PHYSICAL REVIEW E **73**, 061508 (2006)

Relaxation rate in the critical dynamics of the micellar isobutoxyethanol-water system with lower consolute point

I. Iwanowski,[1] S. Z. Mirzaev,[1,2] and U. Kaatze[1,*]
[1]*Drittes Physikalisches Institut, Georg-August-Universität, Friedrich-Hund-Platz 1, D-37077 Göttingen, Germany*
[2]*Heat Physics Department, Uzbekistan Academy of Sciences, Katartal 28, 700135 Tashkent, Uzbekistan*

(Received 28 February 2006; published 23 June 2006)

For the isobutoxyethanol-water mixture of critical composition acoustical scaling function measurements have been performed at frequencies between 200 kHz and 3 MHz. Using the relaxation rates of concentration fluctuations as resulting from dynamic light scattering and shear viscosity measurements, the acoustical data can be well represented by the Bhattacharjee-Ferrell empirical scaling function. The theoretically predicted scaled half attenuation frequency $\Omega_{1/2}^{BF}=2.1$ follows from the experiments. However, the adiabatic coupling constant, derived from the amplitude of the Bhattacharjee-Ferrell critical term, displays an unreliably strong temperature dependence. It is shown that this dependence upon temperature likely reflects a hidden low-frequency and low amplitude relaxation term in the acoustical spectra of the mixture of critical composition.

DOI: 10.1103/PhysRevE.73.061508 PACS number(s): 83.80.Qr, 62.60.+v, 64.60.Ht, 43.35.+d

I. INTRODUCTION

Much attention has been directed towards nonionic surfactants of the type $CH_3(CH_2)_{i-1}(OCH_2CH_2)_jOH$, abbreviated as C_iE_j $(i=1,2,\ldots;j=0,1,\ldots)$, because of their many applications in surface chemistry, chemical engineering, and the life sciences as well. Scientific interest springs particularly from the fascinating complexity of the solution properties of such molecules of dual nature [1,2]. C_iE_j-water mixtures display a rich phase behavior, reflecting the delicate hydrophilic-hydrophobic balance of the amphiphilic molecules [3].

The higher homologues in the C_iE_j series self-aggregate in aqueous solutions to form micellar structures above a critical micelle concentration cmc. In addition, the C_iE_j-water systems exhibit closed loop coexistence curves [4–11]. Close to the critical demixing points the individual molecular dynamics of the binary liquids is largely screened by the fluctuations in the local surfactant concentration, covering vast ranges of size and tending to mask specific molecular interactions thereby. These phenomena are well understood for binary mixtures in which the constituents are molecularly dispersed [12–17]. An interesting aspect of the micellar systems is their critical dynamics near a consolute point. Conflicting results have been reported for micellar systems with critical demixing point, namely, nonuniversal near-critical demixing properties and also universal behavior [18–21]. Recent broadband ultrasonic studies of a variety of C_iE_i-water systems [22,23] suggested the idea of a fluctuation controlled monomer exchange [24,25]. Butoxyethanol isomers are the first in the series of the C_iE_j molecules forming closed loop coexistence curves in water. These short chain ethylene glycol monoalkyl ethers are not typical nonionic surfactants. Nevertheless, their aqueous solutions indicate concentrations above which aggregates are formed [26–31]. These aggregates will be named premicellar structures or (nonproper) micelles further on. From the critical micelle mass fractions Y_{cmc} of higher C_iE_i homologues the critical micelle mass fraction $Y_{cmc}=0.07$ has been extrapolated for the system isobutoxyethanol-water (i-C_4E_1-H_2O), corresponding with a critical micelle concentration cmc=0.6 mol/ℓ at room temperature [24,25]. The cmc of this short chain amphiphile is, of course, no sharply defined concentration but characterizes a transition range from predominantly molecularly dispersed solutions to mixtures containing micellar structures. For the system i-C_4E_1-H_2O this transition regime is located well below the lower critical demixing mass fraction: $Y_c=0.318$ [11] and $Y_c=0.330$ [32] have been found according to the equal-volume criterion. Hence micelles and concentration fluctuations exist simultaneously near the critical demixing point. The question of whether or not the fluctuations in the local concentration interfere with the micelle formation and decay kinetics has not been answered so far [33].

A broadband ultrasonic spectrometry study of the i-C_4E_1-H_2O system has been performed recently [32] in order to investigate the aggregation kinetics as well as the critical dynamics at the lower demixing point. The relaxation rate Γ of concentration fluctuations, derived from the critical contribution to the ultrasonic spectra, did not follow the power law,

$$\Gamma(t) = \Gamma_0 t^{Z_0\tilde{\nu}} \tag{1}$$

as did the Γ values from static and dynamic light scattering [34]. In Eq. (1) Γ_0 is an amplitude,

$$t = |T - T_c|/T_c \tag{2}$$

is the scaled (reduced) temperature, with T_c denoting the critical temperature, and Z_0 as well as $\tilde{\nu}$ are universal critical exponents. The value of the exponent $\tilde{\nu}$ of the fluctuation correlation length is $\tilde{\nu}=0.63$ [12], whereas the value of the dynamical critical exponent Z_0 follows as $Z_0=3.054$ from mode-mode coupling theory and as $Z_0=3.065$ from renormalization group theory [12]. The deviations of relaxation rates, derived from the sonic attenuation coefficient measurements, from universal power law behavior may be consid-

---

[*] Electronic address: uka@physik3.gwdg.de

©2006 The American Physical Society

ered an indication of the simultaneous presence of two parallel pathways in the acoustical relaxation, of which the second pathway shortcuts the extremely slow concentration fluctuation dynamics near the critical point. Likely the second pathway is provided by the micelle formation and decay processes. This idea implies that the broadband ultrasonic spectra have been incompletely described by a model spectral function in which relaxation terms due to the concentration fluctuations and the micelle formation and decay kinetics are linearly superposed. An alternative view proceeds from an incomplete consideration of other relaxation processes, such as reflecting i-C_4E_1 dimerizations, in the analytical treatment of the experimental spectra.

In order to cast light on the discrepancy in the relaxation rates of the i-C_4E_1-H_2O mixture of critical composition, as resulting from different experimental methods, we have determined the low-frequency part of the ultrasonic spectra at 11 temperatures close to T_c, applying a special mode of measurements. These measurements allow for a re-evaluation of the relaxation rate Γ or order parameter fluctuations and for a detailed discussion of alternative relaxation models for the complex i-C_4E_1-water mixture.

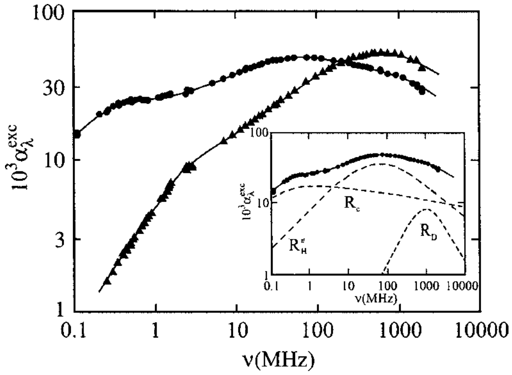

FIG. 1. Acoustical attenuation-per-wavelength spectra for i-C_4E_1-H_2O mixtures at 25 °C. •: mass fraction of surfactant $Y=0.330(=Y_c)$; ▲: $Y=0.57$. The inset shows the subdivision of the former spectrum into three relaxation terms. The subdivision follows from nonlinear least-squares regression analysis of the experimental spectrum in terms of Eqs. (3) and (5). In the fitting procedure the inverse experimental errors were used as weighing factors. The B value followed as 49.2 ps. The values for the other parameters from this regression analysis are given in Ref. [32], where also data at other i-C_4E_1 concentrations are presented.

II. EXPERIMENT

Isobutoxyethanol [$(CH_3)_2CHCH_2(OCH_2CH_2)OH$, i-C_4E_1, $\geq 97\%$] was purchased from Wako (Japan) and was purified by fractional distillation at 318 K and at a reduced pressure of 23 mbar. A concentric tube column of 64 real plates was used. In order to avoid unwanted water uptake from the air, the purified surfactant was always kept under dry nitrogen. The i-C_4E_1-H_2O mixtures of critical composition ($Y_c=0.330$, Y_c=mass fraction of surfactant) were prepared by weighing appropriate amounts of the constituents into suitable flasks. Deionized water was used that was additionally purified in a quartz bi-distil and subsequently degassed. The critical temperature of the mixture of critical composition, visually determined according to the equal volume criterion, was $T_c=298.10$ K. It was smaller than the critical temperature in previous experimental studies of the i-C_4E_1-H_2O system: $T_c=299.60$ K [11], $T_c=298.75$ K [28], $T_c=299.51$ K [32], $T_c=298.85$ K [35]. The differences in the T_c values are assumed to be predominantly due to the influence from traces of impurities in i-C_4E_1.

At frequencies between 200 kHz and 3 MHz the ultrasonic attenuation coefficient $\alpha(\nu,T)$ of the i-C_4E_1-H_2O mixture of critical composition has been measured at various temperatures $T \leq T_c$ using a cavity resonator method [36,37]. This method utilizes multiple reflections of the ultrasonic waves at the faces of the circular cylindrical cell in order to increase the path length of acoustical field interactions with the sample and thus the sensitivity in the measurements. For the consideration of intrinsic cell losses the attenuation coefficient of the sample was determined relative to water, used as reference liquid with sufficiently well matched sound velocity c_s and density ρ. We utilized the focusing effect of concavely shaped cell face (finely ground quartz transducer disc, $2R_q=80$ mm, radius of curvature $R_c=2$ m) to reduce disturbances of the resonator properties as might result from mechanical stress at varying temperature. Only the principal resonator modes were evaluated in terms of $\alpha(\nu,T)$. In order to take into account the effects from higher order modes of the acoustical cavity field the complete transfer function of the cell has always been recorded in an appropriate frequency range around a principal resonance peak. For this purpose a sensitive network analyzer was operated in a computer controlled mode. The transfer function was subsequently analyzed in terms of relevant analytical expressions [37,38] to obtain the unaffected principal resonance curve, the half power bandwidth of which yielded the attenuation coefficient α. As a byproduct of the resonator measurements the sound velocity c_s of the samples has been determined from the series of principal resonance frequencies, taking into account their nonequidistant distribution [37]. The temperature of the resonator cells was controlled to within 0.03 K and measured with an error of less than 0.02 K. The error in the frequency of measurements was negligibly small. The attenuation coefficient was determined to within $\Delta\alpha/\alpha=0.05$. The error in the sound velocity was $\Delta c_s/c_s=0.005$.

III. RESULTS AND DISCUSSION

In Fig. 1 a broadband ultrasonic spectrum of the i-C_4E_1-H_2O mixture of critical composition ($Y_c=0.330$) at $T-T_c=1.36$ K is shown in the format α_λ^{exc} vs ν. Here

$$\alpha_\lambda^{exc} = \alpha_\lambda - B\nu, \tag{3}$$

where $\alpha_\lambda=\alpha\lambda$ and $\lambda=c_s/\nu$ is the sonic wavelength. In Eq. (3) the term $B\nu$ denotes the asymptotic high frequency contribution which, according to [39,40]

$$B = \frac{2\pi^2}{\rho c_s^2}\left(\frac{4}{3}\eta_s + \eta_v\right) \tag{4}$$

is related to the shear viscosity η_s and volume viscosity η_v of the liquid. The excess-attenuation-per-wavelength data of the mixture of critical composition display significant contributions over an extended frequency range. A careful analysis of the broadband sonic i-C_4E_1-H_2O spectra at Y_c lead to the conclusion that at least three relaxation terms,

$$\alpha_\lambda^{exc} = R_c(\nu) + R_H^\#(\nu) + R_D(\nu), \tag{5}$$

are required for an adequate analytical representation of the experimental data [32]. The critical term, due to the fluctuations in the local concentration, has been assumed to be given by the Bhattacharjee-Ferrell dynamic scaling theory [41,42]

$$R_c(\nu) = c_s(\nu,T)A(\nu,T)F(\Omega) \tag{6}$$

with an amplitude factor $A(\nu,T)$ weakly depending on frequency ν and temperature T and with the empirical scaling function [42]

$$F(\Omega) = [1 + 0.414(\Omega_{1/2}^{BF}/\Omega)^{1/2}]^{-2}. \tag{7}$$

Herein

$$\Omega = \omega/\Gamma(t) = 2\pi\nu/\Gamma(t) \tag{8}$$

is the scaled frequency and $\Omega_{1/2}^{BF}$ is the scaled half attenuation frequency, defined by $F(\Omega_{1/2}^{BF})=0.5$. $\Omega_{1/2}^{BF}=2.1$ has been predicted theoretically and has been experimentally found for various critical binary liquids with simpler ultrasonic spectra [43–46]. The restricted Hill term [47,48]

$$R_H^\#(\nu) = 2^{1/s_H}A_H^\#\omega\tau_H[1 + (\omega\tau_H)^{2s_H}]^{-3/4s_H} \tag{9}$$

in Eq. (5) reflects the micelle formation and decay kinetics [49]. In this term $A_H^\#$ is an amplitude factor, τ_H is the principal relaxation time, and the parameter s_H controls the width and shape of the underlying continuous relaxation time distribution function. Finally, a Debye-type relaxation term [50]

$$R_D(\nu) = A_D\omega\tau_D[1 + (\omega\tau_D)^2]^{-1} \tag{10}$$

exists already in i-C_4E_1 without water added [32]. It seems to reflect a chemical equilibrium of i-C_4E_1 molecules, likely a step in the isobutoxyethanol isomerization scheme [32]. In Eq. (10) A_D is the relaxation amplitude and τ_D is the discrete relaxation time.

The subdivision of the excess attenuation spectrum of the i-C_4E_1-H_2O mixture of critical composition is indicated by dashed lines in Fig. 1. The sum function [Eq. (5)], shown by the full line in the figure, reveals a satisfactory description of the experimental α_λ^{exc} data by the three spectral terms R_c, $R_H^\#$, and R_D.

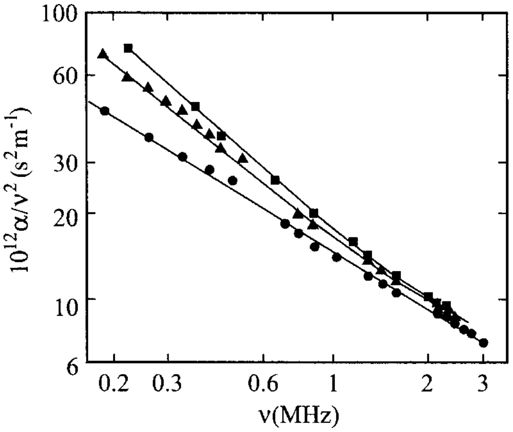

FIG. 2. Low-frequency part of frequency normalized acoustical attenuation spectra for the i-C_4E_1-H_2O mixture of critical composition. •: 20.2 °C; ▲: 23.64 °C; ■: 24.95 °C.

The spectrum for the i-C_4E_1-H_2O mixture with off critical composition, displayed in Fig. 1 for comparison, exhibits substantially smaller α_λ^{exc} values at frequencies below 100 MHz, indicating the enormous contribution of the critical term $R_c(\nu)$ to the attenuation coefficient data of the mixture of critical composition. A shoulder in the spectrum of the noncritical mixture points at an additional relaxation term in the frequency range around some MHz. All spectra of noncritical mixtures with $0.09 \leq Y < 1$ could be satisfactorily described only by a sum of four relaxation terms [32],

$$\alpha_\lambda^{exc}(\nu) = R_D^*(\nu) + R_c(\nu) + R_H^\#(\nu) + R_D(\nu), \tag{11}$$

with

$$R_D^*(\nu) = A_D^*\omega\tau_D^*[1 + (\omega\tau_D^*)^2]^{-1} \tag{12}$$

denoting another Debye-type relaxation function. This function has been also tentatively assigned to a step in the i-C_4E_1 dimerization scheme including linear and cyclic dimers [32]. It cannot be excluded, however, that this term also exists in the spectra of the mixture of critical composition but that it remains unnoticed because it is masked by the predominating critical term.

The low-frequency part in the excess attenuation spectra of the i-C_4E_1-H_2O mixture of critical composition is displayed at some temperatures in Fig. 2. At a given frequency ν the α_λ^{exc} values increase significantly with increasing temperature ($T \leq T_c$), indicating near T_c the sonic attenuation coefficients at $\nu < 3$ MHz to be predominantly given by the critical contribution $R_c(\nu)$. The scaling function $F(\Omega)$ in the critical term [Eq. (6)] may thus be favorably determined from the low-frequency attenuation data. According to the Bhattacharjee-Ferrell model [42] $F(\Omega)$ may be determined as the ratio [44]

$$F(\Omega) = F^*(T)\frac{\alpha_\lambda^c(\nu,T)}{\alpha_\lambda^c(\nu,T_c)} \tag{13}$$

from the experimental α values. In this relation

$$\alpha_\lambda^c(\nu,T) = \alpha_\lambda(\nu,T) - \alpha_\lambda^b(\nu,T) \tag{14}$$

is the critical contribution to the attenuation-per-wavelength data and α_λ^b denotes the noncritical background contribution. Factor

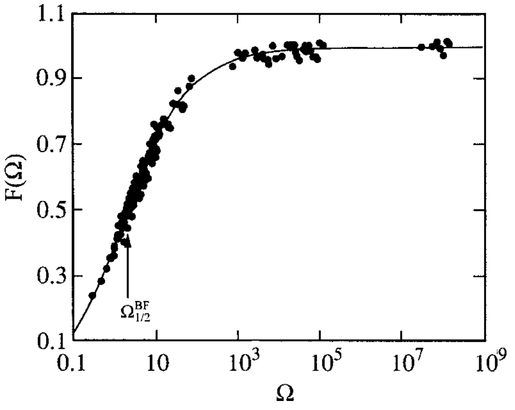

FIG. 3. Scaling function data for the i-C_4E_1-H_2O system. The line is the graph of the Bhattacharjee-Ferrell scaling function Eq. (7) with $\Omega^{BF}_{1/2}=2.1$.

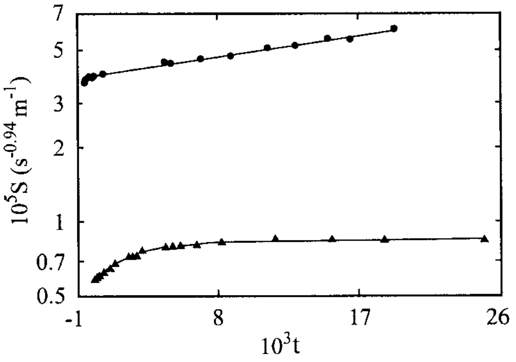

FIG. 4. Amplitude S of the Bhattacharjee-Ferrell theoretical model [Eq. (19)] as a function of reduced temperature t for the i-C_4E_1-H_2O (•) and the nitroethane-cyclohexane (▲ [44]) mixtures of critical composition. The lines are drawn to guide the eyes.

$$F^*(T) = c_s(T_c)A(T_c)/[c_s(T)A(T)] \tag{15}$$

considers the weak temperature dependence of the sound velocity c_s and amplitude factor A in Eq. (6). Normally, $F^* \equiv 1$ is assumed near T_c.

In order to reduce the number of unknown parameters in the treatment of the experimental data we used

$$\alpha^b_\lambda(\nu,T) = R^\#_H(\nu,T) + R_D(\nu,T) + \nu B(T) \tag{16}$$

as followed from the broadband ultrasonic spectra. With the values for the parameters of the $R^\#_H$ and R_D terms and for B, as obtained from interpolation and extrapolation of the previous data [32], the experimental scaling function displayed in Fig. 3 has been obtained. Calculating the scaling function data the relaxation rate Γ of concentration fluctuations has been assumed to follow a power law [Eq. (1)] with $\Gamma_0=5.3\times 10^9$ s^{-1}, as resulting from

$$\Gamma_0 = 2D_0/\xi_0^2 \tag{17}$$

with amplitudes $D_0=2.66\times 10^{-10}$ m^2 s^{-1} and $\xi_0=0.32$ nm for the mutual diffusion coefficient and fluctuation correlation length, respectively, from static and dynamic light scattering [11].

The only adjustable parameter in this treatment of experimental attenuation coefficients is the factor $F^*(T)$ and thus, because $c_s(T)$ is known from measurement, the amplitude factor $A(T)$. The scaling function data resulting from this procedure nicely fit to the empirical form [Eq. (7)] of the scaling function, the graph of which is also shown in Fig. 3. The agreement between experiment and theoretical predictions is, in particular, almost perfect at low Ω values. It is therefore concluded that $\Omega^{BF}_{1/2}=2.1$ is the correct value for the scaled half attenuation frequency of the critical i-C_4E_1-H_2O system.

Use of the relaxation rates from light scattering experiments, however, results in $F^*(T)$ values which lead to a distinct temperature variation in the Bhattacharjee-Ferrell amplitude [42]

$$S(T) = A(T)\nu^\delta, \tag{18}$$

where $\delta=\alpha_0/(Z_0\tilde{\nu})=0.06$ is a universal critical exponent. Here $\alpha_0=0.11$ is the specific heat exponent [51,52]. The temperature dependence in S, as following from our treatment of attenuation coefficient data in terms of the Bhattacahrjee-Ferrell scaling function, is almost as large (Fig. 4) as for the nitroethane-cyclohexane critical system [44]. Theory predicts the amplitude [42]

$$S(T) = \frac{\pi^2 \delta C_{pc} c_s^2(T)}{2T_c C_{pb}^2(T)}\left[\frac{\Omega^{BF}_{1/2}\Gamma_0}{2\pi}\right]^\delta g^2(T) \tag{19}$$

to be related to the temperature dependent heat capacity

$$C_p(t) = C_{pc}t^{-\alpha_0} + C_{pb}(t) \tag{20}$$

and adiabatic coupling constant [42]

$$g(T) = \rho(T_c)C_p(T)\left(\frac{dT_c}{dP} - \frac{T\alpha_p(T)}{\rho(T_c)C_p(T)}\right). \tag{21}$$

Herein, dT_c/dP is the slope in the pressure dependence of the critical temperature along the critical line, ρ is the density, and α_p is the thermal expansion coefficient at constant pressure.

Using $C_{pc}=55.45$ J/kg K and $C_{pb}=3740$ J/kg K [53], the g values from the amplitudes S decrease from $g=1.77$ at $t=0.0012$ to $g=1.33$ at $t=0.019$. In contrast, almost constant $g=1.35$ results from the thermodynamic relation Eq. (21). In deriving this value $dT_c/dP=39.8\times 10^{-3}$ K/bar [54] has been used, $\rho(T_c)=0.9715$ g/cm^3 [32], and

$$\alpha_p = \alpha_c t^{-\alpha_0} + \alpha_b \tag{22}$$

with

$$\alpha_c = \frac{\rho(T_c)C_{pc}}{T_c}\frac{dT_c}{dP} \tag{23}$$

as well as

$$\alpha_b = x\alpha_{biso} + (1-x)\alpha_{bw}. \tag{24}$$

In this mixture relation x denotes the mole fraction of isobutoxyethanol in the mixture of critical composition

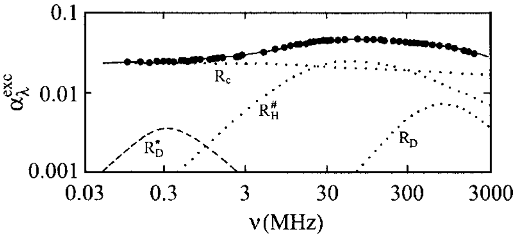

FIG. 5. Ultrasonic attenuation-per-wavelength spectrum for the i-C_4E_1-H_2O mixture of critical composition at the critical temperature. Dashed and dotted lines indicate the subdivision of the spectrum into the hidden term and the obvious relaxation terms, respectively. The full line shows the sum of these terms.

$(x=0.070)$ and $\alpha_{biso}=0.968\times10^{-3}\ \mathrm{K}^{-1}$ as well as $\alpha_{bw}=0.257\times10^{-3}\ \mathrm{K}^{-1}$ are the thermal expansion coefficients of the constituents [55].

Hence treating the ultrasonic spectra of the i-C_4E_1-H_2O mixture of critical composition, assuming three relaxation terms [Eq. (5)] leads to conflicting results. As mentioned before, a temperature independent coupling constant, as suggested by the thermodynamic relation Eq. (21), results in relaxation rates Γ that substantially differ from those obtained from quasielastic light scattering and shear viscosity measurements [32]. If the relaxation rates from the latter measurements are fixed in the evaluation of ultrasonic spectra, the adiabatic coupling constant of the Bhattacharjee-Ferrell term exhibits an unrealistically strong dependence upon temperature. We thus conclude that Eq. (5) does not satisfactorily represent the ultrasonic spectra. Rather an additional relaxation term, as characteristic for the mixtures of noncritical composition [32] seems to also exist in the spectra of the critical mixture. Applying Eq. (12) to the latter allows for an excellent representation of the experimental data even if the relaxation rate Γ is fixed at the values following from Eqs. (1) and (17) and if, in addition, the amplitude of the critical term is calculated with the almost temperature independent coupling constant g obtained from Eq. (21). As indicated by the subdivision of the ultrasonic spectrum of the critical mixture shown in Fig. 5 the additional low-frequency Debye-type relaxation term $R_D^*(\nu)$ is largely masked by the predominating Bhattacharjee-Ferrell term $R_c(\nu)$. Because of its comparatively small amplitude $R_D^*(\nu)$ remains unnoticed in the analysis of the broadband spectra in terms of relaxational contributions. Omission of the Debye term, however, leads to inconsistent conclusions from the scaling function data.

ACKNOWLEDGMENTS

Financial assistance provided by the VolkswagenStiftung (Hannover, Germany) and the Deutsche Forschungsgemeinschaft (Bonn, Germany) are gratefully acknowledged.

---

[1] B. Jonsson, B. Lindmann, K. Holmberg, and B. Kronberg, *Surfactants and Polymers in Aqueous Solution* (Wiley, Chichester, 1998).

[2] D. F. Evans and H. Wennerström, *The Colloidal Domain. Where Physics, Chemistry, and Biology Meet* (Wiley-VCH, New York, 1999).

[3] J. Israelachvili, *Intermolecular and Surface Forces*, 2nd ed. (Academic, New York, 1992).

[4] M. Corti, C. Minero, and V. Degiorgio, J. Phys. Chem. **88**, 309 (1984).

[5] *Physics of Amphiphiles: Micelles, Vesicles and Microemulsions*, edited by V. Degiorgio and M. Corti (North-Holland, Amsterdam, 1985).

[6] W. Biana-Limbelé, N. M. Van Os, L. A. M. Rupert, and R. Zana, J. Colloid Interface Sci. **144**, 458 (1991).

[7] K.-V. Schubert, R. Strey, and M. Kahlweit, J. Colloid Interface Sci. **141**, 21 (1991).

[8] M. Frindi, B. Michels, and R. Zana, J. Phys. Chem. **96**, 6095 (1992).

[9] A. Zielesny, L. Belkoura, and D. Woermann, Ber. Bunsenges. Phys. Chem. **98**, 579 (1994).

[10] S. Kato, S. Harada, and H. Sahara, J. Phys. Chem. **99**, 12570 (1995).

[11] S. Limberg, L. Belkoura, and D. Woermann, J. Mol. Liq. **73-74**, 223 (1997).

[12] M. A. Anisimov, *Critical Phenomena in Liquids and Liquid Crystals* (Gordon and Breach, Philadelphia, 1991).

[13] J. J. Binney, N. J. Dowrick, A. J. Fisher, and M. E. J. Newman, *The Theory of Critical Phenomena* (Clarendon, Oxford, 1992).

[14] C. Domb, *The Critical Point: A Historical Introduction to the Modern Theory of Critical Phenomena* (Taylor and Francis, London, 1996).

[15] M. E. Fisher, Rev. Mod. Phys. **70**, 653 (1998).

[16] H. E. Stanley, Rev. Mod. Phys. **71**, 5358 (1999).

[17] A. Onuki, *Phase Transition Dynamics* (Cambridge University Press, Cambridge, England, 2002).

[18] V. Degiorgio, R. Piazza, M. Corti, and C. Minero, J. Chem. Phys. **82**, 1025 (1985).

[19] M. Corti and V. Degiorgio, Phys. Rev. Lett. **55**, 2005 (1985).

[20] G. Dietler and D. S. Cannell, Phys. Rev. Lett. **60**, 1852 (1988).

[21] K. Hamano, N. Kuwahara, I. Mitsushima, K. Kubota, and T. Kamura, J. Chem. Phys. **94**, 2172 (1991).

[22] T. Telgmann and U. Kaatze, J. Phys. Chem. A **104**, 1085 (2000).

[23] T. Telgmann and U. Kaatze, J. Phys. Chem. A **104**, 4846 (2000).

[24] T. Telgmann and U. Kaatze, Langmuir **18**, 3068 (2002).

[25] E. Hanke, T. Telgmann, and U. Kaatze, Tenside, Surfactants, Deterg. **42**, 23 (2005).

[26] D. Woermann, Prog. Colloid Polym. Sci. **84**, 165 (1991).

[27] U. Kaatze, R. Pottel, and A. Schumacher, J. Chem. Phys. **96**, 6017 (1992).

[28] G. Perron, F. Quirion, D. Lambert, J. Ledoux, L. Gheicha, R. Bennes, M. Privat, and J. E. Desnoyers, J. Solution Chem. **22**, 107 (1993).

[29] U. Kaatze, K. Menzel, and S. Schwerdtfeger, Z. Phys. Chem.

(Munich) **186**, 141 (1994).
[30] B. Knecht and D. Woermann, Ber. Bunsenges. Phys. Chem. **99**, 1067 (1995).
[31] K. Menzel, A. Rupprecht, and U. Kaatze, J. Phys. Chem. B **101**, 1255 (1997).
[32] K. Menzel, S. Z. Mirzaev, and U. Kaatze, Phys. Rev. E **68**, 011501 (2003).
[33] M. Lesemann, A. Martin, L. Belkoura, and D. Woermann, Ber. Bunsenges. Phys. Chem. **101**, 228 (1997).
[34] F. Höhn, Ph.D. dissertation, Universität zu Köln, Köln, 1992.
[35] D. F. P. Rudd and B. Widom, J. Chem. Phys. **33**, 1816 (1960).
[36] U. Kaatze, R. Behrends, and K. Lautscham, Ultrasonics **39**, 393 (2001).
[37] F. Eggers, U. Kaatze, K. H. Richmann, and T. Telgmann, Meas. Sci. Technol. **5**, 1131 (1994).
[38] R. Behrends, F. Eggers, U. Kaatze, and T. Telgmann, Ultrasonics **34**, 59 (1996).
[39] G. Herzfeld and T. Litovitz, *Absorption and Dispersion of Ultrasonic Waves* (Academic, New York, 1959).
[40] U. Kaatze, T. O. Hushcha, and F. Eggers, J. Solution Chem. **29**, 299 (2000).
[41] J. K. Bhattacharjee and R. A. Ferrell, Phys. Rev. A **24**, 1643 (1981).
[42] R. A. Ferrell and J. K. Bhattacharjee, Phys. Rev. A **31**, 1788 (1985).
[43] R. Behrends, U. Kaatze, and M. Schach, J. Chem. Phys. **119**, 7957 (2003).
[44] R. Behrends, I. Iwanowski, M. Kosmowska, A. Szala, and U. Kaatze, J. Chem. Phys. **121**, 5929 (2004).
[45] R. Behrends and U. Kaatze, Europhys. Lett. **65**, 221 (2004).
[46] I. Iwanowski, A. Sattarow, R. Behrends, S. Z. Mirzaev, and U. Kaatze, J. Chem. Phys. **124**, 144505 (2006).
[47] R. M. Hill, Phys. Status Solidi B **103**, 319 (1981).
[48] K. Menzel, A. Rupprecht, and U. Kaatze, J. Acoust. Soc. Am. **104**, 2741 (1998).
[49] T. Telgmann and U. Kaatze, J. Phys. Chem. B **101**, 7766 (1997).
[50] P. Debye, *Polare Molekeln* (Hirzel, Leipzig, 1929).
[51] A. C. Flewelling, R. J. DeFonseka, N. Khaleeli, J. Partee, and D. T. Jacobs, J. Chem. Phys. **104**, 8048 (1996).
[52] P. F. Rebillot and D. T. Jacobs, J. Chem. Phys. **109**, 4009 (1998).
[53] W. Würz, M. Grubic, and D. Woermann, Ber. Bunsenges. Phys. Chem. **96**, 1460 (1992).
[54] H. J. Zimmer and D. Woermann, Ber. Bunsenges. Phys. Chem. **95**, 533 (1991).
[55] H. Doi, K. Tamura, and S. Murakami, J. Chem. Thermodyn. **32**, 729 (2000).