# Diffraction of Light by Ultrasound in Anharmonic Crystals

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Due to anharmonic effects an initially sinusoidal ultrasonic wave of finite amplitude gradually distorts as it passes through a crystal. In the present paper we calculate the asymmetry in the diffraction pattern formed by passing monochromatic light through a cubic crystal perpendicular to the direction of propagation of such a distorted longitudinal ultrasonic wave. The solution of the nonlinear wave equation for the crystal, obtained by iteration to first order in the nonlinear terms, is used in the diffraction integral to obtain an expression for the light amplitude in the diffraction pattern. The first-order intensities are computed for NaCl. In a typical case, a wave of strain amplitude $2\times10^{-5}$ produces a 10% difference in intensity between the first positive and the first negative orders. These results suggest a method for obtaining the third-order elastic constants of transparent crystals.

## I. INTRODUCTION

THE distortion of finite amplitude ultrasonic waves in liquids is well known.[1–4] As an initially sinusoidal ultrasonic wave progresses through a liquid its waveform distorts. The distortion increases both with increasing sound amplitude and with increasing distance from the source. Consequently, a diffraction pattern formed by passing a monochromatic light beam through the liquid perpendicular to the direction of sound propagation reflects this distortion by becoming asymmetric. For example, the intensity of the first positive diffraction maximum is not equal to the intensity of the first negative maximum. We have performed a calculation which indicates that, as suggested,[3] a similar effect might be observable in transparent crystals. Furthermore, if the distortion of the waveform in crystals is assumed to be due to the departures from Hooke's Law, then the asymmetry in the intensity of the diffraction pattern can be related to the third-order elastic constants of the solid. Thus, measurements of the third-order elastic constants for transparent crystals by optical techniques might be feasible. Since determinations of the third-order elastic constants are with few exceptions[5,6] nonexistent, this apparently simple though, perhaps, not very accurate technique might be useful.[7]

In the present paper an expression for the light amplitude in the diffraction pattern is derived for longitudinal waves in cubic crystals. The wave equation with anharmonic terms included is solved to lowest order in the nonlinear terms. Numerical values of the intensities of diffraction maxima for NaCl are computed as a function of sound amplitude and the distance of the light beam from the sound source.

## II. THE WAVE EQUATION

Consider the propagation of an ultrasonic plane traveling wave in crystal rod which has a transducer attached at $x=0$. The nonlinear wave equation for longitudinal waves is a special case of the equation derived by Seeger and Buck[3]:

$$\frac{\partial^2 u}{\partial t^2}=c^2\frac{\partial^2 u}{\partial x^2}+a\frac{\partial u}{\partial x}\frac{\partial^2 u}{\partial x^2}, \tag{1}$$

where $u$ is the particle displacement in the $x$ direction. If the coordinate axis are parallel to the cubic crystal axis (i.e., the $x$ direction is the [100] direction), then we have

$$c^2=C_{11}/\rho, \quad a=(3C_{11}+6C_{111})/\rho,$$

where $\rho$ is the density of the undeformed crystal and the $C$'s the elastic constants (Voigt notation).

Since the effect of the nonlinear term in Eq. (1) is small, an approximate solution can be obtained by iteration. Let $u_0$ be a solution of Eq. (1) with $a=0$. Then let $u_1$ be a solution of

$$\frac{\partial^2 u_1}{\partial t^2}=c^2\frac{\partial^2 u_1}{\partial x^2}+a\frac{\partial u_0}{\partial x}\frac{\partial^2 u_0}{\partial x^2}.$$

Thus, an approximate solution of Eq. (1) is $u=u_0+u_1$. This procedure can be repeated any number of times. If we assume a boundary condition at $x=0$ corresponding to a sinusoidal driving amplitude, $u(0,t)=-A\sin\Omega t$,

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[1] E. A. Hiedemann and K. L. Zankel, Acustica **11**, 213 (1961).
[2] L. K. Zarembo and V. A. Krasil'nikov, Usp. Fiz. Nauk. **68**, 687 (1959) [translation: Soviet Phys.—Usp. **2**, 580 (1959)].
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[5] T. Bateman, W. P. Mason, and H. J. McSkimin, J. Appl. Phys. **32**, 928 (1961).
[6] R. F. S. Hearmon, Acta Cryst. **6**, 331 (1953).
[7] The ordinary elastic constants can be determined optically by the Schaefer-Bergmann method. The diffraction pattern in this case is formed by a three-dimensional elastic grating in the crystal, whereas in the case we will study it is formed by a traveling longitudinal wave. L. Bergmann, *Ultrasonics* (John Wiley & Sons, Inc., New York, 1938), Chap. IV.

then the solution of Eq. (1) iterated twice is

$$u(x,t)=A\ \sin(Kx-\Omega t)-\frac{(AK)^2ax}{8c^2}\cos2(Kx-\Omega t)$$
$$-\frac{A^3K^4a^2x^2}{32c^4}[\sin3(Kx-\Omega t)+\sin(Kx-\Omega t)]$$
$$+\frac{(AK)^3a^2x}{16c^4}[\tfrac{2}{3}\cos3(Kx-\Omega t)+\cos(Kx-\Omega t)]. \quad (2)$$

This solution holds for $x$ small enough so that $AK^2x\ll1$. Thus, as a result of the nonlinearity higher harmonics of the fundamental driving frequency appear in the solution.

### III. DIFFRACTION OF LIGHT

The interaction of ultrasonic waves with light in a crystal involves the relation between the dielectric constant and the strain. The elasto-optical coefficients $p_{ijkl}$ are defined by[8]

$$\Delta B_{ij}=p_{ijkl}\eta_{kl},$$

where $\eta_{kl}$ is the strain and $B_{ij}$ is the dielectric impermeability tensor, which is related to the dielectric constant $\epsilon_{ij}$ by $\epsilon_{ij}B_{jk}=\delta_{ik}$. (The summation convention that repeated indices be summed from 1 to 3 is implied.) Let $\epsilon$ be the dielectric constant of the unstrained crystal. For the case of cubic crystals $\epsilon$ is isotropic and, thus, $\Delta\epsilon_{ij}=-\epsilon^2\Delta B_{ij}$, and

$$\Delta\epsilon_{ij}=-\epsilon^2p_{ijkl}\eta_{kl}. \quad (3)$$

The strain tensor is related to the displacement by

$$\eta_{ij}=\frac{1}{2}\left(\frac{\partial u_i}{\partial x_j}+\frac{\partial u_j}{\partial x_i}+\frac{\partial u_k}{\partial x_i}\frac{\partial u_k}{\partial x_j}\right). \quad (4)$$

A rigorous discussion of the propagation of electromagnetic radiation through a medium with a varying dielectric constant can be given in terms of Maxwell's Equations.[9] However, a simpler procedure based on the diffraction integral[10] has been shown to yield the same results for small orders in the diffraction pattern.[4,9,11,12] The amplitude of diffracted light of wavelength $\lambda$ is proportional to (see Fig. 1)

$$E=\frac{1}{p}\int_{x_0-p/2}^{x_0+p/2}\exp\left\{\frac{2\pi i}{\lambda}[lx-L\mu(x)]\right\}dx, \quad (5)$$

[8] J. F. Nye, *Physical Properties of Crystals* (Oxford University Press, New York, 1957), pp. 243–255.
[9] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press Inc., New York, 1959), pp. 590–596.
[10] C. V. Raman and N. S. N. Nath, Proc. Indian Acad. Sci. **A2**, 406 (1935).
[11] C. V. Raman and N. S. N. Nath, Proc. Indian Acad. Sci. **A3**, 119 (1936).
[12] K. L. Zankel and E. A. Hiedemann, J. Acoust. Soc. Am. **31**, 44 (1944).

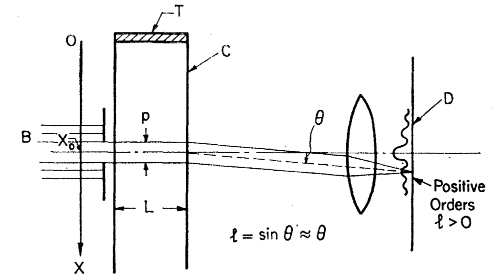


FIG. 1. Experimental arrangement for observing the diffraction effect discussed. The crystal $C$ has a transducer $T$ bonded to it at $x=0$. A beam $B$ of monochromatic light passes through the crystal producing a diffraction pattern $D$.

where $p$ is the width of the light beam, $\mu(x)$ the index of refraction, $L$ the thickness of the crystal, $x_0$ the position of the light beam along the $x$ axis, and $l$ the sine of the angle between the direction of the incident beam and the direction of observation.

The effect of the ultrasonic wave is contained in $\mu(x)$:

$$\mu(x)=\mu_0+\Delta\mu(x)=\mu_0+\Delta\epsilon(x)/2\epsilon^{1/2}. \quad (6)$$

Now $\Delta\epsilon(x)$ is obtained by substituting the solution (2) into Eq. (4) (keeping only terms of order $A^2K^2$) and using the result in Eq. (3):

$$\Delta\epsilon_{33}(x)=-\epsilon^2p_{3311}\left[AK\cos Kx+\frac{A^2K^2a}{4c^2}Kx\sin2Kx+\frac{A^2K^2}{4}\left(1-\frac{a}{2c^2}\right)\cos2Kx\right]. \quad (7)$$

The time dependence has been omitted since the velocity of light is about 5 orders of magnitude larger than the velocity of sound.

If the result Eq. (7) is substituted into Eq. (6) and this result in turn is substituted into the diffraction integral Eq. (5), we have

$$E=\frac{1}{Kp}\int_{K(x_0-p/2)}^{K(x_0+p/2)}\exp i\left[\frac{k}{K}l\xi+k_1L\cos\xi+k_2L\xi\sin2\xi+k_3L\cos2\xi\right]d\xi, \quad (8)$$

where

$$\xi=Kx,\quad k=2\pi/\lambda,\quad k_1=\tfrac{1}{2}(k\epsilon^{3/2}p_{3311}AK),$$
$$k_2=k_1(AKa/4c^2),\quad k_3=k_1(\tfrac{1}{4}AK)(1-a/2c^2). \quad (8')$$

Using the identities

$$e^{iz\sin\varphi}=\sum_{n=-\infty}^{\infty}J_n(z)e^{in\varphi}$$

$$e^{iz\cos\varphi}=\sum_{n=-\infty}^{\infty}J_n(z)i^ne^{in\varphi},$$

Eq. (8) can be rewritten

$$E=\frac{1}{Kp}\sum_{m,n,q=-\infty}^{\infty} i^{m+q}J_m(k_1L)J_q(k_3L)\times\int_{K(x_0-p/2)}^{K(x_0+p/2)} J_n(k_2L\xi)\times\exp i\xi\left[\frac{k}{K}l+m+2n+2q\right]d\xi. \quad (9)$$

Since $p/2\ll x_0$ except when the light beam is very close to the source of sound ($x=0$), and since $J_n(k_2L\xi)$ is a slowly varying function of $\xi$ ($k_2L$ is small), the integral can be approximated by expanding about the point $\xi=Kx_0$:

$$J_n(k_2L\xi)=J_n(k_2LKx_0)+\tfrac{1}{2}k_2L(\xi-Kx_0)\times[J_{n-1}(k_2LKx_0)-J_{n+1}(k_2LKx_0)]+\cdots.$$

The integration in Eq. (9) is then easily performed, and we have

$$E=\sum_{m,n,q=-\infty}^{\infty} i^{m+q}J_m(k_1L)J_q(k_3L)e^{iKx_0\alpha(l,m,n,q)}\times\left\{J_n(k_2LKx_0)\frac{\sin(Kp/2)\alpha(l,m,n,q)}{(Kp/2)\alpha(l,m,n,q)}+ik_2L[J_{n-1}(k_2LKx_0)-J_{n+1}(k_2LKx_0)]\times\left[\frac{\sin(Kp/2)\alpha(l,m,n,q)}{Kp[\alpha(l,m,n,q)]^2}-\frac{\cos(Kp/2)\alpha(l,m,n,q)}{2\alpha(l,m,n,q)}\right]\right\}, \quad (10)$$

where $\alpha(l,m,n,q)=(k/K)l+m+2n+2q$. The diffraction maxima occur when $\alpha(l,m,n,q)=0$. In this case the second term vanishes. The expression for the light amplitude at the diffraction maxima is

$$E_{\max}=\sum_{m,n,q=-\infty}^{\infty} i^{m+q}J_m(k_1L)J_n(k_2LKx_0)J_q(k_3L). \quad (11)$$

The zeroth-order maximum occurs when $l=0$ and $m+2n+2q=0$ and has amplitude

$$E_{\max}{}^{(0)}=\sum_{n,q=-\infty}^{\infty}(-1)^n i^{-q}J_{2n+2q}(k_1L)\times J_n(k_2LKx_0)J_q(k_3L). \quad (12)$$

The $\pm1$st orders occur for $l=\pm K/k$ and $m+2n+2q\pm1=0$, and the corresponding amplitude is

$$E_{\max}{}^{(\pm1)}=\sum_{n,q=-\infty}^{\infty} i^{\mp1-q}(-1)^n J_{\mp1-2n-2q}(k_1L)\times J_n(k_2LKx_0)J_q(k_3L). \quad (13)$$

## IV. NUMERICAL RESULTS

The intensity of light at various diffraction maxima can be calculated from Eq. (11). (Intensity$=|E_{\max}|^2$.) We consider, for example, longitudinal waves in the [100] direction in NaCl. Direct measurements of the third-order elastic constants of NaCl have not been made. However, Hearmon[6] has computed three linear combinations of third-order elastic constants from measurements of the variation of the ordinary elastic constants with hydrostatic pressure.[13] Thus, for NaCl, he finds that $6C_{111}+4C_{112}=-100\times10^{11}$ dyn cm$^{-2}$. If the ratio of $C_{111}$ to $C_{112}$ in NaCl is assumed to be the same as in Ge,[5] then the value of $C_{111}$ can be estimated as $C_{111}=-8.8\times10^{11}$ dyn cm$^{-2}$. The values of the other constants for NaCl needed in Eqs. (8′) are [14–16]

$$p_{3311}=0.178,$$
$$C_{11}=4.87\times10^{11}\text{ dyn cm}^{-2},$$

thus, $a/c^2=-7.87$,

$$\mu=\sqrt{\epsilon}=1.54.$$

Let us choose the following reasonable values for $k$, $K$, and $L$:

$$k=2\pi/\lambda=10^5\text{ cm}^{-1},\quad K=2\pi/\Lambda=126\text{ cm}^{-1},\quad L=2\text{ cm}.$$

In the range of interest the strain amplitude $AK$ is approximately $3\times10^{-5}$. Hence, one obtains $k_3L\simeq10^{-5}$, and in the summation over $q$ in Eq. (11) all terms except the zero-order one are negligible. Using Eq. (13) we have computed the intensity of the first positive and first negative maxima as a function of amplitude $AK$, Fig. 2, and as a function of distance from the source $x_0$, Fig. 3.

For directions of propagation in a cubic crystal other than [100], one simply performs a rotation of axis in both the wave equation and in the relation between the dielectric constant and the strain. The wave equation for longitudinal waves in the [110] or the [111] direction is the same as Eq. (1). The definitions of $c^2$ and $a$, however, are changed as follows: For propagation in the [110] direction we have

$$c^2=(C_{11}+C_{12}+2C_{44})/2\rho,$$
$$a=(3/2\rho)(C_{11}+C_{12}+2C_{44}+C_{111}+C_{112}+C_{166}),$$

and for the [111] direction,

$$c^2=(1/3\rho)(C_{11}+2C_{12}+4C_{44}),$$
$$a=(1/\rho)(C_{11}+2C_{12}+4C_{44})+(2/3\rho)(C_{111}+2C_{112}+C_{144}+2C_{166}+\tfrac{1}{3}C_{123}+\tfrac{2}{3}C_{456}).$$

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[13] D. Lazarus, Phys. Rev. **76**, 545 (1949).
[14] R. S. Krishnan, *Progress in Crystal Physics* (S. Viswanathan, Madras, 1958), Vol. I, p. 128.
[15] Reference 14, p. 80. We use the room-temperature value for $C_{11}$.
[16] *American Institute of Physics Handbook* (McGraw-Hill Book Company, Inc., New York, 1959), pp. 6–23.

The strain optic constants in the rotated coordinate system are derived from

$$p_{ijkl}' = a_{i\alpha} a_{j\beta} a_{k\delta} a_{l\gamma} p_{\alpha\beta\delta\gamma},$$

where the element $a_{ij}$ is the cosine of the angle between the primed $x_i$ axis and the unprimed $x_j$ axis. For the [110] direction this yields

$$p_{3311}' = p_{3311},$$

for the [111] direction,

$$p_{3311}' = \tfrac{1}{3}p_{1111} + \tfrac{2}{3}p_{3311} - \tfrac{2}{3}p_{2323}.$$

The expression for the light amplitude, however, retains the same form.

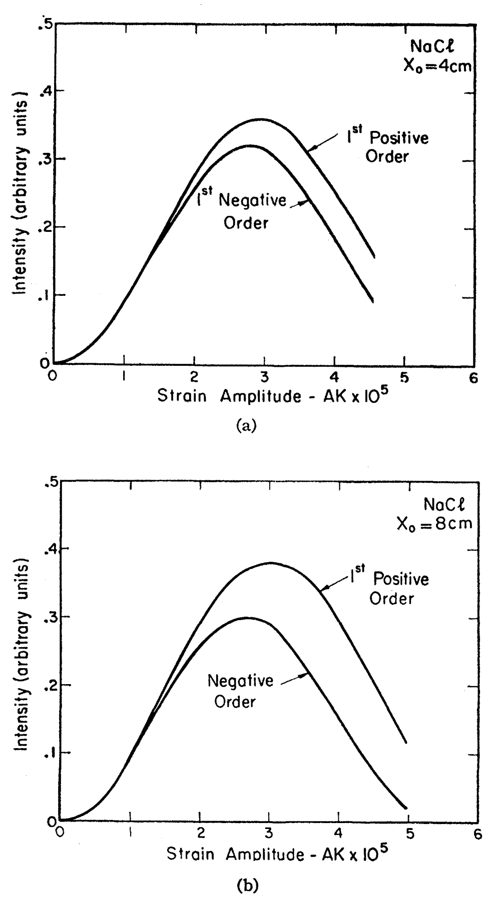


FIG. 2. (a) A plot of the intensity of light in the first positive and negative maxima as a function of strain amplitude for a light beam located 4 cm from the source of sound, i.e., $x_0=4$ cm. The numerical values used are for NaCl. (b) Same as (a) except $x_0=8$ cm.

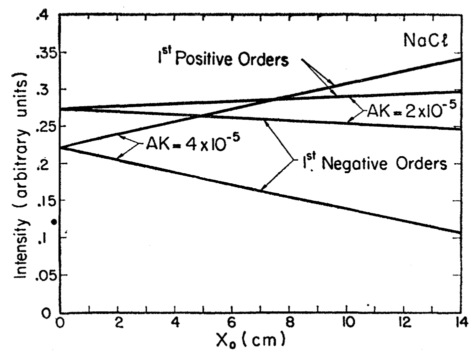


FIG. 3. The intensities of the first positive and negative maxima are plotted as a function of $x_0$, the distance between the light beam and the source of sound, for two values of the strain amplitude.

## V. SUGGESTED EXPERIMENT

To measure the third-order elastic constants with an experimental arrangement such as shown schematically in Fig. 1, a sound wave of sufficient amplitude must be propagated through the crystal. The sound should be sufficiently intense to produce a strain amplitude $AK$ of about $2\times10^{-5}$ or more. The strain amplitude can easily be measured as follows: If the light beam is passed through the crystal near the transducer ($x_0\approx0$), then the ratio of either the first positive or first negative diffraction maximum (in this case they are equal) to the central, zero order, maximum is given by $[J_1(k_1L)/J_0(k_1L)]^2$. This determines $k_1$ $L$, and, since the other constants in the definition (8′) of $k_1$ are known, the strain amplitude $AK$ can be computed.

When a sufficient amplitude is available, plots such as Fig. 2(a) or (b) and Fig. 3 can be obtained experimentally and fitted by Eq. (13) using the arguments of of $J_m$ and $J_n$ as adjustable parameters. This is particularly easy for Fig. 3. The intercept at $x_0=0$ determines $k_1L$, and the slope determines $k_2LK$. These two parameters in turn determine the two unknowns, the strain amplitude $AK$ and the combination of third order of elastic constants.

D. I. Bolef and E. F. Kelly of the Westinghouse Research Laboratories are investigating experimentally the diffraction of light by ultrasound in NaCl and KCl. So far, the experimental picture seems to be roughly what one would expect from our calculations. However, quantitative comparisons have not yet been possible.

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