Microwave Ultrasonics

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SUMMARY. Ultrasonic waves injected into a crystal undergo interactions with the crystal lattice and the lattice vibrations, with conduction electrons and electron magnetic moments and with defects in the lattice. The generation of ultrasonic waves at microwave frequencies has allowed the study of such interactions to be extended to above 1 GHz and has provided data which could previously only be estimated from measurements averaged over the whole spectrum of the thermal vibrations of the lattice. At the same time, investigations of the ultrasonic propagation at these frequencies may lead to useful applications in devices.

1. Introduction

A crystal lattice consists of a regular array of atoms or ions each of which is continually vibrating about a mean position in the crystal. These vibrations may be described in terms of longitudinal or transverse waves of various frequencies propagating through the crystal lattice and resulting in the displacement of the ions. For those waves at frequencies of less than about $10^{11}$ Hz (i.e. angular frequency, $\omega$, less than $6 \times 10^{11}$ radians/sec), adjacent ions in the lattice are displaced in the same direction and the lattice is said to be vibrating in an ‘acoustical’ mode. However, at frequencies a little greater than this, ions of opposite charge situated in adjacent lattice sites may suffer displacements in opposite directions when the lattice is said to be supporting an ‘optical’ mode. These modes are illustrated in fig. 1 which also shows the

Fig. 1. (a) Modes of vibration in an ionic crystal. (b) Relation between $\omega$ and $k$ for the two modes. Although the velocity, taken as $\omega/k$, is seen to vary with the frequency, the waves at microwave frequencies will lie on the linear part of the acoustical branch and have a velocity $s$.

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form of the relations between the angular frequency $\omega$ and the propagation vector $k$ (i.e. the dispersion relations) for the two modes of vibration. Thermal excitation within the crystal causes the generation of lattice waves over the whole frequency spectrum including both optical and acoustical modes of vibration. The lattice waves at frequency $\nu$ are quantized in discrete amounts of vibrational energy each of magnitude $\hbar\nu$ where $\hbar$ is Planck's constant, and these quanta are known as 'phonons' by analogy with the 'photons' of electromagnetic energy.

Many of the non-equilibrium properties of solids, such as thermal and electrical conductivity, are known to depend on the presence and interactions of these thermal phonons. Therefore, a knowledge of their behaviour would enable the better understanding of such properties and of their dependence on the crystal lattice and its defects. However, any study of properties affected by the presence of thermal phonons generally involves some complex averaging over the whole spectrum of thermal phonon frequencies. It would obviously be far simpler to devise experiments for the understanding of the properties of the phonons if it were possible to cause their artificial generation in coherent beams having controlled frequency, polarization and direction. It would then be possible to study the interactions of these phonons with, for example, the natural thermal phonons of the lattice, conduction electrons and lattice defects. The amount of information which may be obtained from such experiments increases rapidly with the frequency range over which coherent phonons may be generated. At the present time, coherent beams of phonons have been generated and propagated through media at frequencies of up to about 100 GHz (Ilukor and Jacobsen-private communication). However, extensive investigations have been carried out only at frequencies of up to about 16 GHz and most of the experimental work has been performed at frequencies below 10 GHz where the ultrasonic generation is relatively simple and convenient. The microwave frequencies are low compared with the mean frequency of about $7 \times 10^{11}$ Hz of thermal phonons in a crystal at room temperature. However, this mean frequency, which is calculated as $kT/\hbar$, and also the numbers of thermal phonons, can be significantly reduced by cooling the crystal to liquid helium temperatures and, in general, the attenuation of any phonons injected into the crystal can be similarly reduced by such cooling.

The velocity of propagation of ultrasonic phonons in single crystals is typically in the region of $8 \times 10^5$ cm/s and so, for microwave frequencies, of around 10 GHz, the corresponding wavelengths of about $8 \times 10^{-5}$ cm (i.e. almost optical wavelengths) are always several orders of magnitude larger than the interatomic lattice spacing. For this reason it is possible, and, frequently convenient, to consider these lattice waves as if they were being propagated in a continuous elastic medium and to use in their description such macroscopic parameters as the bulk Young's modulus and the crystal density.

Recent solid-state investigations in the field of microwave ultrasonics, while necessarily involving the interactions which occur between coherent injected phonons and the existing thermal phonons of the lattice, have largely been concerned with the interactions between the coherent phonons and conduction electrons, magnetic moments in both para- and ferromagnetic materials and other lattice defects. The motivation for the work has been practical application as well as the purely scientific interest. This review will be mainly concerned with those the conven frequencies have been gradually has been g such as a c

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with those ultrasonic experiments which have been carried out in that part of the conventional microwave region where phonons have been generated with frequencies between about 1 GHz and 16 GHz. The techniques to be described have been developed during the last decade and the upper frequency limit has gradually been extended. The practical application of the information which has been gained from these studies has led to the successful operation of devices such as acoustic delay lines and delay line amplifiers.

2. Experimental techniques

The production of electromagnetic radiation at microwave frequencies has long been established. The generation of ultrasonic waves at these frequencies relies on the conversion of such microwave electromagnetic energy into acoustic energy by means of a transducer, a device in which the application of a varying electric or magnetic field causes a corresponding elastic strain. For example, one of the most commonly employed transducer materials is natural crystalline quartz which exhibits piezoelectricity such that, along certain crystallographic directions, the application of an electric field produces a strain in the quartz directly proportional to the applied field. However, while it is possible to generate low frequency phonons by placing the quartz between the capacitor plates of a conventional resonant circuit, at microwave frequencies it is necessary to employ transmission line or microwave cavity techniques.

![Diagram of microwave ultrasonic generator](image)

Fig. 2. Generation of microwave ultrasound by means of a non-resonant quartz transducer. For an experiment within the frequency range 10-16 GHz the cavity would have an internal volume of, typically, 1 cm³.

A typical experimental arrangement used for the generation of coherent longitudinal microwave phonons is shown in fig. 2. A single crystal rod of X-cut quartz is inserted into a re-entrant coaxial microwave cavity such that a high intensity electric field, acting between the end of the central post and
the cavity wall, induces a strain along the X-axis of the quartz. When electromagnetic energy is supplied to the cavity at its resonant frequency, an ultrasonic wave of the same frequency will be launched into the quartz rod. The efficiency with which the ultrasound is generated has been shown by Bommel and Dransfeld (1960) to be a function of cavity design and to be inversely proportional to the ultrasonic frequency. Typically, for a frequency of 10 GHz, a quartz rod of 3 mm diameter and a cavity having a Q-factor of 1000, the fraction of the electromagnetic energy which is converted into ultrasonic energy is about one part in ten thousand. For this reason, the electromagnetic energy is usually supplied to the cavity in the form of high energy pulses of about 1 μs duration and with a peak power, at 10 GHz, of 10-20 watts. As the efficiency decreases with increasing frequency the peak power requirement increases rapidly.

Once the ultrasonic wave, or pulse, has been launched into the quartz rod, the amplitude of the wave will decrease as a function of distance, the ideal attenuation having the form $A = A_0 \exp (-\alpha l)$ where $A_0$ and $A$ are the injected amplitude and the amplitude after a length $l$ respectively and $\alpha$ is the attenuation constant. Since a significant part of the attenuation is due to interaction with the thermal phonons there should be a marked dependence of $\alpha$ on temperature, and the experimentally determined temperature variation of the ultrasonic attenuation for longitudinal ultrasonic waves in quartz is shown in fig. 3 for a range of microwave frequencies.

![Fig. 3. Ultrasonic attenuation in quartz as a function of temperature and frequency. Because of the increase in attenuation it is not generally feasible to propagate ultrasound in quartz above 1 GHz at room temperature.](image)

Because of this temperature-dependent attenuation the majority of experiments with microwave ultrasonics have been carried out at or near liquid helium temperatures. Here the attenuation of the ultrasound is controlled, not by the interactions with thermal phonons, but by the scattering from defects and by diffraction and scattering effects which occur when the ultrasound is reflected to and fro between the end faces of the crystal.

**Echo patterns**

When an ultrasonic pulse has travelled the length of the quartz rod shown in fig. 2, it will be reflected from the end face and, on returning to the original end of the rod, it will re-excite an electromagnetic pulse in the cavity by means of the inverse piezoelectric effect. Due to the very low conversion efficiency of the quartz transducer this re-excited pulse or ‘echo’ will be considerably smaller than their initial frequency.
than the initial excitation pulse by at least a factor of $10^{-8}$ for an operating frequency of 10 GHz. Moreover, because so little of the ultrasonic energy is lost by the reconversion process, it is possible for the pulse to be reflected to and fro between the ends of the rod and to produce a whole series of echoes. For there to be an appreciable number of such echoes it is necessary that the attenuation shall be low, that the ends of the quartz rod are polished flat to about one-fifth of an acoustic wavelength and that these faces are parallel to within a few seconds of arc. If these small echo signals from the cavity are fed into a high-gain, low-noise amplifier capable of detecting about 1 pW

![Echo patterns formed by multiple reflections of a longitudinally polarized ultrasonic pulse within a quartz rod (16 GHz pulses at 4.2 °K).](image)
and then to an oscilloscope, the resulting echo train will ideally show an exponential decay due to ultrasonic attenuation in the quartz. Unfortunately from the point of view of attenuation measurements, this ideal form of echo envelope is rarely, if ever, obtained in practice due to the non-parallelism of the end faces of the quartz. Typical echo patterns are shown in fig. 4. Fig. 4 (a) shows an experimental echo pattern which is nearly exponential, although the first few echoes appear to have the same amplitude because they were each of sufficient magnitude to saturate the detection system, while fig. 4 (b) is much more typical of the echo patterns generally seen. (Care must be taken to generate only a single ultrasonic mode if patterns as simple as fig. 4 (b) are to be obtained. If, for instance, shear waves are generated as well as longitudinal waves, and this can occur in X-cut quartz if there is a component of cavity electric field acting in a direction parallel to the surface of the quartz, the pattern of detected echoes becomes much more complicated.)

Although a full explanation of the shape of the echo pattern envelope involves many complex parameters (see, for example, Taylor 1968), the basic theory is similar to that for the Fraunhofer diffraction of a plane wave at a circular aperture. This may be seen qualitatively from fig. 6. When the reflected ultrasonic wavefront returns to the generating face of the quartz, it will, in general, be at an angle to the surface. Since the electric field generated in the cavity depends on the net charge induced on the surface of the quartz, it is possible for the ultrasonic phase difference across the surface to produce a maximum or a minimum in the observed echo amplitude. Then, as the wave is reflected to and fro between the slightly non-parallel end faces of the rod, the angle of incidence of the wavefront increases and the echo pattern envelope exhibits a number of maxima and minima. Calculations based on this theory, and taking into account the shift of the ultrasonic wavefront across the end faces, yield echo patterns containing many features similar to those obtained experimentally, although there are too many other factors, such as reflections from the side walls and inhomogeneities in the quartz rods themselves, for the whole pattern to be explained on such a simple basis.

A variety of techniques, other than that of the non-resonant quartz transducer described above, can be employed for the generation of microwave ultrasound. For example, the more efficient resonant quartz transducers

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**Fig. 5.** Reflection of an ultrasonic wavefront within a quartz rod. Note the variation of charge across the end face due to the angle of the wavefront which gives compression over part of the detecting surface and rarefaction over the other.
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which the thickness of the quartz is one or two half-wavelengths of the ultrasound have been employed successfully at ultrasonic frequencies of up to 10 GHz (Stewart and Stewart 1962). However, at microwave frequencies, the transducer itself is extremely thin even when used in a high harmonic mode, and the technical difficulties involved in the preparation of such thin wafers, with the criteria for surface polish and parallelism, have prevented their regular use and the extension of their application to even higher frequencies.

For the latter it is simpler to develop the resonant transducer technique with piezoelectric materials which may be deposited in the form of thin films. For example, ultrasonic generation has been achieved in cadmium sulphide which, when vacuum deposited on a suitable substrate, is found to align itself in such a way that its principal axis, the C-axis, has a preferred orientation to the substrate surface. With this material, it is possible for pure longitudinal or pure shear waves to be generated piezoelectrically in the cadmium sulphide, and hence in the substrate. Similar generation has been observed in zinc oxide and it has been suggested that, besides rendering the resonant transducer technique at microwave frequencies more reliable, the use of these materials may enable it to be extended to frequencies well above 10 GHz (Paige and Maines 1968).

The generation of ultrasound has also been achieved by the application of the magnetostrictive effect both in bulk specimens and in thin films of ferromagnetic materials. Here, the ferromagnetic material is placed in the position of maximum r.f. magnetic field in a microwave cavity and, with a static magnetic field applied normal to the polished surface, the magnetization vector is caused to precess about this static field, the amplitude of precession being a maximum when the field is adjusted for resonance. Due to the magnetostrictive effect, the precessing magnetization will be accompanied by a rotating shear strain which results in the propagation of a circularly polarized shear wave along the magnetic field direction. This method of ultrasonic generation has been largely confined to thin films of nickel, the initial experimental work being performed by Bommel and Dransfeld (1969) at 1 GHz while the technique has been extended to 9 GHz by Pomerantz (1961) and Lewis et al. (1962). The limit to the application of the technique is merely the magnitude of the magnetic field required to cause resonance, although there are obvious difficulties in producing uniform thin films of the required half-wavelength thickness. The generation of microwave ultrasound in bulk ferromagnets has been achieved in non-metallic materials and this will be discussed further in Section 3.

Generation of ultrasound in 'inert' materials

In most materials there is no significant direct coupling between an electromagnetic field and the ultrasonic vibrations of the lattice. Therefore, if it is desired to generate coherent ultrasonic vibrations in these 'inert' materials, it is necessary for them to be joined to one of the active types of transducer described above. This may be carried out either by direct deposition of the transducer as a thin film or by bonding a bulk transducer on to the inert material by means of some adhesive.

For ultrasonic frequencies greater than about 1 GHz, the non-resonant quartz transducer has been more commonly employed where experimental conditions have allowed. Quartz is a stable, easily obtainable material for which
preparation techniques have been well established. With such a quartz transducer the bonded pair of crystals has to be able to withstand thermal cycling between room temperature and liquid helium temperatures besides propagating ultrasound. Various adhesives have been employed as bonding agents between the transducer and the specimen under investigation and it has been found experimentally that, for any acceptable degree of transmission, the bond thickness should not exceed one-half of an acoustic wavelength. Furthermore, the bond should be uniform to at least one-fifth of an ultrasonic wavelength over the contact area. Bonding materials successfully employed for the transmission of longitudinal waves have included a variety of greases, cements and metals. The principal metal used has been indium (with which some degree of success has also been achieved for the transmission of transverse waves), although this has been mainly used at the low frequency end of the microwave spectrum. 'Nonaq' D.C. 200 silicone oil and silicone high vacuum grease have been used successfully at frequencies up to about 10 GHz, while 'Araldite' and 'Glyptal' GE7031 low temperature varnish have been used successfully at frequencies up to 16 GHz. The maximum ultrasonic transmission efficiency obtained through a bond has been about 10 per cent even with the optimum conditions of a very thin, uniform bond and with highly polished crystal surfaces, flat to about one-tenth of an acoustic wavelength. The most successful materials reported to withstand the thermal cycling have been forms of Araldite. Further, although it has been possible to generate shear waves in 'inert' crystals by means of directly deposited thin film transducers such as nickel, the use of bonded quartz transducers for this purpose has only been marginally successful. Even then, the success has been limited to low microwave frequencies.

**Fig. 6.** Schematic echo pattern from a bonded pair of crystals. The pulse echoes marked Q1, Q2, Q3 have been reflected entirely within the quartz while, for example, Q2S2 has made two round trips in the specimen and two in the quartz.

It is possible to account substantially for the poor ultrasonic transmission efficiency of the bonds at low microwave frequencies in terms of the acoustic mismatch which exists between the pair of crystals and the bond material.
However, consideration of the scattering of ultrasound by surface imperfections shows that, in the presence of such imperfections, the fractional transmission through the surfaces decreases rapidly as the ultrasonic frequency increases. Thus extreme care is necessary in the preparation of crystals for ultrasonic experiments at the higher frequencies, particularly above 10 GHz. (A comprehensive account of methods employed for crystal surface preparation, including alignment, polishing, flatness, parallelism and assessment, is to be published shortly—Taylor et al.)

The form of the observed echo pattern which arises from the reflection of ultrasonic pulses within a bonded pair of crystals will depend on the relative lengths of the quartz transducer and the specimen, and on the relative ultrasonic velocities in the two crystals. The non-resonant quartz transducer is generally chosen to be longer than the bonded specimen, while, at the same time, the ultrasonic velocity in the quartz is generally found to be less than that in the specimen. For these particular conditions, the expected form of the observed echo pattern can be deduced from the schematic representation of the crystals shown in fig. 6 (a), the form of the pattern being illustrated in fig. 6 (b). There are no separate echoes observed from pulses which might be reflected within the bond, and the reduced amplitudes of those echoes returning from the specimen relative to those returning from the transducer occur as a direct result of the poor ultrasonic transmission efficiency of the bond.

**Ultrasonic velocity and elastic constants**

The most elementary physical quantity which may be determined from the ultrasonic experiments is the velocity of the ultrasound within the crystal. With the crystal orientated so that the ultrasound is propagating along a specific axis, the measurement of the length of the crystal and the determination of the transit time of the ultrasonic pulse will enable the ultrasonic velocity along the particular axis to be computed. From this velocity, and the velocity along other crystal axes, some of the elastic constants of the crystal may be calculated (see, for example, Kittel 1967; Pointon and Taylor 1968). If the case of a longitudinal wave propagating along the (100) axis of a cubic crystal is considered, then the ultrasonic velocity $v_{11}$ is related to the second-order elastic constant $C_{11}$ by the equation:

$$v_{11} = \sqrt{\frac{C_{11}}{\rho}}$$

where $\rho$ is the macroscopic crystal density. Similarly, the longitudinal ultrasonic velocity $v_{12}$ along the (110) axis of a cubic crystal is related to the three second-order elastic constants $C_{11}$, $C_{44}$ and $C_{12}$ by the equation:

$$v_{12} = \sqrt{\frac{1}{3} \left[ C_{11} + C_{12} + 2C_{44} \right] / \rho}.$$  

The velocity of a transverse wave along the (100) axis of a cubic crystal is given by $(C_{44}/\rho)^{1/4}$ and so it is possible to determine three elastic constants of a cubic crystal from three measurements of ultrasonic velocity. Many other relations exist between second-order elastic constants and the ultrasonic velocities of longitudinal and transverse waves, and a complete analysis of

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† Here $C_{11}$ is more generally known as the Young’s modulus of the crystal. The ‘second-order’ elastic constants arise from the simple harmonic (or Hooke’s Law) terms in the energy of distortion of the crystal.
all pure mode directions for crystal point groups belonging to the orthorhombic, tetragonal, cubic, rhombohedral and hexagonal systems has been given by Brugger (1965).

Measurements of the changes in ultrasonic velocity that occur when a crystal is subjected to a stress give information about the third-order elastic constants which express the deviation of the lattice potential energy from the simple harmonic form. Similarly, the variation of the ultrasonic velocity with temperature indicates the variation of these anharmonic effects as the crystal expands. However, a full discussion of such effects is well beyond the scope of this review.

3. Phonon interactions

There are two main parameters which are measured in experiments with microwave ultrasonics. These are the ultrasonic velocity and the amplitude of the observed pulse echo. The variations of these two parameters with the ultrasonic frequency, crystal temperature and an externally applied magnetic field have been investigated for a number of materials.

Lattice defects such as impurity atoms, vacancies and dislocations can produce local variations in the elastic properties of the medium which result in a scattering of incident ultrasonic waves. The interaction of ultrasonic phonons with isolated electronic magnetic momenta, with conduction electrons and with the thermal phonons of the lattice, can produce an attenuation and dispersion (i.e. velocity change) of the ultrasound. Interactions can also occur between ultrasonic phonons and the spin-waves (magnons) which exist in a ferromagnetic material.

Although the effect of dislocations on ultrasonic propagation can be substantial at megahertz frequencies and the scattering of ultrasound by the effective mass-changes of an impurity or a vacancy is important for ultrasonic waves at very short wavelengths, both these phenomena have a negligible effect on the propagation of microwave ultrasound. For this reason the microwave ultrasonic attenuation in a non-magnetic insulator is usually normalized to be zero at 4.2 °K where the attenuation due to thermal phonons is also negligible.

**Phonon-phonon interactions**

In a non-magnetic insulator such as quartz or magnesium oxide it has been established that the dominant cause of the attenuation of a microwave ultrasonic pulse is phonon-phonon interactions. That is, the ultrasonic phonons interact with the thermal phonons of the lattice so that energy is lost from the coherent phonon pulse. The dominant interaction is the three-phonon process, the two possible forms of which are illustrated in fig. 7. The conditions imposed in these processes are that both the energy and the momentum of the phonons be conserved as illustrated in the diagram.

The results of the interactions are most conveniently discussed for two well-defined sets of conditions. For the case of microwave phonons having a low angular frequency ω in a crystal lattice at a temperature well above 4.2 °K the mean lifetime τ between the creation and annihilation of a thermal phonon can be very much less than the period of the ultrasonic wave, so that ωτ ≪ 1. The uncertainty in the angular frequency of a thermal phonon which is given, on average, by Δω~ 1/τ is thus very much greater than the
ultrasonic angular frequency. Any scattering processes between the ultrasonic and thermal phonons which necessarily involve the conservation of energy and momentum may therefore occur for a wide spectrum of thermal phonons. Thus, the ultrasonic phonons may be considered to interact in a macroscopic manner with the whole vibrating crystal lattice. The process by which the equilibrium distribution of thermal phonons is disturbed by the introduction of the ultrasonic phonons has been considered by Akheizer (1939), Woodruft and Ehrenreich (1961) and others. Their thermodynamic treatment of the rate of energy loss,

![Fig. 7. Three-phonon scattering processes between the ultrasonic phonon with propagation vector $k_{us}$ and two thermal phonons $k_i'$ and $k_i''$.](image)

expressed in terms of a phenomenological relaxation time, shows the attenuation over the range of application to be substantially independent of temperature in agreement with the experimental results of Bommler and Dransfeld (1960). (The 'thermoelastic' loss mechanism proposed by Zener (1937), in which the relatively slowly changing regions of ultrasonic compression and rarefaction lead to an irreversible heat flow, is important in metals -q. Lucke (1956)- but accounts for only some 4 per cent of the ultrasonic attenuation in insulators.)

The second condition occurs if the crystal lattice is cooled to a few degrees Kelvin and an ultrasonic wave having a frequency greater than about 10 GHz is introduced into the lattice. In this case, with the increase in the thermal phonon lifetime, the condition $\omega \tau > 1$ is satisfied. This means that the spectrum of thermal phonons which may take part in a given three-phonon process is strictly limited. A value for the reduced attenuation in this case has been derived by Landau and Rummer (1937) who predicted that the ultrasonic attenuation coefficient would be proportional to $\omega T^4$. This is in quite good agreement with some of the experimental results for transversely polarized ultrasound in quartz (e.g. Maria 1963). However, more recent measurements by Lewis and Patterson (1967) have shown that, although the attenuation of the slow transverse waves in quartz is determined by the Landau-Rummer mechanism, the attenuation of fast transverse phonons is determined by three-phonon processes which are only allowed as a result of the finite lifetime of the thermal phonons.

The basic mechanism responsible for the attenuation of longitudinal ultrasonic phonons remains in some doubt. In all the materials studied so far the measured attenuation for longitudinal phonons is of the same order of magnitude as that of transverse phonons, and varies with frequency as $\omega^n$, where $0 < n < 1$, 

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and varies with temperature as $T^m$ where $4 < m < 9$ (Lewis and Patterson 1967 and references cited therein). This result is surprising because of restrictions placed on the three-phonon processes in which longitudinal phonons may take part if they are to conserve energy and momentum. However, calculations by Maris (1964) take account of the finite lifetime of the thermal phonons and consider a number of nearly collinear three-phonon interactions which would be forbidden in the absence of the energy uncertainty. The predictions are in quite good agreement with both the temperature dependence and the magnitude of the experimentally determined attenuation.

The interaction between two sets of coherent ultrasonic phonons has been demonstrated by Shiren (1964) who has shown that it is possible to produce an effective parametric amplification through the non-linearity (or anharmonicity) of a crystal lattice. When pulses of coherent phonons at frequencies of 16.45 GHz and 8.5 GHz were introduced simultaneously into a magnesium oxide crystal lattice, it was found that the higher frequency pulses acted as a 'pump' and caused amplification of the lower frequency ultrasound.

**Spin-phonon interactions**

The interactions that can occur when an ultrasonic wave passes through a crystal containing a very low concentration of paramagnetic ions are of particular interest. Microwave phonons, like microwave photons, can excite transitions between the energy levels of a paramagnetic ion when these are split by the application of an external, static magnetic field of the appropriate magnitude. The interaction occurs because, when ultrasonic phonons are incident upon a paramagnetic ion embedded in a host crystal, the crystalline electric field at the ion is modulated at the phonon frequency. In general this modulation will cause a variation in the relative energies of the outer electrons of the ion and, provided the ultrasonic phonons have the correct energy, they can be absorbed by the ion in causing the spin of the electron to 'flip' relative to the magnetic field. This transfer of energy from the phonons to the electron spins, the phonon-induced electron spin resonance (E.S.R.), is effectively the reverse of the direct spin-lattice relaxation process which broadens the resonance linewidth in conventional photon-induced E.S.R. It follows therefore that phonon-induced E.S.R. is particularly easy to observe in those cases where the spin-lattice broadening of the linewidth prevents the observation of the photon-induced resonance. The energy level structure of the particular paramagnetic ion can thus be investigated in a similar manner to that employed for conventional E.S.R. while, at the same time, the strength of the coupling between the lattice and the ion may be determined.

The experimental observations of electron spin resonance are interpreted, where possible, by expressing the lowest energy levels of the paramagnetic ion as an effective spin Hamiltonian. In the presence of a principal symmetry axis, which may be taken to be the z-axis, this Hamiltonian is usually written in the form

$$\hat{H}_S = \beta g_1 H_x S_x + \beta g_2 (H_x S_x + H_y S_y) + D (S_z^2 - \frac{1}{2} S(S+1))$$

where $H_x, H_y$ and $H_z$ are the components of the applied magnetic field, $\beta$ is the Bohr magneton, $g_1$ and $g_2$ are the appropriate components of the g-factor, $S_x, S_y$ and $S_z$ are the components of the spin operator $\mathbf{S}$, and $D$ is a zero-field splitting of the magnetic hyperfine strains.
spreading term determined by the spin-orbit coupling and the presence of lattice strains. Any deviation from axial symmetry can be taken into account by adding a term of the form $E(\tilde{S}_z^2 - \tilde{S}_y^2)$ and replacing $g_L$ by the separate coefficients $g_x$ and $g_y$. Once the form of the Hamiltonian has been established the E.S.R. measurements are used primarily for the determination of the quantities $g_{||}$, $g_{\perp}$, $D$ and $E$. The interaction between the ultrasonic wave and the paramagnetic ion arises from a modulation of the term $D$ due to lattice strains induced by the ultrasonic wave. It is therefore possible to define a spin-phonon coupling constant $\gamma_{||}$ so that, if a lattice strain $\epsilon$ is introduced along a (100) axis of a cubic crystal, the perturbation of $D$ caused by the strain may be expressed in the form

$$\delta(D) = \gamma_{||} \epsilon.$$

The corresponding ultrasonic attenuation resulting from transitions induced between the various spin-states has been calculated by Dobrov (1964) and others. For a magnetic field applied at an angle $\theta$ to a longitudinal wave travelling along the (100) axis of a cubic crystal, the attenuation due to the transition in which the spin angular momentum changes by one unit of $\hbar$ (i.e. the $\Delta M = 1$ transition) is given by

$$\alpha(\omega) = F(\omega, T)g_{||}^2 \sin^2 2\theta,$$

where $F(\omega, T)$ is a function of the frequency and the temperature as well as of the effective spin, the number of paramagnetic ions and the magnetic field. The value of $F(\omega, T)$ can be determined by measurement of the absorption line shape as a function of applied field and temperature, and hence the measured value of the ultrasonic attenuation can lead to the determination of $\gamma_{||}$.

The experimental observations of phonon-induced E.S.R. began as soon as techniques for the generation of the microwave ultrasound were established. Some of the first experiments included those of Jacobsen et al. (1969) who observed ultrasonic paramagnetic resonance absorption by divalent manganese ions and F-centres in quartz, and of Mattuck and Strandberg (1960) and Jacobsen (1960) who demonstrated the saturation of the chromic (Cr$^{3+}$) ion resonance in magnesium oxide. Measurements of spin-phonon coupling constants for various iron group ions, present as impurities in good cubic crystals such as magnesium oxide, have been made by the ultrasonic E.S.R. technique (e.g. Rampton 1965) and the results obtained are in fair agreement with those of Watkins and Feller (1962) obtained by other methods. Although the ferric (Fe$^{3+}$) ion is only weakly coupled to the crystal lattice because of its electronic structure, there is strong spin-phonon coupling of the ferrous (Fe$^{2+}$) ion and a typical ultrasonic E.S.R. curve showing the absorptions which occur due to ferrous ions as impurities in magnesium oxide is shown in fig. 8. The corresponding diagram of the lowest energy levels as a function of magnetic field is also shown. Observations of these resonances at two different ultrasonic frequencies by the authors (to be published) have enabled new estimates to be made of the constants $g(=g_x, =g_y)$ and $E$. (The ferrous ion resonance shown as $\Delta M = 2$, i.e. corresponding to a change in angular momentum of two units of $\hbar$, is observable only with phonon-induced E.S.R.)
unsuccessful, but the ultrasonic E.S.R. spectrum of this ion, which is extremely complex has been observed at 9.5 GHz by Marshall and Rampton (1968) and at 9.5 and 16 GHz by the authors (to be published). A typical set of ultrasonic E.S.R. spectra for the $^{57}$Fe$^{2+}$ ions are shown in fig. 9. As with photon-induced E.S.R., the ultrasonic resonance absorption is accompanied by a change in

Fig. 8. The ferrous ion resonances in magnesium oxide at 16 GHz. The absorptions arising from the various transitions appear in (a) as a reduction of the height of the echo of a pulse which has passed through the magnesium oxide. The actual transitions are shown on the energy level diagram (a). It will be seen that the relative positions of the energy levels, the asymmetry of which is due to lattice strain, causes the two $\Delta M = 1$ transitions to occur at different magnetic fields. Consequently this resonance line is much broader than the $\Delta M = 2$-line. (The lengths of the arrows showing the transitions each represent the energy $\hbar \nu$ for a 16 GHz phonon.)

Fig. 9. Ultrasonic E.S.R. spectrum of the $^{57}$Fe$^{2+}$ in magnesium oxide at 16 GHz. The complexity of the spectrum arises largely from the strong coupling of the spins to the lattice. The ferrous ion resonance shown arises from the iron impurities always present in magnesium oxide.
ultrasonic velocity. These velocity changes have been investigated by Shiren (1962) for the divalent nickel ion and ferrous ion resonances in magnesium oxide and have been used (see, for example, Taylor 1968) in order to estimate the spin-phonon coupling constants. Also Guermeur et al. (1968) have observed a rotation of the plane of polarization of a transverse ultrasonic wave passing through a crystal of magnesium oxide doped with nickel ions. This is in effect an ultrasonic Faraday rotation.

An interesting experiment involving E.S.R. induced by thermal phonons has been carried out by Challis and Williams (1966) who showed that there were effective resonances in the thermal conductivity when a large magnetic field was applied to chromium-doped magnesium oxide at low temperatures.

**Phonon interactions in semiconductors**

The injection of ultrasonic phonons into semiconductors at room temperature results in the transfer of energy and momentum from the phonons to the conduction electrons (or holes). However, there is not the rapid ultrasonic attenuation which exists in a metal and it is possible to observe an electron drift current in the direction of the phonon beam as the momentum transfer occurs. Acousto-electric currents were observed in germanium by Weinreich et al. (1959) and in cadmium sulphide by Wang (1962). The inverse effect, in which drifting electrons give up energy to a phonon beam travelling in the same direction, was predicted by Weinerich (1956) and was observed in cadmium sulphide by Hutson et al. (1961). More recently, the effect has been observed in zinc oxide by workers at the Royal Radar Establishment, Malvern, Worcs.

The acousto-electric effects are naturally most pronounced in semiconductors which are also piezoelectric, such as cadmium sulphide and zinc oxide, because of the stronger coupling which exists in these materials between the phonons and the electric field of the lattice. Conveniently, when it is required to inject phonons into these materials, they may be employed as their own transducers.

When the wavelength of the injected phonons is greater than the mean free path of the conduction electrons, i.e. \( \lambda > l \), these electrons are able to bunch in the regions of low electrostatic potential energy produced by the phonons. There will thus be a tendency for the electrons to be carried along with the phonons. However if, by the application of an electric field, the electrons are given a drift velocity \( v \) greater than the ultrasonic velocity \( s \), there will be a tendency for the electrons to drag the phonons and to produce amplification of the ultrasound. This amplification will have a maximum for some value of the velocity difference \( (v - s) \)--see, for example, Paige (1966). For \( v < s \) there will be an attenuation of the ultrasound which is symmetrical with the amplification about the point \( v = s \). If the phonon wavelength is very much less than the electron mean free path (i.e. \( \lambda < l \)) the amplification or attenuation depends linearly on the quantity \( (v - s) \) and the variation of the amplification for the two cases is shown in fig. 10. In the microwave frequency region the predicted ultrasonic gains in cadmium sulphide are as high as 100 dB per cm of the ultrasonic path for frequencies of 1-2 GHz. With such gains, ultrasonic delay lines become a useful proposition, although it may be necessary for large powers, rising to several kW per cm², to be dissipated in the material. It is predicted that, with zinc oxide, the range of useful frequency application may be extended to 14 GHz although, at present, satisfactory operation has been reported only to
5 GHz (Maines and Paige 1968). This compares with a maximum frequency of 3 GHz for cadmium sulphide.

Because the amplification maximum in fig. 10 will occur at different values of \( v \) for different phonon frequencies, a given applied electric field will lead to the selective amplification of a limited range of thermal phonon frequencies. Consequently, if the field is chosen to give amplification in the microwave region, the piezoelectric semiconductor will act as a generator of the appropriate ultrasonic frequency. At the same time, the drift velocity will not vary linearly with electric field and the material will show non-ohmic behaviour (again, see Paige 1966). Finally we note that, since the density of conduction electrons in the semiconductor may be varied by illuminating the material with light of the appropriate wavelength, the amplification of the ultrasound may be conveniently modulated by varying the light intensity.

**Phonons in metals**

Very little work has been published regarding the propagation of microwave ultrasound in metals. This is due, presumably, to the large attenuation which exists in even the purest metals and, also, to the wider interest which has been shown in the ultrasonic properties of semiconductors. While, at room temperature, the ultrasonic attenuation caused by free electrons in a metal is small (Stewart and Stewart 1962), the attenuation due to phonon-phonon interactions is here generally prohibitive. At low temperatures, where the phonon-phonon effects are reduced, Pippard (1960) has shown that due to electron-phonon interactions, there is a large attenuation which may be many hundreds of dB per cm. Furthermore this electronic attenuation is approximately proportional to the ultrasonic frequency in the microwave region and it has therefore been necessary for attempts to investigate the electron-phonon interactions in metals at these frequencies to be made, in general, with very thin specimens. For example, Tepley (1966) has made limited observations on single crystals of nickel crystal.
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Magnon-phonon effects

In a ferromagnetic insulator such as yttrium iron garnet (YIG) there exists a magnetostrictive coupling between the lattice vibrations and the spin waves (magnons) which are formed by variations in the alignment of the electron spins. This means that transverse vibrational modes propagating along the direction of an applied magnetic field will not be pure phonons but will rather consist of hybrid 'magnetoelastic' waves. In such waves the variations of lattice position and magnetization are combined (see, for example, Le Craw and Comstock 1965). In the absence of any magnetoelastic coupling, the dispersion relations for the two types of wave are as shown in fig. 11 (a) where

$$\omega_{\text{phonon}} = \omega_k$$

and

$$\omega_{\text{spin wave}} = Dk^4 + \gamma H_1$$

with $D$ being an exchange constant, $\gamma$ the gyromagnetic ratio and $H_1$ the magnetic field acting on the electron. However, when the magnetoelastic coupling is present, the modes in the 'cross-over' region become complex magnetoelastic modes and the dispersion relation takes the

Fig. 11. Dispersion relations for magnetic and elastic waves. In (a) the magnetostrictive coupling is assumed negligible and there is no interaction between the transverse acoustic waves and the spin waves. The magnetostatic modes indicated on the diagrams are simply long wavelength spin waves, whose parameters are mainly determined by the dimensions of the specimen. (There is, in any case, very little interaction between spin waves and longitudinal acoustic waves propagated along the magnetic field direction.)
form illustrated in fig. 11 (b). For a crystal having the magnetoelastic dispersion relation shown in fig. 11, a microwave magnetic field of frequency $\omega_{\text{rf}} > \gamma H_s$ induces a spin wave in the crystal on the branch (1). Thus, when the magnetic field $H_i$ is reduced, it is possible to reach a point where there are no allowed spin waves of the frequency $\omega_{\text{rf}}$, and the disturbance originally induced as a spin wave will pass through the magnetoelastic modes and may be converted into a pure acoustic wave.

Experimentally, the conversion of spin waves to acoustic waves via the magnetoelastic interaction has been achieved in a cylindrical rod specimen of yttrium iron garnet. When an axial magnetic field $H_a$ is applied to the rod, the effective magnetic poles generated at the end faces give rise to a non-uniform magnetic field as shown in fig. 12. The value of the magnetic field at

![Fig. 12. Magnetoelastic waves in a magnetic material.](image)

the ends of the rod is $H_a - 2\pi M$, where $M$ is the saturation magnetization and the field $H_i$ rises almost to the value $H_a$ near the centre of the rod. At a given microwave frequency the spin waves will be generated at the positions $S$, where ferromagnetic resonance occurs. Since there are no allowed spin waves at higher fields for this frequency, the waves will travel to regions of lower magnetic field and, on passing through the cross-over region at the field $H_c$, will convert ultimately to acoustic waves. These acoustic waves will then be reflected back from the polished crystal end face $E$ and be redetected as spin waves at $S$.

With a crystal such as YIG the conversion of electromagnetic energy to spin wave energy is much more efficient than is the direct conversion to acoustic energy in a quartz transducer. However, the propagation losses are higher because the spin waves are attenuated more strongly than are ultrasonic waves.
A limit to the magnitude of the detected echo is set by spin wave instabilities, the maximum usable drive power being about 1 mW. On the other hand, the non-linear characteristics of the material have been used to produce parametric amplification at 1 GHz at room temperature and at 9 GHz at 4.2 °K, this amplification more than compensating for the attenuation of the waves. When the applied magnetic field is varied, the position of $S$ will vary along the rod and, since the major part of the delay between the injection of the pulse and its return is in the time spent in the acoustic mode, the delay time of the pulse can be altered by a few microseconds.

4. Conclusion

The development of techniques for ultrasonic generation at microwave frequencies has extended the range of investigations of paramagnetic phenomena not amenable to conventional E.S.R. studies. The utilization of the magnetoelastic and acoustoelectric effects may lead to the production of useful high frequency delay lines, delay line amplifiers and frequency memories. Further investigations of phonon interactions and the clarification of the crystal processes which involve their interaction should provide a valuable field of research for some time to come.

**General References**


**References**


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