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DIFFRACTION OF A HIGH INTENSITY LIGHT BY ULTRASOUND IN NONLINEAR LIQUIDS*

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The paper presents some experimental and theoretical results obtained in the examination of high power light diffraction (Nd^{3+} laser) by ultrasound (4 MHz) in nonlinear liquids. The diffraction patterns differ from the usual results for the light of small intensity. The distributions of the light can be explained by the influence of the optical nonlinearity of the medium (3-rd harmonic light generation and frequency mixing). The theoretical description proposed appears to explain the experimental results.

1. Introduction

Since coherent sources of light of high intensity became available, many experimental and theoretical papers have appeared on the nonlinear effects accompanying light propagation in material media. One of the phenomena, the diffreaction of high intensity light by ultrasound was first examined in 1971 [8] and has been further investigated [9, 11, 12, 13]. This paper gives a review of the topic, including some new results and theoretical explanations. We start with a description of some elements of nonlinear optics required for further acousto-optic considerations.

The intensities of classical sources of light are small (0.1-100 W/cm^2), but in laser sources intensities from some mW/cm^2 to some decades of GW/cm^2 [14] are available experimentally, depending on the kind of the laser device used. The electric field intensities of the optical vector corresponding to such

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light energy densities are 10^5 V/cm to 10^8 V/cm (by comparison, the electric field intensity of sun light is barely 10 V/cm) [10]. Such high electric field strengths are comparable to the electric fields existing in the interior of matter (e.g. the electric field from the nucleus on an electron in a hydrogen atom is of the order of 10^9 V/cm, internal fields in crystals are of the order of 10^8 V/cm, in liquids 10^5-10^7 V/cm).

Thus, one can see that in the strong electric fields created with laser beams some properties of atoms and molecules and, consequently, of the whole material media can be changed during the propagation of light through them. Under such conditions the electric polarization of a medium is no longer proportional to $\boldsymbol{E}$, i.e. the relation

$$\boldsymbol{P}_L(\boldsymbol{r}, t) = \chi_1^e \boldsymbol{E}(\boldsymbol{r}, t) \tag{1}$$

is not adequate for a description of physical phenomena. An additional nonlinear polarization $\boldsymbol{P}_{NL}$ occurs and

$$\boldsymbol{P}_{NL}(\boldsymbol{r}, t) = \chi_2^e : \boldsymbol{E}^2(\boldsymbol{r}, t) + \chi_3^e : \boldsymbol{E}^3(\boldsymbol{r}, t) + \ldots, \tag{2}$$

where χ_1^e, χ_2^e and χ_3^e are tensors of the electric susceptibility of the first, second and third rank, respectively.

In linear optics the refractive index of a medium for light has a constant value, and in the range of linear phenomena the principle of superposition is valid, i.e. no interaction among various light beams takes place. The electric fields of very intense light cause changes in the refractive index of the medium and nonlinear optical effects occur. In this case the principle of superposition is not valid: the electric fields of different light beams interact with one another, and they also interact with the material medium in which they are propagating (the nonlinear polarization mentioned above).

A special effect in nonlinear phenomena consists in the generation of higher harmonics of $2\omega, 3\omega, \ldots$, where ω is the frequency of the fundamental component. These harmonics correspond to the consecutive orders of nonlinearity of the electric polarization of the medium in formula (2).

It may be shown [10] that the tensor of the electric susceptibility of the second rank, χ_2^e, has components different from zero only in the case of crystals belonging to the crystallographic classes which have no centre of symmetry. For crystals possessing a centre of symmetry and for isotropic bodies the second order nonlinear phenomena do not occur, but at very high light intensities the cubic nonlinearity plays a dominant role. For the last case one can write the dependence of polarization on the electric field as follows:

$$\boldsymbol{P} = \chi_1^e \cdot \boldsymbol{E}(\boldsymbol{r}, t) + \chi_3^e : \boldsymbol{E}^3(\boldsymbol{r}, t) + \ldots \tag{3}$$

For isotropic media this gives

$$P = \alpha E + \beta E^3 + \ldots, \tag{4}$$

where α and β denote coefficients corresponding to χ_1^e and χ_2^e for isotropic media.

Thus, in a medium with 3-rd order nonlinearity the 3-rd harmonic of light can be generated during propagation. This results from the following consideration.

If the original beam has the form

$$\boldsymbol{E} = \boldsymbol{E}_0 \cos(\boldsymbol{kr} - \omega t), \tag{5}$$

where $\boldsymbol{E}_0$ is the amplitude of light, $\boldsymbol{k}$ — wave vector, $\boldsymbol{r}$ — vector of coordinates, ω — fundamental frequency, t — time, then, due to the nonlinear interaction, the 3-rd order polarization will occur:

$$\boldsymbol{P}^{(3)} = \beta \boldsymbol{E}_0^3 \cos^3(\boldsymbol{kr} - \omega t). \tag{6}$$

Formula (6) may be written as the sum of two terms (in view of the relation $\cos^3 x = \frac{1}{4}(3\cos x + \cos 3x)$):

$$\boldsymbol{P}^{(3)} = \tfrac{3}{4}\beta \boldsymbol{E}_0^3 \cos(\boldsymbol{kr} - \omega t) + \tfrac{1}{4}\beta \boldsymbol{E}_0^3 \cos(3\boldsymbol{kr} - 3\omega t). \tag{7}$$

Using this formula one can estimate the change in refractive index introduced by the nonlinear polarization.

Generally, for the electric induction D in the medium, we have

$$D = D_0 + P, \tag{8}$$

where $D_0 = \varepsilon_0 \boldsymbol{E}$ is the electric induction in a vacuum, ε_0 — permeability of a vacuum. From the definition one can write

$$\varepsilon_E \varepsilon_0 = \frac{dD}{dE}, \tag{9}$$

where ε_E is the electric permeability which is a function of the electric field E. From (9), using (8) and (7), we get

$$n_E = n + \gamma E^2 \quad \text{or} \quad \Delta n = n_E - n = \gamma E^2, \tag{10}$$

where $\gamma = 3\beta/2\varepsilon_0 n$, $n = \sqrt{\varepsilon}$, $n_E = \sqrt{\varepsilon_E}$.

Thus the variation of refractive index due to the 3-rd harmonic is proportional to the squared amplitude of the fundamental. γ is the characteristic constant responsible for the 3-rd order nonlinear polarization of the medium.

In the literature [9] the electrostriction of the medium is considered as the main reason for the nonlinear polarization and the coefficient γ is expressed by the formula

$$\gamma = \frac{\varrho(\partial\varepsilon/\partial\varrho)_s \beta_s}{16\pi n_0}, \tag{11}$$

where ϱ is the density, β_s — the adiabatic compressibility, ε — the electric permeability, n — the refractive index of the undisturbed medium.

The quantities β_s and $\varrho(\partial\varepsilon/\partial\varrho)_s$ are known for many substances and it was possible to calculate Δn from formulae (10) and (11) for a chosen value of E. For a laser beam of power density 10^{12} W/cm^2, corresponding to $E = 2.7 \cdot 10^7$ V/m, γ and Δn were calculated for a few liquids. Results are presented in paper [9] (see Table 1).

Table 1

Substance	n_0	$\varrho \cdot 10^3$ [kg/m^3]	$\varrho(\partial\varepsilon/\partial\varrho)_s$	$\beta_s \cdot 10^{13}$ [N/m^2]	$\gamma \cdot 10^{20}$ [m^2/V^2]	$\Delta n \cdot 10^6$
water	1.3397[c]	0.997[c]	0.82[c]	45.7[c]	0.06	0.44 (0.38)[a]
carbon disulfide	1.674[a]	1.262[a]	2.39[a]	49.5[a]	0.42	3.19 (2.91)[a]
benzene	1.522[a]	0.879[a]	1.56[a]	52.6[a]	0.21	1.61 (0.04)[a]
carbon tetrachloride	1.4720[c]	1.595[c]	1.15[a]	58.0[c]	0.14	0.99
toluene	1.5130[c]	0.865[c]	1.60[c]	70.0[c]	0.30	2.24
ethyl alcohol	1.369[c]	0.789[c]	0.95[c]	92.8[c]	0.15	1.14
methyl alcohol	1.338[c]	0.791[c]	0.82[c]	100.5[c]	0.14	0.95
nitrobenzene	1.553[b]	1.203[a]	40.0[a]	34.8[a]	2.21	670.0

a – [22], *b* – [23], *c* – [24]

2. Conditions for the third harmonic generation

The conditions required for the 3-rd harmonic generation [1] are the following:

a. The isotropic medium must be nonactive optically.

b. The electronic absorption of light must be absent or very small.

c. Performance of phase matching conditions must be present, i.e. the value of the phase velocity for the fundamental and the 3-rd harmonic of light in the medium must be the same, i.e. $\Delta k \approx 0$, in the relation

$$\Delta k = k_{3\omega} - 3k_{\omega} = \frac{3\omega}{c}(n_{3\omega} - n_{\omega}), \tag{12}$$

where $n_{3\omega}$, n_{ω} and $k_{3\omega}$, k_{ω} are the refractive indices and the wave numbers for the 3-rd harmonic and the fundamental one, respectively.

d. There must be an interaction along the coherence length:

$$l_s = \frac{\pi}{\Delta k}.$$

The formula for the intensity of the 3-rd harmonic of light is as follows [1]:

$$I(3\omega, l) = (\chi^{3\omega}_{xxyy})^2 \left\{ \frac{r \sin \frac{1}{2} l \Delta k}{\frac{1}{2}\Delta k} \right\}^2 I^3. \tag{14}$$

The first experimental steps in obtaining the 3-rd harmonic generation were performed by TERHUME et al. [15] in 1962. They used a ruby laser and a crystal of calcite. In 1969 GOLBERG and SCHUUR [7] examined the 3-rd harmonic generation in liquids, particularly in liquid crystals. The change in the refractive index and other interesting nonlinear effects were described by BUCKINGHAM [4] and BLOMBERGEN [3].

Condition b may be achieved by introducing an abnormal dispersion for the frequency range between the fundamental and harmonic [6] as was first experimentally realized by BEY et al. [2] (see Fig. 3).

The first calculations on the diffraction of intense light generating the 3-rd harmonic by ultrasound were performed in 1969-72 by JÓZEFOWSKA et al. [8, 9] (see Figs. 1 and 2). The amplitude distributions are described by the formulae

$$E(\omega, l, x, t) = \exp(2\pi i\nu t) \sum_{k=-\infty}^{+\infty} \Phi_k^0(a) \exp\left(-i\frac{n_0}{\delta n}a\right) \exp\left(2\pi i k \frac{x}{\Lambda}\right) \times$$

$$\times \exp(-2\pi i N t) + \exp[3(2\pi i\nu t)] \sum_{k'=-\infty}^{+\infty} [\Phi_{k'}^1(3a) + A_{k'}(a)] \times \tag{15}$$

$$\times \exp\left[-i\frac{n_0}{\delta n} \cdot 3a\right] \exp\left(2\pi i k' \frac{x}{\Lambda}\right) \exp(-2\pi i N t) \quad \text{for } k' = 3k,$$

where ν and λ are the frequency and wavelength of light, respectively, N and Λ are the frequency and wavelength of the ultrasound, n_0 is the optical refractive index for the undisturbed medium, δn is the amplitude of variation in the index due to the ultrasound and is proportional to the acoustic pressure of the wave, t is the time, $\Phi_k^0(a)$ are Bessel functions of argument $a = 2\pi\delta n l/\lambda$, $\Phi_{k'}^1(3a)$ are Bessel functions of argument $3a$, and $A_{k'}$ are complicated linear combinations of the products of Bessel functions of different orders [8].

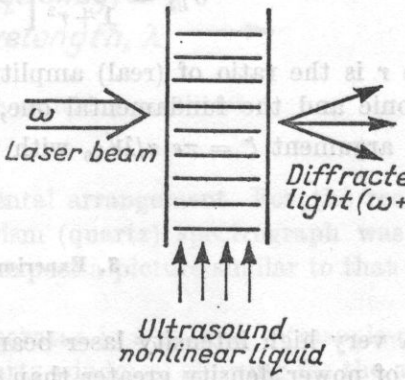


Fig. 1. Geometrical situation of the diffraction of light by ultrasound generating the 3-rd harmonic in a nonlinear liquid medium

The light intensity distributions are given by

$$J_0 = (1-r)^2 \Phi_0 \Phi^* + E^2 r^2 [\Phi_0(3a) + A_0(a)][\Phi_0(3a) + A_0(a)]^* \tag{16}$$

(* denotes the complex conjugate) and for the 3-rd harmonic for the orders $k' = \pm 1, \pm 2, \ldots$ we have

$$J_{\pm 1} = E^2 r^2 [\Phi_{\pm 1}(3a) + A_{\pm 1}(a)][\Phi_{\pm 1}(3a) + A_{\pm 1}(a)]^*. \tag{17}$$

Some experimental work on this problem was performed by KOSMOL [11].

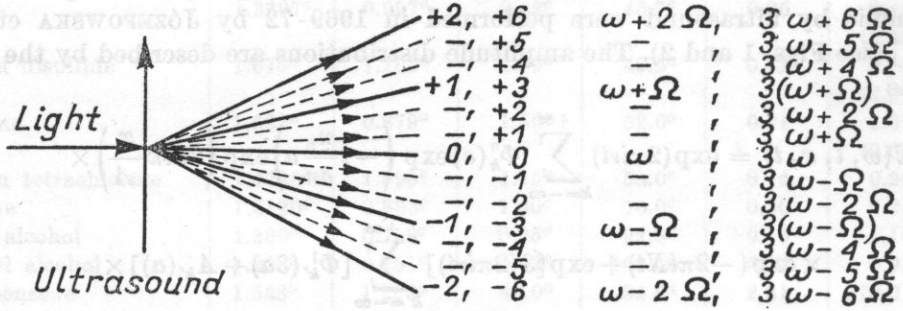


Fig. 2. Spectrum of diffraction pattern in the phenomenon of the diffraction of light by ultrasound in a nonlinear liquid medium

Lastly, MERTENS and LEROY [13] recalculated the results of Józefowska et al. and obtained an exact solution using a generating function method which gave simple expressions for the intensities of the fringes in the diffraction pattern,

$$J_j = \frac{1}{1+r^2}\left\{J_j^2(\zeta) + \left[r^2 + \frac{9\beta^2\zeta^2}{\varepsilon^2(1+r^2)^2}\right] J_{3j}^2(3\zeta)\right\}, \tag{18}$$

$$J_{l/3} = \frac{1}{1+r^2}\left[r^2 + \frac{9\beta^2\zeta^2}{\varepsilon_1^2(1+r^2)^2}\right] J_l^2(3\zeta), \tag{19}$$

where r is the ratio of (real) amplitudes of the light for the generated third harmonic and the fundamental one, and $J_j(\zeta)$ is a Bessel function of order j and argument $\zeta = \pi\varepsilon_1 z/\lambda\sqrt{\varepsilon_0}$ with the condition $l \neq 3j$.

3. Experimental conditions

A very high intensity laser beam is required in the experiments. A laser beam of power density greater than 10^{12} W/m^2 was used, corresponding to an electric field amplitude of the light wave in the beam of $2.7 \cdot 10^7$ V/m. As our first investigations with a ruby laser $\lambda = 6943$ Å, and the 3-rd harmonic at

about 2300 Å, were not satisfactory, we used an Nd-glass laser with $\lambda = 10600$ Å (and the 3-rd harmonic about 35300 Å) which proved more convenient for the experiment. Using a switching cell with an Eastman Kodak Q-switch 9860, we obtained gigantic laser pulses of tens of nanoseconds duration.

Material suitable for the 3-rd harmonic generation must be relatively transparent both for the fundamental and for the 3-rd harmonic. The generation takes place along the coherence length of the light beam l_s, given by the formula (derived from [12] and [13])

$$l_s = \frac{\lambda_\omega}{6(n_{3\omega} - n_\omega)}, \tag{20}$$

where λ_ω is the wavelength for the fundamental, and $n_{3\omega}$ and n_ω are the refractive indices of the fundamental and the 3-rd harmonic, respectively.

Conditions for matching after FRANKEN et al. [6] are presented in Fig. 3. An ultrasonic wave of 4 MHz was used in the experiments.

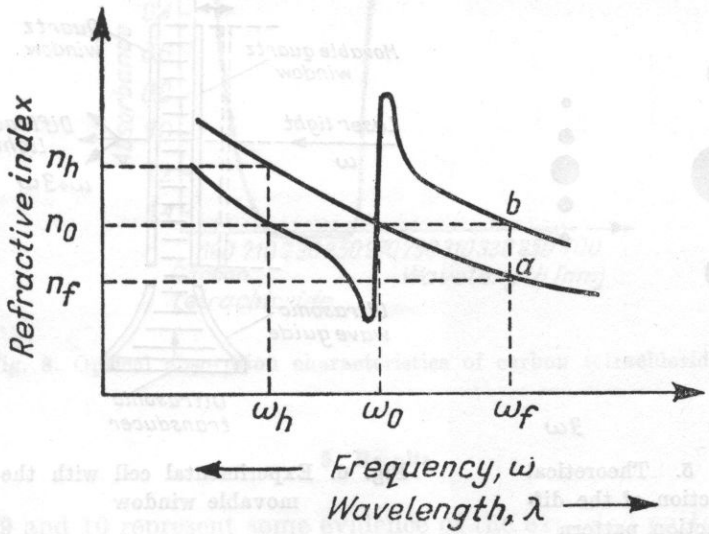


Fig. 3. Description of phase matching conditions

Fig. 4 presents a schematic experimental arrangement. For the recording of the diffraction spectra a simplified prism (quartz) spectrograph was used. From the theoretical predictions one can expect a picture similar to that shown in Fig. 5.

The most important part of the apparatus is a special ultrasonic-optical cell which has parallel movable walls (quartz windows) allowing for the adjustment of the interaction distance for the proper matching of refractive indices

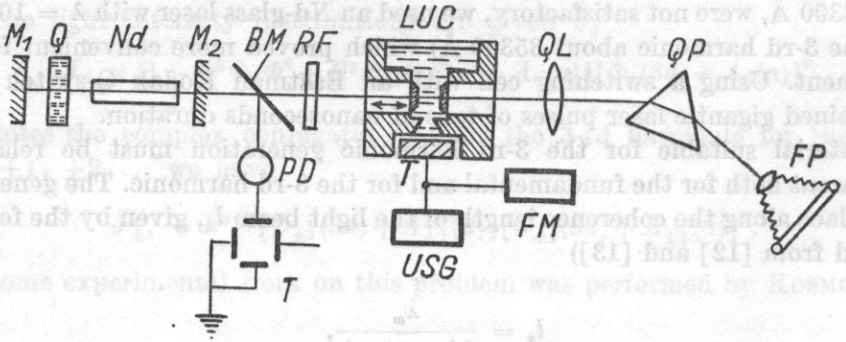


Fig. 4. Experimental arrangement

M_1 – mirror $R = 100\,\%$ M_2 – mirror $R = 50\,\%$, Q – Q-switcher, Nd – Nd^{3+} glass laser, BM – beam-splitter, PD – photodiode, RF – red filter LUC – light ultrasonic cell with liquid, T – piezoelectric transducer, USG – ultrasonic supply generator, FM – frequency meter, QL – quartz lens, QP – quartz prism spectrograph, FP – photographic plate

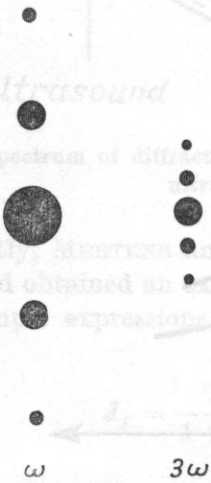


Fig. 5. Theoretical prediction of the diffraction pattern

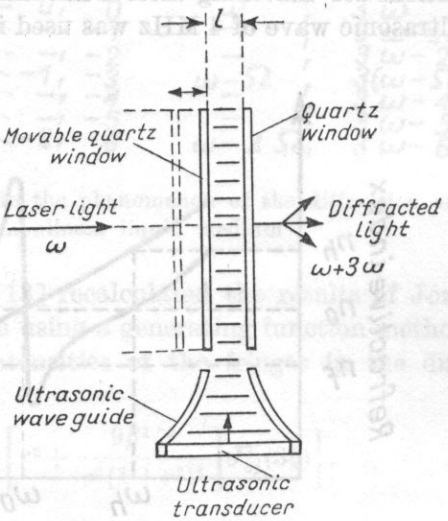


Fig. 6. Experimental cell with the movable window

and for determination of the coherence length. Fig. 6 presents the structure of the cell. The thickness of the cell can be regulated from 0.05 to 6.27 mm with a precision of 0.001 mm.

4. Choice of liquids

Experiments were performed in carbon tetrachloride for comparison wih the calculations in [9] and in a 130 g/liter solution of methylene blue (the substance used in [5]) in methyl alcohol. Figs. 7 and 8 show the absorption characteristics for these substances, obtained with a Carl Zeiss Jena Specord 71 and 72 IR.

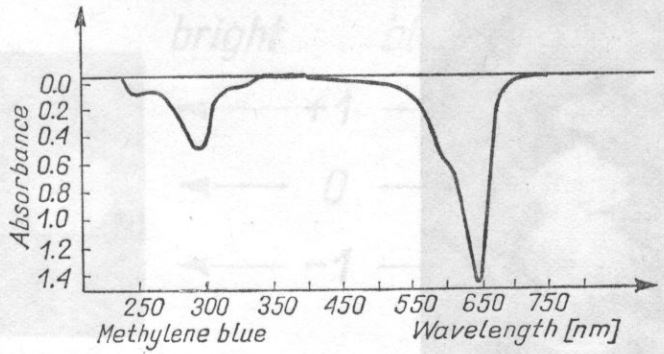


Fig. 7. Optical absorption characteristics of methylene blue

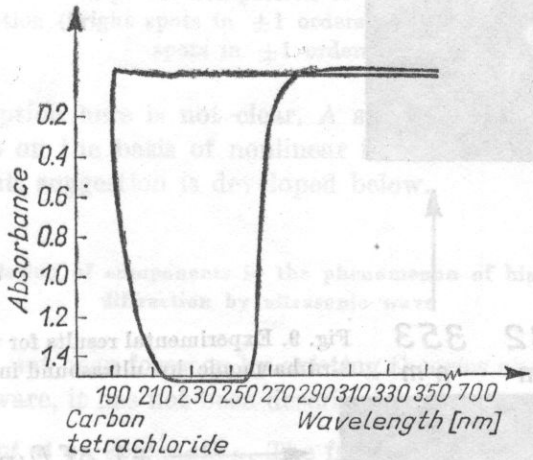


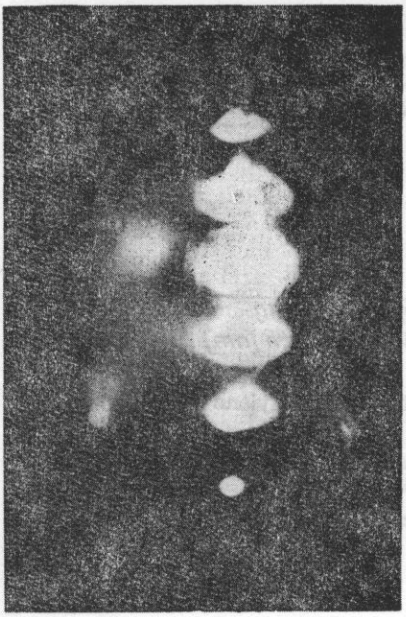
Fig. 8. Optical absorption characteristics of carbon tetrachloride

5. Results

Figs. 9 and 10 represent some evidence of the expected pattern, where the third harmonic was diffracted by the ultrasonic beam.

In addition to obtaining such a pattern, in the experiments of M. Kosmol a new effect of the appearance of a black hole in the first of diffraction order instead of the expected light maximum was discovered (Fig. 11, a, b). (The diffraction of the 0-order beam to an area large compared with those of the ± 1 order spots is connected with some additional effects of light scattering associated with obtaining a letter collimation of pulse beam with a duration of about 40 ns.)

The situation is closely analogous to the observation of absorption lines in a light spectrum, e.g. Fraunhofer lines in the spectrum of the sun. The reason



1058 nm 632 nm 353 nm

Fig. 9. Experimental results for the diffraction of the 3-rd harmonic by ultrasound in carbon tetrachloride

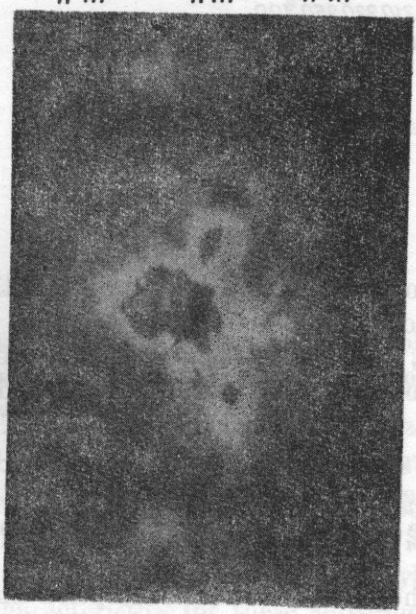


Fig. 10. Experimental diffraction pattern in carbon tetrachloride for a light pulse of duration about 40 ns and 20 MW/cm^2

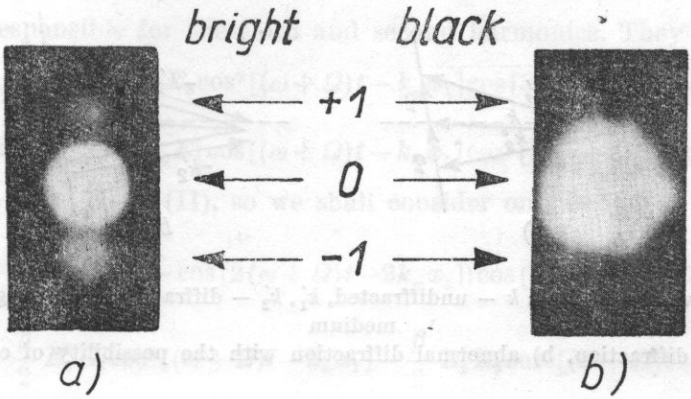


Fig. 11. Comparison of two patterns
a) for normal diffraction (bright spots in ± 1 orders), b) for abnormal diffraction (black spots in ± 1 orders)

for such an absorption here is not clear. A suggestion has been proposed to explain this effect on the basis of nonlinear interaction in a multidiffraction effect [12], and this suggestion is developed below.

6. Selective annihilation of components in the phenomenon of high-power light pulse diffraction by ultrasonic wave

The effect has not been foreseen by existing theories and, as far as the present authors are aware, it has not been described in the literature.

6.1. *An attempt at an explanation.* The fact of the cancellation of coherent beams diffracted at angles corresponding to the ± 1 orders may be interpreted only as a result of the appearance of other coherent beams propagating at the same angles but in opposite direction and with phases able to cancel the emission line, thus giving an absorption line (the medium is absorbing this wave) (Fig. 12).

Let us consider the generation of the third harmonic in a nonlinear medium with the 3-rd order nonlinearity and the diffraction of the light of this harmonic into the first order (± 1 order), for instance.

Further, let us assume that the $+1$ (or -1) beam has still sufficiently high power to interact nonlinearly with the medium: in particular, the 3-rd harmonic beam will interact with the fundamental. Assuming that the light diffraction by ultrasound can be multiple (reiterated) as BRILLOUIN suggested in 1922, we may consider an intermediate plane in the ultrasonic beam (Fig. 13), where there is a second process of diffraction. The vibrations of the light vectors of the overlapping and interacting beams of the fundamental and the 3-harmonic must there satisfy the formula

$$E_{\pm 1}^3 = \beta [E_\omega^{(+1)} + E_{2\omega}^{(+2)}]^3 = E_\omega^{(+1)^3} + E_{3\omega}^{(+3)^3} + 3E_\omega^{(+1)^2} E_{3\omega}^{(+3)} + 3E_\omega^{(+1)} E_{3\omega}^{(+3)^2}. \qquad (21)$$

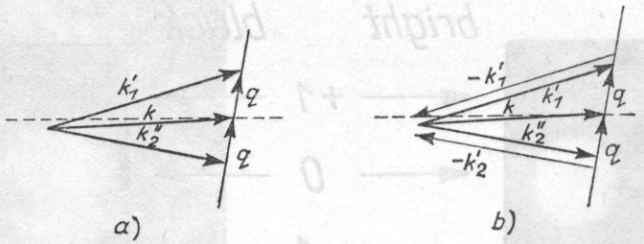


Fig. 12. Wave vectors of light: k – undiffracted, k_1', k_2' – diffracted interacting in a nonlinear medium
a) normal diffraction, b) abnormal diffraction with the possibility of cancellation

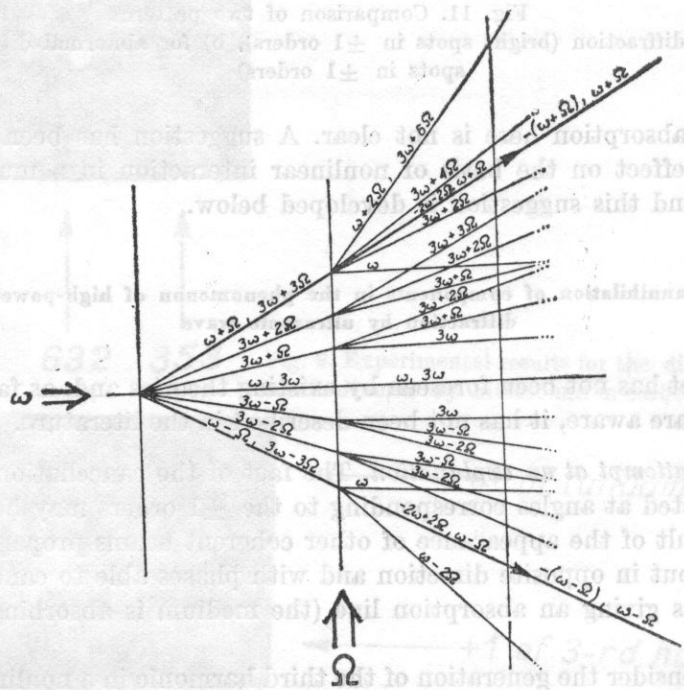


Fig. 13. Diffraction of a very high power light beam by an ultrasonic wave when the third harmonic of the light is generated as a multiple (reiterated) process considered for two-fold case

Putting

$$E_\omega^{(+1)} = E_0 \cos[(\omega+\Omega)t - k_\omega x_{+1}],$$
$$E_\omega^{(+3)} = E_3 \cos[3(\omega+\Omega)t - k_{3\omega} x_{+1}], \tag{22}$$

where $k_{3\omega} = 3k_\omega$, we obtain (among others) expressions such as

$$\cos^3 x = \tfrac{1}{4}(3\cos x + \cos 3x), \qquad \cos^2 x = \tfrac{1}{2}(1 - \cos 2x),$$

which are responsible for the third and second harmonics. They are:

$$\text{(I)}\ E_\omega^2 \cdot E_{3\omega} = \beta E_0^2 \cdot E_3 \cos^2[(\omega+\Omega)t - k_\omega x_1]\cos[3(\omega+\Omega)t - 3k_\omega x_1], \tag{23}$$

$$\text{(II)}\ E_\omega \cdot E_{3\omega}^2 = \beta E_0 E_3^2 \cos[(\omega+\Omega)t - k_\omega x_1]\cos^2[3(\omega+\Omega)t \cdot 3k_\omega x_1]. \tag{24}$$

But we have (I) $\gg$ (II), so we shall consider only

$$\text{(I)} = \frac{\beta}{2} E_0^2 E_3 \{(1 - \cos[2(\omega+\Omega)t - 2k_\omega x_1])\cos[3(\omega+\Omega)t - 3k_\omega x_1]\}$$

$$= \frac{\beta}{2} E_0^2 E_3 \cos^3[(\omega+\Omega)t - k_\omega x_1] - \frac{\beta}{2} E_0^2 E_3 \cos^2[(\omega+\Omega)t - k_\omega x_1] \times$$

$$\times \cos 3[(\omega+\Omega)t - k_\omega x_1]. \tag{25}$$

Since

$$\cos\alpha\cos\beta = \tfrac{1}{2}[\cos(\alpha+\beta) + \cos(\alpha-\beta)], \quad \text{where } x = [(\omega+\Omega)t - k_\omega x_1],$$

we have

$$\cos 2[(\omega+\Omega)t - k_\omega x_1]\cos 3[(\omega+\Omega)t - k_\omega x_1] \Rightarrow$$

$$\Rightarrow \cos\{2[(\omega+\Omega)t - k_\omega x_1] + 3[(\omega+\Omega)t - k_\omega x_1]\} +$$

$$+ \cos\{2[(\omega+\Omega)t - k_\omega x_1] - 3[(\omega+\Omega)t - k_\omega x_1]\} \Rightarrow$$

$$\cos 5[(\omega+\Omega)t - k_\omega x_1] + \cos\{-[(\omega+\Omega)t - k_\omega x_1]\} \Rightarrow$$

$$\Rightarrow \cos 5[(\omega+\Omega)t - k_\omega x_1] + \cos\{-[(\omega+\Omega)t + k_\omega x_1]\} =$$

$$= \cos 5[(\omega+\Omega)t - k_\omega x_1] + \cos[(\omega+\Omega)t + k_\omega x_1]. \tag{26}$$

A similar result may be obtained for (II).

Returning to Fig. 12 (b) and attributing the expressions for the wave numbers k_1' and $-k_1'$,

$$\left.\begin{aligned} k_1' &\Rightarrow \cos[(\omega+\Omega)t - k_\omega x_{+1}] \\ -k_1' &\Rightarrow \cos[(\omega+\Omega)t + k_\omega x_{+1} s] \end{aligned}\right\} +1, \tag{27}$$

$$\left.\begin{aligned} k_2' &\Rightarrow \cos[(\omega-\Omega)t - k_\omega x_{-1}] \\ -k_2' &\Rightarrow \cos[(\omega-\Omega)t + k_\omega x_{-1}] \end{aligned}\right\} -1 \text{ order}, \tag{28}$$

we see that the beams which can cancell one another appear as a result of the nonlinearity of the medium.

6.2. *Discussion and conclusions.* It is worthwhile to consider the physical mechanism of this interaction. Let us consider the induced Mandelsztam-Brillouin effect as a possible explanation. Under the influence of light nonlinear polarization, the density of a medium is chan-

ged due to an electrostrictive effect (see formula (11)) and, as a consequence, an elastic wave (M. B. effect) is induced.

The energy from a light beam of frequency $\omega+\Omega$ is pumped into the ultrasonic wave of frequency Ω (this corresponds to the vector q in Fig. 12) but with opposite phase, thus giving the strong absorption of this particular component. We can also say that this light induced (pumped) elastic energy forms an elastic wave propagating in the opposite direction ($-q$) which gives the appearance of a light beam in exactly the opposite direction (opposite in phase) which is coherent to the first beam and is cancelling it out. The first beam gives the emission line (or bright spot) but the second transformed beam gives the absorption line (or black spot) as the result of its interaction with the first beam.

This is only a qualitative conception, which requires much more examination both in experiment as in theory to evaluate the effect quantitatively.

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