XII

Diffraction of light by ultrasonic waves

In Chapters I and II it was shown that the propagation of electromagnetic waves may be studied either by using Maxwell's equations, supplemented by the material equations, or by means of certain integral equations which utilize the polarization properties of the medium. In particular, either of these methods may also be applied to the study of the propagation of light through a medium whose density depends on space coordinates and on time. Though the former method has been used extensively in the past, the latter has only more recently been applied to such studies. In this chapter we shall apply the integral equation method to the problem of diffraction of light by a transparent homogeneous medium, disturbed by the passage of ultrasonic waves. It will be useful, however, to give first a qualitative description of this diffraction phenomenon and a brief summary of the theoretical work on this problem based on Maxwell's differential equations.

12.1 Qualitative description of the phenomenon and summary of theories based on Maxwell's differential equations

12.1.1 Qualitative description of the phenomenon

Ultrasonic waves are sound waves whose frequencies are higher than those of waves normally audible to the human ear. The angular frequencies of the ultrasonic waves produced in laboratories lie from about $10^5\ \mathrm{s}^{-1}$ to about $3 \times 10^9\ \mathrm{s}^{-1}$, the former value representing the limit of audibility of the human ear. The corresponding range of wavelengths Λ of course depends on the velocity v of these waves in the medium in which they travel. For example, in water $v = 1.2 \times 10^5$ cm/s and the above frequency range corresponds to the wavelength range $\Lambda = 7.5$ cm to 2.5×10^{-4} cm.*

In 1921 Brillouin† predicted that a liquid traversed by compression waves of short wavelengths, when irradiated by visible light, would give rise to a diffraction phenomenon similar to that due to a grating. In order to see this, consider a fluid lying between two infinite planes $y = 0$ and $y = d$, and let a plane compression wave of wavelength Λ progress through it along the positive x direction. This creates periodic

* For methods of generation of ultrasonic waves and their many uses, see for example, L. Bergmann, *Der Ultraschall* (Zürich, Hirzel, 1954).

† L. Brillouin, *Ann. de Physique*, **17** (1921), 103.

stratifications of matter along the x-axis, the distance between two successive planes of maximum density being Λ.

Let a monochromatic plane light wave of angular frequency ω and wavelength $\bar{\lambda}$ inside the medium be incident, with its wave-normal lying in the x,y-plane and making an angle $\bar{\theta}$ with the y-axis (see Fig. 12.1). Further let $\bar{\phi}$ denote the angle which a diffracted ray makes with the y-axis. Since the velocity v of the compression waves is always very much smaller than the velocity of light, we may, to a first approximation, consider the stratification of matter to be stationary. Then the directions $\bar{\phi}$ in which there is an appreciable intensity are determined by the condition that the optical path difference between the rays from two successive planes distance Λ apart shall be an integral multiple of $\bar{\lambda}$. This condition gives a relation between $\bar{\lambda}$, $\bar{\theta}$ and the directions of propagation $\bar{\phi}_l$ of the waves of various orders in the diffracted spectrum:

$$BC - AD = \Lambda(\sin\bar{\phi}_l - \sin\bar{\theta}) = l\bar{\lambda} \qquad (l = 0, \pm1, \pm2, \ldots), \tag{1}$$

AB and CD being portions of wave-fronts associated with the refracted and diffracted rays. It will be convenient to rewrite (1) in terms of the angles θ and ϕ, and the wavelength λ outside the medium. If we use in (1) the law of refraction

$$\frac{\sin\bar{\theta}}{\sin\theta} = \frac{\sin\bar{\phi}}{\sin\phi} = \frac{\bar{\lambda}}{\lambda},$$

we obtain

$$\Lambda(\sin\phi_l - \sin\theta) = l\lambda \qquad (l = 0, \pm1, \pm2, \ldots). \tag{2}$$

From (2) one has for the angular separation between successive orders

$$\sin\phi_l - \sin\phi_{l-1} \simeq \phi_l - \phi_{l-1} = \frac{\lambda}{\Lambda}.$$

Thus, for a given λ, the angular separation decreases with increasing Λ. If Λ is sufficiently large, the principal lines will be so close together that they will not be resolved in the observing instrument, and for this reason diffraction effects are not observed when ordinary sound waves are irradiated by visible light.

It was nearly a decade after Brillouin's prediction that Debye and Sears,* and Lucas

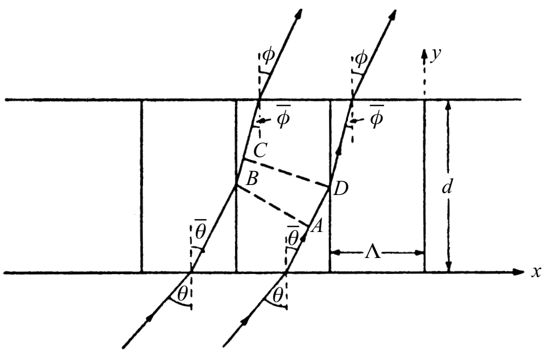

Fig. 12.1 Ultrasonic waves acting as a diffraction grating.

* P. Debye and F. W. Sears, *Proc. Nat. Acad. Sci., Wash.*, **18** (1932), 409.

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and Biquard,* independently observed the diffraction of light by ultrasonic waves. Since then, many investigators have studied this phenomenon under a variety of experimental conditions obtained by varying one or more of the following quantities: (a) the angle of incidence θ, (b) the wavelength Λ of the ultrasonic wave, (c) the wavelength λ of the incident light, (d) the amplitude of the ultrasonic waves, (e) the width d of the ultrasonic beam.

Naturally the positions of the various orders on the screen, and their number and relative intensities, depend on one or more of these factors.† Fig. 12.3, p. 692, shows in a typical case the number of orders appearing on either side of the transmitted beam at different angles of incidence θ. The usual experimental arrangement for studying diffraction spectra is schematically drawn in Fig. 12.2.

It will be convenient at this stage to define some of the symbols and sign conventions used in this chapter.

The number density of molecules (atoms) of a medium will be denoted by $N(\mathbf{r}, t)$. For an isotropic homogeneous medium traversed by a plane compression wave propagated in the positive x direction, $N(\mathbf{r}, t)$ may be written in the form‡

$$N(\mathbf{r}, t) = N_0[1 + \Delta \cos(Kx - \Omega t)], \tag{3}$$

where N_0 is the average number density of the medium, $N_0\Delta$ (usually of the order of $10^{-4}N_0$) the amplitude of the compression wave, $K = 2\pi/\Lambda$ its wave number (the magnitude of the wave-vector) and $\Omega = Kv$ the angular frequency of the ultrasonic disturbance. In such a medium the dielectric constant ε will also be a function of the space and time coordinates; this dependence may be assumed to be of the form

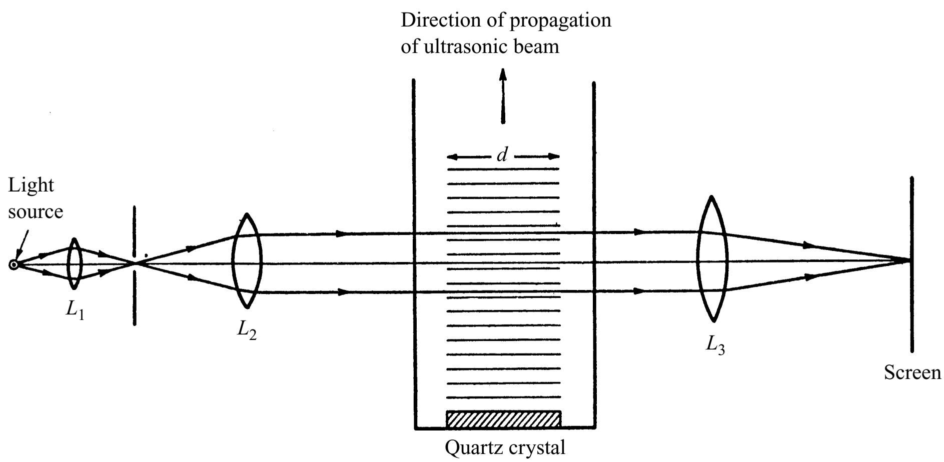

Fig. 12.2 Experimental arrangement for observing diffraction of light by ultrasonic waves.

* R. Lucas and P. Biquard, *J. Phys. Radium*, **3** (1932), 464.

† For a semiquantitative discussion of the dependence of the diffracted spectrum on these factors, see G. W. Willard, *J. Acoust. Soc. Amer.*, **21** (1949), 101.

‡ For simplicity, we shall be concerned only with plane progressive ultrasonic waves. The diffraction of light by standing waves has also been studied experimentally (see Bergmann, *loc. cit.*); the corresponding generalization of the theory is straightforward.

It should be mentioned that completely plane ultrasonic waves are hard to produce experimentally; in general, however, it is possible to regard the wave-fronts to be plane over regions of linear dimensions much greater than Λ.

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$$\varepsilon = \varepsilon_0 + \varepsilon_1 \cos(Kx - \Omega t). \tag{4}$$

There is, of course, a relation between ε_1 and Δ which we shall write in the form

$$\varepsilon_1 = \gamma\Delta. \tag{5}$$

If we assume the Lorentz–Lorenz law (see §2.3 (17))

$$\frac{\varepsilon - 1}{\varepsilon + 2}\frac{1}{N} = \text{constant}, \tag{6}$$

differentiate the logarithm of (6) and remember that both Δ and $\varepsilon_1/\varepsilon_0$ are very much less than unity, we obtain

$$\varepsilon_1\left(\frac{1}{\varepsilon_0 - 1} - \frac{1}{\varepsilon_0 + 2}\right) - \frac{N_0\Delta}{N_0} = 0,$$

or

$$\gamma = \tfrac{1}{3}(\varepsilon_0 - 1)(\varepsilon_0 + 2); \tag{7}$$

γ is of the order of magnitude of unity for most liquids.

We also set

$$\left.\begin{aligned} n &= \sqrt{\varepsilon_0}, & k &= \frac{2\pi}{\lambda}, & \bar{k} &= nk, \\ \delta &= \frac{\Delta\Lambda^2}{\lambda^2}, & \xi &= \frac{\Lambda}{\lambda}\sin\theta, & \beta &= \frac{\pi\lambda}{n\Lambda^2}. \end{aligned}\right\} \tag{8}$$

Finally the angles θ, ϕ, etc., will be measured clockwise from the positive y direction to the direction along which light advances (see Fig. 12.1). It may be assumed that $0 \leqslant \theta < \pi/2$.

12.1.2 *Summary of theories based on Maxwell's equations*

In regions free of currents and charges Maxwell's equations for a nonmagnetic, nonconducting medium, whose dielectric constant ε may be a function of space and time coordinates, are

$$\operatorname{curl}\mathbf{E} = -\frac{1}{c}\frac{\partial\mathbf{H}}{\partial t}, \qquad \operatorname{curl}\mathbf{H} = \frac{1}{c}\frac{\partial\mathbf{D}}{\partial t}, \tag{9a}$$

$$\operatorname{div}\mathbf{H} = 0, \qquad \operatorname{div}\mathbf{D} = 0. \tag{9b}$$

Eliminating $\mathbf{H}$ from (9a), and making use of the relations $\mathbf{D} = \varepsilon\mathbf{E}$, $\operatorname{div}\mathbf{D} = 0$ and $\operatorname{curl}\operatorname{curl} \equiv -\nabla^2 + \operatorname{grad}\operatorname{div}$, we find that

$$\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\varepsilon\mathbf{E}) = \nabla^2\mathbf{E} + \operatorname{grad}(\mathbf{E}\cdot\operatorname{grad}\ln\varepsilon). \tag{10}$$

If we now use (4) and consider $\mathbf{E}$ as a superposition of plane waves of wavelengths $\bar{\lambda} \sim \lambda/n$, we find that the second term on the right-hand side of (10) is of the order $\varepsilon_1(\lambda/\Delta)$ times the first term. Since under the usual experimental conditions both ε_1 and λ/Λ are very much less than unity, we may neglect this term in (10), and obtain

$$\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\varepsilon \mathbf{E}) = \nabla^2 \mathbf{E}. \tag{11}$$

Now consider the physical situation illustrated in Fig. 12.1, and let the incident monochromatic plane electromagnetic wave be linearly polarized with its electric vector perpendicular to the plane of incidence (E-polarization), i.e. along the z-axis. It then follows from the preceding discussion that inside the medium the components E_x and E_y of $\mathbf{E}$ will be small quantities of order $\varepsilon_1(\lambda/\Lambda)$ times E_z and may, therefore, be neglected. Consequently E_x and E_y outside the medium are also negligible.

From the symmetry of the problem it is clear that E_z will be independent of the z coordinate; hence using (4) and (11), we obtain for E_z the equation

$$\frac{\partial^2 E_z}{\partial x^2} + \frac{\partial^2 E_z}{\partial y^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\{[\varepsilon_0 + \tfrac{1}{2}\varepsilon_1(\mathrm{e}^{\mathrm{i}(Kx-\Omega t)} + \mathrm{e}^{-\mathrm{i}(Kx-\Omega t)})]E_z\} = 0. \tag{12}$$

In order to solve (12) assume E_z to be of the form

$$E_z = \sum_l V_l(y)\mathrm{e}^{\mathrm{i}[(k\sin\theta + lK)x - (\omega + l\Omega)t]}, \tag{13}$$

where the summation is over all integral (positive, negative, and zero) values of l.

By substituting (13) into (12) and equating the coefficient of each exponential to zero, we obtain the following recurrence relations for $V_l(y)$

$$\begin{aligned} &V_l''(y) + \{\varepsilon_0 c^{-2}(\omega + l\Omega)^2 - (k\sin\theta + lK)^2\}V_l(y) \\ &\quad = -\tfrac{1}{2}\varepsilon_1 c^{-2}(\omega + l\Omega)^2\{V_{l-1}(y) + V_{l+1}(y)\} \qquad (l = 0, \pm 1, \pm 2, \ldots), \end{aligned} \tag{14}$$

where a prime on $V_l(y)$ denotes differentiation with respect to y. These equations have to be solved subject to the boundary conditions*

$$\left.\begin{aligned} &V_0(0) = B, \quad \text{the amplitude of the incident light wave} \\ \text{and} \quad &V_l(0) = 0 \quad \text{for all } l \neq 0. \end{aligned}\right\} \tag{15}$$

Before solving (14) we observe that (13) represents a superposition of waves of frequencies $\omega_l = \omega + l\Omega$ $(l = 0, \pm 1, \pm 2, \ldots)$. Moreover, the x component of the wave-vector for the wave of frequency ω_l is $k\sin\theta + lK$. Therefore the sine of the angle ϕ_l which the wave of frequency ω_l makes with the y-axis beyond the scattering medium is given by

$$\begin{aligned} \sin\phi_l &= \frac{c(k\sin\theta + lK)}{\omega + l\Omega} \\ &\sim \sin\theta + l\frac{\lambda}{\Lambda}, \qquad \text{since } \frac{\Omega}{\omega} \ll 1, \end{aligned} \tag{16}$$

in agreement with (2). Further the intensity of a particular order l may be taken to be $|V_l(d)|^2$.

We first solve (14) by assuming that $|V_0| \gg |V_{\pm 1}| \gg |V_{\pm 2}| \ldots$. Remembering that

* These boundary conditions are, of course, correct only if the intensities of the reflected waves are negligible; this is so in the present problem since the angle of incidence θ, for which the amplitudes of the diffracted waves are appreciable, is at most about 3° (see also §12.2.4).

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one has to consider only those solutions of (14) which correspond to light waves travelling in directions along which y increases, we may put all V_l, except V_0, equal to zero in (14), make use of (15), and have in the first approximation

$$V_0^{(0)}(y) = B\,\mathrm{e}^{\mathrm{i}\sqrt{\varepsilon_0 - \sin^2\theta}\,ky} + O(\delta^2). \tag{17}$$

Similarly, by putting all V_l, except $V_{\pm 1}$ and V_0, equal to zero in (14), we obtain after a straightforward calculation

$$V_{\pm 1}(y) = \tfrac{1}{4}\gamma\delta\,\frac{1 - \mathrm{e}^{-2\mathrm{i}\beta(\xi \pm \frac{1}{2})y}}{\xi \pm \frac{1}{2}}\,V_0^{(0)}(y). \tag{18}$$

Here use has also been made of (5) and (8). If we now substitute (18) into the equations (14) for $l = 0$ and for $l = \pm 2$, we obtain a correction term to $V_0{}^{(0)}(y)$ and expressions for $V_{\pm 2}(y)$; both are proportional to δ^2 in this approximation. In this manner we obtain expressions for the intensities of any order l in the form of a series in ascending powers of δ. Solutions in terms of such power series were first derived by Brillouin,* who used somewhat more intricate analysis than outlined here, while David,† following the above procedure, gave explicit expressions for the intensities of the first- and second-order lines on the assumption that the intensities of the higher orders are negligible. The latter formulae are quoted in §12.2.5 (38).

Brillouin's approximation (and also David's) is convenient if either $\delta \ll 1$ or $\delta/\xi \ll 1$, for then the method of successive approximations and the power series converge rapidly. When these conditions are satisfied, it is clear that only the first few orders will have appreciable intensity.

The explanation of the simultaneous appearance of many orders and approximate expressions for their intensities were first given by Raman and Nath.‡ They solved (14) in the following manner. Setting in (14)

$$V_l(y) = \mathrm{e}^{\frac{1}{2}\mathrm{i}\,l\pi} \times \mathrm{e}^{\mathrm{i}\bar{k}(\cos\bar{\theta})y} \times U_l(y), \tag{19}$$

and remembering that $\Omega/\omega \sim 10^{-5}$ or less, $\bar{k} = nk$ and $k\sin\theta = \bar{k}\sin\bar{\theta}$, we obtain the following recurrence relations for $U_l(y)$

$$\begin{aligned} &U_l''(y) + 2\mathrm{i}\bar{k}\cos\bar{\theta}\,U_l'(y) - (2l\bar{k}K\sin\bar{\theta} + l^2K^2)U_l(y) \\ &\quad + \tfrac{1}{2}\mathrm{i}(\bar{k})^2\varepsilon_1\varepsilon_0{}^{-1}(U_{l+1}(y) - U_{l-1}(y)) = 0 \qquad (l = 0, \pm 1, \pm 2, \ldots). \end{aligned} \tag{20}$$

Introducing a new variable

$$\chi = \tfrac{1}{2}y\bar{k}\varepsilon_1\varepsilon_0{}^{-1}\sec\bar{\theta},$$

(20) becomes

* L. Brillouin, *La Diffraction de la Lumière par des Ultrasons* (Paris, Hermann, 1933).
† E. David, *Phys. Z.*, **38** (1937), 587.
‡ C. V. Raman and N. S. N. Nath, *Proc. Ind. Acad. Sci.* A, **2** (1935), 406, 413; *ibid.*, **3** (1936), 75, 119.
It may be mentioned that Raman and Nath in their first two papers (*loc. cit.*), following the work of Rayleigh (*Proc. Roy. Soc.*, A, **79** (1907), 399) on phase gratings, found that the amplitudes of the diffracted waves can be represented by Bessel functions. This method considers only the changes in phase of the plane light wave as it traverses the ultrasonic beam.

$$2U_l'(\chi) + U_{l+1}(\chi) - U_{l-1}(\chi) = (\tfrac{1}{2}\mathrm{i}\varepsilon_0{}^{-1}\varepsilon_1)\sec^2\overline{\theta}\,U_l''(\chi) - 2\mathrm{i}\varepsilon_0\varepsilon_1{}^{-1}\{2lK(\overline{k})^{-1}\sin\overline{\theta} + l^2K^2(\overline{k})^{-2}\}U_l(\chi), \quad (21)$$

where now the prime on U denotes differentiation with respect to χ. Since $\lambda/d \ll 1$, the first term on the right is usually of the order $\varepsilon_1 U$, i.e. $10^{-4}U$, and may be neglected. Moreover if, following Raman and Nath, we put the second term also equal to zero, the resulting set of equations are the recurrence relations* satisfied by Bessel functions of integral order. On making use of the boundary conditions (15), we have for the intensity of the lth order wave the expression $B^2J_l^2(\tfrac{1}{2}\varepsilon_1\varepsilon_0{}^{-1}\overline{k}d\sec\overline{\theta})$.

It will be noticed that the approximation made by Raman and Nath essentially consists in neglecting l^2/δ and $l\xi/\delta$ for all l. Therefore, if δ is sufficiently large compared to unity, this approximation will be a good one for the intensities of the lower orders. The Bessel function expressions, however, overestimate the intensities of higher orders. This has been shown by Extermann and Wannier† by numerical calculation of the intensities for three values of the parameter δ. In the work of these authors, the solution of (12) is ultimately determined by equations essentially similar to (18) and (19) of the next section (§12.2).

Finally we mention that Nath‡ and Aggarwal§ have obtained solutions of (21) in the form of power series in ascending powers of $1/\delta$; these series appear to have rather limited applicability as they converge very slowly. Another treatment based on Maxwell's equations in which the diffraction is treated as a boundary value problem was given by Wagner.||

12.2 Diffraction of light by ultrasonic waves as treated by the integral equation method

It was pointed out in §2.4 that the integral equations §2.4 (4) for the effective electric field $\mathbf{E}'(\mathbf{r}, t)$ and the accompanying formula §2.4 (5) for $\mathbf{H}'$ are equivalent to Maxwell's equations for isotropic nonmagnetic substances. It was assumed there that the density of the medium is independent of time but an extension to cover the more general case of time dependence may easily be made. As before, we shall assume the medium to be nonmagnetic and nonconducting.

We recall that the main content of the integral equation method is that the influence of matter on the propagation of an electromagnetic wave is equivalent to the effect of electric dipoles embedded in vacuum, the dipole moment induced in any physically infinitesimal volume element $\mathrm{d}\mathbf{r}'$ of linear dimensions much smaller than λ¶ being proportional to the field $\mathbf{E}'(\mathbf{r}', t)$ acting on it and to the number of molecules (atoms) in that volume. Associated with such a dipole at $\mathbf{r}'$ is the Hertzian vector

* See, for example, E. T. Whittaker and G. N. Watson, *A Course of Modern Analysis* (Cambridge, Cambridge University Press, 4th edition, 1946), p. 360.
† R. Extermann and G. Wannier, *Helv. Phys. Acta*, **9** (1936), 520.
‡ N. S. N. Nath, *Proc. Ind. Acad. Sci.*, A, **4** (1936), 222; *ibid.*, A, **8** (1938), 499.
§ R. R. Aggarwal, Ph.D. thesis, Delhi University, India (1954).
|| E. H. Wagner, *Z. Phys.*, **141** (1955), 604, 622.
¶ Such volume elements, namely elements much larger than the volume of an individual atom (molecule) but of linear dimensions small compared to λ, can be nearly always chosen for optical wavelengths.

$$\mathbf{\Pi}_e = \alpha N\left(\mathbf{r}',\, t - \frac{R}{c}\right)\frac{\mathbf{E}'(t - R/c,\, \mathbf{r}')}{R}\,\mathrm{d}\mathbf{r}',$$

from which the field at a point $\mathbf{r}$ and at time t may be derived by operating on it with (see §2.2 (43))

$$-\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + \operatorname{grad}\operatorname{div}.$$

Here the various symbols have the same significance as in §2.4; thus $R = |\mathbf{r} - \mathbf{r}'|$ and the operator grad div acts on the variables $\mathbf{r}(x,\, y,\, z)$. Then by an argument similar to that which led to §2.4 (4), one now obtains the following integral equation for $\mathbf{E}'$ within the medium*

$$\mathbf{E}'(\mathbf{r},\, t) = \mathbf{E}^{(i)}(\mathbf{r},\, t) + \alpha\iiint\left(-\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + \operatorname{grad}\operatorname{div}\right)\left[N(\mathbf{r}',\, t - R/c)\frac{\mathbf{E}'(\mathbf{r}',\, t - R/c)}{R}\right]\mathrm{d}\mathbf{r}'. \qquad (1)$$

As in §2.4 (4), the integration extends throughout the whole medium, except for a small domain occupied by the atom at the point of observation $\mathbf{r}(x,\, y,\, z)$.

This is the basic integral equation of the present theory. When it is solved for $\mathbf{E}'$ at all points inside the medium, the field outside the medium is calculated by adding together the incident field $\mathbf{E}^{(i)}(\mathbf{r},\, t)$ and the dipole field $\mathbf{E}^{(d)}(\mathbf{r},\, t)$ given by the integral in (1) but extending now throughout the whole medium. It will be noted that this treatment of the propagation of light through media, in contrast to the usual method in which one sets up Maxwell's equations for the medium and the vacuum, avoids explicit introduction of boundary conditions at the refracting surfaces but instead brings in the dimensions of the medium through the process of integration throughout the medium. Moreover, whereas in the Maxwell equations the variation in density of the medium is taken into account through its influence on the dielectric constant ε, in the integral equation (1) the density function $N(\mathbf{r},\, t)$ occurs explicitly.

Eq. (1) is valid only under certain restrictive conditions. First, the polarizability α per molecule in general depends on the frequency of $\mathbf{E}'$, so that $\mathbf{E}'$ should be strictly monochromatic. However, unless one is too near the dispersion frequencies, the variation of α with the frequency of the external field is small. Hence, provided that all the component frequencies of $\mathbf{E}'$ lie close to each other, we may still use (1). Even when the incident field $\mathbf{E}^{(i)}$ is strictly monochromatic, the field $\mathbf{E}'$ acting on a molecule and producing a dipole will not necessarily be monochromatic when $N(\mathbf{r},\, t)$ depends on time, on account of thermal agitation or some other cause of disorder; the spread of frequencies depends on the time variations of $N(\mathbf{r},\, t)$. Hence only when the variation in time of N is slow in comparison with that of $\mathbf{E}^{(i)}$ may (1) be used with confidence;† fortunately this restriction is not severe, since in problems of scattering and diffraction of light the required condition is almost always satisfied.

* Eq. (1) differs from §2.4 (4) in that the operator curl curl in the latter has been replaced by $(-(1/c^2)(\partial^2/\partial t^2) + \operatorname{grad}\operatorname{div})$ and that the density N which now depends on space and time coordinates has also to be taken — just like $\mathbf{E}'$ — at the retarded times $t - R/c$.

† The use of the relation $\mathbf{D} = \varepsilon\mathbf{E}$ in Maxwell's equations for heterogenous media is in fact also justified only under similar restrictions on N.

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Further, we have taken α to be a scalar, an assumption fully justified for atoms and for molecules having special symmetries, but holding also more generally whenever the molecules are oriented at random as already mentioned in §2.3. Finally, the absorption of light by the medium has been assumed to be negligible here; it could be taken into account by allowing α to be complex.

We shall now apply the integral equation method to the problem of diffraction of light by a fluid traversed by ultrasonic waves, following the analysis of Bhatia and Noble.*

12.2.1 Integral equation for E-polarization

Let us again consider the physical situation described in §12.1.1, and assume the incident light to be linearly polarized with its electric vector perpendicular to the plane of incidence; then the components of the electric vector $\mathbf{E}^{(i)}(\mathbf{r}, t)$ of the incident light wave are (the real part, as usual, representing the physical quantity),

$$\left.\begin{aligned} E_x^{(i)} &= E_y^{(i)} = 0, \\ E_z^{(i)} &= B\mathrm{e}^{\mathrm{i}(kx\sin\theta + ky\cos\theta - \omega t)}. \end{aligned}\right\} \tag{2}$$

From the arguments given on p. 675, it may be concluded that the effective field $\mathbf{E}'(\mathbf{r}, t)$ inside the medium will then be nearly parallel to the z-axis, so that we may assume $E'_x = E'_y = 0$, and the vector integral equation (1) for $\mathbf{E}'$ reduces to a single integral equation for E'_z. Remembering that $N(\mathbf{r}, t)$ is now given by §12.1 (3), we may write the integral equation for E'_z in the form

$$\begin{aligned} E'_z(\mathbf{r}, t) = B\mathrm{e}^{\mathrm{i}(kx\sin\theta + ky\cos\theta - \omega t)} &+ \frac{\tau_0}{4\pi}\iiint\left(-\frac{1}{c^2}\frac{\partial^2}{\partial t^2} + \frac{\partial^2}{\partial z^2}\right) \\ &\times\left\{\frac{1}{R}[1 + \tfrac{1}{2}\Delta(\mathrm{e}^{\mathrm{i}[Kx' - \Omega(t - R/c)]} + \mathrm{e}^{-\mathrm{i}[Kx' - \Omega(t - R/c)]})] \times E'_z(\mathbf{r}', t - R/c)\right\}\mathrm{d}\mathbf{r}', \end{aligned} \tag{3}$$

where for convenience we have written†

$$4\pi N_0\alpha = \tau_0.$$

From §2.3 (17)

$$\tau_0 = \frac{3(n^2 - 1)}{n^2 + 2}. \tag{4}$$

12.2.2 The trial solution of the integral equation

Since all planes perpendicular to the z-axis are physically equivalent, we take as a trial solution of our integral equation (3) an expression of the form‡

* W. J. Noble, Ph.D. thesis, University of Edinburgh (1952); A. B. Bhatia and W. J. Noble, *Proc. Roy. Soc.*, A, **220** (1953), 356, 369.

† The macroscopic quantity $\tau(\mathbf{r}, t) = 4\pi\alpha N(\mathbf{r}, t)$ is sometimes called the *scattering index* of the medium.

‡ No confusion should arise between the amplitudes N_{lm} and the number density $N(\mathbf{r}, t)$ of the molecules since from here onwards the latter quantity does not appear explicitly in our equations.

$$E'_z = \sum_{l,m} N_{lm} \mathrm{e}^{-\mathrm{i}(\omega_{lm} t - p_l x - q_m y)}, \tag{5}$$

where l and m are integers (positive, negative and zero). Eq. (5) is seen to represent a doubly infinite sheaf of plane waves; this form of a possible solution is suggested by the multiple reflections and refractions to be expected in an infinite slab of stratified medium with parallel plane faces. It will be seen presently that the various unknowns N_{lm}, ω_{lm}, p_l, and q_m may be determined from the condition that (5) satisfies the integral equation (3).

To solve (3), integrals $\mathcal{J}(\omega, p, q)$ must be evaluated, defined by

$$\begin{aligned}
&\mathcal{J}(\omega, p, q) \times \mathrm{e}^{[-\mathrm{i}(\omega t - px - qy)]} \\
&= \frac{1}{4\pi} \iiint_{V'} \left[-\frac{1}{c^2} \frac{\partial^2}{\partial t^2} + \frac{\partial^2}{\partial z^2} \right] \frac{\mathrm{e}^{-\mathrm{i}[\omega(t - R/c) - px' - qy']}}{R} \, \mathrm{d}x' \, \mathrm{d}y' \, \mathrm{d}z' \\
&= \frac{1}{4\pi} \iiint_{V'} \mathrm{e}^{-\mathrm{i}(\omega t - px' - qy')} \left\{ \frac{\omega^2}{c^2} + \frac{\partial^2}{\partial z^2} \right\} \frac{\mathrm{e}^{\mathrm{i}\omega R/c}}{R} \, \mathrm{d}x' \, \mathrm{d}y' \, \mathrm{d}z',
\end{aligned} \tag{6}$$

where $\omega^2 > c^2 p^2$. If the point of observation x, y, z is outside the scattering medium, the volume V' extends throughout the medium ($-\infty < x' < \infty$, $0 \leqslant y' \leqslant d$, $-\infty < z' < \infty$). If the point of observation is within the scattering medium, the integral extends over the same volume, except for a small sphere of radius a (which is eventually taken to the limit $a \to 0$) around the point of observation. Setting

$$x_1 = x' - x, \qquad y_1 = y' - y, \qquad z_1 = z' - z,$$

(6) may be written in the form, after cancelling the factor $\mathrm{e}^{-\mathrm{i}\omega t}$ on both sides,

$$\mathcal{J}(\omega, p, q) = \mathcal{J}_1 + \mathcal{J}_2, \tag{7}$$

where

$$\mathcal{J}_1 = \frac{1}{4\pi} \iiint_{V_1} \left[\mathrm{e}^{\mathrm{i}(px_1 + qy_1)} \frac{\partial^2}{\partial {z_1}^2} \left(\frac{\mathrm{e}^{\mathrm{i}\omega R/c}}{R} \right) \right] \mathrm{d}x_1 \, \mathrm{d}y_1 \, \mathrm{d}z_1, \tag{8}$$

$$\mathcal{J}_2 = \frac{1}{4\pi} \frac{\omega^2}{c^2} \iiint_{V_1} \left[\mathrm{e}^{\mathrm{i}(px_1 + qy_1)} \left(\frac{\mathrm{e}^{\mathrm{i}\omega R/c}}{R} \right) \right] \mathrm{d}x_1 \, \mathrm{d}y_1 \, \mathrm{d}z_1, \tag{9}$$

the scattering medium now extending over the volume V_1: $-\infty < x_1 < \infty$, $-y \leqslant y_1 \leqslant d - y$, $-\infty < z_1 < \infty$. These integrals are evaluated in Appendix X and give

(a) when (x, y, z) lies inside the scattering medium*

$$\begin{aligned}
\mathcal{J}(\omega, p, q) = {} & \frac{1}{\sigma(\omega, p, q)} - \frac{\omega^2}{2c^2} \frac{\exp[-\mathrm{i}g(\omega, p, q)y]}{g(\omega, p, q)(\omega^2 c^{-2} - p^2)^{1/2}} \\
& + \frac{\omega^2}{2c^2} \frac{\exp[-\mathrm{i}h(\omega, p, q)(y - d)]}{h(\omega, p, q)(\omega^2 c^{-2} - p^2)^{1/2}};
\end{aligned} \tag{10a}$$

* Throughout §12.2 the positive square root of an expression is to be taken, unless otherwise stated.

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(b) when (x, y, z) lies beyond the scattering medium,

$$\mathcal{J}(\omega, p, q) = \frac{\omega^2 \exp[-\mathrm{i}g(\omega, p, q)y]}{2c^2 g(\omega, p, q)(\omega^2 c^{-2} - p^2)^{1/2}} \{\exp[\mathrm{i}g(\omega, p, q)d] - 1\}; \qquad (10b)$$

(c) when (x, y, z) lies in front of the scattering medium (i.e. on the same side as the incident light),

$$\mathcal{J}(\omega, p, q) = \frac{\omega^2 \exp[-\mathrm{i}h(\omega, p, q)y]}{2c^2 h(\omega, p, q)(\omega^2 c^{-2} - p^2)^{1/2}} \{\exp[\mathrm{i}h(\omega, p, q)d] - 1\}. \qquad (10c)$$

In these expressions,

$$\left.\begin{aligned} \sigma(\omega, p, q) &= 3(p^2 + q^2 - \omega^2 c^{-2})(p^2 + q^2 + 2\omega^2 c^{-2})^{-1}, \\ g(\omega, p, q) &= q - (\omega^2 c^{-2} - p^2)^{1/2}, \\ h(\omega, p, q) &= q + (\omega^2 c^{-2} - p^2)^{1/2}. \end{aligned}\right\} \qquad (11)$$

If we now substitute (5) into (3) and make use of the relations (6) and (10a), we obtain in a straightforward manner

$$\begin{aligned}
&-\sum_{l,m} N_{lm} \exp[-\mathrm{i}(\omega_{lm} t - p_l x - q_m y)] + B \exp[(-\mathrm{i})(\omega t - kx\sin\theta - ky\cos\theta)] \\
&+ \tau_0 \sum_{l,m} N_{lm} \left[\frac{\exp[(-\mathrm{i})(\omega_{lm} t - p_l x - q_m y)]}{\sigma(\omega_{lm}, p_l, q_m)} \right. \\
&+ \tfrac{1}{2}\Delta \sum_{+,-} \frac{\exp\{(-\mathrm{i})[(\omega_{lm} \pm \Omega)t - (p_l \pm K)x - q_m y]\}}{\sigma(\omega_{lm} \pm \Omega, p_l \pm K, q_m)} \\
&- \frac{\omega_{lm}^2 \exp\{(-\mathrm{i})[\omega_{lm} t - p_l x - (\omega_{lm}^2 c^{-2} - p_l{}^2)^{1/2} y]\}}{2c^2 g(\omega_{lm}, p_l, q_m)(\omega_{lm}^2 c^{-2} - p_l{}^2)^{1/2}} \\
&- \tfrac{1}{2}\Delta \sum_{+,-} \frac{(\omega_{lm} \pm \Omega)^2 \exp[(-\mathrm{i})\{(\omega_{lm} \pm \Omega)t - (p_l \pm K)x - [c^{-2}(\omega_{lm} \pm \Omega)^2 - (p_l \pm K)^2]^{1/2} y\}]}{2c^2 g(\omega_{lm} \pm \Omega, p_l \pm K, q_m)[c^{-2}(\omega_{lm} \pm \Omega)^2 - (p_l \pm K)^2]^{1/2}} \\
&+ \frac{\omega_{lm}^2 \exp[(-\mathrm{i})\{\omega_{lm} t - p_l x + [\omega_{lm}^2 c^{-2} - p_l{}^2]^{1/2} y\}] \times \exp[\mathrm{i}h(\omega_{lm}, p_l, q_m)d]}{2c^2 h(\omega_{lm}, p_l, q_m)[\omega_{lm}^2 c^{-2} - p_l{}^2]^{1/2}} \\
&+ \tfrac{1}{2}\Delta \sum_{+,-} \frac{(\omega_{lm} \pm \Omega)^2 \exp[(-\mathrm{i})\{(\omega_{lm} \pm \Omega)t - (p_l \pm K)x + [c^{-2}(\omega_{lm} \pm \Omega)^2 - (p_l \pm K)^2]^{1/2} y\}]}{2c^2 h(\omega_{lm} \pm \Omega, p_l \pm K, q_m)} \\
&\left. \times \frac{\exp[\mathrm{i}h(\omega_{lm} \pm \Omega, p_l \pm K, q_m)d]}{[c^{-2}(\omega_{lm} \pm \Omega)^2 - (p_l \pm K)^2]^{1/2}} \right] \equiv 0, \qquad (12)
\end{aligned}$$

where the summation symbol $\sum_{+,-}$ in front of any expression is to be interpreted as follows:

$$\sum_{+,-} F(a \pm b, c \pm d) \equiv F(a + b, c + d) + F(a - b, c - d).$$

In order that (12) shall be satisfied at all times and at all points within the scattering medium, the coefficient of each exponential differing from all the others in any of the variables (x, y, t) must separately vanish. We see from (12) that ω_{lm} changes in steps

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of Ω and is always accompanied by a change of p_l in steps of K. The coefficients of y in the various exponentials, however, either remain unchanged (q_m) or are always the same function of the corresponding ω's and p's. Hence we may take ω_{lm} to depend only on the index l. Moreover, since we may assume without loss of generality that ω_0 is the frequency ω of the incident light, we have

$$\left.\begin{aligned} \omega_0 &= \omega, \qquad & \omega_l &= \omega + l\Omega, \\ p_0 &= k\sin\theta, \qquad & p_l &= k\sin\theta + lK \end{aligned}\right\} \quad (l = 0, \pm 1, \pm 2, \ldots). \qquad \begin{aligned}(13a)\\(13b)\end{aligned}$$

Using these relations in (12) and regrouping the various terms, (12) becomes

$$\begin{aligned} &\sum_{l,m}[N_{lm}(\tau_0/\sigma_{lm} - 1) + \tfrac{1}{2}\Delta(\tau_0/\sigma_{lm})(N_{l-1,m} + N_{l+1,m})] \times \exp[(-\mathrm{i})(\omega_l t - p_l x - q_m y)] \\ &\quad + \sum_l (B\delta_{l,0} - G_l)\exp\{(-\mathrm{i})[\omega_l t - p_l x - (\omega_l^2 c^{-2} - p_l^2)^{1/2} y]\} \\ &\quad + \sum_l H_l \exp\{(-\mathrm{i})[\omega_l t - p_l x + (\omega_l^2 c^{-2} - p_l^2)^{1/2} y]\} \equiv 0, \end{aligned} \tag{14}$$

where $\delta_{l,l'}$ is the Kronecker delta symbol* (i.e. $\delta_{l,l'} = 0$ when $l \neq l'$, and $\delta_{ll} = 1$) and G_l and H_l are given by

$$G_l = \tau_0\omega_l^2 \sum_m [N_{lm} + \tfrac{1}{2}\Delta(N_{l-1,m} + N_{l+1,m})] \times [2c^2 g_{lm}(\omega_l^2 c^{-2} - p_l^2)^{1/2}]^{-1}, \tag{15}$$

$$H_l = \tau_0\omega_l^2 \sum_m [N_{lm} + \tfrac{1}{2}\Delta(N_{l-1,m} + N_{l+1,m})][\exp(\mathrm{i}h_{lm}d)][2c^2 h_{lm}(\omega_l^2 c^{-2} - p_l^2)^{1/2}]^{-1}. \tag{16}$$

Here we have also used the abbreviation (see (11))

$$\sigma_{lm} = \sigma(\omega_l, p_l, q_m), \qquad g_{lm} = g(\omega_l, p_l, q_m) \quad \text{and} \quad h_{lm} = h(\omega_l, p_l, q_m). \tag{17}$$

Equating to zero the coefficient of each exponential in (14), we obtain the following sets of equations for the permissible values of q_m and the amplitudes N_{lm}:

$$N_{lm}(1 - \sigma_{lm}/\tau_0) + \tfrac{1}{2}\Delta(N_{l-1,m} + N_{l+1,m}) = 0, \qquad \text{for all } l \text{ and } m, \tag{18}$$

$$\left.\begin{aligned} B\delta_{l,0} - G_l &= 0, \\ H_l &= 0 \end{aligned}\right\} \quad \text{for all } l. \qquad \begin{aligned}(19)\\(20)\end{aligned}$$

* No confusion should arise between the Kronecker symbol $\delta_{l,l'}$, and the parameter δ introduced in §12.1 (8) since the former always occurs with subscripts.

12.2.3 Expressions for the amplitudes of the light waves in the diffracted and reflected spectra

Before discussing the solution of (18)–(20), we write down the expressions for the total light disturbance at a point (x, y, z) beyond the scattering medium. For this purpose one has to substitute (5) into the integrand on the right-hand side of (3), integrate over the scattering medium, and add to this result the incident field. One again encounters the integrals $\mathcal{J}(\omega, p, q)$ discussed in the preceding section; remembering that for a point beyond the scattering medium $\mathcal{J}(\omega, p, q)$ is given by (10b) and making use of (18), we obtain the following expression for the only nonvanishing component of the total transmitted electric field:

$$E_z = \sum_l B_l \exp\{(-\mathrm{i})[\omega_l t - p_l x - (\omega_l^2 c^{-2} - p_l^2)^{1/2} y]\}, \tag{21}$$

where

$$B_l = \omega_l^2 \sum_m \sigma_{lm} N_{lm}[\exp(\mathrm{i} g_{lm} d)] \times [2c^2 g_{lm}(\omega_l^2 c^{-2} - p_l^2)^{1/2}]^{-1}. \tag{22}$$

According to (21) and (22) the transmitted wave may be regarded as consisting of many plane waves, each with a different frequency and a different direction of propagation. By substituting (13) in the exponential of (21), the expressions for the frequencies ω_l and the angles ϕ_l may be easily obtained; these are the same as those given in §12.1.2.

With the help of (10c) one may similarly write down the expressions for the amplitudes $B_l^{(r)}$ in the reflected spectrum also. $B_l^{(r)}$ are given by

$$B_l^{(r)} = -\omega_l^2 \sum_m \sigma_{lm} N_{lm}[2c^2 h_{lm}(\omega_l^2 c^{-2} - p_l^2)^{1/2}]^{-1}. \tag{23}$$

We shall, however, be concerned here only with points beyond the scattering medium, since in the present problem the intensities of the waves of the various orders in the reflected spectrum are generally very small.

12.2.4 Solution of the equations by a method of successive approximations

In §12.2.4 and §12.2.5, on the assumption that the amplitude Δ of the ultrasonic wave is small, (18)–(20) will be solved and approximate expressions for the intensities of the first- and second-order lines in the transmitted spectrum will be obtained. The case for which this approximation fails will be qualitatively discussed in §12.2.6, and finally, in §12.2.7, (18)–(20) will be solved by an approximation essentially equivalent to that of Raman and Nath.

First consider the set (18). The suffix m in these equations distinguishes quantities referring to different permissible values of q. Therefore, we may suppress this suffix and write (18) in the form

$$f_l(q^2)N_l(q^2) - \tfrac{1}{2}\Delta[N_{l-1}(q^2) + N_{l+1}(q^2)] = 0 \qquad (l = 0, \pm 1, \pm 2, \ldots), \tag{24}$$

with

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$$f_l(q^2) = \frac{\sigma_l(q)}{\tau_0} - 1 = \frac{3(p_l^2 + q^2 - n^2\omega_l^2 c^{-2})}{(n^2-1)(p_l^2 + q^2 + 2\omega_l^2 c^{-2})}. \tag{25}$$

Eqs. (24) form an infinite set of linear homogeneous equations for the amplitudes $N_l(q^2)$. The condition for the existence of a nontrivial solution, i.e. $N_l \not\equiv 0$ for all l, is that the determinant formed by the coefficients of N_l vanishes. The roots of this determinantal equation give the permissible values of q^2; let these be denoted by q_m^2 ($m = 0, \pm 1, \pm 2, \ldots$). Corresponding to each such q^2 there are, of course, two values of q, viz. $+|q|$ and $-|q|$ and two sets of amplitudes, i.e. $N_l(+|q|) \equiv N_l^+(q^2)$ and $N_l(-|q|) \equiv N_l^-(q^2)$, ($l = 0, \pm 1, \ldots$). Then for a given permissible value of q^2, say q_m^2, the recurrence relations (24) determine all the $N_l^+(q_m^2)$, ($l = 0, \pm 1, \ldots$) in terms of one of them, say $N_m^+(q_m^2)$. In this way, one may obtain all the amplitudes $N_l^\pm(q^2)$ in terms of $N_m^\pm(q_m^2)$ ($m = 0, \pm 1, \pm 2, \ldots$). These latter sets of amplitudes are to be determined from (19) and (20), which are just enough in number for this purpose. (Note that in expressions (5), (12), (15), etc., the symbol $\sum_m$ implies sum over both $N^+_{\ldots,m}$ and $N^-_{\ldots,m}$ terms.)

We shall obtain here approximate solutions of (24) by using a perturbation method. Regarding Δ as a small parameter and following the usual perturbation procedure, we expand η ($= q^2$) and N_l in powers of $\frac{1}{2}\Delta$:

$$N_l(\eta) = N_l^{(0)} + \tfrac{1}{2}\Delta N_l^{(1)} + (\tfrac{1}{2}\Delta)^2 N_l^{(2)} + \cdots, \tag{26a}$$

$$\eta = \eta^{(0)} + \tfrac{1}{2}\Delta\eta^{(1)} + (\tfrac{1}{2}\Delta)^2\eta^{(2)} + \cdots. \tag{26b}$$

Making use of (26b), $f_l(\eta)$ may be written as

$$f_l(\eta) = f_l(\eta^{(0)}) + \tfrac{1}{2}\Delta\eta^{(1)} f_l'(\eta^{(0)}) + (\tfrac{1}{2}\Delta)^2[\eta^{(2)} f_l'(\eta^{(0)}) + \tfrac{1}{2}(\eta^{(1)})^2 f_l''(\eta^{(0)})] + \cdots, \tag{27}$$

where a prime on f denotes differentiation with respect to η. (Note that $f_l'(\eta)$, $f_l''(\eta) \ldots$ are nonzero for every positive real value of η.) Substituting (26) and (27) in (24), we have ($l = 0, \pm 1, \pm 2, \ldots$)

$$\begin{aligned}
&\{f_l(\eta^{(0)}) + \tfrac{1}{2}\Delta\eta^{(1)} f_l'(\eta^{(0)}) + (\tfrac{1}{2}\Delta)^2[\eta^{(2)} f_l'(\eta^{(0)}) + \tfrac{1}{2}(\eta^{(1)})^2 f_l''(\eta^{(0)})] + \cdots\}\\
&\times [N_l^{(0)} + \tfrac{1}{2}\Delta N_l^{(1)} + (\tfrac{1}{2}\Delta)^2 N_l^{(2)} + \cdots]\\
&- \tfrac{1}{2}\Delta(N_{l-1}^{(0)} + \tfrac{1}{2}\Delta N_{l-1}^{(1)} + \cdots + N_{l+1}^{(0)} + \tfrac{1}{2}\Delta N_{l+1}^{(1)} + \cdots) = 0.
\end{aligned} \tag{28}$$

Equating first to zero those terms in (28) which are independent of Δ, we obtain in the zero-order approximation

$$f_l(\eta^{(0)}) N_l^{(0)} = 0 \qquad (l = 0, \pm 1, \pm 2, \ldots). \tag{29}$$

Eqs. (29) have as solutions,

$$\text{either } f_l(\eta^{(0)}) = 0, \quad N_l^{(0)} \neq 0 \quad \text{or} \quad N_l^{(0)} = 0, \quad f_l(\eta^{(0)}) \neq 0. \tag{30}$$

Denoting by $\eta_l^{(0)}$ the value of $\eta^{(0)}$ given by $f_l(\eta^{(0)}) = 0$, we find that (30) gives

$$\eta_l^{(0)} = n^2\omega_l^2 c^{-2} - p_l^2, \qquad N_l^{(0)}(\eta_l^{(0)}) \equiv N_{ll} \neq 0, \tag{31}$$

and

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$$N^{(0)}_{l'}(\eta^{(0)}_l) \equiv N^{(0)}_{l'l} = N_{ll}\delta_{l'l}. \tag{32}$$

Next equating to zero the coefficient of Δ in (28), we obtain

$$f_l(\eta^{(0)}_m)N_l^{(1)}(\eta^{(0)}_m) + \eta^{(1)}_m f_l'(\eta^{(0)}_m)N^{(0)}_l(\eta^{(0)}_m) = N^{(0)}_{l-1}(\eta^{(0)}_m) + N^{(0)}_{l+1}(\eta^{(0)}_m). \tag{33}$$

Putting $l = m,\ m+1,\ m-1,\ m+2,\ m-2, \ldots$ successively in (33) and making use of (32), we find

$$\eta^{(1)}_m = 0, \tag{34a}$$

$$N^{(1)}_{m\pm1,m} = \frac{N_{mm}}{f_{m\pm1}(\eta^{(0)}_m)}, \tag{34b}$$

and

$$N^{(1)}_{m\pm j,m} = 0 \qquad \text{for } j \geqslant 2. \tag{34c}$$

Similarly, by equating to zero the coefficient of Δ^2 in (28), we obtain the following expressions for the correction to $\eta^{(0)}_m$ and for the amplitudes N_{lm} up to second order:*

$$\eta^{(2)}_m = \frac{1}{f_m'(\eta^{(0)}_m)}\left[\frac{1}{f_{m+1}(\eta^{(0)}_m)} + \frac{1}{f_{m-1}(\eta^{(0)}_m)}\right], \tag{35a}$$

$$N^{(2)}_{m\pm2,m} = \frac{N_{mm}}{f_{m\pm1}(\eta^{(0)}_m)f_{m\pm2}(\eta^{(0)}_m)}, \tag{35b}$$

and

$$N^{(2)}_{m\pm1,m} = 0, \qquad N^{(2)}_{m\pm j,m} = 0 \qquad \text{for } j \geqslant 3. \tag{35c}$$

It will be seen from the foregoing calculations that in the zero-order perturbation calculation only the quantities N_{mm} ($m = 0, \pm1, \ldots$) are different from zero; in the first-order calculation N_{mm} and $N_{m\pm1,m}$ differ from 0, while, in the second order, $N_{m\pm2,m}$ are also different from zero. Likewise, by pursuing the perturbation calculation to still higher orders one obtains more and more nondiagonal amplitudes (i.e. amplitudes whose two suffixes are not equal) which are different from zero. These calculations are lengthy and cannot be given here; but it may be assumed that whenever the perturbation procedure is valid one may neglect the higher-order terms.

The nondiagonal amplitudes (34b) and (35b) are completely determined once the diagonal amplitudes which form the zero-order solution of (24) are known. We determine the latter from (19) and (20). First, however, it will be instructive to examine their solutions in the simple case $\Delta = 0$, for which an exact solution of (18)–(20) is easily obtained. In this case the only possible nonzero amplitudes are the diagonal amplitudes $N^{\pm}_{m,m}$ ($m = 0, \pm1, \ldots$). If we put all the nondiagonal amplitudes equal to

* The formulae given here are valid only if $f_{m\pm1}(\eta^{(0)}_m)$, $f_{m\pm2}(\eta^{(0)}_m)$, ... are all different from zero. If this is not so, the above perturbation procedure has to be modified to take this degeneracy into account. In perturbation calculations up to second order, the degeneracy comes into play when light is incident at angles $\theta = 0$ and $\theta = \sin^{-1}(\lambda/2\Lambda)$. However, we shall not pursue this matter further here, but will give the results for these two cases in §12.2.5.

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zero in (19) and (20), we find that all the amplitudes $N^{\pm}_{m,m}$ are identically equal to zero except $N^{\pm}_{0,0}$, which are given by*

$$N^{+}_{0,0} = (2B/\sigma_{0,0})\cos\theta[(n^2 - \sin^2\theta)^{1/2} - \cos\theta](1 + \rho^2 - 2\rho\cos 2q_0 d)^{-1/2}\,\mathrm{e}^{\mathrm{i}\psi},$$

$$N^{-}_{0,0} = N^{+}_{0,0}(g_{0,0}/h_{0,0})\mathrm{e}^{2\mathrm{i}q_0 d},$$

where

$$\rho = \left|\frac{N^{-}_{0,0}}{N^{+}_{0,0}}\right|^2, \qquad \psi = \tan^{-1}\left[\frac{\rho\sin 2q_0 d}{1 - \rho\cos 2q_0 d}\right]. \tag{36}$$

For normal incidence ($\theta = 0$), (11), (31) and (36) give

$$\rho_0 = \left|\frac{N^{-}_{0,0}}{N^{+}_{0,0}}\right|^2_{\theta=0} = \frac{(n-1)^2}{(n+1)^2}. \tag{37}$$

With the help of (36), (27) and (23), one easily obtains for the reflectivity at normal incidence on a plane parallel plate the expression

$$\left|\frac{B_0^{(r)}}{B}\right|^2 = \frac{4\rho_0\sin^2 q_0 d}{1 + {\rho_0}^2 - 2\rho_0\cos 2q_0 d},$$

in agreement with §7.6 (9), since $q_0 = nk$ for $\theta = 0$.

When Δ differs from zero but is still sufficiently small for the perturbation method to apply, the above solution for $\Delta = 0$ suggests that

$$|N_{0,0}| \gg |N_{\pm1,\pm1}| \gg |N_{\pm2,\pm2}| \ldots .$$

We may, therefore, solve (19) and (20) for the diagonal amplitudes by successive approximations. Moreover, since for normal or near normal incidence (θ is at most about 3° in the ultrasonic diffraction experiments) the ratio of a given N^- to the corresponding N^+ will be small (see (37)), we may neglect the N^- altogether and determine the N^+ from equations (19) alone.† It will be recalled that a similar approximation is also implicit in the use of the boundary conditions §12.1 (15).

The expressions for the diagonal amplitudes $N^{+}_{0,0}$, $N^{+}_{\pm1,\pm1}$ and $N^{+}_{\pm2,\pm2}$ may now be obtained by using (34b), (34c), (35b) and (35c) in (19). Further, the expressions for the intensities of the first- and second-order lines in the transmitted spectrum can be easily written down with the help of (22). These expressions are given in the next section.

12.2.5 Expressions for the intensities of the first and second order lines for some special cases

(a) $\delta/\xi \ll 1$ and ξ large compared to unity

This is the case considered in detail in the previous section. The intensities $I_{\pm1}$ and $I_{\pm2}$ of the first and second order lines are respectively

* There are some misprints in the expressions for $N^{+}_{0,0}$ and $N^{-}_{0,0}$ given by Bhatia and Noble (*loc. cit.*); these have been corrected here.

† For a detailed discussion on these points see Bhatia and Noble (*loc. cit.*). In particular, it is shown there that the effect of the N^- on the amplitudes $B_l^{(r)}$ of the reflected spectrum is generally not negligible.

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$$I_{\pm 1} = |B_{\pm 1}|^2 = \tfrac{1}{4} B^2 \gamma^2 \delta^2 \frac{\sin^2[\beta d(\xi \pm \frac{1}{2})]}{(\xi \pm \frac{1}{2})^2}, \tag{38a}$$

and

$$I_{\pm 2} = |B_{\pm 2}|^2 = \tfrac{1}{64} B^2 \gamma^4 \delta^4 \left[\frac{1}{2(\xi \pm 1)(\xi \pm \frac{1}{2})(\xi \pm \frac{3}{2})}\right]$$
$$\times \left\{\frac{\sin^2[\beta d(\xi \pm \frac{1}{2})]}{(\xi \pm \frac{1}{2})} + \frac{\sin^2[\beta d(\xi \pm \frac{3}{2})]}{(\xi \pm \frac{3}{2})} - \frac{\sin^2[2\beta d(\xi \pm 1)]}{2(\xi \pm 1)}\right\}. \tag{38b}$$

These equations are to be understood with either all upper signs or all lower signs.

We quote without proof the expressions for the intensities for two other cases considered by Bhatia and Noble*

(b) $\xi \sim \frac{1}{2}$, $\delta \ll 1$

$$I_0 = \frac{1}{4} \frac{B^2 \delta^2 \gamma^2}{(\xi - \frac{1}{2})^2 + \frac{1}{4}\delta^2\gamma^2} \left(\frac{(\xi - \frac{1}{2})^2}{\frac{1}{4}\delta^2\gamma^2} + \cos^2\{\beta d[(\xi - \tfrac{1}{2})^2 + \tfrac{1}{4}\delta^2\gamma^2]^{1/2}\} \right), \tag{39a}$$

$$I_{-1} = \frac{1}{4} \frac{B^2 \delta^2 \gamma^2}{(\xi - \frac{1}{2})^2 + \frac{1}{4}\delta^2\gamma^2} \sin^2\{\beta d[(\xi - \tfrac{1}{2})^2 + \tfrac{1}{4}\delta^2\gamma^2]^{1/2}\}. \tag{39b}$$

The expressions for I_1 and I_{-2} are more complicated. For $\xi = \frac{1}{2}$, however, they take the simple form

$$I_1 = \tfrac{1}{16} B^2 \delta^2 \gamma^2 \left\{ -\sin^2\left(\frac{\pi d \Delta \gamma}{2\lambda n}\right) + 2 \sum_{+,-} \sin^2[\beta d(1 \pm \tfrac{1}{4}\delta y)] \right\} \tag{39c}$$

and

$$I_{-2} = \tfrac{1}{16} B^2 \delta^2 \gamma^2 \sin^2\left(\frac{\pi d \Delta \gamma}{2\lambda n}\right). \tag{39d}$$

(c) Normal incidence ($\xi = 0$), $\delta \ll 1$

$$I_1 = I_{-1} = B^2 \delta^2 \gamma^2 \sin^2[\tfrac{1}{2}\beta d(1 + \tfrac{1}{8}\delta^2\gamma^2)], \tag{40a}$$

$$I_2 = I_{-2} = \tfrac{1}{48} B^2 \delta^4 \gamma^4 \{ -\tfrac{1}{4}\sin^2 2\beta d + \sin^2[\tfrac{1}{2}\beta d(1 + \tfrac{1}{8}\delta^2\gamma^2)]$$
$$+ \tfrac{1}{3}\sin^2[\tfrac{3}{2}\beta d(1 - \tfrac{1}{24}\delta^2\gamma^2)]\}. \tag{40b}$$

If we neglect the quantity $\delta^2\gamma^2$ occurring in the argument of the sines in (40), the resulting expressions may also be obtained from (38) by putting $\xi = 0$ in the latter.

As already mentioned, Brillouin† and David,‡ and also Rytov,§ derived expressions

* Bhatia and Noble (*loc. cit.*).
† L. Brillouin, *La Diffraction de la Lumière par des Ultrasons* (Paris, Hermann, 1933).
‡ E. David, *Phys. Z.*, **38** (1937), 587.
§ S. M. Rytov, *Diffraction de la Lumière par les Ultrasons* (Paris, Hermann, 1938).

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(38) for the intensities of the first- and second-order lines in the manner outlined in §12.1. Aggarwal* has derived expressions (38) from the Raman and Nath differential equations §12.1 (21). Phariseau† has shown that expressions (39) for the intensities of the first- and second-order lines when $\xi \sim \frac{1}{2}$ can also be derived from (21).‡ This is, of course, as it should be, since the method based on Maxwell's differential equations and the integral equation method of the present section are equivalent.

12.2.6 Some qualitative results

It is clear from the expressions for the intensities given in the previous section that, for values of δ and ξ such that either (a) $\delta \ll 1$ or (b) $\delta/\xi \ll 1$, only the first few lower orders will appear on each side of the transmitted beam and their intensities will diminish rapidly with increasing orders. When, however, neither (a) nor (b) is satisfied, i.e. when δ and δ/ξ are both large compared to unity, many more orders will in general appear. For this case the solution of (18)–(20), and hence the calculation of the intensities of the various orders, is more difficult. By examining the conditions for the validity of the perturbation method of §12.2.4, it may be shown§ that in solving (18)–(20), one may consider only those amplitudes N_{lm} as nonzero for which both the suffixes l and m lie between the numbers $-M_1$ and M_2 defined approximately by $(0 \leqslant \xi < \delta^{2/3})$

$$\left.\begin{aligned} M_1 &\sim \xi + \delta^{2/3} + 1, \\ M_2 &\sim -\xi + \delta^{2/3} + 1. \end{aligned}\right\} \tag{41}$$

Since, in general, Δ cannot be increased much beyond 10^{-4} and the maximum value of λ/Λ for which diffraction phenomena can be observed is restricted by practical limitations of resolving power, etc., the maximum possible value of δ $(= \Delta\Lambda^2/\lambda^2)$ is about 100. Hence, even under extreme experimental conditions, one has only to solve at most about twenty linear simultaneous equations from each of the infinite sets (18), (19), (20). But even with this simplification the calculations are necessarily tedious and have not been performed.

Qualitatively, the numbers M_1 and M_2 also represent the number of orders likely to appear on the two sides of the direct transmitted beam. According to (41), the number of orders appearing on the two sides of the direct transmitted beam should become different as $\xi = (\Lambda \sin\theta)/\lambda$ increases from zero, more lines appearing on the side which can be reached by light reflected from the wave-fronts of the ultrasonic wave. Parthasarathy‖ has studied experimentally the diffracted spectrum as a function of the angle of incidence θ, and we reproduce in Fig. 12.3 a plate from his paper. In this experiment $(\lambda/\Lambda) = 3 \times 10^{-3}$; assuming $\Delta \sim 10^{-4}$, we have from (41) for normal incidence $M_1 = M_2 = 5$. Table 12.1 gives the number of lines actually observed to appear on each side of the direct transmitted beam at different angles of incidence. Within brackets are the corresponding theoretical numbers as given by (41). (Of

* R. R. Aggarwal, *Proc. Ind. Acad. Sci.*, A, **31** (1950), 417.
† P. Phariseau, *Proc. Ind. Acad. Sci.*, A, **44** (1956), 165.
‡ A discussion of the experimental results in relation to the various expressions for the intensities given in this section may be found in the papers by Bhatia and Noble (*loc. cit.*).
§ Bhatia and Noble (*loc. cit.*).
‖ S. Parthasarathy, *Proc. Ind. Acad. Sci.*, A, **3** (1936), 442.

Table 12.1. *Diffraction of light by ultrasonic waves: number of orders observed, and (in brackets) the number predicted by theory, for different values of the angle of incidence* θ
($\lambda/\Lambda = 3 \times 10^{-3}$, $\Delta \sim 10^{-4}$).

θ	0	0° 06′	0° 22′	0° 39′	1° 01′	1° 23′	1° 45′	2° 07′
ξ	0	0.6	2	4	6	8	10	13
M_1	5 (5)	5 (6)	6 (7)	6 (9)	3	2	1	1
M_2	5 (5)	5 (4)	3 (3)	2 (2)	2	1	1	1

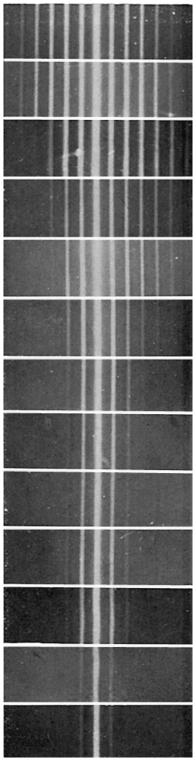

Fig. 12.3 Diffraction of light by ultrasonic waves: spectra observed with different angles θ of incidence ($\lambda/\Lambda = 3 \times 10^{-3}$, $\Delta \sim 10^{-4}$). (After S. Parthasarathy, *Proc. Ind. Acad. Sci.*, A, **3** (1936), 442.)

course, once δ/ξ becomes much less than unity one or two orders only will appear on each side of the direct transmitted beam.)

Similar experimental results have also been obtained by Nomoto.* His curves of M_1 and M_2 against θ, though in qualitative agreement with (41), show also a slight

* O. Nomoto, *Proc. Phys. Math. Soc. Japan*, **24** (1942), 380, 613.

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periodic variation with ξ in the range $0 \leqslant \xi < \delta^{2/3}$. Eqs. (41) are based on too crude a consideration to explain this latter feature.

12.2.7 The Raman–Nath approximation

Finally it will be shown that the Bessel function expressions for the intensities obtained by Raman and Nath may also be deduced from the solution of (18)–(20). Neglecting the small variation of the frequencies ω_l and l and remembering that $q_m^2 \simeq (n^2\omega_m^2 c^{-2} - p_m^2)$, we can take, to a good approximation, $p_l^2 + q^2 + 2\omega_l^2 c^{-2} \simeq k^2(n^2 + 2)$ in the denominator of (25). Hence (24) may be written in the form

$$\frac{q^2 - n^2k^2 + k^2\sin^2\theta}{\frac{1}{2}\Delta\gamma k^2} N_l(q) - N_{l+1}(q) - N_{l-1}(q) = -\frac{l^2 + 2l\xi}{2\gamma\delta} N_l(q)$$

$$(l = 0, \pm 1, \pm 2, \ldots). \quad (42)$$

Now the Bessel function expressions for the intensities were obtained by Raman and Nath by neglecting the terms $[(l^2 + 2l\xi)/\delta] \times U_l$ in their equations §12.1 (21). To the same approximation we may also neglect the right-hand side of (42). Eqs. (42) may then be written as

$$\frac{q^2 - b^2}{\mu^2} N_l(q) = N_{l+1}(q) + N_{l-1}(q) \qquad (l = 0, \pm 1, \ldots), \quad (43)$$

where

$$b^2 = k^2(n^2 - \sin^2\theta) \qquad \text{and} \qquad \mu^2 = \tfrac{1}{2}\Delta\gamma k^2. \quad (44)$$

To solve (43)* let us assume $N_l(q)$ to be of the form

$$N_l(q) = N\,\mathrm{e}^{2\pi \mathrm{i} lm/M}, \quad (45)$$

where M is some very large integer and m is an integer such that $0 \leqslant m < M$. (As will be seen presently the final results are independent of M.) Substituting from (45) in (43), we have for the permissible values of q^2,

$$q_m{}^2 = b^2 + 2\mu^2\cos(2\pi m/M), \qquad 0 \leqslant m < M, \quad (46)$$

or remembering that $\mu^2 \ll b^2$,

$$\pm|q_m| = \pm[b + (\mu^2/b)\cos(2\pi m/M)]. \quad (47)$$

Thus for each m there are two values of q, namely $\pm|q_m|$; the corresponding amplitudes are given by

$$N_l(\pm q_m) \equiv N_{lm}^{\pm} = N^{\pm}\,\mathrm{e}^{2\pi \mathrm{i} lm/M}. \quad (48)$$

The two constants N^+ and N^- in (48) are now to be determined from (19) and (20). As in §12.2.4, we shall neglect the amplitudes $N_{lm}{}^-$ and determine the constant N^+ from (19) alone. Noting that for not too large a value of l, $[\sigma_l(q)/g_l(q)] \sim [\sigma_0(q_0^{(0)})/g_0(q_0^{(0)})]$, we may verify that (19) are identically satisfied by taking

* Eqs. (43) are similar to the relations which determine the normal modes of vibration of a linear chain of atoms (see M. Born and Th. v. Kármán, *Phys. Z.*, **13** (1912), 297).

$$N^+ = \frac{1}{M} 2Bc^2\omega^{-2}(\omega^2 c^{-2} - p_0^2)^{1/2}(g_{0,0}/\sigma_{0,0}) \tag{49}$$

in (48). Substituting (48) and (49) in (22) and putting $({\omega_l}^2 c^{-2} - {p_l}^2) = (\omega^2 c^{-2} - p_0^2)$, we obtain for the amplitude B_l of the lth-order diffracted wave

$$B_l \simeq \frac{B}{M} \sum_{m=0}^{M-1} \mathrm{e}^{\mathrm{i}[2\pi lm/M + (\mu^2 d/b)\cos(2\pi m/M)]}. \tag{50}$$

Here a phase factor independent of m has been omitted. Since by hypothesis M is a very large integer, we may replace the series in (50) by an integral. Putting $2\pi m/M = \psi'$ and $\mathrm{d}\psi' = 2\pi/M$, we may write (50) as

$$B_l \simeq \frac{B}{2\pi} \int_0^{2\pi} \mathrm{e}^{\mathrm{i}[l\psi' + (\mu^2 d/b)\cos\psi']}\,\mathrm{d}\psi', \tag{51}$$

which is independent of M. By splitting the integral in (51) into two parts (i) from 0 to $\frac{3}{2}\pi$ and (ii) from $\frac{3}{2}\pi$ to 2π, and putting in them $\psi' = \frac{1}{2}\pi - \psi$ and $\psi' = \frac{5}{2}\pi - \psi$ respectively, we obtain*

$$\begin{aligned} B_l &\simeq \frac{B}{2\pi} \int_{-\pi}^{\pi} \mathrm{e}^{\mathrm{i}[\frac{1}{2}l\pi - l\psi + (\mu^2 d/b)\sin\psi]}\,\mathrm{d}\psi \\ &= B\,\mathrm{e}^{\frac{1}{2}\mathrm{i}l\pi} J_l(\mu^2 d/b). \end{aligned} \tag{52}$$

Hence the intensities $I_l = |B_l|^2$ are just $B^2 J_l^2(\mu^2 d/b)$. Moreover, it may be seen with the help of (44), §12.1 (5) and §12.1 (8) that the argument $(\mu^2 d/b)$ of the Bessel functions J_l is the same as that occurring in the expressions of Raman and Nath, given on p. 680.

* H. and B. S. Jeffreys, *Methods of Mathematical Physics* (Cambridge, Cambridge University Press, 1946), p. 547.