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MAGNETIC FIELD INDUCED FAR INFRARED
TRANSMISSION IN BISMUTH

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Magnetic Field Induced Far Infrared

Transmission in Bismuth*

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ABSTRACT

It has been demonstrated that high fields render the semi-metal bismuth sufficiently transparent at far infrared frequencies to study effects not previously seen in reflection experiments. The frequency interval $10 - 400 \text{ cm}^{-1}$ was explored at fields up to 100 kG using Fourier transform spectroscopic techniques and He^3 -cooled bolometer detectors, and samples were cooled to liquid helium temperatures. Transmission was observed in all three principal crystal directions when the magnetic field was sufficiently large to drive one or more sets of electrons into the extreme quantum limit. The reduced absorptivity at these fields results from a large reduction in the relaxation rate.

Efforts were concentrated on the geometry where the magnetic field and Poynting vectors were both parallel to the crystal bisectrix axis, and measurements at the highest

fields indicated that the relaxation rate increases as the 1.4 ± 0.1 power of the frequency and that $\omega\tau \approx 700$ at 30 cm^{-1} . The temperature dependence of the relaxation rate continues to be quadratic at these fields.

Three resonances were observed at the hole cyclotron resonance frequency ω_c and at $2\omega_c$ and $3\omega_c$, and they appear to arise from small departures of the hole Fermi surface from a perfect ellipsoid. Since these harmonics occur in the normal mode where the electric vector and the holes are rotating in opposite senses, they correspond to the $n = -1, -2$ and -3 Fourier components of the motion in a labelling scheme where the cyclotron resonance itself is the much stronger $n = +1$ component. Other resonances were seen, and one was identified as a magnetoplasma resonance.

SECTION I.

INTRODUCTION

It has been pointed out by Buchsbaum and Galt¹ that Alfvén waves, first proposed to explain certain phenomena in astrophysics,² could also propagate in solid state plasmas such as bismuth. This has led to several investigations of Alfvén wave propagation in bismuth, primarily at microwave frequencies,³ but also at megacycle frequencies.⁴ Alfvén waves have also been studied in antimony⁵ and graphite.⁶

The lack of similar experiments at far infrared frequencies can be presumed to stem from some or all of the following difficulties: (1) Microwave results suggested that the required condition $\omega\tau \gg 1$ would not be met at higher frequencies; (2) the Alfvén wave theory restricts itself for simplicity to frequencies much lower than that of the lowest cyclotron resonance; (3) experiments at far infrared frequencies are hampered by a dearth of strong sources.

Nevertheless, some transmission studies have been carried out on bismuth at far infrared frequencies. Boyle and Brailsford⁷ have carried out zero-field and low-field investigations at frequencies above 150 cm^{-1} , using samples less than 0.05 mm thick. Burgiel and Hebel⁸ have explored transmission in the trigonal

direction of bismuth in the frequency range $13 - 85 \text{ cm}^{-1}$ at fields up to 50 kG, using samples up to 0.02 mm thick. Carter and Picard⁹ have used microwave techniques at 20 cm^{-1} to study transmission in fields up to 60 kG, again in the trigonal direction, but using samples up to 2 mm thick. Only the last of these experiments gave an indication that higher fields would result in greatly improved transmittance at far infrared frequencies.

While carrying out reflection measurements on a sample of bismuth, we found that increasing the magnetic field above 10 kG produced a 13% drop in the reflected power. This prompted an attempt to measure the transmission through the 3 mm sample, and the result was a spectrum rich in details as well as interesting in itself.¹⁰ This success was followed by a series of transmission experiments on samples of all orientations and thicknesses at fields up to 100 kG. The results of these measurements constitute the subject of this article.

Sections II and III describe the details of the instrumentation and methods used in these experiments. Section IV reviews what is known about the electronic structure of bismuth, including the changes brought about by the application of a large uniform static magnetic field. Section V is devoted to the general theory for electromagnetic radiation propagating through an anisotropic plasma such as bismuth, without making the simplifying

approximations used in the Alfvén-wave limit. Section VI is a presentation of the various results of our investigations in terms of this theory.

SECTION II.

INSTRUMENTATION

A. General Description

The experimental setup is summarized in Figure 1, and has been described elsewhere.¹¹⁻¹³ Radiation from a mercury arc lamp is modulated by a brass chopper wheel, passes through either a Michelson or Strong lamellar-type interferometer and through various filters to remove high frequency components of the radiation, and finally passes through the sample of interest before being detected. The detector is a crystal bolometer of germanium doped with indium and antimony,¹⁴ which is cooled to 0.35°K by helium-3 refrigeration. The electrical output of the detector is combined with a reference signal from the chopper wheel by a lock-in amplifier and then digitalized for computation by an on-line computer. The computer is a Digital Equipment Corporation PDP 11/20 minicomputer with 8K words of core memory, Teletype keyboard and printer, high-speed paper tape reader and punch, and an extended arithmetic unit. The display units are an oscilloscope and an x-y point plotter, driven by digital-to-analog converters.

B. Magnet Cryostat

Our initial far infrared transmission measurements on bismuth were carried out using a magnet cryostat consisting of a 50 kG superconducting solenoid into which was inserted a long light pipe complete with sample and filters, and below which was mounted a He^3 -cooled bolometer operating at 0.35°K.

To carry out experiments in fields up to 100 kG, it was necessary to design a cryostat which could be inserted through the 1½-inch bore of a General Electric 100 kG superconducting magnet. This insert is shown in Figure 2, and is based in part on earlier cryostats built in our laboratory.^{15,16} Far infrared radiation enters the light pipe at the upper right of the figure, passes through the sample and filters, and then passes through a vacuum window to a detector cooled by a noncycling liquid- He^3 evaporator. The light pipe assembly is removable to facilitate sample changes.

A noteworthy innovation was the concentric arrangement of a sample section and the He^3 pumpline, with the sample section now doubling as the 1.2°K evaporator for condensing the He^3 gas during startup. The concentric arrangement made it possible for the sample section to accommodate samples up to an inch in diameter, as well as providing a large surface area for heat transfer during condensation.

A second innovation was the addition of an air lock system to prevent air from entering the sample section and forming "ice" on the window at the bottom. A large ball valve with a one-inch bore was mounted directly on the topplate, with a second flange above it. A removable polyethylene bag about five feet long could then be clamped onto the upper flange with a large hose clamp to form a tolerably good seal. A system of valves and pumps allowed the gas bag to be purged of air and re-filled with helium prior to opening the ball valve and lowering the sample and light pipe assembly. This system permitted upwards of twenty sample changes with little deterioration in signal strength, an important advantage for magnetic field experiments where there is no room for more than one sample at a time.

This cryostat differs from earlier ones in other ways as well. First, because space restrictions made it impossible to mount a quartz window complete with flange and indium o-ring, a $\frac{1}{2}$ -inch diameter sapphire blank brazed with copper into a stainless steel ring (0.825" o.d.) was purchased from Linde Division of Union Carbide.

Second, the detector is about 18 inches below the sample, thus assuring good thermal isolation for the detector and facilitating temperature-dependence studies. In fact, the bottom of the sample section is typically at about 2°K even when

the needle valve is opened and liquid helium is allowed to fill the sample section from the outside bath. Apparently the rising He^3 gas cools the sample section on its way up the pumpline. This allows one to raise the temperature of the sample with minimal effect on the detector's operating temperature.

SECTION III.

EXPERIMENTAL TECHNIQUES

A. Sample Preparation

The bismuth samples were prepared from 99.9999% pure rods and ingots (69 Grade) purchased from Cominco American, Inc. of Spokane, Washington, and were grown in a zone levelling furnace consisting of an evacuable quartz tube and a mechanically driven cylindrical furnace element.

Thick single crystal samples were grown in the graphite boat shown in Figure 3, which incorporates features used by H. D. Drew and U. Strom at the University of Maryland. In particular, a stack of glass microscope slides is used to provide thermal isolation of the seed crystal from the graphite itself. Seventy grams of starting material is placed in regions C and D, and then is melted down so that it fills regions B, C and D entirely and overflows some into region A. After cooling, an oriented seed crystal, if one is available, is placed adjacent to the melt in region A. The furnace element is then brought in from the left and the portions in regions D, C and B are successively melted. Finally, the entire melt and a portion of the seed crystal are both molten, but surface tension can prevent the two from joining. To avoid this, the seed can be inserted just under the melt at the outset. Alternatively, the furnace can be tilted or subjected to violent movements in order to

effect fusion of the two.

The furnace is then slowly withdrawn to the left so that a single crystal is grown from the seed crystal. The cross-section of the liquid solid interface where regions A and B join was made small (0.1 cm^2) because seed crystals are sometimes small and because in the absence of a seed only one crystallite is likely to exist in a constricted region. In region B the vertical dimension grows to a half inch, and in region C the horizontal dimension expands to its full size as well. Separating the regions of vertical and horizontal growth simplified the work of machining the graphite boat. It should be noted that regions B and C are not interchangeable, because the strong surface tension of bismuth makes it difficult to fill a region which is wide but shallow. Also, region E should be at least as long as the furnace element, so that the heat input remains stable as the element is slowly withdrawn during crystal growth.

Bismuth expands upon solidification, and the boat was designed so that it would not confine the crystal and thereby strain it. The free surface at the top prevented vertical stresses, and the split mold design avoided stresses between the two parallel sides. Longitudinal stresses were minimal because the crystal was grown in this direction; in any event, the one end is tapered. The split mold also facilitated extraction

of the grown crystal from the mold.

Two different etching solutions were used when preparing samples. For analysis, the most useful was a 50-50 solution of concentrated nitric acid and water. This solution attacks the different crystal faces with varying effect, so that regions of substantially different orientation are easily identified by sudden changes in the appearance of the etched surface across a boundary. Absence of such patterns is a good indication that the sample is a single crystal.

During the various stages of sample preparation, concentrated nitric acid alone was a useful etchant. For the final preparation of a shiny surface free of salt residues, a 50-50 solution of concentrated nitric acid and glacial acetic acid was used. Rapid agitation was required to prevent the accumulation of a layer of salt.

Crystals were oriented by analyzing Laue x-ray reflection photographs. It was possible to align a crystal to within 0.5° in this manner, and it is believed that all samples were aligned to within one degree of the designed orientation when the actual transmission measurements were made. The exception was "Bi-sectrix no. 3," which was consistently misaligned by about 5° .

Following orientation, the samples were spark cut at a low rate to minimize risk of straining the crystals. Samples up to 1.5 cm thick were prepared, and they are listed in Table 1. No

Table 1
Bismuth Samples

<u>Orientation</u>	<u>Sample No.</u>	<u>Thickness (mm)</u>
Bisectrix	1	0.15
Bisectrix	2	1.30
Bisectrix	3*	3.10
Bisectrix	4	9.70
Bisectrix	5**	11.1
Binary	1	0.15
Binary	2	0.80
Binary	3	1.33
Binary	4	15.45
Trigonal	1	2.65

* Provided by H. D. Drew, University of Maryland.

**This sample was later cut into Bisectrix No. 2 and No. 4.

attempt was made to produce optically smooth faces; spark cutting typically left a slight ridged structure which was smoothed by etching but not removed entirely by any means. However, "Bisectrix No. 3," which was prepared with reflection studies in mind, had faces of optical quality.

Audio frequency skin effect measurements¹⁷ indicated that the resistivities of our samples at helium temperatures were

at least 200 times lower than at 300°K.

B. Sample Mounting

For the transmission experiments, it was important that no radiation be allowed to leak around the sample. It was also important to mount the crystal in a stress-free environment. Early experiments were performed with the sample lightly pressed against the end of a metal light pipe with a soft steel spring. The metal-to-metal light seal proved to be quite unsatisfactory, since the polarization with electric vector normal to the surface propagates rather easily. The smallness of the gap greatly enhances the absorption by the metal faces, but not sufficiently well to prevent a substantial leak signal from reaching the detector.

To eliminate the leakage problem a light-absorbing material was introduced, as shown in Figure 4. Charcoal was ground into powder, mixed with vacuum grease, and applied to the brass surface. It was then covered with a thin mylar ring. The plastic cover prevented the sample from sticking to the vacuum grease and becoming strained when cycled between 300°K and 4°K. The thin mylar sheet with the charcoal on the one side transmitted very little radiation, thus solving the problem of leakage for all but the very lowest of signals.

Measurements in the Voigt geometry required a more elaborate sample mount, since the radiation had to be redirected so that

it would traverse the sample at right angles to the magnetic field. Figure 5 shows how this was accomplished within the one-inch-diameter cylindrical sample section of the cryostat. A long thin mirror reduced the cross-section of the light pipe by half, and another brass mirror reflected the radiation through a sample mounted vertically between two brass plates. This was not a strain-free mounting, but by surrounding the sample disk with a brass ring which was slightly thicker than the sample itself, a satisfactory mount was obtained.

Still another mount was used to measure transmission at a skew angle, and it consisted of a sample sandwiched between the two halves of a very thick brass ring which had been cut at an oblique angle. This had the effect of tilting both the sample and the direction of propagation, since the large refractivity of metals causes all radiation to enter at near-normal incidence. Therefore, the geometry was midway between the Faraday and Voigt geometries.

A final mount was used on occasion when the signal levels were very low. It consisted of soldering the sample to a 0.001-inch thick copper foil with a bismuth-rich low temperature solder (Cerrolow). This method eliminated leakage altogether, whereas the charcoal-and-grease method still allowed a detectable amount of radiation through. The sample did not appear to be adversely affected by the experience; however, no quantitative measurements were made to verify this.

C. Far Infrared Measurements

Using the system described in Section II, spectra showing transmitted power as a function of frequency (in cm^{-1}) were obtained. With one sample in place, the applied magnetic field was varied to obtain several spectra. Also, spectra were taken through samples of varying thickness. Both techniques were employed because transmitted power is a function of both the absorptivity and reflectivity of the sample, as well as its thickness. For example, if the attenuation coefficient α is small and the reflectivity is large then the fractional power attenuation per unit sample length is not constant because of multiple reflections. For frequencies greater than 10 cm^{-1} , the wavelength in a medium with index of refraction 10 is 0.1 mm or less, whereas the sample thickness typically varied by 0.2 mm or more over the sample. Consequently, the equation to use in describing multiple reflections combines the intensities to give the transmittance

$$T = \frac{(1-R)^2 \exp(-\alpha x)}{1-R^2 \exp(-2\alpha x)},$$

where R is the reflectivity. The $(1-R)^2$ term takes into account reflection losses at the two surfaces, and the denominator incorporates the effect of multiple reflections. To analyze data, it is useful to obtain an experimental attenuation coefficient α' by measuring the intensities passing through two

samples of differing thickness and calculating α' as follows:

$$\alpha' = (x_2 - x_1)^{-1} \ln(I_2/I_1).$$

This experimental quantity is related to the actual attenuation coefficient by

$$\alpha' = \alpha + \frac{1}{x_2 - x_1} \ln \frac{1 - R^2 \exp(-2\alpha x_2)}{1 - R^2 \exp(-2\alpha x_1)}.$$

For α' to approximate α , it is important that the source intensity and detector sensitivity remain constant for the two measurements. Note that the $(1-R)^2$ terms drop out, but that an additive correction term arises from the differing effect of multiple reflections on samples of different thicknesses.

Efforts were concentrated on bisectrix samples with propagation parallel to the bisectrix axis and the applied magnetic field (Faraday geometry), and the bulk of the ensuing analysis relates to these measurements.

SECTION IV.

ELECTRONIC PROPERTIES OF BISMUTH

A. Crystal Structure

Metals are characterized by free electron densities of 10^{22} cm^{-3} , corresponding to something on the order of one electron per atom. Bismuth on the other hand is characterized by a free charge carrier density of about one electron and one hole per 100,000 atoms, and is therefore designated a semimetal. Together with the semimetals arsenic and antimony, bismuth occupies the Group VA column in the periodic table, and its crystal structure can be visualized as follows: (1) take a cubic lattice with an atom on each site; (2) separate it into two face-centered cubic sublattices and allow one of them to move slightly with respect to the other along a body diagonal of the original simple cube; (3) elongate the structure along that same body diagonal, hereafter referred to as the trigonal axis because it still retains its three-fold rotational symmetry. At right angles to the trigonal axis are three binary and three bisectrix axes, with each bisectrix axis 30° from a binary axis on either side. An orthogonal system for describing the crystal is formed from a binary (1) axis, the bisectrix (2) axis at 90° to it, and the trigonal (3) axis. The unit cell is shown in Figure 6.

The distortion of the crystal structure from cubic symmetry is small, on the order of 5%. Hence, there are still pseudo-threefold and pseudofourfold axes present in Laue x-ray photographs of the ionic structure. However, the electronic structure is profoundly altered by the distortion. There are now two atoms per unit cell, with five electrons each. These fill the first five Brillouin zones of the reciprocal lattice, with a small number spilling over into the sixth zone and leaving behind a pocket of holelike carriers in the fifth zone. In the reduced zone scheme, the holes are located in a pocket along the trigonal axis and straddling the zone boundary. The electrons occupy three pockets located at the L points of the zone.¹⁹ See Figure 7.

B. Anisotropic Bandstructure

To a first approximation, the highly anisotropic electronic structure of bismuth can be described by introducing into the free electron model an inverse mass tensor

$$\alpha_{\approx} = (\hbar)^{-2} \nabla_k^2 E(k)$$

such that

$$E = \frac{1}{2} \mathbf{p} \cdot \alpha_{\approx} \cdot \mathbf{p}.$$

The notation used throughout this article is summarized in Appendix I. The hole pocket is an ellipsoid of revolution about the trigonal (3) axis, and

$$\alpha^{(h)} = \begin{pmatrix} 1/M_1 & 0 & 0 \\ 0 & 1/M_1 & 0 \\ 0 & 0 & 1/M_3 \end{pmatrix}$$

One of the pockets of electrons is an ellipsoid with its three axes along the 1, 2 and 3 axes, but tilted 6° away from the bisectrix axis toward the trigonal axis. For this pocket,

$$\alpha = \begin{pmatrix} \alpha_1 & 0 & 0 \\ 0 & \alpha_2 & \alpha_4 \\ 0 & \alpha_4 & \alpha_3 \end{pmatrix}$$

The inverse mass tensor for the other two pockets is obtained by $\pm 120^\circ$ rotations about the 3 axis.

This model is rather good for describing the energy-momentum relationship for the holes. However, the electron bands are highly nonparabolic. A model which enjoys considerable success plus minimal added complexity is the two-band model developed by Kane²⁰ for semiconductors, and applied to bismuth by Lax and collaborators.²¹ In this case, the presence of a second band nearby modifies the quadratic energy-momentum relationship so that

$$E(1 + E/E_G) = \frac{1}{2} \tilde{p} \cdot \tilde{\alpha} \cdot \tilde{p} ,$$

where E_G is the gap energy. See Figure 8. The parabolic model is recovered formally by letting E_G become infinite. Since the hole band is also slightly non-parabolic, a large but finite

gap energy has been estimated for it.³ Some typical published values for these constants are given in Table 2.

Cohen²² has introduced a modification of the two-band model which incorporates the effect of other bands by adding terms which are quartic in the momentum. The result is a Fermi surface which is no longer ellipsoidal. The model is aptly referred to as the non-ellipsoidal nonparabolic (NENP) model. Experimentally, however, ellipsoidal models have proved adequate for describing most results. (An exception is the analysis by Dinger and Lawson²³ of the cyclotron resonance measurements of Edel'man and Khaikin²⁴ at limiting points on the Fermi surface.) The two-band model has the further advantage that its energy-momentum relation is formally equivalent to that of the relativistic electron. Wolff²⁵ exploited this fact to obtain matrix elements and selection rules for the two-band model.

Table 2

Bismuth Bandstructure Parameters

Energy Band Parameters*

E_F	28 meV
E_G	15 meV
E_o	39 meV
$E_G^{(h)}$	225 meV

Mass Tensor and Inverse Mass Tensor at Fermi Surface**

m_1	0.0071	α_1	141.0
m_2	1.50	α_2	1.85
m_3	0.0301	α_3	92.3
m_4	0.170	α_4	-10.5
M_1	0.067	β_1	14.9
M_3	0.76	β_3	1.32

Equations Relating m and α

$$m_1 = \frac{1}{\alpha_1}$$

$$m_2 = \frac{\alpha_3}{\alpha_2 \alpha_3 - \alpha_4^2}$$

$$m_3 = \frac{\alpha_2}{\alpha_2 \alpha_3 - \alpha_4^2}$$

$$m_4 = \frac{-\alpha_4}{\alpha_2 \alpha_3 - \alpha_4^2}$$

*From Smith, Baraff and Rowell.²⁶

**From Takano and Kawamura.²⁷

Effects of Magnetic Field on Bandstructure

The application of a large magnetic field dramatically changes the properties of a free electron gas. The uniform distribution of states in momentum space condenses into a system of concentric cylinders with discrete values of angular momentum, known as Landau levels.²⁸ For a magnetic field H ,

$$E = (n + \frac{1}{2})\hbar\omega_c + p_H^2/2m,$$

where $\omega_c = |e|\hbar/mc$ is the cyclotron frequency of the electrons, p_H is the component of the momentum parallel to the field, and $n = 0, 1, 2, \dots$.

The spin of the electron adds another term

$$\pm \mu_B H = \pm (eh/2mc)H = \pm \frac{1}{2}\hbar\omega_c$$

so that

$$E = (n + \frac{1}{2} \pm \frac{1}{2})\hbar\omega_c + p_H^2/2m.$$

Therefore, the spin-splitting does not reduce the degeneracy of any except the lowest Landau level. For example, the $0+$ and $1-$ levels are degenerate.

For electrons in bismuth, the masses must be replaced by suitable combinations of the effective mass tensor components:

$$m^* \approx \alpha^{-1}$$

$$m_H^* \approx H \cdot m^* \cdot H/H^2$$

$$m_c^* \approx (\det m^*/m_H^*)^{\frac{1}{2}}.$$

For the two-band model, we now have

$$E(1 + E/E_G) = (n + \frac{1}{2} \pm \frac{1}{2})\hbar\omega_c^* + p_H^2/2m_H^*,$$

where

$$\omega_c^* = |e|\hbar/m_c^*c.$$

It should be noted that m_c^* and m_H^* are the effective masses at the bottom of the band only. Differentiation yields the result that masses at the Fermi surface are related to those at the bottom by the relation

$$\tilde{m}_c^*(E_F) = (1 + 2E_F/E_G)\tilde{m}_c^*.$$

Table 2 contains experimentally determined values for the components of $\tilde{m}^*(E_F)$ in the various crystal directions.

The two-band model does not predict any spin splitting of the levels. However, large spin splittings have been observed in bismuth by Smith, Baraff and Rowell.²⁶ Sakai, Matsumoto and Mase²⁹ have introduced a spin splitting factor γ defined by the equation

$$E(1 + E/E_G) = (n + \frac{1}{2} \pm \frac{1}{2}\gamma)\hbar\omega_c^* + p_H^2/2m_H^*.$$

γ is roughly the ratio of spin splitting to orbital (Landau) splitting, and measured values are found to deviate substantially from unity as seen from Table 3.

Table 3

Spin Splitting Factors in Bismuth*

<u>Carrier</u>	<u>Bisectrix</u>	<u>Binary</u>	<u>Trigonal</u>
Electrons (A)	1.10	0.36	0.6
Electrons (B,C)	1.08	1.10	0.6
Holes	0.14**	0.14**	1.86

*From Sakai, Matsumoto and Mase.²⁹

²⁶
**From Smith, Baraff and Rowell.

The densities of the electrons in bismuth are about $1 \times 10^{17} \text{ cm}^{-3}$ per ellipsoid, and that of the holes is about $3 \times 10^{17} \text{ cm}^{-3}$. However, the densities of all four groups of carriers is known to vary strongly with the applied magnetic field, and a calculation for the bisectric direction was carried out as prescribed by Smith, Baraff and Rowell,²⁶ but with the bottom Landau levels moving linearly with field as predicted by Baraff³⁰ and observed by Takano and Kawamura.²⁷ For each field, the positions and degeneracies of the Landau levels were calculated and a new Fermi energy was found by requiring charge neutrality. The results are plotted in Figure 9. The corresponding densities of the electrons and holes are plotted in Figure 10. It can be seen that all of the carrier densities increase markedly at high fields.

For a free electron gas, the Landau levels are evenly spaced as in Figure 11 and all of the allowed transitions are of frequency ω_c . In the two band nonparabolic model, the Landau levels are no longer evenly spaced. This has no effect at low fields, since the transitions are all between states close to the Fermi surface and the effective cyclotron mass is simply

$$m_c^*(E_F) = (1 + 2E_F/E_G)m_c^*,$$

where m_c^* is the cyclotron mass at the bottom of the band.

The situation at high fields is quite different. As the

applied field is increased, the energy separating adjacent Landau levels becomes comparable to the Fermi energy, and the number of levels that remain occupied become fewer and fewer. The degeneracy of the levels, however, compensates for this,³¹ so that the Fermi surface remains relatively fixed until the last few levels pass through it. When a field has been attained such that only the bottommost level is still occupied, the electrons are said to have passed into the extreme quantum limit (EQL). Because of the large effective masses and small Fermi energies in bismuth, this limit can be attained with fields as low as 15 kG for the light-mass electrons in the binary and bisectrix directions.

Figure 12 shows the Landau levels for the heavier electrons at 25 kG. Since these electrons are in the extreme quantum limit and only the lowest Landau level is occupied, all cyclotron transitions are between the 0- and 1- levels ($\Delta n = \pm 1$, $\Delta s = 0$). But due to the nonparabolic bandstructure, the energy involved in the transition is a strong function of the longitudinal component of the momentum. Thus the single cyclotron frequency ω_c is replaced by a band of frequencies. Figure 13 shows a plot of this band of frequencies as a function of the applied magnetic field, together with a straight line representing the value of ω_c obtained from the low field cyclotron mass. It is apparent that the straight line does not represent the high

field cyclotron frequencies well at all. To obtain a more representative value, we observe that the density of states contributing to each frequency in the band is fairly uniform, so that a weighted average of the frequencies yields a value $\bar{\omega}_c$ close to the middle of the band. This average is indicated in the figure by the dashed line, and it is found empirically to vary roughly as the two-thirds power of the magnetic field.

This situation changes quickly as the field drops below the threshold of the extreme quantum limit. Figure 14 shows the situation at 23 kG. The 0+ to 1+ and the 1- to 2- transitions are now possible, and they supplant the largest 0- to 1- transitions. The results are summarized in Figure 15, which shows a rapid narrowing of the band of frequencies, together with a concentration of the density of states at the lower end of the band. The effect of the two is to cause the average cyclotron frequency to drop rapidly to its low field value as the electrons pass out of the extreme quantum limit.

For calculational purposes, we used

$$\bar{\omega}_c = \begin{cases} \omega_c & (H < H_{EQL}) \\ \omega_c (H/H_{ref})^{2/3} & (H > H_{EQL}) \end{cases}$$

to approximate the behavior of the electrons. The actual parameters used in calculations were $H_{ref} = 50$ kG for the

heavier electrons and $H_{\text{ref}} = 25 \text{ kG}$ for the lighter electrons. For the holes, the EQL is never attained at available fields, and in any event the hole bands are much more parabolic.

SECTION V.

ELECTROMAGNETIC WAVES IN METALS

A. Classical Theory

In this section, we will begin with a general discussion of the propagation of electromagnetic radiation through a charged gas, and work progressively toward an accurate description of propagation of far infrared frequencies through bismuth.

For an electromagnetic (EM) wave of the form $\exp(i\omega t - i\mathbf{q} \cdot \mathbf{r})$ travelling in a medium of complex dielectric constant $\epsilon(\mathbf{q}, \omega)$, Maxwell's equations yield the following relation for the electric field:

$$-\mathbf{q} \times (\mathbf{q} \times \mathbf{E}) = \frac{\omega^2}{c^2} \epsilon \cdot \mathbf{E}.$$

This equation constitutes three simultaneous homogeneous equations for the three Cartesian components of $\mathbf{E}$. For a nontrivial solution, the determinant of the coefficients must vanish, and this constitutes the dispersion relation. For $\mathbf{q}$ directed along the z-axis, the determinantal equation is

$$\begin{vmatrix} \epsilon_{xx} - N^2 & \epsilon_{xy} & \epsilon_{xz} \\ \epsilon_{yx} & \epsilon_{yy} - N^2 & \epsilon_{yz} \\ \epsilon_{zx} & \epsilon_{zy} & \epsilon_{zz} \end{vmatrix} = 0$$

where $N = cq/\omega$. This equation has solutions for two values of N^2 , corresponding to two normal modes of differing polarization. The objective, therefore, is to obtain an expression for ϵ in terms of the known properties of bismuth.

There are two contributions to the dielectric constant. The first is due to the polarizability of the ions and the static polarizability of the electron gas,³ and is denoted by ϵ_l . The free electron contribution is $-i4\pi\sigma/\omega$, where σ is the conductivity tensor relating the free charge current density to the applied electric field. Thus,

$$\epsilon = \epsilon_l - i4\pi\sigma/\omega.$$

For an isotropic plasma of non-interacting electrons moving without resistance through a uniform background of positive charge to insure electrical neutrality, $\sigma = ne^2/im\omega$, and hence

$$\epsilon = 1 - \frac{\omega_p^2}{\omega^2},$$

where $\omega_p^2 = 4\pi ne^2/m$ is the plasma frequency. In this case, the two solutions are degenerate and

$$N^2 = \frac{\omega^2}{c^2} \left(1 - \frac{\omega_p^2}{\omega^2}\right).$$

If we express N as $n - i\kappa$, where n is the usual index of refraction, we see that the wave propagates as $\exp(i\omega t - in_0 z - \kappa\omega z/c)$, where $q_0 = \omega/c$ is the wavevector in free space

($\epsilon = 1$) and $\kappa\omega/c$ represents the amplitude attenuation. The corresponding power attenuation coefficient is $\alpha = 2\kappa\omega/c$, and this is the quantity measured in our experiments. The reflection coefficient is given by the usual formula

$$R = \left| \frac{(n-i\kappa) - 1}{(n-i\kappa) + 1} \right|^2 = \frac{(n-1)^2 + \kappa^2}{(n+1)^2 + \kappa^2}.$$

If a large static magnetic field is applied in the z -direction, the dielectric function is no longer isotropic. The results are summarized below, with the magnetic field dependence contained in the cyclotron frequency $\omega_c = |e|H/mc$.

$$\epsilon_{xx} = \epsilon_{yy} = 1 - \frac{\omega_p^2}{\omega^2 - \omega_c^2}$$

$$\epsilon_{xy} = -\epsilon_{yx} = -i \frac{\omega_p^2}{\omega^2 - \omega_c^2} \frac{\omega_c}{\omega}$$

$$\epsilon_{zz} = 1 - \frac{\omega_p^2}{\omega^2}$$

$$\epsilon_{xz} = \epsilon_{zx} = \epsilon_{yz} = \epsilon_{zy} = 0$$

If we consider a wave propagating along the magnetic field (Faraday geometry), the solutions to the determinantal equation are

$$\epsilon_{\pm} \equiv \epsilon_{xx} \pm i\epsilon_{xy} = 1 - \frac{\omega_p^2}{\omega(\omega \pm \omega_c)}.$$

For frequencies $\omega < \omega_c$, ϵ_+ is positive and propagation is possible for the circularly polarized wave whose electric vector rotates in the same direction as the electrons. These are called helicon waves. Figure 16(b) is a graph of ϵ_+ for all frequencies. Since $\epsilon_-(\omega) = \epsilon_+(-\omega)$, the negative frequencies correspond to the other propagating mode.

If there are two or more species of charge carriers present, their conductivities combine additively, so that other regions of positive ϵ are formed. Figure 16(c) is a graph of ϵ for a plasma with two species of negative carriers present. In the region between the two cyclotron resonance frequencies the function passes through zero, and at a slightly higher frequency it takes on the free space value of $\epsilon = 1$. The corresponding drop in the reflectivity due to this "dielectric anomaly" permits substantial penetration of the EM wave into the plasma. This technique has recently been exploited by Drew and Strom³² to measure relaxation times of electrons in bismuth.

If in addition to electrons there are positive free carriers, there is a corresponding cyclotron resonance, but in the other mode. The divergence at $\omega = 0$ remains, and its sign depends on the relative dominance of negative and positive carriers. If the charge densities of all the free carriers sums to zero, the plasma is said to be compensated. In this case, the divergence at $\omega = 0$ vanishes. The proof³³ employs the method of

partial fractions:

$$\begin{aligned}\epsilon_+ &= 1 - \sum_{i=1}^N \frac{\omega_{pi}^2}{\omega(\omega - \omega_{ci})} \\ &= 1 - \sum_{i=1}^N \frac{\omega_{pi}^2}{\omega_{ci}(\omega - \omega_{ci})} + \sum_{i=1}^N \frac{\omega_{pi}^2}{\omega \omega_{ci}}.\end{aligned}$$

Now $\omega_{pi}^2 \sim n_i e^2 / m_i$ and $\omega_{ci} \sim e / m_i$; therefore $\omega_{pi}^2 / \omega_{ci}$ is proportional to the charge density. Since $\sum n_i e = 0$ for a compensated plasma, the second sum vanishes and with it the divergence at $\omega = 0$.

Under these conditions, both circular polarizations of EM waves propagate at frequencies low compared with the cyclotron resonance frequencies of the carriers, as in Figure 16(d). The low-frequency limit was first explored theoretically by Alfvén,² and his results are restricted to frequencies much lower than the lowest cyclotron resonance frequency. However, each normal mode only resonates with charge carriers of one sign, so that the ϵ_+ mode in Figure 16(d), for instance, continues to propagate at frequencies at and above the cyclotron resonance frequency of the positive carriers.

Note that if the material is slightly uncompensated due to contamination or alloying, the last term in the equation for ϵ_+ above does not vanish and therefore a divergence at $\omega = 0$ remains. However, for the large magnetic fields and low impurity

levels encountered in this study, the effects of this divergence are confined to frequencies far below the far infrared.

B. Quantum Mechanical Treatment

We are now ready to present the results of a quantum mechanical treatment of EM wave propagation in a solid state plasma such as bismuth. We will restrict ourselves to the Faraday geometry, in which the wave propagates parallel to a large uniform static magnetic field. This theory has been developed in a series of papers³⁴⁻³⁸ and is based on the free electron approximation. Free electron wavefunctions are used, and the gross bandstructure effects are incorporated into an effective mass which depends on direction and energy. Initially, the electrons are assumed to be noninteracting. Then an energy dependent phenomenological relaxation time is introduced to approximate the effects of various possible scattering mechanisms. The theory omits nonlocal effects; it is assumed for instance that a dielectric tensor can be defined, relating the response of the plasma to the local value of the field alone. Also, the plasma is assumed to be infinite, with boundary effects omitted.

In their 1962 paper, Quinn and Rodriguez³⁴ treat the simplest case, that of an isotropic plasma in a uniform magnetic field with no relaxation time. Solving to first order in the EM field intensities and assuming that the response varies as

$\exp(i\omega t - i\mathbf{q} \cdot \mathbf{r})$, they obtain a gauge invariant result which for simplicity can be expressed in the gauge in which

$$\tilde{A}(\mathbf{q}, \omega) = (ic/\omega) \tilde{E} \exp(i\omega t - i\mathbf{q} \cdot \mathbf{z})$$

$$\varphi(\mathbf{q}, \omega) = 0.$$

Using the notation that prevails in the later papers, they obtain

$$\tilde{g}(\mathbf{q}, \omega) = \frac{\omega^2}{4\pi i \omega} \left\{ 1 + \frac{m}{N} \sum_{\nu\nu'} \frac{f(E_{\nu'}) - f(E_{\nu})}{E_{\nu'} - E_{\nu} - \hbar\omega} \langle \nu' | \tilde{V}(\mathbf{q}) | \nu \rangle \langle \nu' | \tilde{V}(\mathbf{q}) | \nu \rangle^* \right\}.$$

Here, $\tilde{V}(\mathbf{q})$ is the velocity operator with matrix elements given by

$$\langle n', k_y', k_z' | \tilde{V}(\mathbf{q}) | n, k_y, k_z \rangle = \delta(k_y', k_y) \delta(k_z', k_z + q) F_{n'n}(\mathbf{q}),$$

and $f(E)$ is the Fermi distribution function for the energy E .

The components of $F(\mathbf{q})$ are given by

$$\left[F_{n'n}(\mathbf{q}) \right]_x = i \left(\frac{\hbar\omega c}{2m} \right)^{\frac{1}{2}} X_{n'n}^{(-)}(\mathbf{q})$$

$$\left[F_{n'n}(\mathbf{q}) \right]_y = \left(\frac{\hbar\omega c}{2m} \right)^{\frac{1}{2}} X_{n'n}^{(+)}(\mathbf{q})$$

$$\left[F_{n'n}(\mathbf{q}) \right]_z = \frac{\hbar}{m} (k_z + \frac{1}{2}q) f_{n'n}(\mathbf{q}),$$

where

$$X_{n'n}^{(+)}(\mathbf{q}) = (n+1)^{\frac{1}{2}} f_{n', n+1}(\mathbf{q}) \pm n^{\frac{1}{2}} f_{n', n-1}(\mathbf{q})$$

and $f_{n'n}(\mathbf{q})$ is an overlap integral between normalized one

dimensional harmonic oscillator wavefunctions $u_n(x)$. For this simple case,

$$f_{n',n}(q) = \delta_{n',n}$$

and is independent of q .

The expression for the dielectric constant can then be written

$$\epsilon(q, \omega) = 1 - \frac{\omega_p^2}{\omega^2} \left\{ 1 + \frac{m}{N} \sum \frac{f[E_{n'}(k_z + q)] - f[E_n(k_z)]}{E_{n'}(k_z + q) - E_n(k_z) - \hbar\omega} F_{n',n}(q) F_{n',n}^*(q) \right\}$$

where the sum is over the Landau level indices n' and n , as well as k_y and k_z , and

$$E_n(k_z) = (n + \frac{1}{2})\hbar\omega_c + \hbar^2 k_z^2 / 2m.$$

Nonzero terms in the sum can only arise for $n' = n \pm 1$. Furthermore, at zero temperature, one state must be above the Fermi surface and the other below it. If the n th state is occupied and the $(n + 1)$ th state is empty then only $1/n$ of the electrons can make an upward transition. However, the magnitude of the transition probability is proportional to n , so that the same numerical result can be obtained by summing over all pairs of n and n' such that at least one of the two states is occupied, with an oscillator strength corresponding to transition between the lowest two oscillator levels. If in addition the kinetic energy of motion along the static magnetic field is ignored so that

$$E_{n'}(k_z + q) - E_n(k_z) = \pm \hbar\omega_c,$$

then the sum consists of N identical terms, and the classical results are recovered.

Returning to consideration of the energy denominator, we can see that the expression for $E_n(k_z)$ yields

$$E_{n+1}(k_z + q) - E_n(k_z) = \frac{\hbar\omega}{2} + (\hbar/m)(k_z + q/2)q.$$

The second term is the product of q and a term which is the average z-component of the initial and final state momenta.

This is then the $\mathbf{q} \cdot \mathbf{v}$ term encountered in classical expressions which takes into account the motion of the electrons along the direction of propagation of the EM wave.

C. Introduction of Relaxation Time

Tosima, Quinn and Lampert³⁶ and Greene, Lee, Quinn and Rodriguez³⁷ have successfully incorporated a relaxation time τ into the above theory in a gauge invariant fashion, and the result is seen to be somewhat more complicated than the simple prescription of everywhere replacing ω by $\bar{\omega} = \omega - i/\tau$. Instead one replaces

$$\frac{1}{E_{\nu'} - E_{\nu} - \hbar\omega}$$

by

$$\left[1 + \frac{\hbar\omega}{E_{\nu'} - E_{\nu} - \hbar(\omega - i/\tau)} \right] \left[\frac{1}{E_{\nu'} - E_{\nu}} \right].$$

The method of partial fractions yields the alternative expression

$$(1 + i\omega\tau)^{-1} \left[\frac{i\omega\tau}{E_{v'} - E_v - \hbar(\omega - i/\tau)} + \frac{1}{E_{v'} - E_v} \right].$$

For $\omega\tau \gg 1$, the second term is much smaller than the first. Also, the second term displays no resonances under the conditions which interest us. As long as $\Delta n = \pm 1$, only a large Doppler shift ($q \cdot v = -\omega_c$) would cause the second term to assume any significance. Otherwise, it assumes the value $(1 + i\omega\tau)^{-1} (\hbar\omega_c)^{-1}$ and can be ignored for our application, as will be discussed later.

As an example, the results for the classical case discussed earlier assume the form

$$\epsilon_{xx} = 1 - \frac{\omega_p^2}{\omega^2 - \omega_c^2} \cdot \frac{\bar{\omega}}{\omega}$$

$$\epsilon_{xy} = -i \frac{\omega_p^2}{\bar{\omega}^2 - \omega^2}$$

$$\epsilon_{\pm} = \epsilon_{xx} \mp i\epsilon_{xy} = 1 - \frac{\omega_p^2}{\omega(\bar{\omega} \mp \omega_c)}$$

D. Introduction of Anisotropy

The above theory has been generalized by Quinn³⁵ to describe an anisotropic plasma, by introducing an inverse mass tensor

$$\alpha = \begin{pmatrix} \alpha_1 & 0 & 0 \\ 0 & \alpha_2 & \alpha_4 \\ 0 & \alpha_4 & \alpha_3 \end{pmatrix}$$

While further generalization does not lead to any new physical effects, this theory as it stands is inadequate for our purposes, since some of the matrix elements are assumed to be zero. If we assume a general inverse mass tensor

$$\alpha \approx \begin{pmatrix} \alpha_{11} & \alpha_{12} & \alpha_{13} \\ \alpha_{12} & \alpha_{22} & \alpha_{23} \\ \alpha_{13} & \alpha_{23} & \alpha_{33} \end{pmatrix}$$

then the cyclotron frequency becomes

$$\omega_c^* = (\alpha_{11}\alpha_{22} - \alpha_{12}^2)^{\frac{1}{2}}\omega_c$$

and the components of $F_{n'n}(q)$ become (assuming $q_x = q_y = 0$)

$$[F_{n'n}(q)]_x = \alpha_{12}\bar{X}^{(+)} + i(\alpha_{11}\alpha_{22} - \alpha_{12}^2)^{\frac{1}{2}}\bar{X}^{(-)} + \frac{\hbar}{2m}\alpha_{13}q f_{n'n}(q)$$

$$[F_{n'n}(q)]_y = \alpha_{22}\bar{X}^{(+)} + \frac{\hbar}{2m}\alpha_{23}q f_{n'n}(q)$$

$$[F_{n'n}(q)]_z = \alpha_{23}\bar{X}^{(+)} + i(\alpha_{13}\alpha_{22} - \alpha_{12}\alpha_{23})(\alpha_{11}\alpha_{22} - \alpha_{12}^2)^{-\frac{1}{2}}\bar{X}^{(-)} + \frac{\hbar}{2m}(2\alpha^*k_z + \alpha_{33}q)f_{n'n}(q)$$

where

$$\alpha^* = (\alpha_{22})^{-1}[(\alpha_{22}\alpha_{33} - \alpha_{23}^2) - (\alpha_{13}\alpha_{22} - \alpha_{12}\alpha_{23})^2(\alpha_{11}\alpha_{22} - \alpha_{12}^2)^{-1}]$$

and

$$\bar{X}^{(\pm)} = (\alpha_{22})^{-\frac{1}{2}}\left(\frac{\hbar\omega_c^*}{2m}\right)^{\frac{1}{2}} X_{n'n}^{(\pm)}(q).$$

The function $f_{n'n}(q)$ assumes a somewhat more complicated form as well.

E. Approximations Applicable to Bismuth

In the process of presenting the foregoing theory, several assumptions, both explicit and implicit, were made. We will now see if they are appropriate for bismuth.

The smallest component of the effective mass at the Fermi surface in bismuth is $0.007m$. This corresponds to a Fermi velocity of $0.004c$. In contrast $\omega/q = 0.1c$ for a typical index of refraction of 10. Under these circumstances, $\mathbf{q} \cdot \mathbf{v} \ll \omega$, and non-local effects such as Doppler-shifted cyclotron resonance are small.

For our case of bismuth at liquid helium temperatures and EM frequencies in the far infrared, electron mean free paths are quite large so that typically $\omega\tau \gg 1$. For example, Drew and Strom³² estimated that $\omega\tau = 128$ for dielectric anomaly absorption near 30 cm^{-1} . Since the frequency range of interest is $10\text{-}100 \text{ cm}^{-1}$ and since τ^{-1} is expected to vary as ω^2 or ω^3 at most, it can safely be assumed that $\omega\tau \gg 1$ for our application.

As for the general validity of the foregoing theory, Zyryanov and Okulov³⁹ found that the results of Tosima, Quinn and Lampert³⁶ gave incorrect results for $\omega \ll \tau^{-1} \ll \omega_c$, and then proceeded to set forth the conditions for their validity. Aside from assumptions about the nature of the scattering process, which are necessary for any theory which lumps the effects of collisions into a single parameter, Zyryanov finds that for

$\omega_c \tau \gg 1$ and $\omega \lesssim \omega_c$, it is sufficient (1) that the Larmor radius be less than the wavelength of the EM wave ($qr_L \ll 1$) and (2) that $q_z v_z / (\omega - i/\tau)$ be small. Recalling that $r_L = v_F / \omega_c$, we see that these conditions are easily satisfied for our application.

The previous section closed with an expression for $F_{\tilde{n}'n}(q)$ which would be considerably simpler if the second term $\alpha \cdot q f_{\tilde{n}'n}(q)$ were much smaller than the first and could therefore be omitted in calculations. From the definition of $f_{\tilde{n}'n}(q)$ as an overlap integral, it is clearly no greater than unity. It remains only to show that

$$\left(\frac{\hbar^2 q^2}{2m} \cdot \frac{1}{\hbar \omega_c} \right)^{\frac{1}{2}} \ll 1,$$

which is easily demonstrated to be the case for bismuth at far infrared frequencies and ten kilogauss fields. Hence the second term may indeed be omitted from the x- and y-components of $F_{\tilde{n}'n}(q)$.

Under these same conditions, $f_{\tilde{n}'n}(q)$ is equal to $\delta_{\tilde{n}'n}$ plus small terms of order

$$\frac{\frac{\hbar^2 q^2}{2m} \cdot \frac{1}{\hbar \omega_c}}{\frac{|n' - n|}{2}}.$$

Appendix II contains the results of this section as applied to the Faraday geometry in the bisectrix direction. Also, it should be noted that ϵ_{ℓ} is anisotropic, and that the values used in our calculations were those suggested by Takano and

Kawamura:²⁷

$$\alpha_l = \begin{pmatrix} 100 & 0 & 0 \\ 0 & 100 & 0 \\ 0 & 0 & 60 \end{pmatrix}$$

SECTION VI

EXPERIMENTAL RESULTS

A. Frequency Dependence of Relaxation Time

1. Lower Frequencies

The foregoing theory will now serve as a framework for analyzing the results obtained from transmission measurements at far infrared frequencies and liquid helium temperatures. Figure 17 contains the results of measurements on two samples of different thickness with a 100 kG magnetic field applied along the direction of propagation (Faraday geometry). The quantity being graphed is the experimental power attenuation coefficient α' discussed in Section III. Recall that

$$\alpha' = - \frac{1}{x_2 - x_1} \ln (I_2/I_1),$$

where I_1 and I_2 are the intensities measured passing through samples of thickness x_1 and x_2 respectively. Apart from some structure which will be discussed later, $\alpha'(\omega)$ displays a typical trend of increased absorption at higher frequencies. The negative values of α' at the lower frequencies in this figure are not surprising when one remembers that the previously discussed problems associated with measuring absolute intensities cause α' to be off by a frequency independent additive constant.

In the bisectrix direction, the low-field cyclotron masses of the electrons (at the Fermi surface) are $m_A^* = 0.008m$ and

$m_B^* = m_c^* = 0.016m$ and that of the holes is $m^* = 0.21m$. Since

$$\omega_c = (0.0933 \text{ cm}^{-1}/\text{kG}) \cdot H \cdot \left(\frac{m}{m^*}\right),$$

the electron cyclotron resonances at 100 kG are well above the region of transmission, whereas the hole cyclotron frequency is in the middle of it. Consequently, the ϵ_- mode, which interacts strongly with the holes, would be strongly absorbed over this entire frequency region, whereas the ϵ_+ mode would propagate easily at these frequencies which are much lower than either of the electron cyclotron frequencies.

To test this interpretation, we wished to calculate the power attenuation coefficient from the foregoing theory. However, the theory contains a parameter about which relatively little is known. This is the relaxation time in bismuth and its frequency dependence at far infrared frequencies. Edel'man and Cheremisin⁴⁰ have analyzed Azbel-Kaner electron cyclotron resonance linewidths between 0.33 cm^{-1} and 2.5 cm^{-1} using microwave techniques, and found that the frequency dependent part of the scattering rate varied as the 2.5 ± 0.2 power of ω at those frequencies. Using an HCN laser at 30 cm^{-1} , Drew and Strom³² analyzed a dielectric anomaly linewidth to obtain $\omega\tau = 128$ at this frequency. By comparing this result with their microwave results at lower frequencies, they found that the scattering rate increased faster than quadratically with frequency.

These were all reflection measurements requiring analysis of line shapes to determine the relaxation times, and were conducted at relatively low magnetic fields. The only work to date which was attempted to measure relaxation times at higher fields and far infrared frequencies is that of Carter and Picard,⁹ where microwave transmission at 20 cm^{-1} was measured through a thick sample at fields up to 60 kG. They mention a value of $\omega\tau \approx 400$.

We therefore allowed τ^{-1} to assume the form

$$\tau^{-1} = \tau_0^{-1} + A\omega^n,$$

where the first term is the sum of a residual relaxation rate and a temperature dependent contribution appropriate for liquid helium temperatures. In so doing, we chose to neglect any variation of τ among the different sets of carriers. As a result, τ takes on the character of an average relaxation time.

Since the frequency dependent term was expected to dominate at far infrared frequencies, the constant term was set equal to zero. For convenience the frequency dependent term was rewritten as

$$\tau^{-1} = \left(\frac{\omega}{\omega_0}\right)^n \frac{\omega_0}{(\omega\tau)_{\omega_0}},$$

where ω_0 is the angular frequency corresponding to our reference frequency of 30 cm^{-1} . The two parameters actually used, then,

were the exponent n and the value of $\omega\tau$ at 30 cm^{-1} .

Calculations were carried out for various values of these two parameters, and it was seen that only one of the two normal modes of electromagnetic wave propagation would correspond to what was measured. The absorption coefficient in the ϵ_- mode would be substantially higher over the entire range of $10\text{-}100 \text{ cm}^{-1}$ because the hole cyclotron resonance was in this frequency interval. The absorption would peak around the cyclotron frequency, but its effects would be felt over the entire range. In contrast, the ϵ_+ mode would be entirely unaffected by the hole cyclotron resonance, even right at the resonance frequency, for its electric vector would be rotating in the opposite direction.

Figure 18 shows the results of fitting the ϵ_+ mode to the 100 kG results of an earlier figure, assuming first an ω^2 and then an $\omega^{3/2}$ dependence for the relaxation rate. In the first case, a value of $\omega\tau = 1000$ at 30 cm^{-1} gave the best agreement over the entire frequency range from 10 to 75 cm^{-1} . However, the $\omega^{3/2}$ fit with $\omega\tau = 700$ gave a superior fit, although such a weak frequency dependence was not expected. The two fits also required slightly different values of the third parameter always present, the additive uncertainty in the measured α' . Similar results were obtained for the 90 kG data, and this is shown in Figure 19.

A third set of data at 30 kG yielded the results in

Figure 20. In spite of the uncertainties in the measurements and the narrow frequency range available ($15 - 60 \text{ cm}^{-1}$), it appears that the $\omega^{3/2}$ dependence seen at higher fields may no longer hold at 30 kG.

These results suggest that a different frequency dependence prevails at higher fields, since our $\omega^{3/2}$ result differs from the $\omega^{5/2}$ result previously reported.

There are no technical difficulties preventing measurements at any field above 30 kG, and additional experiments are called for to explore the region between 30 and 90 kG as well as to confirm the 30, 90 and 100 kG results. This is especially so because the two samples used to obtain these three measurements were prepared in different laboratories, although grown from similar starting material under similar conditions, and were measured on different days. However, these factors are not believed to invalidate the conclusions we have drawn regarding the $\omega^{3/2}$ vs. $\omega^{5/2}$ frequency dependence. Indeed, further support for an $\omega^{3/2}$ dependence over an $\omega^{5/2}$ dependence has been provided by measurements at higher frequencies as discussed in the next section.

2. Higher frequencies

By using our Michelson interferometer with a 0.00025-inch mylar beamsplitter to improve the throughput at higher frequencies, we were able to measure substantial transmission through our

thinner samples in the frequency range from 200 to 350 cm^{-1} . Figure 21 shows a series of transmission spectra taken at 90 kG through three samples of varying thickness. The vertical scales are different for the three spectra. However, it can be seen that the spectrum is divided into two pass bands separated by a stop band, and that in general the higher frequencies are much more rapidly attenuated than lower ones.

Figure 22 is a plot of the experimental attenuation coefficient at 100 kG, together with a calculated theoretical fit. It can be seen that the pass band at higher frequencies is due to radiation propagating in the ϵ_- mode. The frequency dependence of $\omega^{1.4}$ was assumed because it had given the best fit at lower frequencies, slightly better than the $\omega^{3/2}$ fits shown earlier, and because it gave the best fit at the higher frequencies as well. Figure 23 is a similar plot for the 80 kG data. A dashed curve in that same figure demonstrates the sensitivity of the theoretical results to the frequency dependence of the relaxation rate. Even a change from 1.4 to 1.3 in the exponent can be seen to change the high frequency results substantially. In fact, the results for an ω^2 dependence and $\omega_T = 1000$ at 30 cm^{-1} gives an attenuation which never drops below a value of 50 cm^{-1} , a result very much at variance with the measured values.

A second look at Figures 22 and 23 show the shape of the high frequency attenuation is not adequately reproduced in our calculations. However, we have been restricting ourselves to one relaxation time for all carriers. This can affect the calculated attenuation coefficient, since the relative influence of the various individual relaxation times varies with frequency. Hartman⁴¹ has measured the zero field relaxation time tensor for the electrons and holes, and in no case do they vary by more than a factor of five; a factor of two or three is typical when the carriers are executing helical motion.

It therefore seemed reasonable to allow the relaxation times to vary over a limited range. The dotted curve in Figure 23 was obtained by retaining the $\omega^{1.4}$ dependence, but setting ω_T at 30 cm^{-1} equal to 600 for the holes and equal to 1000 for both groups of electrons. The substantially better fit suggests that the data can in principle be duplicated by suitable choices of the three relaxation times, all with a common frequency dependence. It is reassuring that this new set of relaxation times continues to yield good values of the experimental attenuation at 30, 90 and 100 kG for the lower frequency band discussed in the previous section. Therefore, it appears that the higher frequencies are the more sensitive to our choice of a set of relaxation times, and could be used to determine the three relaxation times more precisely. However, measurements

which are sensitive to the relaxation time of just one set of carriers are much easier to analyze. Cyclotron resonance linewidth measurements such as those of Edel'man and Cheremisin⁴⁰ meet this requirement.

Additional measurements were taken with two samples cut from one single crystal, and they gave the same high frequency attenuation as measurements taken where one sample was the one grown in another laboratory and used in the low frequency measurements. Thus, the general absorption does not seem to be sensitive to the conditions under which the samples are prepared.

It can therefore be concluded that the high frequency results to confirm the previously drawn conclusions regarding the relaxation time and its weak frequency dependence. Furthermore, these measurements together cover a factor of 25 in frequency, or over four and one-half octaves, making them quite sensitive to the frequency dependence. Therefore, the relaxation rate is estimated to vary as the 1.4 ± 0.1 power of the frequency.

B. A Magnetoplasma Resonance

We now move on to the details of the observed transmission spectra. In particular, a strong absorption ($\alpha > 10 \text{ cm}^{-1}$) was seen in bisectrix spectra, and it was found to move in a non-linear fashion as the field was varied, as shown in Figure 24. It occurred at frequencies intermediate between the hole cyclotron frequency and that of the heavier electrons has its origin in

the component of the dielectric constant along the magnetic field. The results of Appendix II indicate that ϵ_{zz} appears in the denominator in the calculation of the complex index of refraction N . Consequently, if ϵ_{zz} should vanish, a singularity results and absorption then occurs. Figure 25 is a graph of ϵ_{zz} at 30 kG, and it can be seen to cross the horizontal axis at 59 cm^{-1} , and again near 240 cm^{-1} and 300 cm^{-1} . Figure 26 is a graph of the attenuation coefficients of the two propagating modes. Resonant absorption is produced in both modes when ϵ_{zz} vanishes, and this is especially clear for the resonance near 240 cm^{-1} . (The resonance near 300 cm^{-1} is extremely weak.)

The vanishing longitudinal component of the dielectric tensor indicates that near the resonance, the oscillating electric field has a very small transverse component and a very large longitudinal component. This would cause the heavier electrons and holes to move in orbits which are inclined to the magnetic field, and they would move in their new orbits at the resonance frequency rather than at their respective cyclotron frequencies. The lighter electrons are affected very little.

The solid lines in Figure 24 show the frequency dependence of the resonance as calculated from theory. At lower fields, there are two lines. The upper line passes through frequencies predicted from the empirical $H^{2/3}$ dependence of the cyclotron frequencies of the electrons in the extreme quantum limit, with

no attempt being made to optimize the fit by varying any parameters. The other line shown at lower fields is based on the low field cyclotron mass of the heavier electrons, and should be used below 25 kG where the heavier electrons pass out of the extreme quantum limit. It is difficult to obtain data on this resonance at low fields, for reasons which will become apparent in the next section. However, the one point at 27 kG falls on the lower line, and will lend support to a later contention that the onset of the EQL for the heavier electrons is closer to 27 kG than to the value of 25 kG predicted from the parameters used in our calculations.

C. Quantum Effects at Low Fields

When Landau levels pass through the Fermi surface, they produce oscillations in many of the electrical and magnetic properties of the metal being studied. Examples are de Haas-van Alfen oscillations in the magnetic susceptibility and Shubnikov-de Haas oscillations in the D.C. resistivity. These effects are accentuated as the last levels pass through the Fermi surface and the electrons pass into the extreme quantum limit.

A similar type of behavior has been seen in the transmission spectra at lower fields, as summarized in Figure 27. The quantity being plotted is the integrated intensity

$$I(H) = \int I(H, \omega) d\omega ,$$

which is a function of the reflectivity and absorptivity as well as the source spectrum. Nevertheless, it indicates the general trend of improved transmission at higher fields, as well as rapid increases in transmission at fields near 14 and 27 kG. The 27 kG field corresponds roughly to onset of the extreme quantum limit for the heavy electrons as the 1^- and 0^+ levels emerge through the Fermi surface. The increase at 14 kG may be due either to the 2^- and 1^+ levels of the heavy electrons emerging, or to the light electrons passing into the EQL.

If we look at the actual measured spectra as plotted in Figure 28, we see that the rapidly decreasing transmission near 27 kG is associated with strong attenuation of the higher frequencies as well as decreased transmission at all frequencies. Generally speaking, higher fields extend transmission to higher frequencies because the cyclotron resonances are moving to higher frequencies. Therefore, one would expect a substantial change at onset of the EQL, because the cyclotron frequency of the electrons involved changes rapidly there, as previously discussed. Figure 29 is a graph of the transmittance at 27 kG assuming the corresponding EQL average cyclotron frequency of 177 cm^{-1} , together with the transmittance at 25.5 kG assuming a cyclotron frequency of 136 cm^{-1} based on the low field cyclotron mass, since these electrons appear to be passing out of the EQL at

about 27 kG. A large change can be attributed to this effect, as much as a 40% reduction at 45 cm^{-1} . However, the actual spectra show a 100% reduction at that frequency.

There is another known effect which we have not yet taken into account, the field-dependent variation in the relaxation time as each Landau level crosses the Fermi surface, which is responsible for the Shubnikov-de Haas effect in D.C. resistivity measurements. Oscillations in which the average relaxation time varies over a factor of two have been measured along the trigonal axis.⁴² If we assume that the relaxation time of the heavy electrons decreases by a factor of 3.5 between 27 kG and 25.5 kG, we obtain the dashed line in Figure 30(a). In Figure 30(b) the ratio of the observed intensities at 25.5 kG and 27 kG has been plotted together with the ratio of the calculated transmittances, and they agree fairly well. Together with the oscillatory behavior present in Figure 27, this result confirms that the relaxation time of the heavier electrons is undergoing large Shubnikov-de Haas oscillations at these fields.

D: Temperature Dependence of Relaxation Time

Many measurements have been made of the temperature dependence of the relaxation rate in bismuth.⁴³ Experiments at microwave frequencies have typically measured Alfvén wave attenuation and cyclotron resonance linewidths. Drew and Strom³² studied the dielectric anomaly lineshape in a 4 kG field, using

an HCN laser operating at 30 cm^{-1} . These measurements, as well as D.C. measurements, have consistently found a quadratic temperature dependence. Candidates for the responsible relaxation mechanism include electron-electron scattering and intervalley scattering by phonons, but compelling evidence for choosing either of them is not available.

We measured the temperature dependence of the low frequency transmission through a thick sample between 2°K and 12°K at 48 kG and found the fractional change with temperature to be essentially frequency independent. Therefore, only the integrated intensity

$$I(T) = \int I(T, \omega) d\omega$$

was recorded at each temperature. Theoretical calculations showed that adding a constant term to the relaxation rate produces a fairly uniform increase in the experimental attenuation coefficient over the frequency range studied ($3 \text{ to } 30 \text{ cm}^{-1}$). Furthermore, the increase is proportional to the magnitude of the constant term added, (except at the lowest frequencies where small attenuation and the resulting multiple reflections produce nonlinearities). It was therefore possible to extract the temperature dependent part of the experimental attenuation coefficient from measurements. Its temperature dependence was then determined by plotting the logarithm of the temperature-dependent part of α' vs. the logarithm of the temperature. To

do this, it was necessary to assume a value for the transmission at absolute zero, an experimentally inaccessible temperature. The data above 4°K were relatively insensitive to the value chosen, but suggested a zero-temperature transmittance several percent higher than at 2°K. A choice 6% higher brought the low temperature data into collinearity with the data above 5°K, and the results are plotted in Figure 31. From these measurements, the relaxation rate is judged to vary as the 2.02 ± 0.04 power of the temperature. The coefficient indicating the strength of the temperature dependent term is

$$B = (3.0 \pm 0.3) \times 10^{-7} \text{ sec}^{-1} \cdot \text{°K}^{-2},$$

and was obtained using a constant of proportionality

$$\frac{d\alpha'}{d(\tau^{-1})} = 4 \times 10^{-10} \text{ cm}^{-1}/\text{sec}^{-1}$$

relating the changes in α' to changes in the relaxation rate.

The value of B thus obtained is seven times smaller than the values obtained by Drew and Stron,³² and is half as large as the values given by Chen, Kirschenbaum and Kao.⁴³ (Also, we did not see the "break" at 5°K reported by Chen, et al.⁴³) However, our measurements were made with a 48 kG field applied, with additional measurements at 36 kG and 30 kG showing very little change in the strength of the T^2 term, while the values reported by others were obtained with applied fields of 10 kG or less. In addition, there is a deviation from a quadratic power dependence as the field approaches the EQL threshold from above; between 36 kG and 30 kG, the exponent changes from

2.0 to 2.3. This is presumably because the 1- and 0+ levels of the heavy electrons are only a few $k_B T$ from the Fermi surface and the additional scattering does not vary quadratically with temperature.

In conclusion, the temperature dependent portion of the scattering rate is substantially reduced as bismuth passes into the EQL, but it retains the same quadratic dependence at fields well above the EQL threshold as has been reported for low fields.

E. A Harmonic Series of Cyclotron-like Resonances

Besides the magnetoplasma resonance discussed earlier, other structure was seen in our spectra. In particular, a series of three evenly spaced lines were consistently observed. Figure 32 shows bisectric transmission spectra through two samples of different thickness at 45 kG. Besides the broad absorption at 75 cm^{-1} due to the magnetoplasma resonance, there are dips at approximately 20, 40 and 60 cm^{-1} .

These three lines were found to move linearly with field, as demonstrated in Figure 33. Also, their strengths did not exhibit any temperature dependence. Furthermore, the effective mass associated with the lowest line is $0.21m$, which is the cyclotron mass of the holes in this direction. However, this absorption is not the cyclotron resonance, since only radiation of the cyclotron-resonance-inactive polarization is seen at these lower frequencies. Nor can it be the phonon-assisted

2

cyclotron resonance seen in indium antimonide,⁴⁴ since the creation of an optical phonon would produce absorption at frequencies given by $n\omega_c + \omega_o$, where ω_o would be 75 cm^{-1} or more for optical phonons in bismuth.⁴⁵ Similarly, plasmon-assisted cyclotron resonance as seen in indium antimonide⁴⁶ would shift the absorption to frequencies near the plasma frequency in bismuth ($\sim 200 \text{ cm}^{-1}$). Only some unknown low-lying excitation ($\omega < 1 \text{ cm}^{-1}$) could assist the observed transitions without producing an observable shift. It should also be noted that none of the above mechanisms would predict a stronger third harmonic.

There is, however, another possibility. Our calculations are based on an ellipsoidal Fermi surface; even the two band model gives constant energy surfaces which are ellipsoidal. Under these circumstances, a transformation can be found for which the hole Fermi surface is spherical and the cyclotron orbits in momentum space are circular, thereby simplifying the problem to that of simple harmonic motion. The motion in velocity space can then be described by uniform motion around a circle in the complex plane at the cyclotron frequency with a normalized velocity given by

$$v(t) = \exp(i\omega_c t).$$

This motion would couple only to radiation of one circular polarization with one angular frequency ω_c . In quantum mechanical terms, the selection rule for transitions between

Landau levels would be the well-known $\Delta n = \pm 1$. Wolff²⁵ confirms that the two band model with its ellipsoidal Fermi surface does indeed still allow only $\Delta n = \pm 1$ transitions.

However, the two band model does not take into account the influence of other bands, and they can cause the Fermi surface to be slightly non-ellipsoidal.⁴⁷ After transformation, the hole Fermi surface is then a slightly distorted sphere and the cyclotron motion is no longer simple harmonic in character. However, it is still periodic and can be Fourier analyzed:

$$v(t) = \sum a_n \exp(in\omega_c t)$$

where the sum is over all integral values of n , both positive and negative. We would expect $a_1 \approx 1$, with the other a_n being much smaller.

Figure 34 shows how small admixtures of some of the a_n affect the motion of the electrons. The harmonics given by $n = -1, -2, -3, \dots$ would give rise to absorption at frequencies $n\omega_c$ in the ϵ_+ mode, the mode which is propagating with little attenuation through bismuth at these lower frequencies. The $n = -1$ mode would produce a resonance at ω_c , but it should not be confused with the fundamental cyclotron resonance ($n = +1$) which occurs in the ϵ_- mode and is so strong that it prevents measurable transmission in that mode at all far infrared frequencies below 200 cm^{-1} .

McClure⁴⁸ has carried out a more rigorous and complete analysis, and concludes that the net effect of the anharmonic motion of the holes is the same as would be obtained by adding a few holes of mass $m_c/2$, $m_c/3$,... and electrons of mass m_c , $m_c/2$, $m_c/3$,... . A rough calculation suggests that number densities on the order of 0.001% to 0.01% the hole density would give the sort of absorption strengths we observed.

McClure also considers the effects of symmetry, and finds that the mirror plane defined by the bisectrix and trigonal axis does not result in any missing harmonics for magnetic fields applied in this plane. This is consistent with our observation of all of the first three resonances of the series.

Experimentally, we observe these resonances to have a full width at half-maximum (FWHM) of about 1.5 cm^{-1} and a peak attenuation on the order of 0.5 to 10 cm^{-1} . This is to be compared with the true cyclotron resonance, for which the calculated attenuation exceeds $10,000 \text{ cm}^{-1}$. The strength of the three lines increases with the magnetic field, and this increase correlates well with the calculated hole density at each field. For example, the $n = -1$ resonance for bisectrix sample no. 3 exhibits a constant peak attenuation of 0.45 cm^{-1} per $10^{17} \text{ holes/cm}^3$ between 35 kG and 100 kG while the hole density itself is increasing by more than a factor of two.

The line shape has not been carefully analyzed yet, but

the $q \cdot v$ term which was omitted in the general calculations produces a 1% Doppler broadening which can not be neglected here. Also remaining to be analyzed are the variations in the relative strength of the lines in two of our samples. For example, the $n = -2$ resonance in "Bisectrix no. 3" was only about 20% as strong as the $n = -1$ resonance, whereas they were of comparable magnitude in "Bisectrix no. 5." This is probably related to the known misalignment of one of the samples (no. 3) by almost 5° toward the nearby pseudotrigonal axis. In all samples, the $n = -3$ resonance was two to three times stronger than the $n = -1$ resonance. Also, no higher resonances were ever seen. However, the circumstances were not favorable for their observation, so that we can only say that the $n = -4$ and $n = -5$ resonances have peak attenuations of less than 1 cm^{-1} . Theoretical calculations are now needed to determine if nearby bands do indeed induce cyclotron harmonics of the observed strengths.

F. Other Results

1. Faraday Geometry: Binary and Trigonal

A survey was made of the far infrared transmittance in the other two crystal directions for the Faraday geometry. The results are described below in terms of the low-field cyclotron masses for these two directions, as listed in Table 4.

Table 4
Cyclotron Masses

Carrier	Bisectrix	Binary	Trigonal
Electrons (A)	0.008	0.12	0.06
Electrons (B,C)	0.016	0.009	0.06
Holes	0.21	0.21	0.065

Unlike the bisectrix direction, the binary direction has both heavy electrons and heavy holes. Consequently, both propagating modes should be strongly absorbed at the lower frequencies. Some transmission was in fact seen at lower frequencies, but it was not reproducible and was therefore believed to be due to light leaking around the samples. At higher frequencies, the binary samples produced spectra very similar in every way to the high-frequency band seen in the bisectrix spectra, and again it would be the ϵ_- mode that is being observed. The similarity is due to the hole mass being the same in the two directions.

In the trigonal direction, both the electron and hole masses are small and nearly equal. Therefore, both modes would transmit well at the lower frequencies, and there would be no high frequency transmission. This is what was observed.

In practice, the onset of the extreme quantum limit for one

or more sets of carriers determined the lowest field at which it was practical to do transmission experiments in bismuth. In the binary direction, this occurred at 15 kG, corresponding to the lighter electrons passing into the EQL. In the trigonal direction, measurements were feasible above 40 kG, but transmission was seen to improve greatly above 90 kG, where the holes pass into the EQL.

2. Voigt Geometry

A few measurements were made of the transmission characteristics of bismuth in the Voigt geometry, where waves propagate across the large static magnetic field instead of along it. These measurements were limited to fields below 50 kG and to frequencies below 100 cm^{-1} . No measurable transmission was observed through the 2.65 mm thick trigonal sample. Measurements of the 0.80 mm thick binary sample were inconclusive, but they suggested that some transmission was occurring.

The 1.30 mm thick bisectrix sample transmitted about 10% as much signal as in the Faraday geometry, but this was sufficient to obtain good spectra. When the sample was oriented with its trigonal axis parallel to the magnetic field, a strong absorption ($\alpha \approx 20 \text{ cm}^{-1}$ and $\text{FWHM} \approx 1 \text{ cm}^{-1}$) was observed. It appeared to move quadratically with the applied field, with a center frequency of 19 cm^{-1} at 48 kG. However, the absorption

could only be studied between 40 kG and 48 kG, so the field dependence is still open to question. The absorption itself was not identified with any mechanism, but additional measurements did reveal that the transmitted radiation in this geometry was plane polarized, with the electric field vector perpendicular to the applied magnetic field. Also, the absorption was not present in bisectrix transmission spectra when the sample was rotated 90° so that the trigonal axis was perpendicular to the applied field.

3. More Resonances

We conclude by mentioning several other absorption lines which appeared in bisectrix spectra in the Faraday geometry, all of which moved linearly with the applied field. Since all the electron bands are nonparabolic and move in a nonlinear way as the magnetic field is increased, these lines are not associated with them. One such line may be a hole combination resonance ($\Delta n = +1$, $\Delta s = -1$). The line was present in our 9.70 mm sample, but was absent altogether in the 3.10 mm sample provided by H. D. Drew. It can be seen as a second line present only in the bottom spectrum in Figure 21.

On the other hand, the 3.10 mm sample had a multitude of lines which were absent in other samples. Figure 35 is a plot of the quantity

$$- \frac{1}{x} \ln (I_2/I_1),$$

where I_2 is the intensity at 90 kG and I_1 is the intensity of 100 kG. (In such a quantity, absorption lines in the denominator spectrum appear as inverted absorption peaks.) The largest absorption near 40 cm^{-1} is due to the $n = -1$ term in the cyclotronlike harmonic series discussed earlier, and the weaker absorption at 80 cm^{-1} is due to the $n = -2$ term of the series. But the remaining three absorptions at 0.40, 0.62, and 1.35 times the hole cyclotron frequency could not be accounted for theoretically.

SECTION VII

SUMMARY

In summary, we have carried out a comprehensive investigation of the far infrared features of bismuth when subjected to magnetic fields above 10 kG. In retrospect, the success of this venture stemmed from the electronic relaxation rate being much lower at high fields. Matsumoto, Sakai and Mase⁴⁹ reported in 1970 that their giant quantum attenuation of sound waves in bismuth yielded a relaxation rate 70 times lower than previous de Haas-van Alfen results, an extraordinary discrepancy for which they had no explanation at that time. We have found that these lower relaxation rates result from the application of large magnetic fields, and that much of the reduction takes place rapidly as one or more sets of carriers pass into the extreme quantum limit. Indeed, the onset of measurable transmission through thick samples usually coincides with onset of the EQL.

Using our generalized equations for electromagnetic waves in an anisotropic metal like bismuth, information about the frequency and temperature dependence of the relaxation rate was obtained. The quadratic temperature dependence previously reported for lower fields was found to hold at these higher fields as well, but the frequency dependence proved to be unexpectedly weak over the studied frequency range from 20 cm^{-1}

to 300 cm^{-1} . These results should shed new light on the underlying carrier scattering mechanisms in bismuth at these low temperatures.

Many features were observed in the transmission spectra. One strong line was identified as a magnetoplasma resonance. Three other lines are tentatively identified as true harmonics of the hole cyclotron resonance, but theoretical calculations based on what is known of the bandstructure of bismuth would help to confirm this. Still other lines remain unidentified at this time. All in all, because of the thick samples employed, far infrared transmission spectroscopy is proving to be a sensitive probe for studying the electronic properties of the solid state plasma.

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APPENDIX I

The following is a list of some of the symbols used in this work and their meanings:

ω = angular frequency

$\bar{\omega} = \omega - i/\tau$

$\nu = 2\pi c\omega$ = wavenumber (reciprocal wavelength in vacuum)

$\tilde{k}$ = crystal wavevector

$\tilde{p} = \hbar k$ = crystal momentum

$\tilde{q}$ = EM wavevector

c = speed of light

m = mass of electron (bare)

e = charge of electron

$\hbar$ = Planck's constant

$\tilde{H}$ = static magnetic field

$\tilde{A}, \varphi$ = EM potentials (vector and scalar)

E_F = Fermi energy

E_G = gap energy

n = Landau level index; also index of refraction

s = spin index

$\omega_c = |e|\hbar/mc$ = cyclotron resonance frequency

$\omega_p = (4\pi ne^2/m)^{1/2}$ = plasma frequency

$\tilde{\sigma}$ = complex conductivity tensor

$\tilde{\epsilon}$ = complex dielectric constant tensor

$N = n - i\kappa$ = complex index refraction

α = (power) attenuation coefficient; also inverse mass tensor

N = number of charge carriers in sample

m_c = cyclotron mass

m_H = longitudinal mass

$f(E)$ = Fermi distribution function

μ_B = Bohr magneton

Numbered subscripts refer to crystal directions (1 = binary, 2 = bisectrix, 3 = trigonal) and lettered ones (x,y,z) refer to orientation with respect to an applied static magnetic field, the direction of the field always determining the z-axis. For example, for wave propagation in the Faraday geometry ($q \parallel H$) along a bisectrix axis, the wavevector q and the field H are in the z-direction, which is also the 2-direction of the crystal. The cyclic pairing (1 = y, 2 = z, 3 = x) will be used for analysis of this case. The α_{ij} 's of Section V are an exception; numbered subscripts are used instead of lettered ones.

APPENDIX II

APPLICATION OF SECTION V TO BISECTRIX

DIRECTION IN BISMUTH

In this section, the general formulas presented in Section V for calculating the dielectric tensor are applied to the bisectrix direction in the Faraday geometry.

We shall define two quantities:

$$\sigma_{xx}^{(i)} = \frac{\omega_{pi}^2}{4\pi i} \cdot \frac{\omega}{\bar{\omega}^2 - \omega_{ci}^2}$$

$$\sigma_{xy}^{(i)} = \frac{\omega_{pi}^2}{4\pi} \cdot \frac{\omega_{ci}}{\bar{\omega}^2 - \omega_{ci}^2}$$

Here, $\bar{\omega} = \omega - i/\tau$. A superscript will be used to indicate which set of carriers is being referred to. The electron ellipsoids will be referred to as A, B and C. The electrons on ellipsoids B and C have the same cyclotron mass and number density for a field applied in the bisectrix direction, so only expression $\sigma_{xx}^{(BC)}$ will serve for both.

The equations for the components of the total conductivity tensor become:

$$\sigma_{xx} = \alpha_3 (\sigma_{xx}^{(A)} + 2\sigma_{xx}^{(BC)}) + \beta_3 \sigma_{xx}^{(H)}$$

$$\sigma_{yy} = \alpha_1 \sigma_{xx}^{(A)} + \frac{\alpha_1 + 3\alpha_3}{2} \sigma_{xx}^{(BC)} + \beta_1 \sigma_{xx}^{(H)}$$

$$\sigma_{xy} = (\alpha_1 \alpha_3)^{\frac{1}{2}} \sigma_{xy}^{(A)} + [\alpha_1 \alpha_3 + 3(\alpha_2 \alpha_3 - \alpha_4^2)]^{\frac{1}{2}} \sigma_{xy}^{(BC)} + (\beta_1 \beta_3)^{\frac{1}{2}} \sigma_{xy}^{(H)}$$

$$\sigma_{xz} = \alpha_4 (\sigma_{xx}^{(A)} - \sigma_{xx}^{(BC)})$$

$$\sigma_{yz} = \alpha_1 \alpha_4 [-(\alpha_1 \alpha_3)^{\frac{1}{2}} \sigma_{xy}^{(A)} + 2(\alpha_1 \alpha_3 + 3(\alpha_2 \alpha_3 - \alpha_4^2))^{\frac{1}{2}} \sigma_{xy}^{(BC)}]$$

$$\sigma_{yx} = -\sigma_{xy}$$

$$\sigma_{zy} = -\sigma_{yz}$$

$$\sigma_{zx} = +\sigma_{xz}$$

$$\begin{aligned} \sigma_{zz} = \frac{[\omega_p^{(A)}]^2}{4\pi i\omega} (\alpha_2 - K_A) + \frac{[\omega_p^{(BC)}]^2}{4\pi i\omega} \left(\frac{3\alpha_1 + \alpha_2}{2} - K_{BC} \right) \\ + K_A \sigma_{xx}^{(A)} + K_{BC} \sigma_{xx}^{(BC)} \end{aligned}$$

where

$$K_A = \frac{\alpha_4^2}{\alpha_3}$$

and

$$K_{BC} = \frac{1}{2} \cdot \frac{(7\alpha_1 - 3\alpha_2)\alpha_4^2 + 3\alpha_3(\alpha_1 - \alpha_2)^2}{\alpha_1 \alpha_3 + 3(\alpha_2 \alpha_3 - \alpha_4^2)}$$

As usual, the dielectric tensor is calculated from the conductivity tensor by

$$\epsilon_{\approx} = \epsilon_{\approx l} + \frac{4\pi}{i\omega} \sigma_{\approx}$$

For the general case where all components of the dielectric tensor are nonzero, the determinantal equation discussed at

the beginning of Section V becomes

$$N_{\pm}^2 = -\frac{b}{2} \pm \sqrt{(-b/2)^2 - c},$$

where

$$-\frac{b}{2} = \frac{1}{2} [\epsilon_{xx} + \epsilon_{yy} - \frac{1}{\epsilon_{zz}} (\epsilon_{xz}\epsilon_{zx} + \epsilon_{yz}\epsilon_{zy})]$$

and

$$c = \epsilon_{xx}\epsilon_{yy} - \epsilon_{xy}\epsilon_{yx} \\ + \frac{1}{\epsilon_{zz}} [(\epsilon_{xy}\epsilon_{yz} - \epsilon_{yy}\epsilon_{xz})\epsilon_{zx} + (\epsilon_{yx}\epsilon_{xz} - \epsilon_{xx}\epsilon_{yz})\epsilon_{zy}] .$$

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Figure Captions

- Fig. 1: Schematic diagram of data acquisition system. The upper row traces the path of the far infrared radiation. The electrical signal from the detector (upper right) is shown entering the amplifier at bottom left.
- Fig. 2: Cryostat and sample insert. The removable sample insert is shown at right. When inserted into cryostat at left, end of light pipe covers vacuum window and sample is centered in the magnetic field.
- Fig. 3: Graphite boat for growing thick single crystals of bismuth. An oriented seed crystal is placed at the left end of region A.
- Fig. 4: Sample mount for Faraday geometry measurements. A large steel spring presses the sample against a light-absorbing sandwich of charcoal powder-and-grease plus Mylar.
- Fig. 5: Sample mount for Voigt geometry measurements. The plane mirror at top reduces the cross section of the light pipe, and a right angle mirror then directs the radiation horizontally through the sample.
- Fig. 6: The rhombohedral unit cell of bismuth. The cell contains two atoms (from J. M. N. van Goor¹⁸).
- Fig. 7: The Brillouin zone of bismuth. The projection shown

here is along the trigonal axis, with the hole ellipsoid exhibiting a circular cross section at the near face. The other half of each of the three electron ellipsoids is not shown, but they would extend inward from the hexagonal faces on the back side of the zone. The ellipsoids in the diagram are not drawn to scale; they should occupy only 0.001% of the zone.

Fig. 8: A model of the bismuth bandstructure. The Fermi energy is measured from the bottom of the electron conduction band.

Fig. 9: The Fermi level versus magnetic field parallel to the bisectrix axis. Values were calculated from the band parameters of Section IV.

Fig. 10: The carrier densities versus magnetic field parallel to the bisectrix axis. These calculated values complement those of Smith, Baraff and Rowell²⁶ for the binary direction.

Fig. 11: Landau levels in a degenerate free electron gas. The allowed transitions ($\Delta n = 1$) at the Fermi surface have been indicated.

Fig. 12: Calculated energy levels of the heavier electrons at 25 kG. These are the electrons which have the lower cyclotron frequency for a magnetic field applied along

the bisectrix axis. Only the cyclotron transition between the 0- and 1- levels is considered here. The numbers indicate the energies involved in wavenumbers (cm^{-1}).

Fig. 13: The band of cyclotron frequencies for the heavier electrons in the extreme quantum limit. The straight line gives the cyclotron frequency calculated from the low-field cyclotron mass. The dashed line is the average cyclotron frequency used in calculations.

Fig. 14: Calculated energy levels of the heavier electrons at 23 kG. At this field, transitions between higher levels are possible. These transitions involve smaller energies, thereby lowering the average cyclotron frequency.

Fig. 15: The band of cyclotron frequencies for the heavier electrons at lower magnetic fields. The shaded region represents the large number of cyclotron transitions contributing to the lower frequencies.

Fig. 16: Various dielectric functions for electrodynamic disturbances in plasmas. The vertical axis is the square of the index of refraction for a plasma with no relaxation effects included. Negative values of ϵ correspond to frequency bands where no propagation occurs. Negative frequencies refer to the second

mode (ϵ_-) with opposite circular polarization.

Fig. 17: The measured attenuation of far infrared frequencies by bismuth at 100 kG. Only the general shape of the graph is of interest here. The sharp features will be treated later.

Fig. 18: The observed and calculated attenuation of far infrared frequencies at 100 kG. The upper graph assumes that the relaxation rate varies as the square of the frequency; the lower graph assumes a $3/2$ dependence (bisectrix samples 3 and 4).

Fig. 19: The attenuation of far infrared frequencies at 90 kG. This is similar to Fig. 18, but at a slightly lower field.

Fig. 20: The attenuation of far infrared frequencies at 30 kG. This is similar to Figs. 18 and 19, but the range of frequencies is more restricted and the measurements are less accurate (bisectrix samples 3 and 5).

Fig. 21: Bisectrix transmission spectra observed at 90 kG for three sample thicknesses. The vertical scale is expanded for the thicker samples.

Fig. 22: The calculated attenuation of higher frequencies at 100 kG. A frequency dependence of 1.4 for the relaxation rate gives the above agreement at the higher frequencies, with the ϵ_- mode being the one that is

transmitted (bisectrix samples 2 and 3).

Fig. 23: The attenuation of higher frequencies at 80 kG.

A frequency dependence of 14 for the relaxation rate gives the solid line; an $\omega^{1.3}$ dependence (the dashed curve) gives a considerably different fit. The dotted curve is an improved $\omega^{1.4}$ fit obtained by letting the relaxation rate differ for electrons ($\omega\tau = 1000$) and holes ($\omega\tau = 600$).

Fig. 24: The center frequency of the magnetoplasma resonance versus magnetic field. The observed values are indicated by circles. The upper solid line was calculated assuming field-dependent cyclotron masses for the electrons, as in Fig. 13. The lower solid line assumes the low-field value for the cyclotron mass of the heavier electrons.

Fig. 25: The longitudinal component of the dielectric tensor at 30 kG. Only the real part of ϵ_{zz} is plotted here. A second magnetoplasma resonance at 240 cm^{-1} is predicted by this graph.

Fig. 26: The calculated attenuation of 30 kG for both propagating modes over a large frequency interval. The hole cyclotron resonance is at 12 cm^{-1} and the "average" cyclotron frequencies of the electrons are at 192 cm^{-1} and 300 cm^{-1} .

Fig. 27: Total transmitted power versus magnetic field parallel to the bisectrix axis. The power is seen to undergo oscillations as well as increase by an order of magnitude at 14 kG and 27 kG. The solid line connects the observed values for sake of clarity. The sample was 3.10 mm thick.

Fig. 28: A series of spectra showing rapid changes near onset of the extreme quantum limit. The spectra were taken at 25.5, 27, 30, 35, and 40 kG. The sharp absorption in the 27 kG spectrum was removed from the higher-field spectra for clarity. The rapid oscillations seen in all spectra originate with the source lamp (bisectrix sample 3).

Fig. 29: Calculated change in transmittance near 27 kG due to changing cyclotron frequencies alone. Most of the calculated change shown above is due to the large increase in the cyclotron frequency of the heavier electrons at onset of the EQL. The dots represent the points actually calculated.

Fig. 30: Calculated change in transmittance near 27 kG, including Shubnikov-de Haas effect. Graph on left is Fig. 29 . with a dashed line added to indicate effect of increasing the scattering rate of the heavier electrons. Graph on right indicates the

extent to which these results agree with the ratio of observed intensities at 25.5 kG and 27 kG.

Fig. 31: The temperature-dependent component of the power attenuation coefficient at 48 kG. The straight line has a slope of 2.02, suggesting a quadratic dependence.

Fig. 32: Cyclotron-like resonances in bisectrix spectra. The lines near 20, 40 and 60 cm^{-1} can be seen in two samples of different thickness. Also visible in the upper spectrum is the magnetoplasma resonance at 75 cm^{-1} .

Fig. 33: Magnetic field dependence of the center frequencies of the cyclotron-like resonances. The straight lines represent a fit to a single cyclotron mass (lowest line) and its submultiples; the mass corresponds to that of the holes. The other points mark the position of the magnetoplasma resonance.

Fig. 34: The effects of various Fourier components on otherwise uniform circular motion. The function graphed was $v(\theta) = \exp(i\theta) + 0.1 \exp(in\theta)$ evaluated for $\theta = 0^\circ, 10^\circ, 20^\circ, \dots$. The presence of a harmonic results in non-uniform as well as noncircular motion.

Fig. 35: Some unidentified resonances seen in one sample. Cyclotron-like resonances are seen near 40 cm^{-1} and 80 cm^{-1} ; the three others are unaccounted for.

Figure 1

Cryostat + Sample Insert

Figure 2

E | D | C | B | A

Figure 3

Sample Holder

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13

Figure 14

Figure 15

Figure 16

Figure 17

Figure 18

Figure 19

Figure 20

Bisectrix Transmission
Spectra at 90 kGauss

Frequency (cm^{-1})

Figure 21

Figure 22

Figure 23

Figure 24

Figure 25

Figure 26

Figure 27

Figure 28

Figure 29

Figure 30

Figure 31

Bisectrix Spectra
45 kGauss

Frequency (cm^{-1})

Figure 32

Figure 33

Figure 34

Figure 35