PHONON INTERACTIONS IN CRYSTALS

Report No. 1 / Contract DA 36-039 AMC-02280 (E)

FIRST QUARTERLY TECHNICAL REPORT

15 MAY 1963—14 AUG. 1963

INTERNATIONAL BUSINESS MACHINES CORP.

Thomas J. Watson Research Center, Yorktown Heights, New York

Work performed for U. S. Army Electronics Research and Development Laboratories, Fort Monmouth, New Jersey

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Program Objective: The object of this research program is to investigate various types of phonon interactions in order to produce new and improved amplifiers, oscillators, and frequency converters of microwave acoustic energy.

Report Prepared by

A. H. Nethercot, N. S. Shiren, M. Pomerantz

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PURPOSE

The purpose of this contract is to investigate, experimentally and theoretically, various interaction processes of phonons in solids with a view towards developing and improving methods of amplifying, generating, frequency converting, and propagating microwave acoustic energy.

A chief approach is to study the parametric amplification and parametric frequency conversion of acoustic waves, especially the forward travelling wave interaction. The use of paramagnetic ions in magnesium oxide is being investigated to maximize such effects. Since the phase velocity of an acoustic wave of a specific frequency can be changed by resonating the paramagnetic ion, advantage can be taken of various constructive and destructive interference effects to enhance these travelling wave parametric mixing effects.

Microwave acoustic masers and electron - phonon interaction effects are also being studied for the same reasons. In addition, various sound propagation effects are under investigation.

ABSTRACT

A significant achievement has been the parametric up-conversion of 9,000 Mc acoustic waves in a crystal of magnesium oxide. Both the harmonics of a single input frequency and the sum frequency of two input signals have been generated. Power conversion efficiencies of up to 80% have been obtained in a two centimeter path for input powers on the order of $\frac{1}{2}$ watt/cm^2. This converted power has not yet been observed directly, but a new technique using parametric ions has shown that the parametric process is indeed that responsible for the observed large power dependent attenuation of the input signals: This large attenuation is absent when the resonance frequency of these paramagnetic ions is adjusted so that the phase velocity of the acoustic waves at the sum frequency is substantially altered. This causes destructive interference effects (phase mismatching) so that the forward travelling wave interaction is effectively suppressed.

Acoustic maser amplification has been observed in magnesium oxide crystals containing Ni^{++}. The acoustic power was amplified by a factor of 25 in traversing a two-centimeter path. This is very much larger than the small percentage increase previously reported by Tucker for Cr^{+++} ions in sapphire (ruby). The reason is that the spin-phonon interaction constant for Ni^{++} ions in MgO is very much larger, and this leads to correspondingly larger maser effects. Attempts have

been made to measure the signal velocity of the acoustic pulse when the Ni^{++} ions have an inverted population, but the effect is small and the shift in pulse arrival time has not yet been measured.

The main interest in acoustic interactions in germanium is that it should be possible to amplify acoustic waves by interactions with a conduction current. The interaction is somewhat similar to that observed in CdS by Hutson, McFee and White. However, in germanium this interaction utilizes the deformation potential rather than the piezoelectric effect and, more important, space charge effects should be absent. Therefore, debunching effects should not cause an upper frequency limitation of the effect. The theory has largely been worked out, but amplification has not yet been definitely established experimentally. In the course of this work, a power dependent attenuation of the input wave was discovered in non-conducting germanium. This is probably the same type of parametric interaction that was discussed above for MgO. Also, the temperature dependence of the ordinary phonon attenuation in germanium has been measured. Low loss transmission at 9,000 Mc is observed to higher temperatures ($\sim 40^{\circ}K$) than is the case for quartz.

A variety of complicated propagation effects for acoustic waves have been predicted in crystals having an odd - fold axis of rotational

symmetry. These effects are somewhat similar to those occuring for light waves in biaxial crystals. In particular, it has been predicted by Waterman that in quartz a sound wave with its phase velocity along the z - axis will have as its direction of energy flow a cone at an angle of 14^{o} from the z - axis (internal conical refraction). The acoustic energy will be found uniformly over this cone. A more careful theoretical consideration of this problem has indicated that the acoustic energy will not be distributed uniformly but will instead be found preferentially at the three azimuthal positions $\phi = 90^{o}$, 210^{o}, and 330^{o}.

PUBLICATIONS, LECTURES, REPORTS, AND CONFERENCES

Dr. Shiren's paper on parametric up-conversion has been published under the title "Non-Linear Acoustic Interaction in MgO at 9 Gc/sec" in the Physical Review Letters 11, 3 (July, 1963).

Dr. Shiren delivered a talk on this same subject at the Sperry-Rand Research Center, Sudbury, Massachusetts, on August 2, 1963.

Drs. Shiren and Pomerantz are both scheduled to deliver invited papers at the Ultrasonics Symposium to be held in Washington, D. C. in December.

Dr. Pomerantz has also submitted an abstract on "Generation of Longitudinal and Transverse Microwave Phonons by Spin Wave Resonance" to the Magnetics Conference to be held in Atlantic City in November.

I. PARAMETRIC UP-CONVERSION

We have observed large power dependent attenuations of 9 Gc/sec longitudinal acoustic wave pulses propagating on the <100> axis of MgO. The causative non-linear interaction is interpreted to be three phonon scattering through the cubic term in the anharmonic lattice energy. Because the frequencies involved were a factor 10^3 higher than those used in the recently reported work of Rollins[1] and Mahler et al.,[2] the interactions were many orders of magnitude larger; power conversions of as much as 70% were observed over a 2cm interaction path for input acoustic power levels $\sim 1/2$ w/cm^2. The generated harmonics and sum frequencies resulting from the use of one and two input signals, respectively, have not been directly detected. However, their presence was ascertained by the

use of ultrasonic paramagnetic resonance dispersion to provide phase velocity mis-matching.

The experimental geometry is shown in Fig. 1. It differs from that previously used[3] only in the addition of a rectangular cavity, positioned so that the bonded surfaces of the crystals lay in the maximum microwave electric field, with the MgO crystal extending through the back of the cavity. The x-cut quartz transducers were 1.75 cm long and 3 mm in diameter. They were bonded to MgO crystals of the same diameter and ~1 cm long, with GE 7031 resin.

In the harmonic generation experiments, only the re-entrant cavity was used to generate a single input. For the experiments utilizing two acoustic inputs the second was generated at the bonded surface of the quartz. The efficiencies of generation and detection in the rectangular cavity were ~ 8 db lower than in the re-entrant cavity. However, this method of obtaining two acoustic inputs obviates the necessity for two bonds, the use of which is particularly disadvantageous at microwave frequencies.

Fig. 2 shows the echo patterns obtained in the harmonic generation experiment at two acoustic power levels differing by 20 db. Several bond echoes are shown; each is followed by a series of four echoes within the MgO. (The third and fourth appear just before the

first and second, respectively, of the next set.) It is seen that at the high power level the MgO echoes are smaller, relative to the bond echoes, than at low power. The measured decrease in amplitude represents ~65% power conversion. A typical input frequency was 9.2 Gc.

In the two input frequency experiments a decrease in the amplitude of the signal MgO echoes (rectangular cavity) was observed when the pump wave (re-entrant cavity) was timed so as to propagate through the MgO simultaneously, and in the same direction, as the signal wave. The pump wave pulse width was made large enough to bracket the 0.5μ sec signal pulses. Signal power conversions of ~70% were observed with pump and signal frequencies differing by as much as 1000 Mc/sec.

If the attenuations are interpreted on the basis of coherent three phonon scattering processes, conserving energy and quasi-momentum in a dispersionless medium (acoustic approximation), then two input co-linear longitudinal waves can only combine to form sum and difference frequency waves which propagate in the same direction as the input waves and are also longitudinally polarized. (Transverse waves are eliminated by the conservation conditions and also by crystal symmetry properties.) Since the waves all travel with the same phase and group velocities, the interaction is extended

over the whole path length and all possible sum and difference combinations will eventually be generated.[4] One then expects the attenuation of the primary wave to increase with distance. In the present experiments, however, one cannot make a meaningful comparison of the observed attenuations of successive passes in the MgO since for a thin film bond the transmission and reflection properties will be frequency dependent; thus the relative harmonic content is not the same immediately after a reflection as it was immediately before. Each successive pass in the MgO must be treated, theoretically, as a separate problem with a specified (but here unknown) set of initial conditions.[5]

In order to confirm the three phonon assumption, it is important to demonstrate that the generated waves are present at the expected frequencies and that they are longitudinally polarized as required by the conservation conditions. We have used the paramagnetic resonance dispersion due to Fe^{2+} impurities in the MgO for the above purposes. The spin phonon coupling for Fe^{2+} is large enough[6] so that, depending on the particular transition, a percentage change in phase velocity $\sim 1\%$ is obtainable at $2^{o}K$ with an impurity concentration of $\sim 10^{18}$ cm^{3}. Thus, by setting the spin resonance frequency near one of the input or generated frequencies, thereby mis-matching the

phase velocities, it was possible to suppress the non-linear interaction. The line width of this suppression effect is determined by the resonance line shape and is proportional to the strength of the spin phonon interaction. The latter varies with the angle, θ, of the magnetic field with respect to the <100> propagation direction. For a given spin resonance transition the functional form of this angular variation is characteristic of the acoustic polarization.[7]

With the pump frequency at ω_1 and the signal at ω_2, Fig. 3 shows the variation in signal amplitude as the magnetic field is scanned through the region about $\gamma H = \omega_3 = \omega_1 + \omega_2$. The dip at the center occurs at exact resonance where the paramagnetic dispersion is zero. The angular variation of the suppression line width, (taking the resonance line shape into account) was found to follow that expected for a longitudinal wave, $(\cos\theta \sin\theta)^2$ for $\Delta M = 1$ transitions, and $\sin^4\theta$ for $\Delta M = 2$ transitions. Similar effects (although not as clearly resolved) were observed at the resonance fields for higher sum frequencies (or harmonics). Typical input frequencies used were $\omega_1 / 2\pi = 9.238$ Gc and $\omega_2 / 2\pi = 9.132$ Gc.

In addition to confirmation of the longitudinal polarization, the presence of phase mis-matching by the impurity ions is proof that the non-linear interaction takes place in the MgO, and not in the bonding material or at the discontinuity between quartz and MgO .

Further evidence is provided by the absence of non-linear effects in quartz to quartz bonds.

In view of the large magnetic effects observed, it was considered necessary to show that the impurities were not themselves the cause of the anharmonicity. Measurements made on a crystal having smaller magnetic ion concentrations by a factor 10^2 showed the same non-linear interaction strength as previously observed and, of course, proportionally smaller magnetic effects. In addition, no temperature dependence of the non-linear attenuation was observed in the range $1.6^{\circ}K$ to $4.2^{\circ}K$.

In Fig. 4 the fractional decrease in amplitude of the signal frequency is plotted against pump power for a signal input power level 20 db less than the maximum pump input power. In the region where the change in signal amplitude is small, and varies linearly with pump power, only the lowest sum frequency, ω_3, is important. For this case Lasher and Nethercot[8] have shown that the fractional decrease of signal strain amplitude is given by,

$$\frac{\Delta e_2}{e_2} = \frac{1}{2}\left(\frac{A_{111}^{111}}{4C_{11}^{11}}\right)^2 e_1^2 \ \omega_2/\omega_3 \ (k_3 x)^2.$$

e_1 and e_2 are, respectively, the appropriate longitudinal pump and

signal strain amplitudes, and the anharmonic lattice energy density is

$$V = \frac{1}{3!} \Sigma_{ijk\ell mn} A_{ijk}^{\ell mn} e_{\ell}^{i} e_{m}^{j} e_{n}^{k} .$$

From an approximate calibration of the efficiency of the ultrasonic generation, we are able to set limits on the pump strain amplitude e_1. For a pump rf drive power of 48 watts, which, from Fig. 4, gives a decrease $\Delta e_2/e_2 = 0.1$, we find

$$0.6 < e_1 \times 10^6 < 2.8.$$

Then, using $k_3 x = 2.7 \times 10^5$, $\omega_2/\omega_3 \simeq 0.5$ the anharmonic elastic constant is,

$$3.2 < \left| A_{111}^{111} \right| / C_{11}^{11} < 16.0$$

This is of the same order as the values of A_{111}^{111} reported for other cubic crystals.[9]

Phonon-phonon scattering involving three longitudinal phonons is generally considered to be forbidden by the conservation conditions, because of the normal dispersion in the crystal. However,

for the simple model of a one dimensional crystal having the same lattice constant as MgO, the fractional change in phase velocity from 10 Gc/s to 20 Gc/s is only $\sim 10^{-8}$; whereas, from measurements of the width of the magnetic phase mis-match, it is found that a fractional change of $\sim 10^{-5}$ is required to suppress the non-linear interaction by a factor 1/2. This represents a phase change (at ω_3) of ~ 2 radians over the total interaction distance.

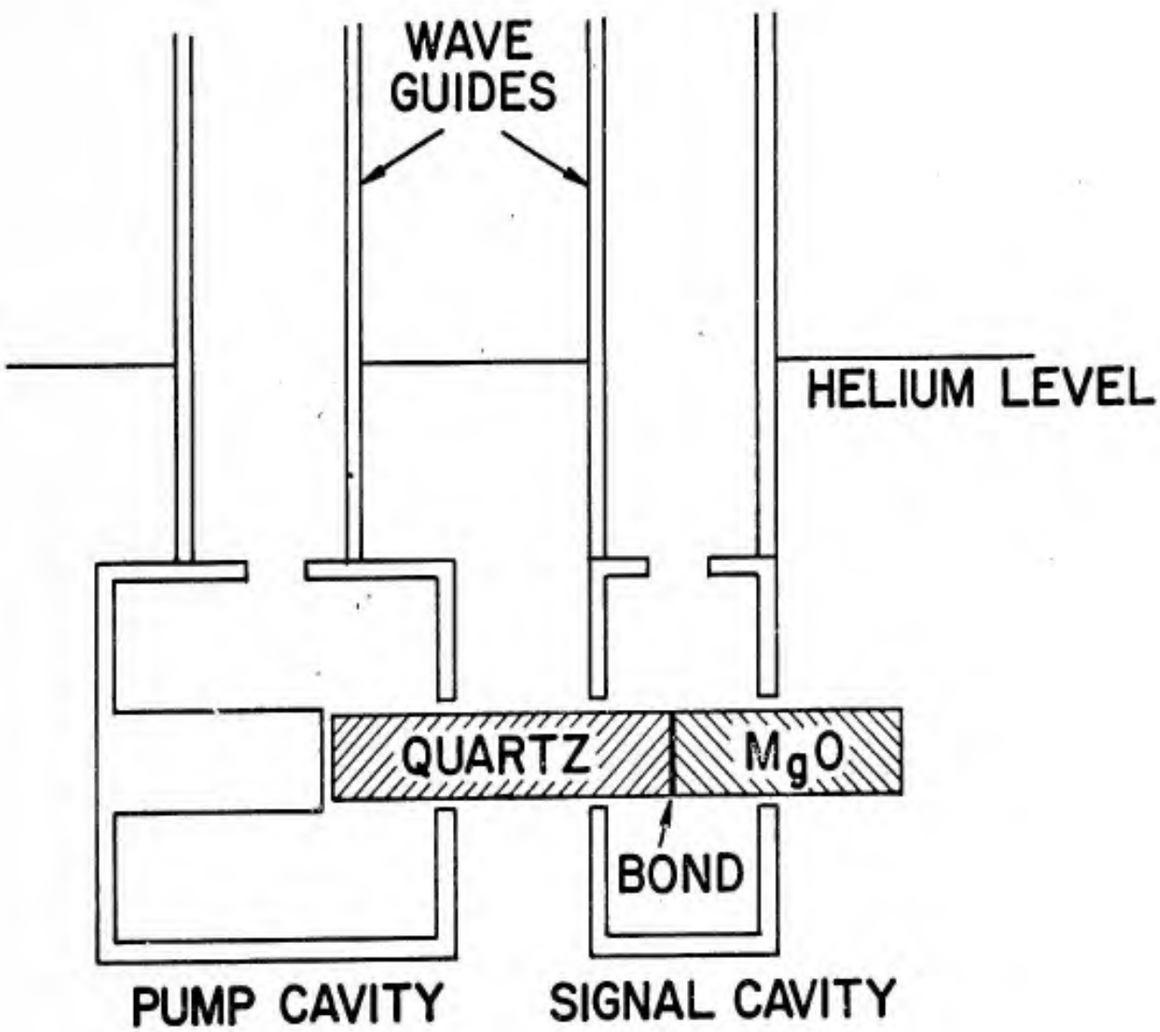

Figure 1 Schematic diagram showing ultrasonic drive cavities and crystal arrangement.

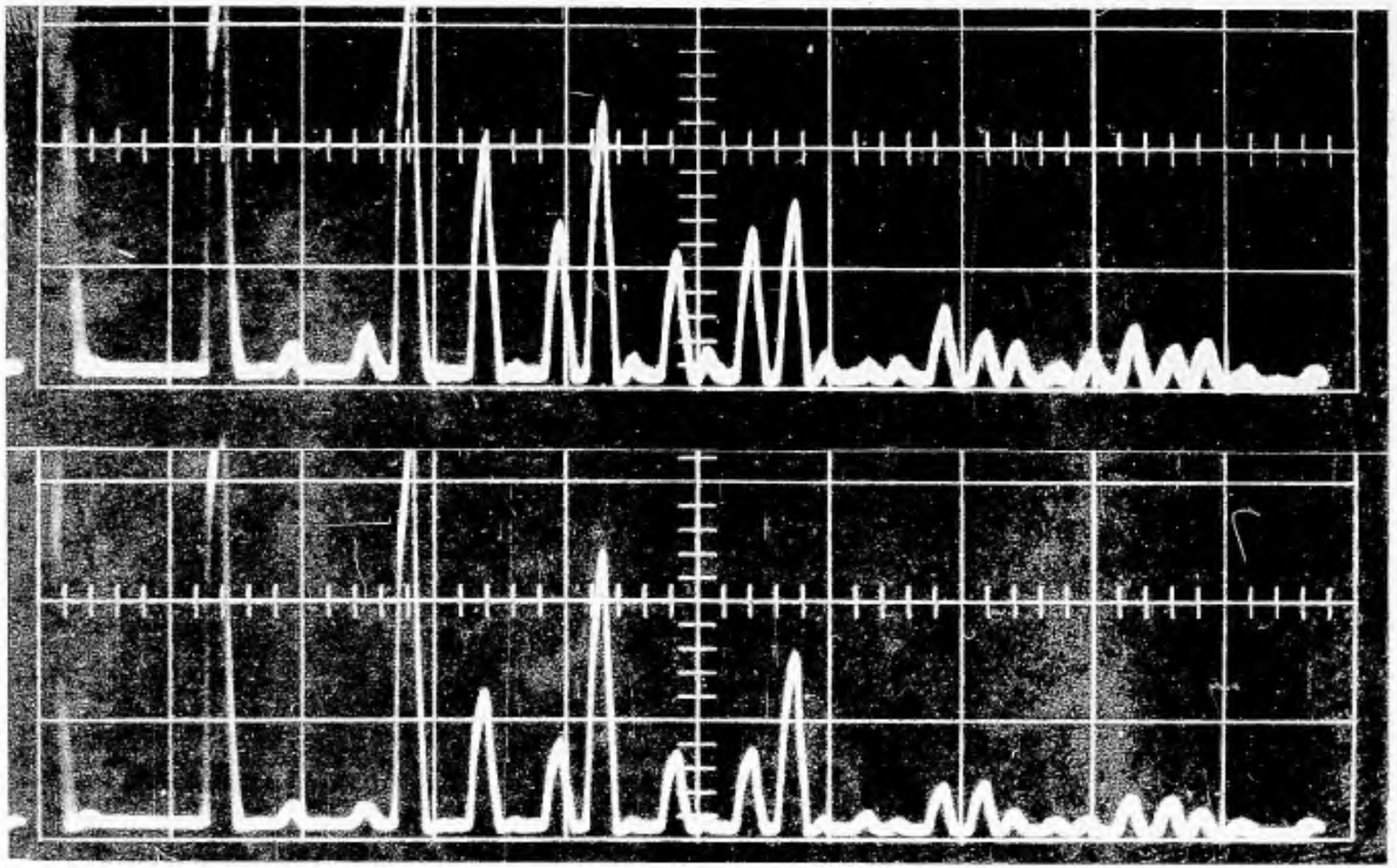

Figure 2 Oscilloscope traces showing relative power dependent attenuation of MgO echoes for a single input. For the lower trace the input power level is 20 db greater than for the upper trace, and the post detection amplification is 20 db smaller. The bond echoes are distinguishable from the MgO echoes by the fact that their amplitudes are the same on both traces.

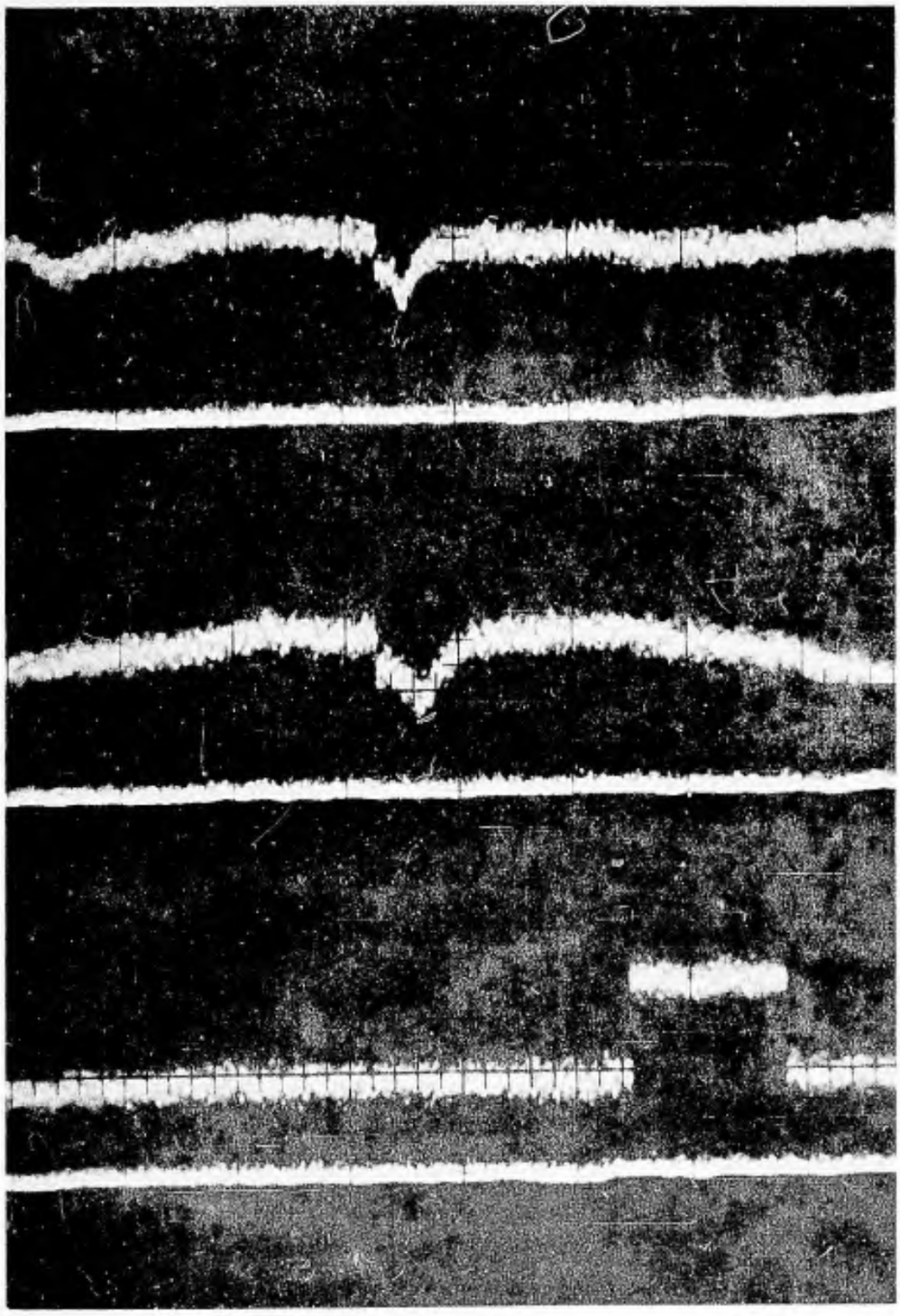

Figure 3 Variation of signal amplitude as a function of magnetic field in the region about $H = \omega_3/\gamma$. Signal (ω_2) input level 16 db smaller than pump (ω_1) input level. $\omega_1/2\pi = 9.20$ Gc/sec, $\omega_2/2\pi$ 9.18 Gc/sec. Total field scan ~ 2500 oe. Top: $\theta = 45^\circ$. Middle: $\theta = 22.5^\circ$. Bottom: $\theta = 0^\circ$, and signal amplitude with pump off.

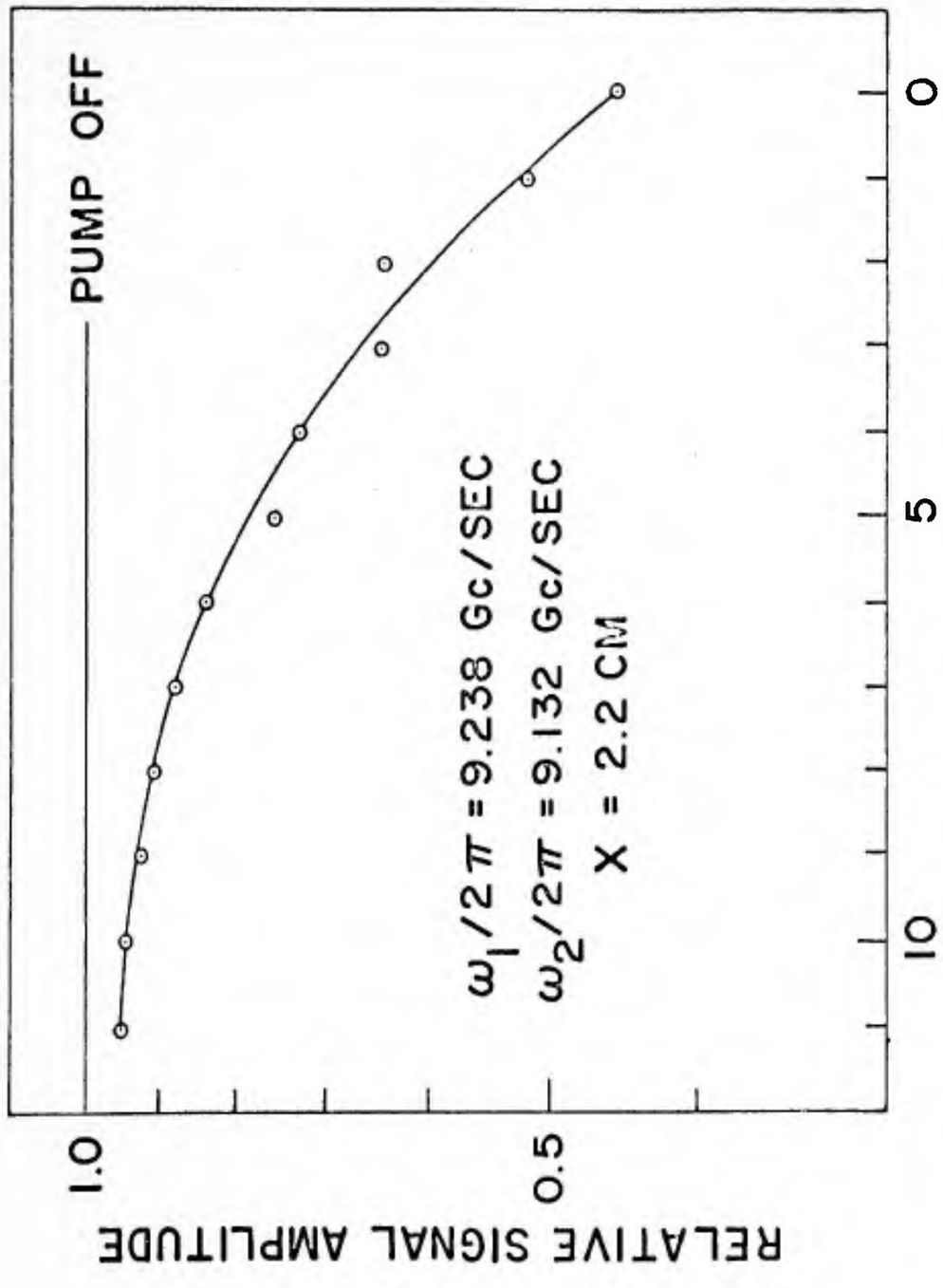

Figure 4 Signal amplitude vs. pump power for input signal level 20 db smaller than maximum pump input level.

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II. SIGNAL VELOCITY OF ACOUSTIC WAVES AND THE ACOUSTIC MASER:

In non-dissipative media, it may be shown that the velocity of energy propagation is equal to the group velocity. However, for wave propagation at frequencies within a narrow absorption band, the group velocity is larger than the characteristic velocity, c , which would be observed in the absence of dissipation. The calculation of the velocity of energy flow (signal velocity) in a medium containing narrow resonance absorption lines was considered by Sommerfeld in 1907.

The actual problem he considered was that of light propagation in a medium consisting of a uniform distribution of Lorentz oscillators. However, the arguments are non-relativistic and apply equally well to any plane wave propagation, provided the definition of c is that given above. Also, Sommerfeld's result that "no signal travels faster than c" is a consequence of causality. As the "Principal of Limiting Distance" it has been shown to be valid for any complex refractive index obeying a causal dispersion relation.

Sommerfeld's calculations were extended by Brillouin and Baerwald, who computed signal velocities valid for small and large propagation distances respectively. The result of these calculations is that the change in signal velocity as a function of frequency is

negative and has the shape of the absorption line. Experimental confirmation of this result was recently reported by Shiren[1] for microwave ultrasonic waves interacting with spin resonance absorption lines.

We are now attempting to make similar measurements of signal velocity for the case of ultrasonic waves propagating in a medium containing an inverted spin population. The interest here arises from the fact that for such interactions the absorption, phase velocity, and group velocity are all inverted with respect to their values in the case of normal populations. However, the signal velocity is expected to be the same as in the absorption case. This is a result of the assumption of micro-causality in the theory and so the experiment provides a possible test of this assumption, however, before any conclusions are drawn from the experimental results, a more detailed theoretical analysis will have to be made.

The experiments so far have been made in MgO containing Fe^{2+} and Ni^{2+} impurities. Adiabatic rapid passage is being used to invert the spin populations. The experimental geometry consists, in addition to the usual ultrasonic generation apparatus, of a cylindrical ceramic cavity resonator operating in the TE_{011} mode. The MgO crystal lies along the cylinder axis, and a coil consisting of several hundred turns of #42 wire is wound around the cavity. By pulsing

1. N. S. Shiren, Phys. Rev., 128, 2103 (1962)

the coil from a thyratron, we have obtained sweep fields of ~400 gauss with a 5 μ sec rise time. In order to invert with such fast sweep rates, high rf fields are required. Therefore, the cavity was driven with a 5 μ sec 100 watt pulse from a magnetron. In this way, the Ni^{2+} line was completely inverted ; however, we were not able to achieve complete inversion of the Fe^{2+} line. The latter has a linewidth of 400 gauss and, therefore, larger sweep fields will be required.

The signal velocity measurements for the interaction with the Ni^{2+} inverted population have, thus far, been inconclusive, the change in velocity being within the noise level. The power amplification of the acoustic wave has, however, been considerable. Gains of ~7 db/cm were obtained. The cw ruby ultrasonic maser reported by Tucker has achieved a gain of only about 1.05 db/cm. The amplification with $MgO : Ni^{2+}$ is large enough to be of technological interest, although it can only be achieved on a pulse basis because of the method used to obtain population inversion. The frequency used was 9.2 Gc.

The apparatus is being modified to achieve larger sweep fields so that velocity measurements may be made when the ultrasonic amplification is occuring in the $MgO : Fe^{2+}$ crystal. Since the Fe^{2+} interaction with the ultrasonic waves is greater than that of Ni^{2+}, larger changes in signal velocity should be produced and a conclusive measurement should be possible.

III PHONON INTERACTIONS IN GERMANIUM

(a) Theory of Phonon Amplification in Germanium

Hutson, Mc Fee and White have amplified ultrasonic waves in the radio frequency range by travelling wave interaction in CdS and other piezoelectric semiconductors. We consider here the amplification of microwave frequency ultrasonic waves, using as the active crystal non-piezoelectric semiconductors such as germanium and silicon. These materials have multi-valley conduction bands which permit a strong interaction between particular acoustic waves and conduction electrons.

It may be asked at the outset whether there is any hope of getting as large an amplification in a non-piezoelectric semiconductor as in a piezoelectric semiconductor. In the former case, the electron-lattice interaction is via the deformation potential, and the electric fields associated with the ultrasonic wave are of order of magnitude

$$E = -\frac{\partial V}{\partial x} = \frac{\partial}{\partial x}(S\bar{\Xi}) = kS_o\bar{\Xi} \quad \exp\ i(kx - \omega t) \quad ,$$

Where k is the wavevector of the strain wave, $S = S_o \exp\ i(kx - \omega t)$, and $\bar{\Xi}$ is the shear deformation potential constant. Electric field/strain is a measure of the electron-phonon coupling ; this quantity $E/S_o = k\bar{\Xi}$ for deformation potentials. At a frequency of

10^{10} sec^{-1}, $k \approx 10^{7}$ m^{-1}. $\overline{\quad}$ is on the order of 10ev for germanium. Thus $E/S_o \approx 10^{8}$ volts/m.

For piezoelectric materials, an equation of state is $D = \epsilon E + eS$, where e is the piezoelectric constant. In an open circuited insulator, $D = 0$, and the quantity $E/S_o = e/\epsilon$. In MKS units, $e \approx .1$ coul./m^2 (typical of GaAs) and $\epsilon \approx 10^{-10}$ farad/m. For piezoelectrics we find $E/S_o \approx 10^{9}$ volts/m. Thus, at 10 Gc/s the electron interaction with the deformation potential fields is typically an order of magnitude less than the interaction with piezoelectric fields. Interaction constants usually enter as squares, so the deformation interaction may be inferior by two orders of magnitude. It seems worthwhile to pursue the study of the deformation potential case for two reasons. First, at higher frequencies, say 100 Gc/s or higher, where amplifiers may be most useful, the deformation interaction becomes comparable to, or exceeds, the piezoelectric interaction. Secondly, there are properties of the crystals other than the coupling constant that enter into the gain. It may be that some of these are more favorable in particular deformation potential materials than in the piezoelectric materials.

Theory:

In their theory of the acoustoelectric effect in Ge[1] Weinreich

et al. (WSW) have analyzed the problem of electron transport in a many-valley semiconductor. WSW were interested in the average (or D. C.) force exerted by a sound wave on conduction electrons. They pointed out that to the extent that the electrons are bunched in the sound wave, there is an interaction between them. It is obvious that the repulsive coulombic effect between these bunched electrons will tend to limit the amount of bunching. This will reduce the amplification that it is possible to achieve.

There is a special case, however, in which the electronic repulsion does not act to reduce the bunching although the density of carriers is large. As pointed out by Holstein(unpublished) , in multi-valley semiconductors certain sound waves destroy the degeneracy of equivalent valleys. An example of this is a shear wave propagating along a (100) direction in Ge. At a given point in real space, the electrons can be subdivided into two classes of particles corresponding to those in the two kinds of valleys made inequivalent by the acoustic strain. At a given point in space, electronic redistribution will occur such that electrons tend to populate the valley whose energy has been lowered , and the population of the raised valley will tend to be depleted by the same amount. There is thus a spatial bunching of electrons in the minima of the wave in each valley, but at each point in real space, the

charge density is uniform. The "movement" of charge is a redistribution in k-space only. In this case, coulombic effects can be ignored to first order. WSW write the equation for the current density in each valley (indicated by ±) as

$$j_{\pm} = -D \left[\frac{\partial n_{\pm}}{\partial x} \pm \left(\beta q n_{\pm}\right) \frac{\partial \phi}{\partial x} \right] \qquad (1)$$

where the second term on the right is the current caused by the electric field of the acoustic waves. If space change entered the problem, this term would be modified to include the electric fields of the bunched charge carried by the wave. The first term is the diffusion current that arises from non-uniform spatial distribution in each valley. $n_{\pm}$ are the charge densities in the valleys, and the Einstein relation for non-degenerate statistics has been used to express the current due to the sonic wave as a diffusion. $q\phi$ is the energy of an electron in the acoustic wave where $q=\Xi/3\sqrt{c}$ is the "acoustic charge" and ϕ is the amplitude of the acoustic wave as defined by WSW. c is an elastic constant. $\beta = (kT)^{-1}$.

To obtain acoustic amplification, the electrons must deliver energy to the sound wave. The energy of the electrons can be provided by an applied electric field. This inclusion of an electric field modifies equation (1) by the addition of a conduction current, $j = \sigma E = n\mu E$

$$j_{\pm} = -D\left[\frac{\partial n_{\pm}}{\partial x} \pm (\beta q n_{\pm})\frac{\partial \phi}{\partial x} - n_{\pm}\, e\beta E\right] \tag{2}$$

Following WSW, we write the continuity equations for the two valleys

$$\frac{\partial n_{\pm}}{\partial t} + \frac{\partial j_{\pm}}{\partial x} = R\left(\begin{matrix}- \rightarrow + \\ + \rightarrow -\end{matrix}\right) \tag{3}$$

where R is the intervalley scattering rate. $n_{\pm}$ is written as the sum of small complex travelling wave perturbation n_1, and a constant background n_o:

$$n_{\pm} = n_o \mp n_1 \exp i(kx - \omega t) \tag{4}$$

We assume that WSW's expression for R (in the absence of applied fields E) also holds in the case of applied fields

$$R(- \rightarrow +) = -(4/3\tau)\left[n_+ - n_o(1 - \beta q\phi)\right] \tag{5}$$

and $R(- \rightarrow +) = -R(+ \rightarrow -)$, where τ is the intervalley scattering time. It is neither necessary nor probably correct to assume that the values of τ in the absence of E are the same as their values in an

applied field. Substituting equations (2), (4) and (5) into equation (3) and solving for n_1, we find

$$n_1 = n_o \beta q \phi_o / (1 + i (\mu E/s - 1) \omega \tau_R) \tag{6}$$

where

$$\tau_R^{-1} = (4/3\tau) + k^2 D , \tag{7}$$

s = velocity of sound and $v = \mu E$ is the electron drift velocity.

The rate of energy transfer from the wave to the particles determines whether the wave is amplified or attenuated by the interaction. The time average energy transfer from the wave to the particles for each valley i can be shown to be

$$\overline{Wi} = s \left\langle \left(- q \frac{\partial \phi}{\partial x} \right) n_i \right\rangle / \langle n_1 \rangle = s \overline{Fi} \tag{8}$$

where $\overline{Fi}$ is the average force of the wave on the particles. (To prove (8), we can either use WSW's equation (2, 6), including the uniform drift current, or apply the general result of wave dynamics which states that the energy of a wave equals the momentum of the wave multiplied by the phase velocity s ; the rate of change of energy is thus s times the rate of change of momentum, or force, as eq. (8)

states). To calculate the time average quantities in eq. (8), we evaluate

$$\left\langle \left(-q\, \partial\phi/\partial x \right) n_i \right\rangle = \tfrac{1}{2}\ \mathrm{Re}\left[\left(-q\, \partial\phi/\partial x \right)\ n_i^{*} \right] \ , \left\langle n_i \right\rangle = n_o \ .$$

Using (4), (6) and assuming a sinusoidal form for ϕ we find that the average energy transfer per particle in each set of valleys is

$$\overline{W} = \overline{W_+} = \overline{W_-} = \frac{q^2 \phi_o^2 \beta \left(1 - v/s \right) \omega^2 \tau_R}{2 \left(1 + (1 - v/s)^2\ \omega^2 \tau_R^{\,2} \right)} \qquad (9)$$

Eq. (9) is the energy transfer <u>from</u> the wave <u>to</u> the particles. $\overline{W} > 0$ for $v<s$, i. e. the wave delivers energy to the particles if the particle velocity v is less than the sound velocity s. This results in an attenuation of the waves. If $v>s$, then $W<0$ and the electrons transfer energy to the wave, which results in an amplification of the waves. Eq. (9) reduces to WSW's equation (3.9) when $v = 0$ (no applied electric field).

The measureable quantity, the attenuation constant α, can be calculated from

$$\begin{aligned} \alpha &= N, \text{ (energy transfer per particle)/incident energy flux} \\ &= N\overline{W} \,/\, \tfrac{1}{2}\, \phi_o^{\,2}\, s \end{aligned} \qquad (10)$$

where N is the density of particles. Substituting (9) in (10) and using the value[1] for $q = \Xi/3s\sqrt{\rho}$ (where s is the velocity of the transverse wave polarized along the <010> direction and propagating in the <100> direction in Ge, Ξ is the shear deformation potential constant, and ρ is the density), the final result is

$$\alpha = \frac{N\Xi^2\beta}{9s^3\rho} \quad \frac{\omega^2\tau_R \quad (1 - v/s)}{\left[1 + (1 - v/s)^2\,\omega^2\tau_R^2\right]} \tag{11}$$

Estimates of the Magnitude of the Amplification:

We estimate the total relaxation time (eq. 7) for Ge at $10^{\circ}K$, using the values $N = 10^{16}$ cm^{-3} (corresponding to .1Ω cm. at room temperature) and the "cold" electron mobility $\mu \approx 10^4$ $cm^2/volt\text{-}sec.$ This gives $Dk^2 \approx 10^{11}$ sec^{-1} for an acoustic frequency of 10^{10} cps. The intervalley scattering rate[1] is $\tau \approx 10^{11}$ sec^{-1}. The total relaxation rate $t_R = Dk^2 + \tau^{-1} \approx 10^{11}$ sec^{-1}. For Ge, Ξ = 16 ev., $s = 5 \times 10^5$ cm/sec & $\rho = s$ gm/cm^3. Substituting these values into eq. (11), we find

$$\alpha \approx 2 \times 10^2 (1 - v/s) \Big/ 1 + .4 (1 - v/s)^2 \text{ nepers/cm.}$$

The maximum amplification occurs when the denominator = 2 or $v \approx 3s$, for which

$$\alpha \text{ max} \approx -200 \text{ nepers/cm}$$

The current density required for maximum amplification is

$$j = Nev = Ne\,(3s) = 3 \times 10^{3}\ \text{amp/cm}^2 .$$

Comparison with Piezoelectric Semiconductors:

The amplification in Ge can be compared to that in a piezoelectric semiconductor using the formulas derived for the latter by White[3]. We quote his eq. (21), which gives the maximum gain (the applied electric field has been optimized):

$$\alpha\ \text{max} = -\frac{e^2}{4\varepsilon c}\ k\ \left(1 + \omega^2/\omega_c\omega_D\right)^{-1}$$

where e is a piezoelectric constant, ε = dielectric constant , $\omega_c = \sigma/\epsilon$ = dielectric relaxation frequency, $\omega_D = s^2/D$, c = elastic constant. Using the value $e^2/2c\varepsilon \approx 10^{-2}$ for CdS given by White and assuming optimum values of ω_c and ω_D , ($\omega_c \approx \omega_D \approx \omega$) , and $\omega = 6 \times 10^{10}\ \text{sec}^{-1}$, we find

$$\alpha\ \text{max} = -\ 200\ \text{nepers/cm.}$$

This similarity of the gains possible with typical piezoelectric and non-piezoelectric semiconductors does not tell the whole story. The power density required to achieve this gain is an important practical consideration. For example, to achieve optimum gain in CdS requires $\omega_c \approx \omega$, which occurs for $\sigma \approx .1\Omega^{-1}\ \text{cm}^{-1}$. The mobility of CdS[4] at the low temperatures required for propagation of microwave phonons is $\mu \approx 5\ \text{cm}^2/\text{volt sec.}$ The optimum density of electrons required is

thus $N \approx 10^{17}$ cm^{-3}. The power density, $P = jE \approx Nes^2/\mu$, is of order 10^4 larger for CdS than for Ge, to get the same gain at microwave frequencies. The low mobility of CdS at low temperature is a serious disadvantage of this material. We can then compare Ge to a piezoelectric material that may have high mobility at low temperatures, e.g., GaAs.

For GaAs, where at low temperatures $\mu = 10^4$ cm^2/volt-sec., and $e^2/2\epsilon c \approx 10^{-3}$, there is a maximum gain of $\alpha \approx -20$ nepers/cm., for a carrier density of $N \approx 10^{14}$ cm^{-3}. This carrier density is that which gives a maximum gain. A gain of 20 nepers/cm. for Ge would require a power density 10-100 times greater than that required for the same gain in GaAs.

Summary:

This study shows that many properties of the amplifier crystal must be considered in evaluating its potentialities as an amplifier. Because microwave phonons propagate only at low temperatures, the mobilities at low temperatures play an important role in determining the power requirements. Thus, although CdS is strongly piezoelectric, its low mobility makes it inferior to Ge. GaAs, on the other hand, seems to be superior to Ge at 10 Gc/s. The Ge amplifier, however,

should be comparable to GaAs at 100 Gc/s because the electron-phonon coupling in Ge increases with frequency.

It should be pointed out that the estimates of the mobilities, relaxation times, etc., used here have been those appropriate to "cold" electrons. In fact, however, in the presence of an applied E field, the electrons will be heated, and there may be considerable changes in the values of the electronic transport parameters. Experiments at the operating conditions will be required to decide which materials are the best amplifiers.

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(b) Experiments on Phonon Amplification in Germanium

Experiments have been conducted to try to observe the phonon amplification predicted by the theory discussed above. As yet, no unambiguous verification has been achieved, although in some cases increased phonon amplitudes have been observed. The difficulty is to obtain a large current density, uniformly distributed in the body of the Ge, and at the same time to maintain a sufficient number of phonons to work with.

The magnitude and uniformity of the current density can be increased by decreasing the cross-sectional area of the samples. Since the total acoustic energy is proportional to the area of the samples, this adversely affects the phonon energy that is observed. We hope to improve the experimental techniques so that an optimum area of samples is achieved. We have also sought to obtain wide area electrical contacts by diffusing As into the surface of the Ge thus making a high conductivity (degenerate) surface. This should improve the uniformity of the current flow which heretofore originated at small area wire contacts soldered to the sample.

Thus far, our largest current densities have been obtained by using a modified SKL pulser. This pulser has delivered 8 amps. peak current,

which for our samples corresponds to a current density of $j = 200$ amp/cm^2. For the sample in this experiment, the theory predicts that about 1000 amp/cm^2 is more nearly the current density required by its impurity content.

(c) Temperature Dependence of Ultrasonic Attenuation

We have made some preliminary measurements in a program of study of the temperature dependence of ultrasonic attenuation. To perform these experiments, ferromagnetic film transducers are evaporated on suitably oriented and polished single crystal samples. The use of ferromagnetic film transducers has the advantages that (1) a variety of modes of vibration can be obtained without changing the transducer (2) the transducer is practically temperature independent and lossless. Quartz transducers, by comparison, become lossy at about 20°K and are opaque above 40°K. The temperature of the samples is varied and the amplitudes of the phonon echoes are observed.

It had been found earlier (by Bömmel, Dransfeld, and Jacobsen) that the attenuation of microwave phonons in quartz was low for temperatures below about 20°K, but in the range of temperatures above 20°K the attenuation increased considerably. This early work was confined to the study of quartz. Thus far, we have re-examined the attenuation

in quartz in order to check our methods and find fair agreement with the published results.

We have made a preliminary measurement of the temperature dependence of attenuation in Ge. The attenuation of the shear wave along the (100) axis in Ge begins at a higher temperature than for shear waves in quartz. For Ge, 3db of attenuation occurs at about $45^{\circ}K$. For x-cut quartz, 3db attenuation for the slow shear wave occurs at about $20^{\circ}K$, and for the fast shear wave, 3db attenuation occurs at about $35^{\circ}K$.

Improvements of the experimental apparatus have been completed which will permit more accurate measurements of the temperature in the range of $4^{\circ}K$ to $77^{\circ}K$. We have begun preparation of samples of GaAs, silicon and CaF_2, and we have available samples of Ge and MgO.

(d) Phonon-Phonon Interaction in Germanium

Non-linear phonon propagation at 9 Gc/s has been reported in MgO; similar effects have also been observed in Ge. It was noticed that the amplitude of longitudinal phonons propagating along the (100) axis in Ge did not increase linearly with the input power. We believe that this non-linearity is probably caused by scattering of 9 Gc/s phonons into 18 Gc/s phonons (harmonic generation). The same

experiment was performed using longitudinal waves along the (110) axis of Ge , but there was no observable non-linearity. Therefore, we believe that the anharmonicity for longitudinal waves propagating along the (110) axis is less than that of longitudinal waves along (100) by at least a factor of three.

This observation of non-linearity can be compared with the measurements of the temperature dependence of phonon attenuation. It is commonly assumed that scattering with thermal phonons dominates the low level attenuation of microwave phonons at temperatures of about $50^{o}K$. Since the anharmonicity of the lattice potential energy is responsible for both effects, one would thus expect a correlation between attenuation and non-linearity: materials for which microwave phonon anharmonicities are large would also be expected to show large microwave phonon attenuation to lower temperatures than those for which the anharmonicities are small. This expectation is <u>not</u> borne out by our relative observations on quartz and Ge. The anharmonicities in quartz seem to be quite small ; despite the relatively large phonon amplitudes possible in quartz , no non-linear effects have yet been observed. On the other hand, such non-linearities in Ge have probably been observed. On this basis, one would guess that quartz should be more harmonic than Ge with respect to thermal phonon scattering and be lossless to higher

temperatures than Ge. As pointed out in the previous section, we observed the opposite to be true: Ge is less lossy than quartz to higher temperatures. The implication is that properties other than the harmonicity of materials must be considered before even a qualitative understanding of the temperature dependence of the phonon attenuation is possible.

Conclusions

Amplification of 10,000 Mc ultrasonic waves should be observable by interaction with drifting conduction electrons in germanium. The use of germanium as compared to CdS or GaAs should become more advantageous as the frequency of the ultrasonic waves is increased. Various experiments on germanium are planned in order to more fully understand its acoustic properties.

IV. PROPAGATION EFFECTS.

The propagation of acoustic waves in crystals having an odd-fold axis of rotation symmetry (e.g. the [0, 0, 1] axis in quartz or the [1, 1, 1] axis in cubic crystals) should show certain unique effects rather similar to those observed with optical radiation in biaxial crystals. In particular, a phenomenon rather similar to internal conical refraction of light in such crystals has been predicted [P. C. Waterman, Phys. Rev. 113 , 1240 (1959)] . Thus, if transverse acoustic waves are sent into a quartz crystal by a transducer such that the equal phase wave fronts are normal to the z - axis, the ray (or energy) direction is not along the z - axis, but is instead inclined at an angle θ which is about $\theta = 14^{\circ}$ for quartz. Waterman has predicted from a first order theory that the acoustic energy will be found uniformly distributed on the cone about the z - axis having this angle if the acoustic waves are unpolarized.

However, we believe that this interpretation is not quite correct because the first order theory ignores for small θ , the variation of the elastic constants with the azimuthal angle, ϕ. Our interpretation is that the acoustic radiation will not be found uniformly over the cone, but instead will preferentially be located at the three azimuthal positions $\phi = 90^{\circ}$, 210°, and 330°. Inasmuch as this prediction is different from Waterman's and also seems to disagree with a possibly incorrect experiment [J. deKlerk and M. J. P. Musgrove, Proc. Phys. Soc.

B 68, 81 (1955)], it would appear to be worthwhile to check this experimentally.

We have also arrived at an understanding of the type of effects to be expected when the phase propagation direction is somewhat misaligned from the [0, 0, 1] direction. The whole field of double refraction of acoustical waves in crystals appears to have been relatively unexplored both experimentally and theoretically to date. Therefore experimental work on these double refraction effects is contemplated, but not immediately planned.

CONCLUSIONS

The large power conversions observed in the parametric up-conversion experiment have demonstrated the potential practical importance of these effects and have also demonstrated that down-conversion and parametric amplification should also be feasible.

The greatly increased gain observed for the acoustic maser utilizing Ni^{++} ions is also a significant step in the understanding and use of acoustic wave interactions.

The other phases of the program are proceeding satisfactorily.

PROGRAM FOR THE NEXT INTERVAL

A major objective is to accomplish parametric down-conversion and amplification of acoustic wave energy. It is planned to convert the acoustic energy produced into microwave energy by a piezoelectric transducer and to detect it externally to the cryostat.

Further work is planned on the measurement of the signal velocity of a pulse of acoustic waves in the presence of an inverted population of paramagnetic ions. The Fe^{++} ion will be used in MgO because the interaction should be greater than for the Ni^{++}.

Further experimental work is planned on the interaction of electrons and phonons in germanium with the objective of clearly demonstrating travelling wave amplification.

A systematic study of the acoustic attenuation of various materials will be initiated. The temperature dependence will be investigated as well as the effects of different directions of propagation and polarization. The thin film ferromagnetic transducer will be used.

IDENTIFICATION OF KEY PERSONNEL

A. H. Nethercot - Research Staff Member 130 hours

Dr. Nethercot has been engaged in research in various aspects of solid state physics and of microwaves.

Dr. Nethercot received the PhD. degree in Physics from the University of Michigan in 1950. From 1950 to 1957, he was a Research Associate at Columbia Univesity and was closely associated with Professor C. H. Townes. His primary duties were developing millimeter wave sources and circuitry and utilizing them in various molecular and solid state investigations.

Since joining IBM in 1957, Dr. Nethercot has studied the switching behavior of subharmonic oscillators, the switching speed of superconducting metals, photoconductive mixing of laser beams, and the behavior of heat pulses in insulators at low temperatures.

Dr. Nethercot has authored or co-authored some eighteen published papers. Dr. Nethercot is currently in charge of a group investigating various optical and microwave effects in solids, particularly in microwave ultrasonics and in the modulating and mixing of light.

N. S. Shiren - Research Staff Member 512 hours

Dr. Shiren has been active in the field of microwave ultrasonics

for approximately six years. He received his PhD. from Stanford University in 1956, having completed his thesis work in high energy Physics in 1951. He then worked in the field of under-water sound detection and in noise theory at the Hudson Laboratories of Columbia University from 1950 to 1955. In 1955, he joined the General Electric Research Laboratories, working on electron spin resonance and on microwave ultrasonics. He joined IBM in 1961 and continued his work on microwave ultrasonics.

Dr. Shiren has contributed particularly greatly to the understanding of the interactions of phonons with paramagnetic ions. and he has many noteworthy papers in this field. He has published some twelve scientific papers on this and on other subjects.

M. Pomerantz - Research Staff Member 353 hours

Dr. Pomerantz studied at the University of California in the allied fields of spin resonance and quadrupole resonance and received his PhD. degree in Physics in 1958. Following a post-doctoral appointment there, he did further post-doctoral work in this field in France under Professor A. Abragam. He joined IBM in 1960 and has concentrated since then principally on microwave ultrasonic investigations. His particular interests and contributions have been on the interactions of ferromagnetic spin wave resonances with phonons and on the

interactions of phonons with conduction electrons in semiconductors. He has published a total of six papers in these various fields.

ABSTRACT CARDS

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