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AMPLITUDE AND PHASE FLUCTUATIONS OF AN ULTRASONIC WAVE IN A NONUNIFORM MEDIUM

MIKOŁAJ ŁABOWSKI

Acoustics Department, UAM (60-769 Poznań)

The paper presents the method of investigation of the results obtained for the fluctuation of quantities of the wave field in CO_2 close to the critical state. The method used the phenomenon of light diffraction by an ultrasonic wave. The analysis of the experimental results was performed by a correlation method, in which the autocorrelation functions and the power spectra of the fluctuation were determined for individual thermodynamic conditions and various frequencies of the ultrasonic wave.

1. Introduction

An ultrasonic wave, propagating in a non-uniform medium, (e.g. in a liquid-liquid or liquid-vapour system under critical conditions) where large fluctuations in concentration or density may occur, undergoes certain deformations. The deformations are seen in the fluctuation of characteristic quantities of the wave field due to the superimposition of the primary wave and the waves scattered by the heterogeneities of the medium.

If a medium with a fluctuation in density is characterized by the refraction index which is subject to fluctuation, a definite relation can be found between the fluctuations of the coefficient and the characteristic quantities of the wave field.

The quantities of the wave field which are subject to fluctuation are primarily amplitude and phase.

Chernov [1] gives two methods for deriving a general expression for the amplitude and phase fluctuations of a wave propagating in a non-uniform medium. One is the method of small perturbations, the other, a more general approach, is Rytov's method.

The basic method permitting the evaluation of diffraction effects in the case of large heterogencities (i.e., $\lambda \ll a$, where a is the size of heterogencity) is the method derived by Rytov [2], and developed by Obukhov [3], Chernov [4] and others. The formulae that they derived give the dependence of the phase and amplitude fluctuations of a wave propagating in a non-uniform medium on the statistical properties of the medium characterised by the function $\mu(\xi, \eta, \zeta)$. The statistical process of the fluctuation of the refraction coefficient was considered to be stationary, so that the dependence of μ on time could be neglected. Describing such a process with a correlation function and knowing its form, the mean squares of the fluctuations of the phase $\overline{S_1^2}$ and of the amplitude $\overline{B^2}$ of the wave can be calculated.

2. Use of the phenomenon of light diffraction by an ultrasonic wave for the investigation of the amplitude and phase fluctuations of the wave propagating a non-uniform medium

Pictures of the diffraction of light by an ultrasonic wave can for such a medium give information on the heterogeneity of the medium and also on the deformation of the ultrasonic wave itself. The phenomenon of light diffraction by an ultrasonic wave propagating in a non-uniform medium is considerably more complicated [5] than the well-known phenomenon in a uniform medium [6-14].

The phenomenon of light diffraction by an ultrasonic wave in a medium where large fluctuations in density or concentration occur is such that the resulting diffraction image is also subject to fluctuation [15-20]. The light intensity fluctuates in the individual diffraction orders, and also does the angle of diffraction of the light wave. The character of the phenomenon is different from that of the RAMAN-NATH theory [6]. ŚLIWIŃSKI [5] attempted to consider the effect of the fluctuations of the medium on the diffraction image. Fluctuations in the density of the medium cause amplitude and phase fluctuations of the ultrasonic wave which causes an additional change in the coefficient of light diffraction. Considering these changes he derived an expression for the instantaneous distribution of the light diffracted over the plane of departure from the ultrasonic layer.

The mean value of the angle of diffraction that is subject to fluctuation is given by the expression [5]:

$$\operatorname{tg} \alpha_p = \frac{p}{k}\left(K + \frac{\beta}{2\gamma}\right), \tag{1}$$

where $k = 2\pi/\lambda$, $K = 2\pi/\Lambda$, p is the order of the spectrum, with β, γ being functions of the mean squares of fluctuation in phase $\overline{S^2}$ and amplitude, $\overline{B^2}$, of the ultrasonic wave, given by the following formulae:

$$\beta = \frac{R_{BS_1}}{2\sqrt{\overline{B^2}}\sqrt{\overline{S_1^2}(1-R_{BS}^2)}}\,; \quad \gamma = \frac{1}{2\overline{S_1^2}(1-R_{BS_1}^2)}.$$

R_{BS_1} is the correlation coefficient between B and S_l which is defined by

$$R_{BS_1} = \frac{\overline{BS_1}}{\sqrt{\overline{B^2}\cdot\overline{S_1^2}}}.$$

2.1. The method of measurement of the fluctuations in the ultrasonic wave. The light intensity in the individual diffraction orders and the angles of diffraction of the light waves are subject to fluctuations. Owing to the fluctuation of the diffraction angles the widths of individual diffraction lines change. The mean width of these lines, defined as the "half" width of the curve of the light intensity distribution over a line, is a measure of the diffraction angle which in turn may be related to the fluctuations of the amplitude and the phase of the ultrasonic wave.

It is possible to develop a method of defining the magnitude of the fluctuations of the amplitude and the phase of the ultrasonic wave propagating in a medium close to the critical point, based on the measurement of the diffuseness of the width of diffraction lines of the light diffracted by the wave [28]. A schematic diagram of the light diffracted by a scattered ultrasonic wave is shown in Fig. 1.

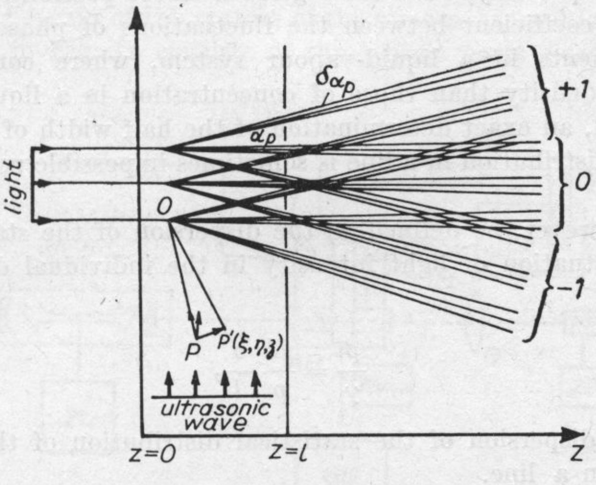

Fig. 1. A diagram of light diffraction by a scattered ultrasonic wave, according to [5]

Relation (1) can be written for small angles in the form

$$\operatorname{tg}(\alpha_p+\delta\alpha_p) \approx \sin(\alpha_p+\delta\alpha_p) = p\left(\frac{\lambda}{\Lambda}+\frac{\beta}{2\gamma}\right), \tag{2}$$

whence

$$\alpha_p+\delta\alpha_p = p\left(\frac{\lambda}{\Lambda}+\frac{\beta}{2\gamma}\right). \tag{3}$$

Since $\alpha_p = p\lambda/\Lambda$, and $\delta\alpha_p = \Delta p/l$, $\delta\alpha_p = p\beta/2\gamma$, where p is the order of the spectrum, $\delta\alpha_p$ is the fluctuation in the diffraction angle, β and γ are functions of the mean squares of the fluctuations of the amplitude and phase of the ultrasonic wave [6, 13, 21, 15], λ is the wavelenght of the light, Λ is the ultrasonic wavelength, Δ_p is the mean width of the pth line, and l is the distance of the objective from the recording photographic film (or photo multiplier).

The ratio β/γ can be expressed in the following way [22]:

$$\frac{\beta}{\gamma} = \frac{\overline{B \cdot S_1}}{\overline{S_1^2}} = R_{\beta S_1} \sqrt{\frac{\overline{B^2}}{\overline{S_1^2}}}\,. \tag{4}$$

From a determination of the width of the diffuseness of a line and with knowledge of the distance l, the ratio β/γ which is defined in terms of the mean squares of the fluctuations of amplitude and phase of the ultrasonic wave, can be found. This ratio can be assumed to be the measure of the fluctuation of an ultrasonic wave propagating in a medium close to the critical state.

From the width of the diffuseness of the diffraction lines, information can be obtained on both the phase and the amplitude fluctuations of the ultrasonic waves, albeit, in an involved form. If we know the fluctuations of the phase and the amplitude separately, their ratio gives a direct possibility of determining the correlation coefficient between the fluctuations of phase and amplitude.

In experiments in a liquid-vapour system, where considerably larger fluctuations in density than those of concentration in a liquid-liquid system can be observed, an exact determination of the half width of the curve of the light intensity distribution in a line is sometimes impossible using photographic methods.

It may, however, be defined as the dispersion of the statistical distribution of the fluctuation of light intensity in the individual diffraction orders [5, 15]. Then

$$\frac{\beta}{\gamma} = \frac{K}{p}\frac{\sigma}{l},$$

where σ is the dispersion of the statistical distribution of the fluctuation of light intensity in a line.

This paper presents the results of the investigation of fluctuations of quantities of the ultrasonic wave field in CO_2 close to the critical state, using a method involving the diffraction of light by an ultrasonic wave. Due to a linear relation between the intensity (not too high an intensity*) of the ultrasonic wave, and the intensity of the light in the diffraction figures, this method permits the fluctuations of quantities of the wave field to be investigated through measurement of the fluctuation of the light diffracted by this wave.

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* When only lines of the ± 1 orders, and none of higher orders occur.

Fluctuations of the intensity of the light diffracted by an ultrasonic wave at a few chosen frequencies (0.8, 1.2, 3.51, 5.76, 27.8 MHz) propagating in CO_2 which was held in a state close to the critical point where distinct fluctuations in density occur, were investigated.

The analysis of the experimental results for a liquid-vapour system was performed by a correlation method, in which the autocorrelation functions and the power spectra of the fluctuation for specific thermodynamic conditions and various frequencies of the ultrasonic wave [20] were determined.

It should be noted that the object of the investigation was the slow ("coarse grained") fluctuations connected with fluctuations of the thermodynamic state close to the critical point, which have also been investigated using the photographic method, by Śliwiński [23] and mentioned by **Brown** [24]. For purely technical reasons the fast ("fine grained") fluctuations of light diffracted by the ultrasonic wave (which also occur when this wave is absent), which cause the well-known phenomenon of optical opalescence, have not been investigated so far.

3. Apparatus and investigation method

A general diagram of the system of the apparatus used is shown in Fig. 1a. The apparatus in Fig. 1a consists of three basic parts closely linked to each other:

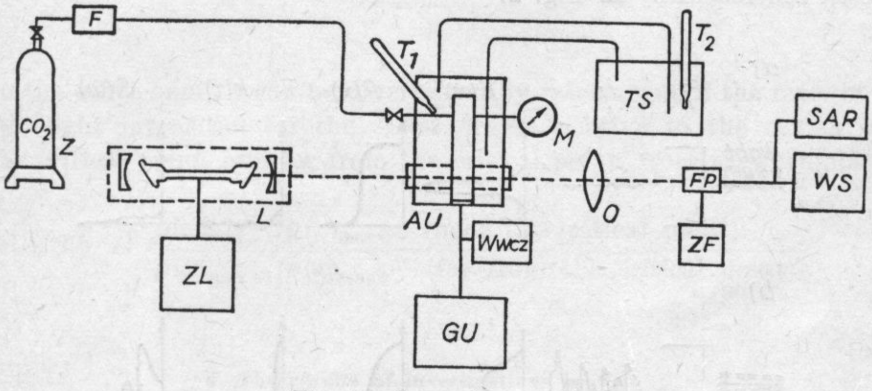

Fig. 1a. General apparatus

Z – container of CO_2, F – drying filter, AU – ultrasonic autoclave (author's design [25]), GU – high frequency generator, WW_{cz} – high frequency voltmeter, TS – Höppler thermostat, T_1, T_2 – thermometers, M – manometer, L – He-Ne laser, ZL – laser power supply, O – objective, FP – photomultiplier, ZF – photomultiplier power supply, WS – voltmeter pen recorder, SAR – statistical distribution analyser

(1) A system for generating adequate thermodynamic conditions, i.e. an adequate ultrasonic autoclave [25] with a thermostat combined with pressure and temperature meters.

(2) A system propagating an ultrasonic wave into the autoclave.

(3) An optical system with photographic or photoelectrical recording.

A laser beam of 2.5 mm diameter passes through the ultrasonic autoclave where there is a hetereogeneous medium maintained under suitable thermodynamic conditions and where an ultrasonic wave generated by a piezoelectric transducer propagates in a direction parallel to the laser beam.

The diffracted waves after being collected by the objective *O* give a diffraction image which can be recorded with a photoelectic system or a photographic method.

The starting point was an exact positioning of the optical system and checking of the linearity of the photoelectrical system, which plays an extremely important role in these cases.

This linearity was confirmed by the agreement of the spectral distributions obtained in liquid CO_2 under thermodynamic conditions far from the critical point with the Raman-Nath distribution [6]. The intensity of the ultrasonic wave close to the critical point was determined using the method described in reference [26].

The recording of the fluctuation of a given diffracted line (of the zeroth or ±1-st order) was made by recording the photoelectrical current. It was essential to place the inlet hole through which a beam of a chosen order fell on the photomultiplier in the correct position. The hole was fixed and thus fluctuations of the diffraction angle caused part of the beam to be extinguished.

A decrease in the light intensity due to fluctuation of the diffraction angle is shown schematically in Fig. 2.

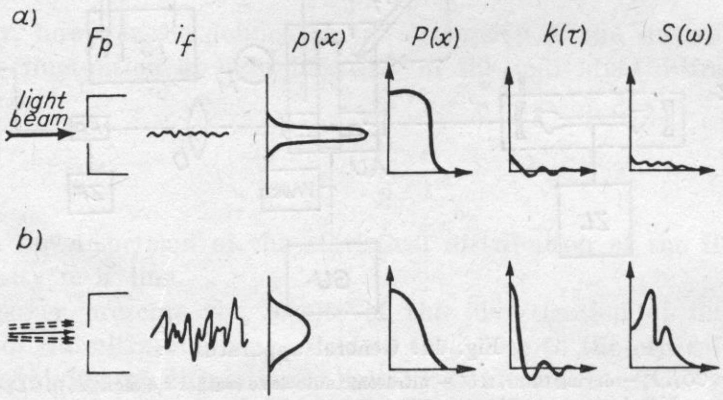

Fig. 2. A schematic representation of the process of recording and analysis of the measurement results

In addition to a decrease due to fluctuation of the diffraction angle, the photomultiplier also recorded fluctuations due to fluctuations of brightness connected with fluctuation of the amplitude of the diffracted light. The process recorded a given diffraction line is a combined phenomenon: of the fluctuation of the diffraction angle (formula (11)) which depends on the ratio, and (thus on the fluctuation of both the amplitude and the phase of the ultrasonic wave), and also on the fluctuation of the light amplitude, which depends also on the fluctuation of the amplitude of the ultrasonic wave, and thus on the quantities $\alpha, \beta,$ and γ defined by the formulae given in reference [15].

Fig. 2. shows schematically successive phases of recording and analysis of this combined process. The apparatus used permits an intensity proportional to the photoelectrical current i_f to be recorded, and a statistical distribution $p(x)$ and the distribution function $P(x)$ to be simultaneously determined automatically. Further processing of the statistical data was performed by calculations based on the fluctuation recording in which the autorrelation function $k(\tau)$ and the power spectrum of the fluctuation process $S(\omega)$, were determined.

The following quantities were used to characterize the fluctuation process: σ (the dispersion of the statistical distribution of the changes of light intensity in a line), τ_0 for $k(\tau) = 0$ (the first zero crossing of the function on the time axis), $S(\omega)_{\max}$ and $\omega_{\max}$ for which $S(\omega) = S(\omega)_{\max}$.

Based on the data obtained from measurements taken near the critical point and far from the critical point, the quantities γ and β which are related to the phase and amplitude of the ultrasonic wave, can be determined. Fluctuations of the diffraction angle can be expressed by the β/γ ratio which is related to σ in the following way:

$$\frac{\beta}{\gamma} = \frac{k}{p} \frac{\sigma}{l}.$$

On the other hand, γ can be determined by calculation of the ratio of most probable light intensities (in the $+1$st orders relative to the zeroth order) near the critical point, and far from the critical point, from the formula [28]:

$$e^{-1/2\gamma} = \frac{p(x)_{\max+1}/p(x)_{\max 0} \quad \text{(near the critical point)}}{p(x)_{\max+1}/p(x)_{\max 0} \quad \text{(far from the critical point)}}. \tag{5}$$

4. The results of investigations in CO_2

The experimental investigations, which were very difficult technically and laborious, permitted a relatively large number of recording to be obtained of the fluctuations of the intensity of light in diffraction figures from an ultrasonic wave under different thermodynamic conditions (far from and near the critical point). It was only partly possible to systematize the experiments, since the thermodynamic states obtained could not be, for purely technical reasons, planned beforehand and subsequently established with precision.

Furthemore for some thermodynamic states, the occurrence of a large absorption of the ultrasonic wave, prevented investigations over longer distances. In order to compare the results of measurements on lines of the same order (0 or $+1$) under different thermodynamic states, a rather severe selection of results was made and only a limited number of them underwent further manipulation and analysis.

The measurement results selected for this paper are shown in Table 1.

In order to explain the method used for the analysis of results, and also to present the differences between the fluctuation processes near and far from the critical point, Figs. 3 and 4 show, as an example, two full sets of results of all the intermediate phases (of. Table 1).

Figs. 3 and 4 present: a) original recordings of the fluctuation processes (for two different paper speeds -10 and 30 mm/s), b) their statistical distributions, $p(x)$, c) their distribution functions $P(x)$, and also d) the autocorrelation function, $k(\tau)$ (continuous line) and e) the density functions of the spectral power fluctuations $S(\omega)$ (discontinuous line), of the processes analyzed. The autocorrelation function has been deliberately given in an unnormalized form, since such a form was used for the calculation of $S(\omega)$.

Table 1 shows the characteristic quantities calculated for five different frequencies of the ultrasonic wave, corresponding to different thermodynamic conditions. The table also includes categories representing the order of the line and the distance from the quartz. In individual groups (of three and four lines) the values are ordered so that the first line corresponds to thermodynamic states close to the critical point; and succeeding lines to states further and further from the critical point. Thus the last line of each group corresponds to thermodynamic states very far from the critical state, for which the fluctuations are already relatively small and it can be assumed that the Raman-Nath distribution is valid for the light diffraction lines. The values in these lines gave a reference for the calculation of γ from formula (5).

All the recorded fluctuations of the light diffracted by an ultrasonic wave at specific frequencies for different thermodynamic conditions were analyzed in the manner mentioned above.

Figs. 5-8 show as an illustration the final calculations of the functions of spectral power of the fluctuation process for four selected cases.

For two of the frequencies of the ultrasonic wave used, (5.76 MHz and 27.8 MHz, Figs. 6 and 8), the behaviour of $S(\omega)$ is characterized by very sharp maxima at low pulsation rates. In particular, close to the critical point (continuous curves) these maxima have large values. It can also be noted that at a frequency of 27.8 MHz, the number of characteristic maxima becomes larger as the thermodynamic conditions diverge further from those of the critical point (discontinuous curves).

For the lines of the 1st order these very characteristic maxima are, considerably shifted (compared with the zeroth order) towards lower pulsation rates.

It can be seen from the diagrams and tables that for all the fluctuations of the ultrasonic wave investigated, the mean value of the power spectrum of the fluctuations becomes higher, as the thermodynamic state approaches the critical point. Also the closer the state comes to the critical point, the greater the shifts from the mean fluctuation power become. These deviations have components lying in the range of low pulsation rates and are thus connected with the occurrence of the "coarse grained" fluctuations.

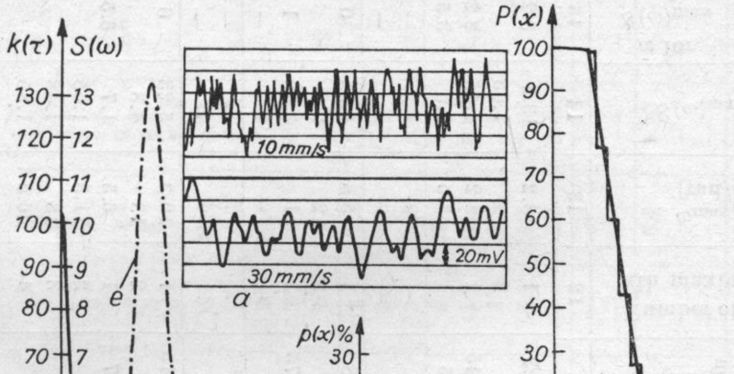

Fig. 3. An example of successive steps in the analysis of the fluctuation of the light diffracted by an ultrasonic wave at a frequency $f = 0.8$ MHz near the critical point ($t = 31.4$°C, $p = 71$ at) for the single line of the zeroth order

a – original fluctuation recordings (for two paper speeds 10 and 30 mm/s), *b* – statistical distribution $p(x)$, *c* – distribution function $P(x)$, *d* – autocorrelation function $k(\tau)$ (continuous line), *e* – fluctuation power spectrum $S(\omega)$ (discontinuous line)

When the thermodynamic conditions are farther from the critical point, the values of the maxima of the fluctuation components in the power spectrum decrease until they vanish completely when no fluctuations of the light diffracted by the ultrasonic wave can be observed.

Table 1

Ultrasonic wave frequency [MHz]	Ultrasonic wave intensity $\left[\frac{\text{Watt}}{\text{cm}^2}\right]$	Thermodynamic conditions		Distance from wave source x [cm]	Spectrum order	Dispersion σ [cm]	$p(x)_{\max}^*$	Autocorrel. function $k(0)$	$\tau \times 33.3$ [ms]	Fluct. power spectrum $S(\omega)_{\max}$	Number of the nth maximum	$\omega_{\max_n}$ [rad/s]	$S(\omega)_{\max_n}$	Pulsation rate		β/γ	γ	β
		t [°C]	p [at]											ω for $S(\omega)_{\max}$ [rad/s]	$\omega_{\max}$ [rad/s]			
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
0.8	0.1	31.4	71	1.3	0	2.4	110	101.1	5	13.3	1	8.2	13.3	8.2	36	0.14	4.01	0.55
											2		1.65					
	0.1	34.5	75	1.3	0	1.8	125	43.8	12	4.45	1	2.2	4.45	2.2	32	0.10	—	—
	0.1	27.4	62	1.3	0	1.1	120	15.1	12	1.5	1	3.5	1.5	3.5	30	0.06	—	—
	0.1	31.6	71	1.3	+1	1.1	70	—	—	—	—	—	—	—	—	0.05	—	—
	0.1	27.6	63	1.3	+1	1.0	55	—	—	—	—	—	—	—	—	—	—	—
	0.1	31.2	72.5	6.5	0	2.7	95	82	37.5	10.8	1	5.8	6.95	0	32	0.15	3.52	0.54
											2	12	2.3					
	0.1	35.1	76	6.5	0	1.3	95	22.1	32.5	3.75	1	1	3.75	1	30	0.07	—	—
	0.1	26.3	61.5	6.5	0	0.8	95	—	—	—	—	—	—	—	—	0.04	—	—
	0.1	32	73.5	6.5	+1	2.8	65	—	—	—	—	—	—	—	—	—	—	—
	0.1	27	62.5	6.5	+1	0.8	75	—	—	—	—	—	—	—	—	—	—	—
	0.1	32.1	73	1.3	0	2.9	75	100	5.3	10.15	1	10.3	5.25	0	40	0.16	6.25	1.03
											2	18	4.8					
	0.1	30	68	1.3	0	1.65	85	41.5	3	1.7	1	6.5	1.7	6.5	45	0.09	—	—
											2	11.5	1.65					
											3	18	1.48					
											4	26.5	1.35					
	0.1	28.6	60.5	1.3	0	0.82	95	—	—	—	—	—	—	—	—	0.05	—	—
	0.1	26.4	59.5	1.3	0	0.8	95	—	—	—	—	—	—	—	—	0.04	—	—
	0.1	31.4	71.5	6.5	0	2.4	75	91	5	6.05	1	6.2	6.05	6.2	43	0.15	12	1.85
											2	11.6	5.06					
											3	15.8	3.45					

											4	19.3	2.75					
1.2	0.1	29.6	67	6.5	0	1.8	80	31.2	8	3.20	1	4.8	3.2	4.8	26	0.09	—	—
											2	9.7	2.65					
	0.1	28	60	6.5	0	1.2	85	27.2	32.5	2.5	—	—	—	0	40	0.09	—	—
	0.1	31.2	71.5	6.5	+1	0.9	55	—	—	—	—	—	—	—	—	—	—	—
	0.1	28.2	60	6.5	+1	0.8	65	—	—	—	—	—	—	—	—	—	—	—
	0.1	32.2	73.5	1.3	+1	2.7	55	185.5	6	17.8	1	6.8	13.2	0	44	—	—	—
											2	13	13.5					
											3	16	5.1					
	0.1	30.3	69	1.3	+1	1.62	45	85.2	15.5	9	1	1.8	9	1.8	38	—	—	—
											2	5.3	6.7					
											3	14	3.35					
											4	21.7	0.78					
	0.1	29.4	63	1.3	+1	1.55	55	38	4.5	2.25	1	6.8	2.25	6.8	34	—	—	—
											2	14.6	1.95					
											3	23	0.75					
	0.1	27.1	60.5	1.3	+1	1.1	75	31	3.5	1.6	1	12	1.6	12	40	—	—	—
3.51	0.1	31.5	73	1.3	0	3.3	95	136.7	15.5	13.15	1	2.2	13.15	2.2	40	0.19	2.6	0.49
											2	5.2	11.1					
											3	14.8	6.25					
											4	27.5	0.8					
	0.1	33.4	74.5	1.3	0	1.9	115	55	30	6.32	1	1.4	6.32	1.4	36	0.11	—	—
											2	8.5	3.15					
											3	18.6	1.8					
	0.1	35.8	80	1.3	0	1.2	120	20	13	2.4	1	2.6	2.4	2.6	32	0.07	—	—
	0.1	31.2	73	1.3	+1	2.65	55	—	—	—	—	—	—	—	—	—	—	—
	0.1	36	79	1.3	+1	0.8	85	—	—	—	—	—	—	—	—	—	—	—
	0.1	32.8	75.5	6.5	0	3.1	90	89.2	40	20.6	1	6.2	5.73	0	34	0.18	8.1	1.43
	0.1	34.1	76	6.5	0	2.8	95	39.7	48	8.8	—	—	—	0	30	0.16	—	—
	0.1	35.6	79.5	6.5	0	1.9	100	37.3	63	7.6	—	—	—	0	34	0.11	—	—
	0.1	34.1	70.5	1.3	0	3.2	85	117.3	5.5	10.86	1	5.6	8.8	10.8	38	0.18	3.57	0.65
											2	10.8	10.86					
											3	16	2.5					
	0.1	30.1	67	1.3	0	2.0	85	59.3	8	4.6	1	2.6	4.2	7.2	38	0.10	—	—
											2	7.2	4.6					

Table 1 ctd.

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
											3	19.6	1.35					
5.76	0.1	27.2	61	1.3	0	1.05	85	9.7	66.5	1.97	1	0	1.97	0	20	0.04	—	—
	0.1	34.2	70	1.3	+1	2.65	65	139.5	6.5	17.1	1	5.6	17.1	5.6	36	—	—	—
											2	10.6	11.03					
	0.1	29.4	66.5	1.3	+1	1.9	55	48.8	4.5	3.83	1	9.4	2.30	14.2	48	—	—	—
											2	14.2	3.83					
											3	23.6	1.05					
	0.1	27.4	61.5	1.3	+1	1.1	75	17.3	29.5	2	1	—	—	0	36	—	—	—
	0.1	32.6	73.5	2	0	3.6	85	152.2	5.5	16.5	1	11.2	15.5	11.2	40	0.20	6.25	1.28
	0.1	30.1	69	2	0	2.9	95	63.8	5	4.05	1	3	4.05	3	40	0.16	—	—
											2	11.4	4					
											3	17.6	2.55					
	0.1	27.5	62	2	0	1.3	85	26	18	2.35	1	4	2.35	4	36	0.07	—	—
27.8											2	12	1.63					
											3	18	0.63					
	0.1	32.5	74.5	2	+1	3.1	65	120	6.5	—	—	—	—	—	—	—	—	—
	0.1	30.6	69.5	2	+1	2.8	70	72.2	5	7.7	1	3.2	3.73	9.6	40	—	—	—
											2	9.6	7.7					
											3	15.2	3.45					
	0.1	36.9	63	2	+1	0.9	70	23.2	75	4.8	—	—	—	0	32	—	—	—

* $p(x)_{\max}$ denotes the most probable value of the photoelectric current (i.e. light intensity).

The distribution of the light intensity in the diffraction spectrum then agrees with the Raman-Nath theory. The characteristic maxima in the fluctuation power spectrum for a line of the zeroth order at a distance of 6.5 cm are shifted towards lower values of pulsation rate, compared to those at a distance of 1.3 cm. A similar shift occurs at the same distance for a line of the 1st order, compared with a line of the zeroth order. The occurrence of specific maxima in the fluctuation power spectrum for a line of the zeroth order under critical conditions shows that the behaviour of the fluctuations is "quasiperiodic". In the case when several characteristic maxima occur in the fluctuation power spectrum, it can be observed that they occur at nearly equal intervals on the scale of pulsation rate ω. Thus the spectrum is "quasiharmonic".

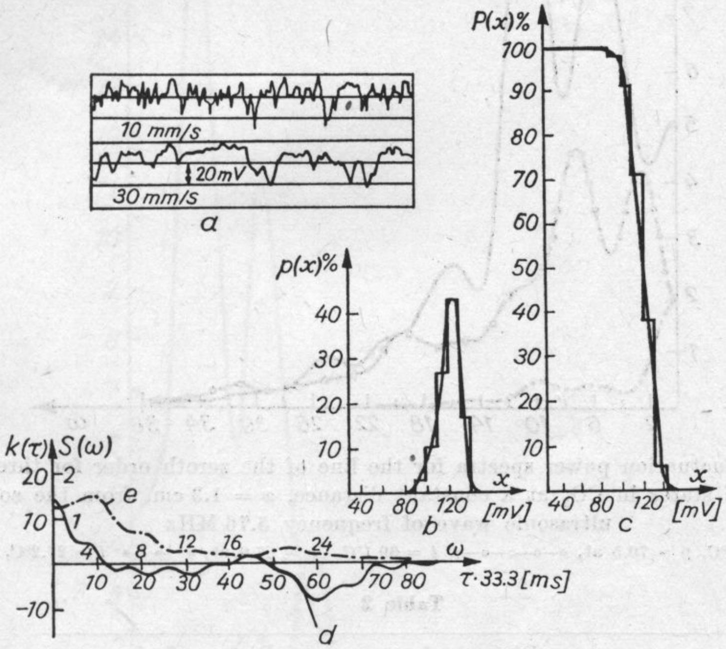

Fig. 4. An example of successive steps in the analysis of the fluctuations of the light diffracted by an ultrasonic wave at a frequency of $f = 0.8$ MHz relatively far from the critical point ($t = 27.4°C$, $p = 62$ at), for the single line of the zeroth order

a – original fluctuation recordings (for two paper speeds – 10 and 30 mm/s), b – statistical distribution $p(x)$, c – distribution function $P(x)$, d – autocorrelation function $k(\tau)$ (continuous line), e – fluctuation power spectrum $S(\omega)$ (discontinuous line)

The β/γ ratio and subsequently γ itself, were calculated, on the basis of previously determined values of the dispersion (δ) of the statistical distribution of the fluctuations of the intensity of the light diffracted by the ultrasonic wave under different thermodynamic conditions. From these results β could be determined. Values corresponding to these quantities are shown in Table 1.

For all frequencies of the ultrasonic wave used, the value of β/γ decreases as the deviation of the thermodynamic conditions from the critical point decreases; thus showing that fluctuations in quantities of the wave field become increasingly smaller. It can be also observed that the β/γ ratio rises sligtly close to the critical point as the frequency of the ultrasonic wave increases. This

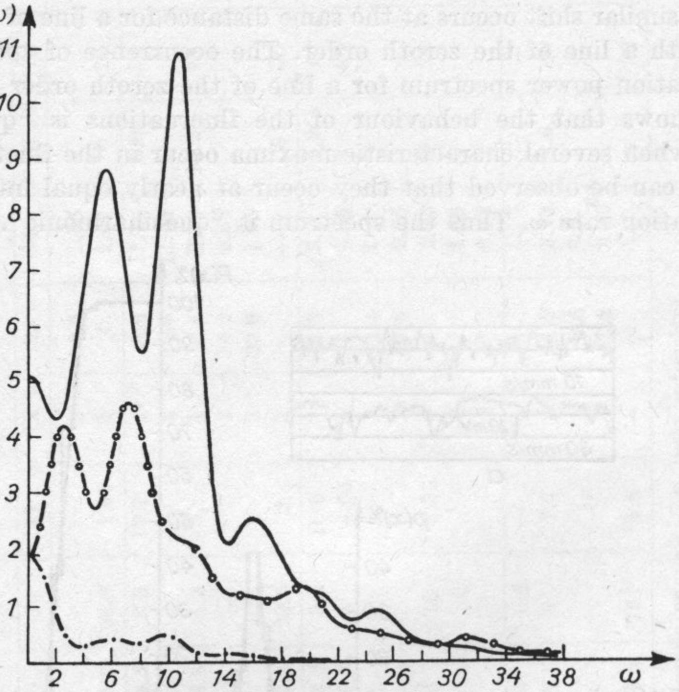

Fig. 5. Light fluctuation power spectra for the line of the zeroth order for three different thermodynamic states in CO_2 at a constant distance, $x = 1.3$ cm, from the source of an ultrasonic wave of frequency 5.76 MHz

——— $t = 34.1$ 1°C, $p = 70.5$ at, o—o—o—o—o $t = 30.1$°C, $p = 67.0$ at, •—•—• $t = 27.2$°C, $p = 61.0$ at

Table 2

Frequency f[MHz]	Distance from source of ultrasonic wave 1.3 [cm]		Distance from source of ultrasonic wave 6.5 [cm]	
	values of γ	values of β	values of γ	values of β
0.8	4.01	0.55	3.52	0.54
3.51	2.6	0.49	8.1	1.43
5.76	3.57	0.65	—	—
27.8	6.25	1.28	—	—

results from the fact that a stronger interaction between the ultrasonic wave and the fluctuations of the medium occurs close to the critical point.

The values of γ and β for five frequencies and two distances from the source of the ultrasonic wave are shown in Table 2.

It follows from the statistical distributions of the light intensity fluctuations of the diffraction spectrum recorded (at the shortest distance from the quartz), that for a line of the zeroth order these distributions are generally symmetrical (differing slightly from normal distributions), while for lines of the 1st order they are very asymmetrical and irregular. As an illustration, Fig. 9 shows two distributions recorded at an ultrasonic frequency of 5.76 MHz close to the critical point, for the two cases. At longer distances from the wave source, deviations from the normal distribution are considerably greater, which means that deformation of the acoustic wave increases with distance for a given intensity.

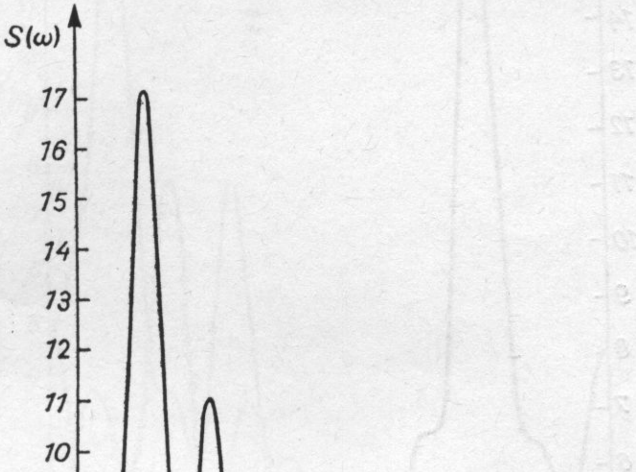

Fig. 6. Light fluctuation power spectra for a line of the first order for three different thermodynamic states in CO_2 at a constant distance, x = 1.3 cm, from the source of an ultrasonic wave of frequency 5.76 MHz

——— t = 34.2°C, p = 70.0 at, ○—○—○—t = 29 4°C, p = 66.5 at, ●—●—● t = 27.4°C, p = 61.5 at

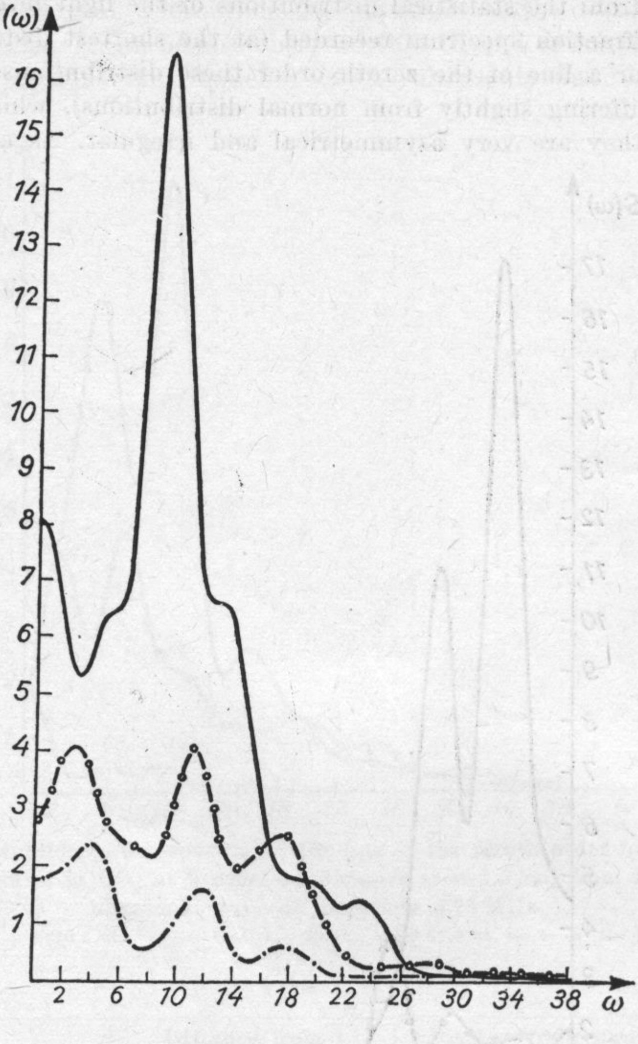

Fig. 7. Light fluctuation power spectra for the line of the zeroth order for three different thermodynamic states in CO_2 at a constant distance, $x = 2$ cm, from the source of an ultrasonic wave of frequency 27.18 MHz

——— $t = 32.6$°C, $p = 73.5$ at, ○—○—○ $t = 30.1$°C, $p = 69.0$ at, ●—●—● $t = 27.5$°C, $p = 62.0$ at

5. Comparison of results for liquid-liquid and liquid-vapour systems

In both a liquid-liquid system [21] and a liquid-vapour system [20] the β/γ ratio takes increasingly higher values as the critical point of the medium is approached, which shows that deformation of a propagating ultrasonic wave increases as the critical point is approached.

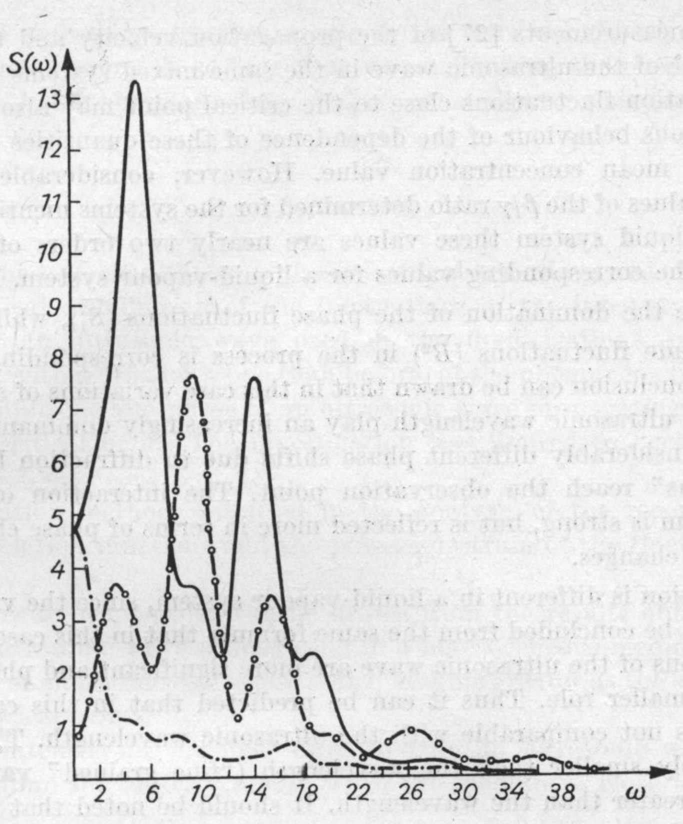

Fig. 8. Light fluctuation power spectra for a line of the first order for three different thermodynamic states in CO_2 at a constant distance, $x = 2$ cm, from the source of an ultrasonic wave of frequency 27.8 MHz

—— $t = 32.5$°C, $p = 74.5$ at, ○—○—○ $t = 30.6$°C, $p = 69.5$ at, —●—●— $t = 26.9$°C, $p = 63.0$ at

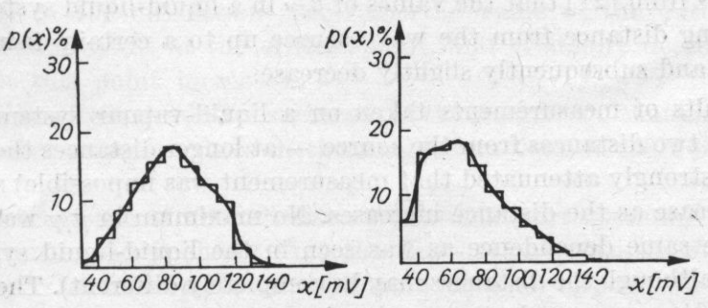

Fig. 9. Statistical distributions of the light fluctuation in a diffraction spectrum near the critical point ($t = 34.1$°C, $p = 70.5$ at)

a — for the line of the zeroth order, b — for a line of the first order

Parallel measurements [27] of the propagation velocity and the attenuation coefficient of the ultrasonic wave in the same mixed systems [21] showed that concentration fluctuations close to the critical point may also be a source of the anomalous behaviour of the dependence of these quantities on temperature and the mean concentration value. However, considerable differences occur in the values of the β/γ ratio determined for the systems mentioned above. For a liquid-liquid system these values are nearly two orders of magnitude greater than the corresponding values for a liquid-vapour system. High values of β/γ indicate the domination of the phase fluctuations ($\overline{S_1^2}$), while the share of the amplitude fluctuations ($\overline{B^2}$) in the process is correspondingly smaller. The indirect conclusion can be drawn that in this case variations of sizes comparable with the ultrasonic wavelength play an increasingly dominant role. Thus waves with considerably different phase shifts due to diffraction by the fluctuation "grains" reach the observation point. The interaction of the wave and the medium is strong, but is reflected more in terms of phase changes than in amplitude changes.

The situation is different in a liquid-vapour system, since the values of β/γ are low. It can be concluded from the same formula that in this case the amplitude fluctuations of the ultrasonic wave are more significant and phase fluctuations play a smaller role. Thus it can be predicted that in this case the size of variations is not comparable with the ultrasonic wavelength. They are either considerably smaller than the wavelength ("fine grained" variations) or considerably greater than the wavelength. It should be noted that the sizes of the variations change considerably close to the critical point, and in each thermodynamic state there is a very wide statistical distribution of sizes, in which only the most probable sizes can be distinquished. The ratio β/γ which is determined is the effect of a combined interaction of the ultrasonic wave and variations of different sozes and the remarks above refer only to interactions with the variations of the dominant size in a given distribution.

It follows from [21] that the values of β/γ in a liquid-liquid system increase with increasing distance from the wave source up to a certain distance, reach a maximum, and subsequently slightly decrease.

The results of measurements taken on a liquid-vapour system (although taken at only two distances from the source — at longer distances the ultrasonic wave was so strongly attenuated that measurement was impossible) show a tendency to increase as the distance increases. No maximum in β/γ was observed. However, the same dependence as was seen in the liquid-liquid system could be expected (although the distances may be completely different). The maximum in the ratio β/γ corresponds to a region in which phase fluctuations become comparable with amplitude fluctuations, $\overline{S_1^2} = \overline{B^2}$ (assuming that the statistical distributions of the fluctuations of these quantities are normal [1]). The region is related with the transition from the so called Fresnel region (near field) to the Fraunhofer region (far. field). Phase fluctuations play an essential role in the near field and a less significant role in the far field. In the far field the distance from the source is so long that waves reaching the observation point have only slight shifts in phase. Amplitude fluctuations are then more dominant.

5. Summary

The method of simultaneous photoelectrical recording and determination of the statistical distribution of the fluctuations of the intensity of the light diffracted by an ultrasonic wave used in the investigations, permitted the investigation, with a broader scope than before, of the phenomena of the fluctuation of the amplitude and phase of an ultrasonic wave propagating in a medium close to the critical point. The investigation was limited to "coarse grained" fluctuations.

A correlation method was used in the analysis of the records obtained. The autocorrelation functions and the power spectrum of the fluctuations were determined.

The following conclusions can be drawn from the results obtained:

(1) An ultrasonic wave propagating in a heterogeneous medium undergoes deformations of amplitude and phase which become greater as the system approaches the critical point.

(2) The intensity of the light diffracted by an ultrasonic wave propagating in a medium fluctuates strongly when the medium is close to the critical point.

(3) Statistical distributions of the fluctuations of the light intensity in the zeroth order are approximately symmetrical (being only slightly different from normal distributions).

(4) Statistical distributions of the fluctuations of the light intensity in diffraction lines of the first order are asymmetrical and irregular.

(5) The dispersion of the statistical distributions of the fluctuations of the light intensity in the diffraction spectrum (the value of the autocorrelation function $\tau(o)$) is greatest at the critical point, and gradually decreases as the distance from this point increases.

(6) It follows from the power spectrum of the fluctuation that characteristic components with pulsation rates in the range from 0 to 35 rad/s occur in the process.

(7) The occurrence of characteristic pulsations (corresponding to maxima in the spectrum of $S(\omega)$) shows that the fluctuation processes observed are "quasiperiodic". In some cases the spectrum of $S(\omega)$ appears to be "quasiharmonic" in character.

(8) The mean values of the power spectrum ($S(0)$) reach their highest values for thermodynamic conditions close to the critical point.

(9) Deviations from the mean value of the fluctuation power spectrum at a long distance from the source of the ultrasonic wave (for a line of the zeroth

order) are much smaller than those at a shorter distance and are in a lower range of pulsation rates (up to about 15 rad/s).

(10) A shift of the characteristic maxima towards lower pulsation rates can be observed in the power spectra of the fluctuations of the light intensity in a line of the 1st order.

(11) The methods of determinating the ratio β/γ, and γ itself, given in this paper, permit the value of β to be calculated. This represents an improvement on the investigations of Śliwiński who gave only the method for determinating the ratio β/γ.

(12) The apparatus used to record the fluctuations permitted only "coarse grained" fluctuations to be investigated. The use of the correlation method in the analysis of the results obtained illustrates its value. The correlation method could also be successfully used for the analysis of processes connected with "fine grained" fluctuations. This would require recording devices with very small time constants to be used. This is a separate problem which could be investigated by an analogous method when suitable apparatus is available.

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