

## ON THE ACOUSTOOPTICS OF AN INTENSE LASERBEAM IN A LIQUID

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The acoustooptic diffraction of an intense laserbeam is described theoretically, when the ultrasonic field is preceded and followed by an undisturbed part of the liquid, as may be the case in experiments. The approach is based on a combination of Bloembergen's theory of third harmonic generation and the generalized Raman-Nath theory for acoustooptic diffraction. The coupled amplitude equations for the fundamental and the third harmonic light waves can be solved exactly in the case of large ultrasonic wavelengths. The spectral character is expressed by Bessel functions containing the width of the ultrasonic field in the argument, while the total light path is included in the amplitudes and phase-shifts. The intensities of the diffracted spectral lines are then computed exactly.

### INTRODUCTION

When an intense laser-light beam is diffracted by an ultrasonic wave in a liquid, nonlinear effects occur besides the usual acoustooptic interaction known from the diffraction of ordinary light. For light of high intensity the whole liquid acts as a nonlinear medium, which gives rise to third harmonic generation (THG). Besides the acoustooptic diffraction of the fundamental light wave, there will be a similar effect on the third harmonic. This results in the appearance of supplementary diffraction lines in the spectrum.

A recent theoretical treatment of this phenomenon was given by the authors (Mertens et al. 1980, Hereman et al. 1981), based on a consistent combination of Bloembergen's theory of THG (Armstrong et al. 1962, Bloembergen 1965) and the generalized Raman-Nath theory of acoustooptic diffraction of ordinary light (Mertens and Hereman 1980). In the combined treatment mentioned above the ultrasonic field was supposed to pervade the whole of the liquid. It is the purpose of the present paper to give a theoretical description of the acoustooptic diffraction of an intense laserbeam, when the ultrasonic field is preceded and possibly followed by an undisturbed part of the liquid, as may be the case in experiments.

The starting point will be a nonlinear wave equation, in which the nonlinear susceptibility is kept constant throughout the whole liquid, whereas the periodic part of the linear susceptibility incorporates the necessary Heaviside functions in order to make it vanish in those regions of the liquid not disturbed by the ultrasonic wave. We then find that the amplitudes of the fundamental and the third harmonic light waves satisfy a coupled system of first-order PDEs, expressing the interaction of those light waves throughout all parts of the liquid. These equations can be solved exactly in the case of large ultrasonic wavelengths. The spectral character of the diffracted waves may be expressed with the help of Bessel functions containing the width of the ultrasonic field in the argument, whereas the total light path (in the

liquid) is included in the amplitudes and the phase-shifts of the different subwaves. At the end, it is then possible to compute in an exact way the intensities of the diffracted spectral lines.

### THIRD HARMONIC GENERATION

We consider a parallel beam of intense laser-light of frequency  $\nu = \omega/2\pi$  and wavenumber  $k$  passing through a liquid column of total width  $L$ . Supposing a normal incidence (as indicated in Figure 1), we put the  $z$ -axis along the direction of the incident laserbeam and take the  $x$ -axis along the direction of propagation of a progressive ultrasonic wave of frequency  $\nu^* = \omega^*/2\pi$  and wavenumber  $k^*$ . The ultrasonic field occupies a region  $z \in [z_A, z_B]$  inside the liquid, such that  $0 < z_A < z_B \leq L$ , giving two undisturbed regions  $z \in [0, z_A]$  and  $z \in [z_B, L]$ . The total wave electric field is in lowest approximation

$$E = \frac{1}{2} A_1(z, k^*x - \omega^*t) \exp i(kz - \omega t) + \frac{1}{2} A_3(z, k^*x - \omega^*t) \exp 3i(kz - \omega t) + \text{c.c.} \quad (1)$$

$E$  contains a fundamental laser-light wave with amplitude  $A_1$  and its third harmonic with amplitude  $A_3$ . Outside the liquid only the fundamental is present; THG occurs inside the liquid viewed as a nonlinear dielectricum. Hence at the plane of incidence:

$$A_1(0, k^*x - \omega^*t) = A_0, \quad A_3(0, k^*x - \omega^*t) = 0. \quad (2)$$

Furthermore,  $A_1$  and  $A_3$  vary slowly with the penetration depth inside the liquid, as well as with the slow periodic disturbance of the medium through the ultrasonic wave in the center region.

The equations for these slow variations are derived in the general treatment by Hereman et al. (1981):

$$\begin{aligned} 8i(1+\chi_0) \frac{\partial A_1}{\partial z} + 4k\chi_1 A_1 + 3k\chi_3 (\bar{A}_1^2 A_3 + A_1^2 \bar{A}_1 + 2A_1 A_3 \bar{A}_3) &= 0, \\ 8i(1+\chi_0) \frac{\partial A_3}{\partial z} + 12k\chi_1 A_3 + 3k\chi_3 (A_1^3 + 6A_1 \bar{A}_1 A_3 + 3A_3^2 \bar{A}_3) &= 0. \end{aligned} \quad (3)$$

Here  $\chi_0$  and  $\chi_1$  respectively stand for the constant and the periodically varying parts of the linear susceptibility, whereas  $\chi_3$  refers to the lowest-order nonlinear susceptibility of the liquid.  $\chi_3$  is supposed to be constant throughout the liquid, because the ultrasonic disturbance of this quantity gives a higher-order effect. Using the substitution

$$\zeta = \frac{3}{8} \frac{k\chi_3}{1+\chi_0} z \quad (4)$$

and putting

$$A_1 = \rho_1 \exp i\beta_1, \quad A_3 = \rho_3 \exp i\beta_3, \quad (5)$$

allows us to split (3) into its real and imaginary parts:

$$\begin{aligned}
\frac{\partial \rho_1}{\partial \zeta} &= \rho_1^2 \rho_3 \sin \phi , \\
\frac{\partial \rho_3}{\partial \zeta} &= -\rho_1^3 \sin \phi , \\
\frac{\partial \beta_1}{\partial \zeta} &= \frac{4\chi_1}{3\chi_3} + \rho_1 \rho_3 \cos \phi + \rho_1^2 + 2\rho_3^2 , \\
\frac{\partial \beta_3}{\partial \zeta} &= \frac{4\chi_1}{\chi_3} + \rho_1^3 \rho_3^{-1} \cos \phi + 6\rho_1^2 + 3\rho_3^2 .
\end{aligned} \tag{6}$$

Here  $\phi$  stands for  $3\beta_1 - \beta_3$ , such that

$$\frac{\partial \phi}{\partial \zeta} = \rho_1 \rho_3^{-1} (3\rho_3^2 - \rho_1^2) \cos \phi - 3\rho_1^2 + 3\rho_3^2 . \tag{7}$$

The equations for  $\rho_1$ ,  $\rho_3$  and  $\phi$  are exactly the same as in the previous treatment (Hereman et al. 1981), so that the solutions for  $\rho_1$  and  $\rho_3$  are

$$\rho_1^2 = \frac{9\rho_0^2}{11 - 2\cos 3\rho_0^2 \zeta} , \quad \rho_3^2 = 2\rho_0^2 \frac{1 - \cos 3\rho_0^2 \zeta}{11 - 2\cos 3\rho_0^2 \zeta} , \tag{8}$$

where  $\rho_0$  is an integration constant related to the initial wave energy:

$$\rho_0^2 = \rho_1^2(0) = A_1(0, k^*x - \omega^*t) \bar{A}_1(0, k^*x - \omega^*t) = A_0 \bar{A}_0 . \tag{9}$$

#### DISTINCTION BETWEEN THE ACOUSTOOPTIC AND UNDISTURBED REGIONS OF THE LIQUID

It is now time to explicitly take into account that the ultrasonic field is only present in  $z \in [z_A, z_B]$  or  $\zeta \in [\zeta_A, \zeta_B]$  :

$$\chi_1(\zeta, k^*x - \omega^*t) = \hat{\chi}_1(k^*x - \omega^*t) H(\zeta - \zeta_A) H(\zeta_B - \zeta) , \tag{10}$$

where  $H$  is Heaviside's function of the appropriate argument. When (10) is substituted into the last two equations (6), the determination of  $\beta_1$  and  $\beta_3$  follows as

$$\beta_1(\zeta) = \alpha_1(\zeta) + \frac{4\hat{\chi}_1}{3\chi_3} F(\zeta) , \quad \beta_3(\zeta) = \alpha_3(\zeta) + \frac{4\hat{\chi}_1}{\chi_3} F(\zeta) , \tag{11}$$

with

$$\begin{aligned}
\alpha_1(\zeta) &= \frac{1}{2}\rho_0^2 \zeta + \gamma(\zeta) + \beta_1(0) , \\
\alpha_3(\zeta) &= 3\rho_0^2 \zeta + 3\gamma(\zeta) + \beta_3(0) ,
\end{aligned} \tag{12}$$

and



$$\gamma(\zeta) = \frac{1}{\sqrt{13}} \arctan\left(\frac{\sqrt{13}}{3} \tan^2 \frac{3}{2} \rho_0^2 \zeta\right), \quad (13)$$

$$F(\zeta) = (\zeta - \zeta_B)H(\zeta - \zeta_A)H(\zeta_B - \zeta) + (\zeta_B - \zeta_A)H(\zeta - \zeta_A).$$

#### DIFFRACTION SPECTRUM

We consider the linear susceptibility of the medium to be modulated by a simple progressive ultrasonic wave, such that

$$\hat{\chi}_1 = \delta \sin(kx - \omega t). \quad (14)$$

Using the expressions (8) for  $\rho_1$  and  $\rho_3$ , (11) for  $\beta_1$  and  $\beta_3$  and applying the formula of Jacobi

$$e^{i\xi \sin \theta} = \sum_{n=-\infty}^{+\infty} e^{in\theta} J_n(\xi), \quad (15)$$

we find the expression for the wave electric field (1), written in the original variables,

$$\begin{aligned} E(x, z, t) = & \sum_{m=-\infty}^{+\infty} 3\rho_0 \left\{ 9 + 4 \sin^2 \left( \frac{9k\rho_0^2 \chi_3}{16 + 16\chi_0} z \right) \right\}^{-1/2} \\ & \cdot (-1)^m J_m \left( \frac{k\delta F(z)}{2 + 2\chi_0} \right) \cos[kz - mkx - (\omega - m\omega^*)t + \alpha_1(z)] \\ & + \sum_{n=-\infty}^{+\infty} 2\rho_0 \left| \sin \left( \frac{9k\rho_0^2 \chi_3}{16 + 16\chi_0} z \right) \right| \left\{ 9 + 4 \sin^2 \left( \frac{9k\rho_0^2 \chi_3}{16 + 16\chi_0} z \right) \right\}^{-1/2} \\ & \cdot (-1)^n J_n \left( \frac{3k\delta F(z)}{2 + 2\chi_0} \right) \cos[3kz - nkx - (3\omega - n\omega^*)t + \alpha_3(z)], \end{aligned} \quad (16)$$

if

$$F(z) = (z - z_B)H(z - z_A)H(z_B - z) + (z_B - z_A)H(z - z_A). \quad (17)$$

It is obvious that the ultrasonic field in the liquid acts as a diffraction grating for the incident laser-light wave. In addition, the argument of the Bessel functions in (16) depends on  $F(z)$  instead of simply on  $z$  as in earlier treatments (Hereman et al. 1981). Two cases are thus to be distinguished concerning the position  $z_S$  of the screen on which the diffraction pattern is observed. As it would be pointless to put the screen before the ultrasonic field, one can have that either  $z_A \leq z_S \leq z_B$  and then  $F(z_S) = z_S - z_A$  or  $z_B \leq z_S \leq L$ , in which case  $F(z_S) = z_B - z_A$ . In both cases we will use  $W$  to denote the effective width of the ultrasonic field from  $z_A$  to the position of the screen  $z_S$ .

The original incident wave is split into a spectrum of sub-waves of two types :

##### i) Ordinary diffraction lines (of order $m$ , $m \in \mathbb{Z}$ )

These diffraction lines are found at the same position as in a linear medium, if ordinary instead of intense laser-light were used, but of course their intensities will be different. Each spectral component of order  $m$  is characterized by a frequency-shift

$$\Delta v_m = -mv^* \quad (18)$$

and a deflection angle  $\theta_m$  with the positive z-axis:

$$\theta_m = -mk^*/k . \quad (19)$$

The intensity of such a line is

$$I_m = \rho_0^2 \frac{9J_m^2 \left( \frac{k\delta W}{2+2\chi_0} \right) + 4J_{3m}^2 \left( \frac{3k\delta W}{2+2\chi_0} \right) \sin^2 \left( \frac{9k\rho_0^2\chi_3}{16+16\chi_0} z_S \right)}{9 + 4 \sin^2 \left( \frac{9k\rho_0^2\chi_3}{16+16\chi_0} z_S \right)} , \quad (m \in \mathbb{Z}) \quad (20)$$

since part of the diffraction pattern due to the third harmonic coincides with the original pattern due to the fundamental.

ii) Intermediate diffraction lines (of order  $n/3$ ,  $n \in \mathbb{Z}$ ,  $n/3 \notin \mathbb{Z}$ )

Such lines do not occur in the diffraction spectrum for an ordinary light beam, because they are exclusively due to THG in the liquid. For these intermediate lines the frequency-shifts are

$$\Delta v_{n/3} = -\frac{n}{3}v^* , \quad (21)$$

with deflections given by

$$\theta_{n/3} = -nk^*/3k \quad (22)$$

and with intensities

$$I_{n/3} = \frac{4\rho_0^2 J_n^2 \left( \frac{3k\delta W}{2+2\chi_0} \right) \sin^2 \left( \frac{9k\rho_0^2\chi_3}{16+16\chi_0} z_S \right)}{9 + 4 \sin^2 \left( \frac{9k\rho_0^2\chi_3}{16+16\chi_0} z_S \right)} , \quad (n \in \mathbb{Z}, \frac{n}{3} \notin \mathbb{Z}) . \quad (23)$$

As a final remark, one can check that the total intensity of the diffraction spectrum exactly equals the intensity of the incident laser-light beam.

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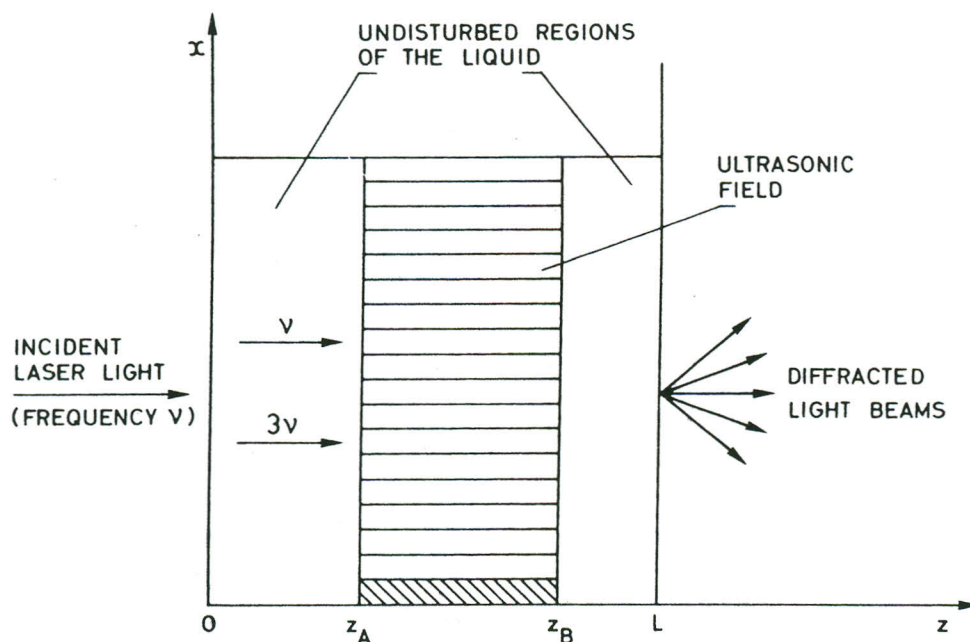


Figure 1. Geometry of the diffraction phenomenon