SOFT MATTER

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RELAXATION TIME OF CONCENTRATION FLUCTUATIONS IN A VICINITY OF THE CRITICAL STRATIFICATION POINT OF THE BINARY MIXTURE n-PENTANOL–NITROMETHANE

The propagation velocity and the absorption coefficient of ultrasound in a frequency range of 5–2800 MHz in a n-pentanol–nitromethane solution in a vicinity of its critical stratification point from the homogeneous state side have been studied. The research make it possible to reveal the influence of concentration fluctuations on the sound propagation velocity. Three regions of dynamical parameters are analyzed: the mean-field ($\omega\tau_{\mathrm{FL}} \ll 1$), fluctuation ($\omega\tau_{\mathrm{FL}} \gg 1$), and transition (crossover, $\omega\tau_{\mathrm{FL}} = 1$) ones. On the basis of experimental data, the temperature dependence of the concentration fluctuation relaxation time $\tau(T)$ is studied, and its magnitude τ_0 is determined. The contribution to the fluctuation part of the sound absorption coefficient at high frequencies ($\omega > 300$ MHz), which is connected with the sound scattering by concentration fluctuations near the critical stratification point is estimated.

K e y w o r d s: ultrasound velocity, ultrasound absorption, critical stratification point, binary solution, concentration fluctuations, relaxation time.

1. Introduction

An extremely unusual and, simultaneously, universal behavior of binary solutions, different by their nature, near their critical stratification points continues to be a matter of scientific discussions for several decades. The observed anomalies of various transport quantities near the critical point are conventionally associated with order parameter fluctuations and explained in the framework of the dynamical theories of critical phenomena [1–8]. In those researches, besides the characteristic correlation length R_{c}, the examined media are also characterized by a characteristic time scale, namely, the relaxation time of fluctuations near

the critical point, τ_{FL}. This parameter can be determined with the help of various, in particular, acoustic methods [9].

The inherent distinction and the undoubted advantage of acoustic spectroscopy consist in the opportunity to study both static properties of the system (e.g., the velocity of sound propagation) and dynamical ones (e.g., the coefficient of sound absorption, which makes it possible to determine relaxation times in the system). The values of corresponding relaxation times for order parameter fluctuations, τ_{FL}, can be obtained by means of various experimental methods (nuclear magnetic resonance, dielectric spectroscopy, neutron scattering, light scattering, *etc*). However, the results obtained with their help can differ from one another by several orders of

magnitude. For example, according to the method of quasielastic neutron scattering, the relaxation time of concentration fluctuations in a close vicinity of the critical stratification point ($\Delta T = 0.5$ K) amounts to $\tau_{\text{FL}} \sim 10^{-7}$ s [10], which coincides with the data of IR spectroscopy [11]. However, these results differ by several orders of magnitude from those obtained with the use of the light scattering technique [12]. Even the results of different acoustic experiments concerning the relaxation time are different [9,13]. This situation is related to the fact that the majority of experimental researches in the critical region [14] were carried out in a narrow interval of frequencies, f, and only for solutions with critical concentrations [15], which diminishes the reliability of the results obtained. The indicated shortcomings can be eliminated by complex acoustic researches of the propagation velocity c_s and the absorption factor αf^{-2} of ultrasound. Those researches should be carried out in a wide frequency interval and in various regions of the parameter $\omega \tau_{\text{FL}}$ ($\omega = 2\pi f$), namely, in the region far from the critical point (at $\omega \tau_{\text{FL}} \ll 1$), where the mean-field theory is actual; in the critical region (at $\omega \tau_{\text{FL}} \gg 1$), where the fluctuation theory is actual, and in the crossover region (at $\omega \tau_{\text{FL}} \approx 1$).

Therefore, the aim of this work was to determine the relaxation time of concentration fluctuations in a vicinity of the critical stratification point. For our researches, we chose the binary solution of n-pentanol–nitromethane at the corresponding critical concentration. The propagation velocity c_s and the absorption coefficient αf^{-2} of ultrasound were studied in a wide frequency range of 5–2800 MHz. The examined solution was in a homogeneous state at a temperature above its critical temperature of stratification, T_c.

2. Experimental Specimens and Technique

The binary solution of n-pentanol–nitromethane selected for experimental researches has the upper critical temperature of stratification T_c at the room-temperature level, which simplifies the experimental procedure. For the preparation of solutions, n-pentanol and nitromethane of the chemically pure grade with various concentrations within an interval of 0.1–0.8 mole fractions were used. The propagation velocity and the sound absorption coefficient in the solutions near the critical temperature of stratification were performed using temperature-controlled

specimens. The temperature stabilization error was $\pm 10^{-3}$ K.

The absorption coefficient and the propagation velocity of sound were studied experimentally using the pulse method, which was described in work [16] in detail. The pulse method with variable distance and the method of resonance excitation of a quartz piezocrystal were used in a frequency interval of 5–150 MHz, and the method of non-resonance excitation of a lithium niobate single crystal in a high-frequency range of 300–2800 MHz were applied. Depending on the experimental conditions, the relative measurement errors for the propagation velocity and the absorption coefficient of ultrasound amounted to 2–5%.

3. Estimation of the Fluctuation Relaxation Time on the Basis of the Data for the Sound propagation Velocity

In the dynamical scaling theory [1, 2], the variation of the fluctuation relaxation time, when approaching the critical temperature of stratification T_c, is given by the formula

$$R_c = R_0\, t^{-\nu}. \tag{1}$$

Here, τ_0 is the amplitude of the temperature-dependent relaxation time for concentration fluctuations; $t = (T - T_c)/T_c$; $Z = D + X_\eta = 3.065$ [2,3,15] is the dynamical critical exponent, which is determined by the space dimensionality D and the exponent in the temperature dependence of the dynamic viscosity $X_\eta = 0.065$ [13]; ν is the critical index in the temperature dependence of the correlation length R_c,

$$R_c = R_0\, t^{-\nu}, \tag{2}$$

and R_0 the amplitude of the temperature-dependent correlation length of concentration fluctuations. For the studied solution, $R_0 = 0.14 \pm 0.8$ nm [14], so that $\nu = 0.630$.

According to the dynamical theory of critical phenomena [1–3], we may write the following expression that connects the relaxation time of concentration fluctuations with the fluctuation correlation length:

$$\tau_{\text{FL}} = \frac{6\pi \eta_s}{k_B T}\, R_c^3. \tag{3}$$

Here, $\eta_s = \eta_0(T)(r_0 q)^{Z_\eta} t^{-\nu Z_\eta}$ is the dynamical viscosity, and η_0 the amplitude in the temperature dependence of the dynamic viscosity. The latter parameter for the examined solution was determined

by means of viscosimetric researches [19]. Its value amounts to $\eta_0 = 0.98 \pm 0.05$ mPa.

From Eqs. (1)–(3), we obtain the following expression for the amplitude τ_0 of the temperature-dependent relaxation time of concentration fluctuations:

$$\tau_0 = \frac{3\pi\eta_0 R_0^3}{k_B T}. \qquad (4)$$

This formula can be used to calculate the parameter τ_0 near the critical stratification point. Then, using the experimental values of R_0 and η_0 [15], we obtain $\tau_0 = 0.56 \times 10^{-11}$ s for the examined n-pentanol–nitromethane solution. This result agrees with the data of works [9, 13, 15].

The amplitude τ_0 of the relaxation time for concentration fluctuations can be determined experimentally with the help of acoustic spectroscopy. For this purpose, we researched the frequency and temperature dependences of the sound propagation velocity in an n-pentanol–nitromethane solution in a frequency range of 5–2800 MHz and a temperature interval of 300.9–320 K. We studied solutions with various concentrations in an interval of 0.1–0.9 mole fractions of n-pentanol, including the solution with the critical concentration. In accordance with work [15], the latter equals $X_c = 0.385$.

In Fig. 1, experimental data for the temperature dependence of the sound propagation velocity in the examined solution at three frequencies – $f_1 = 5$ MHz, $f_2 = 150$ MHz, and $f_3 = 2800$ MHz – are depicted. One can see that, for the frequency $f_1 = 5$ MHz, the sound velocity in the n-pentanol–nitromethane solution with the critical concentration $X_c = 0.385$ decreases almost linearly as the temperature grows. On the contrary, at the frequency $f_2 = 150$ MHz and provided the same solution concentration, a deviation from the indicated linear dependence is observed, when approaching the critical temperature of stratification. The analysis of the obtained experimental data testifies that the observed deviation from the linear temperature dependence increases with the sound frequency. In particular, the deviation maximum was registered at the frequency $f_3 = 2800$ MHz (see Fig. 1).

In our opinion, the linear dependence $c = c(T)$ taking place at a certain frequency testifies that, at this frequency, the studied solution "does not feel" its approach to the critical state, i.e., in accordance with the dynamical scaling theory, $\omega\tau_0 \ll 1$ in this case. In
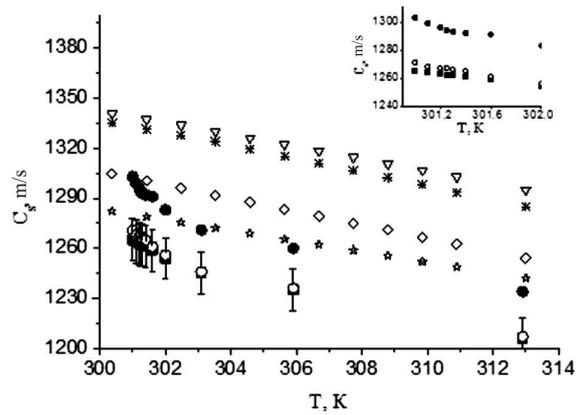
Fig. 1. Temperature dependences of the sound propagation velocity in the n-pentanol–nitromethane solution with the concentration $X_c = 0.385$ at various frequencies: $f_1 = 5$ MHz (■), $f_2 = 150$ MHz (○), and $f_3 = 2800$ MHz (●); and temperature dependences of the sound propagation velocity in the solution components: in pentane at the frequencies $f_1 = 5$ MHz (★) and $f_3 = 2800$ MHz (∗), and in nitromethane at the frequencies $f_1 = 5$ MHz (◇) and $f_3 = 2800$ MHz (▽)

other words, the obtained linear dependence $c = c(T)$ confirms that the mean-field theory is valid in this $\omega - T - X$ parameter region. The deviation from the linear dependence $c = c(T)$, which is observed at $f_2 = 150$ MHz, testifies that the experimental conditions in this case correspond to the crossover region, where $\omega\tau_{FL} \sim 1$. A substantial deviation from the indicated linear dependence, when approaching the critical temperature, was observed in our experiment at the frequency $f_3 = 2800$ MHz. This fact means that the relevant experimental conditions correspond to the fluctuation region of the dynamical scaling theory.

The presented analysis of the temperature dependence of the sound propagation velocity makes it possible to estimate the amplitude for the relaxation time of concentration fluctuations in the n-pentanol–nitromethane solution, when it approaches its critical point of stratification at a distance of 0.1 K from above. According to our evaluation, which was carried out with the use of the relation $\omega\tau_0 \approx 1$, this parameter equals $\tau_0 \approx 10^{-11}$ s.

4. Estimation of the Fluctuation Relaxation Time on the Basis of the Data on Sound Absorption

In order to calculate the relaxation time for concentration fluctuations with the use of the data on

the coefficient of sound absorption in the binary n-pentanol–nitromethane solution at the critical concentration $X_c = 0.385$ (this is the mole fraction of n-pentanol), it is necessary, first of all, to determine the fluctuation part of the absorption coefficient. The latter is connected with concentration fluctuations, being a component of the total absorption coefficient normalized by the squared frequency [20]:

$$\left(\alpha f^{-2}\right)_{\text{FL}} = \left(\alpha_{\text{EF}} f^{-2}\right) - \left(\alpha f^{-2}\right)_{\text{REG}}. \tag{5}$$

Here, $\alpha_{\text{EF}} f^{-2}$ is the total effective coefficient of sound absorption along the isoconcentration curves, which is a function of the frequency and the temperature; $\left(\alpha f^{-2}\right)_{\text{REG}}$ is the corresponding regular part associated with structural features of the studied solutions at insignificant fluctuations; and $\left(\alpha f^{-2}\right)_{\text{FL}}$ is the fluctuation part of the absorption coefficient, which is associated with the processes that occur near the critical temperature of stratification. Note that the fluctuation part of the effective coefficient of sound absorption $\alpha_{\text{EF}} f^{-2}$ takes into account both the real sound absorption and the sound scattering by concentration fluctuations in the frequency interval, where the experimental sound wavelength becomes comparable with the correlation length of fluctuations, i.e. at frequencies $f \geq 300$ MHz.

As one can see from formula (5), in order to find the fluctuation part of the absorption coefficient, we have to determine the regular component. The latter dominates in the interval far from the critical point, i.e. in the hydrodynamic region. Therefore, the experimental researches of solutions with various concentrations were carried out at temperatures exceeding T_c by 20–30 K. The magnitude of the regular part was found at a considerable distance from the critical point and extrapolated into the critical region. Then $\left(\alpha f^{-2}\right)_{\text{FL}}$ was determined, by using formula (5).

The extracted fluctuation part of the absorption coefficient was analyzed in the framework of the phenomenological theory of heat capacity relaxation [20, 21]:

$$\left(\alpha_{\text{EF}} f^{-2}\right)_{\text{FL}} = A f^{-\left[1 + \frac{\alpha_t}{z_\nu}\right]} F(\Omega), \tag{6}$$

where $F(\Omega)$ is a scaling crossover function of the relative frequency $\Omega = 2\pi f \tau_{\text{FL}}$, τ_{FL} the relaxation time of concentration fluctuations, and α_t the critical index of isochoric heat capacity ($\alpha_t = 0.091$ [19]). The quantity A is determined by specific features of the

examined solution. It can be calculated by the formula [15, 20]

$$A = \frac{\pi^2 \alpha_t C_{pc}}{2Z\nu T_c} \left(\frac{\Omega_{1/2}}{2\pi \tau_0}\right)^{\frac{\alpha_t}{z_\nu}} \frac{c_c g^2}{C_{pr}^2}, \tag{7}$$

where g is the adiabatic coupling constant (for the n-pentanol–nitromethane solution, $g = 0.106$ [15]), $\Omega_{1/2} = 2.1$ is a constant that is determined by analyzing the form of the scaling crossover function for the solution concerned, $c_{s,c}$ is the sound velocity at $T = T_c$ (according to the Ferrell–Bhattacharjee theory of relaxing heat capacity [20], this parameter is finite for binary solutions; in particular, for the solution concerned, $c_c = 982$ m/s), C_{pr} is the regular part of the specific heat capacity at a constant pressure (according to work [22], $C_{pr} = 2.61 \times \times 10^3$ J/(kg K)), and C_{pc} the specific heat capacity at $T = T_c$ ($C_{pc} = 8.2 \times 10^5$ J/(kg K) [22]).

The calculation by formula (7) gives rise to the value $A = (2.0 \pm 0.5) \times 10^{-5}$ s$^{0.94}$/m, which coincides with the literature data [15] within the calculation errors.

In view of the expression for the scaling crossover function [3],

$$F(\Omega) = \frac{1}{\left[1 + 0.414 \left(\frac{\Omega_{1/2}}{\Omega}\right)^n\right]^2}, \tag{8}$$

Eq. (6) reads

$$\left(\alpha_{\text{EF}} f^{-2}\right)_{\text{FL}} =$$
$$= A f^{-\left[1 + \frac{\alpha_t}{z_\nu}\right]} \left[1 + 0.414 \left(2\pi f \,\Omega_{1/2} \tau_{\text{FL}}\right)^n\right]^{-2}. \tag{9}$$

This formula makes it possible to determine the relaxation time of concentration fluctuations in the researched solution on the basis of experimental data on the effective coefficient of sound absorption at various temperatures near the critical stratification point.

There are a few works [21, 23, 24], in which their authors determined the magnitudes of the parameters that characterize the behavior of the scaling crossover function. In particular, Bhattacharjee and Ferrell in work [21] found the values of the exponent $n = 0.50$ and the constant $\Omega_{1/2} = 2.1$. Folk and Moser in work [23] reported that $n = 0.63$ and $\Omega_{1/2} = 3.1$. On the other hand, Onuki [24] calculated that $n = 0.50$ and $\Omega_{1/2} = 6.2$. Our analysis of the obtained experimental data shows that the Ferrell–Bhattacharjee theory

[21] with $n = 0.52$ and $\Omega_{1/2} = 1.9$ provides the best agreement with the experiment.

Formula (9) was used to approximate the fluctuation part of the absorption coefficient (see Eq. (5)) in the n-pentanol–nitromethane solution with the critical concentration. Then we determined the relaxation time of concentration fluctuations, τ_{FL}, for various temperatures near the critical point of stratification. The results obtained are quoted in Table 1.

The relaxation time of concentration fluctuations in the solution can also be found in a different way, namely graphically. For this purpose, let us plot the dependence of the fluctuation part of the absorption coefficient (5) on the quantity $\ln\left(f^{-\left[1+\frac{\alpha t}{Z\nu}\right]}\right)$ for several temperatures (see Fig. 2) and analyze them. One can see that, when the sound frequency increases (the quantity $\ln\left(f^{-\left[1+\frac{\alpha t}{Z\nu}\right]}\right)$ decreases at that), the sound absorption coefficient, in accordance with formula (9), almost linearly decreases within the interval from the lowest experimental frequency (5 MHz) to a certain "threshold" frequency f_p. At frequencies above the threshold one, the behavior of the temperature dependence of the sound absorption coefficient considerably changes. Namely, it does not decreases so rapidly and also forms an almost linear dependence of the effective absorption coefficient on the quantity $\ln\left(f^{-\left[1+\frac{\alpha t}{Z\nu}\right]}\right)$, but with a different slope. This phenomenon can be explained by the fact that, at a certain threshold frequency, the ultrasound wavelength becomes comparable with the correlation length maximum for those concentration fluctuations that are typical at this temperature in a vicinity of the critical point. The further increase of the frequency results in that the correlation length of other fluctuations near the critical point (we assume that they have a certain distribution over the dimensions) become comparable with the sound wavelength. This model allows the maximum value of fluctuations in their distribution over the correlation length to be evaluated. In particular, the correlation length equals $R_c = 25$ nm at the temperature $T = 301.1$ K. Using these data for the correlation length near the critical point and formula (3), we can determine the relaxation time for concentration fluctuations.

Thus, the experimental value of the effective sound absorption coefficient $\left(\alpha_{\text{EF}}f^{-2}\right)_{\text{FL}}$, which "reflects" sound attenuation at the sound propagation from a
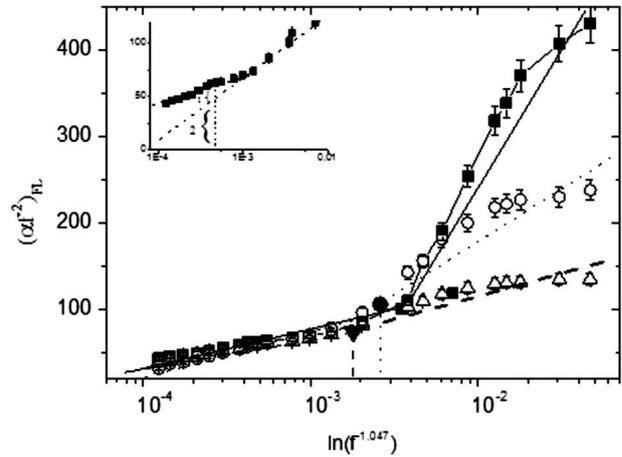
Fig. 2. Dependences of the fluctuation component in the effective coefficient of sound absorption in the n-pentanol–nitromethane solution with the critical concentration on the quantity $\ln\left(f^{-1.047}\right)$ at various temperatures $T = 301.1$ (■), 302 (○), and 313 K (△). The components of the effective coefficient of sound absorption are shown in the inset: the contribution associated with the ultrasound scattering by concentration fluctuations (1) and the coefficient of intrinsic sound absorption normalized by the squared frequency (2)

source to a detector through the solution concerned, contains, besides the term $\left(\alpha f^{-2}\right)_{\text{FL}}$ describing the intrinsic sound absorptions, one more term, B_p, which corresponds to the sound scattering by concentration fluctuations, i.e.

$$\left(\alpha_{\text{EF}}f^{-2}\right)_{\text{FL}} = \left(\alpha f^{-2}\right)_{\text{FL}} + B_p. \tag{10}$$

The ratio between the terms in formula (10) can be evaluated by analyzing the inset in Fig. 2. Namely,

Relaxation time τ_{FL} of concentration fluctuations in a vicinity of the critical stratification point of the n-pentanol–nitromethane solution

Temperature, K	τ_{FL}, ns
301.1	27000
301.2	8000
301.3	3600
301.4	2000
301.5	1300
301.6	800
302	10
303	70
313	3
323	0.5
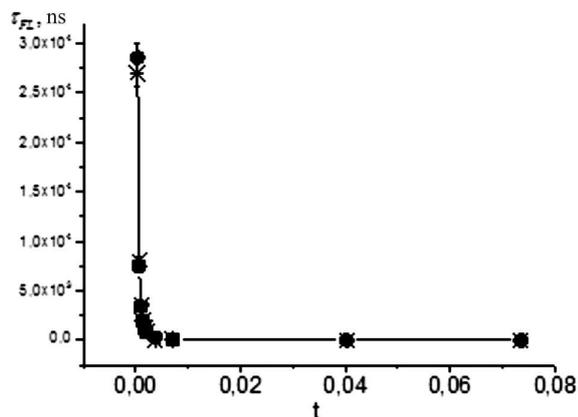
Fig. 3. Temperature dependence of the relaxation time of concentration fluctuations. Circles correspond to the results obtained, by using formula (9), asterisks to the results obtained graphically, and the solid curve to the approximation of experimental data by formula (1)

the scattering of 150-MHz ultrasound by concentration fluctuations in the n-pentanol–nitromethane solution at the temperature $T = 301.1$ K (this value exceeds the critical temperature by 0.1 K) equals 8% of the effective sound absorption coefficient. This means that, at this frequency, the actual sound absorption amounts to 92% of the effective absorption coefficient. At the same time, at $f = 2800$ MHz, the share of ultrasound scattering increases and amounts to 28%, so that the share of ultrasound absorption equals 72%.

Figure 3 demonstrates the relaxation time $\tau_{\rm FL}$ of concentration fluctuations found for the n-pentanol–nitromethane solution, by using two methods. The data obtained were approximated with the help of formula (1). One can see that the temperature dependence of the relaxation time $\tau_{\rm FL}$ for concentration fluctuations is described by relation (1). This fact confirms the correctness of our calculations.

5. Conclusions

1. The propagation velocity and the absorption of ultrasound with the frequencies 5–2800 MHz in the n-pentanol–nitromethane solution in a vicinity of its critical state with respect to both the concentration and the temperature (from the homogeneous-state side) have been studied.

2. The influence of concentration fluctuations on the sound propagation velocity is revealed. Three do-

mains of the dynamical parameters were examined; with the mean-field theory being valid in one of them, the fluctuation theory in the other, and the third domain is a transition (crossover) region.

3. The data for the sound propagation velocity are used to determine the amplitude of the temperature dependence of the relaxation time for concentration fluctuations near the critical point of stratification in the n-pentanol–nitromethane solution.

4. The data on the sound absorption coefficient in the examined solution are used to determine the temperature dependence of the relaxation time for concentration fluctuations.

5. The contribution of the component associated with the sound scattering by concentration fluctuations near the critical point of stratification to the total effective sound absorption coefficient is estimated.

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ЧАС РЕЛАКСАЦІЇ ФЛУКТУАЦІЙ
КОНЦЕНТРАЦІЙ ПОБЛИЗУ КРИТИЧНОЇ
ТОЧКИ РОЗШАРУВАННЯ БІНАРНОГО
РОЗЧИНУ Н-ПЕНТАНОЛ-НІТРОМЕТАН

Р е з ю м е

Проведені дослідження швидкості поширення та коефіцієнта поглинання звуку в розчині н-пентанол-нітрометан в інтервалі частот 5–2800 МГц при підході до його критичної точки розшарування з боку гомогенного стану дозволили визначити вплив флуктуацій концентрації на акустичні параметри. В термінах динамічних параметрів досліджено три області: середнього поля $\omega\tau_{\mathrm{FL}} \ll 1$, флуктуаційна $\omega\tau_{\mathrm{FL}} \gg 1$, та перехідна – кросоверна $\omega\tau_{\mathrm{FL}} = 1$. На основі експериментальних даних в роботі досліджено температурну залежність часу релаксації флуктуацій концентрації $\tau(T)$, визначено його амплітудне значення τ_0. На високих частотах ($\omega > 300$ МГц) оцінено внесок у флуктуаційну частину коефіцієнта поглинання звуку, який пов'язаний із розсіянням звуку на флуктуаціях концентрацій поблизу критичної точки розшарування.