TEMPERATURE DEPENDENCE OF THE RESISTIVITY AND OF THE HALL COEFFICIENT OF SIZE-QUANTIZED BISMUTH FILMS

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The results are reported of an experimental investigation of the temperature dependence of the resistivity ρ and of the Hall coefficient R_H of Bi films in the temperature range 42--200°K. The $\rho(T)$ dependence is interpreted on the basis of a theory of the quantum size effect. The temperature dependence of the carrier density n(T) is calculated. It is found that if the carrier mobility is assumed to be independent of temperature, the dependence n(T) does not explain all the detailed features of the experimental curve $\sigma(T) = 1/\rho(T)$. Allowance for the temperature dependence of the mobility in the case of scattering by impurities with a δ-shaped potential gives better agreement between $\sigma(T)$ and experiment. The dependences of σ on the film thickness are calculated for various temperatures. The theory predicts stronger damping of the "thickness" oscillations with temperature than the damping observed experimentally.

WE have reported earlier [1,2] the observation of oscillations of the dependence of the resistivity ρ, the Hall coefficient R_H, and the magnetoresistance $\Delta\rho/\rho$ on the thickness of bismuth films. The presence of oscillations and their damping when the temperature and film thickness are increased are interpreted as manifestations of the quantum size effect. The size quantization effect in the spectrum of carriers in Bi has also been observed in tunnel systems, containing size-quantized bismuth films. [3]

Determination of the characteristic features of the temperature dependence of the resistivity and Hall coefficient of size-quantized films is of considerable interest. The present paper reports the results of such a determination.

The film preparation technique and the results of the structural anaylsis have been reported in [1,2]. Measurements of the resistivity and Hall coefficient were carried out by the usual null method. The investigated samples were placed in a helium-cooled Dewar flask, placed between the poles of an electromagnet. After the evaporation of the helium, temperature increased and ρ and R_H were measured. At $T > 30$°K, the rate of change of temperature, $\approx$ 3 deg/min, was controlled with a special heater placed in the helium-cooled Dewar. The temperature of the samples was measured with a platinum thermistor. The accuracy of the temperature measurement was $\approx$1°K.

Before presenting the experimental results and interpreting them, we must recall that bismuth is a semimetal, for which the overlap of the conduction and valence bands is of the order of 30 meV. Moreover, the valence band of bismuth has extrema located 20 meV from the bottom of the conduction band. We can easily show that, up to 200°K, the number of electrons generated at these extrema is slight and, therefore, their contribution to conduction will be ignored (in the 4.2—200°K range).

The main results obtained in the determination of the dependences $\rho_T/\rho_{4.2}$ and $R_H(T)/R_{Hmax}$ are presented in Fig. 1.

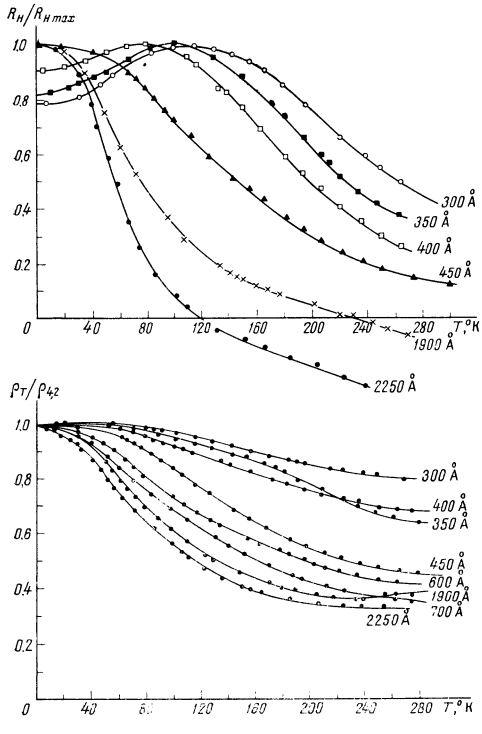

FIG. 1. Plots of $R_H(T)/R_{Hmax}$ and $|\rho_T/\rho_{4.2}$ for Bi films.

We shall consider first the results obtained in the determination of the dependence $R_H(T)/R_{Hmax}$. When temperature is increased the Hall coefficient of films thicker than 1000 Å decreases at first, passes through zero at some temperature, and then varies slowly with temperature. The thinner the film the higher is the temperature at which R_H passes through zero. In the thickness range $d < 1000$ Å, the Hall coefficient does not change its sign when temperature is varied in the range 4.2—300°K: it remains positive in this range (this indicates p-type conduction; bulk samples of the

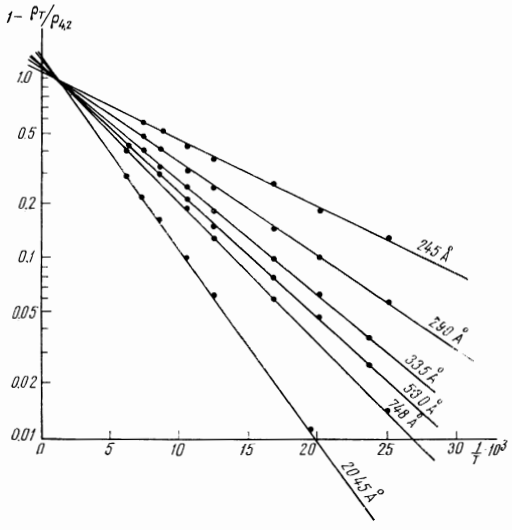

FIG. 2. $(1 - \rho_T/\rho_{4.2}) = f(1/T)$ plotted on logarithmic scale for Bi films.

same orientation have n-type conduction). The thinner the film, the higher is the position of the $R_H(T)/R_{Hmax}$ curve. The $R_H(T)/R_{Hmax}$ curve of films thinner than 400 Å has a maximum. The temperature at which this maximum is observed increases when the thickness is reduced.

The observed positive sign of the Hall coefficient of Bi films may be explained by the presence of local electron acceptor states, which are filled at low temperatures. In this case, conduction by holes is somewhat stronger and this gives rise to a positive Hall coefficient. When the temperature is raised, the number of electrons and holes increases. However, since the electron mobility is greater than the hole mobility, the Hall coefficient should change its sign at some temperature. In thinner films, the density of local states is higher and, therefore, the change of sign of R_H takes place at higher temperatures (this change of sign of R_H may not be observed at all for thin films in the 4.2–300°K range). The presence of a maximum in the $R_H(T)/R_{Hmax}$ dependence of films whose thicknesses are $d \leq 400$ Å cannot yet be interpreted satisfactorily.

The $\rho_T/\rho_{4.2}$ dependence has the following features: when the temperature is increased, the value of $\rho_T/\rho_{4.2}$ decreases monotonically and saturation is observed at $T < 60°K$ and $T > 200°K$.

The effect of the film thickness on the dependence $\rho_T/\rho_{4.2}$ becomes clear if we plot the experimental curves using the coordinates

$$\ln\left(1-\frac{\rho_T}{\rho_{4.2}}\right)=f\left(\frac{1}{T}\right).$$

Figure 2 shows these dependences for several thicknesses. It is evident from this figure that the dependences are linear and intersect at one point. The slopes of the straight lines depend on the film thickness.

Figure 3 shows the dependence of the tangent of the slope angle α of these straight lines (in relative units) on the film thickness. The characteristic feature of this dependence is the presence of oscillations which are damped by an increase of the film thickness. The distance Δd between the minima increases somewhat when

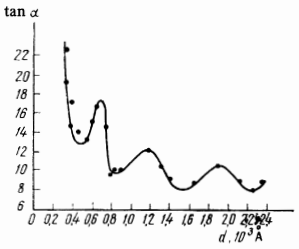

FIG. 3. Slope (in relative units) of straight lines $\ln(1 - \rho_T/\rho_{4.2}) = f(1/T)$ vs. the thickness of Bi films.

the thickness is increased ($\Delta d \approx 400$ Å for thin films). The value of tan α in the region $d < 500$ Å increases rapidly when the thickness is reduced.

The presence of oscillations in the tan $\alpha = f(d)$ curve, the damping of these oscillations when the thickness is increased and the identity of the oscillation period of tan $\alpha = f(d)$ with the period of quantum oscillations of $\rho(d)$, reported in [1,2], may all be attributed, with a fair certainty, to the size quantization of the energy spectrum of carriers in bismuth films. We shall now give a more detailed interpretation of the dependence $\rho(T)$ using the theory of quantum size effects proposed in [4].

We shall attempt to explain the observed dependences $\rho(T) = 1/\rho(T)$ by assuming that only the carrier density is affected by temperature while the mobility remains constant. This may happen, for example, when the mean free path is limited by the average size of crystallites and remains less than the lattice mean free path in the investigated range of temperatures.

The expression for the electron density under quantum size effect conditions has the following form for one minimum: [4]

$$n(T)=\gamma\sum_{s=1}^{\infty}\ln\left(1+\exp\frac{F_n-E_0s^2}{kT}\right),$$
$$\gamma=\frac{m_nkT}{\pi\hbar^2d}, \qquad (1) \qquad (1)$$

where F_n is the Fermi level for electrons, measured from the bottom of the conduction band of a bulk sample; E_0 is the zero energy for electrons; m_n is the effective density-of-states mass; k is Boltzmann's constant.

To calculate the dependence n(T), it is necessary to know the function $F_n(T)$, which is found from the electrical neutrality equation n = p (p is the density of holes). We shall carry out a calculation for the band scheme of Bi (three minima in the conduction band and one maximum in the valence band). The components of the effective masses of electrons and holes are assumed to be:

$$m_{n1}=0{,}007m_0, \quad m_{n2}=1{,}8m_0, \quad m_{n3}=0{,}02m_0,$$
$$m_{p1}=m_{p2}=0{,}007m_0, \quad m_{p3}=m_0$$

(the subscripts represent, respectively, the binary, bisector, and trigonal axes). The band overlap is assumed to be $\Delta = 0.030$ eV.

Since the experimentally observed direction of "quantization" coincides with the trigonal axis, we may assume that only electrons are quantized, because $m_{n3} \ll m_0$ and $m_{p3} \approx m_0$. Then, the expression for p has the form

$$p=N_vF_{1/2}\left(\frac{\Delta-F_n}{kT}\right), \qquad (2)$$

where N_V is the effective density of states in the valence band and $F_{1/2}$ is a Fermi integral.

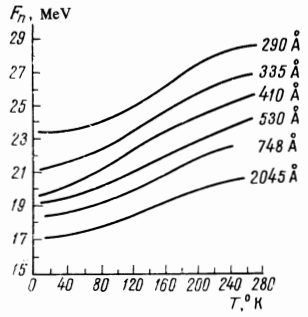

FIG. 4. Plot of $F_n(T, d)$.

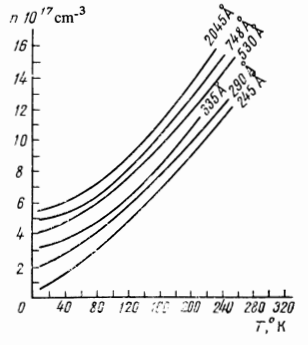

FIG. 5. Plot of n(T, d).

A numerical solution of the equation n = p gives the dependences $F_n(T, d)$ shown in Fig. 4. Then, using Eq. (1), we plot the n(T, d) curves shown in Fig. 5. For comparison with experiment, Fig. 6 gives the dependences

$$\ln\left(1-\frac{n_{20}}{n_T}\right)=f\left(\frac{1}{T}\right)$$

(it has not been possible to carry out calculations for $T < 20^\circ K$ because of the absence of suitable tables of Fermi integrals). We can see that, in the temperature range 40–150°K, these dependences are approximated well by straight lines with a slope angle depending on the film thickness. To compare the dependence of this angle on the thickness with the experimental data, we plot tan δ = f(d) as shown in Fig. 7. Comparison of the curve in Fig. 7 with the experimental curve in Fig. 3 shows that, in spite of the great similarity (the presence of oscillations, the damping of oscillations when the thickness is increased, and the good agreement between the absolute values), there are also differences between these two curves.

First of all, the amplitude of the oscillations of the experimental curve is greater than the theoretical amplitude and, secondly, at low values of d the theoretical tan α curve decreases rapidly when d is increased, while the experimental curve shows a rise. It must be mentioned that a correct interpretation of the results for films of thicknesses $d < 400$ Å requires a detailed consideration of the lifting of the band overlap; on the other hand, the quality of such thin films is much poorer. Therefore, a comparison of the experimental and theoretical curves in this range of thicknesses should be carried out with caution.

We shall now consider the dependence σ(T, d) for the case of scattering by impurities with δ-shaped potentials. If we use this model, the mobility is found to depend on temperature because the carrier relaxation time depends on the carrier energy; when the temperature is increased, carriers are redistributed between different sub-bands.

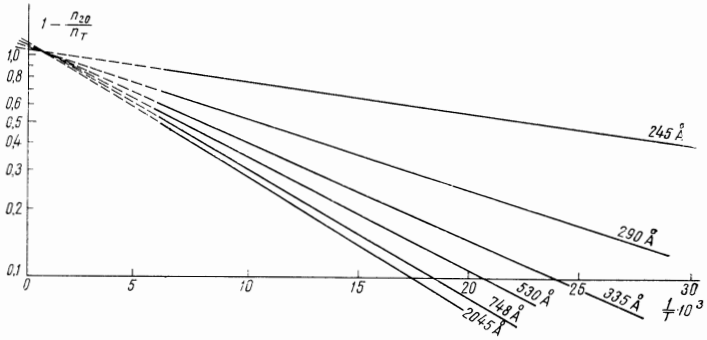

FIG. 6. Plot of $(1 - n_{20}/n_T)$ vs. 1/T on logarithmic scale.

The expression for the contribution of electrons in one minimum to the electrical conductivity has the form [4]

$$\sigma=\gamma_1\sum_{s,\,\mathbf{q}}k_z^2\frac{\partial f_0/\partial E}{[\sqrt{E/E_0}]+1/2},\qquad(3)$$

$$\mathbf{q}=(k_y,\ k_z),\quad s=1,\ 2,\ 3,\ldots,$$

where γ_1 is independent of temperature; $f_0(E)$ is the Fermi distribution function; k_Z is the crystal momentum along the direction of the current in the plane of the film; [...] is taken to the nearest integer.

This formula can be reduced by certain transformations to a form convenient for the calculation of σ(T, d):

$$\sigma=\gamma_2\sum_{s=1}^{\infty}\frac{1}{4s^2-1}\left\{kT\ln\left(1+\exp\frac{F_n-s^2E_0}{kT}\right)\right.$$

$$\left.-\frac{E_0}{3}(s-1)s(4s+1)\left(1+\exp\frac{s^2E_0-F_n}{kT}\right)^{-1}\right\},\qquad(4)$$

where γ_2 is independent of temperature.

We used this formula to plot the dependences tan α = f(d) shown in Fig. 8. We can see that the curve in Fig. 8 agrees better with experiment than the curve in Fig. 7.

Next, using Eq. (4) we plotted the dependences σ(d) for T = const (Fig. 9). These curves should be compared with the experimental dependences reported in [1,2].

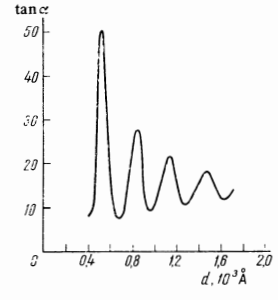

FIG. 8. Plot of tan α vs. d [Eq. (4)].

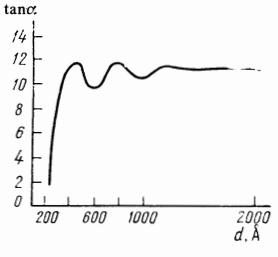

FIG. 7. Plot of tan α vs. d [Eq. (1)].

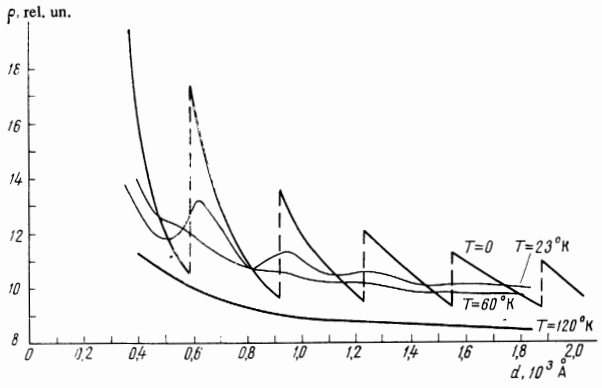

FIG. 9. Plots of $\rho(d, T) = 1/\sigma(d, T)$.

Such a comparison shows that the theoretical and experimental curves are very similar. However, there is one definite difference between them: when the temperature is increased oscillations of the theoretical curves are damped out more rapidly than those of the experimental ones.

This comparison indicates that the experimental dependences $\sigma(T, d)$ for Bi films can be explained on the basis of the quantum size effect.

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138