Piezoelectric Ultrasonic Harmonic Generation in Cadmium Sulfide

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Theory and experiment are presented for the strong acoustic harmonics produced in photoconductive cadmium sulfide as a result of nonlinearities in the electron-lattice interaction responsible for ultrasonic amplification. Data have been taken at fundamental shear-wave frequencies of 11.5 and 30 Mc on a 7-mm crystal and at 30 Mc on a 3-mm crystal. The second-harmonic output was measured as a function of input power at the crossover point (where the electron drift velocity is equal to the sound velocity) for various sample resistivities. The harmonic power is shown to be proportional to the square of the input power, and the largest harmonic obtained was 4 dB below the fundamental for an input acoustic intensity of approximately 1 W/cm^2.

I. INTRODUCTION

STRONG ultrasonic harmonic generation has been observed in photoconductive cadmium sulfide. The mechanism which produces the harmonics is the nonlinearity of higher order terms in the electron-lattice interaction responsible for ultrasonic amplification.[1] The second harmonic power can be as large as 4 dB below the fundamental for an input acoustic intensity of approximately 1 W/cm^2. The data have been taken at crossover (where the electron drift velocity is equal to the sound velocity) in order to simplify the interpretation of the experimental results. At crossover, the change in the amplitude of the fundamental during propagation down the crystal is due solely to losses to higher harmonics while at other drift voltages the fundamental and the harmonics are amplified or attenuated at different rates. However, dispersion as a function of frequency is a maximum here, giving rise to coherence phenomena similar to the optical harmonic case.[2]

The possibility of achieving large harmonic generation and parametric interaction of acoustic waves in piezoelectric semiconductors was first realized by Hutson.[3] Harmonic generation in CdS has recently been reported by Ishiguro, Uchida, and Suzuki,[4] Tell,[5] and Kroger.[6]

The physical situation is that the self-consistent field produced by the interaction of the electrons with the traveling wave contains higher harmonics which, since the crystal is strongly piezoelectric, give rise to large strains at the harmonic frequencies.

In the original explanation offered by Hutson[3] in regard to the saturation of acoustic flux, the space-charge wave at one frequency would interact with the piezoelectric field at a second frequency giving rise to strains at the sum and difference frequencies. This interaction would take place by virtue of the nonlinearities in the electron lattice interaction. In particular, the nonlinearities arise from the term $(\partial/\partial X)\{E(\partial D/\partial X)\}$ in Eq. (2) below. This term is omitted in the ordinary treatment of the ultrasonic amplification process.[1,7,8]

We have, in effect, an equivalent third-order elastic constant which can be larger than 10^{14} dyne/cm^2 compared to the usual lattice anelastic constants of 10^{11} to 10^{12} dyne/cm^2.[9]

II. THEORY

Following Hutson,[3] the electric field E and the strain S are expanded in Fourier series

$$E=E_0+\sum_m E_{m\omega}\sin(k_mX-m\omega t+\varphi_{Em}), \quad (1a)$$

$$S=\sum_m S_{m\omega}\sin(k_mX-m\omega t+\varphi_{Sm}), \quad (1b)$$

where E_0 is the applied drift field. Using Eq. (7) in White[7] with the notation previously used[7]

$$-\frac{\partial^2 D}{\partial X\partial t}=\mu\frac{\partial}{\partial X}\left\{\left(qn_0-\frac{\partial D}{\partial X}\right)E\right\}-D_n\frac{\partial^3 D}{\partial X^3}, \quad (2)$$

where D is the displacement vector, μ the mobility, and D_n the diffusion constant. Substituting for $D=\epsilon E+eS$ (ϵ is the dielectric constant and e the piezoelectric constant) and solving for $E_{m\omega}$ in terms of $S_{m\omega}$ at crossover, with the assumptions that the second harmonic is small compared to the fundamental, higher harmonics are negligible, and dispersion can be neglected in the above equation yields

$$E_\omega=-\frac{e}{\epsilon}\frac{\omega/\omega_D}{(\omega_c/\omega+\omega/\omega_D)}S_\omega \quad (3)$$

and

$$E_{2\omega}=-\frac{e}{\epsilon}\frac{2\omega/\omega_D}{(\omega_c/2\omega+2\omega/\omega_D)}S_{2\omega}+\frac{\frac{1}{4}f(\mu e^2/V_0\epsilon^2)(\omega_c/\omega)(\omega/\omega_D)}{(2\omega/\omega_D+\omega_c/2\omega)(\omega/\omega_D+\omega_c/\omega)^2}S_\omega^2, \quad (4)$$

[1] A. R. Hutson, J. H. McFee, and D. L. White, Phys. Rev. Letters **7**, 237 (1961).
[2] P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963).
[3] A. R. Hutson, Phys. Rev. Letters **9**, 296 (1962).
[4] T. Ishiguro, I. Uchida, and T. Suzuki, 1964 IEEE International Convention (unpublished).
[5] B. Tell, Bull. Am. Phys. Soc. **9**, 478 (1964).
[6] H. Kroger, Appl. Phys. Letters **4**, 150 (1964).
[7] D. L. White, J. Appl. Phys. **33**, 2547 (1962).
[8] A. R. Hutson and D. L. White, J. Appl. Phys. **33**, 40 (1962).
[9] J. M. Ziman, *Electrons and Phonons* (Oxford University Press, New York, 1960), p. 152.

where $\omega_c=\sigma/\epsilon$ is the conductivity frequency and $\omega_D=V_0^2/D_n$ is the diffusion frequency. Substituting for E in the wave equation [Eq. (2) from White[7]]

$$\rho\frac{\partial^2 u}{\partial t^2}=c\frac{\partial^2 u}{\partial X^2}-e\frac{\partial E}{\partial X} \tag{5}$$

gives

$$\rho(\partial^2 u_\omega/\partial t^2)=c_\omega(\partial^2 u_\omega/\partial X^2)=0 \tag{6}$$

where c_ω is defined as

$$c\left[1+\frac{e^2}{c\epsilon}\left(\frac{\omega/\omega_D}{\omega_c/\omega+\omega/\omega_D}\right)\right]$$

and

$$\rho\frac{\partial^2 u_{2\omega}}{\partial t^2}-c_{2\omega}\frac{\partial^2 u_{2\omega}}{\partial X^2}=-\frac{f\mu e^3}{2V_0\epsilon^2}\times\frac{(\omega_c/\omega)(\omega/\omega_D)}{(\omega_c/\omega+\omega/\omega_D)^2(\omega_c/2\omega+2\omega/\omega_D)}\frac{\partial u_\omega}{\partial X}\frac{\partial^2 u_\omega}{\partial X^2}, \tag{7}$$

where

$$c_{2\omega}=c\left[1+\frac{e^2}{\epsilon c}\frac{2\omega/\omega_D}{(\omega_c/2\omega+2\omega/\omega_D)}\right].$$

$(\partial u_\omega/\partial X)(\partial^2 u_\omega/\partial X^2)$ is the driving term for the second harmonic, and its coefficient is an effective third-order elastic constant which we define as $c_{\mathrm{III}}^{\mathrm{e.l.}}$. The superscript e.l. denotes electron-lattice to distinguish it from the usual lattice anelastic constant c_{III}.

The solution to these equations with the boundary condition that $u=A\sin(-\omega t)$ at $X=0$ is

$$u=u_\omega+u_{2\omega},$$

$$u=A\sin(k_1X-\omega t)-\frac{A^2k_1}{8}\left(\frac{c_{\mathrm{III}}^{\mathrm{e.l.}}}{c}\right)\times\frac{\sin[(\omega X/V_0)(V_{2\omega}-V_\omega)/V_0]}{(V_{2\omega}-V_\omega)/V_0}\cos(k_2X-2\omega t). \tag{8}$$

This approximation is valid for

$$\frac{Ak_1}{8}\left(\frac{c_{\mathrm{III}}^{\mathrm{e.l.}}}{c}\right)\frac{\sin[(\omega X/V_0)(V_{2\omega}-V_\omega)/V_0]}{(V_{2\omega}-V_\omega)/V_0}\ll 1.$$

In terms of the acoustic intensities $P_\omega=\frac{1}{2}c_\omega V_\omega(k_u u_\omega)^2$ and $P_{2\omega}=\frac{1}{2}c_{2\omega}V_{2\omega}(k_{2\omega}u_{2\omega})^2$ and here taking $c=c_\omega=c_{2\omega}$, $V_\omega=V_{2\omega}=V_0$ and $k_{2\omega}=2k_\omega$, the final result is

$$P_{2\omega}=\frac{P_\omega^2}{8V_0c}\left(\frac{c_{\mathrm{III}}^{\mathrm{e.l.}}}{c}\right)^2\frac{\sin^2[(\omega X/V_0)(V_{2\omega}-V_\omega)/V_0]}{[(V_{2\omega}-V_\omega)/V_0]^2}. \tag{9}$$

The harmonic consists of a wave which is locked to the fundamental which therefore exists only in presence of the driving term and has frequency 2ω and propagation vector $2k_1$. There is also a free wave of frequency 2ω and propagation vector k_2 which is a solution to the homogeneous equation. The sum of the two solutions, obeying the boundary condition, results in the form obtained in Eq. (8). At crossover, the dispersion is given by

$$V_{m\omega}=\left(\frac{c_{m\omega}}{\rho}\right)^{1/2}=V_0\left[1+\frac{K^2}{2}\frac{1}{1+(\omega_c\omega_D/m^2\omega^2)}\right], \tag{10}$$

with $V_0=(c/\rho)^{1/2}$ and $K^2=e^2/c\epsilon$ is the electromechanical coupling constant. This dispersion is the result of the difference in the screening, at different acoustic frequencies, of the longitudinal electric field which accompanies the sound wave in a piezoelectric crystal.[7,8] Equation (9) exhibits the coherence effects similar to optical harmonics,[2] and reduces to the result obtained by Melngailis *et al.*[10] for the usual anelastic case in which there is negligible dispersion.

III. EXPERIMENT

The experimental arrangement was similar to that of Hutson, McFee, and White. Data were taken at crossover for various light levels at the fundamental shear wave frequencies of 11.5 and 30 Mc on a 7-mm crystal and at 30 Mc on a 3-mm crystal. The crossover voltage was determined at low input power levels for each resistivity. The fundamental and second harmonic were then measured as a function of input power at the low input crossover voltage.

The harmonics could be reduced by greater than 30 dB by removing the illumination and returning the crystal to its insulating state. This appears to establish the presence of the harmonics as due to the electron-lattice interaction and not to other effects which should not be appreciably affected by illumination. Furthermore, working at crossover eliminates the possibility that a small spurious signal could be amplified.

The low input level was chosen for determining the crossover point because it was felt that in the absence of nonlinearities the crossover should be independent of the power level. In the dark or insulating condition, the fundamental output power was linearly proportional to the input up to the highest power used, whereas in the illuminated state the output deviated from its dark value at high input levels. The depletion of the fundamental in the conducting state appears to be a further check on the origin of the harmonics.

The major experimental problem was the measurement of absolute acoustic intensities which is necessary in order to determine the effective anelastic constants. The absolute intensity can be determined from insertion loss measurements if the input and output terminals of the system are electrically and acoustically symmetric. Identical transducers were used for transmitter and receiver, and were inductively tuned and then shunted by a 100-Ω resistor which made the input and output

[10] J. Melngailis, A. A. Maradudin, and A. Seeger, Phys. Rev. 131, 1972 (1963).

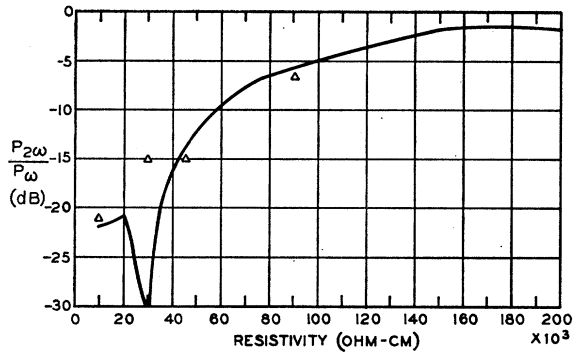

FIG. 1. $\frac{P_{2\omega}}{P_\omega}=P_\omega\left[\frac{1}{8V_0c}\left(\frac{c_{III}^{e\cdot l\cdot}}{c}\right)^2\frac{\sin^2[(\omega X/V_0)(V_{2\omega}-V_\omega)/V_0]}{[(V_{2\omega}-V_\omega)/V_0]^2}\right]$ at 30 Mc for $P_\omega=0.18$ W/cm² as a function of sample resistivity on 7-mm crystal.

impedances effectively 100 Ω. The resistors considerably increased the insertion losses, but made the transmitting and receiving efficiency of the transducers equal and also matched the transmitting transducer to the rf pulse generator. The ratio of the input to output power (insertion loss) was determined by inserting an attenuator with 100-Ω characteristic impedance in place of the sample while maintaining the same input voltage and output signal level, and the input power was determined by measuring the voltage across the 100-Ω terminating resistor. The acoustic intensity was then found by assuming all of the electrical power was absorbed in the terminating resistors, and therefore the acoustic intensity is the input power diminished by half of the insertion loss. Allowing for some electrical and acoustic asymmetry (which can be estimated by flipping the sample) the acoustic intensities are believed reliable to within a factor of 3.

In order to check for harmonics which may exist in the electrical input, a band pass filter, centered at the fundamental, was sometimes used. However, checking

TABLE I. Effective third-order elastic constants.[a]

$$c_{III}^{e\cdot l\cdot}=\frac{f\mu e^3}{2V_0\epsilon^2}\frac{(\omega_c/\omega)(\omega/\omega_D)}{(\omega_c/\omega+\omega/\omega_D)^2(\omega_c/2\omega+2\omega/\omega_D)}$$

Resistivity (Ω-cm)	$c_{III}^{e\cdot l\cdot}$ (dyne/cm²)
11.5 Mc/sec	
30 000	4.8×10^{12}
60 000	1.7×10^{13}
90 000	3.4×10^{13}
180 000	1.0×10^{14}
270 000	1.8×10^{14}
30 Mc/sec	
10 000	8.3×10^{12}
30 000	4.9×10^{13}
90 000	1.6×10^{14}
200 000	3.2×10^{14}
270 000	2.4×10^{14}

[a] The values of the constants used in the calculations are $\omega_c=1.25\times10^{12}$ $\sigma(\Omega^{-1}\text{ cm}^{-1})$, $\omega_D=6.0\times10^9$ sec^{-1}, $\epsilon=0.8$ cgs, $e=6.5\times10^4$ cgs, and $c=1.5\times10^{11}$ dyne/cm² and $\mu=200$ cm²/V sec.

TABLE II. The ratio of fundamental to second harmonic power for the maximum fundamental intensity is given in dB.

Resistivity (K Ω-cm)	Experimental (dB)	Theoretical (dB)
7 mm—30 Mc/sec Maximum fundamental intensity is 0.85 W/cm²		
10	18	15
30	9	[a]
90	4	0
7 mm—30 Mc/sec Maximum fundamental intensity is 0.18 W/cm²		
10	21	22
30	14	[a]
45	15	15
90	7	6
270	11	5
3 mm—30 Mc/sec Maximum fundamental intensity is 0.13 W/cm²		
90	18	8
270	14	2
7 mm—11.5 Mc/sec Maximum fundamental intensity is 0.25 W/cm²		
30	25	28
60	17	19
90	14	14
180	19	6
270	21	1

[a] Theory predicts the second harmonic should be zero.

the dark value of the harmonic (where the interaction was turned off) proved the filter to be unnecessary. It was also pointed out to the author by White that a sine-wave incident on a nonlinear medium will in general reflect a harmonic, thereby changing the boundary condition used in deriving Eq. (8). However, calculation of the reflected harmonic at the interface between quartz and cadmium sulfide leads to the belief that this effect should be negligible.[11] This calculation does, however, neglect any effects due to the acoustic bonds.

Another cause of experimental error could be inhomo-

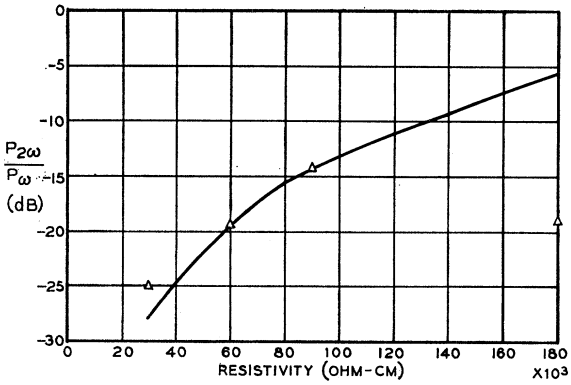

FIG. 2. $\frac{P_{2\omega}}{P_\omega}=P_\omega\left[\frac{1}{8V_0c}\left(\frac{c_{III}^{e\cdot l\cdot}}{c}\right)^2\frac{\sin^2[(\omega X/V_0)(V_{2\omega}-V_\omega)/V_0]}{[(V_{2\omega}-V_\omega)/V_0]^2}\right]$ at 11.5 Mc for $P_\omega=0.25$ W/cm² as a function of sample resistivity on 7-mm crystal.

[11] N. Bloembergen and P. S. Pershan, Phys. Rev. 128, 606 (1962).

geneities in the crystal such as a nonuniform distribution of trapping centers and in the illumination which might smear out coherence effects. Furthermore at low-carrier concentrations trapping effects may become important,[4] thereby partly accounting for the discrepancy between theory and experiment at high resistivities.

IV. RESULTS

The effective anelastic constants $c_{III}{}^{e.l.}$ are given as a function of frequency and resistivity in Table I. In Table II, the experimental results are compared with the theory as given by Eqs. (9) and (10), and some of the results are plotted against the theoretical curve in Figs. 1 and 2. It is seen from Table I and Figs. 1 and 2 that the general fit is good except where coherence effects should produce a null in the harmonic output and at high resistivities. The causes of these discrepancies are not entirely clear, although possible reasons have been mentioned at the end of the preceding section.

The plot of fundamental and harmonic power against input power for the case of maximum harmonic production is given in Fig. 3. It is seen from this figure that both the fundamental and harmonic deviate at high power from their low power slopes. The deviation becomes significant for inputs greater than 25 dB ($\approx$0.12 W/cm^2). The criteria for the validity of the theory as given by Eq. (8) was

$$\frac{Ak_1}{8}\frac{c_3}{c}\frac{\sin[(\omega X/V_0)(V_{2\omega}-V_\omega)/V_0]}{(V_{2\omega}-V_\omega)/V_0}\ll 1.$$

At 0.12 W/cm^2 this quantity is approximately 0.2, so that for lower input levels or cases of less harmonic production, the theory should be valid.

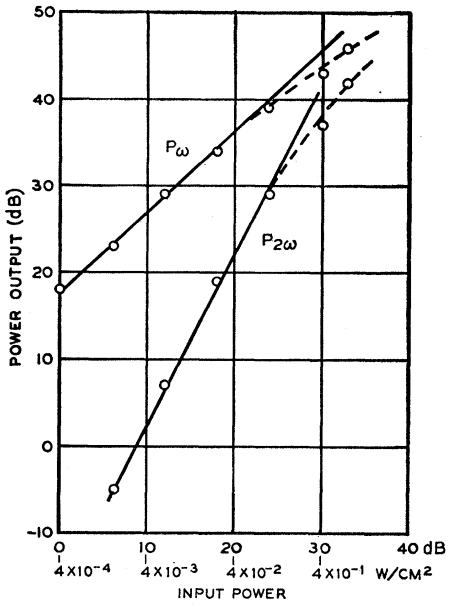

FIG. 3. Fundamental and harmonic intensities as a function of input intensity at 30 Mc and 90-k Ω-cm on a 7-mm crystal.

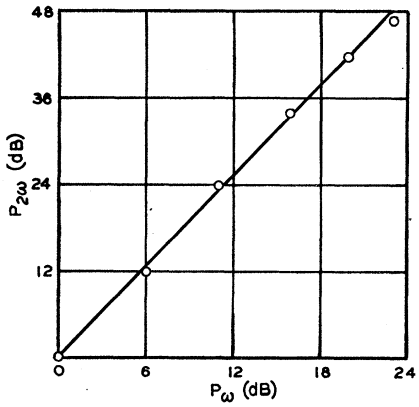

FIG. 4. Graph showing $P_{2\omega}$ proportional to $P_\omega{}^2$.

In Fig. 4, it is shown that the harmonic power is proportional to the square of the fundamental power even at the highest power level.

V. CONCLUSIONS

This work has given an absolute measurement of the electron-lattice nonlinearity as originally predicted by Hutson.[3] A theory, in terms of equivalent third-order elastic constants, has been developed which is in reasonable agreement with the experimental data except for the above mentioned discrepancies. It is felt that the unambiguous appearance of coherence effects and agreement at high resistivities would require work on select samples of various lengths under extremely homogeneous lighting conditions, which may be beyond our present technology.

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