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PHYSICAL REVIEW B VOLUME 6, NUMBER 2 15 JULY 1972

Temperature Dependence of Alfvén-Wave Amplitudes and Carrier Relaxation Times in Bismuth

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Alfvén-wave propagation in parallel-plate Bi samples has been studied using a 24-GHz microwave reflection spectrometer. The attenuation of Alfvén-wave amplitudes in a magnetic field up to 10 kG was measured at different temperatures ranging from 2.4 to 11°K. From the amplitude of the weakly damped waves, effective relaxation times of charge carriers were measured directly without using any band-structure parameter. The temperature dependence of the effective relaxation times can be fitted to a T^{-2} variation with different slopes for different crystal orientations.

I. INTRODUCTION

The propagation of low-frequency electromagnetic waves in solid-state plasmas has been the subject of many recent investigations.[1] In the presence of a dc magnetic field, waves can propagate in metals and semiconductors with little attenuation when the wave frequency ω is much lower than the cyclotron frequency ω_c of the charge carriers in the solid which can be regarded as a stable plasma. Various modes of wave propagation have been extensively investigated in several laboratories. If the solid contains equal numbers of electrons and holes, as in Bi and other group-V semimetals, the magnetoplasma wave propagates in a linearly polarized mode, known as the Alfvén wave.

In the Alfvén mode, the propagation vector of the wave is determined by the diagonal magnetoresistance term in the conductivity tensor. As a result, the power absorption in the solid-state plasma is directly proportional to the dc magnetic field H and the wave is heavily damped except for $\omega\tau \gg 1$ (τ is the carrier relaxation time). However, when the conditions for Alfvén-wave propagation are satisfied, measurements of the wave characteristics can be used in complement with the ordinary magnetoresistance studies to obtain information on the electronic structure of the solids.

To date, Alfvén waves have been used to study the carrier concentration and effective masses of electrons and holes in Bi. More recently, from

attenuation of Alfvén-wave amplitudes, the average carrier relaxation time τ in Bi has been determined and shown to be anisotropic and magnetic field dependent.[2,3] The field dependence was consistent with the Shubnikov–de Haas or de Haas–van Alphén-effect data. It has been pointed out[2] that the Alfvén-wave method may offer a more direct means for measuring the variations of τ than the ordinary galvanomagnetic or size-effect methods.

This paper reports studies of the temperature dependence of the Alfvén-wave amplitude and τ in pure Bi for two different orientations. We have investigated the microwave power absorption in the sample by a cavity method for a dc magnetic field up to 10 kG in the temperature range (2.4–11)°K.

II. MAGNETOPLASMA WAVES IN Bi

The propagation of electromagnetic waves in a solid in the presence of a dc magnetic field H is determined by

$$\vec{k}\times(\vec{k}\times\vec{E}) = -(4\pi i\omega/c^2)\vec{j} \tag{1}$$

and

$$\vec{j} = \sigma(\vec{H})\vec{E}, \tag{2}$$

where (1) is a general result from Maxwell's equations while in (2) the magnetoconductivity tensor $\sigma(H)$ is determined by the structure of the solid. In general, there are two different modes of Alfvén waves which can propagate in a solid. This can be seen easily by an examination of the eigenvectors of the wave equations obtained from (1) and (2). In one mode, $\vec{E}$ is perpendicular to $\vec{H}$ but lies in the plane of $\vec{H}$ and $\vec{k}$. This is referred to as the "slow wave." The other mode, in which E is at right angles to both $\vec{H}$ and $\vec{k}$, is termed the "fast wave." In this experiment, we choose to work with the configuration where $\vec{H}$, $\vec{E}$, and $\vec{k}$ are mutually perpendicular to each other so that only the fast wave will propagate. This simplifies the analysis of data considerably.

It can be shown that for ellipsoidal Fermi surfaces, assuming an isotropic relaxation time τ, k satisfies

$$k^2 = \frac{\omega^2 4\pi n f(m)}{H^2}\left(1+\frac{i}{\omega\tau}\right), \tag{3}$$

where n is the carrier concentration and $f(m)$ is a function of the effective masses. For the specific case of Bi, with three electron ellipsoids and one hole spheroid, the function $f(m)$ has been tabulated.[2] It was shown in Ref. 2 that for normal incident Alfvén waves propagating through a parallel-plate sample of thickness d, the apparent power absorption A (sum of power absorbed and transmitted by the sample) can be written

$$A = D + \alpha(4V/c)[1+2\cos(4\pi d/\lambda)\,e^{-d/V\tau}] \tag{4}$$

for $\omega_c\tau \gg \omega\tau \gg 1$. D and α are undetermined constants characteristic of the experimental microwave system. V is the Alfvén velocity $H/[4\pi n f(m)]^{1/2}$ and $\lambda = 2\pi V/\omega$. The significance of this result was discussed in Ref. 2. The absorption oscillates with H, being maximum when $d = \frac{1}{2}p\lambda$, where p is an integer. It was also pointed out there that τ in (3) can be determined directly from a single trace of the A-vs-H plot obtained experimentally making use of the expression given in (4).

The point we would like to emphasize is that once the quantity τ is defined as in (3), it can be determined from a microwave absorption measurement without any other information about the solid. That is, τ in (3) can be measured directly, independent of the functional form of $f(m)$.

The above argument can be generalized to the case of anisotropic relaxation times. One can easily visualize that, at least for simple closed ellipsoidal Fermi surfaces, for the case of anisotropic scattering, it is possible to define an effective relaxation time τ such that k^2 takes the form of (3) when $\omega_c\tau \gg \omega\tau \gg 1$, although the τ and $f(m)$ may involve more complicated combinations of m's and τ's owing to different carriers. As a simple example, consider two isotropic masses m_1 and m_2 with different relaxation times τ_1 and τ_2, respectively. k^2 can be written as in (3) with $f(m) = m_1 + m_2$ and $1/\tau = (m_1/\tau_1 + m_2/\tau_2)/(m_1+m_2)$. Thus the effective τ is a "mass-weighted" average relaxation time. We therefore regard the τ in (3) as a formal definition of the effective relaxation time determined by the Alfvén-wave experiment.

To analyze the anisotropic scattering for each type of charge carrier in the solid, one has, naturally, to make use of the knowledge of electronic band structure.[4] However, the direct determination of the effective τ by the Alfvén-wave method has the advantage for studying the variation of τ which may be common to all anisotropic charge carriers, such as the temperature or impurity dependence.

The average relaxation times measured by the Alfvén-wave method may be compared with those obtained from line-shape measurements of the cyclotron resonance and of the Gantmakher effect. In the latter cases, although the cyclotron or geometrical resonance peaks are determined by the selective resonant carriers, nevertheless, other nonresonant carriers present in the metal may have significant contributions to the observed line shape. By and large, the effective τ measured by the Alfvén-wave method and that by the resonance methods are similar in nature; however, the local current-field relationship commonly used in the Alfvén-wave case, in contrast to the nonlocal relationship in the resonance effects, makes the phenomenological approach and analysis of the anisotropic relaxation

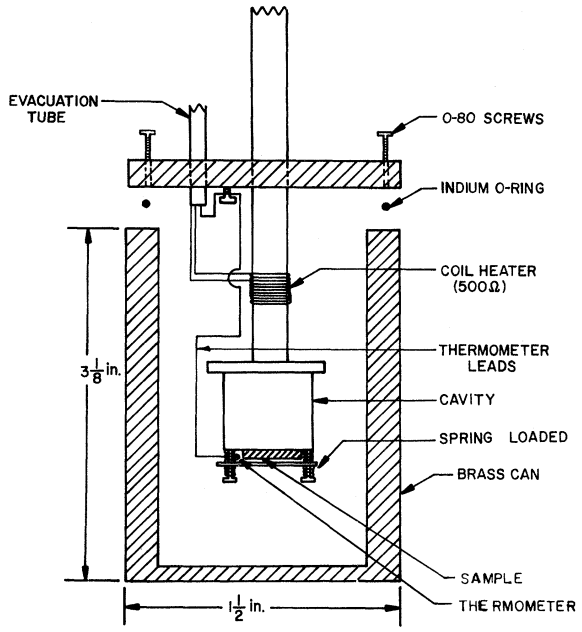

FIG. 1. Experimental sample holder.

times more tractable.

III. EXPERIMENTAL

The bismuth samples were prepared from a single crystal ingot, grown in a zone-leveling apparatus. A thin slab $1\times1\times0.1$ cm was cut by electric discharge machining with its large faces parallel to each other and perpendicular to the bisectrix axis. This orientation was chosen since previously reported data[2,5] have shown that it is the one for which $1/\tau$ is the smallest among all the possible orientations of the sample with respect to the applied magnetic field and microwave electric field directions. Though the Alfvén signal is large at low temperatures, it decreases rapidly with increasing temperature as τ decreases, and the initial signal strength ultimately determines the highest temperature at which measurements of τ can be achieved. The sample was chemically polished and then annealed for 36 h at 10^{-6} Torr and 130 °C. The finished samples have mirrorlike surfaces.

A microwave reflection spectrometer employing a TE102 rectangular resonance cavity of frequency 24 GHz was used to determine changes in the absorption of the sample. The measuring technique is quite standard as described elsewhere.[2]

The cavity and lower portion of the waveguide were contained in an evacuable brass can (Fig. 1). The can pressure was kept at about 10^{-3} Torr. The temperature was raised above 4.2 °K by passing a regulated current through a 500-Ω constantan coil heater wound around the portion of waveguide inside the can near the cavity. The temperature was determined by means of a calibrated carbon resistance thermometer placed next to the sample and smeared with Apiezon N grease for good thermal contact. The temperature was stable to better than 0.005 °K. The estimated error in the temperature measurement was at worst ±0.1 °K.

The Alfvén mode propagates strongest with the applied magnetic field perpendicular to the microwave electric field, and therefore this was the geometry used. Small errors in the parallelism of the faces of the sample could affect the absolute magnitude of the measured relaxation times but not the temperature dependence.

IV. RESULTS AND DISCUSSION

We begin by presenting in Fig. 2 a typical recorder tracing, showing the variation in crystal detector output which is proportional to the fraction of radiation absorbed and transmitted through the sample. Oscillations in the absorption coefficient of the sample become noticeable at about 3 kG, and increase in amplitude as the magnetic field increases. Such curves were obtained at various temperatures from 2.4 to 11 °K. In each case it was possible to draw the upper and lower envelopes of the oscillations and to measure the differences between them at the position of each extremum. The differences are directly proportional to the oscillation

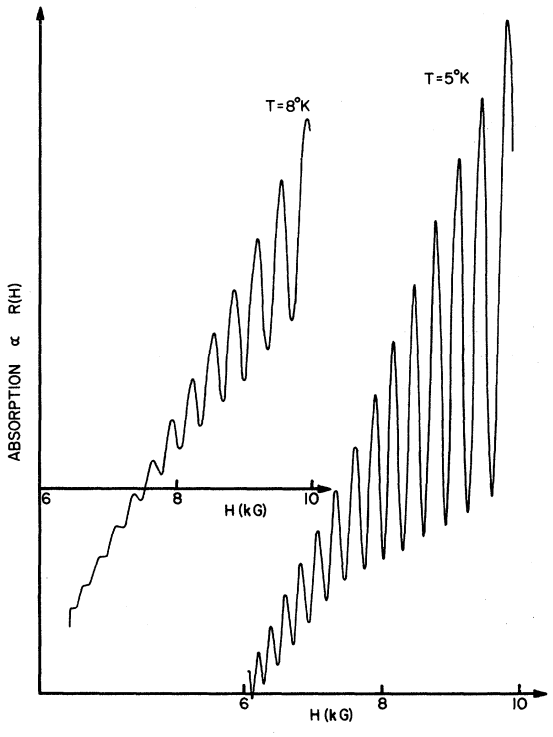

FIG. 2. Typical standing-wave Alfvén oscillations. The absorption is proportional to the crystal detector output. Both curves are plotted on the same scale.

amplitude $E = 16\alpha(V/c)e^{-d/V\tau}$. The field at which the Alfvén oscillations first become apparent, increased with temperature. This is due to an overall decrease in the oscillation amplitude with increasing temperature.

The data allow us to determine the integer p, the number of Alfvén half-wavelengths contained in the sample at each maximum. For $d = \frac{1}{2}p\lambda$, it is apparent that p is inversely proportional to the magnetic field H. The plot of $1/H$ at each successive maximum against successive integers p should yield a straight line, whose slope gives us $\omega d[4\pi mf(m)]^{1/2}/\pi$. The determination of the Alfvén velocity and τ is independent of the absolute value of p, although p may be determined by appropriate assignment of the "fringe index."

In the present experiment, we studied two different crystal orientations: (i) $\vec{H} \| $ binary, $\vec{E} \|$ trigonal and (ii) $\vec{H} \|$ trigonal, $\vec{E} \|$ binary. The propagation vector k is always along the bisectrix direction and perpendicular to the sample. The slope of the p-vs-$1/H$ plot can be determined quite accurately and the only significant source of error is in the measurement of $H(\pm 2\%)$. The measured Alfvén-wave velocities are in agreement with those reported in Ref. 2 and will not be repeated here.

We attempted to determine τ as defined in (3) and (4) by the method described in Ref. 2. To this end, we plotted $\ln(E/H)$ vs $1/H$ at the absorption maxima, the result is a straight line whose slope gives us $d[4\pi mf(m)]^{1/2}/\tau$. Since $d[4\pi mf(m)]^{1/2}$ is known from the p-vs-$1/H$ plot for the same crystal orientation, this method will allow us to determine τ without knowing the functional form of $f(m)$.

Above 8 °K, the ln plot of (E/H) vs $1/H$ does give a good straight line from which τ can be determined quite accurately. At lower temperatures, however, all the log-plot curves contain invariably oscillations periodic in $1/H$. These oscillations are due to the well-known Shubnikov–de Haas effect (SdH) as have been studied in the previous experiments.[2,3] The presence of these oscillations sometimes makes an accurate determination of τ difficult. For the range of magnetic field where we observed the Alfvén oscillations (3–10 kG), there are enough SdH oscillations in the ln plot at low temperatures [(2–4) °K], so that an average value of τ can be determined. However, at slightly higher temperatures, typically around 6 °K, the SdH oscillations diminish in amplitude and the Alfvén oscillations start at higher field values; consequently, the In plot gives a smooth curve with less than one SdH period up to 10 kG. In this case, the average slope is subject to large error and we are not at a position to determine τ following the method described above.

To surmount such a difficulty, we have looked at the data in a slightly different manner. Assuming that the microwave system is insensitive to temperature changes in the range from 2 to 10 °K (which has been checked by a background run), we measured the $\ln E$ values for different temperatures at the same magnetic field. Since the Alfvén velocity is independent of temperature in our model and the temperature-dependent part of SdH oscillations makes very little contribution to the temperature variations in Alfvén amplitude (estimated to be less than 10%), the difference in $\ln E$ at two different temperatures gives us the difference in $1/\tau$, since $\ln E = \ln\alpha(16V/c) - d/V\tau$. Although this method does not give us the values of τ, it does give the temperature variation. As a consistency check, we used the τ value at the lowest temperature in this experiment obtained from $\ln E$-vs-$1/H$ plot as a reference point and then computed all the τ values at higher temperatures by finding the differences in $\ln E$ at a constant H around 9 kG. These values are compared with those determined from the $\ln E$-vs-$1/H$ plots in the temperature range for which an accurate τ could be obtained. The two methods give results which agree to within 10%. We therefore employed the measurement of $\ln E$ at constant H as a convenient way for determining the temperature dependence of $1/\tau$. The constant H value was chosen at a pronounced maximum in the Alfvén-oscillation curve; throughout the temperature range under investigation the position of a given peak is essentially independent of temperature.

We plot the temperature dependence of $1/\tau$ for two Bi crystal orientations in Figs. 3 and 4. We assume that the temperature variation of $\tau(T)$ can be written

$$1/\tau(T) = 1/\tau_i + BT^{\gamma}, \tag{5}$$

where τ_i is the temperature-independent impurity limit of τ, B and γ are constants. For each crystal orientation, we plot $\Delta(1/\tau) = 1/\tau(T) - 1/\tau(T_0) = BT^{\gamma} - BT_0^{\gamma}$ against T^{γ} for $\gamma = 1, 2$, and 3, T_0 is the lowest temperature at which τ has been measured for that particular orientation.

To decide on the choice of γ, we may first rule out $\gamma = 1$ in the temperature region we have studied [curve(c)]. The T^3 dependence appears to fit the data reasonably well at lower temperatures, but deviates appreciably for T higher than about 8 °K. If the scattering is due to electron-phonon interaction and the small angle scattering is equally effective as the large angle scattering, as in the case of resonance effects, we would expect a T^3 dependence for $1/\tau$. Since the phonon density increases with increasing temperature, the agreement of data with a T^3 plot should improve with an increase in temperature. As this is not observed, we tend to rule out the possibility of T^3 dependence. It should be pointed out, however, that phonons at tempera-

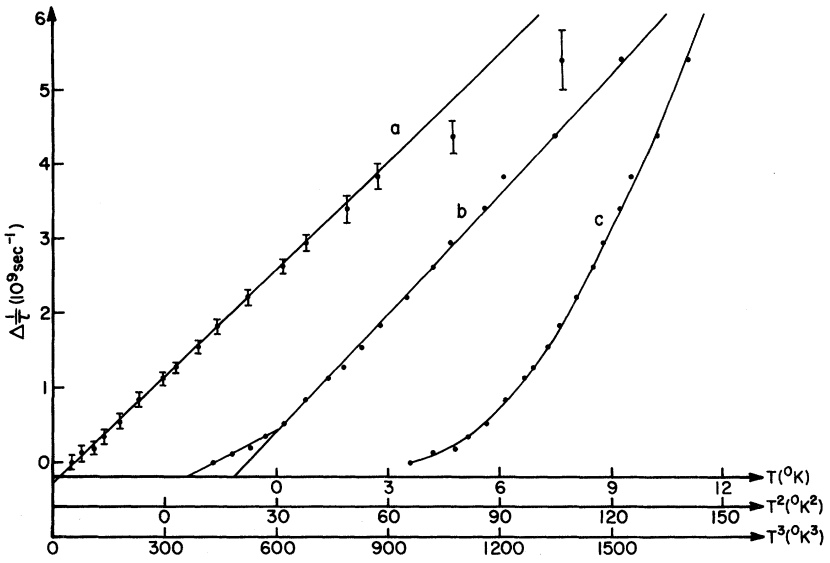

FIG. 3. Temperature variations of $1/\tau$; $\Delta(1/\tau) = 1/\tau(T) - 1/\tau(T_0)$, $T_0 = 3.6$ °K. Curve (a): $\Delta(1/\tau)$ vs T^3; (b): $\Delta(1/\tau)$ vs T^2; and (c): $\Delta(1/\tau)$ vs T. All the data were taken at an oscillation maximum at $H = 9.72$ kG. $\tau(T_0 = 3.60) = 1.33 \times 10^{-10}$ sec. In curve (b), the two slopes are $(5.20 \pm 0.2) \times 10^7$ and $(2.40 \pm 0.2) \times 10^7$ sec^{-1}°K^{-2}, respectively. $H \parallel$ binary, $E \parallel$ trigonal.

tures near 10 °K do not provide large enough momentum to cause intervalley scattering of electrons in Bi. Intravalley electron-phonon scatterings may still be possible at low temperatures, but this process gives rise to a T^2 dependence in Bi when the geometry of the highly prolate Fermi surfaces is taken into account.[6] The absence of a T^3 dependence is therefore consistent with these considerations.

As we see in Figs. 3 and 4, the T^2 dependence fits the data quite well. This is in accord with the measurements obtained by different methods.[6-13] Furthermore, we notice the presence of a rather marked change in slope at a temperature near 5 °K. This effect was also found in some resistivity measurements.[11] The presence of a break may indicate the onset of a new scattering process although the nature of this scattering mechanism is still unclear. In any event, its presence in two independent sets of measurements indicates that this effect would warrant further study.

Gantmakher and Leonov[6] have pointed out that the intravalley electron-phonon scattering may result in a T^2 dependence of $1/\tau$, this mechanism, though plausible in low temperatures, may be ruled out by our observation of a constant-slope T^2 dependence from 5 to 11 °K. To scatter an electron within the same ellipsoid, a phonon with energy $\lesssim 6$ °K is needed. At about 6 °K, a phonon has a momentum equal to the largest dimension of an electron ellipsoid.[6] Therefore, we would expect an increase in $1/\tau$ with T up to 6 °K and then it should level off. In contrast to this prediction, we

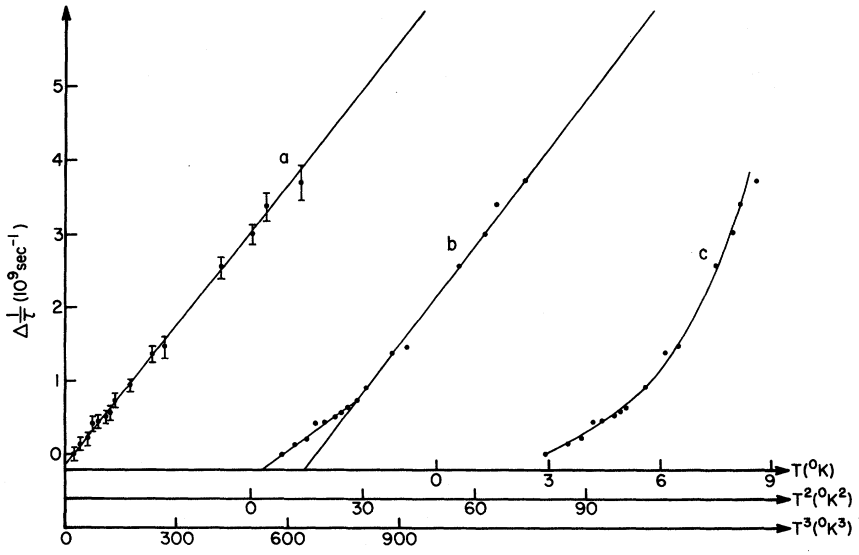

FIG. 4. Temperature variations of $1/\tau$; $\Delta(1/\tau) = 1/\tau(T) - 1/\tau(T_0)$, $T_0 = 2.94$ °K. Curve (a): $\Delta(1/\tau)$ vs T_3; (b): $\Delta(1/\tau)$ vs T^2; and (c): $\Delta(1/\tau)$ vs T. All the data were taken at an oscillation maximum at $H = 9.35$ kG. $\tau(T_0 = 2.94) = 2.40 \times 10^{-10}$ sec. In curve (b), the slopes are $(6.30 \pm 0.4) \times 10^7$ and $(4.0 \pm 0.3) \times 10^7$ sec^{-1}°K^{-2}, respectively. $H \parallel$ trigonal, $E \parallel$ binary.

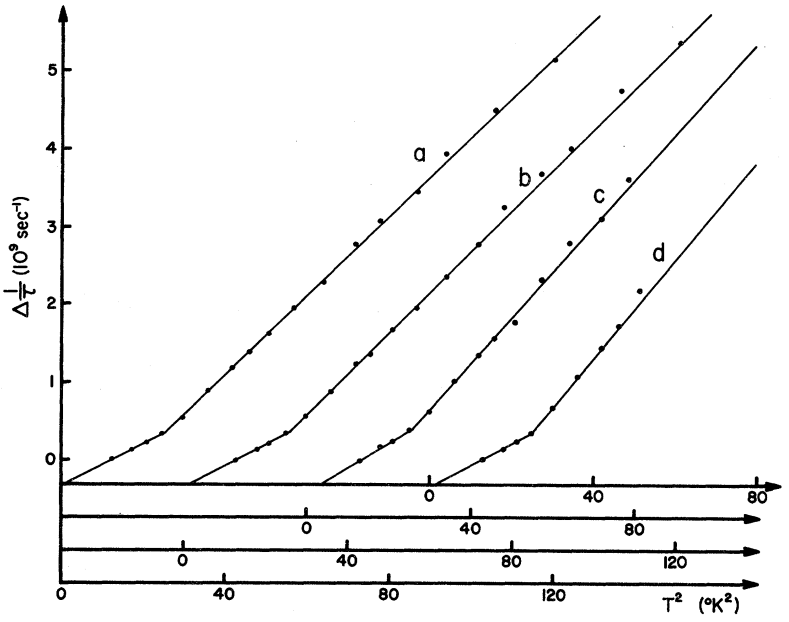

FIG. 5. $\Delta(1/\tau)$ vs T^2 for different values of H. Curve (a): $H=9.72$ kG; (b): $H=9.00$ kG; (c): $H=8.12$ kG; and (d): $H=7.20$ kG. $E\parallel$ trigonal, $H\parallel$ binary.

found *no decrease* in slope in the $1/\tau$-T^2 plot and that T^2 dependence remains valid up to 11 °K, indicating that the intravalley electron-phonon scattering is probably not an important mechanism.

The T^2 dependence may be attributed to interband or intraband carrier-carrier scattering due to direct Coulomb interaction or via phonons. It is difficult to compare the relative contributions arising from each process without a theoretical quantitative estimate. No such a theoretical calculation for Bi is available at this time.

In addition to the normal processes, there are possible carrier-carrier umklapp processes which may also be responsible for the T^2 dependence of of $1/\tau$ in Bi and was not accounted for in the analysis of previous works. To obtain a rough estimate of the magnitude of this U process, we employed a formula given by Ziman[14] and modified it for the case of Bi. The T^2-dependence part yields $1/\tau \approx 2.0 \times 10^{14} G^2 T^2$ sec^{-1}, where G is an interference factor depending on the carrier wave functions in the unit cells involved with the U process.

Our measured values for the slope B in the expression $1/\tau(T) = 1/\tau_i + BT^2$ are (i) $B=(5.20\pm0.1)\times10^7$ sec^{-1} °K^{-2} for $\vec{H}\parallel$binary, $\vec{E}\parallel$trigonal and (ii) $B=(6.30\pm0.2)\times10^7$ sec^{-1} °K^{-2} for $\vec{H}\parallel$trigonal, $\vec{E}\parallel$binary. These values are in the same range as those obtained from cyclotron resonance by Drew and Strom,[13] $B=(2.02\pm0.07)\times10^8$ sec^{-1} °K^{-2} at low frequency by extrapolation for $\vec{E}\parallel$binary and $\vec{H}\parallel$bisectrix and $B=(2.30\pm0.08)\times10^8$ sec^{-1} °K^{-2} using the dielectric anomaly at 890.7 GHz. Although their values are about four times larger than ours, the frequency and angular dependence as well as different experimental accuracies may account for the differences.

Using our measured values of B's, we estimated the interference factor G in the U process to be approximately 5×10^{-4}.

We have studied the T^2 dependence of $1/\tau$ at various H values. Figure 5 shows a plot of $\Delta(1/\tau)$ vs T^2 for four different magnetic fields. It is seen that B as well as the kink at ≈ 5 °K are insensitive to H although B seems to show a small decrease as H is increased, indicating that the scattering mechanism responsible for the T^2 behavior is insensitive to the change of H. This also tends to rule out the possibility of transition between different Landau levels in a quantizing magnetic field.

The occurrence of a break at ≈ 5 °K appears to be quite interesting. Although its origin is unknown at this time, we may at least state that apparently more than one scattering process must be present in Bi. We speculate that a rather abrupt increase in scattering probability may occur when either a phonon emission has taken place or when the screening effect of charges has been reduced. More evidence is needed to exploit this new effect.

V. CONCLUSION

We have measured the temperature dependence of $1/\tau$ in Bi between $T=2.4$ °K and $T=11$ °K. The scattering probability is fitted quite well by T^2 variations with a change of slope at a temperature near 5 °K. It is difficult at present to assess the different contributions to this temperature dependence due to various carrier scattering mechanisms. However, we ruled out the possibilities of electron-phonon interactions even though the peculiar Fermi-surface geometry may give rise to a T^2 dependence.

It seems that intraband and interband Coulomb scatterings are weak because of the effective screening in the high-density limit, however, we feel that their contributions should not be precluded before more careful quantitative estimates are made. The phonon-mediated carrier-carrier interaction and U processes seem highly probable. So far we have only made quantitative estimates for the U process based on our data while the other conceivable carrier-carrier scattering processes will require more detailed theoretical assessment. The break at about 5 °K strongly suggests that more than one scattering process is present in Bi; the origin of this break remains unknown.

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Effect of Hydrostatic Pressure and Pd Doping on the Fermi Surface of $AuGa_2$ †

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The effect of hydrostatic pressure on most of the reported cross-sectional areas of the Fermi surface of $AuGa_2$ for fields along principal symmetry directions is reported. Pressure derivatives of these cross sections were obtained using the fluid-He phase-shift technique. de Haas–van Alphen frequencies were also measured for $\vec{H} \parallel [111]$ to maximum pressures of ~9 kbar generated in solid He. The results in general are found to be in good qualitative agreement with the band model of Switendick and Narath. A striking departure from linearity in the pressure dependence of the cross section associated with the arms of the third-zone hole surface occurs near 6 kbar. Also reported are some results of the effect of adding Pd to $AuGa_2$ on these same third-zone arms which are in rough qualitative agreement with a rigid-band picture.

I. INTRODUCTION

The isomorphous compounds $AuAl_2$, $AuGa_2$, and $AuIn_2$ have received considerable attention in the past several years. A large part of this interest was stimulated by the so-called "$AuGa_2$ dilemma" which stems from the fact that $AuGa_2$ displays a strongly temperature-dependent ^{71}Ga Knight shift, spin relaxation time, and bulk susceptibility while the analogous quantities in $AuIn_2$ and $AuAl_2$ are relatively temperature independent.[1] On the other hand, many of the electronic properties such as the Fermi-surface topologies,[2] the electronic specific heats,[3] and the Hall coefficients[4] are characterized by similarities across the series.

Switendick and Narath[5] advanced an explanation for this situation based on a striking difference in the band structure of $AuGa_2$ from that of the other two compounds. In $AuAl_2$ and $AuIn_2$ the second band intersects the Fermi energy giving rise to a closed-hole surface centered at the point Γ in the Brillouin zone. This band possesses strong Ga, In, or Al character in the respective compounds. In $AuGa_2$ this band is found to be extremely flat and lies entirely below the Fermi energy at absolute zero. A reproduction of Switendick's band picture for $AuGa_2$ is shown in Fig. 1. The anomalous temperature dependence of the ^{71}Ga NMR data is then explained in terms of depopulation of this band as the temperature increases.

The Fermi surfaces of the series have been shown to agree fairly well with the free-electron picture for a valence-seven face-centered-cubic metal.[2] Figure 2 shows the second-zone hole sur-