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Critical concentration fluctuations of the ionic binary mixture ethylammonium nitrate–*n*-octanol: An ultrasonic spectrometry study

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Between 200 kHz and 130 MHz, the ultrasonic attenuation spectrum of the ionic ethylammonium nitrate–*n*-octanol mixture of critical composition has been measured at various reduced temperatures ($1.5\times10^{-4}\leqslant t\leqslant5.7\times10^{-2}$). Using static and dynamic light scattering data as well as viscosity and heat capacity data from the literature, the experimental spectra have been evaluated to yield the scaling function, with the background contribution to the spectra as the only adjustable parameter. Agreement, within the limits of experimental error, of the measured scaling function with that of the nonionic binary system ethanol–dodecane and with the theoretical predictions of the Bhattacharjee-Ferrell dynamic scaling model is found. The amplitude of the fluctuation correlation length ξ_o ($=0.47$ nm) and the amount of the coupling constant $|g|$ ($=1.3$) are rather high as compared to nonionic binary critical mixtures. The amplitude of the relaxation rate of order parameter fluctuations $\Gamma_o(=2.6\times10^8\ \mathrm{s}^{-1})$ exhibits an unusual small value, likely to the most part a reflection of the high viscosity and thus small diffusion coefficient of the ionic liquid.

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I. INTRODUCTION

Critical demixing in nonionic binary fluids is governed by short-range van der Waals forces, hydrogen bonding, and entropy controlled solvophobic effects. Despite the relative importance of these interactions for a given system, Ising-type criticality normally results. Close to the critical point, the properties of such binary mixtures reflect large fluctuations in the concentration, largely masking the identity of the system. The phase transition is characterized by power laws determined by universal critical exponents, of which, in many cases, only two are linearly independent. The universal properties of the liquid-liquid phase transition have been intensively investigated in the past both experimentally and theoretically [1–6].

More recently, much attention has been directed towards ionic fluids [7–34] in which long-range Coulombic forces might lead to a special critical behavior. An unusual crossover from Ising-type to classical mean-field behavior has been found [35–37] and has been attributed to the simultaneous presence of two crossover parameters, related to two independent characteristic spatial scales of the liquid [38,39].

Since acoustical fields couple to the spatial Fourier components of the long-range concentration fluctuations and to many noncritical elementary processes as well, ultrasonic spectrometry is a powerful tool for the study of binary liquids near critical points. For this reason, we measured the ultrasonic attenuation coefficient of the ethylammonium nitrate (EAN)–*n*-octanol (n-$\mathrm{C_8OH}$) mixture of critical composition as a function of frequency and temperature near the critical temperature T_c. The results of our measurements are combined with heat capacity [40], shear viscosity [25], and static [16,27] and dynamic [16] light scattering data in order to minimize the number of unknown parameters in the evaluation of spectra. As we are interested in the importance of Coulombic interactions for the criticality of the system, attention is particularly paid to indications of effects from this interparticle force in the critical behavior of the ionic liquid.

II. EXPERIMENT

Ethylammonium nitrate was supplied by Weingärtner, Ruhr-Universität Bochum, Germany. The salt had been synthesized and dried as described previously [15]. Immediately before use, it was desiccated again by keeping it in a drying cabinet for three days at 65 °C under vacuum. The nonionic constituent *n*-octanol (⩾99.5%, Fluka, Deisenhofen, Germany) was used as delivered by the manufacturer. Mixtures of critical composition ($x_c=0.766$ [25], $x_c=$mole fraction of EAN) were prepared under dry nitrogen by weighing appropriate amounts of the constituents into suitable flasks. In order to avoid water uptake, all subsequent sample manipulations were also performed under dry nitrogen. All samples were carefully stirred at 70 °C well above the critical temperature T_c. Despite cautious handling of the samples, different mixtures of critical composition exhibited different visually determined critical temperatures T_{cvis}, thus reflecting the well-known high sensitivity of the critical temperature of ionic binary mixtures against small traces of impurity such as water [29]. Here we report results for three mixtures of critical composition with visually determined critical temperatures $T_{cvis}=47.70$, 44.29, and 42.37 °C, respectively. We mention that in the measurements of the heat capacity C_p, the critical temperature of different samples of the EAN–n-$\mathrm{C_8OH}$ system varied between 41 and 47 °C without any noticeable effect in the structure of the C_p versus T profiles [40]. It turned out that the ultrasonic spectra of the mixtures with different critical temperature also agree with one another if comparison is made of the excess attenuation coefficient

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$$\alpha_{exc}=\alpha-\alpha_{bg}, \tag{1}$$

at the same reduced temperature $t=|T-T_c|/T_c$. Here α_{bg} denotes the noncritical background attenuation coefficient at T. $T_{cvis}=42.37\ °\mathrm{C}$ is close to the lowest upper critical temperature reported so far for the system EAN–n-C_8OH ($T_c=40.42$ [25]).

The ultrasonic attenuation coefficient α of the mixtures of critical composition has been measured as a function of frequency f between 200 kHz and 2 MHz and reduced temperature t at $1.5\times10^{-4}\leqslant t\leqslant5.7\times10^{-2}$. In order to appropriately account for the asymptotic high frequency background contribution α_{bg} to the acoustical attenuation coefficient, one mixture has been additionally investigated at frequencies between 40 and 130 MHz. At some temperatures ethylammonium nitrate and n-octanol have been measured between 200 kHz and 15 MHz. In the lower frequency band ($f<15\ \mathrm{MHz}$), a resonator method has been utilized in which the sample is contained in a cylindrically shaped cavity cell. We used a planoconcave [41] and a biplanar [42] version of the resonator. Resonator measurements yield the attenuation coefficient of the sample relative to a reference liquid. We used appropriately chosen methanol–water mixtures as a reference. In the high frequency band ($f>40\ \mathrm{MHz}$), a pulse-modulated wave transmission method was applied that because of variation of the sample length, enables absolute α measurements [43]. The sound velocity V_c of the mixtures of critical composition has been determined from successive resonance frequencies of the cavity resonator cells. The temperature of the cells was controlled to within 0.03 K and measured with an error of less than 0.02 K. The frequency of measurement was known and kept constant with a negligibly small error. The error in the attenuation data was less than 5% at $f<15$ MHz and smaller than 2% at $f\geqslant40$ MHz. The sound velocity has been measured to within ±0.1%.

III. RESULTS

In Fig. 1, for an EAN–n-C_8OH mixture of critical composition, a double logarithmic plot is given of the α/f^2 data as a function of frequency f. Within each series of measurement, the attenuation coefficient decreases with f. The negative slope $-d(\alpha/f^2)/df$ of the curves increases on approach to the critical temperature ($t\rightarrow0$). For all reduced temperatures it is distinctly smaller than the negative slope of a Debye-type relaxation term

$$\frac{\alpha}{f^2}=\frac{A_D}{1+(2\pi f)^2\tau_D^2}, \tag{2}$$

at $f\gg(2\pi\tau_D)^{-1}$. At frequencies well above the relaxation frequency $f_D=(2\pi\tau_D)^{-1}$, such term is indicated by the dashed line in Fig. 1.

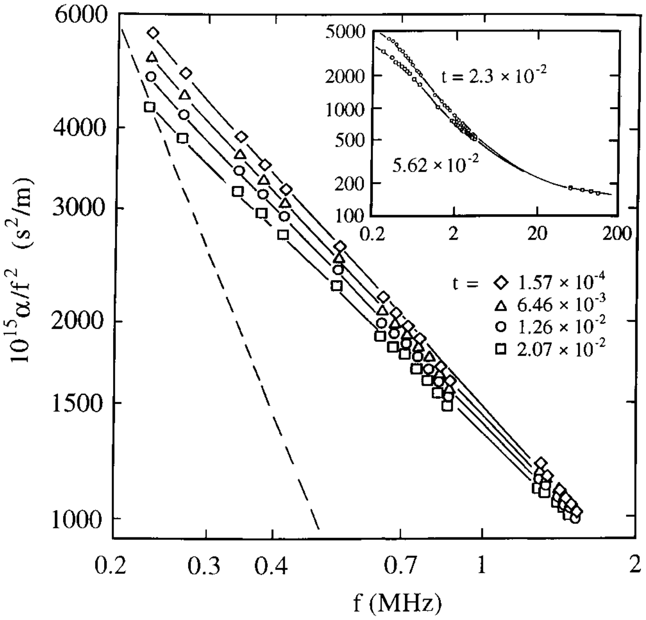

FIG. 1. Ultrasonic attenuation spectra at several reduced temperatures t for an ethylammonium nitrate–n-octanol mixture of critical composition. Full curves are drawn to guide the eyes. The dashed line is the graph of a Debye relaxation term [Eq. (2)] well above the relaxation frequency $(2\pi\tau_D)^{-1}$.

In Eq. (2) $A_D=2\alpha(f_D)/f_D^2$ is the amplitude and τ_D is the discrete relaxation time. The comparatively small negative slope in the α/f^2 versus f relations of the EAN–n-C_8OH mixture of critical composition points at an underlying broad relaxation time distribution, as characteristic for spectra due to critical fluctuations in concentration. As illustrated by the data shown in the inset of Fig. 1, towards high frequencies, the α/f^2 data tend toward a constant high frequency value B' that is taken to represent the noncritical background contribution to the spectrum. Hence, according to the relation

$$\frac{\alpha}{f^2}=\left(\frac{\alpha}{f^2}\right)_{crit}+B', \tag{3}$$

our spectra are assumed to be composed of a critical and a noncritical part.

In order to make sure that the low-frequency contribution in the α/f^2 spectra of the mixture of critical composition (Fig. 1) does not mask relaxation behavior of the constituents themselves let us inspect the ultrasonic attenuation coefficient data of n-octanol and EAN themselves. As shown by Fig. 2 for the alcohol at 40 °C, the α/f^2 values are constant within the relevant frequency range ($f<2$ MHz). This is in conformity with a recent broadband ($300\ \mathrm{kHz}\leqslant f\leqslant3\ \mathrm{GHz}$) ultrasonic study of alcohols, which for n-octanol at 25 °C, revealed Debye-type relaxation terms with relaxation frequencies as high as 420 MHz and 1.7 GHz [44]. The frequency normalized attenuation coefficient data of EAN may be taken to indicate a small dispersion at $f>7$ MHz (Fig. 2). These dispersion characteristics $[d(\alpha/f^2)/df<0]$ may reflect an ion complexation mechanism as has been suggested from broadband dielectric spectrometry of EAN [45]. The existence of a high-frequency ultrasonic relaxation process in pure EAN has still to be verified by additional measurements at $f>15$ MHz. Such process could, at least in parts, explain the rather high α/f^2 values of the room temperature molten salt at low frequencies ($\alpha/f^2=195\times10^{-15}\ \mathrm{s^2\,m^{-1}}$, 40 °C) but it is irrelevant in the measuring range of this study.

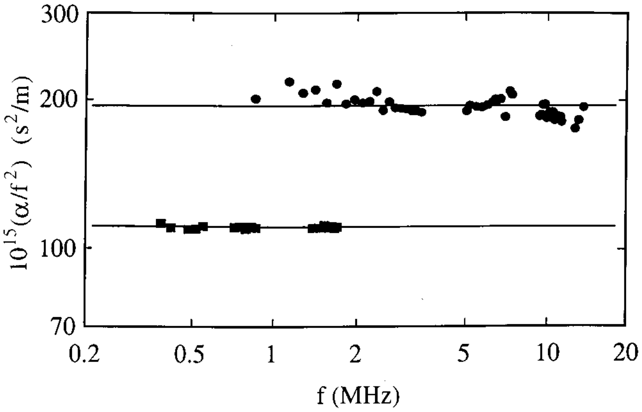

FIG. 2. Frequency normalized ultrasonic attenuation data versus frequency for ethylammonium nitrate (●) and *n*-octanol (full squares) at 40 °C.

IV. DISCUSSION

Let us consider the critical contribution (α/f^2) to the ultrasonic attenuation spectra in the framework of the Bhattacharjee-Ferrell dynamic scaling theory (BF) [46–48]. We are aware of more recent theoretical models [49–55] of critical sound attenuation in binary liquids. As will become obvious below, however, this ionic binary system is less suitable for an experimental examination of the fine details in the scaling function predicted by the different theoretical models. For reasons of a clear comparison with previous results for nonionic critical mixtures we thus prefer the BF treatment here.

The Bhattacharjee-Ferrell theory predicts the frequency normalized attenuation at the critical temperature to follow the relation

$$\left(\frac{\alpha}{f^2}\right)_{crit}(T_c)=Sf^{-(1+\delta)}+B'(T_c). \tag{4}$$

Herein

$$S=\frac{\pi^2\delta A V_c(T_c)}{2T_c}\left[\frac{\Omega_{1/2}\Gamma_o}{2\pi}\right]^{\delta}\frac{g^2}{C_p^2}, \tag{5}$$

is an amplitude that only weakly depends upon frequency, B' is the background contribution to the attenuation coefficient [Eq. (3)], $\delta=\alpha_o/(Z_o\nu)=0.06$, and α_o, Z_o, ν are the specific heat critical exponent, the dynamical critical exponent, and the critical exponent of the correlation length of order parameter fluctuations. In Eq. (5), A is the amplitude describing the singular behavior of the heat capacity [56]

$$C_p=\frac{A}{\alpha_o}t^{-\alpha_o}+Et+B, \tag{6}$$

$\Omega_{1/2}$ is the dimensionless half-attenuation frequency [46], Γ_o is the amplitude of the relaxation rate Γ of order parameter fluctuations, $V_c(T_c)$ is the sound velocity at the critical point, and g is the adiabatic coupling constant. According to

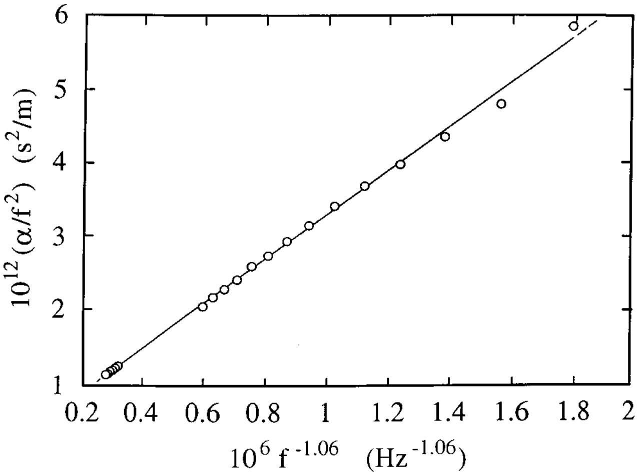

FIG. 3. Frequency normalized ultrasonic attenuation coefficient at $t=1.57\times10^{-4}$ for an EAN–n-C_8OH mixture of critical composition plotted as a function of $f^{-1.06}$.

$$g=\rho_c C_p\left(\frac{dT_c}{dP}-\frac{T\alpha_p}{\rho C_p}\right), \tag{7}$$

the coupling constant g is related to density ρ_c of the mixture at T_c, to the slope dT_c/dP in the pressure dependence of the critical temperature, and to the thermal expansion coefficient α_p at constant pressure.

In Fig. 3, at a temperature as close as possible to the critical, the frequency normalized attenuation coefficient of an EAN–n-C_8OH mixture of critical composition is plotted versus $f^{-1.06}$. Within the limits of experimental error, the data follow the predictions of Eq. (4), with frequency independent $S=(3.01\pm0.04)\times10^{-6}$ $s^{0.94}\,m^{-1}$ and $B'=(281\pm0.5)\times10^{-15}$ $s^2\,m^{-1}$. This result indicates the frequency dependent ultrasonic attenuation of the critical mixture to be well represented by the BF model. The small error in the asymptotic high frequency part B' should not be overestimated since B' just results from an extrapolation of low-frequency data to high frequencies. The B' value may thus contain contributions from noncritical relaxation processes.

At $T\neq T_c$, the Bhattacharjee-Ferrell theory yields an empirical scaling function [47]

$$F_{BF}(\Omega)=\frac{\alpha_{\rm crit}(f,T)}{\alpha_{\rm crit}(f,T_c)}=\frac{\Omega}{(1+\Omega^{1/2})^2}, \tag{8}$$

where $\alpha_{\rm crit}$ denotes the critical contribution to the total attenuation coefficient [Eq. (3)]. A more recent form of the scaling function [48]

$$F_{BF}(\Omega)=[1+0.414(\Omega_{1/2}/\Omega)^{1/2}]^{-2}, \tag{9}$$

is defined with the aid of the half-attenuation frequency $\Omega_{1/2}$ and with the reduced frequency $\Omega=2\pi f/\Gamma$. A half-attenuation value $1.7\leqslant\Omega_{1/2}\leqslant2.4$ with a weighted mean of $\Omega_{1/2}=2.0$ [49] had been derived from ultrasonic attenuation data of the critical systems 3-methylpentane–nitroethane [57] and cyclohexane–nitroethane [58], whereas $\Omega_{1/2}=1.2\pm0.2$ followed for ethanol–dodecane [59].

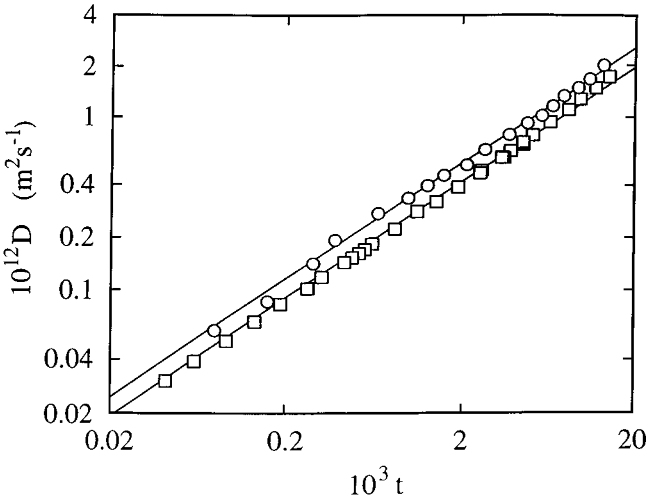

FIG. 4. Mutual diffusion coefficient of an EAN–n-C_8OH mixture of critical composition versus reduced temperature. One set of data has been obtained from static and dynamic light scattering data (○), [16], the other one (□) from static light scattering [27] and shear viscosity data [25].

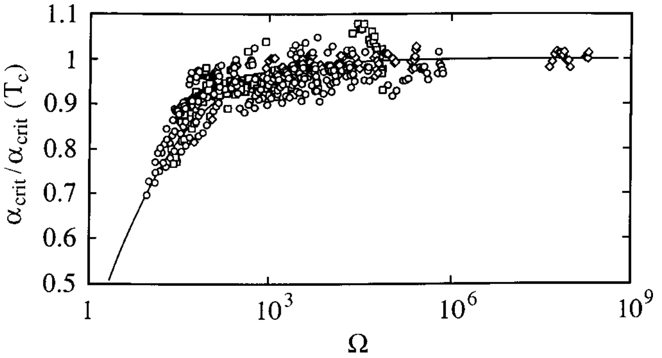

FIG. 5. Linear/log plot of the normalized attenuation coefficient excluding background contributions of three EAN–n-C_8OH samples of critical composition displayed as a function of reduced frequency. The curve is the graph of the empirical scaling function $F_{BF}(\Omega)$ defined by Eq. (9) with $\Omega_{1/2}=2$. Figures symbols indicate different samples.

In order to plot the experimental $\alpha_{\mathrm{crit}}(f,T)/\alpha_{\mathrm{crit}}(f,T_c)$ data as a function of the reduced frequency Ω, the relaxation rate Γ of the order parameter fluctuations has to be known for each pair of frequency-temperature values. We utilize the relation

$$\Gamma = 2D/\xi^2, \tag{10}$$

to calculate Γ from the mutual diffusion coefficient D and the fluctuation correlation length

$$\xi = \xi_o t^{-\nu}, \tag{11}$$

with the amplitude ξ_o and the critical exponent $\nu = 0.63$. Two sets of diffusion coefficients are available for the EAN–n-C_8OH system. One has been obtained from a recalculation of static and dynamic light scattering data [16]. The other one has been derived from shear viscosity data η_s [25] and $\xi_o = 0.48$ nm [27] using the equation

$$D = \frac{k_B T}{6\pi\eta_s\xi}. \tag{12}$$

In Fig. 4 both sets of data are displayed as a function of reduced temperature to show that the diffusion coefficient follows power law behavior

$$D = D_o t^{\nu}, \tag{13}$$

with $\nu^* = 0.664$, as predicted by the mode coupling theory [60]. Slightly different amplitudes $D_o = 3.3\times10^{-11}$ $\mathrm{m^2\,s^{-1}}$ and $D_o = 2.5\times10^{-11}$ $\mathrm{m^2\,s^{-1}}$ follow from the dynamic light scattering measurements and from the other set of diffusion coefficient data, respectively. The amplitudes of the fluctuation correlation length are also most consistent. $\xi_o = 0.47$ nm [16] and $\xi_o = 0.48$ nm [27] have been found in two independent series of light scattering experiments, $\xi_o = (0.50\pm0.04)$ nm results from heat capacity measurements [40] according to the two-scale-factor universality relation [61].

$$\xi_o = 0.27(k_B/A)^{1/3}, \tag{14}$$

with A defined by Eq. (6). Using a mean of these ξ_o data

$$\Gamma = \Gamma_o t^{Z_o\nu}, \tag{15}$$

follows with $\Gamma_o = (2.6\pm0.4)10^8$ $\mathrm{s^{-1}}$.

The $\alpha_{\mathrm{crit}}/\alpha_{\mathrm{crit}}(T_c)$ data obtained from measurements of three mixtures of critical composition at frequencies between 200 kHz and 130 MHz and at reduced temperatures between 1.5×10^{-4} and 5.7×10^{-2} are displayed as a function of reduced frequency Ω in Fig. 5. Also shown in the figure is the graph of the scaling function $F(\Omega)$ defined by Eq. (9), with a half-attenuation frequency $\Omega_{1/2}=2$. The experimental data do not deviate from the $F(\Omega)$ function by more than 10% and there are no systematic deviations of the $\alpha_{\mathrm{crit}}/\alpha_{\mathrm{crit}}(T_c)$ data from the theoretical predictions. In view of the fact that, except the ultrasonic background attenuation B', no adjustable parameters have been used in the evaluation of the measured attenuation coefficients and that only the half-attenuation frequency $\Omega_{1/2}$ in the scaling function may be varied within some narrow limits, the agreement between the experimental findings and the predictions of the BF theory is excellent. Due to the unusual small amplitude $\Gamma_o = 2.6\times10^8$ $\mathrm{s^{-1}}$ of the relaxation rate Γ, however, no Ω values smaller than 10 are reached with the ethylammonium nitrate–n-octanol system. In Fig. 6, therefore, the $\alpha_{\mathrm{crit}}/\alpha_{\mathrm{crit}}(T_c)$ values for one EAN–n-C_8OH mixture of critical composition are shown along with the corresponding data for the nonionic binary system ethanol–dodecane. Both sets of data nicely fit to one another and, with the exception of the region of small reduced frequencies ($\Omega < 0.3$), the scaling function defined by Eq. (9) represents the experimental data satisfactorily. It is only mentioned that the Folk-Moser scaling function [54] predicts $F(\Omega)$ values smaller than the measured $\alpha_{\mathrm{crit}}/\alpha_{\mathrm{crit}}(T_c)$ values at $\Omega < 0.3$ [16]. This

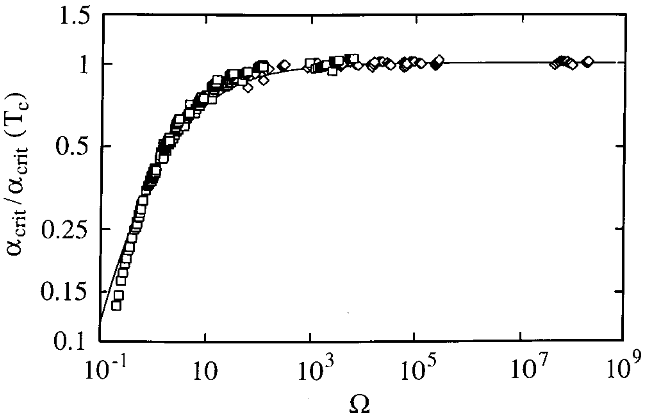

FIG. 6. Normalized attenuation coefficient, excluding background contributions, bilogarithmically plotted versus reduced frequency for one ethylammonium nitrate–*n*-octanol mixture of critical composition (◇) and for the ethanol–*n*-dodecane mixture of critical composition (□), [59]). The curve is the graph of the Bhattacharjee-Ferrell scaling function [Eq. (9)].

difference between the scaling functions at different theoretical models needs further inspection including additional experimental data at $\Omega < 0.1$.

The focus of this paper is potential special characteristics in the ultrasonic attenuation of the ionic binary critical system. We, thus, also evaluated the amplitude *S* of the spectrum at the critical point [Eq. (4)] for the amount of the coupling constant *g*. Using Eq. (5) with $S = 3.01 \times 10^{-6}\ \mathrm{s}^{0.94}\,\mathrm{m}^{-1}$, $T_c = 315.52\ \mathrm{K}$, $\delta = 0.06$, $V_c(T_c) = 1395.7\ \mathrm{ms}^{-1}$, $A = 1.82 \times 10^{-3}\ \mathrm{J/(gK)}$, $C_p = 2.111\ \mathrm{J/(gK)}$, $\Gamma_o = 2.6 \times 10^8\ \mathrm{s}^{-1}$, and $\Omega_{1/2} = 2.0$, the rather high amount $|g| = 1.3$ follows for the coupling constant of the EAN–n-C_8OH system. The errors in the parameter values yield an uncertainty $\Delta |g| = 0.2$. This uncertainty includes the use of $\Omega_{1/2} = 1.2$ instead of 2.0. In the above evaluation of the amplitude *S*, the *A* and C_p values have been calculated from the heat capacity data per unit volume [40] using the density $\rho_c = 1.0678\ \mathrm{g\,cm}^{-3}$ at the critical point.

In Table I, the system specific parameters Γ_o, ξ_o, $|g|$, and *S* are listed for various binary mixtures with critical demixing point. The ionic system exhibits by far the smallest amplitude Γ_o in the relaxation rate of order parameter fluctuations. For the system methanol–*n*-heptane, Γ_o is 31 times larger than for EAN–n-C_8OH and for all other systems listed in Table I the Γ_o is even larger. The amplitude ξ_o in the fluctuation correlation length of the EAN–n-C_8OH mixture is rather large, only that of the polypropylene glycol–polyethylene glycol system is somewhat larger. Also the amount of the coupling constant $|g| = 1.3$ is the second largest one within the series of binary critical mixtures compiled in Table I. The isobutyric acid–water system shows an even somewhat larger $|g| (= 2.1)$ than the system EAN–n-C_8OH. Hence, the most striking parameter of the various liquids displayed in Table I is the small Γ_o value of the ionic system, which seems to reflect its high viscosity. In addition, due to the Coulombic interactions, relaxation from a local nonequilibrium distribution of electrical charges into thermal equilibrium will involve extensive redistribution of ions and may, therefore, proceed with a smaller relaxation rate than a molecular liquid mixture at the same reduced temperature.

V. CONCLUSIONS

The ultrasonic attenuation spectra for an ionic binary system with critical demixing point can be well represented by the Bhattacharjee-Ferrell dynamic scaling theory. At the critical point the attenuation data (Fig. 3) nicely follow the theoretically predicted frequency dependence [Eq. (4)] and near T_c the spectra for the mixture of critical composition can be well represented by the Bhattacharjee-Ferrell scaling function $F_{BF}(\Omega)$ [Eqs. (8,9)]. No crossover corrections [48] have

TABLE I. Amplitudes of the relaxation rate and fluctuation correlation length, amount of the coupling constant, and amplitude of the ultrasonic attenuation spectrum at the critical point for EAN–n-C_8OH and some nonionic binary critical mixtures.

	Γ_o, $10^9\ \mathrm{s}^{-1}$	ξ_o, nm	$\|g\|$	S, $10^{-6}\ \mathrm{s}^{0.94}\,\mathrm{m}^{-1}$
Ethylammonium nitrate–*n*-octanol	0.22	0.47	1.3	3.0
Methanol–*n*-heptane	6.9 [62]	0.26 [63]	0.22 [62]	2.2 [62]
Ethanol–dodecane	8.6 [59]	0.34 [59]	0.1 [59]	0.72 [59]
Isobutyric acid–water	9.1 [64]	0.36 [64,65]	2.1 [64]	65 [64]
Nitrobenzene–isooctane	15 [66]	0.30 [66]		2.6 [66]
Nitrobenzene–*n*-hexane	20 [67,68]	0.35 [69]		
$HO(C_3H_6O)_nH$–$HO(C_2H_4O)_nH$	22 [70]	0.6 [70]	0.3 [70]	2.4 [70]
2,6-dimethylpyridine–water	24 [71]	0.23 [71]	0.22 [71]	8.2 [71]
Methanol–cyclohexane	35 [62]	0.32 [72]	0.24 [62]	2.1 [62]
Benzonitrile–isooctane	39 [73]	0.27 [73]	0.4 [73]	27 [73]
Perfluoromethylcyclohexane–CCl_4	47 [74]	0.23 [75]	0.16 [74]	10 [74]
Cyclohexane–nitroethane	82 [58]			5.3 [58]
3-methylpentane–nitroethane	122 [57]	0.23 [76]	0.33 [77]	29 [57]
Triethylamine–water	140 [78]	0.10,0.13 [79,80]	0.7 [78]	232 [78,81]
n-pentanol–nitromethane	181 [82]	0.14 [82]	0.11 [82]	

been applied because the ethylammonium nitrate–*n*-octanol system has been measured in a reduced temperature range ($1.5\times 10^{-4}\leqslant t\leqslant 5.7\times 10^{-2}$) in which such corrections are small [16]. Due to the unusually small amplitude $\Gamma_o(=2.6\times 10^8\ \mathrm{s}^{-1})$ in the relaxation rate of order parameter fluctuations, however, experimental data for the scaling function were attainable for reduced frequencies $\Omega\geqslant 10$, so that the agreement with the scaling function is known for the upper part of the Ω range only. In that part, the data for the ionic binary system also agree with those for the nonionic critical system ethanol–dodecane that had been studied recently over an extended Ω range.

The amplitude of the fluctuation correlation length $\xi_o=0.47$ nm and the amount $|g|=1.3$ of the coupling constant of the EAN–n-C_8OH system are rather large, but are within the range of data covered by nonionic critical mixtures. The unusually small amplitude Γ_o of the relaxation rate is to the most part due to the large viscosity of the ionic ethylammonium nitrate–*n*-octanol mixtures.

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