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PHONON-PHONON
INTERACTION IN CRYSTALS

SIXTH QUARTERLY PROGRESS REPORT
1 September 1962 - 30 November 1962

Signal Corps Contract No. DA-36-039-sc-87209
Order No. 39589-PM-61-91-91
PR and C No. 61-ELP/R-4122

Placed by U.S. Army Signal Research
And Development Laboratory
Fort Monmouth, New Jersey

GENERAL ELECTRIC

HEAVY MILITARY ELECTRONICS DEPARTMENT
SYRACUSE • NEW YORK

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PHONON-PHONON INTERACTION IN CRYSTALS

Report No. 6
Contract No. DA 26 039-sc 87209
Order No. 39589 PM-61-91-91
PR & C No. 61-ELP/R-4122

Sixth Progress Report

1 September 1962 - 30 November 1962

The objective of this project is to study the generation, propagation and interaction of phonons with emphasis on the phonon interactions in crystals.

U. S. Army Signal Research and

Development Laboratory

Fort Monmouth, New Jersey

ELECTRONICS LABORATORY
GENERAL ELECTRIC COMPANY
SYRACUSE, NEW YORK

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1. PURPOSE

The work performed under this contract is aimed at studying the generation, propagation and interaction of phonons in solids by means of microwaves. The main interest is centered on phonon-phonon interaction in crystals.

1.1 Task I Analytical Study

Investigate the generation and propagation of phonons in solids. Study the phonon interactions in one and three dimensional media.

1.2 Task II Experimental Study of Phonon Generation and Propagation

Investigate experimental techniques of generating phonons. Develop methods for enhancement of the transducer coupling. Evaluate the properties of phonon propagation in crystals.

1.3 Task III Experimental Study of Phonon Interactions

Study and evaluate the various types of phonon interactions in solids experimentally.

1.4 Task IV Material Evaluation

Measurement of the elastic properties of various materials and search for materials suitable for phonon propagation and interactions.

2. ABSTRACT

The objective of this project is to study the generation, propagation and interaction of phonons with emphasis on the phonon interactions in crystals.

Theoretical curves based on the selection rules have been drawn for the various modes of operation for phonon-phonon interaction in solids. These are then analyzed with regard to the various types of parametric interactions that are possible.

Experimental results and observations are discussed pertaining to phonon generation and attenuation in various types of single crystals.

Experimental studies involving the search for phonon interactions are discussed.

3. PUBLICATIONS, LECTURES, REPORTS, AND CONFERENCES

1. A conference was held at the Electronics Laboratory between W. G. Matthei, S. Wanuga and W. Brouillette on November 20, 1962 to discuss the status of the project.

2. A paper titled, "Field Enhancement Techniques for the Generation of Microwave Phonons" was presented at the 1962 Ultrasonic Symposium on November 30, 1962 at Columbia University. (H. Hsu, W. Brouillette and S. Wanuga.) The paper has been submitted for publication in the forthcoming issue of PGUE of IRE Proceedings.

4. FACTUAL DATA

4.1 Phase I Theoretical Aspects

During the past period, we have cataloged all the possible interactions that can take place among colinear phonons. These fall broadly into two categories. The first comprises all the interactions that involve direct amplification of the signal wave with gain, with an idler corresponding to a lower sideband. The second comprises all the interactions where the idler corresponds to the upper sideband, thus the interaction falls in the category of up-conversion with gain. The interactions can also, of course, be characterized as to whether they involve forward wave interaction or backward wave interaction.

In discussing these interactions, it is necessary to point out that, for any frequencies presently obtainable, the relation between frequency and wave vector is essentially linear. The selection rules usually discussed in texts in relation to the absorption of sound by thermal phonons are arrived at by considering the entire dispersion curve. The low energy phonons considered in the present waves are not subject to the restriction that an interaction cannot take place where all phonons lie on the same polarization curve. This also presents the disadvantage that for such an interaction, energy may go to the upper sideband rather than the lower.

We also point out that the selection rules we then arrive at are necessary, but not sufficient conditions for interaction to take place. These rules are arrived at by observing that in an interaction, both phonon

energy and momentum are conserved, if we exclude Umklapp processes. In the latter interactions, the phonons must have sufficient energy to carry them to the band edge, which will not occur for the frequencies under discussion. The conservation rules are not the only conditions that must be met. There must also exist appropriate interactions probabilities, arising from the anharmonic Hamiltonian terms.

The selection rules are easily depicted in terms of a transition diagram such as Figure (1). This diagram shows nine possible backward wave interactions where signal and pump energy propagate in opposite direction, the idler is the lower sideband, and the signal is amplified as it propagates through the medium. The polarization branches are marked "L" for the longitudinal wave, and "T_s" and "T_f" for the transverse slow and fast waves respectively. The smaller diagram above indicates the relative position of signal, pump and idler wave for all cases. The transition marked "I" corresponds to a signal phonon which is a slow transverse wave, a pump which is a transverse wave, and idler which is also slow transverse. The segment from "O" to "A" is the signal, that from "O" to "B" the pump, and that from "A" to "B" the idler. Each of the nine cases can be derived from the diagram. Similar diagrams have been prepared for the backward wave case with up conversion, and for the forward wave mixed mode cases for both up and down conversion. The up and down conversion on a single branch is obvious without a diagram.

4.1.1 Non-linear Analysis

In addition to the above considerations, we have examined

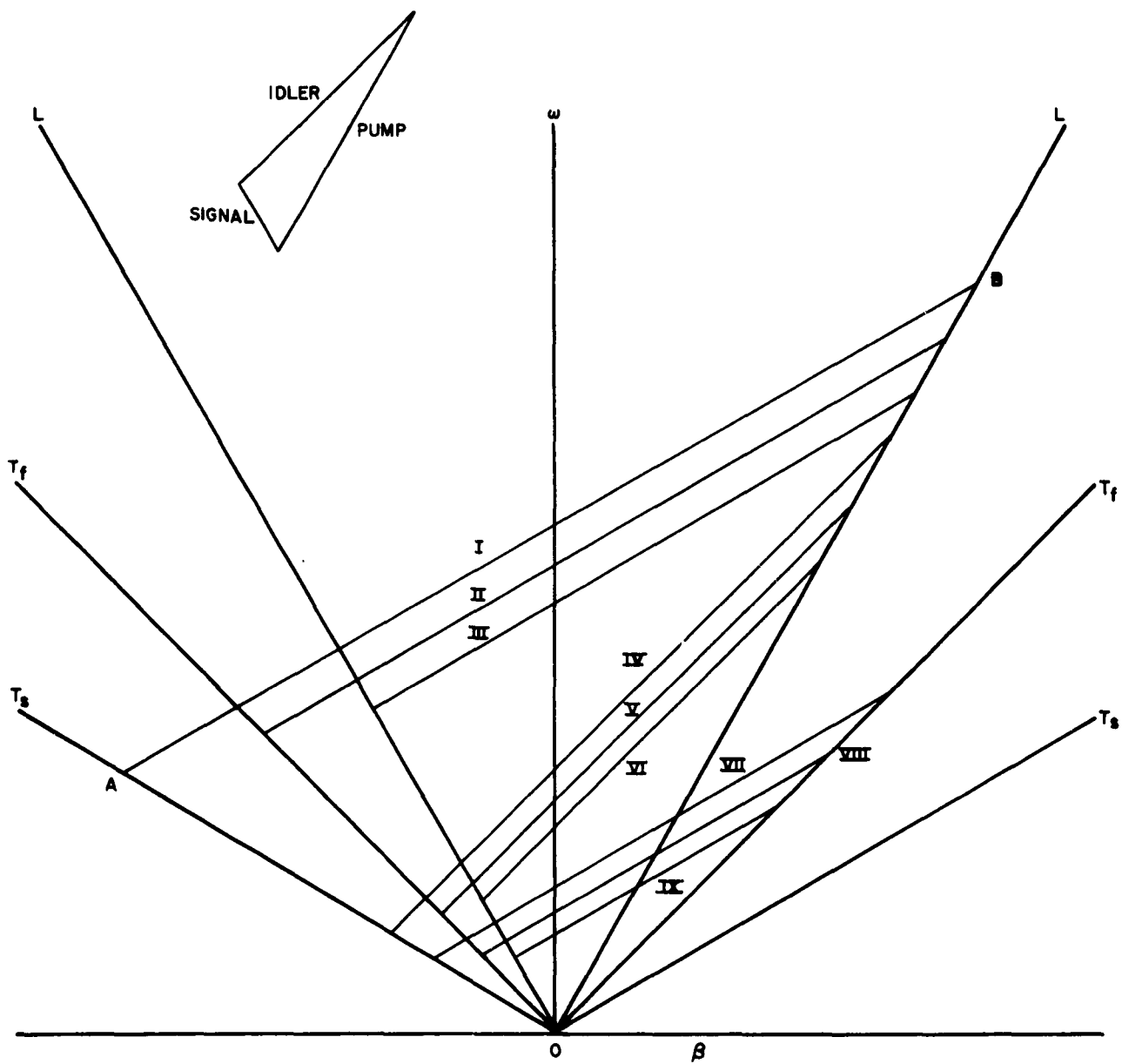


FIGURE 1

ω - β Diagram of Acoustic Lattice Waves in a Crystal, Showing Some Allowed Interactions.

various methods of analysing the non-linear interaction. In these analyses, it is commonly assumed that only those modes of interest can be propagated. For direct amplification, one thus assumes that the only modes propagated are signal, pump, and an idler corresponding to the lower sideband. This type of condition can often be met in an electromagnetic structure by using waveguides or helices with cut-off frequencies. In the solid crystalline medium, modes of all frequencies can be propagated. This restricts the validity of previous analysis and efforts will be continued for a method of determining what effect this may have on traveling wave interactions.

4.1.2 Velocities in Crystals

Numerous types of crystals are being continually studied both from the standpoint of possible use for phonon interaction studies and characteristics of phonon propagation. Two crystals have been investigated during this period. They are hexagonal CdS and ZnO. There are five independent elastic moduli for hexagonal symmetry. Both have higher electromechanical coupling coefficients (k) greater than quartz. A. R. Hutson⁽¹⁾ reports the coupling coefficients as 0.095 for quartz, 0.2 for CdS and 0.4 for ZnO. CdS and ZnO may be useful for phonon interaction studies, since the power flow in a crystal is proportional to k^2 . Velocity equations for this type of crystal are given in section 4.1.3 of the Second Quarterly Progress Report of this contract.

(1) A. R. Hutson, Phys. Rev. Letters, 4, May 14, 1960, pp. 505 - 507.

Elastic Constants of Hexagonal CdS at 27° C. taken from K. E. Bolef⁽²⁾ et al.

$$\begin{aligned} c_{11} &= 84.32 \times 10^{10} \text{ dynes/cm}^2 \\ c_{12} &= 52.12 \times 10^{10} \text{ dynes/cm}^2 \\ c_{13} &= 46.38 \times 10^{10} \text{ dynes/cm}^2 \\ c_{33} &= 93.97 \times 10^{10} \text{ dynes/cm}^2 \\ c_{44} &= 14.89 \times 10^{10} \text{ dynes/cm}^2 \\ c_{66} &= \left(\frac{c_{11} - c_{12}}{2} \right) \end{aligned}$$

The value of density (ρ) used was 4.823 g/cm³.

Table I gives calculated velocities for the C direction.

TABLE I

C-axis CdS <001>

<u>Mode</u>		<u>Calculated Velocity</u>
v_1	=	$4.38 \times 10^5 \text{ cm/sec.}$
$v_{1t} = v_{2t}$	=	$1.73 \times 10^5 \text{ cm/sec.}$

<100> Direction or any other direction perpendicular to <001> axis.

v_1	=	$4.18 \times 10^5 \text{ cm/sec.}$
v_{1t}	=	$1.76 \times 10^5 \text{ cm/sec}$
v_{2t}	=	$1.83 \times 10^5 \text{ cm/sec.}$

(2) D. I. Bolef, N. T. Melamed and M. Menes, Bul. Am. Phys. Soc. 5, 169, (1960).

Values for the calculated velocities for ZnO are given in table II.
The elastic constants were taken from T.B. Bateman⁽³⁾, at 25° C.

Elastic Constants of ZnO

C_{11}	=	209.71×10^{10} dynes/cm ²
C_{12}	=	121.14×10^{10} dynes/cm ²
C_{13}	=	105.13×10^{10} dynes/cm ²
C_{33}	=	210.94×10^{10} dynes/cm ²
C_{44}	=	42.44×10^{10} dynes/cm ²
C_{66}	=	$(\frac{C_{11} - C_{12}}{2})$

The density used was 5.676 g/cm³

TABLE II

C-axis ZnO <001>

V_1	=	6.09×10^5 cm/sec.
$V_{1s} = V_{2s}$	=	2.73×10^5 cm/sec.

<100> direction as any other direction perpendicular to <001> axis.

V_1	=	6.07×10^5 cm/sec.
V_{1s}	=	2.73×10^5 cm/sec.
V_{2s}	=	2.79×10^5 cm/sec.

(3) T. B. Bateman, J.A.P., 33, 3309, (1962).

4.2 Phase II Experimental Studies

4.2.1 General Considerations

Preliminary experiments have been carried out and theoretical studies have been made for determining the types of phonon interactions that are possible. The conditions necessary for interaction have been determined from the selection rules and are continuously being investigated for experimental verification.

Phonon-phonon interaction studies are based on the anharmonicity in the inter-atomic potential of the crystal. It is this phenomenon which contributes a large share of sonic attenuation in a solid. It is, therefore, necessary to continually study the generation and propagation of ultrasonic waves in various media in order to determine which crystals may be better suited for phonon interaction studies. For each crystal orientation and mode of operation the attenuation and propagation characteristics will differ. For these reasons new crystals and crystal orientations are always being studied and data being obtained for use in the present phonon interaction studies now being carried out. Data obtained from some of these studies are presented in following sections.

As shown in previous reports, acoustic-phonon interactions can take place either for forward wave or backward wave type of operation. Experiments presently attempted were directed toward the forward type of interaction involving phonons of the same polarization, i.e., all longitudinal or all transverse.

4.2.2 Complete Cryogenic and Microwave Assembly

The main experimental assemblies for phonon generation and detection remain the same. A small Dewar for use at liquid nitrogen temperature was made for experimental studying at these temperatures. Figure (2) shows the unit placed in our portable electromagnet. This electromagnet is used mainly for experimental study with magnetostrictive transducers at both liquid nitrogen and room temperatures. The complete assembly offers a better technique of experimental study since there is more ease in handling the microwave structures and the unsilvered Dewar offers direct observation of the set up.

4.2.2.1 Microwave Structures used for Piezoelectric Excitation of Phonons.

Modifications have been made in the microwave cavities by replacing the resonant posts with the chisel shaped posts described in our Fifth Quarterly Progress Report. These have been made mainly where transverse mode generation in X-cut quartz rods was desirable.

4.2.2.2 Microwave Structures used for Magnetostrictive Excitation of Phonons

A wideband structure consisting of a shorted stainless steel coaxial line was built for use with the new Dewar described in section 4.2.3. The structure operated satisfactorily over a frequency tuning range of 1 to 1.8 kMc/s.

4.2.3 Materials

Quartz, cadmium sulfide, germanium, silicon and cadmium telluride were the materials used during this period.



FIGURE 2

PORTABLE ELECTROMAGNET WITH DEWAR ASSEMBLY USED FOR EXPERIMENTING
AT LIQUID NITROGEN AND ROOM TEMPERATURES

4.2.4 Experimental Results

4.2.4.1 Generation of Phonons

Magnetostrictive excitation was used mainly in the experimental testing during this period. A few successful tests were made using the self piezoelectric excitation of cadmium sulfide at S- and X-band frequency.

4.2.4.2 Phonon Propagation in Quartz

Several different orientations of quartz crystal were investigated. Rods 3 mm. in diameter and 2 cm. long of AC and BC oriented quartz were plated with magnetostrictive films on one edge. These two cuts are of interest in the phonon interaction study since they are the only two orientations of rotated Y-cuts where transverse waves have no cross coupling to other modes. The acoustic energy travels parallel to the normal of the wave fronts.

The BC-cut crystal is made by a rotation about the X-axis making an angle of -59° with respect to the Z-axis. Figure (3) shows the echo pattern observed for the BC-cut crystal at 1.15 kMc/s and room temperature. The velocity was 5.03×10^5 cm/sec. A measurement of attenuation for these conditions gave a figure of about 1.4 db/cm.

Figure (4) shows the echo pattern observed for an AC-cut rod. This cut is made by a rotation about the X-axis of 31° with respect to the Z-axis. The echo pattern is not clear since there appears to be other modes being generated. The transverse velocities for this particular cut are very close, being 3.84 and 3.32×10^5 cm/sec. The crystal will



FIGURE 3

TRANSVERSE WAVE ECHO PATTERN IN BC QUARTZ

MAGNETOSTRICTIVE EXCITATION

1.15 kMc/s - ROOM TEMPERATURE

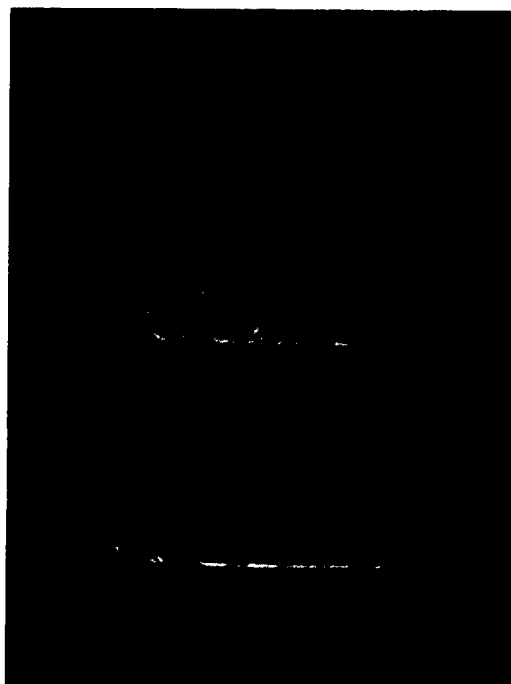


FIGURE 4

ECHO PATTERN IN AC QUARTZ

MAGNETOSTRICTIVE EXCITATION

1.15 kMc/s - ROOM TEMPERATURE

be tested at liquid helium temperature in order to distinguish the proper modes. The attenuation appears to be significantly larger for this cut of crystal compared to the BC-cut rod.

4.2.4.3 Phonon Propagation in CdS

Cadmium sulfide crystal is of the hexagonal class and is piezoelectric. Small bars of C-axis oriented crystal were processed on our polishing equipment and tested at various frequencies. Figure (5) shows the echo pattern observed at 3.3 kMc/s and at liquid helium temperature. The rod was approximately 5 mm. square and 1.4 cm. long with a resistivity of approximately 10^5 ohm-cm. The crystal itself was used as the transducer. The echo pattern is completely undistinguishable as to mode designation. Figure (6) shows the same rod tested at X-band frequency under the same conditions.

A piece the same block of CdS was cut to approximately .7 cm. length and the ends were polished flat and parallel. The ends were then plated with magnetostrictive films. The crystal was then tested at liquid helium temperature and 1.15 kMc/s. Figure (7) shows the transverse mode observed. Velocity is 1.7×10^5 cm/sec. A measurement of attenuation under these conditions gave a figure of approximately 5.9 db/cm.

4.2.4.4 Phonon Propagation in Non-piezoelectric Media

Magnetostrictive transducers were used for phonon generation in non-piezoelectric materials. Figure (8) shows the transverse mode observed in a $\langle 100 \rangle$ oriented germanium crystal 3 mm. diameter by 2 cm. length. The test was made at 1.15 kMc/s and liquid helium temperature. Velocity measured was 3.56×10^5 cm/sec.

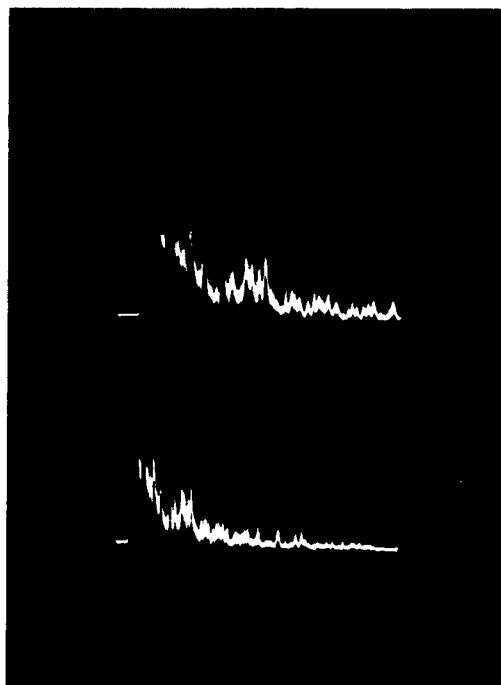


FIGURE 5

ECHO DISPLAY IN C-AXIS CUS

PIEZOELECTRIC EXCITATION

3.3 km/s - LIQUID HELIUM TEMPERATURE

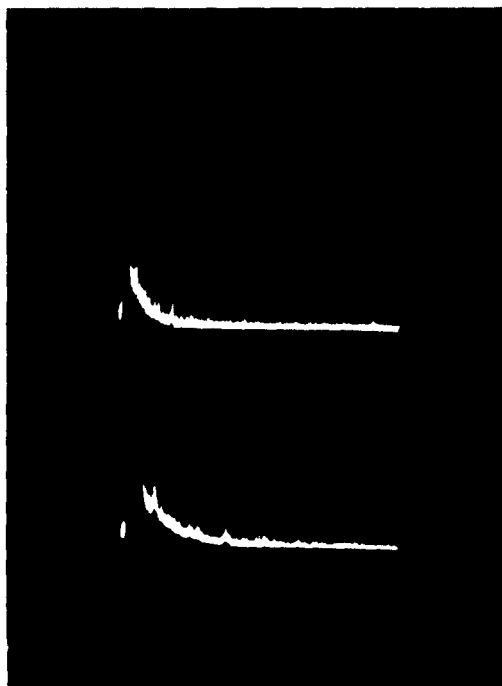


FIGURE 6

ECHO DISPLAY IN C-AXIS CdS

PIEZOELECTRIC EXCITATION

9.4 kMc/s - LIQUID HELIUM TEMPERATURE

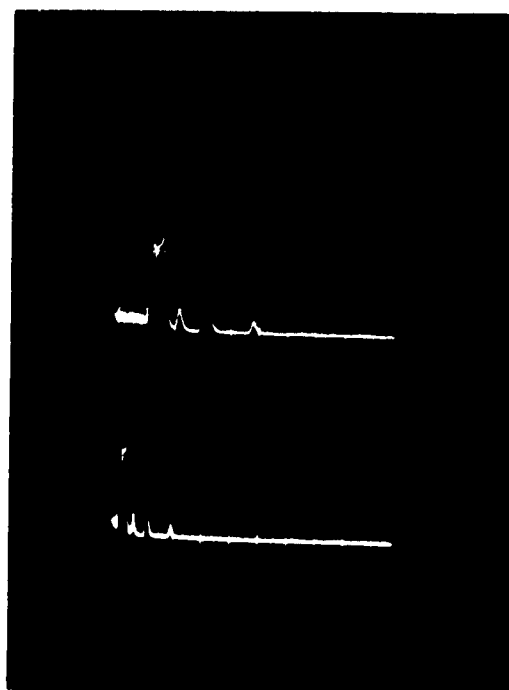


FIGURE 7

C-AXIS CdS

MAGNETOSTRICTIVE EXCITATION

TRANSVERSE MODE. VEL. = 1.7×10^5 cm/sec.

1.15 kMc/s - LIQUID HELIUM TEMPERATURE

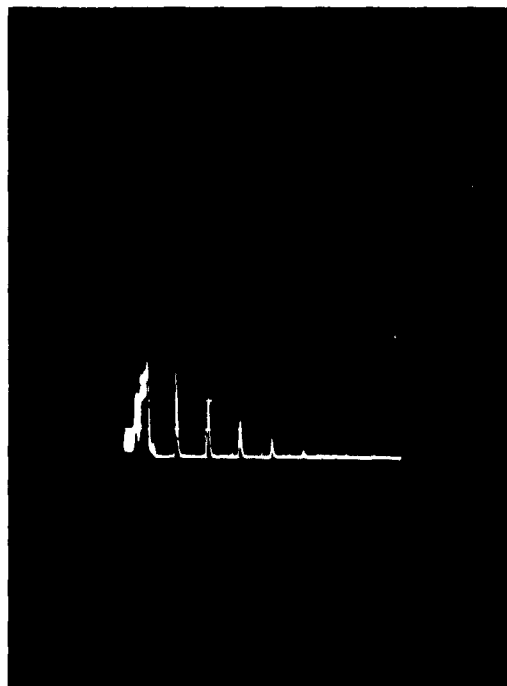


FIGURE 8

$\langle 100 \rangle$ GERMANIUM

MAGNETOSTRICTIVE EXCITATION

TRANSVERSE MODE. VEL. = 3.56×10^5 cm/sec.

1.15 kMc/s LIQUID HELIUM TEMPERATURE

Additional samples of CdTe were also plated with magnetostrictive films and tested for phonon propagation. In each case the material appeared very soft and thin sheets peeled off in the 110 plane.

5. PHONON-PHONON INTERACTION

During this period, a series of experiments were performed, seeking evidence of phonon-phonon interaction in one of various forms. These include direct interaction, yielding gain at the signal frequency, upper sideband conversion, with consequent increased attenuation in the signal mode and appearance of a sum frequency, and production of a third harmonic signal.

The first test involved an X-cut quartz rod. A thin film transducer of nickel-cobalt was applied. At room temperature, echoes were observed at 1.16 gcps. The observed attenuation was about 1 db/cm for the fast transverse mode.

A further test was made in the X-cut rod, generating a signal at 3.3 gcps and looking for the third harmonic at 9.9 gcps. The rod was cooled to liquid helium temperature, pulses were generated at 3.3 gcps with a thin film, using an X-band cavity to detect a possible 9.9 gcps, but no third harmonic was observed.

Another run was made on an X-cut quartz rod, driving it at 1 gcps magnetostrictively and attempting to detect the third harmonic with negative results.

Three tests were performed on a silicon rod cut along the $\langle 111 \rangle$ axis. In these experiments one end of the rod was filmed so that it would operate at one gcps, and the other filmed for three gcps. Both experiments seeking third harmonic generation of the 1 gcps signal and interaction between a 1 gcps and a 3 gcps signal were performed. The sample was cooled in liquid helium. No third harmonic could be detected when the crystal was driven at 1 gcps.

In the second tests of these series, one gcps echoes were obtained by driving the appropriate cavity, and a 3 gcps pump signal was applied to the other cavity. The time lapse between the pulses could be controlled so that the pump signal was applied just as the signal pulse was being reflected from the end of the sample being driven at the pump frequency. When this adjustment was made, a very slight decrease in the amplitude of the signal echoes was observed. This could be due to a direct feed-through from pump transmitter to signal receiver.

A similar test was performed on a $\langle 100 \rangle$ germanium crystal, using a magnetostrictive film transducer. The crystal was cooled in liquid helium, and the filmed end placed in a cavity that could be excited both at 1.1 gcps and 3.3 gcps, by using a $1/4$ and $3/4$ wave length mode respectively. Both longitudinal echoes, with a velocity of 5×10^5 cm/sec and transverse echoes with a velocity of 3.56×10^5 cm/sec. could be observed at both frequencies.

The sample was excited simultaneously with 1.1 and 3.3 gcps energy. The 1.1 gcps echoes were observed to show a slight gain in amplitude

when the 3.3 gcps signal was applied. However, this was definitely proved to be RF feed-through from the S-band transmitter to the L-band receiver.

If interaction occurs with up-conversion, then the signal echoes should show an increased attenuation with the addition of a pump signal, and energy should appear at the sum frequency.

One experiment was performed during this period to test this. A silicon $\langle 111 \rangle$ rod was used with thin films at liquid helium. A single cavity operating at 1 gcps and 3 gcps was used to excite one end with pump and signal energy. A cavity tuned to the sum was placed on the other end. We were not able to observe interaction in this one experiment, and some difficulty was encountered with feed-through. We are presently improving this experiment so that more trials may be made using other samples and other transducers and cavities.

6. CONCLUSIONS

All possible interactions that can take place among colinear phonons have been studied and used for determining the most promising experiments. Tests were performed in order to verify experimentally phonon-phonon interactions in crystals. Although no positive evidence has been observed, difficulties encountered in each test are removed so that the experimental procedures are being optimized.

Numerous orientations of quartz crystals have been studied. Some of these cuts appear to be promising for use in the phonon-phonon interaction studies. CdS was also investigated for phonon propagation at kMc/s frequencies. Although the attenuation is greater than quartz, other advantages such as higher coupling coefficient may be useful.

7. PROGRAM FOR NEXT INTERVAL

The study of phonon-phonon interactions will continue. Emphasis will be placed on the forward wave type of interaction searching for frequency conversion. The microwave structures will be modified for improving experimental techniques, mainly for solving r.f. feed-through problems. Generation and propagation of acoustic waves in various materials and crystal orientations will be carried out. These will include testing at various temperatures and frequencies.

8. IDENTIFICATION OF PERSONNEL

Dr. Hsuing Hsu - 24

Dr. J. Walter Brouillette - 148

Stephen Wanuga - 360

AD	General Electric Co., Electronics Laboratory, Syracuse, N.Y. PHONON-PHONON INTERACTION IN CRYSTALS BY Hsiung Hsu and Stephen Wanuga. First Quarterly Progress Report 1 May 1961-31 July 1961. 61 p. incl. line drawings and photographs. (Contract DA-36-039-sc-87209) The objective of this project is to study the generation, propagation, and interaction of phonons in solids by means of microwaves with emphasis on the phonon-phonon interaction in crystals. These phonon interactions are interpreted as parametric interactions due to crystal anharmonicity. In this quarterly report, the concept of three dimensional parametric interactions is discussed. The mechanism of various types of traveling wave parametric interactions are analyzed. The selection rules of coupled phonon modes and various experimental approaches are described. Detailed discussions on the low temperature microwave structures and experimental results on the generation and propagation of microwave phonons are presented.	UNCLASSIFIED 1. Generation and propagation of hypersonic phonons 2. Phonon-phonon interaction 3. Parametric interaction I. Hsu, Hsiung II. Wanuga, Stephen III. Contract DA-36-039-sc-87209	AD General Electric Co., Electronics Laboratory, Syracuse, N.Y. PHONON-PHONON INTERACTION IN CRYSTALS BY Hsiung Hsu and Stephen Wanuga. First Quarterly Progress Report 1 May 1961-31 July 1961. 61 p. incl. line drawings and photographs. (Contract DA-36-039-sc-87209) The objective of this project is to study the generation, propagation, and interaction of phonons in solids by means of microwaves with emphasis on the phonon-phonon interaction in crystals. These phonon interactions are interpreted as parametric interactions due to crystal anharmonicity. In this quarterly report, the concept of three dimensional parametric interactions is discussed. The mechanism of various types of traveling wave parametric interactions are analyzed. The selection rules of coupled phonon modes and various experimental approaches are described. Detailed discussions on the low temperature microwave structures and experimental results on the generation and propagation of microwave phonons are presented.	UNCLASSIFIED 1. Generation and propagation of hypersonic phonons 2. Phonon-phonon interaction 3. Parametric interaction I. Hsu, Hsiung II. Wanuga, Stephen III. Contract DA-36-039-sc-87209	UNCLASSIFIED
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