FINITE-AMPLITUDE ELASTIC WAVES IN SOLIDS AND DEVIATIONS FROM HOOKE'S LAW

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It is shown that when longitudinal ultrasonic waves of sufficient intensity (alternating pressures of the order of 1 atm) are propagated in solids, the originally sinusoidal wave becomes distorted. The second harmonic increases with the distance from the radiator, passes through a maximum, and then decreases owing to dissipative losses, as in the case of propagation of finite-amplitude elastic waves in liquids. The distortion is mainly due to the asymmetry of the potential well, i.e., the nonlinearity of the molecular interaction which leads to anharmonicity of the lattice. By measuring the ratio of the pressure amplitudes of the second harmonic and the fundamental frequency we can determine the ratio of the coefficients of the quadratic and linear terms in the equation of state of the solid. This ratio which is measured here dynamically using the distortion of the waveform, agrees quite well with the ratio obtained from static measurements under hydrostatic pressure.

1. INTRODUCTION

NONLINEAR phenomena occurring when strong ultrasonic waves are propagated in gases and liquids have been studied quite intensively. Owing to the nonlinearity of the equations of motion and of the equation of state of gases and liquids, a weak periodic shock wave may form under some conditions, nonlinear interaction of finite-amplitude waves may occur, combination frequencies may appear, and other nonlinear phenomena may be observed (see, for example, [1]).

There has not yet been a single serious attempt to find similar nonlinear effects for solids, and the authors are not aware of any work reporting observations of such effects.

The authors have briefly reported[2] that nonlinear phenomena in solids with low attenuation of ultrasonic waves are quite strong, owing to a characteristic cumulative effect with the same mechanism as in gases and liquids, even for longitudinal ultrasonic waves of low intensity. Later it was shown[3] that such effects are mainly due to the asymmetry of the potential well, i.e., due to the nonlinearity of the intermolecular interaction which leads to anharmonic properties of the crystal lattice. In the present paper we shall give a more detailed description of the experimental results for the propagation of finite-amplitude ultrasonic waves in solids, and shall show that by measuring the ratio of the pressure amplitudes of the second harmonic and the fundamental frequency

we can determine the ratio of the coefficients of the quadratic and linear terms in the equation of state for a solid; this ratio is in good agreement with the ratio obtained from static measurements of the hydrostatic compressibility reported in numerous papers by Bridgman and his followers.

2. EXPERIMENTAL APPARATUS AND METHOD OF MEASURING THE SECOND HARMONIC

A spectroscopic method was used to determine the harmonics of a finite-amplitude elastic wave; the harmonics were selected using resonance piezoelectric transducers and a resonance amplifier. This method is very sensitive to small distortions of the waveform; it permits measurement of the pressure amplitude of the second harmonic down to several hundredths of one per cent of the fundamental-frequency amplitude.

The experimental apparatus is in principle very simple. A square-wave generator modulates a radio-frequency oscillator and triggers simultaneously an oscilloscope sweep. Radio pulses with a carrier frequency of 5 Mc are applied to an X-cut quartz plate with the same natural frequency; the silvered part of this plate is 16 mm in diameter. Acoustical contact with the polished end of the test rod of 16 mm diameter is obtained via a thin layer of transformer oil. A quartz receiver plate of 10 Mc natural frequency is located at the other end of the test rod. The voltage from the receiver plate is applied to a band-elimination filter tuned to the

fundamental frequency of 5 Mc. This filter attenuates the 5 Mc fundamental frequency by a factor of 150—200 without affecting greatly the 10 Mc signal. From the band-elimination filter the voltage is applied to the input of a resonance amplifier with a gain of the order of 10^3 (at the 10 Mc frequency) and the signal from the amplifier output is fed to a cathode-ray oscilloscope. The pulse method is used to avoid the effect of standing waves; the pulse duration is 10—50 μsec, the repetition frequency is 0.2—1 kc.

The experiments showed that apart from the 5 Mc fundamental frequency there was also a signal at 10 Mc (the second harmonic). A series of control experiments proved that the second harmonic was not due to secondary effects (overloading of the amplifier by the fundamental-frequency signal, nonlinear distortion factor of the generator, etc.) but was the result of waveform distortion during propagation in the solid. The increase of amplitude of the second harmonic with distance from the radiator is an even more convincing proof of the presence of nonlinear effects in solids.

Figure 1 gives the dependence of the voltage amplitude of the second harmonic (which is proportional to the acoustic pressure produced by this harmonic) on the distance from a radiator for the magnesium-aluminum alloy MA-8 [1) when the voltage across the quartz radiator was 1000 V (peak value); the second-harmonic amplitude was obtained from the first of the reflected pulses for rods of various lengths (with the other experimental conditions unchanged), and from the second and third pulses in the case of rods of 30 and 45 cm length. The curve in Fig. 1 shows that the amplitude of the second harmonic gradually increases,
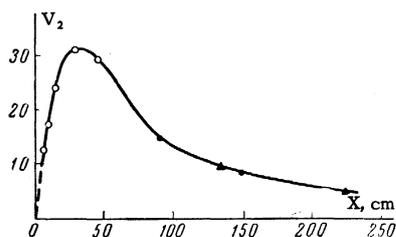
FIG. 1. Dependence of the pressure amplitude (the voltage V_2 on the receiver plate, in mV) of the second harmonic (10 Mc) on the distance from the radiator in the alloy MA-8 with 1000 V (peak) across the radiator: o — from the amplitude of the first pulse for rods of various lengths; ● — from the second and third pulses for a rod of 30 cm length; ▲ — from the second and third pulses for a rod of 45 cm length.

1)The alloy MA-8 has sufficient uniformity and a small absorption coefficient for ultrasound (4-5 dB/m at 5 Mc).

reaches a maximum at a distance of 35 cm (the stabilization distance), and then decreases, owing to dissipation of its energy. Measurements have shown that at the maximum (at the stabilization distance) the second-harmonic amplitude amounts to 2% of the fundamental-frequency amplitude. A similar curve was obtained for the behavior of the second harmonic in duraluminum rods.

The nature of the dependence of the second harmonic on distance shows that a nonexponential series of reflected pulses should be observed in sufficiently short samples (samples of lengths which are considerably smaller than the stabilization distance L_{st} of the second harmonic). This did in fact occur. Figure 2 shows a photograph of a series of 10-Mc pulses (for a fundamental frequency of 5 Mc) in an MA-8 alloy rod of 7.5 cm length and 16 mm diameter (the peak voltage across the radiator was 1000 V). The envelope of the reflected pulses is affected somewhat by the lack of parallelism of the rod ends and by the nonuniformity of contact with the receiver. A check of this effect was obtained by applying small-amplitude pulses of 10 Mc carrier frequency to the same sample and recording them with the same receiver. The oscilloscope screen then showed, as usual, a series of pulses decreasing exponentially in amplitude (Fig. 3).

Figure 2 shows that the third pulse, which traveled a distance of 37.5 cm, had the maximum amplitude. This result is in good agreement with the stabilization distance determined from measurement of the first pulse in samples of various length (Fig. 1).

3. DETERMINATION OF NONLINEAR CORRECTIONS TO HOOKE'S LAW FROM THE WAVEFORM DISTORTION

From the macroscopic point of view the cause of the formation of harmonics in a wave of finite amplitude is the nonlinearity of the equation of state of the solid and the geometrical characteristics of finite deformations. Mathematically the first effect leads to the appearance of terms containing cubes of the deformation tensor components u_{ik}, in addition to squares of these components, in the expression for the elastic energy E per unit volume of an isotropic solid. This expression has the form [4]

$$E = \mu u_{ik}^2 + \left(\tfrac{1}{2}K - \tfrac{1}{3}\mu\right) u_{ll}^2 + \tfrac{1}{3} A u_{ik} u_{il} u_{kl}$$
$$+ B u_{ik}^2 u_{ll} + \tfrac{1}{3} C u_{ll}^3. \tag{1}$$

Here μ is the shear modulus, K is the bulk modu-
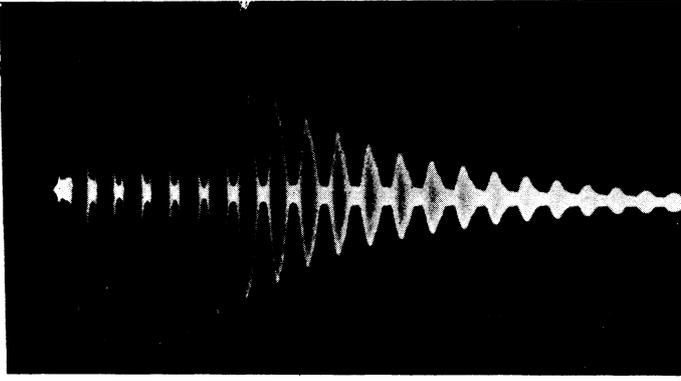
FIG. 2. Series of second-harmonic (10 Mc) pulses arising and then decaying in amplitude in an MA-8 rod of 7.5 cm length. The voltage across the radiator (5 Mc) was 1000 V (peak).
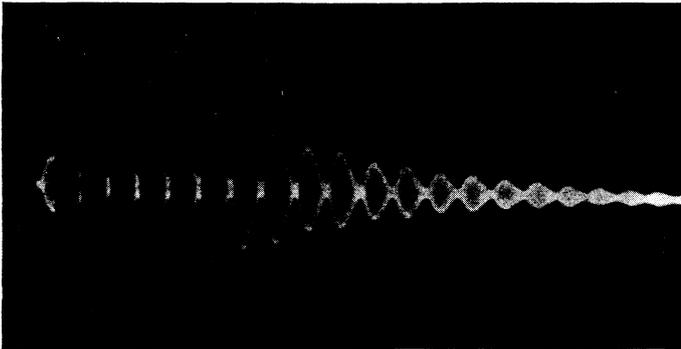
FIG. 3. Series of pulses exponentially decaying in amplitude for the same sample as in Fig. 2 but when 30 V were applied to a radiator plate with a natural frequency of 10 Mc; the signal was received using a plate with the same natural frequency 10 Mc.

lus for hydrostatic compression, and A, B, C are some scalar coefficients [2].

The second effect is of the nature of a "geometrical nonlinearity," manifesting itself by a nonlinear relationship between the deformation tensor components u_{ik} and the derivatives of the deformation vector components u_i with respect to the coordinates x_k:

$$u_{ik} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_k} + \frac{\partial u_k}{\partial x_i} + \frac{\partial u_l}{\partial x_i}\frac{\partial u_l}{\partial x_k}\right). \tag{2}$$

In the case of deformation by unidirectional compression and dilatation [3] along the x axis, which occurs when a plane longitudinal wave is propagated along the x direction, we have

$$u_x(x, t) \neq 0, \qquad u_y = u_z = 0, \tag{3}$$

and consequently among all the deformation tensor components only $u_{xx} \neq 0$. Using the conditions in Eq. (3) we can obtain an expression for the "stress" σ_{xx} [4]:

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[2] As usual, the double repeated subscripts denote summation over values 1, 2 and 3.

[3] The unidirectional compressional (dilatational) deformation means the compression (dilatation) of a rod whose lateral surfaces do not move.

[4] Strictly speaking the quantity σ_{xx} defined in this way is not a stress; this is related to the difference of the coordinates of points in a body before and after deformation. [4]

$$\sigma_{xx} = \frac{\partial E}{\partial(\partial u_x/\partial x)} = \beta\frac{\partial u_x}{\partial x} + \gamma'\left(\frac{\partial u_x}{\partial x}\right)^2, \tag{4}$$

$$\beta = K + \tfrac{4}{3}\mu, \qquad \gamma' = \gamma + 3\beta, \qquad \gamma = A + 3B + C. \tag{5}$$

The nonlinear coefficient γ' consists of two components: a coefficient γ corresponding to the nonlinearity of the equation of state (the "physical nonlinearity," according to [5]) and a coefficient 3β which allows for the "geometrical nonlinearity."

As will be shown later, we are interested in the ratio of the coefficient γ (which we shall call the nonlinear coefficient) and the coefficient in front of the linear term in Eq. (4) (the linear coefficient):

$$\gamma/\beta = (A + 3B + C)/(K + \tfrac{4}{3}\mu). \tag{6}$$

This ratio (and therefore the coefficient γ, because β is known) can be found from measurements of the second-harmonic amplitude. The equation of motion for a plane longitudinal wave in an isotropic medium without allowance for dissipation and including quadratic terms has the form [6]

$$\rho_0\frac{\partial^2 u_x}{\partial t^2} - \beta\frac{\partial^2 u_x}{\partial x^2} = \delta\frac{\partial^2 u_x}{\partial x^2}\frac{\partial u_x}{\partial x}, \tag{7}$$

where ρ_0 is the density of the undeformed body, and

$$\delta = 3\beta + 2\gamma. \tag{8}$$

The solution of the equation of motion shows that the originally sinusoidal wave becomes dis-

torted upon propagation and the pressure amplitude of the second harmonic is given by

$$p_2 = \left(n\omega p_{01}^2 / 2\rho_0 c_l^3\right) x, \qquad n = -\left(^3/_2 + \gamma/\beta\right), \qquad (9)$$

where ω is the angular frequency, p_{01} is the pressure amplitude of the fundamental frequency at the radiator, x is the distance from the radiator, and c_l is the velocity of propagation of longitudinal waves in an infinite medium.

Using the value of n in Eq. (9) we find that

$$\frac{\gamma}{\beta} = -\left[\frac{2\rho_0 c_l^3}{\omega p_{01} x} \frac{p_2}{p_{01}} + \frac{3}{2}\right]. \qquad (10)$$

The above formula applies, strictly speaking, to media where there is no dissipation of energy. However, it can be used for dissipative media over small distances (smaller than the stabilization distance) such that the attenuation is still small. Thus, by measuring the ratio of the amplitudes of the second harmonic and the fundamental frequency at a distance shorter than the stabilization distance, we can determine γ/β. The value of p_{01} can be calculated by finding the intensity of radiation from a piezoelectric quartz plate with a given radio-frequency voltage applied to it. This intensity was found from the formula for the radiation from one side of a piezoelectric quartz plate[7] (this corresponds to the structure of the contact between the plate and the sample). This formula has been checked experimentally for liquids; although its applicability to radiation in a solid is self-evident, the numerical coefficient in the formula may be somewhat different[5]. With a peak voltage of 1000 V across a quartz plate the alternating 5-Mc pressures amount, according to this calculation, to ±6 atm for a single crystal of aluminum.

The results of measurements of the ratio γ/β for some cubic single crystals and for the alloy MA-8, obtained using Eq. (10) with the measured values of p_2/p_{01} substituted and from the formula for the intensity of radiation p_{01} from one side of a piezoelectric plate, are listed in the table.

From experimental measurements of the second harmonic in a plane longitudinal elastic wave we obtain, according to Eq. (6), the nonlinear coefficient γ, which is a linear combination of three (for an isotropic body) nonlinear coefficients A, B, C. For cubic crystals there are, in general, six coefficients in Hooke's law for quadratic deformations.

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[5] We are now investigating this problem.

Ratios of the nonlinear and linear coefficients in Hooke's law found from measurements of the waveform distortion (γ/β) and from measurements of hydrostatic compressibility (γ_1/β_1)

Solid	$-\gamma/\beta$	$-\gamma_1/\beta_1$
Al (single crystal)	7	6.9
NaCl	9	7.4
KCl	6.5	6.2
LiF	6.5	8.7
Magnesium-aluminum alloy MA-8	4.3	—

Note. In single crystals the direction of wave propagation coincided with the crystallographic axis.

4. COMPARISON WITH BRIDGMAN'S DATA

In order to compare our dynamically obtained values of γ/β with the results of Bridgman's static measurements, we must bear the following in mind.

It is known that the relationship between stress and deformation depends strongly, even in the first approximation, on the conditions at lateral surfaces; consequently the coefficient of the quadratic term depends on the type of deformation. From our experiments one can determine the nonlinear coefficient γ for the case of unidirectional compression—dilatation; this type of deformation occurs upon propagation of a plane longitudinal wave in an infinite medium, represented in our case by the rod whose transverse dimensions are considerably greater than the wavelength.

On the other hand Bridgman et al investigated the behavior of solids under uniform omnidirectional (hydrostatic) compression. Bridgman[8,9] showed that for the majority of solids at pressures P up to several tens of thousands of atmospheres the dependence of the relative change of volume on the pressure under uniform hydrostatic compression is satisfactorily given by the formula

$$-\Delta V/V_0 = aP - bP^2, \qquad (11)$$

where a and b are temperature-dependent coefficients characteristic of a given substance; a = 1/K.

In the case of linear deformation under uniform hydrostatic compression we obtain the formula, accurate to the quadratic term

$$P = \beta_1 \Delta l/l_0 + \gamma_1 (\Delta l/l_0)^2, \qquad (12)$$

where $\Delta l/l_0$ is the relative change of length, β_1 is the linear coefficient, γ_1 is the nonlinear coefficient for this type of deformation, and

$$\gamma_1/\beta_1 = -(3b/a^2 - 1). \qquad (13)$$

The table lists values of γ_1/β_1 calculated from the data of Bridgman and Slater for coefficients a and b taken from [10].

We cannot expect better agreement between γ/β and γ_1/β_1 than that in the table because the natures of the deformations in our case and in Bridgman's case were different.

Moreover, the precision of the measurements of γ/β was not high, bearing in mind that we used a formula for the radiation from one side of a piezoelectric quartz plate into a solid to calculate p_{01}; the error, according to our estimates, amounted to 20—30%. Increase of the accuracy of measurement and the possibility of three independent experiments should allow us in future to determine separately the three coefficients A, B, and C for an isotropic body.

In the case of liquids, measurement of the ratio of the nonlinear coefficient B/2 in the equation of state for liquids and the linear coefficient A (analogous to our γ/β) by a dynamic method (from nonlinear distortions of the elastic waveform) and by a static method gave results which in general agreed satisfactorily (see [1]), as they did in our case for solids.

5. CONCLUDING REMARKS

The table lists the values of γ/β for solids in which attenuation is weak and the stabilization distance sufficiently large. For solids with stronger attenuation we can only indicate the per cent content of the second harmonic taken with respect to the first at some distance from the radiator. Thus, for a Plexiglas rod at a distance of 5 cm from the radiator this ratio amounts to 0.2% (because of strong absorption of sound at $L_{st} < 5$ cm in Plexiglas) while for polystyrene at the same distance this ratio is 5%. An even higher ratio is obtained for a slab of 45° X-cut Rochelle salt (for an ultrasonic wave propagated at right angles to the X axis). In solids with strong nonlinearity (polystyrene, Rochelle salt, etc.) the third harmonic is easily observed. When two longitudinal waves of different frequency are propagated simultaneously along the same direction, combination (sum and difference) frequencies are observed, i.e., there is parallel interaction.

So far we have studied in detail only one type of nonlinear interaction for the case of longitudinal waves (distortion of the waveform). However, other types of interaction connected with transverse waves are possible; we are at present investigating them. The notion of phonon-phonon interactions (or nonlinear interactions of elastic waves in solids) has been, following Debye, very widely used as the basis of theories of various transport phenomena (including, for example, the absorption of sound in ideal crystals) and is among the principal ideas of solid-state physics. However, as far as we know, there has not yet been any direct experimental proofs of the occurrence of such interactions. For this reason the proof of the existence of nonlinear interactions of elastic waves in solids (so far for longitudinal waves at ultrasonic frequencies only) is of interest and the technique developed here can be used for further studies in the same field.

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[1] L. K. Zarembo and V. A. Krasil'nikov, UFN 68, 687 (1959), Soviet Phys. Uspekhi 2, 580 (1960).

[2] V. A. Krasil'nikov and A. A. Gedroïts, Vestnik, Moscow State Univ. ser. III 2, 92 (1962).

[3] Gedroïts, Zarembo, and Krasil'nikov, ibid. 3, 92 (1962).

[4] L. D. Landau and E. M. Lifshitz, Mekhanika sploshnykh sred (Mechanics of Continuous Media), Gostekhizdat, 1954.

[5] G. Kauderer, Nonlinear Mechanics (Russ. Transl.) IIL, 1961.

[6] Z. A. Gol'dberg, Akust. zh. 6, 307 (1960), Soviet Phys.-Acoustics 6, 306 (1961).

[7] L. Bergmann, Ultrasonics and Their Scientific and Technical Applications, Edwards Bros., 1944.

[8] P. W. Bridgman, The Physics of High Pressure, Bell, London, 1931.

[9] P. W. Bridgman, Recent work in the Field of High Pressures, Revs. Modern Phys. 18, 1 (1946).

[10] F. Birch, Phys. Rev. 71, 809 (1947).