

Shear viscosity studies above and below the critical consolute point in a nitrobenzene-decane mixture

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The shear viscosity has been studied in a nitrobenzene-decane critical mixture above the critical consolute temperature T_C , in the homogeneous phase, and below T_C , in coexisting phases. The form of background viscosity for coexisting phases has been postulated. The same value of the critical exponent ϕ has been obtained in the lower (L), upper (U), and homogeneous (H) phases. The pretransitional amplitudes ($A^{L,U}$) in coexisting phases are approximately the same, whereas $A^{L,U}/A^H \approx 0.965$. In the homogeneous phase the possibility of the appearance of the quasinematic, field-induced structure of critical fluctuations has been discussed.

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INTRODUCTION

In the homogeneous phase of critical mixtures the shear viscosity (η) exhibits a marked critical anomaly which may be portrayed by the relation ([1–14], and references therein)

$$\eta = \eta_B A_\eta t^{-\phi}, \quad (1)$$

where ϕ is the critical exponent, A_η is the critical amplitude, index B denotes the noncritical background effect, $t = (T - T_C)/T_C$ is the dimensionless distance from the critical consolute point, and η_B is the noncritical background viscosity.

This relation is also valid for the critical consolute point in micellar mixtures [10,11], the ionic critical mixtures [12], and on approaching the critical-end-point in a four-component mixture [6,13]. The theory proposes an analogy to the above relation [6,15,16]:

$$\eta(\xi) = \eta_B (\Lambda \xi)^{z_\eta} = \eta_B (\Lambda \xi_0)^{z_\eta} t^{-z_\eta \nu} = \eta_B A_\eta t^{-\phi}; \quad (2)$$

where Λ is the Debye cutoff number, $\xi = \xi_0 t^{-\nu}$ is the correlation length, and ξ_0 is the correlation length amplitude. A_η and $\nu \approx 0.63$ are the critical amplitude and the critical exponent, respectively. z_η is the universal critical exponent. The comparison with relation (1) gives $A_\eta = (\Lambda \xi_0)^{z_\eta}$ and $z_\eta = \phi/\nu$.

The mode-coupling theory predicts the values $z_\eta \approx 0.051 - 0.053$ ($\phi = 0.032 - 0.033$) [17,18], while the dynamic renormalization group gives the value $z_\eta \approx 0.065$ ($\phi \approx 0.041$) for the second-order approximation [19,20] or $z_\eta \approx 0.054$ and 0.053 for ε expansion to the first [20] and third-order term [21], respectively. No conclusive support for the theory has been obtained so far. A similar ambiguity occurs in the experimental where values of the critical exponent are of the range $0.051 \leq z_\eta \leq 0.065$ [7–14,22,23]. However, recent tests are strongly in favor $z_\eta \approx 0.065$ [13,14,24,25]. In the vicinity of the critical point the shear gradient effect has been taken into account, e.g., as proposed by Oxtoby [26], Onuki and Kawasaki [27,28]. The shear-corrected data are described by a slightly shifted value of z_η and T_C [10–14,23]. There is no viscosity anomaly in the strong shear regime ($z_\eta \approx 0$) [24,27], but this is not the case discussed in

this paper. Remote from T_C the noncritical background effect dominates. It is usually described by means of the Arrhenius equation although other relations are also used [1–14]. Generally, the noncritical background and the critical effect may be linked additively or multiplicatively [13]. The experimental results quoted above are in favor of the latter, in agreement with relation (2). In order to connect the possible scaling behavior of viscosity close to T_C with the background behavior the crossover function was also proposed [12].

Tests on shear viscosity in the two-phase region are scarce. To the best of the author’s knowledge they are limited to the analysis of the temperature evolution of the mean viscosity in the upper and lower coexisting phases [4,13]. This can be related to problems of separate estimation of the background term in coexisting phases. For mean viscosity the same equation (1) was used to parametrize experimental data above and below the critical consolute temperature. It was found that within the limit of experimental error $\phi^- \approx \phi^+$ and $A_\eta^+/A_\eta^- \approx 0.95$, in agreement with theoretical prediction [29]. For mean viscosity below T_C the Arrhenius temperature dependence of the background term was also applied. In the opinion of the author such an assumption neglects the fact of the possible violation of the rectilinear diameter law for the coexisting phase for the viscosity [9]. There is a clear experimental evidence of the weak distortion from the law of rectilinear diameter for the order parameter related properties, as for instance density or concentration [9].

Measurements of shear viscosity in the nitrobenzene–decane critical mixture carried out above and below the critical consolute point are reported here. In the coexisting phases the analysis was conducted separately for the lower and upper phase, respectively. This enabled the comparison of the values of critical exponents and amplitudes for the three paths of approaching T_C . The advantage of the chosen mixture is the value of $dT_C/dP \approx -0.02$ (KMPa⁻¹) [30] which made it possible to neglect the possible pressure induced shift of critical concentration x_C [31,32].

EXPERIMENT

The viscosity measurements at a given temperature were performed simultaneously in the upper and the lower coex-

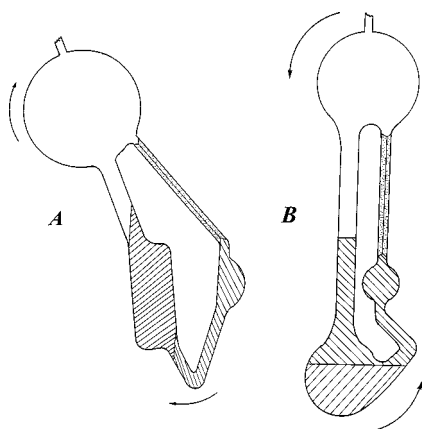


FIG. 1. Diagrams of viscometers: *A* is for measurements in the homogeneous and in the lower phase, *B* is for measurements in the homogeneous and in the upper phase. Arrows indicate directions of turning. The diameter of the capillary was 0.3 mm and the length 9 cm.

isting phases using the two capillary flow viscometers shown in Fig. 1. Calibration was carried out by recording the flow through times for three liquids: nitrobenzene, benzene, and carbon tetrachloride, in a temperature range from 7 to 50 °C. The following relation was applied to calculate the viscosity:

$$\frac{\eta}{\rho} = A t_f + \frac{B}{t_f}, \quad (3)$$

where η and ρ are values taken from [33], t_f denotes the flow through time of a determined volume of liquid, and the constants A and B are viscometer constants. The fact that calibration was conducted in the same range of temperatures made it possible to prepare over 100 pairs of equations from which the coefficients A and B were estimated. The studies focused on the nitrobenzene-decane solution of critical concentration $x_C = 0.57$ mole % fraction of nitrobenzene below and above the critical point ($T_C = 23.05$ °C), in agreement with results given in [34]. All liquids used in the experiments were purified by standard methods [35]. The tested solution was first placed in the viscometers and then frozen to temperature of ca. -80 °C, i.e., considerably lower than the melting temperature of both compounds. Then viscometers were sealed with a torch under vacuum and placed in a large volume (about 200 l of water) double-wall glass thermostat. This use of a relatively large volume made it possible to maintain (passive) temperature stabilization of $\pm 0.01^\circ/24$ h. The temperature was measured by means of a platinum resistor (DIN 43 260) with resolution ± 0.005 K and Keithley 195 A multimeter. Four thermocouples monitored the temperature gradient in viscometers. Measurements of viscosity were carried out simultaneously by means of both viscometers. Within the limits of the experimental error the obtained values were comparable. The time that elapsed before equilibrium was achieved depended on the distance from the critical point and had a value of about 2 h for temperatures remote from T_C and up to 10 h in the immediate vicinity of T_C . The viscometers were next turned to the position in which the measurement was carried out. For the viscometer measuring the viscosity of the upper phase the rotation was 180° and for the viscometer measuring the lower phase the

rotation was about 90° until the capillary was set in the vertical position. Arrows in Fig. 1 show the directions of rotation. The upper level of the liquid and the position of the meniscus dividing the phases are also indicated. If the rotation was performed smoothly, without jerks, then no mixing of the phases took place during measurement. After completing the measurements the viscometers were turned back to their previous position so that the solution could reach the state of equilibrium at the next temperature.

In the homogeneous region the density was measured pycnometrically. Parameters characterizing the region of coexisting phases were obtained by applying a visual cathetometric method developed in Ref. [36] and were as follows:

$$\text{The homogeneous phase: } \rho^H = 0.9826 - 0.1903 \times t, \quad (4a)$$

$$\begin{aligned} \text{The upper phase: } x_{\text{Nb}}^U &= 0.57 - 1.305t^{0.316} \text{ and} \\ \rho^U &= 0.9826 - 0.443 \times t^{0.35}, \end{aligned} \quad (4b)$$

$$\begin{aligned} \text{The lower phase: } x_{\text{Nb}}^L &= 0.57 + 0.81t^{0.36} \text{ and} \\ \rho^L &= 0.9826 - 0.344 \times t^{0.32}, \end{aligned} \quad (4c)$$

ρ denotes the density (g cm^{-3}) and the concentration x_{Nb} is in the mole fraction of nitrobenzene. Values of exponents in these equations are comparable to the so-called effective exponent β [36]. The visual cathetometric method also made possible the independent estimation of the coordinates of the critical point: within the limits of experimental error they were in agreement with results of Ref. [34]. The application of shear gradient corrections mentioned above involves parameters which are unknown for the tested mixture, particularly in the two-phase region. Hence, they were applied only for the homogeneous phase. The analysis of data was conducted using ORIGIN 5.0 software.

RESULTS AND DISCUSSION

Figure 2 shows the obtained behavior of shear viscosity in the tested mixture. It is clearly seen that, on approaching the critical consolute temperature along the critical isopleth and along both branches of the coexistence curve, the pretransitional anomalies appear. For their parametrization the estimation of the noncritical background effect is essential. In the one-phase region remote from T_C the temperature evolution of the viscosity can be portrayed by the simple Arrhenius dependence, which extrapolated up to T_C forms the background effect (Fig. 2): $\ln(\eta/cP) = -4.52 + 1423.8/T$. Below T_C the background is influenced by both the temperature and the composition of the coexisting phases. This can be taken into account by applying the isothermic, concentrational dependence for a perfectly miscible mixture [37]:

$$\ln \eta^U = x_{\text{Nb}}^U \ln \eta_{\text{Nb}} + (1 - x_{\text{Nb}}^U) \ln \eta_{\text{D}}, \quad (5a)$$

$$\ln \eta^L = x_{\text{Nb}}^L \ln \eta_{\text{Nb}} + (1 - x_{\text{Nb}}^L) \ln \eta_{\text{D}}, \quad (5b)$$

where η_{Nb} , η_{D} are the viscosities of constituents of the mixture in cP and x_{Nb} , x_{D} are for the upper (U) and the lower (L) phase, respectively.

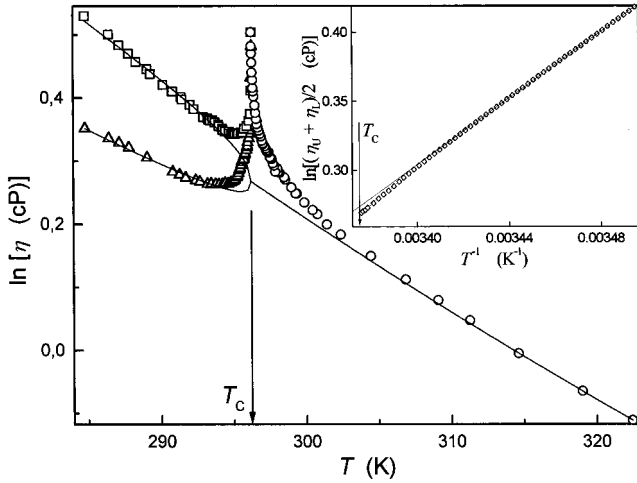


FIG. 2. The shear viscosity data for the tested nitrobenzene-decane mixture. The fitted by means of Eqs. (5a), (5b), and (6) noncritical background effects are shown by solid lines. The inset shows the form of the temperature behavior of the mean of viscosities in coexisting phases. The solid line represents the Arrhenius dependence.

For the tested critical mixture the shift of temperature changes both the composition of the coexisting phases and the viscosity of the mixture constituents. These factors can be taken into account by inserting the following equations into relations (5):

$$\eta_{\text{Nb, D}} = K_{\text{Nb, D}} \exp\left[\frac{D_{\text{Nb, D}}}{T}\right], \quad (6a)$$

$$x_{\text{Nb, D}} = x_C \pm C_{\pm} t^{\beta}, \quad (6b)$$

where $K_{\text{Nb, D}}$, $D_{\text{Nb, D}}$, and C_{\pm} denote constants depending on the type of substance and the sign \pm describes the left or right branch of the coexistence curve. The values of coefficients in Eq. (6b) are given in relations (4a) and (4b).

In the two-phase region the background coefficients K and D are given by the $\ln \eta_{\text{Nb}} = -4.81 + 1518.2/T$, $\ln \eta_{\text{D}} = -3.05 + 974.7/T$. They are in a good agreement with values for pure compounds [33]. The obtained background effects are shown by solid lines in Fig. 2. The inset in Fig. 2 presents the temperature dependence of the mean value of the fitted background viscosities in the coexisting phases, i.e., the diameter of the ‘‘background, noncritical, viscosity coexistence curve.’’ It follows the Arrhenius dependence only for $T - T_C > 2$ K. This deviation from the Arrhenius relation near T_C may indicate a violation of the Cailliet-Mathew law of the rectilinear diameter [9] for the background mean viscosity.

Figure 3 show the critical effects obtained if the above procedure of determining the background effect is applied. A power-law-type, critical behavior, with almost the same value of the critical exponent above and below T_C is clearly visible. It is worth noting that there is a quantitative agreement of critical effects in the lower, nitrobenzene-rich, and in the upper, decane-rich, coexisting phases. Fitted parameters describing critical effects are given in the caption of Fig. 3. In the immediate vicinity of T_C the obtained values of critical exponents are almost the same in all tested phases:

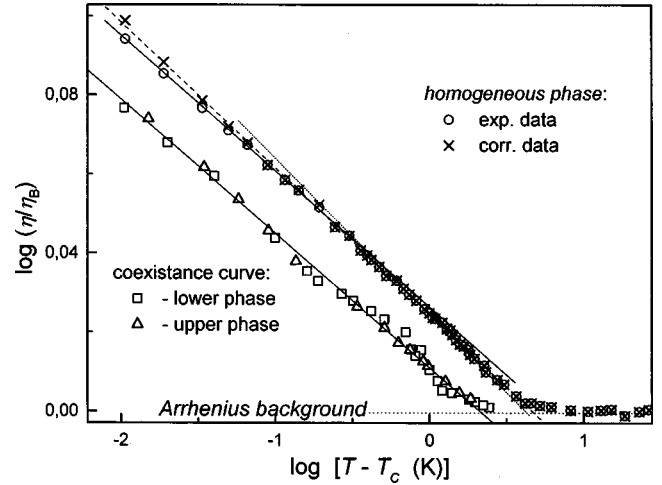


FIG. 3. Critical effects in the lower, upper, and homogeneous phases of a near-critical nitrobenzene-decane critical mixture. The horizontal line presents the Arrhenius background. The dashed line shows results of the fit with shear rate correction [10,23]: $\eta_{\text{corr}} = 1.058t^{-0.038}$. The solid and the dotted line describes results without the correction, for the separate fit of data near T_C and remote from T_C : $\eta = 1.062t^{-0.0332}$ ($T - T_C < 0.5$ K) and $\eta = 1.051t^{-0.041}$ ($1 \text{ K} < T - T_C < 7$ K). The bottom solid line is for the critical effect in the upper and in the lower coexisting phases: $\eta_{\text{lower}} = 1.026t^{-0.0334}$ and $\eta_{\text{upper}} = 1.026t^{-0.0331}$.

$\phi \approx 0.033$. A slight discrepancy from the value of the exponent ϕ mentioned above takes place in the homogeneous phase remote from T_C as shown in Fig. 3. If only data far from T_C are fitted $\phi \approx 0.041$ (dashed line). The application of the shear gradient correction [10,23] practically removes the discrepancy between the behavior near and remote from T_C , giving $\phi \approx 0.038$ (dotted line). The relations for shear gradient corrections were first proposed in the phenomenological model of Oxtoby and next Douglas which derived a simple expression convenient in experimental analysis. These expressions agree with results of Onuki-Kawasaki [28] and Onuki [27] mesoscopic-scale theoretical analysis of a critical solution under shear flow. For the tested critical mixture $z_{\eta} \approx 0.038/\nu \approx 0.062_6$ were obtained (Fig. 3). This value is close to the renormalization group estimation ($\phi \approx 0.065$ [20]) and in agreement with a series of papers [8–14,23–25]. One of the significant problems occurring when the shear corrections are taken account is the estimation of the correlation length amplitude ξ_0 . For the tested solution this coefficient is unknown, and hence it was assumed to be the same as in similar critical mixtures, i.e., $\xi_0 = 2A$ [9–13]. It should be noted that the shear correction gave only subtle distortion of experimental data near T_C (4–5 points in Fig. 3). On approaching T_C the value of the Deborah number $De = S\tau$ (τ denotes the relaxation time) gradually increased from almost zero remote from T_C up to 16 in the immediate vicinity of T_C . The mentioned corrections made it possible to obtain all data for unified shear rate $S = 0$ [8,10,23,26–28]. For very large values of De the local slope of data should decrease as De approaches $\phi = \nu z_{\eta} \approx 0$ [38,39]: Hamamo *et al.* reported even $De \approx 680$ [39]. This behavior is in an agreement with the theory of Onuki and Kawasaki [28,40,41].

In the opinion of the author the results for light scattering (I_L) and ordered parameter (M) studies in a critical solution

under shear flow [42,43] are worth recalling. It has been found that the type of the critical behavior is related to the value of the Deborah number: classical exponents $\gamma \approx 1$ (I_L) and $\beta \approx 0.5$ (M) were found in the vicinity of T_C where $De = S\tau > 1$. Result of these experiments delivered a broad experimental confirmation of the theoretical analysis of Onuki-Kawasaki [28,40,41]. Classical behavior near T_C was also found for a critical solution under strong electric field in nonlinear dielectric effect (NDE) [44–47], electro-optic Kerr effect (EKE) [44,48], and I_L [49] studies. The assumption that strong external field (S , E) induces the quasinematic structure of critical fluctuations and the isotropic, nonclassical correlation length $\xi \propto t^{-\nu} \approx t^{-0.63}$ may change to $(\xi_{\parallel}, \xi_{\perp}, \xi_{\perp})$ with ξ_{\parallel} remaining nonclassical and $\xi_{\perp} \propto t^{-0.5}$ (classical dependence) made the puzzling discrepancy between the theory and the experiment for EKE and NDE possible to explain [44–48]. The discussed behavior under external disturbance agrees with the complex liquid nature of the critical mixture [50]. Assuming the significance of the quasinematic picture also for the shear viscosity anomaly and taking $\eta \propto (\xi_{\perp})^{z_{\eta}}$ one can obtain in the vicinity of T_C :

$$z_{\eta} = \phi / \nu_{\text{class}} \approx 0.033 / 0.5 = 0.066 \pm 0.002,$$

$$\nu_{\perp} = 0.5 \quad \text{for } T \rightarrow T_C.$$

On moving away from T_C the order parameter fluctuations are less affected by the shear and their elongation decreases ($De < 1$):

$$z_n = \phi \nu_{\text{non-class}} \approx 0.041 / 0.63 = 0.065 \pm 0.002,$$

$$\nu_{\perp} = \nu \approx 0.063, \quad \text{for } T > T_C + 1K.$$

The obtained values of z_{η} , the same near and remote from T_C , agrees with the shear corrected values (i.e., for $S=0$) in this paper and in Refs. [11–14,24,25]. The subtle shear-rate decrease of the critical exponent ϕ on moving close to T_C was not found in the two-phase region perhaps due to the complex form of the background effect in the coexisting phases.

CONCLUSIONS

Results presented above show the analysis of the critical effect of shear viscosity in coexisting phases and in the homogeneous phase. Within the limit of experimental error the following relations were obtained for apparent critical exponents: $\phi_{+} = \phi_{-}^U = \phi_{-}^L \approx 0.033$ and amplitudes $A_{\eta}^L = A_{\eta}^U$ and $A_{\eta}^{-}/A_{\eta}^{+} \approx 0.965 \pm 0.10$. The latter value is close to the one predicted theoretically [29]. For all tested regions the multiplicative background effect was successfully applied. The Arrhenius temperature dependence of the mean value of the background viscosity in coexisting phases suggested previously [5] seems to be not valid in the immediate vicinity of T_C . In the homogeneous phase a slight increase of the apparent exponent ϕ from 0.033 (close to T_C) to the value 0.041 (data only remote from T_C) takes place. The application of shear rate corrections enables the description of all experimental data by a single-term temperature dependence with the effective exponent $\phi \approx 0.038$. When discussing the critical anomaly of dynamic physical properties, worth mentioning are also recent results for the relaxation time evolution. On the whole, critical mixtures obey the Einstein-Stokes formula $\tau / \eta \propto t^{-y}$ with $y = z\nu \approx 1.9$, where $z \approx 3.07$ is the dynamical exponent for the conserved order parameter [9]. However, in a strong electric field $y = 1.1 - 1.3$ [47]. This has been associated with the external field-induced semiclassical, calamitic-dislike, quasinematic structure. For prolate fluctuations, which seem to appear for a solution under shear flow [40], basing on Ref. [47], one can expect $y = z(\nu_{\perp} + z_{\eta}) \approx 1.1$ with $z = 2$. The latter value for the nonconserved order parameter [9] appears, for instance, in the isotropic phase of rodlike nematogens where recently a fluidlike description [51–53] was proposed.

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- [1] Y. Izumi and Y. Miyake, Phys. Rev. A **16**, 2120 (1977).
 [2] Y. Izumi, A. Dondos, C. Picot, and H. Benoit, J. Phys. (Paris) **42**, 353 (1981).
 [3] H. C. Burstyn, J. V. Sengers, J. K. Bhattacharjee, and R. A. Ferrell, Phys. Rev. A **28**, 1567 (1983).
 [4] S.-H. Chen, C.-C. Lai, J. Rouch, and P. Tartaglia, Phys. Rev. A **27**, 1086 (1983).
 [5] I. L. Pegg and I. A. McLure, Mol. Phys. Rep. **53**, 897 (1984).
 [6] S. J. Rzoska, J. Chrapeć, and J. Ziolo, J. Chem. Phys. **84**, 1735 (1986).
 [7] R. F. Berg and M. R. Moldover, J. Chem. Phys. **93**, 1926 (1990).
 [8] J. F. Douglas, Macromolecules **25**, 1468 (1992).
 [9] M. A. Anisimov, *Critical Phenomena in Liquids and in Liquid Crystals* (Gordon and Breach, New York, 1994).
 [10] A. Zielesny, S. Limberg, and D. Woermann, Ber. Bunsenges. Phys. Chem. **98**, 195 (1994).
 [11] S. Limberg, L. Belkoura, and D. Woermann, J. Mol. Liq. **73**, 74 (1997); **73**, 223 (1997).
 [12] M. Kleemeier, S. Wiegand, T. Derr, V. Weiss, W. Schroer, and H. Weingaertner, Ber. Bunsenges. Phys. Chem. **100**, 27 (1996).
 [13] I. A. McLure and P. J. Clements, Ber. Bunsenges. Phys. Chem. **101**, 114 (1997).
 [14] R. F. Berg, M. R. Moldover, and G. A. Zimmerli, Phys. Rev. Lett. **82**, 920 (1999).
 [15] K. Kawasaki, *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 5A.
 [16] P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).
 [17] R. Perl and R. A. Ferrell, Phys. Rev. Lett. **29**, 51 (1972); Phys. Rev. A **6**, 2358 (1972).

- [18] T. Ohta and K. Kawasaki, Prog. Theor. Phys. **55**, 1384 (1976).
- [19] E. D. Siggia, B. I. Halperin, and P. C. Hohenberg, Phys. Rev. B **13**, 2110 (1976).
- [20] B. I. Halperin, P. C. Hohenberg, and E. D. Siggia, Phys. Rev. Lett. **32**, 1289 (1974).
- [21] J. K. Bhattacharjee and R. A. Ferrell, Phys. Rev. A **28**, 2363 (1983).
- [22] J. V. Sengers, Int. J. Thermophys. **203**, 3 (1985).
- [23] J. C. Nieuwoudt and J. V. Sengers, J. Chem. Phys. **90**, 457 (1989).
- [24] S. Wiegand, R. F. Berg, and J. M. H. Levelt Sengers, J. Chem. Phys. **109**, 4533 (1998).
- [25] J. Luettmmer-Strathmann, J. V. Sengers, and G. A. Olchowy, J. Chem. Phys. **103**, 7482 (1995).
- [26] D. W. Oxtoby, J. Chem. Phys. **62**, 1463 (1975).
- [27] A. Onuki, Phys. Lett. **70A**, 31 (1979).
- [28] A. Onuki and K. Kawasaki, Ann. Phys. (N.Y.) **131**, 217 (1979).
- [29] P. Calmettes, Phys. Rev. Lett. **39**, 1151 (1977).
- [30] P. Urbanowicz, S. J. Rzoska, M. Paluch, B. Sawicki, A. Szulc, and J. Ziolo, Chem. Phys. **201**, 575 (1995).
- [31] D. T. Jacobs, J. Chem. Phys. **91**, 560 (1989).
- [32] P. Urbanowicz and S. J. Rzoska, Phase Transit. **56**, 239 (1996).
- [33] J. Timmermans, *The Physico-Chemical Constants of Pure Chemical Compounds* (InterScience, New York, 1959).
- [34] A. Latos, E. Rosa, S. J. Rzoska, J. Chrapeć, and J. Ziolo, Phase Transit. **10**, 131 (1987).
- [35] A. Vogel, *Vogel's Textbook of Practical Organic Chemistry* (Longman, London, 1978).
- [36] S. J. Rzoska, Phase Transit. **27**, 1 (1990).
- [37] R. C. Reid, J. M. Prausnitz, and T. K. Sherwood, *The Properties of Gases and Liquids* (McGraw-Hill, Duesseldorf, 1977).
- [38] S. P. Lee and A. J. Purvis, Chem. Phys. **24**, 191 (1977).
- [39] Hamano *et al.*, Int. J. Thermophys. **16**, 355 (1995).
- [40] A. Onuki and K. Kawasaki, Ann. Phys. (N.Y.) **121**, 456 (1979).
- [41] A. Onuki, J. Phys.: Condens. Matter **9**, 6119 (1997).
- [42] D. Beysens, M. Gbadamassi, and B. Moncef-Bouanz, Phys. Rev. A **28**, 2491 (1983).
- [43] D. Beysens, R. Gastaud, and F. Decruppe, Phys. Rev. A **30**, 1145 (1984).
- [44] S. J. Rzoska, Phys. Rev. E **48**, 1136 (1993).
- [45] S. J. Rzoska, A. Drozd-Rzoska, M. Górny, and J. Ziolo, Phys. Rev. E **52**, 6325 (1995).
- [46] J. Ziolo and S. Rzoska, Phys. Rev. E **60**, 4983 (1999).
- [47] S. J. Rzoska, A. Drozd-Rzoska, and J. Ziolo, Phys. Rev. E **61**, 960 (2000).
- [48] S. J. Rzoska, V. Degiorgio, and M. Giardini, Phys. Rev. E **49**, 5234 (1994).
- [49] D. Wirtz, K. Berend, and G. G. Fuller, Macromolecules **25**, 7234 (1992).
- [50] F. Yonezawa, K. Tsuji, K. Kaji, and M. Doi, <http://coral.t.u-tokyo.ac.jp/complx1/What.html> (1999).
- [51] A. Drozd-Rzoska, Liq. Cryst. **24**, 835 (1998).
- [52] A. Drozd-Rzoska and S. J. Rzoska, Mol. Cryst. Liq. Cryst. Sci. Technol., Sect. A **330**, 29 (1999).
- [53] A. Drozd-Rzoska, Phys. Rev. E **59**, 5556 (1999).