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CONTRIBUTION TO THE THEORETICAL STUDY
OF THE DIFFRACTION OF ORDINARY
AND LASER LIGHT BY AN ULTRASONIC WAVE
IN A LIQUID

BY

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INTRODUCTION

The prediction that a sound field in a fluid or solid behaves as a volumetric optical phase grating came from Brillouin already in 1922. That grating behaviour manifests itself by redistributing the energy of the incoming light beam into scattered beams that correspond to the grating spectral orders. Besides the spatial distribution, the frequency of the scattered light is up- or down shifted by a multiple of the sound frequency (Doppler shift).

The diffraction of light by sound is simple to explain qualitatively. Exciting an acoustic compression wave in an acoustic-wave-supporting material creates a periodic strain pattern with spacing equal to the acoustic wave length. The variation of the index of refraction, caused by its internal strain (elasto-optic effect), is proportional to the magnitude of the sound wave and of course also dependent on the elasto-optic characteristics of the particular medium. Compared to the case without sound, the phase velocity of the light is larger in layers of decreased sound pressure and smaller in zones of increased density. Hence, the various light rays emerge with different phase angles, thus producing spectral orders which contain both amplitude and phase information about the acoustic wave.

One had to wait till 1932 for an experimental verification of the acousto-optic (in short AO) diffraction effect by Debye and Sears in the USA and by Lucas and Biquard in France.

During the past fifty years a large number of papers has been devoted to theoretical as well as to experimental aspects of this interesting phenomenon. The history of the theoretical analysis of light-sound interaction, however, has been described so often that we will not repeat it here ; instead, we refer the interested reader to the excellent reviews listed in earlier work [9, 12, 24].

Since the mid-sixties, there is a revived interest in the study of the AO diffraction, mainly for 3 reasons :

- (i) The development of coherent light sources (lasers), the advances in high-frequency acoustic techniques and the discovery of several superior AO materials have lead to new devices based on the old Debye-Sears or Lucas-Biquard effect. Such AO devices often make use of transparent solids, and include modulators, deflectors, signal correlators, tunable filters, spectrum analyzers, switchers, etc... They are used in a variety of new commercial and military applications involving RF signal processing, optical communication, ultrasonic imaging and nondestructive testing, to name only a few.
- (ii) From a theoretical point of view, the mathematical approaches successfully exploited in the treatment of AO diffraction, are equally applicable to many

other similar processes, such as diffraction of X-rays by crystals, scattering of light by hologram gratings, optical wave guiding in thin films and fibers (integrated optics) and various parametric effects in microwave technology. Last but not least, the development of holography has strongly stimulated the search of 3D diffraction gratings either holographically or acoustically induced.

- (iii) Since its breakthrough at the end of the sixties, the study of AO interaction with surface acoustic waves (SAW) has advanced to the level where a number of SAW devices can be built for signal processing applications and optical communication technology. Profound analyses reveal a high degree of isomorphism between the theories of diffraction of light by bulk ultrasonic waves and SAW. The extensive theoretical and experimental research of light-sound interaction in piezo-electric crystals, ferroelectric materials and anisotropic solids, has become a new discipline, nowadays commonly called *acousto-optics*.

In this paper two main sections may be distinguished. In the first one, we focus on the phenomenon of AO diffraction of intense laser light in a liquid. This problem is essentially *nonlinear* since for strong laser light the intensity of the applied electric field is so high that the nonlinear terms in the polarization are no longer negligible. The presence of cubic electric-field terms in the wave equation gives rise to third harmonic generation (THG). Besides the AO diffraction of the fundamental light wave, there will be a similar effect on its third harmonic. Phenomenologically, this results in the appearance of supplementary lines in the diffraction pattern. Our theoretical treatment of this complicated problem is based on a consistent combination of Bloembergen's theory of THG [1, 3, 27] and the generalized Raman-Nath (henceforth RN) theory of AO diffraction (for refs. see e.g. [7, 9, 24]).

In the second part, we will examine the AO diffraction of ordinary light or laser light of low intensity in an isotropic medium. This problem may be considered as a special case of the one mentioned above, since it requires the integration of a scalar wave equation with only *linear* terms referring to the diffracted electric field. Of particular interest is our alternative reformulation of the generalized RN theory, that lends itself to a straightforward derivation of well-known results for Raman-Nath and Bragg diffraction regimes. Our approach, known as the *modified generating function method* (MGFM), indeed, is applicable to a large class of AO interactions. To avoid unnecessary complication, however, we will not fully discuss the results obtainable from the most general analysis with the MGFM. Instead, we deal with the solutions for some idealized cases, giving interpretable and practical-manageable formulae.

NOMENCLATURE

λ_0, k_0, c	wave length, wave number, speed of light (in vacuum)
ϵ_0, μ_0	permittivity, magnetic permeability of vacuum
$\lambda, k, \nu, \omega, u$	wave length, wave number, frequency, angular frequency, speed of <i>light</i> (in medium)
Λ, K, f, Ω, V	wave length, wave number, frequency, angular frequency, speed of <i>sound</i> (in medium)
n_0, ϵ_{rL}	refractive index, linear relative permittivity (undisturbed medium)
ϵ_1	maximum variation of the linear relative permittivity (disturbed medium)
χ_{NL}	cubic relative susceptibility of the nonlinear medium
E_0	amplitude of the (incident) electric field (disturbed medium)
E	amplitude of the (scattered) electric field (disturbed medium)
L	acousto-optic interaction length along the z-axis
x	direction of sound propagation
z	direction of light propagation (at normal incidence)
t	time
φ	angle of incidence of light (in medium)
n	order of diffraction
$\varphi_{BR}^{(n)}$	Bragg angle of order n (in medium)
θ_n	deflection angle of order n (in medium)
ψ_n	complex amplitude of diffracted light wave of order n
φ_n	real amplitude of diffracted light wave of order n
I_n	intensity of spectral line of order n
$J_n(z)$	Besselfunction of the first kind and order n
ν	Raman-Nath parameter or peak phase deviation
ρ	regime parameter
Q	Klein-Cook parameter
\rightarrow	vector symbol
$^*, \text{c.c.}$	complex conjugate
AO	acousto-optic
RN	Raman-Nath
MGFM	modified generating function method
THG	third harmonic generation

I. Acousto-optic diffraction of intense laser light in an isotropic medium (nonlinear case)

1. Basic principles

We consider the configuration as shown in fig. 1 and neglect any sound field diffraction due to the bounded column. Let the bulk acoustic wave, with wave length Λ , wave number $K = 2\pi/\Lambda$ and frequency f , be travelling along the x -direction in the isotropic medium (e.g. a liquid). The monochromatic light beam of wave length λ , wave number $k = 2\pi/\lambda$ (both in the medium) and frequency ν is propagating in the (x, z) -plane at an angle φ with respect to the z -axis. The angle φ , also measured in the liquid, is called the angle of incidence.

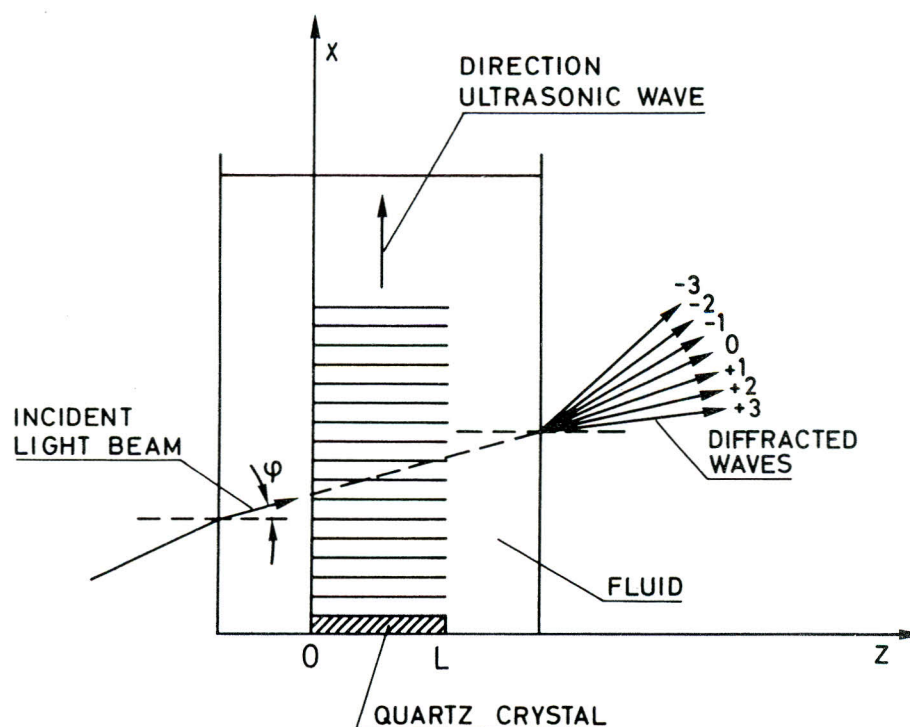


Fig. 1. Geometry of AO diffraction.

As long as one uses sound waves in a lower range of ultrasonic frequencies (e.g. $f \approx 10$ MHz), the angle φ is kept small. For sufficiently small width L of the sound column, the diffracted light appears on both sides of the primary beam in the form of equally spaced lines. This case of multiple-order scattering is commonly called *Raman-Nath diffraction*, shown in fig. 2a. In the range of sound frequencies above e.g.

400 MHz, the angle φ is mostly adjusted to the so-called Bragg angle φ_{BR} ($1^\circ \rightarrow 10^\circ$), which we will define later. After passing through a sound field of sufficiently large width L , light incident at the Bragg angle is — so to speak — partially reflected on the moving sound wave fronts [24], and thus strongly diffracted in a single spot. This case, which is extremely important in applications, is called single-order diffraction or *Bragg diffraction*, schematically shown in fig. 2b.

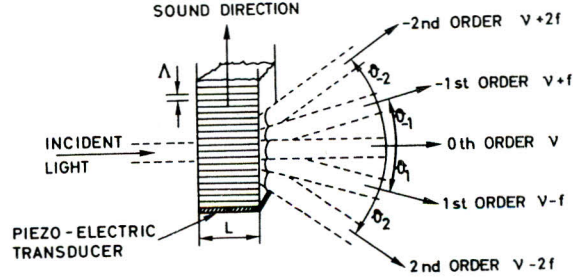


Fig. 2a. Multi-order Raman-Nath diffraction.

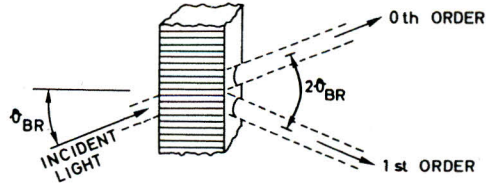


Fig. 2b. Single-order Bragg diffraction.

Since we have to describe the propagation of an e.m. wave through a dielectric, nonconductive medium, let us start from Maxwell's equations without source terms,

$$\begin{aligned}\vec{\nabla} \times \vec{E} + \frac{\partial \vec{B}}{\partial t} &= \vec{0}, \quad \vec{\nabla} \cdot \vec{B} = 0, \\ \vec{\nabla} \times \vec{H} - \frac{\partial \vec{D}}{\partial t} &= \vec{0}, \quad \vec{\nabla} \cdot \vec{D} = 0.\end{aligned}\tag{1}$$

Eliminating the magnetic field \vec{H} and the magnetic induction \vec{B} ($= \mu_0 \vec{H}$), we readily obtain

$$\nabla^2 \vec{E} = \mu_0 \frac{\partial^2 \vec{D}}{\partial t^2} + \vec{\nabla} \vec{\nabla} \cdot \vec{E}, \quad \vec{\nabla} \cdot \vec{D} = 0.\tag{2}$$

The magnetic permeability μ_0 is linked to the permittivity ϵ_0 and the speed of light c (all in vacuum) by $\epsilon_0 \mu_0 c^2 = 1$. In order that the first equation in (2) be a true

wave equation, a relation between the electric displacement \vec{D} and the electric field \vec{E} must be given. In general dielectric materials

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P}(\vec{E}) = \epsilon_0 \vec{E} + \vec{P}_L(\vec{E}) + \vec{P}_{NL}(\vec{E}), \quad (3)$$

where the polarization \vec{P} is split into its linear and nonlinear parts [4, 5].

The linear polarization vector may be written [5] as

$$\vec{P}_L(\vec{E}) = \chi_L^{(1)} : \vec{E}, \quad (4)$$

wherein $\chi_L^{(1)}$ represents the ordinary linear susceptibility tensor of rank 2 (and order 1).

The nonlinear polarization may be expanded [4, 5] in a series of powers of the electric field

$$\vec{P}_{NL}(\vec{E}) = \chi_{NL}^{(2)} : \vec{E}\vec{E} + \chi_{NL}^{(3)} : \vec{E}\vec{E}\vec{E} + \dots, \quad (5)$$

where $:$ and $:$ are the inner products symbols.

The coefficients $\chi_{NL}^{(2)}$, $\chi_{NL}^{(3)}$ stand for the quadratic and cubic susceptibility tensors, respectively of rank 3 and 4 (orders 2 and 3).

In general dielectric materials the components of the susceptibility tensors act as frequency-dependent operators upon the components of the electric field. Following Bloembergen's easiest model [1, 3], we only consider frequency-independent quantities, no longer taking into account their operator character. Furthermore, the optical susceptibilities being very small, we cut off the power series (5) after the second term.

Needless to say that the symmetry properties of the AO material will reflect themselves in symmetry relations between the tensor components. So, in order that the tensor $\chi_{NL}^{(2)}$ will not vanish, the material must lack a center of inversion. Lastly, the tensor components will be time and space dependent if the medium is inhomogeneous or becomes so due to e.g. an ultrasonic disturbance. Although, in our case, the ultrasonic wave will propagate in one particular direction, this will not harm the *optical isotropy* of the medium.

Now, knowing all this, one easily can show [7, 8, 9] that for an isotropy medium

$$\begin{aligned} \chi_{L,ij}^{(1)} &= \tilde{\chi}_L \delta_{ij}, \\ \chi_{NL,ijk}^{(2)} &= 0, \\ \chi_{NL,ijkl}^{(3)} &= A \delta_{ij} \delta_{kl} + B \delta_{ik} \delta_{jl} + C \delta_{il} \delta_{jk}, \quad (i, j, k, l = 1, 2, 3), \end{aligned} \quad (6)$$

where $\tilde{\chi}_L$, A, B and C are scalar functions of space and time. Remark that from the 81 tensor components $\chi_{NL,ijkl}^{(3)}$ only 21 are not identically zero. Among these 21 non-vanishing components only 3 are independent, e.g. $\chi_{NL,1122}^{(3)} = A$; $\chi_{NL,1212}^{(3)} = B$ and $\chi_{NL,1221}^{(3)} = C$.

Taking into account (6), the i-th component of the total polarization, up to terms in E^3 , becomes

$$\begin{aligned}
P_i &= P_{L,i} + P_{NL,i} \\
&= \tilde{\chi}_L \delta_{ij} E_j + (A \delta_{ij} \delta_{kl} + B \delta_{ik} \delta_{jl} + C \delta_{il} \delta_{jk}) E_j E_k E_l \\
&= \tilde{\chi}_L E_i + A E_k E_k E_i + B E_j E_j E_i + C E_j E_j E_i \\
&= [\tilde{\chi}_L + (A + B + C) E^2] E_i, \quad (i = 1, 2, 3),
\end{aligned} \tag{7}$$

where the convention of summing for repeated indices is adopted. Hence, the constituent relation for the optical isotropic medium reads

$$\vec{P} = \epsilon_0 (\chi_L + \chi_{NL} E^2) \vec{E}, \tag{8}$$

introducing the relative susceptibilities $\chi_L = \tilde{\chi}_L / \epsilon_0$ and $\chi_{NL} = \tilde{\chi}_{NL} / \epsilon_0 = (A + B + C) / \epsilon_0$.

Finally, since $\chi_{NL} \ll \chi_L$ the influence of the ultrasonic field on χ_{NL} is a too small effect to take into account. Thus, χ_{NL} is treated as a scalar constant from now on, but due to the presence of a simple progressive ultrasonic wave, we have

$$\chi_L(x, t) = \chi_0 + \epsilon_1 \sin(\Omega t - Kx). \tag{9}$$

χ_0 is the constant value of χ_L for the undisturbed medium, ϵ_1 is the maximum variation of χ_L . Regarding (3), (8) and (9), \vec{D} can be written as

$$\vec{D} = \epsilon_0 [1 + \chi_0 + \epsilon_1 \sin(\Omega t - Kx) + \chi_{NL} E^2] \vec{E} \tag{10}$$

or briefly $\vec{D} = \epsilon(x, t) \vec{E}$, with

$$\epsilon(x, t) = \epsilon_0 [\epsilon_{rL} + \epsilon_1 \sin(\Omega t - Kx) + \chi_{NL} E^2], \tag{11}$$

where $\epsilon_{rL} = 1 + \chi_0$, denotes the constant linear permittivity of the medium without disturbing sound wave.

Expression (11), taken as a starting point by many authors (cf. refs. in [13]), indicates that ϵ_1 indeed is the maximum variation of the linear part of the relative permittivity of the disturbed fluid.

Advoiding unnecessary complications, the sound field is supposed to pervade the whole of the liquid. The theoretical analysis of the problem where the ultrasonic field is preceded and followed by an undisturbed part of the fluid may be found elsewhere [9, 14].

2. Basic equations for AO interaction and THG

We now consider a strong laser light beam impinging orthogonally on the ultrasonic field. In that case $\varphi = 0$ and we speak of normal incidence of the light. Furthermore, we suppose the plane monochromatic light wave to be linearly polarized along the y-axis perpendicular to the (x, z)-plane. Then the incident electric field has the form

$$\vec{E}_0 = \left\{ \frac{1}{2} A_0 \exp i \varphi(z, t) + \text{c.c.} \right\} \vec{e}_y, \tag{12}$$

where A_0 is the constant complex amplitude, and where the phase

$$\phi(z, t) = \omega t - kz, \quad (13)$$

corresponding to light propagation in the z -direction, contains the angular frequency ω and the wave number k (in the medium); c.c. stands for complex conjugate.

Expressing that (12) is a solution of (2), without ultrasonic disturbance ($\epsilon_1 = 0$) and without nonlinear terms ($\chi_{NL} = 0$ in (10)), gives the linear dispersion law

$$\frac{\omega^2}{c^2} = k_0^2 = \frac{k^2}{1 + \chi_0} = \frac{k^2}{\sqrt{\epsilon_{TL}}}, \quad (14)$$

$k_0 = 2\pi/\lambda_0$ is the wave number of the light with wave length λ_0 in vacuum.

Whereas in an optical isotropic medium a change in the state of light polarization is excluded, the perturbed optical field can be represented by $\vec{E} = E(x, z, t) \vec{e}_y$. From $\vec{\nabla} \cdot \vec{D} = 0$ (in (2)), indeed follows that E must be independent of the y -coordinate.

Assuming that \vec{E} is linearly polarized, the vector wave equation in (2), simplifies to the scalar wave equation

$$\frac{\partial^2 E}{\partial x^2} + \frac{\partial^2 E}{\partial z^2} = \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \{ [\epsilon_{TL} + \epsilon_1 \sin(\Omega t - Kx)] E + \chi_{NL} E^3 \}, \quad (15)$$

after substitution of (10) and use of (14). The cubic term in (15) is responsible for the generation of the third harmonic (THG) in the optical field, that thus can be represented by

$$E(x, z, t) = \frac{1}{2} \Phi_1(x, z, t) \exp i \phi(z, t) + \frac{1}{2} \Phi_3(x, z, t) \exp 3i \phi(z, t) + \text{c.c.} \quad (16)$$

wherein Φ_1 and Φ_3 are the complex amplitudes of the fundamental light wave and its third harmonic respectively; for both perfect phase matching is assumed.

The analysis of the AO interaction relies on the basic assumption that the stratification of the medium as produced by the sound wave is stationary in space. This is a fair assumption since the velocity of sound is roughly 10^5 times smaller than the speed of light. Hence, one makes the slowly varying envelope approximation, dropping second order spatial derivatives with respect to z and all the time derivatives of the slowly varying amplitudes Φ_1 and Φ_3 .

With these simplifying assumptions, substitution of (16) into (15) results in two coupled equations

$$\begin{aligned} \frac{\partial^2 \Phi_1}{\partial x^2} - 2ik \frac{\partial \Phi_1}{\partial z} + k_0^2 \epsilon_1 [\sin(\Omega t - Kx)] \Phi_1 \\ = - \frac{3}{4} k_0^2 \chi_{NL} (\Phi_1^2 \Phi_1^* + 2\Phi_1 \Phi_3 \Phi_3^* + \Phi_1^* \Phi_3^2), \end{aligned}$$

$$\begin{aligned}
& \frac{\partial^2 \Phi_3}{\partial x^2} - 6ik \frac{\partial \Phi_3}{\partial z} + 9k_0^2 \epsilon_1 [\sin(\Omega t - Kx)] \Phi_3 \\
& = -\frac{9}{4} k_0^2 \chi_{NL} (\Phi_1^3 + 6\Phi_1 \Phi_1^* \Phi_3 + 3\Phi_3^2 \Phi_3^*),
\end{aligned} \tag{17}$$

wherein * stands for the complex conjugate. Remark that we have neglected terms referring to the generation of harmonics higher than the third, this to be consistent with the basic principles in Section 1.

Before transforming these mode-coupling equations, we consider the boundary conditions at $z = 0$. Supposing that there is no THG before the light enters the disturbed liquid column, the boundary conditions simply are

$$\Phi_1(x, 0, t) = A_0, \quad \Phi_3(x, 0, t) = 0. \tag{18}$$

We restrict ourselves to solutions of the system (17) which incorporate in an intrinsic way the periodicity properties of the ultrasonic waves [7]. Therefore we introduce the coordinate transformation $(x, z, t) \rightarrow (\xi, \zeta)$ defined by

$$\xi = \frac{1}{2} (Kx - \Omega t + \frac{3\pi}{2}), \tag{19}$$

$$\zeta = \frac{k_0 \epsilon_1 z}{2\sqrt{\epsilon_{rL}}}. \tag{20}$$

Denoting Φ_1 and Φ_3 in the new variables by Ψ_1 and Ψ_3 , (17) transforms into

$$\begin{aligned}
\frac{\partial \Psi_1}{\partial \zeta} - i(\cos 2\xi) \Psi_1 &= -\frac{i\rho}{8} \frac{\partial^2 \Psi_1}{\partial \xi^2} - i\gamma (\Psi_1^2 \Psi_1^* + 2\Psi_1 \Psi_3 \Psi_3^* + \Psi_1^{*2} \Psi_3), \\
\frac{\partial \Psi_3}{\partial \zeta} - 3i(\cos 2\xi) \Psi_3 &= -\frac{i\rho}{24} \frac{\partial^2 \Psi_3}{\partial \xi^2} - i\gamma (\Psi_1^3 + 6\Psi_1 \Psi_1^* \Psi_3 + 3\Psi_3^2 \Psi_3^*),
\end{aligned} \tag{21}$$

where the regime parameter

$$\rho = \frac{2K^2}{k_0^2 \epsilon_1} = \frac{2\lambda_0^2}{\Lambda^2 \epsilon_1}, \tag{22}$$

and the nonlinearity parameter

$$\gamma = \frac{3\chi_{NL}}{4\epsilon_1}, \tag{23}$$

have been introduced. The periodicity conditions, obtained from the mentioned physical argument, can now easily be summarized as

$$\Psi_1(\xi + \pi, \zeta) = \Psi_1(\xi, \zeta), \quad \Psi_3(\xi + \pi, \zeta) = \Psi_3(\xi, \zeta). \tag{24}$$

The functions $\Psi_1(\xi, \zeta)$ and $\Psi_3(\xi, \zeta)$ are called generating functions, a name which will be justified in Section 4 of this first part.

Concluding this Section, we want to stress that the derivation of the coupled partial differential equations (21) was essentially based on the idea that there are two

small effects in this AO diffraction problem, one being the weak acoustic scattering of the fundamental, the other the nonlinear generation of the third harmonic. A more rigorous analysis [7 App. B, 13] has shown that requiring these two effects to be of the same order of magnitude, (21) is still correct provided $\rho = 0$. This corresponds to the well-known Raman-Nath limit, which we will continue with in the next Section.

3. Exact solution in the Raman-Nath limit

Without entering into the details, we now show how the nonlinear coupled equations (21), with appropriate boundary and periodicity conditions, can be exactly solved for $\rho = 0$. Experimentally this situation can be achieved by using a strong ultrasonic wave (ϵ_1 reasonably large) of rather long wave length Λ . For nonzero values of ρ the integration of system (21) is still under investigation.

With the second order derivative terms dropped, the structure of the equations (21) is such that the ξ -dependent coefficients can be split off by the substitution

$$\begin{aligned}\Psi_1(\xi, \zeta) &= Z_1(\zeta) \exp(i\zeta \cos 2\xi), \\ \Psi_3(\xi, \zeta) &= Z_3(\zeta) \exp(3i\zeta \cos 2\xi).\end{aligned}\tag{25}$$

This yields

$$\begin{aligned}\frac{dZ_1}{d\zeta} + i\gamma(Z_1^2 Z_1^* + 2Z_1 Z_3 Z_3^* + Z_1^* Z_3) &= 0, \\ \frac{dZ_3}{d\zeta} + i\gamma(Z_1^3 + 6Z_1 Z_1^* Z_3 + 3Z_3^2 Z_3^*) &= 0.\end{aligned}\tag{26}$$

together with the boundary conditions

$$Z_1(0) = A_0, \quad Z_3(0) = 0.\tag{27}$$

For solving the equations (26) we will use Bloembergen's method [1, 3, 27], originally developed for the theoretical study of wave mixing and harmonic generation in dynamic nonlinear optics. Here, we only indicate the main steps of the technique, for more details the reader is referred to the pioneering paper of Armstrong *et al.* [1], which was corrected and reprinted in Bloembergen's book [3, pp. 170-192].

The basic idea is to split the coupled equations (26) into their real and imaginary parts by substituting

$$Z_1 = \rho_1(s) \exp(i\alpha_1(s)), \quad Z_3 = \rho_3(s) \exp(i\alpha_3(s)),\tag{28}$$

where $s = \gamma\zeta$. Thus, we obtain

$$\begin{aligned}\frac{d\rho_1}{ds} &= -\rho_1^2 \rho_3 \sin\psi, \\ \frac{d\rho_3}{ds} &= \rho_1^3 \sin\psi, \\ \frac{d\psi}{ds} &= \frac{\rho_1}{\rho_3} (\rho_1^2 - 3\rho_3^2) \cos\psi + 3(\rho_1^2 - \rho_3^2),\end{aligned}\tag{29}$$

with $\psi = 3\alpha_1 - \alpha_3$. Subject to the boundary conditions $\rho_1(0) = |A_0| = \rho_0$, $\rho_3(0) = 0$, $\alpha_1(0) = 0$ and $\alpha_3(0) = -\pi/2$; from (29) two first integrals may readily be found [9]:

$$\begin{aligned}\rho_1^2 + \rho_3^2 &= \rho_0^2, \\ \rho_1 \cos\psi + \frac{3}{2} \rho_3 &= 0.\end{aligned}\tag{30}$$

These invariants allow complete integration of the equations (29) by quadratures only. Indeed, introducing $u = \rho_1^2$, the system (29) can be reduced to only one ordinary differential equation

$$\frac{du}{ds} = \pm \sqrt{f(u)},\tag{31}$$

where $f(u) = u^2(\rho_0^2 - u)(13u - 9\rho_0^2)$ is only strictly positive between the two single roots $u = 9\rho_0^2/13$ and $u = \rho_0^2$. Hence, u will oscillate between these two values and consequently there will be a periodic (but partial) interchange of energy from the fundamental to the third harmonic wave and vice versa [33].

Remarkably, the integration of (31) does not involve any elliptic integral [6, p. 84], one simply obtains

$$u = \rho_1^2(s) = \frac{9\rho_0^2}{11 - 2\cos 3\rho_0^2 s}, \quad \rho_3^2(s) = \frac{2\rho_0^2(1 - \cos 3\rho_0^2 s)}{11 - 2\cos 3\rho_0^2 s}.\tag{32}$$

Using these results together with the invariants (30), allows us to determine the real phases α_1 and α_3 [9]. Since they do not effect the intensities, though they may be used to obtain further refinements in the determination of the deflection angles of the scattered light waves, we can safely ignore them here (if necessary see [7, 9]).

4. Diffraction spectrum

Even without knowing the explicit expressions of Ψ_1 and Ψ_3 for $\rho \neq 0$, one can draw far-reaching conclusions from their periodicity properties. Indeed, the conditions (24) allow the expansion of these functions into Fourier series

$$\begin{aligned}\Psi_1(\xi, \zeta) &= \sum_{n=-\infty}^{+\infty} \varphi_n^{(1)}(\zeta) i^n \exp(2in\xi) \exp[i\alpha_n^{(1)}(\zeta)], \\ \Psi_3(\xi, \zeta) &= \sum_{m=-\infty}^{+\infty} \varphi_m^{(3)}(\zeta) i^m \exp(2im\xi) \exp[i\alpha_m^{(3)}(\zeta)],\end{aligned}\tag{33}$$

with real amplitudes $\varphi_n^{(1)}$, $\varphi_m^{(3)}$ and real phases $\alpha_n^{(1)}$ and $\alpha_m^{(3)}$. Returning to the old variables via (19) and (20), and substituting these expansions into (16), results in

$$\begin{aligned}
E(x, z, t) = & \sum_{n=-\infty}^{+\infty} \frac{1}{2} \varphi_n^{(1)}(z) \exp[i(\omega - n\Omega)t] \exp[-i(kz - nKx - \alpha_n^{(1)}(z))] \\
& + \sum_{m=-\infty}^{+\infty} \frac{1}{2} \varphi_m^{(3)}(z) \exp[i(3\omega - m\Omega)t] \exp[-i(3kz - mKx - \alpha_m^{(3)}(z))] + \text{c.c.}
\end{aligned} \quad (34)$$

It becomes obvious that the incident light wave will be split into different plane subwaves, in other words the disturbed medium acts as an optical phase grating. To each integer n there corresponds a diffraction order which has a maximum intensity along the direction θ_n (with the z -axis) given by the familiar grating equation

$$\theta_n = -\frac{nK}{k} = -\frac{n\lambda}{\Lambda} = -\frac{n\lambda f}{V}, \quad (35)$$

wherein the phase angle correction $\alpha_n^{(1)}$ is neglected and where $V = \Lambda f$ denotes the sound velocity in the medium. It is also evident from the first series in (34) that the n -th order light beam is frequency (up or down) shifted by an amount

$$\Delta v_n = -nf. \quad (36)$$

In the second series of (34) the term corresponding the $m = 3n$ also gives a contribution to the n -th order diffraction line. Hence, the intensity of this spectral line will be

$$I_n(\zeta) = [\varphi_n^{(1)}(\zeta)]^2 + [\varphi_{3n}^{(3)}(\zeta)]^2. \quad (37)$$

Such lines (of order n , n integer) are called *ordinary diffraction lines*, since they are found at the same places as if the medium were linear ($\chi_{NL} = 0$, $\Psi_3 \equiv 0$). As we will see in Part II this leads to diffraction lines still with the characteristics (35) and (36) but with other intensities.

Regarding the second series in (34), for $m \neq 3n$ ($n \in \mathbb{Z}$), there is an additional set of diffracted subwaves, approximately at angles

$$\theta_{m/3} = -\frac{m}{3} \frac{K}{k} = -\frac{m}{3} \frac{\lambda}{\Lambda} = -\frac{m}{3} \frac{\lambda f}{V}, \quad (38)$$

with Doppler shifts

$$\Delta v_{m/3} = -\frac{m}{3} f, \quad (39)$$

and intensities

$$I_{m/3}(\zeta) = [\varphi_m^{(3)}(\zeta)]^2, \quad m \in \mathbb{Z}, \quad m/3 \notin \mathbb{Z}. \quad (40)$$

These so-called *intermediate diffraction lines* (of order $m/3$, $m/3$ not integer) only appear for sufficiently strong laser light, when the nonlinearity manifests itself. In other words, these intermediate lines, which have been observed in experiments of Sliwinski and his collaborators [18, 19], are exclusively due to THG in the medium.

In passing, note that according to the simple formulae (35) or (38), by keeping the incident light beam fixed, but varying the sound frequency f , the diffracted light beams may be deflected continuously.

From the above analysis it must be clear why Ψ_1 and Ψ_3 are called generating functions. If one succeeds to find their explicit forms, one simply has to expand these into Fourier series of type (33). Next, one can read off the amplitudes and finally calculate the intensities with (37) and (40).

Let us exemplify this procedure by calculating the intensities of the two types of lines in the diffraction pattern in the RN limit.

It follows from the formulae in the preceding section that, for $\rho = 0$,

$$\begin{aligned}\Psi_1(\xi, \zeta) &= 3\rho_0 [9 + 4 \sin^2(\frac{3}{2}\rho_0^2\gamma\zeta)]^{-1/2} \exp(i\alpha_1(\zeta)) \exp(i\cos 2\xi), \\ \Psi_3(\xi, \zeta) &= 2\rho_0 |\sin(\frac{3}{2}\rho_0^2\gamma\zeta)| [9 + 4\sin^2(\frac{3}{2}\rho_0^2\gamma\zeta)]^{-1/2} \\ &\quad \times \exp(i\alpha_3(\zeta)) \exp(3i\cos 2\xi).\end{aligned}\quad (41)$$

Applying Jacobi's generating formula for the Bessel functions $J_n(\zeta)$ of integer order [34, p. 22], i.e.

$$\exp(i\zeta\cos 2\xi) = \sum_{n=-\infty}^{+\infty} J_n(\zeta) i^n \exp(2in\xi); \quad (42)$$

and comparing the thus obtained explicit expressions of Ψ_1 and Ψ_3 with their series expansions as in (33), one gets

$$\begin{aligned}\varphi_n^{(1)}(\zeta) &= 3\rho_0 [9 + 4\sin^2(\frac{3}{2}\rho_0^2\gamma\zeta)]^{-1/2} J_n(\zeta), \\ \varphi_m^{(3)}(\zeta) &= 2\rho_0 |\sin(\frac{3}{2}\rho_0^2\gamma\zeta)| [9 + 4\sin^2(\frac{3}{2}\rho_0^2\gamma\zeta)]^{-1/2} J_m(3\zeta).\end{aligned}\quad (43)$$

Now, the closed form intensities, evaluated at the plane $z = L$, follow readily from (37) and (40):

$$\begin{aligned}I_n(v) &= \frac{\rho_0^2 [9J_n^2(v) + 4J_{3n}^2(3v) \sin^2(\frac{3}{2}\rho_0^2\gamma v)]}{9 + 4\sin^2(\frac{3}{2}\rho_0^2\gamma v)}, \quad n \in \mathbb{Z}, \\ I_{m/3}(v) &= \frac{4\rho_0^2 J_m^2(3v) \sin^2(\frac{3}{2}\rho_0^2\gamma v)}{9 + 4\sin^2(\frac{3}{2}\rho_0^2\gamma v)}, \quad m \in \mathbb{Z}, \quad m/3 \notin \mathbb{Z};\end{aligned}\quad (44)$$

where $v = \zeta L/z = k_0 \epsilon_1 L / 2 \sqrt{\epsilon_{rL}}$ is the so-called Raman-Nath parameter or peak phase deviation. Note that only the argument of the sine function depends on the nonlinearity parameter γ , defined in (23).

Using the Bessel function identities [34, p. 15 & p. 30], $J_{-n} = (-1)^n J_n$ and

$$J_0^2 + 2 \sum_{n=1}^{\infty} J_n^2 = 1, \quad (45)$$

one can immediately check that the total intensity of the spectrum equals the intensity ρ_0^2 of the incident laser light. Furthermore, with (44) one can verify that the spectrum is completely symmetric with respect to the zeroth order line, i.e. $I_{-n}(v) = I_n(v)$ and $I_{-m/3}(v) = I_{m/3}(v)$. This is always the case for normal incidence of the light as will be discussed in more detail in Part II.

II. Acousto-optic diffraction of ordinary light or weak laser light in a liquid (linear case)

1. The modified generating function method

We have already introduced the idea of a generating function in the Sections 2 and 4 of Part I. Here, we will present a detailed study of the AO diffraction problem in the absence of nonlinearities, i.e. when ordinary light or weak intensity laser light is used. Emphasis will be on the modified generating function method (MGFM), originally developed by Hereman for the theoretical investigation of AO diffraction due to an amplitude-modulated ultrasonic wave [10], but giving full scope to its applicability when a simple progressive ultrasonic wave is used (cf. (9)).

To incorporate Bragg diffraction as well as Raman-Nath diffraction regimes (see Section 1 of Part I), we initially take an oblique angle of incidence φ , as depicted in fig. 1 (not excluding the limiting case $\varphi = 0$). Hence, the phase (13) of the incident light beam must be replaced by

$$\varphi(x, z, t) = \omega t - k(x \sin \varphi + z \cos \varphi). \quad (46)$$

In the absence of nonlinear terms ($\chi_{NL} = 0$), the wave equation (15) is still the starting point and the subsequent steps are still valid, although all the references to the third harmonic must be dropped ($\gamma = 0$, $\Psi_3 = 0$). It is straightforward to show [7, 9, 25] that the generating function $\Psi_1(\xi, \zeta) \equiv \Psi(\xi, \zeta)$ must be the solution of the partial differential equation

$$\frac{\partial \Psi}{\partial \zeta} - i(\cos 2\xi) \Psi = -\frac{1}{8} i \rho \frac{\partial^2 \Psi}{\partial \xi^2} - \frac{1}{2} a \sin \varphi \frac{\partial \Psi}{\partial \xi}, \quad (47)$$

where $a = 2K\xi_r/\varepsilon_1 k$, together with the appropriate boundary and periodicity conditions:

$$\Psi(\xi, 0) = 1, \quad (48)$$

$$\Psi(\xi + \pi, \zeta) = \Psi(\xi, \zeta). \quad (49)$$

For simplicity we have normalized the intensity of the incident light ($\rho_0 = 1$) and for notational convenience we have dropped the indices 1 and L. It is now possible to make the following statements:

(1) The AO diffraction is entirely formulated in terms of one — so far unknown — generating function Ψ , which turns out to be a powerful approach.

First, let us repeat that the Fourier expansion of Ψ , i.e.

$$\Psi(\xi, \zeta) = \sum_{n=-\infty}^{+\infty} \psi_n(\zeta) i^n \exp(2in\xi), \quad (50)$$

will generate the (complex) amplitudes $\psi_n(\zeta)$ of the diffracted light waves. Furthermore, rewriting (50) in the original variables (x, z, t), the formulae (35) (however with the deflection angle θ_n replaced by $\theta_n - \varphi$) and (36) readily follow. Hence, deflection and Doppler shift are comprised in the present formulation of the problem.

(2) One can prove that the problem is *well-posed*, this means that (47) with (48) and (49) has one and only one solution. An explicit proof of these theorems would carry us to far (cf. however [7, 9]).

(3) The famous system of Raman-Nath equations

$$2 \frac{d\psi_n}{d\zeta} - (\psi_{n-1} - \psi_{n+1}) = in(\rho n - 2a \sin\varphi) \psi_n, \quad (51)$$

for the complex amplitudes ψ_n ($n \in \mathbb{Z}$), satisfying the boundary conditions $\psi_n(0) = \delta_{n0}$ (δ_{n0} denotes the Kronecker delta), is equivalent to our problem (47) - (48) - (49). Indeed, substitution of the Fourier expansion (50) into (47) yields (51), while the boundary conditions for ψ_n follow from (48).

Consequently, there is a unique set of complex amplitudes satisfying the RN equations, explicitly solving the equations is something else !

(4) As an immediate result of almost trivial transformations and the existence of a *unique* solution, one can derive symmetry properties of the diffraction spectra. Let us give an easy example. Changing ξ into $-\xi$ into $\Psi(\xi, \zeta)$ and denoting $\Psi(\xi, \zeta)$ by $\Psi^+(\xi, \zeta)$ and $\Psi(-\xi, \zeta)$ by $\Psi^-(\xi, \zeta)$, from (47) and (50) readily follows that

$$\begin{aligned} \Psi^-(\xi, \zeta) &= \sum_{n=-\infty}^{+\infty} \psi_n(\zeta) i^n \exp(-2in\xi) \\ &= \sum_{n=-\infty}^{+\infty} (-1)^n \psi_{-n}(\zeta) i^n \exp(2in\xi) \end{aligned} \quad (52)$$

satisfies

$$\frac{\partial \Psi^-}{\partial \zeta} - i(\cos 2\xi) \Psi^- = -\frac{i\rho}{8} \frac{\partial^2 \Psi^-}{\partial \xi^2} + \frac{a}{2} \sin\varphi \frac{\partial \Psi^-}{\partial \xi} \quad (53)$$

and $\Psi^-(\xi, 0) = 1$, provided Ψ^+ is the unique solution of the original problem. Comparison of both problems, clearly reveals that for normal incidence of the light ($\varphi = 0$) $\Psi^+(\xi, \zeta) = \Psi^-(\xi, \zeta)$ and from (50) and (52), we thus obtain

$$\psi_{-n}(\zeta) = (-1)^n \psi_n(\zeta). \quad (54)$$

Regarding the definition of the intensity of order n , i.e.

$$I_n(\zeta) = \psi_n(\zeta) \psi_n^*(\zeta), \quad (55)$$

we then may conclude that

$$I_{-n}(\zeta) = I_n(\zeta). \quad (56)$$

In words : for normal incidence the spectrum is symmetric with respect to the zeroth order line (central line). For oblique incidence of the light ($\varphi \neq 0$) and for arbitrary nonzero values of ρ , (47) and (53) are never identical, hence, the spectrum shows asymmetry. However for $\rho = 0$ (RN limit) one can show [9] that the spectrum is again symmetric, even for oblique incidence of light. Other symmetry properties and the reciprocity property may be proved similarly [7, 9].

(5) Again without solving the problem explicitly, one can demonstrate that the sum of all intensities always equals 1. Indeed, multiplying (47) by Ψ^* , adding the complex conjugate equation term by term and integrating the result with respect to ξ over the period π , yields

$$\int_0^\pi \Psi(\xi, \zeta) \Psi^*(\xi, \zeta) d\xi = \pi, \quad (57)$$

where we have taken into account (48). Substitution of (50) into (57) leads, after explicit integration of the left hand side, to

$$\sum_{n=-\infty}^{+\infty} \psi_n(\zeta) \psi_n^*(\zeta) = \sum_{n=-\infty}^{+\infty} I_n(\zeta) = 1. \quad (58)$$

This result should not surprise us for we assumed that there is no loss of energy in the medium.

(6) It can not be denied that the present approach in an economizing modification of an original GFM, established by Kuliasko, Mertens and Leroy [20, 26] and further developed by Plancke-Schuyten *et al.* [29, 30, 31].

Indeed, by starting directly from Maxwell's equations, and systematically introducing the necessary approximations, the present approach avoids the detour along the RN equations (51). From our point of view that infinite system is now redundant, except when it is truncated in accordance to special cases of Bragg diffraction [7, 9, 11, 25, 28].

In the following two sections, we will deal with approximate solutions of (47) for different values of the parameters ρ and φ . In the last section we draw special attention to its exact solution.

2. Raman-Nath diffraction

From physical arguments (see e.g. [16, 17]), RN diffraction is only possible for small AO interaction lengths L (thin phase grating approximation). Mathematically

speaking, the condition $\rho \ll 1$ essentially defines the RN regime, provided the RN parameter v is kept sufficiently small as well. Quite often, after Klein and Cook [15], the criterion $Q = \rho v = K^2 L / k \ll 1$ is used. Today there is a big controversy about the validity of this classical criterion [7, 32], but a profound discussion of that would deviate us too far from our subject.

In the RN limit $\rho = 0$, intensities of the squared Bessel function form may be obtained. Indeed, the π -periodic solution of (47), satisfying the boundary condition (48), reads

$$\begin{aligned} \Psi(\xi, \zeta) &= \exp \frac{i}{b} [\sin(b\zeta) \cos(2\xi - b\zeta)] \\ &= \sum_{n=-\infty}^{+\infty} i^n \exp(2in\xi) \exp(-inb\zeta) J_n\left(\frac{\sin b\zeta}{b}\right) \end{aligned} \quad (59)$$

with $b = (a \sin \varphi) / 2$. The diffraction intensities, evaluated in the plane $z = L$, are then given by

$$I_n(v) = J_n^2\left(\frac{2 \sin\left(\frac{1}{2}av \sin \varphi\right)}{a \sin \varphi}\right). \quad (60)$$

Remark that the first order diffraction intensity is largest when $\varphi = 0$, i.e. at normal incidence of the light. The formula (60) for $\varphi = 0$ reduces to

$$I_n(v) = J_n^2(v), \quad (61)$$

a classical result, first obtained by Raman and Nagendra Nath with a geometric optics theory (see e.g. [24]). The formula (61) follows from (44) by simply putting $\gamma = 0$.

Theoretically, all the light would be diffracted when $I_0(v) = J_0^2(v) = 0$, i.e. for $v \cong 2.4$ (the first zero of the zeroth-order Bessel function).

Also remark that with the aid of (45), the total intensity of the spectra corresponding to (60) or (61) is easily proved to equal the initial (normalized) intensity.

For $\rho \ll 1$, i.e. in the RN regime, correction terms to either (60) or (61) may be calculated by using a perturbation expansion approach. On substituting the series expansion

$$\Psi(\xi, \zeta) = \sum_{n=0}^{+\infty} (i\rho)^n \Phi_n(\xi, \zeta), \quad (62)$$

into (47) and equating equal orders of ρ , one finds the following perturbation scheme

$$\frac{\partial \Phi_n}{\partial \zeta} - i(\cos 2\xi) \Phi_n = -\frac{1}{8}(1 - \delta_{n0}) \frac{\partial^2 \Phi_{n-1}}{\partial \xi^2}, \quad n \in \mathbb{N}, \quad (63)$$

for the unknown π -periodic functions $\Phi_n(\xi, \zeta)$, subject to the boundary conditions $\Phi_n(\xi, 0) = \delta_{n0}$.

Solving for Φ_0 , Φ_1 and Φ_2 , inserting these into (62) and expanding into Fourier series, we finally obtain [9]

$$\begin{aligned} \psi_n(\zeta) = J_n(\zeta) + \frac{i\rho\zeta}{6} (n^2 J_n + \frac{\zeta}{2} J'_n) \\ - \frac{\rho^2 \zeta^2}{1440} \{ [n^2 (20n^2 + 7) - 9\zeta^2] J_n + 3\zeta (12n^2 - 1) J'_n \}, \end{aligned} \quad (64)$$

where $J'_n = dJ_n/d\zeta$. The intensity in $z = L$, is then calculated from (55), up to terms in ρ^2 :

$$I_n(v) = J_n^2(v) - \frac{\rho^2 v^2}{720} [(7n^2 - 9v^2) J_n^2(v) + v (6n^2 - 3) J_n(v) J'_n(v) - 5v^2 J_n'^2(v)]. \quad (65)$$

Some comments on the validity of this result may be found elsewhere [9, 20, 24].

For $\rho \gg 1$, we will consider two different cases. The first, corresponding to normal incidence, will result in a symmetric spectrum with respect to the central line and only two sidelines with significant intensity. Here again, the interaction length L must be kept small, so that $Q = \rho v \ll 1$.

According to Klein-Cook's criterion, this spectrum is of the RN type. In order to calculate the intensities of this particular spectrum, we introduce the new variable $\theta = \rho\zeta$ (remark that $Q = \theta L/z$). Hence (47) can be written as

$$\frac{\partial \Theta}{\partial \theta} - \frac{i}{\rho} (\cos 2\xi) \Theta = -\frac{1}{8} i \frac{\partial^2 \Theta}{\partial \xi^2}, \quad (66)$$

where $\Theta(\xi, \theta) = \Psi(\xi, \zeta)$ is subject to the conditions $\Theta(\xi, 0) = 1$ and $\Theta(\xi + \pi, \theta) = \Theta(\xi, \theta)$. For $\rho \gg 1$, obviously we try to find a solution of the form

$$\Theta(\xi, \theta) = \sum_{n=0}^{+\infty} \left(\frac{i}{\rho}\right)^n \Theta_n(\xi, \theta). \quad (67)$$

Substitution of this expansion into (66) and equating equal orders in $1/\rho$, yields

$$\frac{\partial \Theta_n}{\partial \theta} + \frac{1}{8} i \frac{\partial^2 \Theta_n}{\partial \xi^2} = (1 - \delta_{n0}) (\cos 2\xi) \Theta_{n-1}. \quad (68)$$

From (48) and (49) follows that $\Theta_n(\xi, \theta)$ ($n = 0, 1, 2, \dots$) must be π -periodic functions in ξ satisfying $\Theta_n(\xi, 0) = \delta_{n0}$. Subsequently calculating Θ_0 , Θ_1 and Θ_2 , substituting these into (67) and expanding the resulting function into its Fourier series, the expressions of the amplitudes ψ_0 , $\psi_{\pm 1}$ and $\psi_{\pm 2}$ follow (up to orders in $1/\rho^2$). Calculation of the intensities through (55), finally gives (for $z = L$)

$$\begin{aligned} I_0(v) &= 1 - \frac{8}{\rho^2} \sin^2 \frac{1}{4} \rho v, \\ I_1(v) &= I_{-1}(v) = \frac{4}{\rho^2} \sin^2 \frac{1}{4} \rho v, \\ I_n(v) &= I_{-n}(v) = 0, \text{ for } n \geq 2. \end{aligned} \quad (69)$$

These compact expressions were formerly obtained by Mertens [23] as the result of a direct calculation (up to the second order) and by Mertens and Kuliasko [26] after derivation of the exact solution of the problem (47) - (48) - (49) in terms of Mathieu functions (see later Section 4) and subsequent approximations. Once again, remark that the sum of the intensities in (69), although neglecting corrections terms in $1/\rho^3$, $1/\rho^4$, etc., exactly equals 1.

3. Bragg diffraction

In the second case, which is far more interesting for applications, the light enters the medium at a specific angle φ_{BR} . That angle satisfies the expression

$$\sin\varphi_{BR} \cong \varphi_{BR} = \frac{K}{2k} = \frac{\lambda}{2\Lambda} = \frac{\lambda f}{2V} = \frac{\rho}{2a}, \quad (70)$$

which is easily obtained from the conservation of momentum for the light and sound wave vectors [16, 17]. In analogy to diffraction of X-rays in crystals the angle φ_{BR} is referred to as the Bragg angle.

For $f < 1$ GHz one may replace $\sin\varphi_{BR}$ by φ_{BR} (as in (70)) and accordingly put $\cos\varphi_{BR} = 1$. More generally, one can define a Bragg angle of order n (n integer) by

$$\sin\varphi_{BR}^{(n)} = \frac{n\lambda}{2\Lambda} = \frac{n\rho}{2a}, \quad (71)$$

for which obviously the right hand side of (51) vanishes. Note that for first order Bragg incidence ($\varphi_{BR}^{(1)} = \varphi_{BR}$), from (35) however with θ_n replaced by $\theta_n - \varphi_{BR}$, we obtain

$$\theta_1 = -\varphi_{BR}. \quad (72)$$

This means that the angle between the incident and diffracted first order beam is $2\varphi_{BR}$. Therefore, light is reflected by the sound wave fronts [24] and practically only one order is significantly diffracted (see fig. 2a). Many authors take this argument as the definition of the Bragg diffraction regime. Physical arguments [16, 17] indicate that Bragg diffraction is only possible if the AO interaction length L is greater than a certain minimum (thick phase grating approximation). In Klein-Cook's terminology Bragg diffraction occurs for $Q \gg 1$.

For exact first order Bragg incidence, $\varphi = \varphi_{BR}$, (47) becomes

$$\frac{\partial\Psi}{\partial\zeta} - i(\cos 2\xi)\Psi = -\frac{1}{8}i\rho\frac{\partial^2\Psi}{\partial\xi^2} - \frac{\rho}{4}\frac{\partial\Psi}{\partial\xi}, \quad (73)$$

Again applying a perturbation method, with an expansion of type (67) in $1/\rho$ ($Q \gg 1$ implies $\rho \gg 1$), readily leads [9] to easy formulae for the output light intensities (in the plane $z = L$):

$$\begin{aligned}
I_0(v) &= 1 - \frac{1}{4} v^2 - \frac{1}{\rho^2} \sin^2 \frac{1}{2} \rho v, \\
I_{-1}(v) &= \frac{1}{\rho^2} \sin^2 \frac{1}{2} \rho v, \\
I_{+1}(v) &= \frac{1}{4} v^2, \\
I_n(v) &= I_{-n}(v) = 0, \text{ for } n \geq 2.
\end{aligned} \tag{74}$$

These expressions, valid up to orders in $1/\rho^2$, serve as a good example of an asymmetric spectrum. For $\rho \gg 1$ and v sufficiently small, the formulae simplify further to

$$\begin{aligned}
I_0(v) &= 1 - \frac{1}{4} v^2, \\
I_{+1}(v) &= \frac{1}{4} v^2,
\end{aligned} \tag{75}$$

all the other intensities being negligible.

The simple expressions (75) are in good agreement with Phariseau's formulae [28],

$$\begin{aligned}
I_0(v) &= \cos^2 \frac{v}{2}, \\
I_{+1}(v) &= \sin^2 \frac{v}{2},
\end{aligned} \tag{76}$$

obtained from the direct integration of a pair of RN equations.

It is sometimes of interest to consider oblique incidence according to higher order Bragg angles, defined in (71), or to take into account small deviations from Bragg angle incidence. Space is lacking here to discuss the various cases explicitly [7, 9, 11]. Instead, $I_1(v)$ and $I_{-1}(v)$ have been tabulated, in Table 1, for a number of cases of ideal Bragg and near Bragg incidence. Note that $I_0 = 1 - I_1 - I_{-1}$ and that introducing the parameter

$$\beta = - \frac{2a \sin \varphi}{\rho}, \tag{77}$$

helps in distinguishing the various cases. For example, $\beta = -n$ corresponds to Bragg angle incidence of order n ($\varphi = \varphi_{BR}^{(n)}$, as defined in (71)). Furthermore, the parameter β will play an important role in the exact integration of the problem as we will discuss now.

Table I. Intensities of order ± 1 in the case $\rho \gg 1$.

Case	$-\beta$	$I_1(v)$	$I_{-1}(v)$
normal incidence $\varphi = 0$	0	$\frac{4\sin^2 \rho v/4}{\rho^2}$	$\frac{4\sin^2 \rho v/4}{\rho^2}$
Bragg angle of even order $\varphi = \varphi_{BR}^{(2k)}$	$2k$ $k \in \mathbb{Z} \setminus \{0\}$	$\frac{4\sin^2(1+2k)\rho v/4}{(1+2k)^2 \rho^2}$	$\frac{4\sin^2(1-2k)\rho v/4}{(1-2k)^2 \rho^2}$
Bragg angle of odd order $\varphi = \varphi_{BR}^{(2k+1)}$	$2k+1, k \in \mathbb{Z}$ $k \neq 0, -1$	$\frac{\sin^2(1+k)\rho v/2}{(1+k)^2 \rho^2}$	$\frac{\sin^2 k \rho v/2}{k^2 \rho^2}$
$\varphi = \varphi_{BR}^{(1)}$	$k = 0$	$\frac{\sin^2 \rho v/2}{\rho^2}$	$v^2/4$
$\varphi = \varphi_{BR}^{(-1)}$	$k = -1$	$v^2/4$	$\frac{\sin^2 \rho v/2}{\rho^2}$
Neighbourhood of Bragg angle of even order $\varphi \approx \varphi_{BR}^{(2k)}$	$2k+\beta_1,$ $k \in \mathbb{Z}$ $\beta_1 \in]0, 1[$	$\frac{4\sin^2(1+2k+\beta_1)\rho v/4}{(1+2k+\beta_1)^2 \rho^2}$	$\frac{4\sin^2(1-2k-\beta_1)\rho v/4}{(1-2k-\beta_1)^2 \rho^2}$
Neighbourhood of Bragg angle of odd order $\varphi \approx \varphi_{BR}^{(2k+1)}$	$2k+1+\beta_1,$ $k \in \mathbb{Z}$ $\beta_1 \in]0, 1[$	$\frac{4\sin^2(2+2k+\beta_1)\rho v/4}{(2+2k+\beta_1)^2 \rho^2}$	$\frac{4\sin^2(2k+\beta_1)\rho v/4}{(2k+\beta_1)^2 \rho^2}$

4. Exact solution of the problem

In this concluding section we will outline how the equation (47), together with (48) and (49), can be solved exactly. A discussion of all the details of this complicated construction is not appropriate for this review, we rather refer to earlier work [7, 9, 24, 25, 26, 29].

Examining (47) we may try to separate the variables by substituting

$$\Psi(\xi, \zeta) = Y(\xi) Z(\zeta), \quad (78)$$

yielding

$$\frac{dZ}{d\zeta} = \frac{1}{2} i \alpha Z, \quad (79)$$

$$\frac{d^2 Y}{d\xi^2} + 2i\beta \frac{dY}{d\xi} + \left(\frac{4\alpha}{\rho} - \frac{8}{\rho} \cos 2\xi \right) Y = 0, \quad (80)$$

where $i\alpha$ is the separation constant. The general solution of (79) reads

$$Z(\zeta) = C \exp\left(\frac{1}{2} i \alpha \zeta\right), \quad (81)$$

while (80) can be transformed into the canonical form of the well-known Mathieu equation [2, 21, 22]:

$$\frac{d^2 y}{d\xi^2} + (a' - 2q \cos 2\xi) y = 0, \quad (82)$$

where

$$y(\xi) = Y(\xi) \exp(i\beta\xi), \quad (83)$$

and with

$$a' = \frac{4\alpha}{\rho} + \beta^2, \quad q = \frac{4}{\rho}. \quad (84)$$

With regard to the periodicity condition (49) and the transformation (83), we must show that there exist solutions of (82) satisfying the condition

$$y(\xi + \pi) = y(\xi) \exp(i\beta\pi). \quad (85)$$

The values of β , being coupled with the angle of incidence φ by its definition (77), will indeed be of paramount importance for the construction of such solutions $y(\xi)$.

Taking into consideration Floquet's theory [2] it may be shown [9] that the only solutions of physical importance are those summarized in Table 2. For a better comparison with the extensive literature on the Mathieu equation, we wrote argument x instead of ξ . The functions $ce_n(x; q)$ and $se_n(x; q)$ represent the Mathieu cosine and sine of order n ($n \in \mathbb{N}$), corresponding to the characteristic values of $a' = a_n$ and $a' = b_n$, respectively. The functions $ce_{n, \beta_1}(x; q)$, $se_{n, \beta_1}(x; q)$, $ce_{n, 1-\beta_1}(x; q)$ and $se_{n, 1-\beta_1}(x; q)$ are Mathieu functions of fractional order ($n \in \mathbb{N}$, $0 < \beta_1 < 1$), where the corresponding characteristic numbers $a' = a_{n, \beta_1}$, $a' = a_{n, 1-\beta_1}$ have to be chosen in stable domains of the stability chart for Mathieu functions [2, p. 123; 21, p. 40-41, p. 98].

Table II. Solutions of the Mathieu equation (82) with parameters (84) for different values of β .

Value of β	Condition	Solution	Characteristic number a'
$\beta = 2k + \beta_1$ $k \in \mathbb{Z}$ $0 < \beta_1 < 1$	$f(x+\pi) = f(x)e^{i\beta_1\pi}$	$me_{2n, \beta_1}(x; q) = ce_{2n, \beta_1}(x; q) + ise_{2n, \beta_1}(x; q)$ or $me_{-(2n+1, 1-\beta_1)}(x; q) = ce_{2n+1, 1-\beta_1}(x; q) - ise_{2n+1, 1-\beta_1}(x; q)$ $n \in \mathbb{N}$	$a_{2n} < a_{2n, \beta_1} < b_{2n+1}$ or $a_{2n+1} < a_{2n+1, 1-\beta_1} < b_{2n+2}$ $n \in \mathbb{N}$
$\beta = 2k + 1 + \beta_1$ $k \in \mathbb{Z}$ $0 < \beta_1 < 1$	$f(x+\pi) = -f(x)e^{i\beta_1\pi}$	$me_{-(2n, 1-\beta_1)}(x; q) = ce_{2n, 1-\beta_1}(x; q) - ise_{2n, 1-\beta_1}(x; q)$ or $me_{2n+1, \beta_1}(x; q) = ce_{2n+1, \beta_1}(x; q) + ise_{2n+1, \beta_1}(x; q)$ $n \in \mathbb{N}$	$a_{2n} < a_{2n, 1-\beta_1} < b_{2n+1}$ or $a_{2n+1} < a_{2n+1, \beta_1} < b_{2n+2}$ $n \in \mathbb{N}$
$\beta = 2k$, $k \in \mathbb{Z}_0$ $\beta = 2k + 1$, $k \in \mathbb{Z}$ (Bragg cases)	$f(x+\pi) = f(x)$ $f(x+\pi) = -f(x)$	$ce_{2n}(x; q)$ or $se_{2n+2}(x; q)$ $n \in \mathbb{N}$ $ce_{2n+1}(x; q)$ or $se_{2n+1}(x; q)$ $n \in \mathbb{N}$	a_{2n} or b_{2n+2} $n \in \mathbb{N}$ a_{2n+1} or b_{2n+1} $n \in \mathbb{N}$
$\beta = 0$ (normal incidence)	$f(x+\pi) = f(x)$ $f(x) = f(-x)$	$ce_{2n}(x; q)$ $n \in \mathbb{N}$	a_{2n} $n \in \mathbb{N}$

As an example let us construct the solution in the case $\beta = 2k + \beta_1$. The equation (47) is linear, hence, the sum of all particular solutions is still a solution of it. Using (78), (81) and (83), together with the solution as listed in the first row of Table 2, we obtain

$$\begin{aligned} \Psi(\xi, \zeta) = & \sum_{n=0}^{\infty} \{ C_{2n, \beta_1} \exp\left(\frac{1}{2} i \alpha_{2n, \beta_1} \zeta\right) \text{me}_{2n, \beta_1}(\xi; q) \\ & + C_{2n+1, 1-\beta_1} \exp\left(\frac{1}{2} i \alpha_{2n+1, 1-\beta_1} \zeta\right) \text{me}_{-(2n+1; 1-\beta_1)}(\xi; q) \} \cdot \\ & \exp[-i(2k + \beta_1)\xi], \end{aligned} \quad (86)$$

where according to (84)

$$\begin{aligned} \alpha_{2n, \beta_1} &= \frac{\rho}{4} [a_{2n, \beta_1} - (2k + \beta_1)^2], \\ \alpha_{2n+1, 1-\beta_1} &= \frac{\rho}{4} [a_{2n+1, 1-\beta_1} - (2k + \beta_1)^2], \end{aligned} \quad (87)$$

Applying the boundary condition (48), then requires

$$\begin{aligned} 1 = & \sum_{n=0}^{+\infty} \exp[-i(2k + \beta_1)\xi] \{ C_{2n, \beta_1} \text{me}_{2n, \beta_1}(\xi; q) \\ & + C_{2n+1, 1-\beta_1} \text{me}_{-(2n+1, 1-\beta_1)}(\xi; q) \}. \end{aligned} \quad (88)$$

Next, taking into account the Fourier expansions [21, p. 79; 22, p. 111-116], i.e.

$$\begin{aligned} \text{me}_{2n, \beta_1}(\xi; q) &= \sum_{r=-\infty}^{+\infty} A_{2r}^{2n, \beta_1}(q) \exp[i(2r + \beta_1)\xi], \\ \text{me}_{-(2n+1, 1-\beta_1)}(\xi; q) &= \sum_{r=-\infty}^{+\infty} A_{-(2r+1)}^{2n+1, 1-\beta_1}(q) \exp[i(2r + \beta_1)\xi], \end{aligned} \quad (89)$$

considering the coefficients C_{2n, β_1} and $C_{2n+1, 1-\beta_1}$ as complex quantities, using the orthogonality relations between Mathieu functions of fractional order (put $\beta_1 = v/w$) [21, p. 24/82], we find that the coefficients in (86) are all real, and given by

$$\begin{aligned} C_{2n, \beta_1} &= A_{2k}^{2n, \beta_1}(q), \\ C_{2n+1, 1-\beta_1} &= A_{-(2k+1)}^{2n+1, 1-\beta_1}(q). \end{aligned} \quad (90)$$

Substituting those constants into (86), and once again using the expansion (89), it is straightforward to obtain the amplitudes $\psi_n(\zeta)$ from which the n -th order intensity readily follows. In $z = L$ we thus obtain

$$\begin{aligned}
I_n(v) = & \delta_{n0} - 4 \sum_{\substack{r,s=0 \\ r < s}}^{+\infty} A_{2k}^{2r,\beta_1} A_{2(n+k)}^{2r,\beta_1} A_{2k}^{2s,\beta_1} A_{2(n+k)}^{2s,\beta_1} \cdot \\
& \sin^2 \frac{1}{16} (a_{2r,\beta_1} - a_{2s,\beta_1}) \rho v \\
& - 4 \sum_{\substack{r,s=0 \\ r < s}}^{\infty} A_{-(2k+1)}^{2r+1,1-\beta_1} A_{-(2n+2k+1)}^{2r+1,1-\beta_1} A_{-(2k+1)}^{2s+1,1-\beta_1} A_{-(2n+2k+1)}^{2s+1,1-\beta_1} \cdot \\
& \sin^2 \frac{1}{16} (a_{2r+1,1-\beta_1} - a_{2s+1,1-\beta_1}) \rho v \\
& - 4 \sum_{r,s=0}^{+\infty} A_{2k}^{2r,\beta_1} A_{2(n+k)}^{2r,\beta_1} A_{-(2k+1)}^{2s+1,1-\beta_1} A_{-(2n+2k+1)}^{2s+1,1-\beta_1} \cdot \\
& \sin^2 \frac{1}{16} (a_{2r,\beta_1} - a_{2s+1,1-\beta_1}) \rho v.
\end{aligned} \tag{91}$$

Similar expressions may be obtained for the other cases (see also [7, 9, 24, 25, 26, 29]).

SAMENVATTING

Dit beknopt overzicht van ons recent onderzoekswerk [9] bestaat uit twee duidelijk te onderscheiden delen.

In het eerste deel behandelen we de diffractie van intens laserlicht door een voortlopende ultrageluidsgolf in een isotrope middenstof (een vloeistof bijvoorbeeld). Dit vraagstuk is *niet-lineair*, vermits er in de golfvergelijking kubische termen voorkomen die verwijzen naar het elektrisch veld. Dit type niet-lineariteit geeft aanleiding tot de creatie van de derde harmoniek voor het licht in het midden. In het diffractiepatroon zelf veruitwendigt zich dit door de aanwezigheid van intermediaire lijnen, die liggen tussen de gewone diffractielijnen, die men op dezelfde plaats aantreft als bij de diffractie van gewoon licht.

Onze theoretische studie van dit vraagstuk steunt op een verzoenbare combinatie van Bloembergen's theorie voor de voortbrenging van de derde harmoniek van licht [1, 3, 27] en de theorie van Raman en Nagendra Nath voor de behandeling van akoesto-optische diffractie (zie o.a. [7, 9, 24]).

Na het aanbrengen van de basisprincipes, waarbij we speciale aandacht schenken aan het verschil tussen Raman-Nath diffractie en Bragg diffractie en het begrip isotroop midden, leiden we de basisvergelijking af voor akoesto-optische diffractie (inclusief de generatie van de derde harmoniek). Deze vergelijkingen lossen we exact op in de Raman-Nath limiet, corresponderend met diffractie van licht door een intense ultrasone golf van voldoende lange golflengte. Tenslotte bespreken we het diffractiepatroon algemeen en berekenen we de intensiteiten expliciet voor de Raman-Nath limiet.

In het tweede deel bestuderen we theoretisch de akoesto-optische diffractie van gewoon licht of voldoende zwak laserlicht door een voortlopende ultrasone golf in een vloeistof. Hierbij is de generatie van de derde harmoniek uitgesloten, zodat het probleem lineair is. Bijzondere aandacht gaat naar de gewijzigde voortbrengende-functiemethode, die leidt tot een elegante en alternatieve formulering van het probleem en die tevens toelaat het vraagstuk benaderd of exact op te lossen zonder expliciet gebruik te moeten maken van de Raman-Nath vergelijkingen.

In een eerste paragraaf zetten we de methode zelf uiteen en wijzen hierbij op de wiskundige trefkracht van deze aanpak (b.v. voor de a priori kennis van symmetrieëigenschappen van het spectra). Nadien berekenen we de intensiteiten van spectra in het Raman-Nath en Bragg diffractie regime. Tenslotte tonen we hoe het vraagstuk exact kan worden opgelost door gebruik te maken van de Floquet theorie voor de differentiaalvergelijking van Mathieu. Als voorbeeld berekenen we de intensiteiten van het spectrum dat bekomen wordt bij schuine inval van het licht onder een invalshoek die nauwelijks afwijkt van een hogere orde Bragg hoek van het even type.

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