Dispersion of sound velocity and the electron relaxation time in bismuth and antimony

A. P. Korolyuk, M. A. Obolenskiĭ, and V. I. Beletskiĭ

Institute of Radiophysics and Electronics, Ukrainian Academy of Sciences
Khar'kov State University
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The temperature dependence of the sound velocity dispersion in bismuth and antimony is measured in the temperature ranges 3.5–7.5°K and 4–10°K, respectively, and in strong magnetic fields for which $KR \ll 1$ (**K** is the sound wave vector, R is the electron orbit cyclotron radius) and under the conditions that $\mathbf{K} \perp \mathbf{H}$ and $\omega\tau \leq 1$ (ω is the sound frequency, τ is the relaxation time). The temperature dependence of the relaxation time in bismuth and antimony is studied. In both cases, the temperature-dependent part of the inverse relaxation time is proportional to T^2. The numerical values of the proportionality coefficients for bismuth and antimony are $\beta = 0.57 \times 10^8$ sec^{-1} deg^{-2} and $\beta = 0.15 \times 10^8$ sec^{-1} deg^{-2}, respectively.

Magnetoacoustic phenomena in metals have long been used with success in the study of their electron energy spectra.[1] It was shown in[2-4] that there is a sharp (resonant) increase in the sound absorption coefficient and sound velocity in strong magnetic fields in the case of deviation of the magnetic field vector **H** by a small angle $\varphi = \varphi_{cr}$ relative to the position $\mathbf{K} \perp \mathbf{H}$ (**K** is the sound wave vector). This phenomenon, which is given the name "oblique effect" in the literature, is due to electrons from the neighborhood of the limiting point on the Fermi surface. The conditions imposed here on the sound frequency and on the magnetic field intensity H should be quite stringent:

$$\omega\tau \gg 1, \quad KR \ll 1. \tag{1}$$

Here τ is the relaxation time and R is the cyclotron radius of the orbit of the electrons.

Interest in the study of the time of free flight of electrons on the Fermi surface stimulates the development of new methods for its measurement. Magnetoacoustic resonance phenomena have already been used under certain specific conditions for the measurement of the electron free path length in metals.[5,6] However, if we are interested in the temperature dependence of the path length (or time) in connection with the study of scattering mechanisms, resonance effects turn out to be less suitable, because of natural limitations (their vanishing) that occur following a comparatively small increase in the temperature and decrease in the path length. It could be expected that investigations of the dispersion and absorption of sound in strong magnetic fields (but not strong enough to be quantizing) would turn out to be preferable from this point of view, and would allow us to raise the temperature limit of the measurements.

It was shown both theoretically and experimentally in[2,3,7,8] that the sound absorption in a transverse magnetic field $\mathbf{H} \perp \mathbf{K}$ changes according to the law

$$\Gamma = A \frac{\omega\tau}{1+\omega^2\tau^2}, \tag{2}$$

while the change in the velocity under these same conditions, referred to the velocity in the absence of the field, is equal to

$$\frac{\Delta s}{s} = A \frac{\omega^2\tau^2}{1+\omega^2\tau^2}. \tag{3}$$

Here A is a constant that is independent of the magnetic field and of the temperature.

Upon increase in τ (decrease in temperature), the sound absorption decreases and, if $\omega\tau \gg 1$, it tends toward zero as $1/\omega\tau$; on the other hand, the dispersion increases, tending toward its maximum value A, the determination of which under the conditions $\omega\tau \gg 1$ would make it possible to calculate $\tau(T)$ for any temperature, regardless of the scattering mechanism of the carriers.

It is seen from the dispersion (3) that the greatest sensitivity to a change in τ is achieved in the interval where $\omega\tau = 1/\sqrt{3}$:

$$\frac{d^2}{d(\omega\tau)^2}\frac{\Delta s}{s} = 2A \frac{1-3\omega^2\tau^2}{(1+\omega^2\tau^2)^3} = 0 \tag{4}$$

and the dispersion is almost completely insensitive to changes in τ if $\omega\tau \gg 1$ (the saturation region).

Various mechanisms of scattering of electrons are usually regarded, in the first approximation, as independent of one another, which allows us to represent τ in the form

$$1/\tau = 1/\tau_0 + 1/\tau(T), \tag{5}$$

where τ_0 is the relaxation time of electrons on impurities and other static distortions of the lattice, and $\tau(T)$ is the time determined by electron-electron and electron-phonon collisions.

For bismuth and antimony, the temperature-dependent part of the relaxation time was measured in[9-12]; it is of interest, by measuring this same quantity over a wider temperature range, to compare the resultant data with already known data, thereby verifying the new method. Inasmuch as there is interest also in the numerical values of the mean time of flight τ, along with interest in the function $\tau(T)$, one could ascertain, by comparing our data with the results of[9-11], whether the quantity τ defined in Eqs. (2) and (3) as the relaxation time of the distribution function at equilibrium[7,8] is identical with the mean free path time obtained from data on the size effect.

EXPERIMENT

The relaxation time can be determined both by dispersion measurements and by measurement of the sound absorption in a strong field. The quantity $\Delta s/s$ is the more convenient for the measurements. The sensitivity of the apparatus for the determination of Γ depends on the loss in joints between the transducers and the sample, and a calibration is necessary for the temperature measurements. On the other hand, the change in the generation frequency[1)] does not depend on the nature of the joint, and calibration is not required for measurements of the relative frequencies $\Delta f/f \sim \Delta s/s$.

Single crystals of bismuth and antimony were used in the experiments. These were obtained from metals of

grade Bi-000 and Sb-000, subjected to additional zone recrystallization in a graphite mold. The resultant bulk single crystals had dimensions of 20 × 15 × 200 mm. The contaminated ends were cut off by an electric spark cutter and the samples were prepared from the middle part. The resistance ratio which characterizes the purity of the samples, was R(300° K)/R(4.2° K) = 300 for bismuth and R(300° K)/R(4.2° K) = 2700—3000 for antimony.

The samples were cut with a tungsten wire of 0.05 mm diameter in the form of discs of 6—8 mm diameter and thickness d = 1—3 mm. Since the samples must themselves act like acoustic resonators of the Fabry-Perot type in the measurements of the dispersion of the sound velocity, it was important that they be plane-parallel. Lapping of the samples to the required degree of plane-parallelism was performed by a fine abrasive with subsequent polishing; the metal surface layer damaged by such a treatment amounted to a depth of about 0.1 mm, i.e., not more than 20% for very thin samples.

The experiments were performed in the temperature range 1.4—12° K; the temperature was measured as usual by the helium vapor pressure in the range 1.4—4.2° K, and with a differential thermocouple[2)] (ELZh-999-copper) at higher temperatures. In this case the sample was placed in a brass container in which a vacuum was created; it was heated by a wire-wound oven. One end of the thermocouple was attached to the sample and the second to the container wall in direct contact with the liquid helium.

Investigations of the temperature dependence of the dispersion of the sound velocity were made on a series of 3—4 samples, prepared from one bulky single crystal. The experimental points had a small scatter from sample to sample, and therefore we give in this paper the results of measurements obtained on one of them.

EXPERIMENTAL RESULTS

It follows from theory[3] that the dispersion of the sound velocity in a transverse magnetic field does not depend on its intensity if the conditions (1) are satisfied. The field intensity, which can be assumed to be strong for given ω and K, was determined from measurement of the dispersion at some constant temperature as a function of the field H. Typical plots $\Delta s/s$ as a function of the intensity of the magnetic field at T = 4.2° K are shown in Fig. 1 for the case **K** ∥ Y, **H** ∥ Z. As usual, we connect the set of coordinates X, Y, Z with the directions of the binary, bisector and trigonal axes of the crystal, respectively. Saturation of the dispersion in the given case takes place in fields H > 2 kOe. Further increase in the field intensity does not affect the magnitude of the dispersion, showing that the criterion (1) is satisfied for the given mutual orientation of the vectors K and H.

The temperature dependence of the dispersion of the sound velocity in bismuth and antimony was studied at two ultrasonic frequencies, 165 and 65.4 MHz. The maximum of $\Delta s/s$ as a function of the angle φ between the magnetic field and the direction of the sound wave vector K was reached, in complete agreement with the theory, at $\varphi = \pi/2$, i.e., $(\mathbf{K}\cdot\mathbf{H}) = 0$, and the curve $\Delta s/s = f/\varphi$ had a bell-shaped form.

Figure 2 shows the results of measurement of $\Delta s/s$ as a function of temperature with the ordinates in logarithmic scale. The upper three curves, designated by

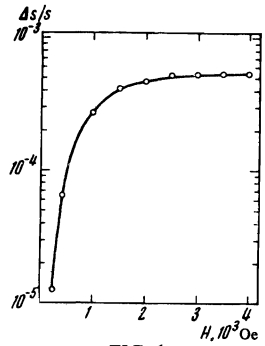

FIG. 1.

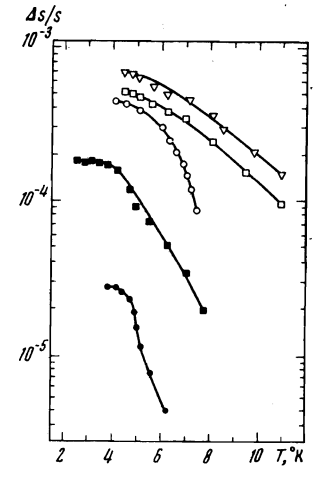

FIG. 2.

FIG. 1. Dispersion of sound velocity $\Delta s/s$ in antimony as a function of the field intensity in the case **K**||Y and **H**||Z.

FIG. 2. Dependence of the sound velocity dispersion $\Delta s/s$ on the temperature in antimony: ▽ – **K**||Y, **H**||X, $\omega/2\pi$ = 165 MHz; □ – **K**||Y, **H**||Z, $\omega/2\pi$ = 165 MHz; ○ – **K**||Y, **H**||Z, $\omega/2\pi$ = 65.4 MHz; in bismuth: ■ – **K**||Y, **H**||X, $\omega/2\pi$ = 165 MHz, ● – **K**||Y, Z ∥ X, $\omega/2\pi$ = 65.4 MHz.

open triangles, squares, and circles, give the dispersion in antimony. The two lower curves, denoted by black squares and circles, give the dispersion in bismuth. The dispersion curves were measured in a magnetic field H = 2400 Oe. At temperatures less than 4.4—5° K, the velocity dispersion tended to a saturation value determined both by the behavior of $\tau(T)$ and by the effect of electron scattering from impurities and structure defects of the lattice.

Figure 3 shows the experimental results in the coordinates $1/\tau$ and T, T^2 and T^3 for bismuth and antimony. The values of $1/\tau$ were calculated from Eq. (3). The constant A was determined for several values of the temperature at two frequencies and then the mean value was taken. The value of A turned out to be equal to 3.5×10^{-4} for bismuth and 9×10^{-4} for antimony.

It is seen from the drawing that the temperature dependence of the inverse relaxation time is best described by the quadratic function

$$1/\tau = 1/\tau_0 + \beta T^2,$$

and the constant $\beta = 0.57 \times 10^8$ sec^{-1}-deg^{-2} for bismuth and $\beta = 0.15 \times 10^8$ sec^{-1}deg^{-2} for antimony.

DISCUSSION OF THE RESULTS

The magnetic field intensity does not enter directly in Eq. (3) for the sound dispersion in a transverse field, but its role is very important. With increase in field, the twisting radius R of the electrons decreases and, if $R \to 0$, all the electrons on the closed Fermi surfaces, moving along the direction of H, slide along the phase fronts of the wave. Collisionless absorption, which is associated with the Landau damping, is impossible in such a geometry. Collisions with the scatterers lead to viscous absorption and sound velocity dispersion. Here the average work performed by the sound field on the electrons becomes different from zero. For purely deformation interaction, the work performed by a longitudinal sound wave on the electrons is proportional to $|\Lambda_{kk}|^2$—the square of the longitudinal component of the

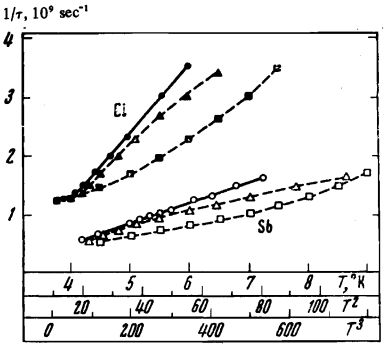

FIG. 3. Inverse relaxation time τ in bismuth and antimony as a function of T (points ■ and □) and T^2 (points ● and ○) and T^3 (points ▲ and △).

deformation potential tensor. The sound velocity dispersion is proportional to this same quantity.

The relaxation time τ, calculated from measurements of the velocity dispersion from Eq. (3) in such multivalley conductors as bismuth and antimony, is a quantity averaged over all equal-energy surfaces. The tensor Λ for bismuth is known.[14] In the set of coordinates connected with the crystal axes, the matrix Λ_{ik} takes the form

$$\Lambda_{ik}^{e}=5.9\ \text{eV}.\begin{vmatrix} -0.37 & 0 & 0 \\ 0 & 1 & 0.25 \\ 0 & 0.25 & -0.29 \end{vmatrix} \qquad (6)$$

for electrons and is diagonal for holes; $\Lambda_{xx}^{h}=\Lambda_{yy}^{h}=1.2$ eV, $\Lambda_{zz}^{h}=1.03\,\Lambda_{yy}^{h}$. We determine the contribution made to the velocity dispersion and to τ by each group of carriers individually. This contribution, as has already been mentioned, is proportional to $|\Lambda_{kk}|^2$. In the case $\mathbf{K}\parallel$ Y, we have

$$\Lambda_{yy}^{(i)}=(\alpha\Lambda\alpha^{-1})_{yy}^{(i)}=\Lambda_{yy}^{(i)}\cos^2\psi+\Lambda_{xx}^{(i)}\sin^2\psi. \qquad (7)$$

Here α is the matrix of rotation about the Z axis, the angle ψ is measured from the Y axis and is equal to zero for the hole and first electron ellipsoids, and $\psi=\pm 120°$ for the turned electron ellipsoids (we use the anisotropic quadratic model of the carrier spectrum in semimetals). Substituting the numerical values of the components, we obtain

$$|\Lambda_{yy}^{h}|^2=1.44\ \text{eV}^2;\quad |\Lambda_{yy}^{Ie}|^2=34.81\ \text{eV}^2;\quad |\Lambda_{yy}^{IIe}|=|\Lambda_{yy}^{IIIe}|^2=0.$$

We can now determine the contribution to the sound velocity dispersion for each equal-energy surface;

$$\left(\frac{\Delta s}{s}\right)_{max}=A=\frac{4\pi\Lambda_{yy}^{2}mp_0}{s^2\rho(2\pi\hbar)^3}, \qquad (8)$$

m is the cyclotron mass, p_0 is the momentum of the electrons from the vicinity of the elliptical limiting point, and ρ is the density. Using the bismuth-spectrum parameters given in[15,16], we obtain the estimates

$$(\Delta s/s)_{max}^{Ie}=3.5\cdot10^{-4},\quad (\Delta s/s)_{max}^{h}=5.8\cdot10^{-6},$$

$$(\Delta s/s)_{max}^{IIe}=(\Delta s/s)_{max}^{IIIe}=0.$$

Thus, $(\Delta s/s)_{max}^{Ie}\gg(\Delta s/s)_{max}^{h}$ and the time τ in bismuth, at $\mathbf{K}\parallel$ Y, is determined by the electrons of a single electron ellipsoid extended along the Y axis.

For antimony, the measurements of the deformation potential tensor are lacking, which makes difficult the exact identification of τ.

In the temperature range studied in semimetals, a whole series of mechanisms are possible which lead to relaxation of the distribution function. The quadratic dependence of the inverse relaxation time on the temperature is typical of processes of electron-electron (or electron-hole) scattering. Along with this, because of the specifically elongated shape of the Fermi surface in bismuth and antimony, such a dependence is possible also for phonons, the limiting frequency of which is equal, for example, to $\omega_0=1.5\times10^{13}$ sec^{-1} for bismuth.[17] It is seen from thermal conductivity data[18] that inelastic scattering of the carriers predominates in bismuth.

	τ_0, sec	β, sec^{-1}·deg^{-2}	Λ_{yy}, eV	Author
Bi	$3.3\cdot10^{-9}$	$0.57\cdot10^{8}$	5.25 *	Present research
	—	$0.77\cdot10^{8}$	—	[9]
	$3.1\cdot10^{-10}$	$2.02\cdot10^{8}$	—	[11]
	—	—	5.9±0.6	[14]
Sb	$3.7\cdot10^{-9}$	$0.15\cdot10^{8}$	—	Present research
	—	$0.22\cdot10^{8}$	—	[10]

*The value of Λ_{yy} was obtained from Eq. (8).

The status of scattering theory at the present time permits us in the best case to determine correctly only the character of the temperature dependence of one process or another. For the determination of the effectiveness of these processes, we require a knowledge of the numerical coefficient in front of the corresponding power of the temperature. Since such calculation does not have the required accuracy, we limit ourselves only to giving the experimental data. It is not possible to determine the predominant mechanism of scattering in semimetals with certainty at the present time.

The table gives the values of the constant β which characterizes the effectiveness of the scattering as determined from measurements of the size effect[9,10] and from a study of the temperature dependence of the amplitude of the cyclotron resonance line,[11] and also the results of the present research.

There is agreement, with good accuracy, between the coefficients β obtained from data on the size effect and those obtained from measurements on the sound velocity dispersion ($\beta_{disp}\approx 0.7\,\beta_{size\ ef}$). Since in the size effect one measures the mean time of flight between collisions, we can assume that the time τ, which is determined from the formula (3), is also the mean time of scattering of the electrons (at least for closed Fermi surfaces). This result, as it appears to us, confirms experimentally the legitimacy of introducing the relaxation time into the theory of sound absorption and of dispersion of its velocity in a strong magnetic field.

In conclusion, we consider it our pleasant duty to thank É. A. Kaner, V. M. Kontorovich and V. L. Fal'ko for useful discussions of the present work.

[1)] A detailed description of the methodology of measurement of the dispersion of the sound velocity, which was used in our experiments, is contained in[13]

[2)] We take this occasion to thank B. A. Merisov for kindly making available his thermocouple and the calibration curves.

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