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Excitation and Attenuation of Hypersonic Waves in Quartz

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A method for the generation and detection of hypersonic waves, which has only been briefly described earlier, together with some absorption measurements in quartz, is discussed in some detail. Further measurements of the hypersonic absorption in quartz at different crystal orientations and after neutron irradiation are reported. The results are in qualitative agreement with a phonon-phonon relaxation process.

INTRODUCTION

We shall follow Rao and others and shall call acoustic waves at frequencies above 10^9 cps hypersonic waves, distinguishing them from sonic and ultrasonic waves at lower frequencies. At low temperature the frequencies of the thermal vibrations, kT/h, are close to or within the hypersonic frequency range. It is obviously of great interest for the study of phonon interactions in solids and liquids to extend acoustic research into this region, where the only information so far available comes from experiments with noncoherent thermal phonons.

The highest acoustical frequencies generated until recently by the standard means of driving thin piezoelectric quartz plates in a high harmonic resonance mode were about 1300 Mc/sec by Ringo *et al.*,[1] and by Sokolov.[2] For frequencies higher than this the plates would have to be extremely thin.

In contrast to these resonance techniques with thin plates, Baranskii[3] placed a very thick quartz plate, in fact several thousand acoustic wavelengths thick, into a high-frequency electric field and could observe longitudinal sound waves by an optical method up to frequencies of 2000 Mc/sec.

As demonstrated by the authors,[4] the results of Baranskii can be explained by assuming that traveling hypersonic waves are excited at the surfaces of the piezoelectric crystal which are exposed to the electric field. More generally it can be shown that ultrasonic waves will always be excited at spatial "discontinuities" of an alternating piezoelectric stress. This is just another statement of the fact that the effect of a uniform piezoelectric stress in a crystal is equivalent to a pair of external forces acting on the "discontinuities," for example a free surface. Such a free surface in a high-frequency electric field can therefore be regarded as a transducer of hypersonic waves while the rest of the crystal acts only as a transmission medium. The inverse effect, namely the reconversion of acoustic into electromagnetic energy which was observed at the same time[4] provides a very sensitive means for the detection of hypersonic waves, applicable to higher frequencies than the optical method.

The actual apparatus consisted of two identical cavities, serving as transmitter and receiver, respectively, acoustically coupled by a piezoelectric quartz rod as will be described in detail later on. With this arrangement we were able to excite and detect hypersonic waves up to 2500 Mc/sec at room temperature and up to 4000 Mc/sec at low temperatures. Very recently Jacobsen,[5] using a very similar apparatus, reported the generation and detection of hypersonic waves at 9400 Mc/sec.

A more detailed investigation of the absorption of hypersonic waves as a function of temperature revealed the following results: a rather large and temperature independent absorption above about 60°K which decreases very rapidly as the temperature is lowered, with only a very small fraction of the high temperature value left below 20°K. The absorption has dropped to about half of its original value at a temperature where the mean free path of the thermal phonons—as derived from heat conductivity data—is of the order of the acoustic wavelength.

In this paper we present some further experiments on the hypersonic absorption in quartz at different crystal orientations and after various doses of neutron irradiation. Our results are in qualitative agreement with a phonon-phonon relaxation mechanism of acoustic absorption which we reported previously[6] and which will be discussed here in more detail.

I. THE EXCITATION OF HYPERSONIC WAVES

In this paragraph we shall try to give a simplified description of the excitation process. Let us consider the arrangement shown in Fig. 1. It consists of the reentrant cavity and the piezoelectric quartz rod, with a cross section q, of which only a small volume shall be inside the cavity. The electric field of the cavity is mainly concentrated in this volume V. The rod axis is parallel to the crystalline x axis. The x component of the field is strongest at the quartz surface, $x=0$, and decays for larger distances x. As will be evident, the form of this decay is of little importance, and for

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[1] G. R. Ringo *et al.*, Phys. Rev. **72**, 87 (1947).
[2] C. Ya. Sokolov, Uspekhi. Fiz. Nauk. **40**, 3 (1950).
[3] K. N. Baranskii, Doklady Akad. Nauk. S.S.S.R. **114**, 517 (1957) [translation: Soviet Phys.—Doklady **2**, 237 (1958)].
[4] H. Bömmel and K. Dransfeld, Phys. Rev. Letters **1**, 234 (1958); **2**, 298 (1959).
[5] E. H. Jacobsen, Phys. Rev. Letters **2**, 249 (1959).
[6] H. Bömmel and K. Dransfeld, Bull. Am. Phys. Soc. **4**, 226 (1959).

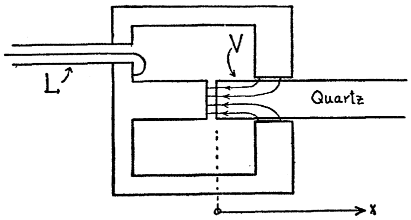

FIG. 1. Cavity with quartz rod, the volume V of which is exposed to the electric field. Lead L critically coupled to cavity.

simplicity we will assume an exponential decay $e^{-\psi x}$. Since the field also oscillates with the resonance frequency $\omega/2\pi$ of the cavity, the x component of the field in the quartz is

$$E = E_0 e^{-\psi x} \cos(\omega t). \tag{1}$$

If a power P is fed into the cavity by means of a matched coaxial lead and if only the x components of the field are taken into account, the energy density of the field can be expressed by

$$\epsilon E_0^2/8\pi = PQ_0/\omega V, \tag{2}$$

where ϵ is the dielectric constant of quartz, Q_0 the quality factor of the unloaded cavity and V the suitably defined "effective" volume of the rod exposed to the electric field. Since the crystal is an x cut—the field causes a piezoelectric stress which in the absence of any strain—is

$$X_x^P = d_{11} c_{11} E, \tag{3}$$

where d_{11} and c_{11} are the appropriate piezoelectric and elastic constants. At the free boundary $x=0$, the total stress X_x must disappear, hence we have a strain

$$x_x = -d_{11} E.$$

The simplest solution, satisfying this boundary condition, is a traveling wave of the form

$$x_x = -d_{11} E_0 \cos(\omega t - 2\pi x/\lambda) e^{-\alpha x},$$

where the acoustical wavelength λ and the absorption coefficient α are assumed to be field independent. The energy transferred from the electric field to such an acoustic wave equals the work done by the strain x_x against the piezoelectric stress X_x^P, and is per period, integrated over the whole crystal

$$F_\tau = q \int_{t=0}^{\tau} \int_{x=0}^{\infty} X_x^P \dot{x}_x dx dt.$$

Using (1), (3), (4) this yields

$$F_\tau = d_{11}^2 c_{11} E_0^2 \lambda q/2 \quad \text{for} \quad (\alpha+\psi)^2 \ll k^2, \tag{5}$$

which is equivalent to the work done in a surface layer of one wavelength thickness. We like to point out, that the amount of energy transferred is quite independent of the field decay across the rest of the sample, and of the acoustic absorption, as long as absorption and decay are small per wavelength. The energy converted per second is accordingly

$$F = \nu F_\tau = (4\pi d_{11}^2 c_{11}/\epsilon)(\epsilon E_0^2/8\pi) v q \quad (v = \text{sound velocity}).$$

Introducing the "piezoelectric coupling factor" $C_x^2 = 4\pi d_{11}^2 c_{11}/\epsilon$, and using (2), this finally becomes

$$F = P(C_x^2 Q_0 \lambda q/2\pi V) = PA_{12}. \tag{6}$$

A_{12} is the fraction of the incident electrical power converted into acoustical energy.

Although the above has been derived for longitudinal waves, the same treatment can be applied to transverse waves. It has already been reported that in order to generate transverse waves in quartz with an arrangement like the one described above one has to use special crystal cuts known as the AC and BC cuts. In these two cuts the rod axis makes an angle of $-59°$ and $+31°$, respectively with the crystallographic z axis. It can be shown that these are the only two orientations of a rotated y cut where transverse waves have at the same time no cross coupling to other modes and are not affected by acoustical birefringence, i.e., where the energy travels parallel to the normal of the wave front. The phenomenon of acoustical birefringence in anisotropic media has been investigated by several authors[7] and is well known. For the generation of transverse waves in the conventional ultrasonic frequency range one uses generally y-cut quartz plates. It can easily be calculated that in a rod whose length is parallel to the crystallographic y axis the acoustical energy will travel in a direction making an angle of about 23° with the normal to the wave front, thus y-cut rods would not be suitable for hypersonic experiments as described above.

II. THE DETECTION OF HYPERSONIC WAVES

The arrangement of Fig. 1 can also be used for the detection of hypersonic waves, i.e., the reconversion of acoustical into electromagnetic energy. It is possible to estimate the efficiency of this process by means of the following thermodynamical argument. The density of longitudinal acoustic modes in the quartz is

$$\Delta Z = 4\pi(\nu^2/v^3)\Delta\nu,$$

at a frequency ν and within a frequency interval $\Delta\nu$ as defined by the cavity. The sound velocities v are assumed to be isotropic for this argument.

Energy exchange between acoustic waves in the rod and the electric field of the cavity will be only noticeable if a large enough area of the quartz surface is vibrating in the same phase. This is the case only for acoustic waves with an angle of incidence smaller than $\lambda/\pi R$, (λ acoustic wavelength, R radius of the quartz rod). Therefore only modes propagating within the solid angle Ω about the rod axis, where

$$\Omega = (4/\pi)(\lambda^2/R^2),$$

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[7] J. W. Price and H. B. Huntington, J. Acoust. Soc. Am. 22, 32 (1950).

can contribute to the energy exchange. Their density is

$$\Delta Z_\Omega = \Omega \Delta Z = \Delta\nu/\pi R^2 v.$$

As each mode carries the energy kT, the effective thermal power F incident on the quartz surface of cross section πR^2 is

$$F = \Delta Z_\Omega kTv\pi R^2 = kT\Delta\nu.$$

Now let us consider the quartz to be in thermal equilibrium with the resistance representing the cavity losses and with the resistance terminating the output lead. See Fig. 2. In equilibrium, the quartz will gain as much energy from both resistances as it loses to them. From the matched output lead the power $kT\Delta\nu$ is fed into the cavity. As we have seen (6) the fraction A_{12} is acoustically transmitted into the quartz. The same energy is incident from the resistance representing the cavity losses. Thus the total energy fed into the cavity is $2kT\Delta\nu$ and the energy transmitted into the quartz rod is

$$E_{12} = 2kT\Delta\nu A_{12},$$

Vice versa, in thermal equilibrium, the same amount E_{12} must be converted from acoustical into electromagnetic energy and fed into the two resistances. If we define with A_{21} the fraction of the incident acoustical power, which can be electrically coupled out and since for critical coupling the same fraction is dissipated in the cavity losses, we have for the power leaving the quartz rod

$$E_{21} = 2kT\Delta\nu A_{21}.$$

From the equilibrium condition $E_{12} = E_{21}$ it follows: $A_{12} = A_{21}$, which means that the efficiency for detection is the same as the efficiency of excitation, and according to (6)

$$\frac{\text{acoustic output}}{\text{electric input}} = \frac{\text{electric output}}{\text{acoustic input}} = (C^2 Q_0 \lambda q / 2\pi V).$$

Because of the dependence on the Q value the sensitivity might in practice be considerably improved by using at least partially superconductive cavities at low temperatures.

III. THE APPARATUS

For the absorption measurements the arrangement shown in Fig. 3 was used between temperatures of 4°K and 300°K. One of the two coaxial cavities with an electrically coupled input lead served as transmitter while the other one with an output lead was the receiver. Both cavities were not only acoustically coupled by the quartz rod but also electrically by a variable coupling loop ABC described later. Since the quartz rod was surrounded by two narrow metal tubes, no detectable electromagnetic power leaked out of the cavities along the rod. The quartz rods were 25 mm long, 3-mm diameter with their two end faces polished optically flat and parallel. If the two end faces were not originally parallel enough or if by a small stress the rod was slightly bent, one could observe a nonexponential decay pattern of the acoustic echoes, familiar from ultrasonic experiments. The central section of the rod was used for the attachment of a thermocouple or carbon resistance thermometer. In order to ensure reliable temperature measurements, and also to keep the cooling liquid out of the cavity, the whole apparatus was enclosed by a tight brass can which was evacuated for the experiment. The whole setup could be cooled down to a temperature of 4°K.

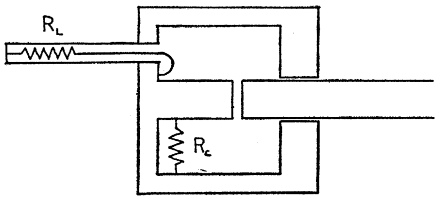

FIG. 2. Cavity with quartz rod. R_L and R_C are resistances representing losses due to the coupling lead and due to dissipation in the cavity, respectively.

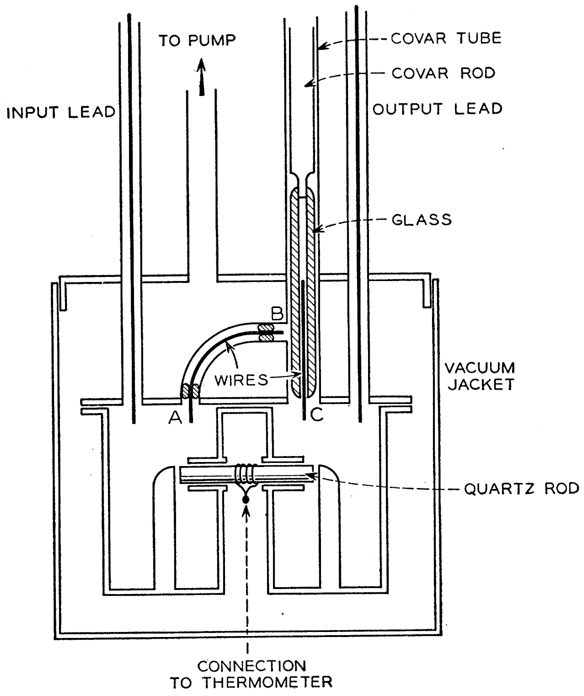

FIG. 3. Experimental arrangement.

The transmitter cavity was coupled to a pulsed high-frequency generator of less than one watt peak power up to a frequency of 4000 Mc/sec, and with a pulse width of 2 μsec. The average power was kept below 1 mw and did not cause a detectable heating of the sample. The signal pulses from the receiver cavity were amplified and displayed on an oscilloscope.

The variable electrical coupling mentioned above consisted of two parts as indicated in Fig. 3: one short

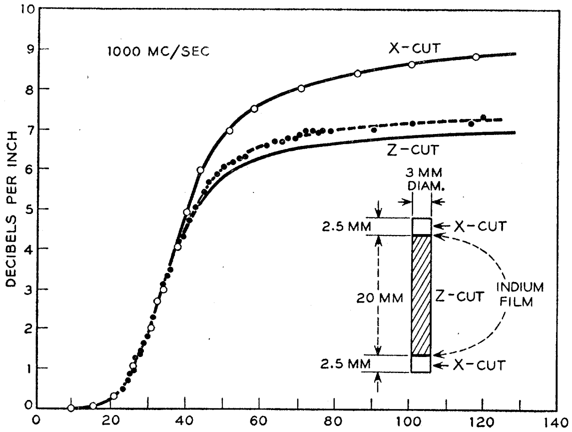

FIG. 4. Absorption of longitudinal waves propagating along the x and z direction.

coaxial bend *A-B* which was rigid with respect to the cavity, and a straight coaxial section *B-C* which could be lowered into the cavity for increased coupling. This latter section was—by means of an isolating piece of glass—fused to a long covar rod which moved inside a covar tube extending out of the cryostat. Its relative motion could be accurately controlled at room temperatures by means of a micrometer screw. Special care was taken to minimize the temperature dependence of the coupling due to differential expansion of the parts involved. Since the amount of power coupled over was very small it did not affect the Q value or the resonance frequency of the cavities.

This cross coupling between both cavities allowed entry into the receiver cavity of a small electrical pulse before the arrival of the first acoustic pulse. On the oscilloscope the first electric and the second delayed acoustic pulse could be seen together. For taking a measurement both pulses were set equal by adjusting the variable coupling which was calibrated in decibels, and at the same time measuring the temperature of the quartz. In this way the *relative* change in absorption with changing temperatures could be observed between helium and room temperature. The *absolute* magnitude of the absorption could be derived from the decay pattern of the acoustic echoes at the lowest temperatures. Since both the electrical and the acoustic signal depend on the same electrical field inside the cavities, this comparison type of measurement was not disturbed by changes of the Q values of the cavities with temperature nor by instabilities of the electronic equipment. Also it was possible to extend the measurements to high absorptions, where only one or two acoustic echoes were visible.

IV. EXPERIMENTAL RESULTS

While in our previous experiments[4] the hypersonic absorption was studied for longitudinal and transverse waves at various frequencies up to 4000 Mc/sec, we are here reporting measurements at one frequency of 1000 Mc/sec but on quartz specimens of different crystal orientations and after various degrees of neutron irradiation.

With the exception of the z-cut crystal all specimens had the same length of 25 mm and diameter of 3 mm. Along the z axis the experiments were made with a compound rod consisting of three parts as shown in Fig. 4. A rod of 20 mm length with its rod axis parallel to the z axis was bonded to two 2.5 mm long x-cut crystals for transmitting and receiving longitudinal waves. After bonding the whole unit had the same dimensions as the other quartz rods. For making the acoustic bonds between two surfaces, both surfaces were polished optically flat, plated in vacuo with a film of indium and then pressed together for a few hours at elevated temperatures. These thermal compression bonds had good mechanical strength in the whole temperature range and showed a satisfactory acoustic transmission at frequencies up to 4000 Mc/sec.

In the Figs. 4–7 for the various specimens the temperature dependent part of the absorption is plotted versus the temperature. The temperature independent part or so-called "residual" absorption (not included in the diagrams) observable at the lowest temperatures was in general less than 1 db/inch, but increased after neutron irradiation of about 10^{19} neutrons/cm^2 to several db/inch. Before irradiation the residual absorption varied from sample to sample often by a factor of two in contrast to the temperature dependent part of the absorption, which for the same specimens (in-

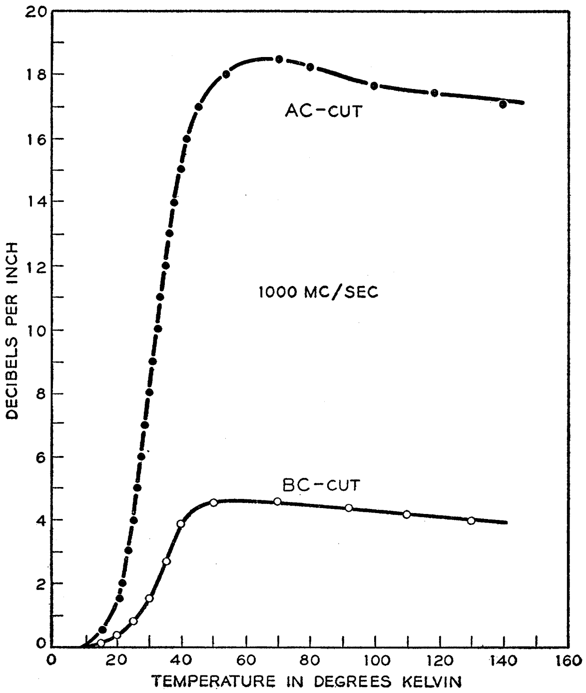

FIG. 5. Absorption of transverse waves propagating along the AC and BC direction.

cluding a crystal of artificial quartz) showed a variation of less than 20% if any. This indicated that the residual part is more structure sensitive than the temperature dependent absorption, at least for small degrees of crystal imperfection.

The absorption of transverse waves propagating in two different crystalline directions, specified as *AC* and *BC* (see first part of this paper for definition) is shown in Fig. 5. The absorption of longitudinal waves along the x and z axis is shown in Fig. 4. Here the points are the measured data of the compound rod including the x-cut pieces, while the full curve has been calculated from it and represents the absorption of a z-cut crystal only of the standard length of 25 mm. The indium bonds apparently did not contribute a temperature dependent absorption, as expected for the used film thickness. Finally, in Figs. 6 and 7, the effect of neutron irradiation is shown for transverse and longitudinal waves. The quartzes were irradiated in the Brookhaven pile.

V. DISCUSSION

Phenomenologically the absorption of sound can be related to a shear viscosity η_S and a compressional viscosity η_K. For transverse waves the absorption per cm is usually described as

$$\alpha_T=(\omega^2/2\rho v_T^3)\eta_S, \tag{7}$$

and for longitudinal waves

$$\alpha_L=(\omega^2/2\rho v_L^3)(\tfrac{4}{3}\eta_S+\eta_K). \tag{8}$$

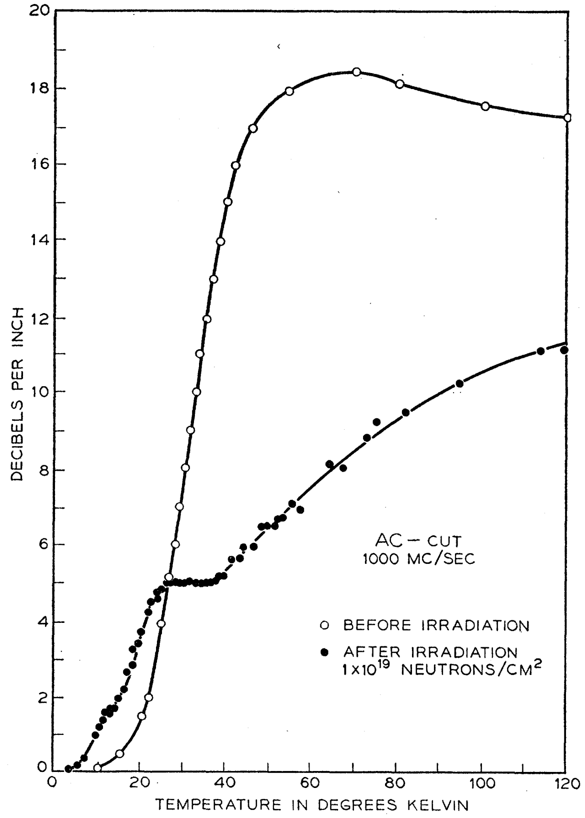

FIG. 6. Absorption of transverse waves propagating along *AC* direction before and after neutron irradiation.

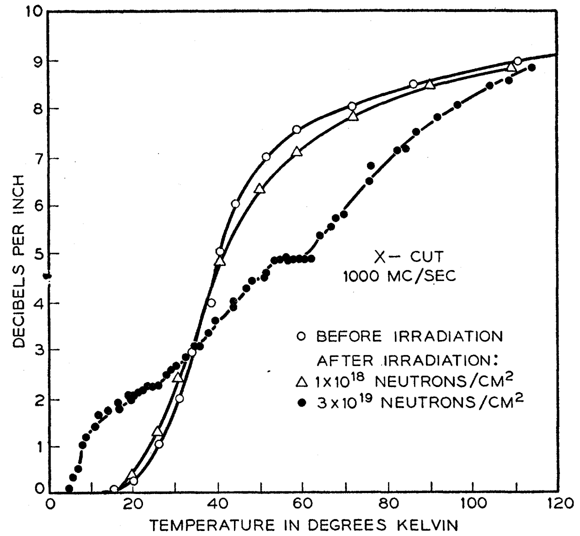

FIG. 7. Absorption of longitudinal waves propagating along the x direction before and after various degrees of neutron irradiation.

It is interesting to compare the viscosities corresponding to the absorption of waves, which propagate in different crystal directions. The following tables show the transverse and longitudinal absorptions at 140°K in two different directions in each case, as well as the product of αv^3. From Table I one sees that the shear viscosity $\eta_S\simeq\alpha_T v_T^3$ is of about the same magnitude for the two directions studied and from Table II it becomes apparent that also the combined shear and compressional viscosity $(\frac{4}{3}\eta_S+\eta_K)\simeq\alpha_L v_L^3$ is practically the same along the x and z axes.

The ratio between compressional and shear viscosity is according to Eqs. (7) and (8)

$$\eta_K/\eta_S=[(\alpha_L v_L^3)/(\alpha_T v_T^3)]-\tfrac{4}{3}.$$

Using average values for $\alpha_L v_L^3$ and $\alpha_T v_T^3$ from Table I, we find

$$\eta_K/\eta_S\cong1.6.$$

This means if we relate the hypersonic absorption to a shear viscosity and to a compressional viscosity that both viscosities are of about the same magnitude and have no strong dependence on the direction of propagation in the crystal, for the four directions studied.

A few experiments were carried out to investigate

TABLE I. The absorption of transverse waves at 140°K.[a]

Transverse waves	Velocity v_T	Abs. α_T	$\eta_S\approx\alpha_T v_T^3$
AC cut	3.32×10^5	17 db/inch	6.3×10^{17}
BC cut	5.04×10^5	3.9 db/inch	5.0×10^{17}

[a] $\omega=6.3\times10^9$, $\rho=2.6$.

TABLE II. The absorption of longitudinal waves at 140°K.[a]

Longitudinal waves	Velocity v_L	α_L	$\alpha_L v_L^3$
X cut	5.6×10^5	9.0 db/inch	15.8×10^{17}
Z cut	6.3×10^5	7.0 db/inch	17.5×10^{17}

[a] $\omega=6.3\times10^9$, $\rho=2.6$.

whether the temperature dependent part of the absorption could be influenced by mechanical or thermal treatment of the quartz rods or if it was related to the origin to the samples. Therefore, one quartz rod was twisted close to fracture for 20 hours at a temperature of 200°C, another was annealed for 112 hours at 500°C. In both cases the absorption at room temperature measured at 1000 Mc was within the accuracy (10%), the same as before the treatment. Furthermore no difference was found within the same limit between a material of different origin including one artificial quartz, (kindly supplied by King of these Labs.). Similarly no effect could be observed after prolonged x-ray treatment. Although these results seem to indicate that the temperature dependent absorption is an intrinsic property of the ideal crystal we are aware of the fact that it is difficult to estimate how much our samples were affected by the treatments.

A well-known absorption process is the one due to heat conduction which arises from the irreversible heat exchange between the compressed and rarefied regions of a longitudinal wave. This absorption has been calculated, for a wave along the x direction in quartz, to be

$$\alpha(\mathrm{cm}^{-1})=\frac{\omega^2}{2\rho v^3}\left(\frac{K}{C_P}\,\frac{c_{11}^{\mathrm{ad}}-c_{11}^{\mathrm{is}}}{c_{11}^{\mathrm{ad}}}\right),$$

where ω=angular frequency, ρ=density, v=longitudinal sound velocity, K=heat conductivity, C_P =specific heat per gram, c_{11}^{ad}=adiabatic value of the elastic constant, and c_{11}^{is} its isothermal value. $c_{11}^{\mathrm{ad}}-c_{11}^{\mathrm{is}}$ can be calculated from the expansion coefficients β_X and β_Z along the X and Z axis, respectively:

$$c_{11}^{\mathrm{ad}}-c_{11}^{\mathrm{is}}=\frac{[\beta_X(c_{11}+c_{12})+\beta_Z c_{13}]^2 T}{\rho C_P},$$

(T=absolute temperature). If one puts actual numbers into this formula it can be seen that this source of attenuation is negligible in quartz at 1000 Mc/sec. This process would, of course, not account at all for the absorption of transverse waves.

It was Akhiezer[8] who introduced a new process quite different from the heat conduction considered above, which could cause the absorption of sound in an ideal crystal, even in the absence of a temperature gradient. Pomeranchuck[9] later on gave a more generalized treatment of this process. Akhiezer pointed out that a sound wave passing through a crystal would cause a disturbance of the distribution of thermal phonons, so that the phonons corresponding to the thermal lattice vibrations, have no longer an equilibrium Planck distribution. The re-establishment of equilibrium in the phonon gas requires an increase of entropy and therefore leads to the absorption of sound at finite temperatures. It will be shown here that it is possible by making a few simplifying assuptions to arrive at an expression for the absorption of sound, which is in fair agreement with our experiments, as far as a comparison is possible at present. To our knowledge no experimental results have been reported previously as an evidence for Akhiezer's process.

We will consider here only temperatures high enough so that the thermal frequencies kT/h are considerably higher than the acoustic frequency. In this case the wavelength of a thermal phonon is so much shorter than the sound wavelength that we can consider the phonons to travel through a uniform medium the strain of which is slowly modulated by the sound wave.

Due to the anharmonicity of the lattice the strain of the sound field, for example a compression, causes a change of the phonon velocity. This change will, in general, differ for phonons with different polarization vector and propagation direction. (In the following we will call a group of phonons with a common polarization j and propagation vector k a "branch".)

The relation between a compression in terms of the relative density change $\Delta\rho/\rho$ and the velocity change $(\Delta v/v)_{jk}$ for a particular branch can be defined by γ_{jk}

$$(\Delta v/v)_{jk}=\gamma_{jk}(\Delta\rho/\rho).$$

We shall call here γ_{jk} the "Grüneisen"-constant of the branch (jk). There are only very few γ values known for quartz. For example Susse[10] measured the effect of hydrostatic pressure[11] on the velocity of transverse waves along the AT, BT, and y direction in quartz, and found

$$(\gamma_{AT}/\gamma_{BT})=-0.53,$$
$$(\gamma_{BT}/\gamma_y)=+0.38,$$
$$(\gamma_y/\gamma_{AT})=-5.0,$$

the absolute magnitude of γ_y being of the order of unity. This may serve as an example for the appreciable variation of Grüneisen's constant between different branches.

We shall neglect dispersion and assume that the γ values do not depend on frequency. Phonons of *all frequencies* which belong to one and the same branch will therefore during a compression change their velocity by the same amount. It can be shown quite generally that a common velocity change $(\Delta v/v)$ for all

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[8] A. Akhiezer, J. Phys. (U.S.S.R.) 1, 277 (1939).

[9] I. J. Pomeranchuck, J. Phys. (U.S.S.R.) 4, 529 (1941).

[10] C. Susse, J. phys. radium 16, 348 (1955).

[11] As yet there are no data available about the effect of shear stress.

phonons of one branch does not disturb the thermodynamic equilibrium within this branch, and it is therefore still proper to attribute a temperature to it. It can be shown further that a compression changes this temperature by an amount which, for the branch (jk), is equal to

$$(\Delta T/T)_{jk}=(\Delta v/v)_{jk}=\gamma_{jk}(\Delta\rho/\rho),$$

if during the compression collisions with phonons of other branches can be neglected. Because of the variety of γ values it is obvious, that after a compression each branch has, in general, a different temperature. In particular, since γ can have negative and positive values, some branches will be cooled down while others are heated up.

Lacking sufficient data on the γ values we can, at present, discuss only the general features of the absorption process. Therefore we shall, at this point, make a very simplifying assumption: after compression we assign all branches to two different groups with different average temperatures, and comparable specific heats. The first group shall contain all branches which have undergone rather large positive temperature changes $(\Delta T/T)_1$ while the second group is made up of all the other branches, i.e., those with small or negative changes $(\Delta T/T)_2$. The average relative temperature difference between both groups is

$$(\Delta T/T)_{\text{Av}}=(\Delta T/T)_1-(\Delta T/T)_2,$$

and defines an average γ value

$$\gamma_{\text{Av}}=\left(\frac{\Delta T}{T}\right)_{\text{Av}}\bigg/\left(\frac{\Delta\rho}{\rho}\right).$$

If a compressional sound wave propagates through the crystal, a periodic temperature difference will therefore be set up between the two phonon groups. In a certain relaxation time θ which is discussed below, heat exchange takes place between them, leading to an increase of entropy and therefore to an absorption of the sound wave. Assuming that both groups have about the same specific heat, the absorption coefficient in (db/inch) can be calculated to be

$$\alpha=4.34\frac{v\int_0^\tau TdS}{I_0}=1.1\frac{cT\gamma_{\text{Av}}^2}{\rho v^3}\left(\frac{\omega^2\theta}{1+(\omega\theta)^2}\right),\qquad(9)$$

where T = temperature, $\omega/2\pi=1/\tau$ = sound frequency, I_0 = sound intensity, v = long. sound velocity, c = spec. heat/cc, and ρ = density of quartz. The relaxation time θ is the time necessary to exchange energy between both groups. It seems reasonable to assume that this exchange takes place by means of phonon-phonon umklapp processes. As we will see, this assumption leads to the right temperature dependence of the absorption. The U process is rather well understood in quartz from heat conductivity data. The heat conductivity

$$K=\tfrac{1}{3}cv^2\theta,$$

where c = spec. heat, and v = sound velocity, above 40°K was found to vary roughly as $1/T$.[12] According to Westrum[13] the specific heat increases linearly with temperature between 40°K and 150°K. Therefore θ changes with temperature as $1/T^2$.

At high enough temperatures the relaxation time θ is much shorter than the sound period, $\omega\theta\ll 1$, and the absorption coefficient (9) reduces to:

$$\alpha=1.1cT\gamma_{\text{Av}}^2\omega^2\theta/\rho v^3.$$

The effect of the temperature dependent factors cancels, so that the product $(cT\theta)$, and therefore the absorption, is practically temperature independent in agreement with our measurements above 60°K. In this same temperature region the absorption is expected to increase with the square of the frequency, which is compatible with our results, although further experiments are planned for a more detailed comparison.

From the measurements of the heat conductivity and specific heat it appears that for $\omega/2\pi=10^9$ (sec^{-1}) the product $\omega\theta$ would be about unity at a temperature of $T=40$°K. At this temperature the absorption coefficient can be evaluated, using Westrum's data, to be

$$\alpha=1.27\gamma_{\text{Av}}^2\text{ (db/inch)},$$

which agrees with the experimental value of 5 (db/inch), if one assumes for the average Grüneisen-constant a value of $\gamma_{\text{Av}}=2$, which does not appear unreasonable.

At still lower temperatures, where $\omega\theta>1$, formula (9) would predict the absorption to disappear rapidly with decreasing temperature and also to become independent of frequency. Both expectations are in qualitative agreement with our measurements below 30°K, although we are aware of the difficulty of applying formula (9) to this temperature region, where the phonon mean free path is larger than the sound wavelength.

We would like to make some remarks about our results with the neutron irradiated specimens: from Berman's study[14] of the effect of neutron irradiation on the heat conduction it is known that the phonon mean free path in quartz can be considerably decreased by irradiation, especially at lower temperatures. The temperature dependent part of the absorption after irradiation should therefore according to formula (9) be higher at low temperatures and lower at high temperatures than the absorption before irradiation. At very high temperatures ($T>180$°K) the effect of ir-

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[12] See for example: W. J. DeHaas and Th. Biermasz, Physica **5**, 620 (1938); **2**, 673 (1935).

[13] We thank Dr. Westrum for permission to use his data prior to publication.

[14] R. Berman, Proc. Roy. Soc. (London) **A208**, 90 (1951).

radiation on the absorption should become negligible, as it is in the case of heat conduction, because U processes are becoming much more frequent than collisions with damage centers. In all these points there seems to be good qualitative agreement with our experiments. (See Figs. 6, 7.) A more quantiative comparison between Berman's results and our data appears difficult, because the mean free path limiting the heat conduction in an irradiated crystal and the mean free path responsible for sound absorption are not necessarily the same.

We would like to emphasize that the relaxation process which we have sketched here has been applied only in the absorption of *longitudinal* hypersonic waves in quartz. Whether it also causes the absorption of transverse waves cannot be answered until data about the effect of shear strain on the elastic constants in quartz are available.

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Nonlocal Current-Field Relationship in Metals*

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It is found that Chambers' formula for the response of the conduction electrons of a metal to an internal transverse electric field must be supplemented by a diffusion current when it is used for longitudinal fields. A new derivation of Chambers' formula is given by means of the Boltzmann transport equation, and the additional diffusion term in the current is exhibited explicitly.

THE purpose of this note is to point out that the well-known and widely accepted formula due to Chambers[1] for the response of a metal to an internal transverse electrical field is actually not immediately applicable to a longitudinal field, but can be used provided that it is supplemented by an additional term. No error exists in the applications of this relationship by Chambers and Pippard[2] to the determination of the anomalous skin depth because of the restriction to transverse fields. But with the increasing interest in the propagation and attenuation of ultrasound in metals,[3] as well as in other phenomena in which longitudinal fields are generated (e.g., screening and slowing down of moving point charges), it seems desirable to improve Chambers' formula so that it is also correct for such fields. Such a diffusion current term is even mentioned by Chambers,[1] but it does not seem to have appeared in the published literature. Chambers' formula, including the additional term, will be exhibited explicitly below, but for the purpose of illustrating the basic idea in the present paper, it will be useful first to consider the special case of a static electric field. If, in addition, the electric field varies only slowly in space (with the scale taken as the mean free path of the conduction electrons in the metal), then Chambers' formula reduces simply to Ohm's law

$$\mathcal{J}_C(\mathbf{x})=\sigma_0\mathcal{E}(\mathbf{x}). \tag{1}$$

This is a completely local relationship between the electric field $\mathcal{E}$ and the current density $\mathcal{J}_C$ at the point $\mathbf{x}$. σ_0 is the dc conductivity of the metal. It is immediately clear, however, that this contribution to the current cannot represent a steady state solution for the response of the electrons to a longitudinal electric

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[1] R. G. Chambers, Proc. Phys. Soc. (London) A65, 458 (1952).

[2] A. B. Pippard, *Advances in Electronics and Electron Physics* (Academic Press, Inc., New York, 1954), Vol. VI. This article gives a brief derivation of Chambers' relationship from kinetic theory and except for the omission of the diffusion current, presents it in the same form as in the present paper.

[3] W. P. Mason and H. E. Bömmel, J. Acoust. Soc. Am. 28, 930 (1956). In the study of the ultrasonics of metals it is essential also to take into account not only the diffusion current, but also the additional current resulting from the collision drag effect. We make no attempt to treat this additional modification here. For detailed treatments of this effect see the work of Steinberg (reference 7), the recent paper of Holstein [T. Holstein, Phys. Rev. **113**, 479 (1959)], and the thesis of J. L. Warren, University of Maryland, 1959 (unpublished). In the last named reference the attenuation and dispersion of phonons is worked out using the dielectric constant approach. The results are identical to those of Steinberg (reference 7).