Acousto-Optic Diffraction of Intense Laser-Light in a Liquid

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Summary

The acousto-optic interaction of an intense laser-light beam with an ultrasonic wave in a liquid is studied theoretically. Starting from Maxwell's equations, a multiple space-time scale formalism is used which incorporates both the electromagnetic nonlinearities and the ultrasonic disturbance of the liquid. As in nonlinear optics, the inclusion of a third-order polarization leads to a set of nonlinear coupled wave equations, but with periodically varying coefficients due to the ultrasonic influence. The amplitudes of the fundamental and the third harmonic are coupled. In the case of large ultrasonic wavelengths this set of nonlinear equations can be solved exactly. Using the generating function method, the intensities of the diffracted spectrum are computed in closed form. This spectrum consists of two types of lines: ordinary lines, as with diffraction of lower-intensity light, and intermediate lines, due to third harmonic generation in the liquid. In contrast with earlier work, the sum of the intensities of all the diffraction lines exactly equals the intensity of the incident light beam.

Akusto-optische Beugung intensiven Laserlichtes in einer Flüssigkeit

Zusammenfassung

Die akusto-optische Wechselwirkung eines starken Laserstrahls mit einer Ultraschallwelle in einer Flüssigkeit wird theoretisch untersucht. Die Maxwellschen Gleichungen werden als Ausgangspunkt genommen, und ein vielfach Raum-Zeit-Skala-Formalismus wird eingeführt, der die elektromagnetischen Nichtlinearitäten und die Ultraschallstörung in der Flüssigkeit umfaßt. Wie in der nichtlinearen Optik führt die Einschließung einer Polarisation dritter Ordnung zu einem System von nichtlinearen gekoppelten Wellengleichungen, die jedoch aufgrund des Einflusses des Ultraschalls periodische Koeffizienten besitzen. Die Amplituden des Grundtons und des zweiten Obertons der Lichtwelle sind gekoppelt. Im Falle größerer Ultraschallwellenlängen ist das System nichtlinearer Gleichungen exakt lösbar. Unter Anwendung der Methode der erzeugenden Funktion lassen sich die Intensitäten des Beugungsspektrums in geschlossener Form berechnen. Dieses Spektrum besteht aus zwei Typen von Linien: gewöhnliche Linien, wie bei der Beugung des Lichtes mit schwacher Intensität, und Zwischenlinien, herbeigeführt durch die in der Flüssigkeit erzeugten dritten Harmonischen. Im Gegensatz zu früheren Arbeiten ist die Summe der Intensitäten aller gebeugten Linien vollkommen gleich der Intensität des einfallenden Lichtes.

Diffraction acousto-optique d'une lumière laser intense à l'intérieur d'un liquide

Sommaire

On fait la théorie de l'inter-action acousto-optique d'un faisceau intense de lumière laser avec un liquide. Partant des équations de Maxwell, on y introduit un formalisme à échelles multiples dans l'espace-temps, ce qui permet de traiter aussi bien les non-linéarités électromagnétiques que la perturbation ultrasonore du liquide traversé. Comme en optique non-linéaire, l'inclusion d'une polarisation du troisième ordre mène à un système d'équations d'onde non-linéaires couplées, mais ici avec des coefficients qui varient périodiquement sous l'influence des ultrasons. L'amplitude de l'onde fondamentale est couplée à celle du troisième harmonique. Dans le cas des ultrasons de plus grandes longueurs d'onde, ce système d'équations non-linéaires peut ête résolu exactement. La méthode de la fonction génératrice fournit les intensités du spectre diffracté en formules finies. Ce spectre se compose de deux types de raies: les raies ordinaires qui sont celles de la diffraction de la lumière aux faibles intensités et les raies intermédiaires qui proviennent de la génération d'un troisième harmonique dans le liquide. En contraste avec des travaux antérieurs, la somme des intensités dans le spectre diffracté est ici égale exactement à l'intensité du faisceau incident.

1. Introduction

The diffraction of ordinary light by an ultra-sonic wave in a liquid has been well explained by the so-called Raman-Nath theory [1]. In this theory, a monochromatic light beam traverses a liquid column, the permittivity of which varies periodically but with a sufficiently low frequency. The light beam is scattered into many spectral harmonic components due to both Doppler shifts and deflection. For the well-separated lines of this diffraction pattern, it is possible to determine the directions, frequencies and intensities [2], [3]. Up to now this kind of diffraction phenomena has been studied quite extensively, from a theoretical [4], [5], [6] as well as from an experimental point of view [7], [8]. Recent and important fields of application are the study of optical image-forming systems [9], [10], acoustical holography [11], acousto-optical light modulation [8], [12] and deflectors [8], [13].

In many of these recent experiments one needs to use strong laser-light beams, but then the intensity of the electric field is so high that besides the acousto-optic interactions several nonlinear effects may occur. Such experiments [14], [15], as well as theoretical arguments from nonlinear optics [16], [17], [18], show that the nonlinear terms in the polarization are no longer negligible and cause e.g. the well-known optical harmonic generation. Due to symmetry properties of isotropic media (in particular a liquid), the term containing the fourth-rank susceptibility tensor is the first important term in the nonlinear polarization. The corresponding third harmonic generation (THG) has been demonstrated first by Terhune et al. [19] for calcite crystals and by Goldberg and Schuur [20] for a liquid medium (in particular a liquid crystal). Such a THG shows up also in the acousto-optical diffraction of an intense laser-light beam [21], [22], with a pattern consisting of two types of lines, in contrast with the spectrum for ordinary light when diffracted by a simple progressive ultrasonic wave. Besides the ordinary diffraction lines, at the same places as if the medium were linear, one observes [14], [15] intermediate lines due to THG in the acousto-optical interaction region (see paragraph 4). Several theoretical attempts to explain this experimental result have been made, which were reviewed by Kosmol and Sliwinski [15]. Although they explain the observed diffraction pattern, none of these theoretical investigations are really satisfactory concerning the calculations of the intensities.

The purpose of the present paper is to present a new approach which relates the theory of THG, established by Bloembergen and coworkers [23], to the generalized Raman-Nath theory for the diffraction of light by ultrasonic waves, recently reviewed by Mertens et al. [24]. Since both these theories start from Maxwell's equations, a global approach is possible, combining different but reconcilable approximations.

In paragraph 2 this is done, using a multiple space-time scale formalism, which is already implicit in both the THG and Raman-Nath theories. In this way special attention is paid to the comparison and relative importance of various small effects in the derivation of the basic equations. A coupled system of first-order but nonlinear PDEs is thus obtained, relating the amplitudes of the fundamental and the third harmonic light waves. This system contains coefficients, which vary slowly in a periodic way due to the ultrasonic influence on the liquid.

The exact integration of this set of coupled equations is performed in paragraph 3, and up to this stage the explicit form of the linear and nonlinear susceptibilities is not required. For the calculation in paragraph 4 of the intensities of the diffraction pattern, in the case of a progressive ultrasonic, however, the explicit form of the disturbing wave pattern must be known. In order to apply the generating function method (GFM), established by Hereman and Mertens [25], the nonlinear susceptibility should be constant.

In contrast with previous approaches, the sum of the calculated intensities of the various lines of the diffraction spectrum is exactly equal to the intensity of the incident light beam. Finally, a review of the earlier theoretical treatments of this problem in comparison to the present approach has been made in paragraph 5.

2. Basic equations

The inclusion of nonlinear effects in the treatment of electro-magnetic wave propagation through media, which are disturbed by ultrasonic waves, requires a consistent derivation of the basic equations, as many different physical approximations are involved. Hence one starts from Maxwell's equations without source terms, written in SI units:

$$\nabla \times \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = 0, \quad \nabla \cdot \boldsymbol{B} = 0,$$

$$\nabla \times \boldsymbol{H} - \frac{\partial \boldsymbol{D}}{\partial t} = 0, \quad \nabla \cdot \boldsymbol{D} = 0. \tag{1}$$

Here E and H represent the electric and magnetic fields, whereas D and B are the electric displacement and magnetic induction, characteristic for the medium under consideration. For the applications we have in mind, the magnetic effects are not important. So we can simply, as in vacuum, replace B by $\mu_0 H$ and eliminate these magnetic quantities from eq. (1) to get a wave equation of the general form

$$\nabla \times (\nabla \times E) + \mu_0 \frac{\partial^2}{\partial t^2} D = 0. \qquad (2)$$

μ_0 is the magnetic permeability coefficient in vacuum, and we will use ε_0 to denote the dielectric constant in vacuum. These quantities are related to the vacuum velocity of light c through $\varepsilon_0 \mu_0 c^2 = 1$. For eq. (2) to be a true wave equation, a relationship between D and E must be given. As we want to include some nonlinear effects, we put in a general way

$$D = \varepsilon_0 (1 + \chi_L + \chi_{NL} E^2) E, \qquad (3)$$

where χ_L and χ_{NL} respectively stand for the susceptibilities, appearing in the linear and nonlinear polarizations of the medium. In general, such susceptibilities would be operators in tensorial form, which for isotropic media reduce to scalar operators. Even though the liquid is disturbed by an ultrasonic wave, which propagates in a given physical direction, we will for simplicity assume that this liquid acts as an isotropic medium. Furthermore, the effect of χ_L and χ_{NL} on E is supposed to be independent of the frequencies of the incident light, so that χ_L and χ_{NL} are scalar functions.

Substitution of eq. (3) into eq. (2) yields

$$\nabla \nabla \cdot E - \nabla^2 E + \frac{1}{c^2} \frac{\partial^2}{\partial t^2}$$
$$\cdot [(1 + \chi_L + \chi_{NL} E^2) E] = 0, \qquad (4)$$

together with the condition

$$\nabla \cdot [(1 + \chi_L + \chi_{NL} E^2) E] = 0. \qquad (5)$$

Basically, there are two series of effects in the perturbation of the medium. One is the change in dielectric properties due to the presence of ultrasonic waves, and on its own this would lead to various forms of the Raman-Nath theory. The other effects arise from the nonlinearity of the medium, which has to be taken into account when dealing with intense laser-light, and this on its own would give THG in nonlinear optics. Clearly, both these effects must be comparable in a combined study, otherwise one is reduced to one of either limiting case. We thus adopt the following ordering

scheme, which includes a multiple space-time formalism [26]:

$$\nabla = \nabla_0 + \varepsilon \nabla_1 + \cdots,$$
$$\frac{\partial}{\partial t} = \frac{\partial}{\partial t_0} + \varepsilon \frac{\partial}{\partial t_1} + \cdots,$$
$$E = E_0 + \varepsilon E_1 + \cdots, \qquad (6)$$
$$\chi_L = \chi_0 + \varepsilon \chi_1 + \cdots,$$
$$\chi_{NL} = \varepsilon \chi_3 + \cdots.$$

Here $r_0 = r$ and $t_0 = t$ represent the fast space and time-scales on which the electromagnetic waves vary in phase, whereas $r_1 = \varepsilon r$ and $t_1 = \varepsilon t$ are the slow scales, on which the wave amplitudes will vary as a result of the presence of the ultrasonic wave and the THG. The various effects are of the same order in ε, characterized essentially by the nonlinear susceptibility. It will be seen later on the resulting equations, that the ordering in eq. (6) leads in the absence of nonlinear effects to the Raman-Nath regime in the usual theories of the diffraction of light by ultrasonic waves [27], [28].

Upon substitution of eq. (6) into eq. (4) one gets, after equating all terms of the same order in ε, the set of equations:

$$L E_0 = 0,$$
$$L E_1 - \frac{2 \chi_3}{1 + \chi_0} \nabla_0 (E_0 E_0 : \nabla_0 E_0) \qquad (7)$$
$$- 2 \nabla_0 \cdot \nabla_1 E_0 + \frac{\chi_1}{c^2} \frac{\partial^2}{\partial t_0^2} E_0$$
$$+ \frac{\chi_3}{c^2} \frac{\partial^2}{\partial t_0^2} (E_0^2 E_0) + 2 \frac{1 + \chi_0}{c^2} \frac{\partial^2}{\partial t_0 \partial t_1} E_0 = 0,$$

where use has already been made of the conditions derived from eq. (5):

$$\nabla_0 \cdot E_0 = 0,$$
$$\nabla_0 \cdot E_1 + \nabla_1 \cdot E_0 + \frac{\chi_3}{1 + \chi_0} E_0 \cdot \nabla_0 E_0^2 = 0, \qquad (8)$$

and the linear operator L has been defined as

$$L = -\nabla_0^2 + \frac{1 + \chi_0}{c^2} \frac{\partial^2}{\partial t_0^2}. \qquad (9)$$

The set (7) contains all the information required, if we specify how χ_1 and χ_3 vary with space and time. If the ultrasonic wave propagates in the x-direction with a velocity c^* (starred quantities refer to the ultrasonic wave), χ_1 and χ_3 can at most depend upon $x_1 - c^* t_1$, because the variations in the ultrasonic waves are slow compared to those in the electromagnetic waves. This again is equivalent to the generalized Raman-Nath theory [1], [24]. The explicit form of χ_1 and χ_3 is not yet needed, and a

further discussion will be postponed until paragraph 4.

3. Third harmonic generation

It is now time to specify the form of E_0, the electric field inside the medium in lowest approximation. We take a linearly polarized light wave which propagates in the z-direction in lowest approximation:

$$E_0 = E_0\,e_y,$$
$$E_0 = \tfrac{1}{2} A_1(z_1, x_1 - c^* t_1)\exp(i\varphi)$$
$$+ \tfrac{1}{2} A_3(z_1, x_1 - c^* t_1)\exp(3i\varphi) + \text{c.c.},$$
$$\varphi = k z_0 - \omega t_0. \tag{10}$$

E_0 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; the third harmonic is generated inside the liquid viewed as a nonlinear dielectricum. Hence at the plane of incidence:

$$A_1(0, x_1 - c^* t_1) = A_0,$$
$$A_3(0, x_1 - c^* t_1) = 0. \tag{11}$$

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 ultrasonic waves. The complex conjugate in eq. (10) is necessary, as we are dealing with nonlinear equations, in which E_0 is thus introduced as a real quantity from the outset. Substitution of eq. (10) into the first equation eq. (7) yields the dispersion law for the electromagnetic waves in the liquid:

$$\omega^2 = \frac{c^2 k^2}{1 + \chi_0}. \tag{12}$$

With the choice of E_0 given in eq. (10), the term $E_0 E_0 : \nabla_0 E_0$ vanishes, and the second equation (7) becomes, after projection on the y-axis:

$$L E_1 + \left\{ -i k \frac{\partial A_1}{\partial z_1} - \frac{\omega^2}{2 c^2}\chi_1 A_1 + i\omega\frac{1+\chi_0}{c^2}c^*\frac{\partial A_1}{\partial x_1} - \frac{3\omega^2}{8 c^2}\chi_3(\bar A_1^2 A_3 + A_1^2 \bar A_1 + 2 A_1 A_3 \bar A_3) \right\}\exp(i\varphi)$$
$$+ \left\{ -3 i k \frac{\partial A_3}{\partial z_1} - \frac{9\omega^2}{2 c^2}\chi_1 A_3 + 3 i\omega\frac{1+\chi_0}{c^2}c^*\frac{\partial A_3}{\partial x_1} - \frac{9\omega^2}{8 c^2}\chi_3(A_1^3 + 6 A_1 \bar A_1 A_3 + 3 A_3^2 \bar A_3) \right\}\exp(3 i\varphi)$$
$$- \frac{75\omega^2}{8 c^2}\chi_3(A_1^2 A_3 + \bar A_1 A_3^2)\exp(5 i\varphi) - \frac{147\omega^2}{8 c^2}\chi_3 A_1 A_3^2 \exp(7 i\varphi) - \frac{81\omega^2}{8 c^2}\chi_3 A_3^3 \exp(9 i\varphi) + \text{c.c.} = 0, \tag{13}$$

where $E_1 = E_1 e_y$; the bar signifies complex conjugation. E_1 will only contain information about harmonics higher than the third, if the slow variation of A_1 and A_3 with z_1 is such that

$$8 i(1 + \chi_0)\frac{\partial A_1}{\partial z_1} + 4 k \chi_1 A_1$$
$$+ 3 k \chi_3(\bar A_1^2 A_3 + A_1^2 \bar A_1 + 2 A_1 A_3 \bar A_3) = 0,$$
$$8 i(1 + \chi_0)\frac{\partial A_3}{\partial z_1} + 12 k \chi_1 A_3 \tag{14}$$
$$+ 3 k \chi_3(A_1^3 + 6 A_1 \bar A_1 A_3 + 3 A_3^2 \bar A_3) = 0.$$

Those equations, obtained by putting equal to zero the coefficients of $\exp(i\varphi)$ and $\exp(3 i\varphi)$ in eq. (13), avoid the appearance of secular terms in E_1, connected with the fundamental and the third harmonic.

Terms with a factor c^*/c, which is always a small parameter, have been neglected. With the substitution

$$\zeta = \frac{3}{8}\frac{k\chi_3}{1 + \chi_0}z_1 \tag{15}$$

eq. (14) becomes

$$\frac{\partial A_1}{\partial\zeta} - i\frac{4\chi_1}{3\chi_3}A_1 = i(\bar A_1^2 A_3 + A_1^2 \bar A_1 + 2 A_1 A_3 \bar A_3),$$
$$\frac{\partial A_3}{\partial\zeta} - i\frac{4\chi_1}{\chi_3}A_3 = i(A_1^3 + 6 A_1 \bar A_1 A_3 + 3 A_3^2 \bar A_3). \tag{16}$$

The structure of these equations is such that if we put

$$A_1 = Z_1(\zeta)\exp\!\left(i\frac{4\chi_1}{3\chi_3}\zeta\right),$$
$$A_3 = Z_3(\zeta)\exp\!\left(i\frac{4\chi_1}{\chi_3}\zeta\right), \tag{17}$$

eq. (16) reduces to

$$\frac{dZ_1}{d\zeta} = i(\bar Z_1^2 Z_3 + Z_1^2 \bar Z_1 + 2 Z_1 Z_3 \bar Z_3),$$
$$\frac{dZ_3}{d\zeta} = i(Z_1^3 + 6 Z_1 \bar Z_1 Z_3 + 3 Z_3^2 \bar Z_3). \tag{18}$$

In order to get real equations, we write

$$Z_1 = \varrho_1 \exp(i\alpha_1), \quad Z_3 = \varrho_3 \exp(i\alpha_3), \tag{19}$$

and split eq. (18) into its real and imaginary

parts:

$$\frac{d\varrho_1}{d\zeta} = \varrho_1^2 \varrho_3 \sin\psi,$$

$$\frac{d\varrho_3}{d\zeta} = -\varrho_1^3 \sin\psi,$$

$$\frac{d\alpha_1}{d\zeta} = \varrho_1 \varrho_3 \cos\psi + \varrho_1^2 + 2\varrho_3^2,$$

$$\frac{d\alpha_3}{d\zeta} = \varrho_1^3 \varrho_3^{-1} \cos\psi + 6\varrho_1^2 + 3\varrho_3^2, \tag{20}$$

where ψ stands for $3\alpha_1 - \alpha_3$. From the last two eqs. (20) one deduces

$$\frac{d\psi}{d\zeta} = \varrho_1 \varrho_3^{-1}(3\varrho_3^2 - \varrho_1^2)\cos\psi - 3\varrho_1^2 + 3\varrho_3^2. \tag{21}$$

The set (20) admits two invariants:

$$\varrho_1^2 + \varrho_3^2 = \varrho_0^2,$$

$$\varrho_1^3 \varrho_3 \cos\psi = \tfrac{3}{4}(\varrho_1^4 + \varrho_3^4) + \gamma, \tag{22}$$

where ϱ_0^2 and γ are integration constants.

In view of the boundary conditions (11), and keeping the transformations (17) and (19) in mind, we find that

$$\varrho_0^2 = \varrho_1^2(0) = Z_1(0)\bar{Z}_1(0)$$
$$= A_1(0, x_1 - c^*t_1)\bar{A}_1(0, x_1 - c^*t_1)$$
$$= A_0\bar{A}_0. \tag{23}$$

The second invariant (22) can be reduced to

$$\varrho_1^3 \varrho_3 \cos\psi = -\tfrac{3}{2}\varrho_1^2 \varrho_3^2. \tag{24}$$

With the help of the invariants (22) one gets instead of eq. (20) a single differential equation of the form

$$\frac{du}{d\zeta} = \pm\sqrt{f(u)}, \tag{25}$$

if we put for simplicity u instead of ϱ_1^2, and where

$$f(u) = u^2(\varrho_0^2 - u)(13u - 9\varrho_0^2). \tag{26}$$

The function $f(u)$ is negative everywhere, except in the interval $[9\varrho_0^2/13, \varrho_0^2]$. Hence u will oscillate between these two values $9\varrho_0^2/13$ and ϱ_0^2 in a periodic way. This is borne out by the integration of eq. (25), which gives

$$u = \frac{9\varrho_0^2}{11 - 2\cos(3\varrho_0^2\zeta)}. \tag{27}$$

Returning to the original variables we have

$$\varrho_1^2 = \frac{9\varrho_0^2}{11 - 2\cos\left(\dfrac{9k\varrho_0^2\chi_{NL}}{8 + 8\chi_0}z\right)},$$

$$\varrho_3^2 = 2\varrho_0^2 \frac{1 - \cos\left(\dfrac{9k\varrho_0^2\chi_{NL}}{8 + 8\chi_0}z\right)}{11 - 2\cos\left(\dfrac{9k\varrho_0^2\chi_{NL}}{8 + 8\chi_0}z\right)}. \tag{28}$$

The period in which the situation would repeat itself spatially in the z-direction is

$$z_P = \frac{16\pi(1 + \chi_0)}{9\varrho_0^2 k\chi_{NL}}. \tag{29}$$

Once the moduli ϱ_1 and ϱ_3 are known, the integration of α_1 and α_3 follows from the last two eqs. (20) as

$$\alpha_1 = \frac{3k\varrho_0^2\chi_{NL}}{16 + 16\chi_0}z + \beta(z) + \alpha_1(0),$$

$$\alpha_3 = \frac{9k\varrho_0^2\chi_{NL}}{8 + 8\chi_0}z + 3\beta(z) + \alpha_3(0), \tag{30}$$

if we put

$$\beta(z) = \frac{1}{\sqrt{13}}\arctan\left[\frac{\sqrt{13}}{3}\tan\left(\frac{9k\varrho_0^2\chi_{NL}}{16 + 16\chi_0}z\right)\right]. \tag{31}$$

Returning now to eq. (17), we have that

$$A_1 = 3\varrho_0\left\{9 + 4\sin^2\left(\frac{9k\varrho_0^2\chi_{NL}}{16 + 16\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot\exp\left[i\left\{\frac{k(\chi_L - \chi_0)}{2 + 2\chi_0}z + \alpha_1(z)\right\}\right],$$

$$A_3 = 2\varrho_0\left|\sin\left(\frac{9k\varrho_0^2\chi_{NL}}{16 + 16\chi_0}z\right)\right|$$
$$\cdot\left\{9 + 4\sin^2\left(\frac{9k\varrho_0^2\chi_{NL}}{16 + 16\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot\exp\left[i\left\{\frac{3k(\chi_L - \chi_0)}{2 + 2\chi_0}z + \alpha_3(z)\right\}\right]. \tag{32}$$

It is worth noting that we succeeded in determining A_1 and A_3 without having to specify the functional dependence of χ_L and χ_{NL} upon $x_1 - c^*t_1$. This will be done in the next section, in order to arrive at the generator description of the diffracted light pattern.

4. Diffraction spectrum

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

$$\chi_L - \chi_0 = \delta\sin(k^*x - \omega^*t), \tag{33}$$

where the slow variation is incorporated in $k^* \ll k$ and $\omega^* \ll \omega$, and such that

$$c^* = \frac{\omega^*}{k^*}. \tag{34}$$

In practice the magnitude of the susceptibility χ_{NL} is so small that only strong laser-light sources are sufficiently intense to visualize any THG at all.

Consequently, the influence of the disturbing wave on χ_{NL} may be ignored and henceforth χ_{NL} will be treated as a constant.

Introducing eq. (33) into the explicit solution (32), it is easy to see that we can immediately apply the following formula of Jacobi [29]

$$e^{i\,\xi\sin\theta} = \sum_{n=-\infty}^{+\infty} e^{i\,n\,\theta} J_n(\xi), \tag{35}$$

which can be used to generate the Bessel functions $J_n(\xi)$ of order n. Thus we obtain from (32):

$$A_1 = 3\varrho_0\left\{9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2} \cdot e^{i\alpha_1}$$
$$\cdot \sum_{m=-\infty}^{+\infty} e^{im(k^*x - \omega^*t)} J_m\left(\frac{k\,\delta z}{2 + 2\,\chi_0}\right) \tag{36}$$

and

$$A_3 = 2\varrho_0\left|\sin\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right|$$
$$\cdot\left\{9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot e^{i\alpha_3} \sum_{n=-\infty}^{+\infty} e^{in(k^*x - \omega^*t)} J_n\left(\frac{3\,k\,\delta z}{2 + 2\,\chi_0}\right). \tag{37}$$

Such a manipulation of the solution (32) is possible just because we are working in the Raman-Nath limit.

The interpretation of the diffraction spectrum can be given when we substitute eqs. (36) and (37) into the electric field (10):

$$E_0(x, z, t) = \sum_{m=-\infty}^{+\infty} \tfrac{3}{2}\varrho_0\left\{9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot(-1)^m J_m\left(\frac{k\,\delta z}{2 + 2\,\chi_0}\right) \exp[i\{kz - mk^*x - (\omega - m\,\omega^*)t + \alpha_1(z)\}]$$
$$+ \sum_{n=-\infty}^{+\infty} \varrho_0\left|\sin\left(\frac{9\,\varrho_0^2\,k\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right|\left\{9 + 4\sin^2\left(\frac{9\,\varrho_0^2\,k\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot(-1)^n J_n\left(\frac{3\,k\,\delta z}{2 + 2\,\chi_0}\right) \exp[i\{3\,kz - nk^*x - (3\,\omega - n\,\omega^*)t + \alpha_3(z)\}] + \text{c.c.} \tag{38}$$

or

$$E_0(x, z, t) = \sum_{m=-\infty}^{+\infty} 3\varrho_0\left\{9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot(-1)^m J_m\left(\frac{k\,\delta z}{2 + 2\,\chi_0}\right) \cos[kz - mk^*x - (\omega - m\,\omega^*)t + \alpha_1(z)]$$
$$+ \sum_{n=-\infty}^{+\infty} 2\varrho_0\left|\sin\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right|\left\{9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot(-1)^n J_n\left(\frac{3\,k\,\delta z}{2 + 2\,\chi_0}\right) \cos[3\,kz - nk^*x - (3\,\omega - n\,\omega^*)t + \alpha_3(z)]. \tag{39}$$

It becomes obvious that the ultrasonic liquid column acts as a diffraction grating for the incident (laser) light wave. The original plane wave is split into a spectrum of plane sub-waves of two types:

4.1. Ordinary diffraction lines (of order m, $m \in Z$)

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

$$\Delta\omega_m = -m\,\omega^* \tag{40}$$

and a deflection angle θ_m with the positive z-axis, given by

$$\theta_m = -m\,\frac{k^*}{k}. \tag{41}$$

Their amplitudes are defined by

$$\varphi_m^{(1)} = 3\varrho_0\left\{9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{NL}}{16 + 16\,\chi_0}z\right)\right\}^{-1/2}$$
$$\cdot(-1)^m J_m\left(\frac{k\,\delta z}{2 + 2\,\chi_0}\right). \tag{42}$$

Furthermore, these diffraction lines have a z-dependent phase-shift α_1, which is not important for the calculation of the intensities but could be used to obtain a further refinement in the determination of the deflection angles θ_m.

4.2. Intermediate diffraction lines (of order $n/3$, $n \in Z$)

Such lines do not occur in the diffraction pattern for an ordinary light beam, because they are due to THG in the acousto-optical interaction region. The frequency-shifts of these lines are given by

$$\Delta\omega_{n/3} = -\frac{n}{3}\,\omega^*, \tag{43}$$

with deflections determined by

$$\theta_{n/3} = -\frac{n\,k^*}{3\,k} \tag{44}$$

and amplitudes

$$\varphi_{n/3}^{(3)} = 2\,\varrho_0 \left| \sin\left(\frac{9\,k\,\varrho_0^2\,\chi_{\mathrm{NL}}}{16 + 16\,\chi_0} z \right) \right|$$

$$\cdot \left\{ 9 + 4\sin^2\left(\frac{9\,k\,\varrho_0^2\,\chi_{\mathrm{NL}}}{16 + 16\,\chi_0} z \right) \right\}^{-1/2}$$

$$\cdot (-1)^n J_n\left(\frac{3\,k\,\delta z}{2 + 2\,\chi_0} \right). \tag{45}$$

A similar remark could be made about the z-dependent phase-shift α_3. For $n/3 = m$, these intermediate lines have the same directions as the ordinary lines, and hence give a contribution to the intensities of these lines as well. With now the complete definition of the intensities of ordinary and intermediate lines given by

$$I_n = \varphi_n^{(1)}\overline{\varphi}_n^{(1)} + \varphi_n^{(3)}\overline{\varphi}_n^{(3)} \quad (n \in Z), \tag{46}$$
$$I_{n/3} = \varphi_{n/3}^{(3)}\overline{\varphi}_{n/3}^{(3)} \quad (n \in Z, n/3 \notin Z),$$

we finally obtain

$$I_n = \varrho_0^2 \frac{9\,J_n^2\left(\dfrac{k\,\delta z}{2 + 2\,\chi_0}\right) + 4\,J_{3n}^2\left(\dfrac{3\,k\,\delta z}{2 + 2\,\chi_0}\right)\sin^2\left(\dfrac{9\,k\,\varrho_0^2\,\chi_{\mathrm{NL}}}{16 + 16\,\chi_0}z\right)}{9 + 4\sin^2\left(\dfrac{9\,k\,\varrho_0^2\,\chi_{\mathrm{NL}}}{16 + 16\,\chi_0}z\right)}, \quad (n \in Z)$$

$$I_{n/3} = \frac{4\,\varrho_0^2\,J_n^2\left(\dfrac{3\,k\,\delta z}{2 + 2\,\chi_0}\right)\sin^2\left(\dfrac{9\,k\,\varrho_0^2\,\chi_{\mathrm{NL}}}{16 + 16\,\chi_0}z\right)}{9 + 4\sin^2\left(\dfrac{9\,k\,\varrho_0^2\,\chi_{\mathrm{NL}}}{16 + 16\,\chi_0}z\right)} \quad (n \in Z, n/3 \notin Z), \tag{47}$$

In the following paragraph we will discuss these results and compare them to the approximate calculations obtained in other papers.

5. Comparison with earlier treatments

The diffraction of intense laser-light by an ultrasonic wave in a liquid has been extensively studied by Sliwinski and his collaborators [14], [15], [21], [30]. They reported several experimental results together with some theoretical attempts at an explanation. Jozefowska [31] started from the assumption that the variation of the refractive index, due to THG, is proportional to the amplitude of the electric field squared. The nonlinear wave equation thus obtained was solved by successive approximations. The intensities of a few central diffraction lines were computed, in the form of complicated linear combinations of products of Bessel functions [21], [30]. A more practical expression for the intensity of a diffraction line of any order was given by Mertens and Leroy [22], who recalculated the results of Jozefowska, but using the nonlinear relative permittivity of the medium instead of the nonlinear refractive index. The electric field was developed in a power series in the small susceptibility χ_{NL} (in our notation) and then the generating function method was used to solve the successive wave equations of the perturbation scheme. A clear advantage of the generating function method is that it gives the intensities of the diffraction lines in closed form.

If we translate the results of Mertens and Leroy [22] in our notation, the intensities of the ordinary and the intermediate diffraction lines are given by

$$I_n = \frac{\varrho_0^2}{1 + r^2}\left\{ J_n^2\left(\frac{k\,\delta z}{2 + 2\,\chi_0} \right) + \left[r^2 + \frac{9\,k^2\,\varrho_0^4\,\chi_{\mathrm{NL}}^2\,r^2}{64(1 + \chi_0)^2(1 + r^2)^2} \right] J_{3n}^2\left(\frac{3\,k\,\delta z}{2 + 2\,\chi_0} \right) \right\} \quad (n \in Z),$$

$$I_{n/3} = \frac{\varrho_0^2}{1 + r^2}\left[r^2 + \frac{9\,k^2\,\varrho_0^4\,\chi_{\mathrm{NL}}^2\,z^2}{64(1 + \chi_0)^2(1 + r^2)^2} \right] J_n^2\left(\frac{3\,k\,\delta z}{2 + 2\,\chi_0} \right) \quad (n \in Z, n/3 \notin Z), \tag{48}$$

where r is the ratio of the real amplitudes of the third harmonic and the fundamental initially. In our treatment, r vanishes as we take the THG to occur only inside the perturbed liquid. The total intensity of the diffracted beam, as computed from eq. (48), is greater than ϱ_0^2, however. This is obviously the consequence of the fact that, in the treatment of Mertens and Leroy, there is no feedback during THG on the amplitude of the fundamental. This means that the fundamental is held at a given initial intensity and then acts as a pump wave for the third harmonic. In this picture, THG requires more energy, and hence a larger total intensity. In the present approach we have derived a set of coupled equations for the amplitudes of the fundamental and the third harmonic, with coupling both ways. This means that whatever energy the third harmonic gains is lost by the fundamental and vice versa, in such a way that the total energy remains constant. However, it should be emphasized that only a partial conversion of the energy from the fundamental to the third harmonic is possible. This point is discussed more in detail by Verheest [32] for THG in general media. As can now be expected, regarding the energy changes of both waves, the total intensity of the diffraction spectrum,

$$\sum_{n=-\infty}^{+\infty} I_n + \sum_{\substack{n=-\infty \\ n/3 \notin Z}}^{+\infty} I_{n/3} = \varrho_0^2 = A_0 \bar{A}_0 \qquad (49)$$

(to be calculated from eq. (47)), is exactly equal to the intensity of the incident light beam.

On the other hand, if we develop the expressions for the intensities (47) in a power series in χ_{NL}, up to terms in χ_{NL}^2, we find

$$I_n = \varrho_0^2 \left\{ \left(1 - \frac{9\,k^2\,\varrho_0^4\,\chi_{NL}^2\,z^2}{64\,(1+\chi_0)^2} \right) J_n^2 \left(\frac{k\,\delta z}{2+2\chi_0} \right) \right.$$
$$\left. + \frac{9\,k^2\,\varrho_0^4\,\chi_{NL}^2\,z^2}{64\,(1+\chi_0)^2} J_{3n}^2 \left(\frac{3\,k\,\delta z}{2+2\chi_0} \right) \right\} \quad (n \in Z),$$
$$I_{n/3} = \frac{9\,k^2\,\varrho_0^6\,\chi_{NL}^2\,z^2}{64\,(1+\chi_0)^2} J_n^2 \left(\frac{3\,k\,\delta z}{2+2\chi_0} \right) \, (n \in Z, n/3 \notin Z).$$
$$(49)$$

If we compare this with eq. (48), we see that the coefficient of $J_n^2 [k\,\delta z/(2+2\chi_0)]$ in the first equation is no longer 1, but diminishes with z, in order to compensate for THG. Otherwise it must be remarked that the intensities of the intermediate lines and the supplementary terms in the intensities of the ordinary lines (coefficients of $J_{3n}^2 [3\,k\,\delta z/(2+2\chi_0)]$) are correct up to terms in χ_{NL}^2. The total intensity, as calculated from eq. (49) is again ϱ_0^2

exactly, so that this property holds even after approximating eq. (47) by eq. (49).

References

[1] Raman, C. V. and Nagendra Nath, N. S., Proc. Indian Acad. Sci. **2 A** [1935], 406; **2 A** [1935], 413; **3 A** [1936], 75; **3 A** [1936], 119; **3 A** [1936], 459.
[2] Kuliasko, F., Mertens, R., and Leroy, O., Proc. Indian Acad. Sci. **68 A** [1968], 295.
[3] Mertens, R. and Kuliasko, F., Proc. Indian Acad. Sci. **68 A** [1968], 303.
[4] Born, M. and Wolf, E., Principles of optics, 5th ed. Pergamon Press, New York 1975, chapter 12.
[5] Berry, M. V., The diffraction of light by ultrasound. Academic Press, New York 1966.
[6] Ahmed, M. and Wade, G., Proc. IEEE **67** [1979], 587.
[7] Damon, R. W., Maloney, W. T., and McMahon, D. H., In: Physical acoustics: principles and methods, vol. 7 (ed. by M. P. Mason and R. N. Thurston). Academic Press, New York 1970, chapter 5.
[8] Guljaev, Y. V., Proklov, V. V., and Shkerdin, G. N., Soviet Phys. Usp. **21** [1978], 29.
[9] Imai, Y. and Ohtsuka, Y., Opt. Commun. **27** [1978], 18.
[10] Ohtsuka, Y., Opt. Commun. **17** [1976], 234; **17** [1976], 238.
[11] See for references: Kock, W. E., In: Physical acoustics: principles and methods, vol. 10 (ed. by M. P. Mason and R. N. Thurston). Academic Press, New York 1973, chapter 5.
[12] Lambert, L. B., In: Modern radar: analysis, evaluation and system design (ed. by R. S. Berkowitz). Wiley, New York 1965, chapter 3.
[13] Fukuda, S., Shiosaki, T., and Kawabata, A., J. Appl. Phys. **50** [1979], 3899.
[14] Kosmol, M., Acta Phys. Polon. **A 50** [1976], 375.
[15] Kosmol, M. and Sliwinski, A., Arch. Acoust. **4** [1979], 117.
[16] Bloembergen, N., Nonlinear optics. W. A. Benjamin, Reading, Massachusetts 1965.
[17] Franken, P. A., and Ward, J. F., Rev. Mod. Phys. **35** [1963], 85.
[18] Nelson, D. F., Electric, optic and acoustic interactions in dielectrics. Wiley, New York 1979, chapter 13.
[19] Terhune, R. W., Maker, P. D., and Savage, C. M., Phys. Rev. Letters **8** [1962], 404.
[20] Goldberg, L. S. and Schuur, J. M., Appl. Phys. Lett. **14** [1969], 306.
[21] Jozefowska, J., Kosmol, M., and Sliwinski, A., J. Phys. C6 supp. XI—XII **33** [1972], 239.
[22] Mertens, R. and Leroy, O., Proc. Ultrasonics Intern. [1977!], 416.
[23] Armstrong, J. A., Bloembergen, N., Ducuing, J., and Pershan, P. S., Phys. Rev. **127** [1962], 1918.
[24] Mertens, R., Hereman, W., and Verheest, F., Conf. Proc. 1st Spring School on Acousto-optics, Gdansk 1980, p. 33.

[25] Hereman, W. and Mertens, R., Wave Motion 1 [1979], 287.

[26] Nayfeh, A. H., Perturbation methods. Wiley, New York 1973, chapter 6.

[27] Moharam, M. G. and Young, L., Appl. Optics 17 [1978], 1757.

[28] Moharam, M. G., Gaylord, T. K., and Magnusson, R., Opt. Commun. 32 [1980], 14; 32 [1980], 19.

[29] Watson, G. N., A treatise on the theory of Bessel functions, 2nd ed. Cambridge University Press, Cambridge 1966, p. 22.

[30] Jozefowska, J. and Sliwinski, A., Proc. 7th Int. Cong. Acoust., Budapest, 21, U5 [1971], 497.

[31] Jozefowska, J., Thesis, A. Mickiewicz University, Poznan 1973.

[32] Verheest, F., Wave Motion [1981], in print.