ABSORPTION AND DISPERSION OF ULTRASONIC WAVES IN N-AMYLIC ALCOHOL – NITROMETHANE CRITICAL MIXTURE*

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Measurement of absorption and propagation velocity of ultrasonic waves as well as shear viscosity in n-amylic alcohol–nitromethane critical mixture are reported. Acoustic measurements were performed in the frequency range from 5 to 25 MHz and temperatures $\Delta T = T - T_c$ from 0.1 to 12.3 K. An analysis of the results was carried out on the basis of a modified version of mode-coupling theory [10], which led to good agreement with experiment. The correlation length $\xi_0 = 0.94$ Å is in agreement with the value obtained from optical measurements.

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1. Introduction

In experimental and theoretical work on dynamic phenomena in the critical point a special role belongs to optical and acoustical studies of liquid media. Due to the absence of stress, such media exhibit very sharp phase transitions making them particularly well adapted for the experimental verification of theoretical predictions.

Essentially, three theories are available for the description of dispersion and absorption of acoustic waves in critical mixtures, namely: renormalisation group theory [1], dynamic scaling theory [2], and mode-coupling theory [3-7]. The last, though initially successful, failed to provide a correct account of the experimental data in a wide range of reduced frequencies ω^*. Its failure was due to two factors: firstly, the shape of the scaling function for acoustic wave absorption $I(\omega^*)$ in two

heat-mode approximation is inadequate for the description of experimental data in the range of high reduced frequencies $\omega^* > 10$ and secondly, the expression for the critical amplitude $A(\varepsilon)$ fails to predict correctly the numerical value of the absorption coefficient if the values taken to calculate $A(\varepsilon)$ result from independent thermodynamic measurements [8–9]. In an earlier paper [10] we proposed a modified expression for the critical amplitude $A(\varepsilon)$ circumventing the above difficulty. In the present paper we make use of it for the description of acoustic wave absorption in n-amylic alcohol-nitromethane critical mixture.

2. An outline of the mode-coupling theory

Within two-heat mode approximation the mode-coupling theory for the acoustic anomaly leads to the following general form of absorption coefficient $\alpha\lambda$ of ultrasonic waves in critical mixtures [3]:

$$\frac{\alpha\lambda}{u^2(\omega)} = \pi A(\varepsilon) I(\omega^*), \tag{1}$$

where $u(\omega)$ is the propagation velocity of the wave, $A(\varepsilon)$ is the critical amplitude, whereas $I(\omega^*)$ is the scaling function. Four versions of the mode-coupling theory serve to describe acoustics wave absorption in critical mixtures; they are due, respectively, to Fixman [4], Kawasaki [5], Mistura [5], and Chaban [7]. In all four the scaling function $I(\omega^*)$ is of the form:

$$I(\omega^*) = \int_0^\infty \mathrm{d}x [xg(x)]^2 \frac{\omega^* K(x)}{K^2(x) + (\omega^*)^2}, \tag{2}$$

where x is the product of the wave number k and correlation length ξ. The function $g(x)$ is related with the derivative of the logarithm of the correlation function $G(k, \xi) = \xi^{-\eta+2} f(x)$ by the expression [5]:

$$g(x) = 1 - \frac{\eta}{2} + \frac{x}{2f(x)} \frac{\mathrm{d}f(x)}{\mathrm{d}x}, \tag{3}$$

whereas $K(x)$ is a function related with the decay time $\Gamma(k, \xi)$ of fluctuations in concentration with the wave number k:

$$\Gamma(k, \xi) = \frac{1}{2}\omega_D K(k\xi). \tag{4}$$

In the theories of Kawasaki, Mistura, and Chaban $K(x)$ is given by the Kawasaki function [3]:

$$K(x) = \frac{3}{4}\left[1 + x^2 + (x^3 - x^{-1})\arctan(x)\right], \tag{5}$$

whereas in Fixman's theory by the formula:

$$K(x) = x^2(1 + x^2). \tag{6}$$

The quantity ω^* is referred to as the reduced frequency. It is defined as follows:

$$\omega^* = \omega/\omega_D, \tag{7}$$

where ω_D is the characteristic (relaxation) frequency of fluctuations in concentration. For critical mixtures, ω_D is given by the formula [9]:

$$\omega_D = 2D\xi^{-2} = 2D_0\xi_0\varepsilon^{1.93}, \tag{8}$$

where $\varepsilon = (T - T_c)/T_c$ — the reduced temperature.

In contradiction to the scaling function the expression for $A(\varepsilon)$ is of different form in each of the four theories. However, none of these expressions leads to satisfactory results in describing the experimental data. Accordingly we reanalyzed the problem of absorption and dispersion of ultrasonic waves in critical mixtures from the viewpoint of the mode-coupling theory [10]. In our analysis, proceeding similarly to Fixman and Mistura, we took into account the critical behaviour of the complex specific heat albeit applying more realistic assumptions concerning the contributions from c_p and c_V. This led to the following general expression for $A(\varepsilon)$ which, in particular cases, reduces respectively to those of Fixman, Kawasaki, Mistura, and Chaban:

$$A(\varepsilon) = \frac{k_B g^2}{\pi^2 \rho c^2 + p} \kappa \left(\frac{\partial \kappa}{\partial T}\right)^2. \tag{9}$$

with k_B Boltzman's constant, ρ the density of the mixture, κ the inverse correlation length, and g the adiabatic coupling constant introduced by Bhattacharjee and Ferrell [2] in their dynamic scaling theory.

3. Measurement of the shear viscosity coefficient

To compare the results of acoustic measurements and those provided by the theories the value of the shear viscosity coefficient η_s of the critical mixture must be known. It permits the determination of the diffusion coefficient and the characteristic frequency of concentration fluctuations. The renormalisation group theory [11] predicts a slight increase in η_s as one approaches the critical point, in accordance with the following law:

$$\eta_s = \eta(T)\xi^y = \eta(T)(\xi_0)^y \varepsilon^{-y\nu}. \tag{10}$$

The renormalization group theory, applied to determine the critical exponent y leads to a value of 0.065.

Equation (10) expresses η_s close to the critical point as the product of the regular part $\eta(T)$ and the singular part $\xi_0^y \varepsilon^{-y\nu}$. This representation poses some difficulties when it comes to the interpretation of the experimental results [12]. The procedure most commonly recurred to in this case resides in assuming that the temperature-dependence of the regular part $\eta(T)$ obeys the Arrhenius law:

$$\eta(T) = A_y \exp(B_y/k_B T), \tag{11}$$

with A_y, B_y — coefficients from viscosity values determined far from the critical temperature. On dividing the experimental values of η_s by $\eta(T)$ as determined from Eq.(11) one obtains the critical part of the shear viscosity. Thus, the quantity $\eta_s/\eta(T)$ can be expressed as follows:

$$\log[\eta_s/\eta(T)] = -\varphi \log \varepsilon + \log \eta_0, \tag{12}$$

where $\varphi = \eta y \nu$.

We measured the shear viscosity coefficient η_S in the critical mixture n-amylic alcohol–nitromethane using a Brookfield LVTD viscosimeter operating on the principle of measurements of the force of resistance due to the reaction of the medium to the rotation of the metal spindle. The accuracy in determining η_s was of the order of 1%.

To determine the coefficient of Eq. (11) we chose values of η_s for three temperatures most remote from the critical temperature. This led to the following values of the parameters:

$$A_y = 0.409 \times 10^{-3} \text{ cP },$$

$$B_y = 3.345 \times 10^{-20} \text{ J }.$$

We then determined $\eta(T)$ for each of the temperatures and plotted $\eta_s/\eta(T)$ versus the reduced temperature ε in log–log coordinates (Fig. 1). By regression analysis we obtained:

$$\eta_0 = 0.99 \pm 0.23 \text{ cP },$$

$$\varphi = 0.038 \pm 0.007$$

at a correlation coefficient of 0.991. The critical exponent value thus obtained was slightly lower than that of $\varphi = 0.041$ predicted by renormalization group theory.

The value of the exponent y is accessible to determination either from the relation $y = \varphi/\nu$ or, independently, by plotting $\eta_s/\eta(T)$ as a function of the correlation length ξ (see Fig. 2). The values of ξ for each of the temperatures was determined from the equation $\xi = \xi_0 \varepsilon^{-\nu}$ on assuming ξ_0 as equal to 1.36 Å — a value obtained from the angular dependence of the scattered light intensity [13, 14] and $\nu = 0.63$. The coefficients of regression now become:

$$\eta_0 = 0.98 \pm 0.32 \text{ cP },$$

$$y = 0.059 \pm 0.007$$

at a correlation coefficient of 0.992. Here, too, critical exponent value y is slightly lower than that predicted by the theory; however, with regard to the accuracy of the experiment, our above results can be said to agree with the theory.

As already stated, the knowledge of the viscosity coefficient η_S permits the determination of the diffusion constant D and the characteristic frequency of fluctuations in concentration ω_D from the formula:

$$D = \frac{k_B T}{6\pi\eta\xi} = \frac{k_B T_c}{6\pi\eta\xi_D}\varepsilon^{(y+\nu)} = D_0\varepsilon^{0.67}. \tag{13}$$
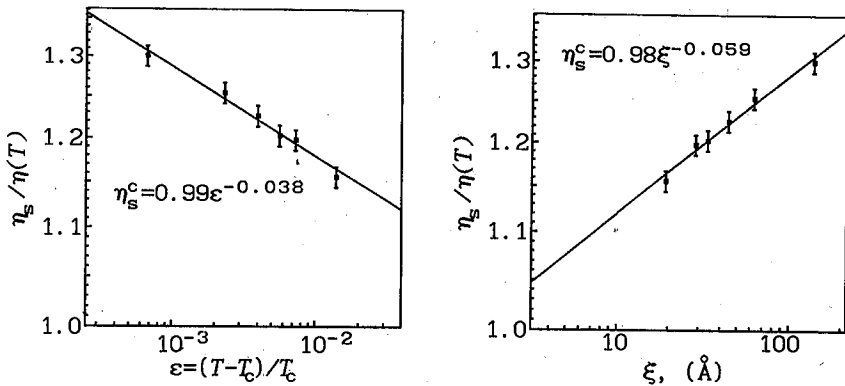
Fig. 1. $\eta_s/\eta(T)$ versus reduced temperature in n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$.

Fig. 2. $\eta_s/\eta(T)$ versus correlation length ξ in n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$.

Insertion of the values $\eta_0 = 0.98$ cP and $\xi_0 = 1.36$ Å [14, 15] leads to

$$D_0 = (1.67 \pm 0.60) \times 10^{-10} \text{ m}^2 \text{ s}^{-1} .$$

In turn, the characteristic frequency ω_D can be determined from the formula:

$$\omega_D = 2D\xi^{-2} = 2D_0\xi_0^{-2}\varepsilon^{(y+3\nu)} = \omega_0\varepsilon^{1.93}. \tag{14}$$

On insertion of these numerical values of D_0 and ξ_0 we get:

$$\omega_0 = (1.81 \pm 0.80) \times 10^{11} \text{ Hz} .$$

4. Measurements of the absorption coefficient and velocity of ultrasonic waves

Measurements of the propagation velocity and absorption coefficient of ultrasonic waves were performed by the pulse method using single emitter–receiver transducer [15]. A block diagram of the setup (mainly involving apparatuses from MATEC) is shown in Fig. 3. An electric signal with well defined frequency, measured with a digital counter, was produced in a high-stability quartz generator. It was then fed to the input of a gate modulator which produced two, mutually time-shifted sequences of rectangular pulses with controlled width and repetition frequency. The first sequence of pulses on amplification to a level of about 100 V was fed by way of an impedance matching circuit to a piezoelectric transducer emitting the ultrasonic pulses into medium. The pulses on reflection from the mirror returned to the transducer; the latter, acting as receiver, transformed them
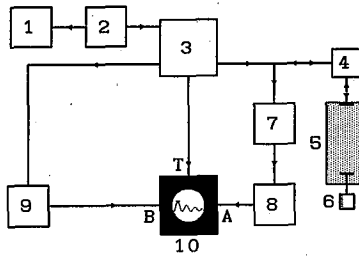
Fig. 3.   Block diagram of setup for measurements of the absorption coefficient and propagation velocity of ultrasonic waves in liquid media: 1 — digital frequency counter; 2 — sine generator; 3 — gate modulator; 4 — impedance matching; 5 — measuring cell; 6 — micrometer screw; 7 — pre-amplifier; 8 — tunable broad-band receiver; 9 — electric attenuator; 10 — oscilloscope.

into a sequence of electric signals which on amplification in a broad-band amplifier were fed to the oscilloscope.

The other sequence of pulses coming from the gate modulator was applied to measure the height of ultrasonic echo on the oscilloscope screen. To this aim, it was transmitted through an attenuator permitting pulse height measurements with an accuracy to 0.1 dB.

The measuring cell of stainless steel was 5.6 cm in height with an inner diameter of 2.5 cm. The piezoelectric transducer with an X-cut gold-coated quartz crystal was situated in the lower part of the cell; the holder in which it was situated was provided with three screws enabling parallel positioning of its surface with respect to the reflector. The latter was fixed in a holder with a micrometric screw enabling measurements of variations in distance to within 0.002 mm.

The measuring cell was placed with a copper thermal shield in which water from a ultrathermostat was made to flow. Temperature stability in the shield was better than 1 mK. The temperature inside the cell was measured with digital thermometer with an accuracy of 10 mK.

The absorption coefficient was determined measuring the height of the first echo on the oscilloscope screen as a function of variations in the distance between the transducer and reflector. Since the relation between the damping of the signal (in dB) and the change in distance Δx is of the form:

$$20 \log(I/I_0) = -\alpha \cdot \log(e)\Delta x, \qquad (15)$$

the absorption coefficient α could be determined by linear regression analysis.

When measuring the absorption coefficient by the pulse method certain factors have to be taken into account with regard to their effect on the accuracy of the results. In our case the most important correction arose from diffraction of the wave on the transducer. The effect has been analyzed in a number of papers (see e.g. [16]) where additional absorption due to diffraction has been shown to amount to about 1 dB for a distance R^2/λ, with R the radius of the transducer

and λ the ultrasonic wavelength. In our measurements the correction for diffraction loss amounted to $\alpha\lambda = 1.8 \times 10^{-4}$ at a frequency of 5 MHz. For higher frequencies the correction is negligible.

Systematic error related with measurements of the absorption coefficient was of the order of 1%; however, particularly, the accuracy was slightly lower as the result of random inaccuracies. The error stated in what follows is taken as the doubled standard deviation from three series of measurements.

The propagation velocity was determined from the formula:

$$u = \frac{2|x_1 - x_n|}{n} f, \tag{16}$$

where n is the number of interference minima on the path $|x_1 - x_n|$, whereas f is the frequency of the ultrasonic wave. The accuracy of our determination of the velocity was of the order of 0.2%.

Our measurements of the propagation velocity and absorption coefficient of ultrasonic waves in the critical mixture of n-amylic alcohol–nitromethane of concentration $x_c = 0.385$ molar fractions of $C_5H_{11}OH$ were carried out in the range of temperatures $\Delta T = T - T_c = 0.1$ K up to 12.2 K for five frequencies, namely 5, 7, 15, 21, 25 MHz.
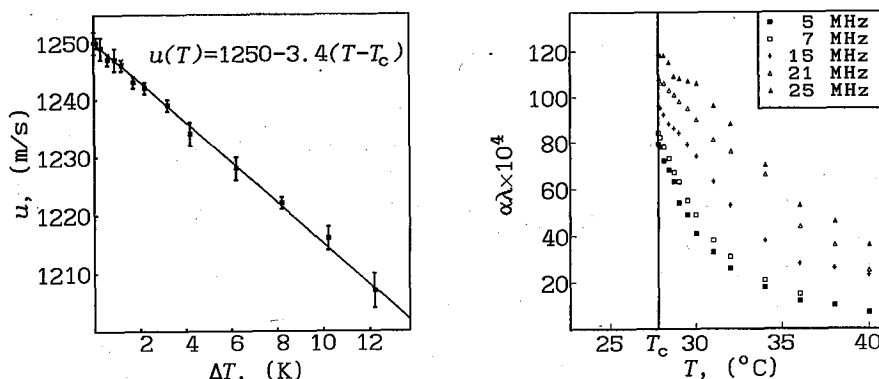

Fig. 4. Propagation velocity u versus temperature for n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$.

Fig. 5. Absorption $\alpha\lambda$ versus temperature for n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$.

The propagation velocity is plotted *vs* temperature in Fig. 4, where it is seen to grow linearly as one approaches the critical point, in accordance with the relation:

$$u = 1250 - 3.4(T - T_c). \tag{17}$$

It exhibits no anomaly in the critical point. Its behaviour is in agreement with the theory [17].

The absorption $\alpha\lambda$ in the critical mixture is shown in Fig. 5. In accordance with the theory $\alpha\lambda$ increases approaching critical point at a rate that is greater the lower is the frequency of the ultrasonic wave.

The experimentally determined absorption coefficient can be expressed as the sum of critical absorption (due to the relaxation of fluctuation in concentration) and classical absorption (due to viscosity and thermal conductivity). The latter is given by the Navier–Stokes formula [18]:

$$\alpha\lambda(NS) = \frac{\pi\omega}{\rho u^2(\omega)}\left[\frac{4}{3}\eta_S + \xi + \Lambda\left(c_V^{-1} + c_{\mathrm{p}}^{-1}\right)\right],\qquad(18)$$

where ξ — the volume viscosity coefficient, and Λ — the thermal conductivity of the medium. The term $\Lambda(c_V^{-1} + c_{\mathrm{p}}^{-1})$ can be neglected since in liquids it contributes less than 0.1% $\left(\frac{4}{3}\eta_s + \xi\right)$.

For a lack of data concerning ξ the determination of $\alpha\lambda$(NS) from Eq. (18) proved impossible. However, $\alpha\lambda$ is known to tend to $\alpha\lambda$(NS) as $f \to \infty$. Since the value of $\alpha\lambda$ is available for hyperacoustic wave of frequency $f_h = 3.5$ GHz [19], we assumed

$$\alpha\lambda_h(\mathrm{NS}) = (0.6 \pm 0.2) \times 10^{-10} f.$$

The classical absorption value can also be estimated from the value of $\alpha\lambda$ in the pure components of the mixture. The data available to us [19, 20] lead to

$$\alpha\lambda(\mathrm{NS}) = (0.87 \pm 0.1) \times 10^{-10} f.$$

Since the critical mixture n-amylic alcohol–nitromethane has hitherto not been studied experimentally, the literature fails to contain the data for $c_{\mathrm{p},x}$ and g necessary for the determination of $A(\varepsilon)$ from Eq. (9). We accordingly had recourse to the least squares method making use of the CURFIT algorithm presented in Ref. [21].

The analysis of the data was performed in four steps. In the first, we assumed for $\alpha\lambda$(NS) the value obtained from hypersonic measurements, whereas in the second step — that obtained from absorption in pure components of the mixture. In both cases similar values were obtained for the coefficients A and ω_0, as well as similar χ^2- values providing a measure of the fitting. Graphs of $\alpha\lambda/\pi u^2(\omega)A(\varepsilon)$ versus ω^* for the two cases are shown in Figs. 6 and 7, where the solid plots represent the scaling function $I(\omega^*)$ given by Eq. (2) calculated on assuming the Ornstein–Zernike expression $g(y) = (1 + y^2)^{-1}$ for the concentration fluctuation relaxation function. The integral in Eq.(2) was calculated within the limits $y_{\min} = 1 \times 10^{-5}$ and $y_{\max} = 1 \times 10^5$ by the Simpson method.

In the third and fourth steps $\alpha\lambda$(NS) as well as A and ω_0 were dealt as fitting parameters in CURFIT procedure. In the first case a lower value of χ^2 was obtained but ω_0 diverged strongly from the values obtained earlier. A graph of $\alpha\lambda/\pi u^2(\omega)A(\varepsilon)$ versus ω^* for this case is given in Fig.8.

In all the above discussed assays, our data analysis admitted only those values of $\alpha\lambda$ for which $\omega^* < 10$. This was dictated by our use of the Ornstein–Zernike formula for the determination of the scaling function. Thus, finally, we decided on
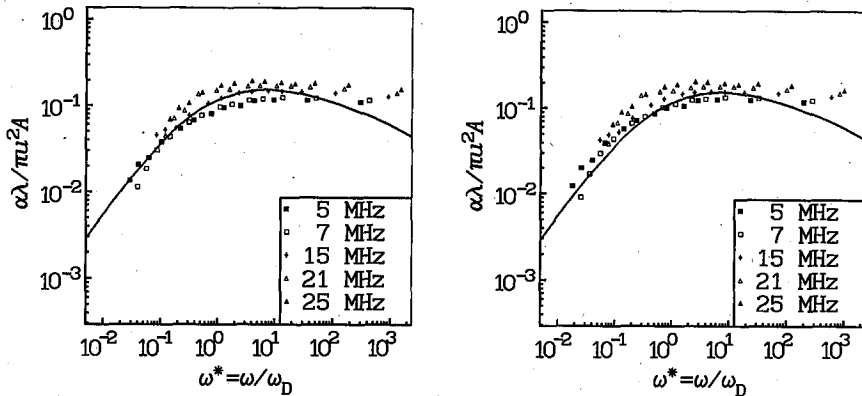
Fig. 6. $\alpha\lambda/\pi u^2(\omega)A$ versus ω^* for five ultrasonic frequencies in n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$; the coefficients $A = 0.585 \times 10^{-12}$ cm^{-2}s^2, $\omega_0 = 5.46 \times 10^{11}$ Hz were determined by CURFIT procedure; classical absorption $\alpha\lambda(NS) = 0.6 \times 10^{-10}f$ was evaluated from hypersonic measurements.

Fig. 7. $\alpha\lambda/\pi u^2(\omega)A$ versus ω^* for five ultrasonic frequencies in n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$; the coefficients $A = 0.307 \times 10^{-12}$ cm^{-2}s^2, $\omega_0 = 4.41 \times 10^{11}$ Hz were determined by CURFIT procedure; classical absorption $\alpha\lambda(NS) = 0.87 \times 10^{-10}f$ was evaluated from absorption in pure components.

an analysis applying as a correlation function the formula used by Roe and Mayer [22] in their analysis of absorption data for ^{3}He:

$$g(y) = \frac{1}{1+y^2} - \frac{1}{2}C_2 y^{-p}\frac{p + C_2 y^{-1}}{[\exp(-C_2/y)] - C_2 y^{-p}}, \qquad (19)$$

where $p \equiv (1 - \alpha)/\nu = 1.413$ and $C_2 = -0.46$. For $y \leq 1$ the function (19) coincides with the Ornstein–Zernike formula, whereas for $y > 1$ it becomes formally reminiscent of the expression proposed by Fisher and Langer [23]. However, the essential difference resides in the value of C_2 which, on theoretical ground, should be posi- tive. Albeit with $C_2 > 0$ agreement between the theory and experi- ment is very bad because the scaling function $I(\omega^*)$ exhibits a local minimum at $\omega^* \approx 20$. In this situation when putting $C_2 < 0$ the formula (19) can no longer be identified with the Fisher –Langer expression. A graph of $\alpha\lambda/\pi u^2(\omega)A(\varepsilon)$ versus ω^* plotted with the formula (19) as a correlation function agrees well with the experimental results in the whole range of reduced frequencies (Fig. 9).

With the values of A and ω_0 available we are able to determine the adiabatic coupling constant g and the correlation length of fluctuations in concentration, respectively. Equation (8) leads to:
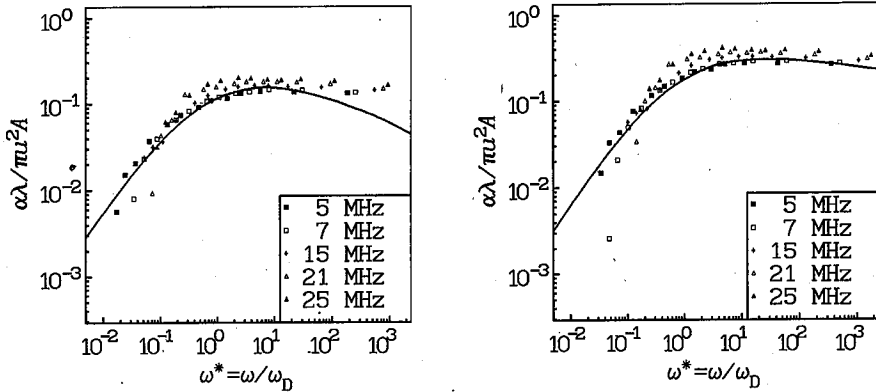
Fig. 8. $\alpha\lambda/\pi u^2(\omega)A$ versus ω^* for five ultrasonic frequencies in n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$; the coefficients $A = 0.467 \times 10^{-12}\,\mathrm{cm}^{-2}\mathrm{s}^2$, $\omega_0 = 9.0 \times 10^{11}\,\mathrm{Hz}$, $\alpha\lambda(\mathrm{NS}) = 1.66 \times 10^{-10}f$ were determined by CURFIT procedure.

Fig. 9. $\alpha\lambda/\pi u^2(\omega)A$ versus ω^* for five ultrasonic frequencies in n-amylic alcohol–nitromethane mixture at critical concentration $x_c = 0.385$; the coefficients $A = 0.239 \times 10^{-12}\,\mathrm{cm}^{-2}\mathrm{s}^2$, $\omega_0 = 4.55 \times 10^{11}\,\mathrm{Hz}$, $\alpha\lambda(\mathrm{NS}) = 1.48 \times 10^{-10}f$ were determined by CURFIT procedure; the scaling function was plotted on the basis of Eq. (2) on assuming Eq. (19) as the correlation function.

$$\xi_0 = [\omega_0/(2D_0)]^{-1/2}, \tag{20}$$

whereas Eq. (9) gives

$$g^2 = \frac{\pi^2 T_c \rho (c_{p,x}^0)^2 \xi_0^3}{A k_B \nu^2}. \tag{21}$$

On insertion of the values $\rho = 958\,\mathrm{kg/m^3}$; $T_c = 300.96$ K, $\nu = 0.63$; $c_{p,x}^0 = 2.61 \times 10^3\,\mathrm{J/kgK}$ (evaluated from the data for c_p in the pure components of the mixture); and $D_0 = 1.67 \times 10^{-10}\,\mathrm{m^2/s}$, we get g and ξ_0 for each pair of values of the coefficients A and ω_0. The results of the analysis of our absorption data obtained by CURFIT procedure with the formulae (20) and (21) are assembled in Table I.

TABLE I
Values of the coefficients and some physical parameters determined
by CURFIT procedure from our experimental results for ultrasonic
wave absorption in n-amylic alcohol–nitromethane critical mixture.

$A \times 10^{12}$ (cm^{-2}s^2)	$\omega_0 \times 10^{-11}$ (Hz)	$\alpha\lambda(NS)$ $\times 10^{10} f^{-1}$	ω_{max}	$\xi_0(\text{Å})$	g	χ^2
0.585 ± 0.002	5.46 ± 0.05	0.6 ± 0.2	10	0.95 ± 0.06	0.133	111.383
0.307 ± 0.002	4.41 ± 0.02	$0.87 + 0.01$	10	1.02 ± 0.05	0.107	114.662
0.467 ± 0.003	9.0 ± 0.2	1.66 ± 0.03	10	0.80 ± 0.08	0.092	26.874
0.239 ± 0.002	4.55 ± 0.04	1.48 ± 0.03	∞	1.00 ± 0.06	0.092	12.243

From Table I we note that the value of the correlation length ξ_0 determined
on the basis of acoustic measurements is but weakly dependent on the analytic
procedure applied. This is so because ξ_0 intervenes in both the formulae for the
critical amplitude A and in that for the characteristic frequency ω_0. Thus, the
differences in A and ω_0 cancel out mutually leaving ξ_0 essentially unaffected.

The average value of the correlation length ξ_0 derived from our acoustic
measurements amounts to:

$$\xi_0 = 0.94 \pm 0.09 \text{ Å} .$$

Since optical measurements led to [14,15]:

$$\xi_0 = 1.36 \pm 0.40 \text{ Å} ,$$

the values obtained for ξ_0 by the two methods can be said to be in agreement to
within experimental errors.

The average value of the adiabatic coupling constant g obtained from our
acoustic measurements amounts to

$$g = 0.106 \pm 0.018.$$

To our knowledge, this is the lowest value of g hitherto reported in the lit-
erature [24–28]. However, since absorption $\alpha\lambda(T_c)$ in the critical system n-amylic
alcohol–nitromethane is also lower that in other systems dealt with in Refs. [24–28]
the above is in agreement with the interpretation of g as a measure of the absorp-
tion intensity in the critical point.

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