

*EXPERIMENTAL INVESTIGATION OF COMBINATION SCATTERING OF SOUND  
BY SOUND IN SOLIDS*

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Under certain (resonance) conditions, nonlinear scattering of sound by sound may occur in solids, in contrast with gases and liquids, resulting in a scattered wave with a combination frequency. The phenomenon can be treated within the framework of three-phonon interactions, as well as from the standpoint of nonlinear elasticity theory. Several types of interactions in polycrystalline aluminum have been observed at ultrasonic frequencies of 1–20 Mc/sec.

THE scattering of sound by sound, treated by Ingard et al.,<sup>[1]</sup> and later in a series of works of different authors,<sup>[2]</sup> may from our viewpoint be correctly called combination scattering, since in such scattering, the scattered wave, if it exists, has a combination frequency. This is an essentially nonlinear effect which can be observed at comparatively high sound intensities. At the present time, it can apparently be regarded as established that there should be no combination scattering, i.e., no wave of combination frequency outside the region of interaction of the sound beams, when two ideally collimated ultrasonic beams intersect in gases and liquids (media with a single velocity of sound propagation); nonideal collimation of the sound beams, which is inevitable under experimental conditions, leads, it is true, to a weak scattering. The situation is quite different in unbounded isotropic solids, where there are two sound propagation velocities, the velocity of longitudinal and the velocity of transverse waves. This leads to the result that combination scattering of sound by sound should be observed upon fulfillment of certain conditions, sometimes called resonance. This follows both from quantum considerations<sup>[3]</sup> and from the considerations given recently<sup>[4]</sup> on the basis of the nonlinear five-constant elasticity theory of Murnaghan.

In our opinion, the experimental study of combination scattering in the region of low energy phonons (ultrasonic phonons) is of considerable interest. In the first place, such a study would give in principle a possibility (the difficulties encountered here will be discussed below) of determining non-

linear moduli (moduli of second order).<sup>1)</sup> At the present time, these moduli are known only for several solids, and with very low accuracy. Furthermore, these characteristics play a very important role in the physics of solids. Although the absorption of ultrasonic and even hypersonic waves cannot be explained by the direct interaction of these waves with thermal vibrations of the lattice, because of the difference in frequencies (according to Akhiezer,<sup>[5]</sup> the ultrasonic wave disrupts the equilibrium distribution of the thermal vibrations of the lattice, which leads in the final analysis to absorption of the sound waves), it is possible that the moduli of second order depend only slightly or even not at all on the frequency; extrapolation of the values of the second order moduli into the region of frequencies of the Debye thermal waves permits us to discuss a whole series of kinetic processes in solids.

The investigation of the nonlinear elasticity of solids is of interest also from the view point of the possibility of the existence of various parametric devices. In media with a single velocity of propagation of sound (gases and liquids) the possibili-

<sup>1)</sup>At the present time various terms are used in the literature for nonlinear moduli. Sometimes on the ground that in these moduli enter into the cubic terms of the expansion of the invariants of the deformation tensor, they are called the moduli of third order; according to this terminology, the usual linear moduli must be called moduli of second order. We have used the more convenient and exact terminology based on the generalized Hooke's law; here the coefficients in front of the terms linear in the deformation are moduli of first order (or the usual linear moduli); the coefficients in front of the quadratic terms are moduli of second order, and so forth.

ties of the existence of parametric devices which employ nonlinear properties of the medium are very limited. In solids, these possibilities are much broader.

In our short previous communication,<sup>[6]</sup> preliminary results were given of an experimental study of the scattering of a transverse wave by a transverse wave of the same frequency, in which case a longitudinal wave of double frequency is obtained (the interaction is  $T(\omega) + T(\omega) \rightarrow L(2\omega)$ , where  $T(\omega)$  is the transverse wave of frequency  $\omega$ ,  $L(\omega')$  is a longitudinal wave of frequency  $\omega'$ ). Results are now given of a more detailed experimental study of different types of interaction:

$$T(\omega) + T(\omega) \rightarrow L(2\omega), \quad L(\omega_1) + T(\omega_2) \rightarrow L(\omega_1 + \omega_2), \\ L(\omega_1) + T(\omega_2) \rightarrow T(\omega_1 - \omega_2)$$

in polycrystalline aluminum. For all the types of interaction mentioned, the displacement amplitude in the scattered wave can, according to<sup>[4]</sup>, be represented in the form

$$a_p = \sigma \frac{a_1 a_2 \omega_1^3 V}{r \rho_0 c_T^4 c_e} |n|, \quad (1)$$

where  $\sigma$  is a coefficient depending on the type of interaction, on the ratios of the frequencies  $\Omega = \omega_2/\omega_1$  and of velocities  $c = c_T/c_e$ , and on the polarization of the shear wave;  $a_1$  and  $a_2$  are the displacement amplitudes of the interacting waves;  $V$  is the volume of the interaction region;  $r$  is the distance from the center of the interaction region to the point of observation;<sup>2)</sup>  $\rho_0$  the density,  $c_T$  and  $c_e$  respectively the velocities of transverse and longitudinal waves;  $n$  a nonlinear parameter for the given type of interaction, representing a linear combination of the moduli of first order (the bulk modulus  $K$  and the shear modulus  $\mu$ ) and the moduli of second order ( $A$ ,  $B$  and  $C$ <sup>[7]</sup>); the expression for  $n$  generally depends also on  $\Omega$  and  $c$ .

The experimental apparatus, a block diagram of which is shown in Fig. 1, differs slightly from that described earlier.<sup>[6]</sup> Transmitting quartz plates  $T_1$  and  $T_2$ , the receiver of the scattered wave  $R$ , and the plates  $R_1$  and  $R_2$  for control of the operation of the radiators were attached to the polished faces of a polycrystalline aluminum block  $A$ . The acoustic contact for the longitudinal waves was made through a thin layer of transformer oil, the detectors and the radiators of transverse waves were glued to the block either by picein or by a solution of resin in acetone. The dimensions of the discs of the Y-cut quartz for transverse waves were  $14 \times 12$  mm, the diameter of the X-cut plate

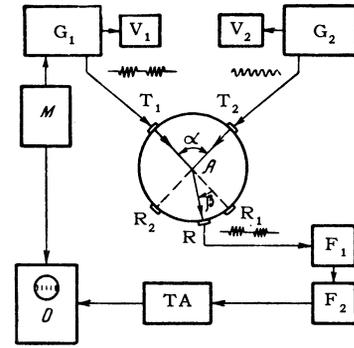


FIG. 1.

for the longitudinal waves was 15–20 mm.

Excitation of the quartz discs can be made either from a single generator  $G_1$  (for the possibility of operating at identical frequencies for the interacting waves), and also from two generators  $G_1$  and  $G_2$  for different frequencies of the interacting waves. One of the generators  $G_1$  operated in a pulse mode (the pulse length was  $\sim 50 \mu\text{sec}$ ), the other,  $G_2$ , in a continuous mode. This naturally led to the result that the scattered signal was pulse modulated. The aluminum blocks had several different dimensions (most of the measurements were carried out on a block of diameter 30 cm and length 2.8 cm.<sup>3)</sup> The investigation of the interaction was carried out in the range 1–20 Mc/sec (the frequencies of interacting and scattered waves).

The detecting system consisted of the receiver  $R$  with a resonant frequency equal or close to the frequency of the scattered wave, two filters—the traps  $F_1$  and  $F_2$  for the frequencies of the interacting waves, the tuned amplifier  $TA$  with gain  $10^3$  at the frequency of the scattered wave, and the oscilloscope  $O$ , the triggering of which is started by the pulse modulator  $M$ . The voltages on the transmitters  $T_1$  and  $T_2$  were determined by the voltmeters  $V_1$  and  $V_2$ . When the interacting waves crossed at an angle of  $\alpha$ , the scattering was observed at the angle  $\beta$  (the angle  $\beta$  was measured in what follows clockwise from the direction of propagation of the first of the interacting waves), which satisfied the resonant condition. For each of the considered types of interaction, the scattering angle  $\beta$  is of course different; as follows from the resonance conditions, it depends on the values of  $\Omega$  and  $c$ . Under experimental

<sup>2)</sup>In<sup>[4]</sup>, the solution for the scattered wave is obtained in the wave zone.

<sup>3)</sup>It must be pointed out that the dimensions of the blocks and the regions of interaction did not allow in their accuracy the existence of conditions corresponding to theory,<sup>[4]</sup> where the scattered wave, as has already been noted, was considered to be in the wave zone.

Nonlinear parameters for different types of interaction in polycrystalline aluminum

Type of interaction	$\Omega = \omega_2/\omega_1$	Angle of interaction $\alpha$	Scattering angle $\beta$	Polarization of shear waves	Nonlinear parameter n	Experimental value of $ n $ , $10^{11}$ dynes/cm <sup>2</sup>
$T(\omega_1) + T(\omega_2) \rightarrow L(\omega_1 + \omega_2)$	1	121°26'	60°43'	⊥	$K - \frac{5}{3}\mu - \frac{1}{4}A + B$	1.8
				∥	$K - \frac{5}{3}\mu - A - 2B$	14
$L(\omega_1) + T(\omega_2) \rightarrow L(\omega_1 + \omega_2)$	0.36	78°	31°50'	∥	$K + \frac{7}{3}\mu + A + 2B$	47
$L(\omega_1) + T(\omega_2) \rightarrow T(\omega_1 - \omega_2)$	0.40	84°	318°30'	⊥	$K - \mu - \frac{1}{2}A + B$	3.3

\*The polarization of the shear wave is indicated relative to the plane passing through the axes of the interacting beams (⊥ - perpendicular, ∥ - parallel)

conditions, the values of  $\Omega$ , as also those of  $\beta$ , were so chosen that the scattered signal was least masked by the interference ("parasitic") signals on various types (interacting waves, directly incident of the receiver, their harmonics, and so forth).

Compared with the study of the nonlinear distortion of waves of finite amplitude,<sup>[8]</sup> the separation of the scattered wave of combination frequency was made somewhat easier here for a number of reasons: first, the difference in the direction of propagation of the incident and scattered waves, and, second, the possibility of control of the interaction by cutting off one of the sources of the interacting waves. The latter lessens the probability of an erroneous identification of "parasitic" signals with the "useful" scattered signal.

1. INTERACTION OF THE TYPE  $T(\omega) + T(\omega) \rightarrow L(2\omega)$

For this interaction, only the one generator  $G_1$  was used (see Fig. 1), operating in a pulsed mode. The voltage from this generator was applied to the two crystals  $T_1$  and  $T_2$ . In contrast with the previous experiments,<sup>[6]</sup> where this interaction was studied only for polarization of both T-waves in the plane passing through the axis of the interacting sound beams (a), in the present work, the interaction was also studied for perpendicular polarization of both T-waves (b) and, in addition, for mutually perpendicular polarization of the two interacting T-waves (c). In the latter case, according to the theory of<sup>[4]</sup>, there is no wave, a fact essentially established experimentally.<sup>4)</sup>

It follows from (i) that when  $a_0 = a_1 = a_2$  we

<sup>4)</sup>A weak scattered signal of the L-wave, with frequency 20 Mc/sec, was observed for one of the thin blocks, with a fundamental frequency of 10 Mc/sec, for the case of mutually perpendicular polarization of the interacting T-waves. This can evidently be attributed by the reflection from the lateral boundaries or to the inhomogeneity of the block (see, for example, [9]).

have  $a_p/a_0 \sim \omega^3$  (the experiment was carried out for different amplitudes of the emitted T-waves). The results of the measurement of the ratio  $|a_p/a_0|$  at three different frequencies (3, 5, and 10 Mc/sec) are shown in Fig. 2 (the curve 1 is obtained for polarization of the T-waves parallel to the plane passing through the axis of the interacting beams, and curve 2 for perpendicular polarization). In carrying out these measurements, the radiating quartz discs  $T_1$  and  $T_2$  (Fig. 2) were attached to the aluminum block (Fig. 1), and the receiving crystals ( $D, R_1, R_2$ ) were changed so that it was possible to work with resonant transducers at each of the frequencies; upon satisfaction of the condition  $a_0 = \text{const}$ , the electrical voltage on the emitters was the same at both frequencies. As seen from Fig. 2, the cubic dependence on frequency is quite closely satisfied, if one keeps in view the difficulties of the experiment. The resonance conditions, and also the expressions for n, are given in the table. We note that the displacement amplitude of the scattered wave amounted to tenths or hundredths of a percent of the amplitude of the interacting waves (see Fig. 2).

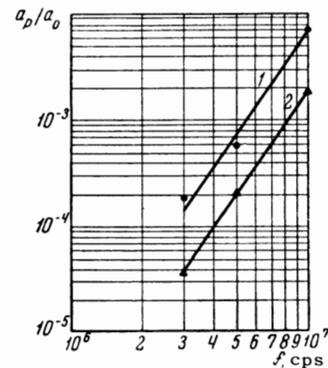


FIG. 2.

2. INTERACTION OF THE TYPE  $L(\omega_1) + T(\omega_2) \rightarrow L(\omega_1 + \omega_2)$

In contrast with the previous interaction, one of the radiators  $T_1$  (Fig. 1) for the L-wave was op-

erated at a frequency of 3.18 Mc/sec in the pulsed mode; the other T-wave radiator  $T_2$  operated in the continuous mode at a frequency of 1.15 Mc/sec; the same voltage of 650 V was applied to each radiator. The receiver for the scattered wave of frequency 4.33 Mc/sec was an X-cut crystal. The presence of the scattered signal (the scattered signal was of course observed in the form of a pulse) was monitored by cutting off one of the generators, just as in the preceding experiment. Moreover, with such an experimental arrangement, there was an additional possibility of monitoring the scattering, for if the frequency of one of the generators (under our conditions, this was  $G_2$ ) is retuned, then (for a fixed position of the radiators) the resonance conditions of interaction are destroyed and the pulse disappears. For ideally collimated interacting beams, the resonance conditions should be destroyed for the slightest deviation from the resonance frequencies. Under the experimental conditions, however, ideal collimation of ultrasonic beams is not possible; therefore, the amplitude of the scattered pulse smoothly decreased with change in the frequency of one of the generators and, finally, at  $\Delta\Omega \approx 0.1$ , the amplitude of the scattered pulse was comparable with the level of the background interference. According to the resonance conditions, such a change of frequency corresponds to the fact that in the radiation there are "oblique beams," which differ in direction from the main beam by  $\Delta\alpha \approx 6^\circ$ , which is reasonable if we recall the dimensions of the radiators and the frequency. For polarization of the T-wave parallel to the plane passing through the axis of the sound beams, the resonance conditions and the value of the nonlinear parameter are given in the table;  $a_p/a_0 = 3 \times 10^{-4}$ . For perpendicular polarization of the T-wave, there is no scattered wave, which agrees with theory.<sup>[4]</sup>

### 3. INTERACTION OF THE TYPE $L(\omega_1) + T(\omega_2) \rightarrow T(\omega_1 - \omega_2)$

Here also one of the radiators  $T_2$  for the T-wave operated in a continuous mode at a frequency of 1.25 Mc/sec; the amplitude of the voltage was 650 V. The other radiator  $T_1$  gave an L-wave of frequency 3.1 Mc/sec; the work was carried out in the pulsed mode, the pulse amplitude being 1500 V. The experimental conditions are shown in the table. Polarization of the scattered T-wave is the same as the polarization of the scattering wave:  $(a_p/a_1) = 3 \times 10^{-5}$ ;  $(a_p/a_1) = 5 \times 10^{-5}$ , where  $a_1$  is the amplitude of the L-wave. Figure 3 shows the oscillograms of the following cases;

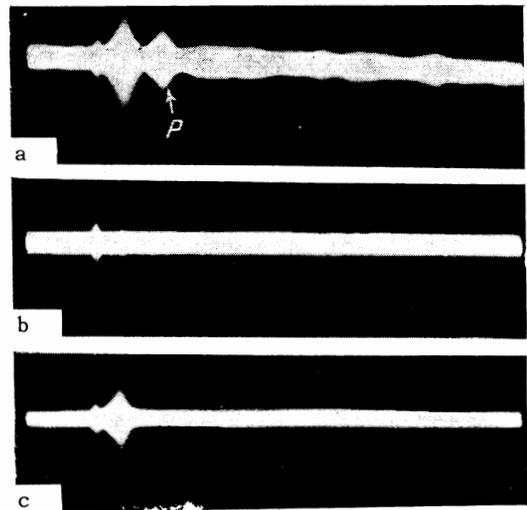


FIG. 3.

a) operation of the radiators  $T_1$  and  $T_2$  (the arrow and the letter P denote the "useful" scattered pulse, in front of which appears a large pulse—the pickup from the pulse generator); b)  $G_1$  is cut off (the pulse generator); c)  $G_2$  is cut off (the generator operating in the continuous mode). We note here that, of all the types of interaction studied, the amplitude of the scattered signal for this interaction was the least (one thousandth of one percent of the amplitude of the interacting waves), thus explaining the low quality of the oscillograms.

The data for the nonlinear parameters of the interactions studied are given in the table. Measurement only of the amplitude of the scattered wave (and not its "phase") allows us to determine the nonlinear parameter  $n$ , apart for its sign. This circumstance makes it difficult to separate the determination of the moduli of second order A, B, and C on the basis of such a measurement. The second circumstance, which lessens the accuracy of determination of  $n$ , is the already noted difference between the experimental conditions and the problem considered for the waveguide in<sup>[4]</sup>. And, finally, the third circumstance is connected with the general difficulties for ultrasonic technology of the determination of the absolute value of the sound pressure, especially in solids. The ratio  $a_p/a_1$  was determined by us as the ratio of the voltage on the receiver R to the voltage on  $R_1$  (for different types of scattered and interacting waves, the assumption was made that the sensitivities at resonance of the X-cut quartz crystal for the longitudinal wave and of the Y-cut for the transverse wave was the same). The two latter circumstances lowered the accuracy of the measurement of  $n$ . According to our estimates, the error of measurement was

~30—50%. At the present time, there is a hypothesis (and even a few experimental results<sup>[9]</sup>), according to which the moduli of second order for different shear or shear-longitudinal nonlinear effects are determined not only by the type of material, but also by its structure, more exactly, by its structural defects. This circumstance (if the purpose of the study is not the investigation of structural defects) allows us to consider as satisfactory the already mentioned comparatively low accuracy of measurement.

In conclusion, it should be noted that in all the phenomena studied, the theoretical predictions<sup>[4]</sup> on the choice of angles and types of interacting waves according to the resonance conditions, on the frequency dependence, etc., are confirmed with complete satisfaction.

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