II. MICROWAVE SPECTROSCOPY*

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A. WORK COMPLETED

1. MAGNETIC FIELD DEPENCENCE OF THE TEMPERATURE VARIATION OF THE HALL COEFFICIENT IN INDIUM ANTIMONIDE

This work has been completed by Stephen N. Bromberg and submitted as a thesis to the Department of Physics, M. I. T., May 1965, in partial fulfillment of the requirements for the degree of Bachelor of Science. An abstract of the thesis follows.

The Hall voltage developed in an InSb thin-film probe was measured as a function of temperature and magnetic field, in order to measure the variations of the temperature dependence of the Hall coefficient with magnetic field. For this purpose, the Hall probe was placed in a small metal can whose temperature was regulated by a thermistor and associated circuitry. Results seem to indicate a decline in temperature dependence with increasing magnetic field, at least for the range 100-4000 gauss, after which the dependence is constant up to 9000 gauss.

M. W. P. Strandberg

2. THIN-FILM BOLOMETER FOR DETECTION OF PHONONS IN QUARTZ AND SAPPHIRE

This work has been completed by Martin C. Graham and submitted as a thesis to the Department of Physics, M. I. T., May 1965, in partial fulfillment of the requirements for the degree of Bachelor of Science. An abstract of the thesis follows.

A superconducting 95% Sn-5% Cu thin-film bolometer was developed which was used to detect heat pulses in quartz and sapphire. It was evaporated to the end of both a quartz and sapphire rod, and was found to be sensitive in a temperature region around 3.5°K. A technique for mounting leads to this bolometer was successfully tested. Several helium-temperature experiments were conducted with a 3/4 inch quartz rod, and preliminary data seem to indicate that phonon dispersion was observed. Dispersion in sapphire was not observed when using this bolometer.

M. W. P. Strandberg

3. EPR STUDIES OF IRRADIATED POLYSTYRENE

This work has been completed by Robert M. Preer, Jr. and submitted as a thesis to the Department of Physics, M. I. T. , May 1965, in partial fulfillment of the requirements for the degree of Bachelor of Science. An abstract of the thesis follows.

Polystyrene samples were irradiated by 3-Mev electrons at the Van de Graaff generator. The total radiation dose was 3 Mrad. The electron paramagnetic resonance spectra of the samples were then obtained after varying amounts of time had elapsed from the time of irradiation. The samples at all times after irradiation were at room temperature and in contact with the air. In the absorption mode and at 6 kc/sec magnetic field modulation, the spectra seemed to agree with those published. With the modulation frequency decreased, the spectra appeared to consist of a single line in each case. The dispersion signal verified this result. The power level used in all experiments was low, being a few tenths of a milliwatt. Finally, an attempt was made to explain the discrepancies with the results on published spectra.

M. W. P. Strandberg

4. A CALCULATION OF THE AMPLITUDE ENVELOPE OBSERVED IN MICROWAVE PHONON GENERATION

This work has been completed by Michael K. Maul and submitted as a thesis to the Department of Electrical Engineering, M. I. T. , May 1965, in partial fulfillment of the requirements for the degree of Master of Science. An abstract of the thesis follows.

Several models have been suggested to explain the deviation in the amplitude envelope observed, for successive microwave acoustic pulse echoes in single crystals of quartz, from that predicted on the basis of phonon-phonon interactions. These models are based respectively on phase averaging over a surface for a wave with non-normal incidence and uniform initial excitation, wave propagation in an isotropic medium with an arbitrary initial excitation, and wave propagation in an anisotropic medium for an arbitrary initial excitation. The amplitude envelope for each of these models has been calculated. Intrinsic attenuation has been ignored. The result of these calculations is to separate the solutions into two classes – one that scales with frequency and one that does not. The calculated frequency dependence is compared with experiment, and it is found that a model based on wave propagation in an anisotropic medium with an arbitrary initial excitation is the only one that will fit the observed echo patterns.

R. L. Kyhl

B. INCOHERENT PHONON PROPAGATION IN X-CUT QUARTZ

Our work on incoherent phonon propagation in x-cut quartz, which was described in detail in Quarterly Progress Report No. 77 has advanced in three areas: (i) additional peaks have been resolved in the infrared phonon signal that arrives at the bolometer-detector; (ii) one of the unknown peaks in the signal has been identified, and (iii) there is evidence that we have observed dispersion effects in the propagation of the infrared phonons.

1. Resolution of Longitudinal and Fast-Transverse Pure Modes

The summary of our pulse-delay data presented in our last report[1] suggested that the first incoherent phonon pulse was actually a superposition of the longitudinal and fast-transverse pure modes whose wave vectors lie along the x crystallographic axis. In a succeeding experiment we have been able to resolve two discrete peaks in place of this single superposition. Figure II-1 shows this improved signal pattern. The labels "L", "FT", and "ST" stand for the longitudinal, fast-transverse, and slow-transverse pure modes, respectively. The "O" stands for an oblique mode that is discussed below. The origin of the two pulses labeled "U" is unknown.

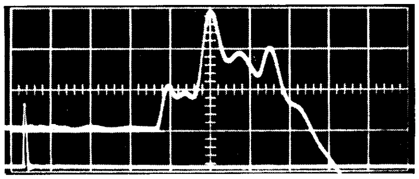

Fig. II-1. Pulses of incoherent phonons in x-cut quartz. The lower trace shows the generating pulse, 0. 1 μsec in duration, which marks zero on the time axis. The sweep rate is 1 μsec/cm.

2. Oblique Mode

In our last two reports,[1,2] we discussed at some length the peculiar characteristics of ultrasonic propagation in anisotropic media wherein the flow of ultrasonic energy does not generally coincide with the direction of the wave vector. This behavior was propounded as a possible origin of the "extra" pulses observed. We have concluded a search for oblique ultrasonic modes whose wave vectors deviate by large angles from their Poynting vectors lying within the solid angle subtended by the bolometer-detector.

Whenever extensive calculations pertaining to the physical properties of crystals are contemplated, it is prudent to begin with an examination of the crystal symmetry in order to avoid redundancy. Quartz belongs to the trigonal system; it is further classified by the point group D_3. The ultrasonic propagation characteristics of a crystal, however, obey the symmetry operations of the point group of the reciprocal lattice, which includes all of the allowed operations of the direct lattice point group plus inversion. Since the product of a twofold rotation about the x-axis and inversion amounts to a mirror reflection in the y-z plane, the ultrasonic propagation characteristics of quartz exhibit the symmetry of the holohedral D_{3d}.

A stereographic projection of the point-group symmetry elements of this class is shown in Fig. II-2. It will be useful for us to define a Cartesian system with the z-axis

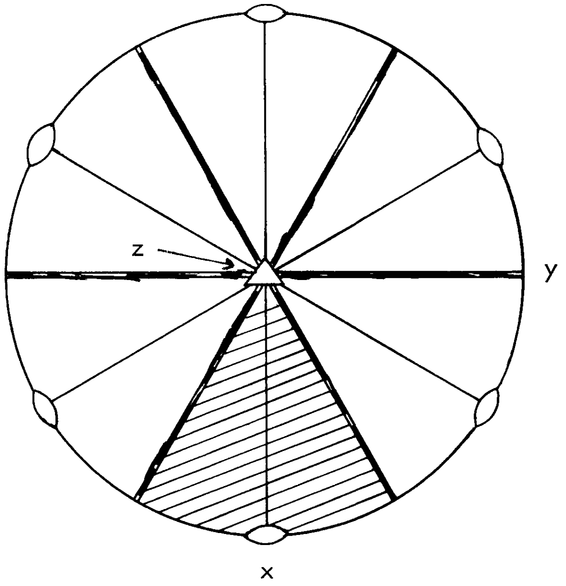

Fig. II-2. Stereogram of the trigonal point group D_{3d}. The z-axis is perpendicular to the plane of the paper; the x-axis can be taken along any of the three twofold axes. The crosshatched area shows an x-y projection of the minimum solid angle from which a knowledge of the entire crystal can be obtained from the allowed symmetry operations.

parallel to the threefold axis and the x-axis parallel to a twofold axis. We shall indicate directions in the crystal with the polar angles θ and ϕ. (The angle between an arbitrary vector and z-axis is θ. If the vector is projected onto the x-y plane, it makes an angle ϕ with the x-axis.)

Our considerations of the symmetry of crystalline quartz lead to a determination of the minimum solid angle that must be studied in order to determine the ultrasonic propagation characteristics of the entire crystal. We specify this solid angle by placing limits on the polar angles:

$$0 \leqslant \theta \leqslant 90 \qquad -30 \leqslant \phi \leqslant +30 \qquad (1)$$

This region is shown crosshatched in Fig. II-2.

Three large, elliptical areas of the unit sphere in wave-vector space are centered on the x-axis and contribute very nearly pure modes that propagate sufficiently close to the x crystallographic direction (within 9°) to be received by our bolometer-detector. The archetypes of these modes are the three rigorously pure modes whose wave and Poynting vectors are precisely collinear with the x-axis. The latter, of course, are the longitudinal, fast-transverse and slow-transverse pure modes, as indicated in Fig. II-1. To take a particular example, the elliptical area of the wave-vector unit sphere enclosing the quasi-longitudinal modes intercepted by our bolometer has a major axis that extends approximately 35° in length, and must be directed 90° to the major axis of the Poynting vector ellipse for the longitudinal mode calculated by Farnell.[3] In addition to these modes, we have found a group of quasi fast-transverse modes whose wave vectors pierce the unit sphere near the point specified by $\theta = 69.19°$ and $\phi = 18.40°$. The particular quasi fast-transverse mode whose wave vector penetrates the unit sphere at the given point has a Poynting vector lying within $\sigma = 0.12°$ of the x-axis and travels with a phase velocity $v_k = 3.89$ km/sec. We shall refer to modes whose wave and Poynting vectors deviate by large angles as oblique. The reason for using the term "quasi fast-transverse" is that the displacement vector of this mode is neither perpendicular nor parallel to the wave vector, so that this mode is neither longitudinal nor transverse in the most strict sense. The direction cosines of the displacement vector are

$$a_1 = -0.651$$
$$a_2 = +0.170 \tag{2}$$
$$a_3 = +0.740$$

The direction cosines of the wave vector are

$$1_1 = \sin\theta\cos\phi = .887$$
$$1_2 = \sin\theta\sin\phi = .295 \tag{3}$$
$$1_3 = \cos\theta = .356$$

If we form the dot product

$$\cos\chi = \sum_i a_i 1_i, \tag{4}$$

we find that the angle between the wave and displacement vector is $\chi = 105°$. The displacement is nearly perpendicular, hence, quasi-transverse.

In order to calculate the time delay of an oblique mode whose phase velocity v_k is known, it is necessary to know the angle ψ between the wave and Poynting vectors. If we represent the direction of the Poynting vector by the polar angles θ' and ϕ', the quantity ψ is given by

$$\psi = \cos^{-1}\left[\cos\theta\cos\theta' + \sin\theta\sin\theta'\cos(\phi-\phi')\right] \tag{5}$$

These quantities are then substituted in the following equation[4] for the total time delay of the k^{th} mode.

$$T_k = \frac{\ell\cos\psi}{v_k\cos\sigma}. \tag{6}$$

For the particular oblique quasi fast-transverse mode that we have just discussed, we calculate the angle ψ and the total time delay sustained in an x-cut quartz rod that is 0.75 inch long.

$$\psi = 27.7°$$
$$T_k = 4.34\ \mu\text{sec}. \tag{7}$$

Within our ability to resolve these heat pulses, this time delay corresponds to the largest pulse, third from the left, in Fig. II-1. We have, therefore, labeled this pulse "O" in the figure, thereby indicating that it is formed by oblique modes of infrared phonons. (This pulse was labeled "X" in our previous report.[1])

There are two pulses that have not yet been identified. These are labeled "U" in Fig. II-1. We are reasonably certain that these are not formed by oblique modes whose Poynting vectors lie sufficiently close to the quartz rod axis to provide direct flow of infrared energy between the generating film and the bolometer-detector. On the other hand, the wavelengths of the infrared phonons forming these heat pulses are of the order of 50 Å; therefore, we had felt justified in assuming that all rays that intercepted the walls of our quartz rod suffered diffuse reflection. In view of the experimental evidence, we now feel that this possibility should be examined in greater detail before the existence of a collective mode of phonon propagation at 3.5°K is postulated.

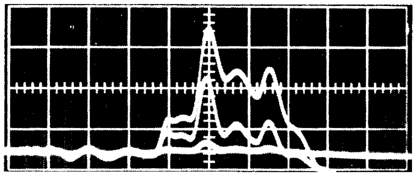

Fig. II-3.

Pulses of infrared phonons in x-cut quartz as a function of input microwave power. Each trace represents a change of 10 db. The 0.1-sec increase in total time delay as the power is changed 20 db is suggestive of the dispersion effects associated with phonons whose wavelengths approach the lattice dimensions.

3. Dispersion

In our last report,[1] we suggested that these heat-pulse experiments may involve phonons whose frequencies are sufficiently high to exhibit dispersion effects. In an attempt to demonstrate dispersion by a shift in the peaks of the heat pulses, we increased the absorption efficiency of our generating film 70 per cent, and raised the microwave input power by a factor of 15. Thus, the heat pulses shown in Fig. II-1 represent an over-all power increase of 25, in comparison with those shown in the last report.[1] The power was then successively decreased by factors of 10 and 100 by means of a microwave attenuator, and the positions of the pulses were observed to shift very slightly. Figure II-3 shows a superposition of the pulses as the power is varied. The power level changes 20 db between the top and the bottom traces. The peak of the third pulse seems to shift approximately 0.1 μsec to the right as the power is increased; this suggests a slight increase in the group velocity of the dominant phonons constituting the pulse.

J. M. Andrews, Jr.

References

1. J. M. Andrews, Jr., "Observations of Incoherent Phonon Propagation in X-Cut Quartz," Quarterly Progress Report No. 77, Research Laboratory of Electronics, M.I.T., April 15, 1965, pp. 7-15.
2. J. M. Andrews, Jr., "Incoherent Phonon Propagation in Anisotropic Media," Quarterly Progress Report No. 75, Research Laboratory of Electronics, M.I.T., October 15, 1964, pp. 5-7.

3. G. W. Farnell, Can. J. Phys. 39, see Fig. 7.
4. J. M. Andrews, Quarterly Progress Report No. 77, *op. cit.*; see Fig. II-3 and Eq. 1.

C. INVESTIGATION OF THE FERMI SURFACE OF GALLIUM BY GEOMETRIC RESONANCE AT MICROWAVE FREQUENCIES

Fermi surfaces of single-crystal Gallium have been investigated by using geometric resonances with L-band ultrasonic waves and a field of 0.1 ≈ 3k Gauss.

Metallic Gallium with 99.9999 per cent purity (purchased from United Mineral and Chemical Corporation, New York) was prepared as a single-crystal grown between the parallel surfaces of two x-cut quartz cylinders. The orientation of the crystal has been verified by x-rays within one degree.

The experimental arrangement is shown in Fig. II-4. Pulsed ultrasonic waves of ~900 Mc, 1000 p.r.f., and 1 μsec in duration, were generated in the transmitting transducer, passed through the sample, and supplied to the receiver through the receiving transducer. To discriminate the transmitted signal from echoes that were generated by the highly parallel end surfaces of the quartz rods, a pulse-gate circuit was used. The detected signal was amplified and the amplitude was recorded against the change of the magnetic field. The cavity and sample were put into liquid Helium in dewar and the whole unit was placed in a magnetic field.

All experiments were made with a longitudinal wave and a magnetic field perpendicular to the direction of wave propagation. In order to study the Fermi surfaces, the sample was rotated in a plane perpendicular to the direction of wave propagation over approximately 180° and measurements were made at 10° intervals.

In Fig. II-5, the transmitted pulse amplitude is plotted as a function of the field for a 883-Mc sound wave travelling along the a-axis at 4.2° K and a magnetic field oriented by θ with respect to the b-axis. Only three cases, $\theta = 65°, 115°$, and $155°$, are presented

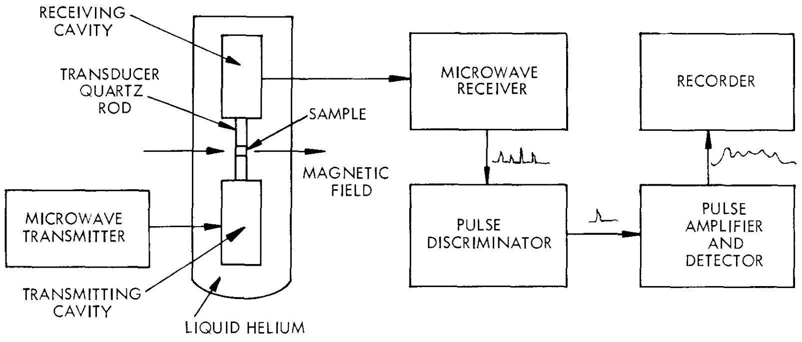

Fig. II-4. Experimental arrangement.

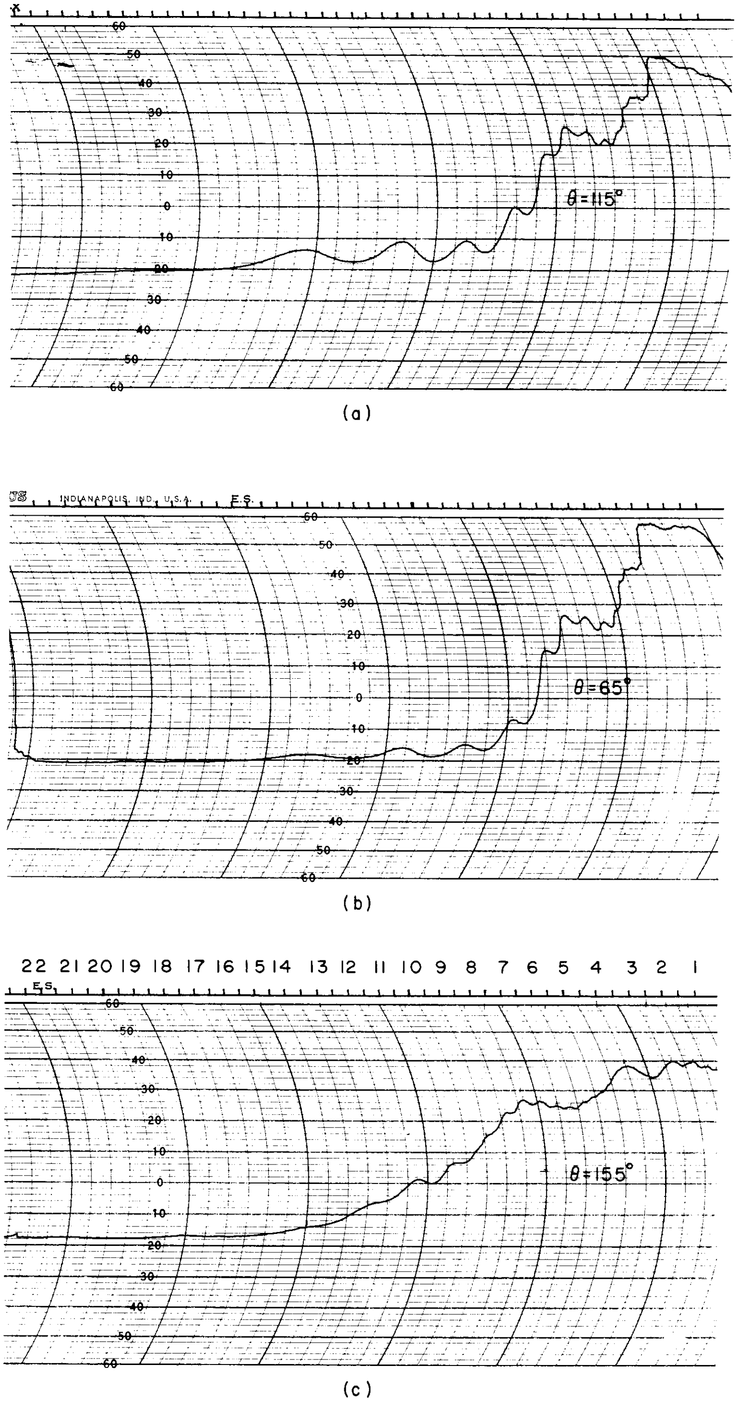

Fig. II-5. Transmitted signal strength vs magnetic field.
(a) θ = 65°. (b) θ = 115°. (c) θ = 155°.

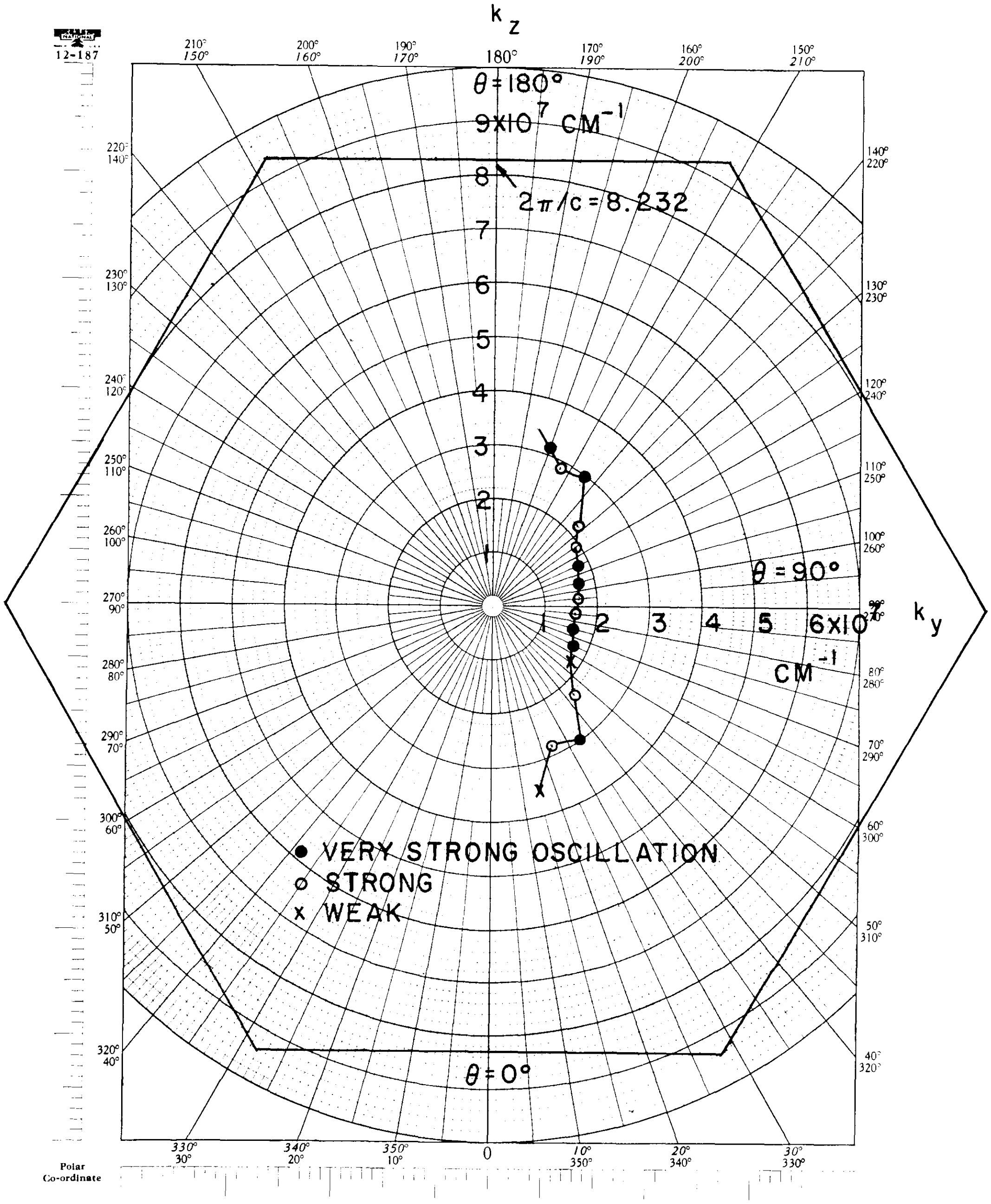

Fig. II-6. Fermi surface derived from data for Series I.

here. The reciprocal field, 1/H, at subsequent maxima and minima shows a periodicity that indicates the geometric resonances. At least two series have been found to be present, one of which, Series I, has been traced continuously with the change of angle θ, while Series II has been traced only at several angles near 5° and 160°.

Using the approximate formula for the subsequent maxima in geometric resonances,

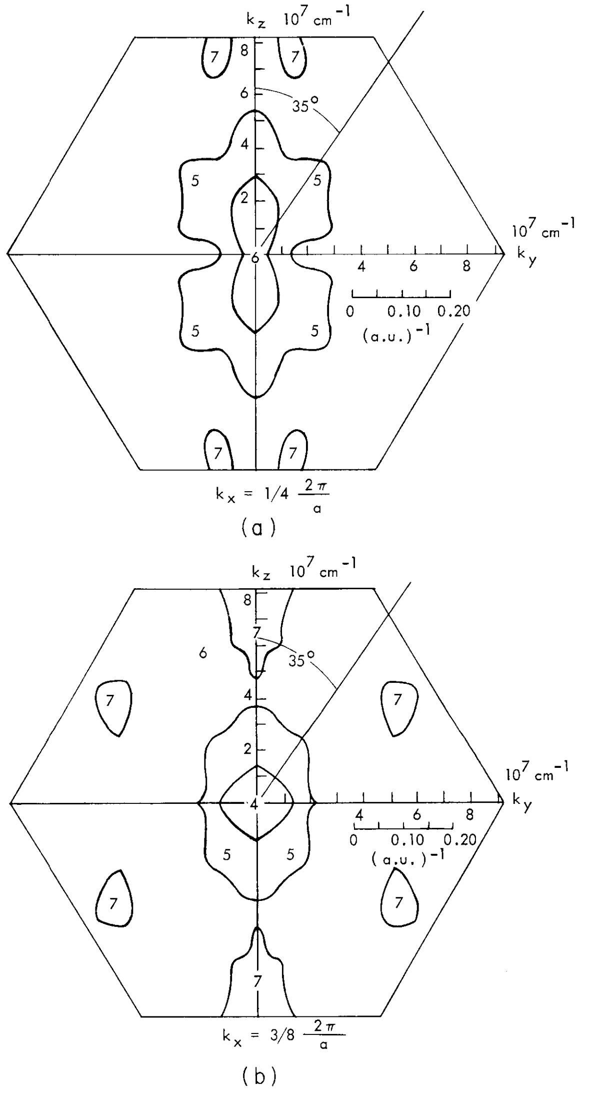

Fig. II-7. Constant energy surface for gallium.

$\Delta\, 1/H = \lambda/2\ e/c\ 1/\hbar k$,[1] we have calculated the wave vector k at the Fermi surface for θ between 5° and 160° for Series I. These vectors are plotted in the first Brillouin zone in Fig. II-6. This shows a two fold symmetry with respect to the k_y-axis which is also obvious from comparing Fig. II-5a and 5b — also, the strength of oscillations has been indicated roughly as shown in the figure. Strong oscillation suggests the existence of a point of high symmetry when the density of states is extremely large.

Wood has recently calculated[2] Fermi surfaces of single-crystal Gallium from an APW method. Some of his results are shown in Fig. II-7. Comparison with the present experiment indicates that band 5, as determined with a Fermi energy of 0.400 Rydberg, shows good agreement with Fig. II-6. It is assumed that as the angle θ changes from 90° to 180° the orbits contributing to the resonance move from point 1 through point 3 in Fig. II-7, with k_x somewhere between $1/4\ 2\pi/a$ and $3/8\ 2\pi/a$ taking extremum geometry successively.

A. Fukumoto

References

1. M. H. Cohen and M. J. Harrison, "Magnetic-Field Dependence of the Ultrasonic Attenuation in Metals," Phys. Rev. 117, 937-952 (February 15, 1960).
2. J. H. Wood, "Constant Energy Surface for Gallium," Quarterly Progress Report No. 55, Solid State and Molecular Theory Group, M. I. T., Cambridge, Mass., January 15, 1965, p. 9.