arithmic divergence in the long-wavelength limit,[13] this smearing is complete and no vestige of long-range order remains.[14]

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$$\omega_k/k = a\omega_p[(-\tfrac{1}{2}\epsilon)\ln(ka)]^{1/2}$$

and the divergence is aggravated. Allowing for plasma oscillations in three dimensions increases the dependence of M upon Δ/ϵ_F to the first power, which, however, is still negligibly small. The effect of retardation on the one-dimensional plasma oscillations, which brings in the vector potential and could alter the phase considerations, is currently under investigation.

HARMONIC GENERATION OF MICROWAVE PHONONS IN QUARTZ

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Bömmel and Dransfeld[1] and Jacobsen[2] have demonstrated that microwave phonons can be generated by the piezoelectric effect at the end surface of quartz placed in the electric field of a reentrant cavity. The present paper shows that second and third harmonics of the fundamental microwave frequency are also generated. The experiment was originally undertaken to measure the higher order elastic constants which are important in the "collinear processes"[3,4] by which a longitudinal acoustic wave interacts with thermal phonons. The nonlinearity due to this interaction, however, was found to be much smaller than that present in the generation of the acoustic waves themselves. Second-harmonic generation in nonpiezoelectric Z-cut quartz has also been observed. It was much smaller than in piezoelectric quartz and of the magnitude to have been caused by the electric stress. This result is contrary to some of Shiren's[5] qualitative measurements of harmonic generation in piezoelectric quartz, which were attributed to the electric stress, and shows that acoustic wave generation is not limited to the relatively rare piezoelectric materials.

Microwave phonons are generated at the end surface of a piezoelectric quartz rod placed in the high electric field region of a re-entrant 4.5-Gc/sec cavity shown in Fig. 1. This cavity is excited by a 0.5-μsec pulse of electromagnetic energy which has passed through a low-pass filter to eliminate any harmonics above 4.5 Gc/sec which might have been present in the pulsed microwave source. The second-harmonic content

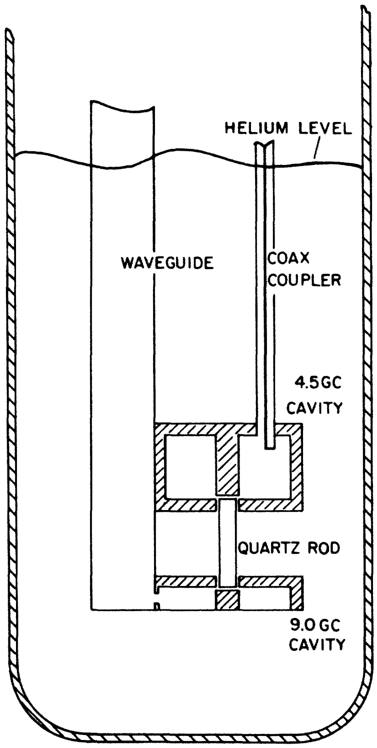

FIG. 1. Cross-sectional view of experiment.

of the acoustic wave packet is detected in the 9-Gc/sec cavity. The series of echoes generated and detected in the 4.5-Gc/sec cavity is shown in Fig. 2 together with the second harmonic detected in the 9-Gc/sec cavity. In the second part of the experiment, microwave phonons are generated by a pulse of electromagnetic radiation in the 9-Gc/sec cavity, and the resulting echoes, also shown in Fig. 2, are detected in the same cavity. The electromechanical conversion efficiency for both cavities was of the order of 10^{-4}.

The fact that the harmonically generated phonons were generated at the end of the rod inserted in the 4.5-Gc/sec cavity was established by noting that they decrease at the same rate as phonons generated at the end surface of the rod in the 9-Gc/sec cavity. If the nonlinearity had occurred as the wave propagated along the rod due to the nonlinearity of the elastic medium, then the harmonic content of the wave would increase, come to a maximum at a distance of the order of the phonon mean free path, and decay exponentially.[6] The mean free path is about 10 cm and the length of the rod 1 cm, so, if the nonlinearity had occurred in the volume, it would

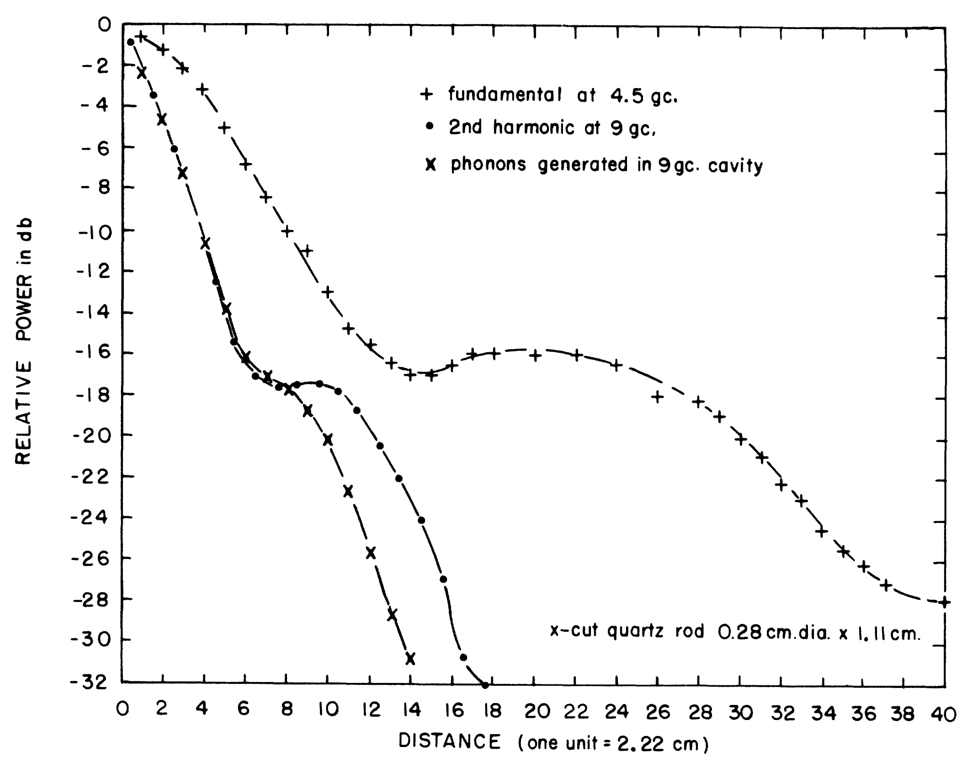

FIG. 2. The measured microwave power of each echo as a function of distance traveled. The beat structure which scales to the acoustic wavelength indicates that the ends are parallel to 1.5 seconds of arc.

have been observed.

The third harmonic was generated by replacing the 4.5-Gc/sec cavity, which has a gap spacing of 0.79 mm, indicated in Fig. 1, by a 3-Gc/sec re-entrant cavity which had a gap of 1.65 mm, and other dimensions chosen to resonate at the desired frequency. There were about five echoes visible above the noise. A measurement of the amplitude of each echo produced a curve similar to that of Fig. 2, and it was again concluded that the nonlinearity producing the third harmonic occurred at the surface of the quartz rod.

The second-harmonic power P_{2f} was found proportional to the square of the fundamental power, and the third, P_{3f}, to the cube of the fundamental. This establishes the nonlinearity of the generating process. The measured values of P_{2f}/P_f^2 and P_{3f}/P_f^3 are tabulated in Table I. The spread reflects the reproducibility of the data. No temperature variation of P_{2f}/P_f^2 was observed in the range from 1.4 to 4.2°K. The rods were all of about 0.3-cm diameter by 1 cm long and the ends were flat and parallel to less than five seconds of arc. The observed piezoelectric constant was found equal to within 50% of that measured by Mason[7] by using themeasured values of P_f and E_γ.[8] E_γ was measured by a perturbation technique.[9]

Second-harmonic generation of longitudinal waves in nonpiezoelectric quartz has also been observed. The fact that no echoes were received in the 4.5-Gc/sec cavity demonstrates that the generation mechanism has no inverse. The harmonic echoes were detected at 9 Gc/sec by a thin-film piezoelectric transducer of cadmium sulfide on the end of the rod in the 9-Gc/sec cavity. Preliminary qualitative results show that the second-harmonic power is about five orders of magnitude smaller than in X-cut piezoelectric quartz. This result for Z-cut quartz is in rough agreement with the calculated discontinuity in the Maxwell stress tensor. The second-harmonic generation in piezoelectric quartz is too large to be generated by this electric stress.[5]

The physical process causing the harmonic generation in piezoelectric quartz could be accounted for in terms of the higher order piezoelectric constants defined by Mason.[7] The process is not the dielectric heating mechanism studied by White,[10] as this mechanism would be the same order of magnitude for both Z-cut and X-cut quartz. The only experimental work known to the author on the nonlinear behavior of the piezoelectric constant is that of Ny,[11] who fitted his measured saturation of the constant to an exponential. The empirical relation involving the exponential had an odd symmetry with respect to the change of sign of the electric field. Only the third harmonic observed here could be related to this. In addition to the exponential, Ny also observed a much smaller term involving the square of the electric field, which he attributed to electrostrictive effects. This term is much too small to account for the observed second-harmonic generation in piezoelectric quartz.

If, in spite of the above, one assumes that the piezoelectric constant is given by $d(E)=d_0e^{-kE}$, it can be shown on expanding the exponential that

$$\frac{(P_{3f}/P_f^{\,3})^{1/2}}{P_{2f}/P_f^{\,2}}=\frac{2}{3}.$$

The observed value in Table I is in rough agreement with this for transverse waves in AC-cut quartz but not in X-cut quartz. Work is continuing to measure the harmonic generation of transverse waves in BC-cut quartz.

The nonlinearity in the generating process places an upper limit for the use of this experimental configuration in the measurement of the higher order elastic constants. The ratio of the second-order elastic constant to the elastic constant[4] is less than 0.1 for longitudinal phonons propagating along the X axis.

Table I. Experimental data.

Polarization	Second harmonic P_{2f}/P_f^2 (W^{-1})	Third harmonic P_{3f}/P_f^3 (W^{-2})	$\frac{(P_{3f}/P_f^3)^{1/2}}{P_{2f}/P_f^2}$ Expt.	Theory
Longitudinal (X-cut)	0.84-2.7	$0.82\text{-}1.9\times10^{-5}$	$1.6\text{-}3.3\times10^{-3}$	0.667
Transverse (AC-cut)	$1.9\text{-}3.2\times10^{-2}$	2.4×10^{-4}	0.48-0.79	0.667

The maximum value of 0.1 W for the acoustic power P_f was set by electric breakdown in the cavity. By increasing the breakdown field, one could make the harmonic of the same magnitude as the fundamental for $P_f = 0.5$ W. Harmonic generation can be useful in extending present techniques to the millimeter range, where high-power microwave sources are presently unavailable.

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OBSERVATION AND POSSIBLE MECHANISMS OF MAGNETOELECTRIC EFFECTS IN A FERROMAGNET

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In this Letter we report the first observation of magnetoelectric (ME) effects in a material which is ferromagnetic. Previous ME experiments in magnetically ordered substances were confined to chromic oxide[1] (Cr_2O_3) and titanium oxide[2] (Ti_2O_3), both of which are antiferromagnetic. In addition, we suggest an atomic model which qualitatively explains our experimental result that the direction of the induced magnetic (electric) polarization is perpendicular to the applied electric (magnetic) field. This is in contrast to earlier theories[3-6] whose applicability is restricted to situations in which the induced polarizations and the applied fields are coaxial.

Our experiments were performed on single crystals[7] of gallium iron oxide having the composition $Ga_{2-x}Fe_xO_3$, where x is around unity. This material is piezoelectric as well as ferromagnetic. Its magnetic structure and even its crystal structure are unknown, but it is known[8] that the crystals are orthorhombic and that the most probable crystallographic space group is $C_{2v}{}^9$. The axes of the unit cell are assigned according to $c < a < b$. If the crystallographic point group is indeed $2mm$, the possible magnetic point groups are $2mm$, $2m'm'$, $2'm'm$, and their subgroups. On the basis of the experimentally established magnetic properties[9] of this material, we eliminate the group $2mm$ because it does not allow ferromagnetism, and the group $2m'm'$ because it allows a spontaneous magnetic moment along the two fold axis (y axis) only. (We take the rectangular coordinates x, y, z to be along a, b, c, respectively.) Since in our experiments the crystal is exposed to a static biasing field H_0 which is applied along the $+z$ or $-z$ direction, and since there is no reason for lowering the symmetry, we suppose the correct magnetic-point group to be $2'm'm$. If we now expand some appropriate thermodynamic potential of the biased crystal in powers of the applied electric-field components E_i and the applied incremental magnetic field components H_j (where i and j denote x, y, z, and we assume $|H_j| \ll |H_0|$), then we easily see that the symmetry permits terms of the type E_yH_z and E_zH_y, both of which lead to linear ME effects. For the experimental configuration used in this work, only the E_yH_z term is applicable, so that the induced magnetization δM_z and the induced electric polarization P_y are given by

$$\delta M_z = \alpha E_y, \quad (1a)$$

$$P_y = \alpha H_z, \quad (1b)$$