system consists of mixtures of water, dodecane, pentanol, sodium dodecyl sulfate. The experiments have been performed at constant temperature in both the one-phase and two-phase regions along different paths. For each critical point investigated, effective critical exponents ν , γ , β , and μ were measured. Although their values are smaller than the Ising one, they verify with a reasonably agreement the following theoretical predictions: $\gamma = 2\nu$, $\mu = 3.88\beta$, and $\mu + \nu = \gamma + 2\beta$. The new results reported in this paper show that the choice of the path followed cannot explain the anomalous behavior observed. In contrast they are consistent with the recent interpretation proposed by Gazeau et al. (*Europhys. Lett.* 1989, 9, 833) of a crossover between two critical phenomena.

I. Introduction

Over the past 15 years or so, a great deal of progress has been made experimentally and theoretically in understanding critical phase separation in fluids. The general concepts of scaling and universality are well established and renormalization group calculations of critical exponents are generally in good agreement with experimental values.¹⁻⁵ The best characterized systems are pure fluids near their liquid-vapor critical points and binary fluids near liquid-liquid consolute point. Both types of critical points belong to the same universality class as the three-dimensional Ising model. For mixtures with three or more components, liquid-liquid phase separation seems also to be relevant of the Ising model.⁶⁻¹³

More recently, the study of critical phenomena in fluids has been extended to binary micellar solutions and multicomponent microemulsion systems.¹⁴⁻¹⁶ The aim of these investigations in

surfactant solutions was to point out differences if they exist between these critical points in these organized mixtures and the liquid-gas critical points of a pure compound. In most of the surfactant solutions the critical exponents are in agreement with those found in pure fluids or usual binary mixtures. However, for three particular cases deviations from the Ising values have been reported.^{16-20,35-38}

A comprehensive light scattering study of aqueous micellar solutions of poly(oxyethylene)nonionic amphiphile $C_{12}E_8$ including temperature, concentration and chain length dependence of the surfactant has been performed by Corti and Degiorgio.¹⁶⁻¹⁹ They found that in these binary mixtures the critical exponents ν and γ depend on the surfactant and the solvent. In particular, for the mixture $C_{12}E_8$ -H₂O the critical exponents are markedly influenced by the isotopic substitution in the solvent and also by the addition of salt. Replacement of pure water by pure D₂O results in an increase of the exponents ν and γ from $\nu = 0.44$ to $\nu = 0.58$ and $\gamma = 0.88$ to $\gamma = 1.20$. The variation of ν and γ is then going from the universal 3D Ising values to values smaller than the meanfield predictions. These results have elicited intense theoretical interest.³⁹⁻⁴² Very recent light scattering measurements have been performed by Dietler and Cannell for $C_{12}E_8$ in D₂O and H₂O.⁴³ Clearly their results for ν and γ are in agreement with the 3D

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Ising values regardless of which solvent is used. On the basis of their data they conclude that this behavior is completely consistent with current theory in the form used to understand the behavior of simple usual molecules. Their experiments suggest that the difference between their results and those of Corti and Degiorgio lies on the sample. This reduces the possibility of known non-Ising cases to multicomponent microemulsions. The rather large value of $\nu = 1.13$ found by Dorshow et al.²⁰ for a five-component microemulsion system has been explained by Sorensen⁴⁴ in terms of an approach to a double critical point. The last known case of anomalous critical behavior has been reported by Bellocq et al.³⁵⁻³⁸ for the quaternary system sodium dodecyl sulfate (SDS)-pentanol-water-dodecane. Their results have provided evidence for a continuous variation of effective critical exponents ν and γ from 3D Ising values to $\gamma = 0.42$ and $\nu = 0.21$ when approaching the critical end point of a critical line. Recently it has been observed that the system exhibits a substantial critical anomaly of the refractive index in the vicinity of the critical end point.⁴⁵ This anomaly cannot be characterized satisfactorily in terms of power law expressions. In view of the anomalous critical behavior of the correlation length and the osmotic compressibility (exponents ν and γ), it appears of interest to characterize the behavior of other properties of the system. In the present work we have investigated how the interfacial tension between the coexisting phases and the difference of density of these phases vanish in various points of the critical line P_C^L . The aim of these experiments was to determine the corresponding critical exponents μ and β and to check if the scaling laws which relate ν , γ , β , and μ are valid all along the critical line P_C^L .

In section II of the paper we briefly recall some general considerations on the critical behavior in multicomponent mixtures. In section III we outline the experimental techniques involved in measuring the critical exponents. In section IV we present our results.

II. General Considerations

The first major feature of critical phenomena is that sufficiently close to a critical point the equilibrium properties A vary as simple power laws of the deviations of the field variables from their critical values.

$$A = A_0 \epsilon^\lambda \quad \text{with} \quad \epsilon = \frac{(X - X_C)}{X_C} \quad (1)$$

In this equation λ is a critical exponent and the coefficient A_0 defines the amplitude of the divergence. The variable X is a field variable and X_C its value at the critical point. The fields are the variables such as temperature, pressure, or chemical potential of the species (μ_i) which have equal values in the coexisting phases. The exponents ν , γ , β , and μ are respectively characteristic of the correlation length, the osmotic compressibility χ , the difference of density $\Delta\rho$ of the coexisting phases, and the interfacial tension σ between phases in equilibrium.¹⁻⁵

The second major important feature of critical phenomena is the concept of scaling. The critical exponents are not independent. There exists relations among the exponents called scaling relations. In particular,

$$\mu + \nu = \gamma + 2\beta \quad (2)$$

Finally the third important feature of critical phenomena highlighted by modern theories is the concept of universality. According to this, the critical exponents associated with the singularities are identical for all the systems within a given universality class. Pure fluids and fluid mixtures near normal critical points belong in the universality class of the three-dimensional Ising model. The renormalization group methods have yielded a detailed and accurate description of the critical thermodynamic behavior of such Ising-like systems. The Ising values for the exponents ν , γ , β , and μ are the following: $\nu = 0.630 \pm 0.001$, $\gamma = 1.240 \pm 0.002$, $\beta = 0.325 \pm 0.001$, $\mu = 1.260$.

The critical exponents are defined along precise paths. As an example, the definition of γ for a pure fluid applies to the path of density equal to the critical density. As pointed out by Griffiths and Wheeler (GW),⁹ the characteristic of this special path is that it is asymptotically parallel to the coexistence line in the space of fields. Along any other path, the exponents become renormalized. GW have generalized these considerations to the case of multicomponent mixtures. They predict that the critical behavior for mixtures will be essentially the same as in pure fluids provided that the critical exponents are measured along equivalent directions in the space of independent variables in both systems. For a system made of n components, a situation equivalent to the case of the pure fluid will be obtained by holding $n - 1$ fields constant. For the systems with $n > 2$, there is in addition along and intersecting the coexistence surface a new way of approaching a critical point, namely parallel to the critical space. GW have predicted that the form of the divergence is determined by the orientation of a path L of approaching a critical point Y with respect to the coexistence surface on one hand and to the critical surface on the other hand. These authors have considered one of the paths of interest is the path asymptotically parallel to the coexistence surface but not parallel to the critical surface. Along such a path one expects to measure exponents characteristic of the phase transition.

Therefore, one of the main difficulties encountered in the study of a multicomponent mixture is to find an appropriate path to approach the critical point. This means that it is necessary to control in addition to temperature and pressure one or more chemical potentials. The detailed experimental study of the phase diagrams of ternary and quaternary microemulsion systems has provided evidence that in the oil-rich part of the phase diagram the ratio X of the water and surfactant concentrations takes the same value in the coexisting phases.³³⁻³⁸ The major consequence of this remarkable property is that the section $X = \text{constant}$ of the phase diagram of the quaternary system can be considered as real ternary systems since the tie-lines and the tie-triangles corresponding to the two and three-phase equilibria are lying in the X section.

The assimilation of X with a field variable in reducing the number of variables of the system leads to a situation equivalent to that of a ternary mixture. In practice, the critical behavior of a ternary mixture is investigated by varying temperature at fixed composition. This leads to a renormalization of the critical exponents which has been considered by Fisher.⁷ The renormalized values of the exponents are 10% higher than the Ising values. They are multiplied by the coefficient $1/(1 - \alpha)$ with $\alpha = 0.11$ (α being the exponent of the specific heat).

The possibility of approaching the critical point of a ternary mixture at fixed pressure and temperature has been studied by Widom and Wheeler^{6,8} who have shown that in this case the critical behavior is identical in character with that in a binary liquid mixture of fixed temperature but variable pressure. They show that as the plait point is approached along the line which is the extension into the one-phase region of the rectilinear diameter of the binodal curve the osmotic compressibility diverges as the $-\gamma/(1 - \alpha)$ power of the distance from the plait point.

III. Experimental Part

III.1. System Phase Diagram. The system studied consists of water-dodecane-pentanol-SDS. A detailed description of its complex phase diagram has been given elsewhere.³⁶ One of the important features found for this system at constant temperature is a critical line which connects a critical end point P_C^E located in the oil rich part of the diagram with a critical point P_C^A located in the limiting ternary face of the system which does not contain oil. A schematic representation of this critical line P_C^L is given in Figure 1, in the space alcohol concentration, dodecane concentration ratio X (X being the ratio between the water and SDS weight fractions). Figure 2 represents two sections of the phase diagram obtained by holding the X ratio constant. In the following, these sections are referred to as an X section. The X values (expressed in weight) corresponding to P_C^E and P_C^A are respectively

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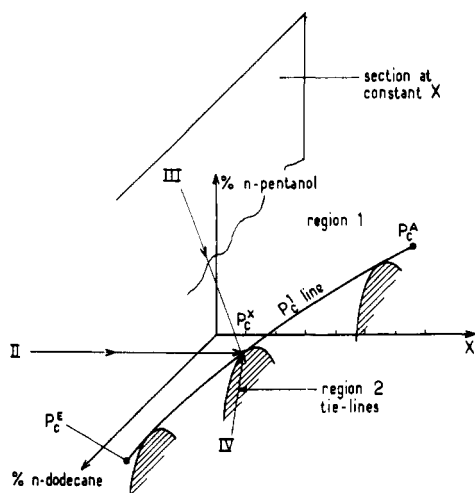


Figure 1. Schematic representation of the critical line P_C^I and of the paths of approach of a critical point P_C^X at fixed temperature (X is the ratio between the water and SDS concentrations). Region 1, microemulsion phase; region 2, two-phase region where two microemulsions are in equilibrium. Path II, approach at variable X ; paths III and IV, approaches in a section at constant X respectively along a direction perpendicular to the tie-lines and in the two-phase region 2.

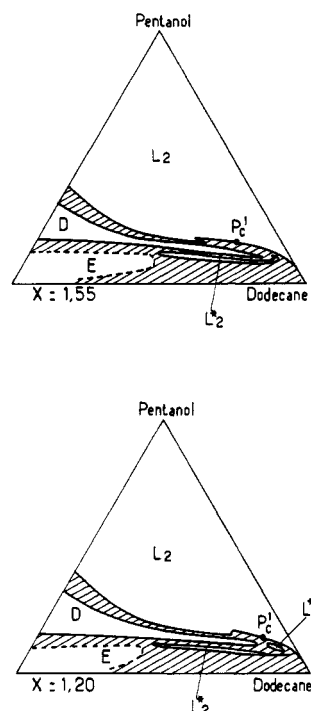


Figure 2. Sections $X = 1.55$ and $X = 1.20$ of the phase diagram of the water-dodecane-pentanol-SDS system at 21 °C. L_2 , microemulsion phase; L_2^* , flow birefringent phase (isotropic at rest); D, lamellar phase; E, hexagonal phase. P_C^I critical point. Hatched domains: multiphase equilibria.

0.95 and 6.6. Any X plane defined by a X value between 0.95 and 6.6 intercepts the critical line at one critical point labeled P_C^X . The critical line is located at the boundary between a microemulsion domain (region 1) and a two-phase region where two microemulsions are in equilibrium (region 2).

Composition analysis of the phases in equilibrium in region 2 has shown that the corresponding tie-lines are located in an X section. Therefore, the pseudoternary diagrams shown in Figure 2 can be considered as ternary ones. In the sections $X = 1.2$ and $X = 1.55$ one identifies in the very oil rich region three distinct phases, L_2 , L_2^* , and D. The isotropic phase L_2 is the classical microemulsion phase, i.e., a dispersion of water droplets in oil. The structure of the flow birefringent phase L_2^* has been recently elicited.⁴⁶ It consists of a sponglike random bilayer continuous

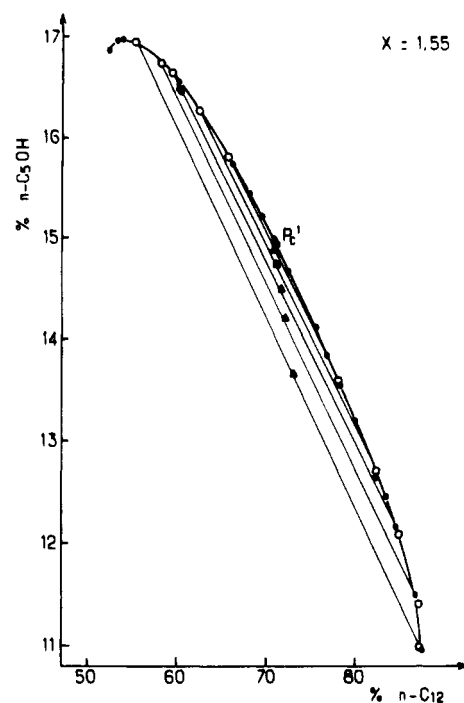


Figure 3. Magnification of the two-phase region 2 around the critical point in the section $X = 1.55$. The segments represent the tie-lines. The filled circles correspond to the visual determination of the coexisting curve, the triangles represent the composition of the overall mixtures, and the open circles represent the ends of the tie-lines. Their positions were deduced from density measurements.

surface separating two oil-continuous regions. The D phase is an oily swollen lamellar phase. One should note that the extent of the region 2 where two microemulsions L_2 and L_2^* are in equilibrium increases with X .

III.2. Determination of the Tie-Lines and Novel Paths of Approach to the Critical Points. In the previous work the critical behavior was observed in several points of the critical line P_C^I .^{35,38} The critical points were approached either by raising the temperature at fixed composition (path I) or by increasing the water to surfactant ratio X at fixed temperature (path II). In this paper we have approached the critical points $X = 1.55$ and $X = 1.207$ along two novel paths (paths III and IV) at constant temperature and pressure. Path III corresponds to an approach of the critical point in the single-phase region L_2 along a straight line perpendicular to the tie-lines and path IV to an approach in the two-phase region 2. The approach along path III requires the determination of the tie-lines in the vicinity of the critical point; this has been done using the densitometry method described by Roux et al.⁴⁷ for the ternary mixture water-pentanol-SDS. In a first step, the coexistence curve limit of the region D_2 in a given X section is accurately located at fixed temperature. A series of samples located along lines cutting the coexistence curve are prepared and equilibrated for several days in a chamber at constant temperature. Then, for each line the density of the last monophasic sample is measured. These data give the variation of density as a function of either dodecane (x_0) or pentanol (x_A) concentrations along the coexistence curve. In a second step, two-phase equilibria are prepared in the region 2 and the density of the both coexisting phases is measured. Using the curve $d = f(x_0)$ or $d = f(x_A)$ we determine the locations of the two conjugate phases (points α and β). Obviously if the points α and β are along the demixing curve in the plane X -pentanol-dodecane, the tie-line $\alpha\beta$ contains the point representative of the overall composition. Figure 3 gives the locations of the tie-lines and those of the overall points for the section $X = 1.55$. The data confirm that the ratio X has the

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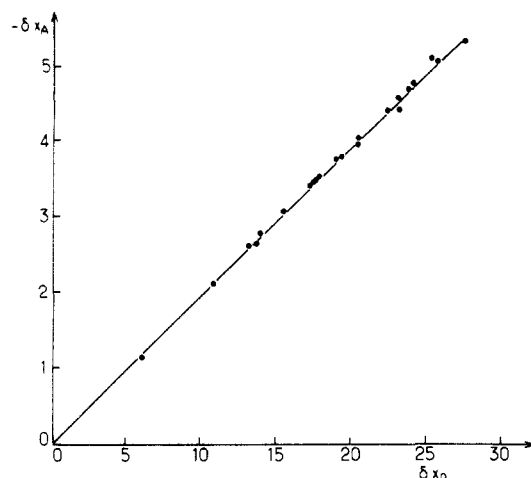


Figure 4. Difference of the pentanol concentration δx_A between the two phases in equilibrium in region 2 ($X = 1.55$) versus the difference of the dodecane concentration δx_0 in the same two-phase equilibrium. The linear behavior of these two differences indicate that the tie-lines are parallel.

same value in the phases in equilibrium. Around the critical point, the tie-lines seem to be parallel; this remark was already pointed out for several ternary mixtures.^{10,12} Indeed the experimental data show that δx_0 varies linearly with δx_A (Figure 4)

$$\delta x_0 = -A\delta x_A \quad \text{with} \quad A = 0.192 \quad (3)$$

$$\delta x_0 = x_0^1 - x_0^2 \quad \text{and} \quad \delta x_A = x_A^1 - x_A^2 \quad (4)$$

where x_0^j and x_A^j are respectively the dodecane and pentanol concentrations in the upper phase ($j = 1$) and the lower phase ($j = 2$).

For each two-phase equilibrium, the function $Y(x_0, x_A)$ is conserved in the two phases 1 and 2

$$Y(x_0^1, x_A^1) = Y(x_0^2, x_A^2) \quad \text{with} \quad Y(x_0, x_A) = Ax_A + x_0 \quad (5)$$

The variable Y takes the same value in the phases in equilibrium, and it behaves as a field variable. We have used this variable, in a similar way to the ratio X , to approach the critical point. Measurements have been performed along path III. This path has been chosen parallel to Y and contains the critical point P_C^X studied. The relations 3, 4, and 5 allow us to define the direction Y , perpendicular to the tie-lines, the angle α between the direction Y and the alcohol axis being given by $\tan \alpha = -1/A$. For each sample, the distance ϵ_Y to the critical point is

$$\epsilon_Y = \frac{Y - Y_C}{Y_C}$$

Y and Y_C are respectively the values of the function Y (see relation 5) at the point considered and at the critical point. Because of the geometry of the tie-lines one can also write

$$|Y - Y_C| = \frac{|x_0 - x_0^C|}{\sin \alpha} = \frac{|x_A - x_A^C|}{\cos \alpha}$$

In these relations x_0 and x_A are the dodecane and pentanol concentrations and x_0^C and x_A^C their values at the critical point. Therefore, in the following the distance to the critical point has been taken as

$$\epsilon_0 = \left| \frac{x_0 - x_0^C}{x_0^C} \right|$$

The measurement of a critical exponent, as the variable used to approach the critical point is a concentration, requires the preparation of a large number of samples close in composition to the critical concentration. That has been carried out by mixing two solutions (one L_2 and one L_2^*) located along the path III. This method allows us to determine the critical concentration (x_0^C, x_A^C) in a concentration range δx defined by the concentrations

TABLE I: Critical Compositions (in wt %) of the Points A and B

	$T_C, ^\circ\text{C}$	X_C	water	SDS	dodecane	pentanol
A	21.36	1.55	7.160	4.620	73.535	14.685
B	21.40	1.20	5.983	4.957	75.955	13.105

of the last monophasic sample and the first diphasic one. One should note that the accuracy on the distance ϵ to the critical concentration is less good as the temperature is used as variable.

III.3. Methods. Bulk light scattering measurements were undertaken in the single phase in order to determine the values of the exponents ν and γ . The samples were held in cylindrical glass cells ($\phi = 8$ mm) and temperature was controlled to within ± 0.01 $^\circ\text{C}$. Measurements of the intensity of the light scattered were performed with an AMTEC setup equipped with Krypton ion laser operating at $\lambda = 6471$ \AA . The angular range covered was from 20° to 150° . The system investigated has been chosen by reason of its very weak turbidity in order to avoid the effect of multiple scattering. Turbidity measurements were conducted on the same samples; the values observed are very weak (around 0.1 cm^{-1} close to T_C) and the contribution of multiple scattering to the measured intensity was found negligible for all the samples. In the analysis of our data we have assumed that the density fluctuations are constant over the concentration range investigated. We have considered that the total intensity is due to the critical concentration fluctuations. In all cases the data were well fitted with the Ornstein-Zernike formula which is expected to be valid in the critical region.

$$I(q) = \frac{I(0)}{1 + q^2\xi^2}, \quad q = \frac{4\pi}{\lambda} n \sin \frac{\theta}{2} \quad (6)$$

ξ is the correlation length of the concentration fluctuation, n is the refractive index, and θ is the scattering angle. The extrapolated scattered intensity at zero scattering angle $I(0)$ is related to the osmotic compressibility of the solution.

The measurements of density were performed with a Parr apparatus; The interfacial tensions were measured with the same spinning drop tensiometer built in the laboratory as in previous studies.³¹ For both types of measurements the temperature was controlled to within ± 0.01 $^\circ\text{C}$. These measurements were carried out along path IV which is the continuation in the two-phase region D of the path III.

IV. Results and Discussion

Two critical points labeled A and B respectively defined by $X = 1.20$ and $X = 1.55$ were approached at fixed temperature, in the single-phase region along path III and in the two-phase region along path IV. The critical compositions of A and B are given in Table I. Light scattering measurements were performed in the single-phase region along path III. The experimental data show typical features of critical phenomenon. Both the correlation length ξ and the scattered intensity $I(0)$ are found to diverge at the critical points A and B. Figure 5 shows log-log plots of $I(0)$ and ξ versus the reduced concentration ϵ_0 for the two critical mixtures. The data are well fitted by the power laws:

$$I(0) = I_0\epsilon_0^{-\gamma} \quad (7)$$

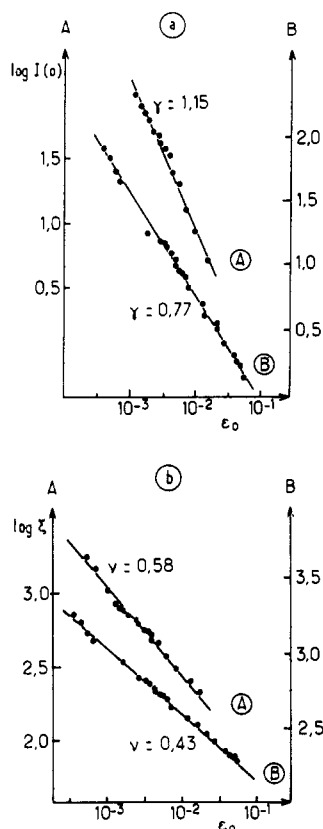
$$\xi = \xi_0\epsilon_0^{-\nu} \quad (8)$$

As ϵ_0 increases above the range presented in Figure 5, a deviation of $I(0)$ and ξ from the linear behavior is found. For both critical points A and B, the smallest values of $I(0)$ are approximately 10 times larger than those measured for the micellar solutions. Therefore, we have attributed the total scattered intensity to critical fluctuations over the entire ϵ_0 range shown in Figure 5. In the fits x_0^C is let free in the concentration range Δx_0 experimentally measured. The values of γ and ν obtained along path III for A and B are given in Table II. They differ from Ising values but are very close to those previously obtained by Honorat et al. along other paths³⁵ (Table II).

In order to determine the values of the exponents β and μ we have measured the density of the coexisting phases in the two-phase

TABLE II: Experimental Values of the Critical Exponents ν , γ , β , and μ Measured for the Two Critical Points A and B along the Paths I–IV

point	path	ν	γ	β	μ
A ($X_C = 1.55$)	I(22)	0.53 ± 0.03	1.01 ± 0.06	0.28 ± 0.03	1.11 ± 0.10
	II(22)	0.57 ± 0.03	1.10 ± 0.06		
	III	0.58 ± 0.05	1.15 ± 0.05		
	IV				
B ($X_C = 1.20$)	I(22)	0.34 ± 0.03	0.63 ± 0.06	0.25 ± 0.03	0.94 ± 0.03
	II(22)	0.38 ± 0.03	0.78 ± 0.06		
	III	0.43 ± 0.03	0.787 ± 0.03		
	IV				

Figure 5. Approach along path III. log-log plots of the total intensity $I(0)$ and the correlation length ξ (Å) vs the reduced variable ϵ_0 for the two critical points A and B.

regions 2 defined by $X = 1.20$ and $X = 1.55$ and the interfacial tension σ between the phases. The Figures 6 and 7 show log-log plots of $\Delta\rho$ and σ versus the reduced variable ϵ_0 . $\Delta\rho$ is the difference between the densities of the lower and upper phases. Both quantities $\Delta\rho$ and σ vanish as one approaches the critical points A and B according to the following power laws:

$$\Delta\rho = \Delta\rho_0 \epsilon_0^\beta \quad (9)$$

$$\sigma = \sigma_0 \epsilon_0^\mu \quad (10)$$

The values of the critical exponents β and μ are smaller than the Ising values (Table II) and depend on the critical point. The values of both exponents β and μ decrease with X .

The critical behavior of the correlation length and the osmotic compressibility for the critical points defined by $X = 1.55$ and $X = 1.20$ has been investigated along three different paths, by using three different variables: temperature, the X ratio, and the dodecane concentration. For each critical point A and B, the values of the two exponents ν and γ found along the three paths are very close and different from the theoretical ones (Table II). They decrease as the X ratio decreases. The values of the critical exponents β and μ measured for the two critical points A and B show the same X dependence as ν and γ . They decrease with X . Having measured for the critical points A and B the four exponents ν , γ , β , and μ it is possible to test several scaling laws. Within the experimental accuracy the obtained numbers of ν , γ , β , and

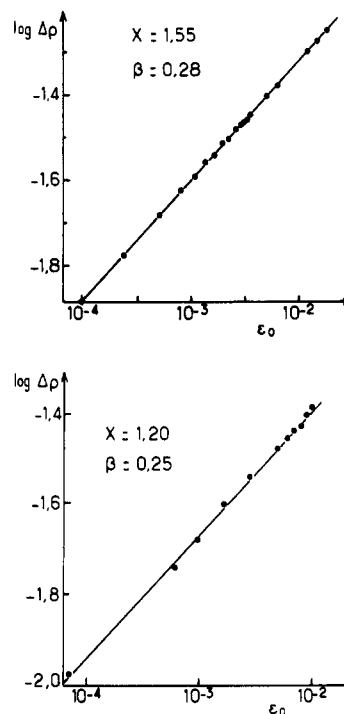
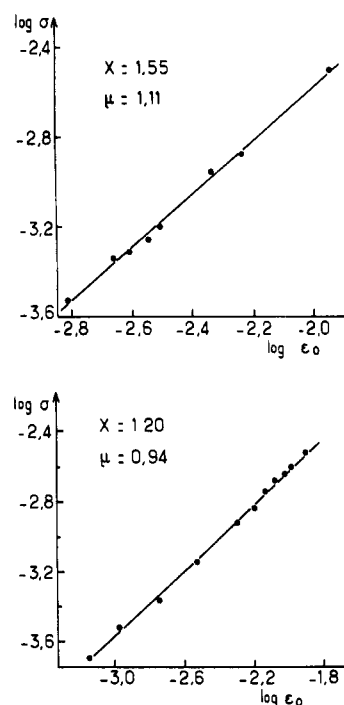
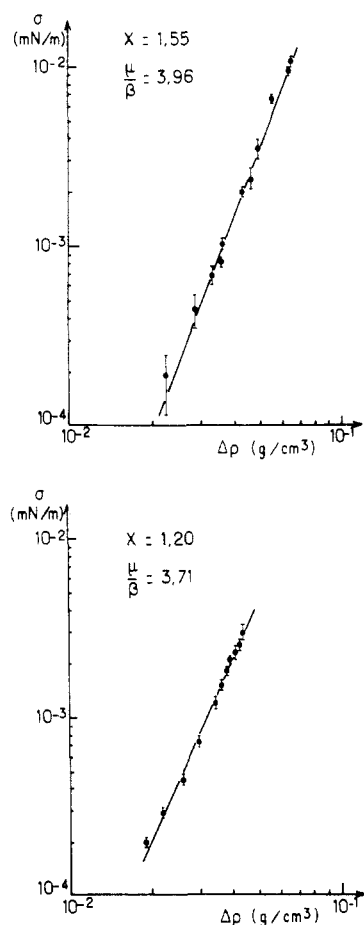
Figure 6. Approach along path IV. log-log plots of the density difference $\Delta\rho$ (g/cm³) vs the reduced variable ϵ_0 for the critical points A ($X = 1.55$) and B ($X = 1.20$).Figure 7. Approach along path IV. log-log plots of the interfacial tension σ (mN/m) versus the reduced variable ϵ_0 for the critical points A and B.

TABLE III: Test of Some Scaling Laws for the Critical Points A and B and Theoretical Ising and Mean Field (MF) Values

X	γ/ν	$(\mu + \nu)/\nu$	$(\gamma + 2\beta)/\nu$	μ/β
MF	2	4	4	3
Ising	1.97	3	3	3.88
1.550	2.0 ± 0.2	2.0 ± 0.3	3.0 ± 0.4	3.96 ± 0.3
1.207	1.8 ± 0.2	3.2 ± 0.4	3.0 ± 0.4	3.76 ± 0.3

**Figure 8.** Interfacial tension versus density difference between the phases for the critical points A and B.

μ satisfy with a reasonable agreement the theoretical predictions (Table III).

As mentioned above, the measurement of the exponents along paths III and IV requires the precise determination of the critical composition. However, it is possible to obtain directly the value of the ratio between μ and β without knowing this concentration by plotting in a log-log scale σ versus $\Delta\rho$. Indeed by eliminating ϵ between the relations 9 and 10 one obtains

$$\sigma = \sigma_0 \left(\frac{\Delta\rho}{\Delta\rho_0} \right)^{\mu/\beta}$$

The slope μ/β of the plots of Figure 8 is in good agreement with the 3D Ising exponents (the mean field value of μ/β is 3). Such behavior was previously found for other systems.^{25-27,32}

In summary, all the new results that we have obtained, power laws, test of scaling laws, confirm that the behavior observed corresponds to critical phenomenon. In a preceding paper³⁵ several possible interpretations were considered to account for the continuous variation of the critical exponents ν and γ . In particular, it was suggested that this behavior could be due to the choice of the specific path followed or to a crossover between two sets of exponents, one of them corresponding to the Ising indices. It was suggested that the second set of exponents should be associated with a "special" critical end point in relation with the existence of the swollen lamellar phase. Our results allow one to definitely eliminate the first hypothesis since an identical variation has been observed along three different paths for the four different critical exponents ν , γ , β , and μ . Recently Gazeau et al.⁴⁸ have performed new light scattering and turbidity measurements in the section of the phase diagram $X = 1.03$. Their data show that the anomalous critical behavior is due as a matter of fact to a crossover between two critical phenomena. Indeed, in addition to the critical point P_C^1 which corresponds to a phase separation between two inverted micellar solutions L_2 and L_2' , Gazeau et al. observe a second critical behavior at the transition between the micellar phase L_2 and the spongelike phase L_2^* . This observation is in agreement with theoretical arguments which state that the "sponge" phase has a particular symmetry resulting from the equivalence of the two continuous media which form it.^{49,50} The sponge to micellar transition breaks this symmetry and corresponds to a first or a second-order phase transition. According to Gazeau et al.⁴⁸ analysis the size of the asymptotic critical domain where the behavior characteristic of the point P_C^1 is expected is very narrow and depends on the distance between the two critical points. Moreover, it appears that the competition between the two critical phenomena produces within some accuracy apparent power laws in the ϵ range investigated (i.e., in the range 10^{-3} – 10^{-1}). The examination of the phase diagram shows that the distance between the two critical points increases with X . Consequently the crossover should be less and less pronounced and effective exponents more and more Ising-like as the X ratio becomes larger. The X dependence of the exponents ν , γ , β , and μ that we have observed are in good agreement with this prediction.

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