Resistivity and transverse magnetoresistance in ultrathin films of pure bismuth

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(Received 24 February 1987; revised manuscript received 16 July 1987)

Electrical resistivity and transverse magnetoresistance were measured at liquid-helium temperatures in thin films of pure bismuth ranging from 101 to 4504 Å in magnetic fields up to a maximum of 80 kG. The films could be divided into two groups, above and below a thickness of about 250 Å. The resistivity (in zero magnetic field) and the magnetoresistance were found to be drastically different, both in magnitude and variation as a function of field strength, for the two groups. Interpretation is given in terms of a transition, due to the quantum size effect, of the charge carriers from states of three-dimensional motion in the thicker films to states of two-dimensional motion in the thinner films.

I. INTRODUCTION

Because of the small effective masses and the low Fermi energy of electrons and holes in bismuth,[1] this material has long been used in the experimental study of quantum effects on the electronic properties of crystals. Thin films of bismuth of large thickness naturally possess energy-band structures similar to those of the bulk crystal. However, for thin films of thicknesses up to several thousand angstroms, the electron localization effect[2] must be taken into account to explain features like the anomalous temperature dependence of the resistivity.[3] Therefore interesting questions are whether bismuth films preserve bulk band structure for ultrathin films where the electron localizations are more likely to be realized, and whether the quantum effects associated with the electronic band structure can still be observed.

Assuming a semimetallic energy-band structure in ultrathin bismuth films similar to that in bulk bismuth, simple calculations regarding the quantum size effect predict that there would be either a semimetallic-semiconducting state transition or a state transition from the three-dimensional motion of the charge carriers to two-dimensional motion, for sufficiently thin films, depending on the appropriate use of a boundary condition.

This work reports the results of a systematic measurement of electrical resistivity and transverse magnetoresistance in bismuth films as a function of thickness, magnetic field, and temperature for film thicknesses from 101 to 4504 Å, with magnetic fields up to 80 kG, and temperatures from 4.2 to 1.5 K. In addition to studies on the line shape of the magnetoresistance and the temperature dependences of the resistivity and magnetoresistance, the prominent changes in these quantities which are believed to be the result of a state transition from three-dimensional to two-dimensional motion of the electrons and holes will be emphasized. No previous analysis of this kind has been reported, and no similar experimental work has been done for such small thicknesses[4] in high magnetic fields.[5]

II. EXPERIMENT

Thin films of high-purity (99.9999%) bismuth were fabricated by thermal evaporation onto heated mica substrates at pressure less than 2×10^{-7} Torr throughout the evaporation process. The evaporation source was a tungsten dimple boat, and it was found that the evaporation rate did not affect epitaxial growth as long as the substrate temperature was kept above 370 K. The thickness of the 1-mm-wide films was monitored with a quartz crystal microbalance with an error generally less than ±1.5 Å. After completion, the films were stored in a dry nitrogen atmosphere and were annealed in vacuum at 425 K for one hour or more.

Electrical measurements were made over a 1 cm length of the film which was placed in the magnetic center, and perpendicular to the axis, of a 35.2-mm bore superconducting solenoid with a field homogeneity greater than 0.1% over the diameter of the bore. Magnetoresistance was measured at constant current in a bridge configuration, as shown in Fig. 1. With the bridge balanced in zero field, the output of the nanovoltmeter is proportional to the change in the resistance of the sample

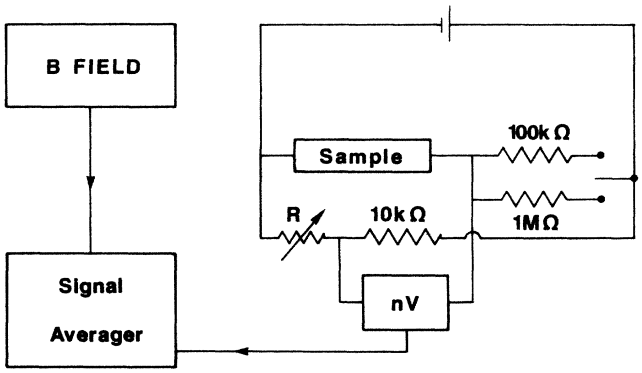

FIG. 1. Schematic diagram for the recording of the magnetoresistance in thin films of pure bismuth.

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due to the transverse magnetic field. Resistivity as a function of temperature at constant fields was measured with a digital ohmmeter, and the temperature was measured with a carbon glass resistance thermometer of negligible magnetic field dependence. Crystallinity of several randomly selected samples was verified by observing the hexagonal symmetry in the diffraction pattern from a transmission electron microscope. This pattern is a result of the growth of the trigonal crystalline axis perpendicular to the plane of the substrate.

III. ENERGY-BAND STRUCTURE IN BISMUTH

A. Energy levels in a three-dimensionally quantized film

The energy-band structure in bismuth consists of three conduction bands (electrons) and one valence band (holes). The electron bands are nonparabolic ellipsoids, while the hole band is parabolic and ellipsoidal. The three electron ellipsoids are symmetrically located about the trigonal axis of the crystal and the single hole ellipsoid has rotational symmetry about the trigonal axis.

By denoting the crystal principal axes (the binary, the bisectrix, and the trigonal) by the x, y, and z axes, respectively, the effective-mass tensor[1] of the electron ellipsoid consists of three diagonal elements m_{11}, m_{22}, and m_{33}, and two off diagonal elements $m_{23}=m_{32}$. The other two electron ellipsoids are obtained by rotating the first one about the trigonal axis through $\pm 120°$. The hole effective-mass tensor consists of diagonal elements only: $M_{11}=M_{22}$ and M_{33}.

The energy levels of the electrons and holes in a bismuth thin film placed in a transverse magnetic field B assume the following forms:

$$E_n(1+E_n/E_g)=(n+\tfrac{1}{2})\hbar\frac{eB}{m_c c}\pm\tfrac{1}{2}\hbar\frac{eB}{m_s c}+\varepsilon_{0n}L_n^2$$

for electrons , (1)

$$E_p=(N+\tfrac{1}{2})\hbar\frac{eB}{M_c c}\pm\tfrac{1}{2}\hbar\frac{eB}{M_s c}+\varepsilon_{0p}L_p^2 \quad \text{for holes} , \tag{2}$$

where E_g is the energy gap between the conduction band and the valence band directly beneath it; m_c and M_c are the orbital cyclotron effective masses; m_s and M_s the spin effective masses; ε_{0n} and ε_{0p} are the energy quantizations due to the size effect; n and N are $0,1,2,\ldots$; L_n and L_p are $1,2,3,\ldots$ if vanishing wave-function boundary conditions are applied; and L_n and L_p are $0,1,2,\ldots$ if the vanishing gradient of the wave function at the boundaries is applied. The left-hand side of Eq. (1) indicates the nonparabolic but ellipsoidal energy-momentum relationship for electrons. The electron energies E_n and the hole energies E_p in Eqs. (1) and (2) are relative to the energy-band edge of the conduction band and that of the overlapping valence band, respectively. The energy separation between the two edges or the energy overlap E_0 is assumed to be constant.

Assuming the direction of the magnetic field (perpendicular to the film plane) is $\mathbf{h}=\mathbf{i}\alpha+\mathbf{j}\beta+\mathbf{k}\gamma$ relative to the binary-bisectrix-trigonal coordinates, the effective masses in (1) and (2) can then be given by

$$m_c=(\det\overleftrightarrow{m}/m_h)^{1/2},\quad M_c=(\det\overleftrightarrow{M}/M_h)^{1/2} , \tag{3}$$

where $\overleftrightarrow{m}$ and $\overleftrightarrow{M}$ are the orbital-effective-mass tensors of electrons and holes respectively, and m_h and M_h are the longitudinal effective masses, i.e.,

$$m_h=\mathbf{h}\cdot\overleftrightarrow{m}\cdot\mathbf{h},\quad M_h=\mathbf{h}\cdot\overleftrightarrow{M}\cdot\mathbf{h} . \tag{4}$$

The longitudinal effective masses appear also in ε_{0n} and ε_{0p}:

$$\varepsilon_{0n}=\pi^2\hbar^2/2m_h d^2,\quad \varepsilon_{0p}=\pi^2\hbar^2/2M_h d^2 , \tag{5}$$

where d is the film thickness. There are similar equations for the spin effective masses, i.e.,

$$m_s=(\det\overleftrightarrow{m}_s/\mathbf{h}\cdot\overleftrightarrow{m}_s\cdot\mathbf{h})^{1/2} ,$$
$$M_s=(\det\overleftrightarrow{M}_s/\mathbf{h}\cdot\overleftrightarrow{M}_s\cdot\mathbf{h})^{1/2} , \tag{6}$$

where $\overleftrightarrow{m}_s$ and $\overleftrightarrow{M}_s$ are the spin-effective-mass tensors.

The direction of the magnetic field relative to the other two energy ellipsoids of electrons can be obtained by rotating $\mathbf{h}$ about the trigonal axis for $\pm 120°$. Thus $\mathbf{h}(\alpha,\beta,\gamma)\rightarrow\mathbf{h}'(\alpha',\beta',\gamma')$, where

$$\alpha'=-\tfrac{1}{2}\alpha\mp\frac{\sqrt{3}}{2}\beta,\quad \beta'=\pm\frac{\sqrt{3}}{2}\alpha-\tfrac{1}{2}\beta,\quad \gamma'=\gamma . \tag{7}$$

Replacing $\mathbf{h}$ by $\mathbf{h}'$ in Eq. (3)–(6), values of m_c, M_c, etc. for the other two electron bands can be evaluated.

Using numerical data from Ref. 1 for the effective-mass tensors, we can calculate the effective masses and thus the

TABLE I. Effective masses in pure bismuth (in units of m_0, the free-electron mass) calculated from the effective mass tensors in Ref. 1.

Electrons	Holes
h parallel to trigonal axis	
$m_c=0.0140$	$M_c=0.064$
$m_s=0.0239$	$M_s=0.033$
$m_h=0.00443$	$M_h=0.69$
h parallel to binary axis	
$m_c{}^{a}=0.0277$	$M_c=0.21$
$m_s{}^{a}=0.0788$	$M_s=2.57$
$m_h{}^{a}=0.00113$	$M_h=0.064$
$m_c{}^{b}=0.00211$	
$m_s{}^{b}=0.00198$	
$m_h{}^{b}=0.195$	
h parallel to bisectrix axis	
$m_c{}^{a}=0.00183$	$M_c=0.21$
$m_s{}^{a}=0.00172$	$M_s=2.57$
$m_h{}^{a}=0.26$	$M_h=0.064$
$m_c{}^{b}=0.00363$	
$m_s{}^{b}=0.00344$	
$m_h{}^{b}=0.0658$	

[a]Single ellipsoid, magnetic field parallel to binary/bisectrix.
[b]Double ellipsoids, magnetic field making $\pm 120°$ with binary/bisectrix.

energy levels in terms of the film thickness and the magnetic field intensity for a given orientation (**h** and **h'**) of the film. For **h** along one of the crystallographic axes, the numerical values of the effective masses to be used in Eqs. (1) and (2) are given in Table I.

B. Three-dimensional to two-dimensional transition

As the film thickness is reduced, the quantum size effect becomes more pronounced and an appropriate boundary condition is necessary in the determination of the size-quantized energy levels of the charge carriers. In reality, the boundary conditions in thin films may be quite complicated. Paskin and Singh[6] have discussed the application of the boundary conditions and concluded that the vanishing wave function may be a better approximation for a perfect metal surface while the vanishing gradient may be better for rough surfaces. The vanishing gradient boundary conditions will be assumed in the following discussion.

At absolute zero, energy levels beyond the Fermi level are unoccupied and the number of occupied levels $L_n=0,1,2,\ldots$ and $L_p=0,1,2,\ldots$ decreases with the film thickness. When the film is sufficiently thin, it may occur that $L_n=0$ and $L_p=0$ are the only occupied longitudinal levels of electrons and holes, respectively. Since the levels of $L_n=0$ and $L_p=0$ correspond to zero longitudinal energies, the motion of the charge carriers may be considered to have become two dimensional. The three-dimensional to two-dimensional transition can be obtained by setting $n=N=0$, $s=S=-1$, and $L_n=L_p=1$ in Eqs. (1) and (2), and using $E_n+E_p=E_0$ (the energy overlap).

In zero magnetic field [$B=0$ in Eqs. (1) and (2)], the electron level E_n for $L_n=1$ can be solved from Eq. (1), while for the hole level, $E_p=\varepsilon_{op}$. Using $E_n+E_p=E_o$, the film thickness d at which the three-dimensional to two-dimensional transition takes place can be evaluated. The numerical results for films perpendicular to the three crystallographic axes are $d=2.55\times10^2$ Å, film perpendicular to trigonal axis; $d=1.37\times10^2$ Å, film perpendicular to binary axis; and $d=1.34\times10^2$ Å, film perpendicular to bisectrix axis.

When a magnetic field B is applied along the trigonal axis perpendicular to the film, the lowest energy levels corresponding to $L_n=L_p=1$ in Eqs. (1) and (2) are

$$E_n=-\frac{E_g}{2}+\left[\frac{E_g^2}{4}+E_g\mu_B B\left(\frac{1}{m_c}-\frac{1}{m_s}\right)+E_g\frac{\pi^2\hbar^2}{2m_0m_hd^2}\right]^{1/2}\quad\text{for electrons},$$

$$E_p=\mu_B B\left(\frac{1}{M_c}-\frac{1}{M_s}\right)+\frac{\pi^2\hbar^2}{2m_0M_hd^2}\quad\text{for holes},$$

where m_0 is the free-electron mass and μ_B is the Bohr magneton and the numerical values of the effective masses are given in Table I. It was shown in the previous paragraph that the crossover of these two energy levels would occur at $d=255$ Å in zero magnetic field. For $d>255$ Å, both levels remain within the Fermi level and the electrons and holes have three-dimensional motion. For $d<255$ Å, however, the charge carriers would move two dimensionally in zero magnetic field (and at low temperatures) since the hole level would be lower than the electron level, taking the conduction-band bottom as reference, and both levels would be unoccupied at zero temperature. As a magnetic field is applied to the films, the hole level would move upward relative to the electron level and would overtake the electron level at certain field intensity, at which the charge carriers would resume three-dimensional motion. Figure 2 shows a transition

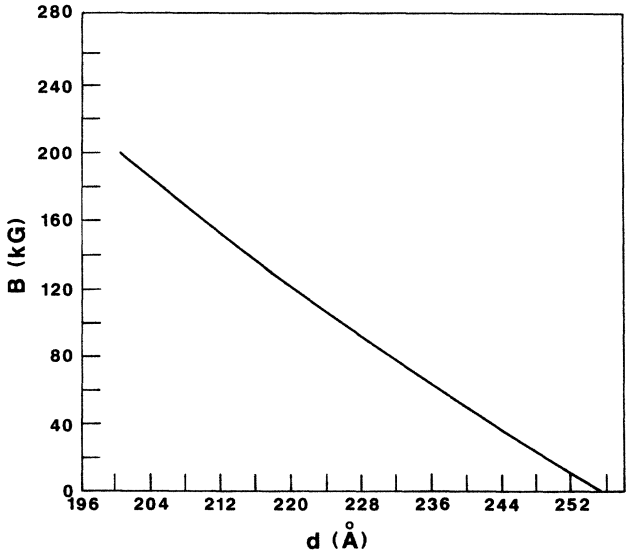

FIG. 2. The predicted transition from two-dimensional to three-dimensional motion in bismuth thin films perpendicular to the trigonal axis.

TABLE II. Resistivity and magnetoresistance for various film thicknesses.

Thickness (Å)	Resistivity in zero field (10^{-3} Ω cm)	Magnetoresistance at $B=78$ kG
101	0.417 53	0.069 71
125	0.395 71	0.131 12
151	0.375 43	0.134 93
200	0.447 86	0.213 65
235	0.692 25	0.210 11
239	0.551 84	0.226 46
245	0.566 09	0.263 43
247	0.613 41	0.240 33
250	0.595 12	0.261 54
254	0.546 91	0.300 14
332	0.759 62	0.381 38
352	0.626 22	0.543 54
399	0.669 60	0.440 60
499	0.675 12	0.945 91
750	0.618 73	2.232 40
1002	0.567 13	3.313 11
1502	0.549 63	4.990 40
4504	1.393 36	7.089 12

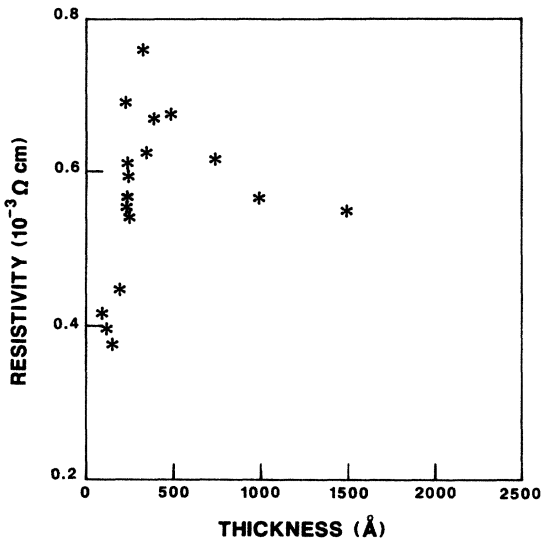

FIG. 3. Electrical resistivity in bismuth thin films at 4.2 K (zero magnetic field).

curve in the *B-d* plane. To the right of the curve is the region for the three-dimensional motion and to the left the two-dimensional motion. For fields parallel to the binary or bisectrix axis, similar transition curves can be obtained.

IV. RESULTS AND DISCUSSION

Both the electric resistivity in zero magnetic field and the magnetoresistance in a field of 78 kG for films of various thicknesses ($T=4.2$ K and film planes perpendicular to the trigonal axis) are presented in Table II. These data are also plotted in Figs. 3 and 4. The magnetoresistances as a function of the (transverse) magnetic field for several of the film thicknesses are shown in Figs. 5 and 6.

Figure 3 shows the electrical resistivity as a function of the film thickness. The decrease in resistivity with thickness from 4504 to 1502 Å is generally consistent with a

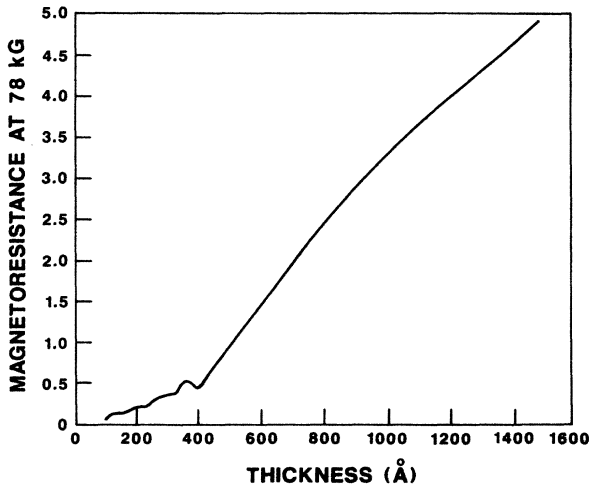

FIG. 4. Magnetoresistance in bismuth thin films at 4.2 K.

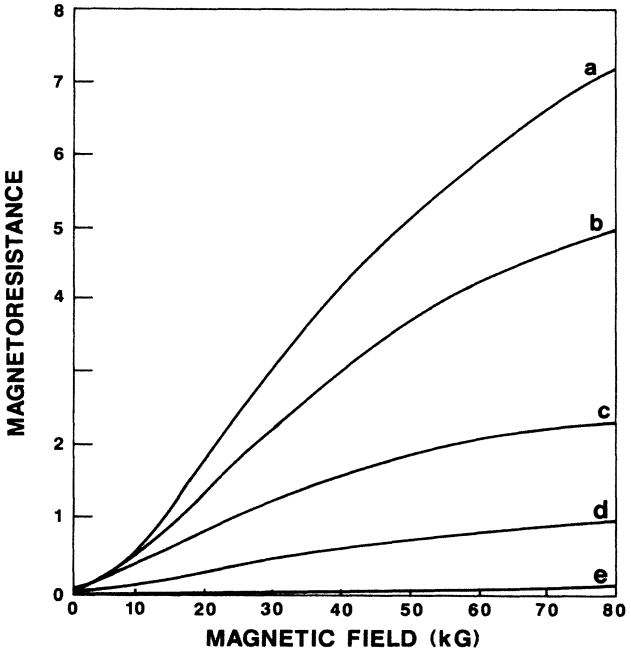

FIG. 5. Magnetoresistance vs magnetic field at 4.2 K. Curve *a*, film of thickness 4504 Å; curve *b*, 1052 Å; curve *c*, 750 Å; curve *d*, 499 Å; and curve *e*, 101 Å.

previous report[3] and is not shown in the figure. Below 1502 Å the resistivity first increases and exhibits some oscillatory variations before it drops down appreciably with (decreasing) thickness, in contrast to the result in Ref. 3, in which a monotonic decrease of the resistivity was reported. The magnitudes of the resistivities in the two works seem to be reasonably close and the overall trend of our data also shows a decreasing resistivity with thinner films. However, in good single crystalline bismuth films, thickness-dependent oscillations in the resistivity due to the quantum size effect ought to be observed. In this work, a large number of good single-crystal films were made for thicknesses below 1000 Å for the purpose of observing the quantum size effect. It can be seen from Fig. 3 and Table II, that (with thickness decreasing from 1502 Å) the resistivity reaches a maximum at about 500 Å and then oscillates a couple of times before undergoing another prominent oscillation in the vicinity of 240 Å. These oscillations correspond to the quantum size effect at the characteristic thicknesses for pure bismuth of about 500 and 250 Å. According to Sec. III, the charge carriers may occupy only the lowest of the size-quantized longitudinal energy levels in films of thickness less than about 250 Å, i.e., $L_n=L_p=0$ only. For thicknesses above about 500 Å, the occupied levels may include $L_n=0,1,2$ and $L_p=0,1,2$; but for those below 500 Å, only $L_n=0,1$ and $L_p=0,1$. Whenever the number of occupied levels L_n or L_p suffers a sudden decrease with decreasing film thickness, the densities of electrons and holes experience a minimum, marking a peak in the resistivity. If the film could be fabricated exactly perpendicular to the trigonal axis, then there would be a clear and single peak in the vicinity of about 500 Å. But if the orientation is not exact, then very likely a doublet or trip-

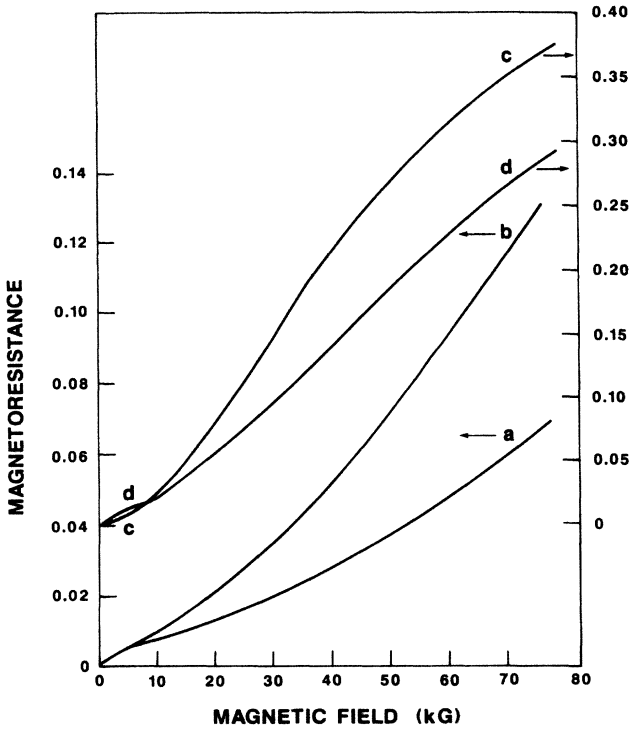

FIG. 6. Magnetoresistance versus magnetic field at 4.2 K. Curve *a*, film thickness 101 Å; curve *b*, 151 Å; curve *c*, 332 Å; and curve *d*, 254 Å.

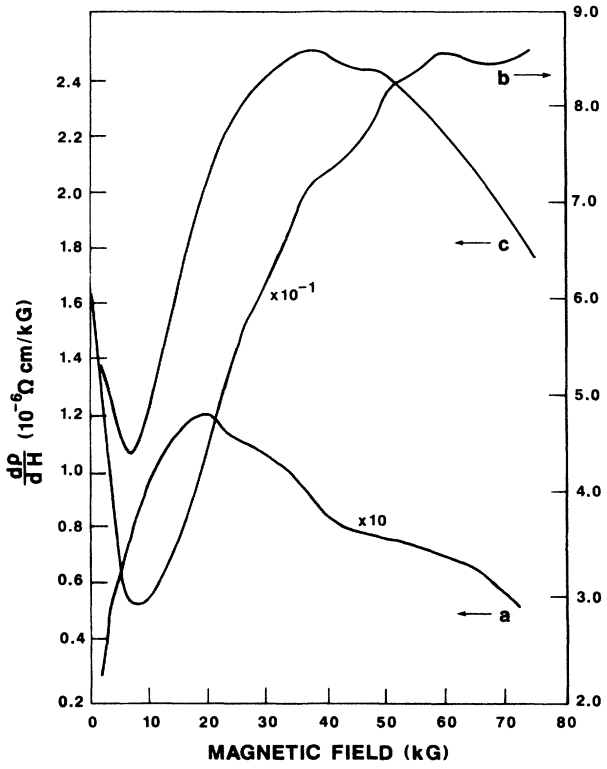

FIG. 7. First-order derivative of magnetoresistance vs magnetic field (4.2 K). Curve *a*, film thickness is 499 Å; curve *b*, film thickness is 125 Å; and curve *c*, film thickness is 254 Å.

let structure in the peak would be observed since there are three electron energy ellipsoids symmetrically located with respect to the trigonal axis. A fairly drastic drop in resistivity was observed in films of 200, 151, 125, and 101 Å, following the resistivity peak at 235 Å. This quantum oscillation is believed to be the result of the transition of charge carriers from occupying the $L_n=0,1$ and $L_p=0,1$ levels to occupying $L_n=0$ and $L_p=0$ levels only.

In Fig. 4 (also see Table II), besides certain oscillations due to quantum size effects, the general shape of the magnetoresistance versus thickness consists of roughly four regions: that for thicknesses below approximately 250 Å having a slow rate of increase, that between 250 and 500 Å having slightly higher rate of increase, that between 500 and 1500 Å having a much steeper slope, and that beyond 1500 Å (not shown) having a slower rate of increase indicating the tendency to approach to bulk limit. This classification of the film thicknesses into several regions is also supported by examining the magnitudes of the magnetoresistance as given in Table II.

In Figs. 5 and 6, the line shapes of the magnetoresistance as a function of the applied magnetic field are shown for some of the films. In the group of films thicker than about 250 Å, the field dependence of magnetoresistance behaves similarly to that in bulk crystals; first a generally quadratic increase with the magnetic field in weak fields, followed by a decrease in slope and a saturation tendency in higher fields. The magnetoresistance in the film group of thickness less than about 250 Å, however, is significantly different. For weak fields these curves are concave downward instead of upwards, as can be seen in Fig. 6. In the field range of 10–20 kG these curves decrease in slope and thereafter the magnetoresistance increases almost linearly with the field up to our maximum field of 80 kG. In Fig. 6, a comparison is also provided of the two different types of the line shape of magnetoresistance.

It was shown in Sec. III that the charge carriers assume three-dimensional motions in the thicker films and two-dimensional motions in the thinner films. Once the electrons and holes are confined to the two-dimensional states, the state transition probabilities of the charge carriers during a scattering process decrease significantly, causing a reduction in the resistivity. In fact, the two-dimensional motion must have altered the scattering mechanism from that of the bulk crystal and thicker films, where large magnetoresistances are common. Numerical calculations indicate that the characteristic thickness for the three-dimensional to two-dimensional transition is about 250 Å for bismuth films perpendicular to the trigonal axis, and that in films in the range of about 250 and 500 Å the electrons and holes perform "quasi" two-dimensional motions, since there is only one occupied longitudinal level ($L_n=1$ or $L_p=1$) in this range for either electrons or holes in addition to the levels $L_n=0$ and $L_p=0$, respectively. This theoretical prediction explains the significantly different line shapes of the magnetoresistance versus applied field in the two thickness groups (above and below about 250 Å), the significantly larger slope beyond roughly 500 Å in Fig. 4, the prominent quantum oscillations in the resistivity versus thick-

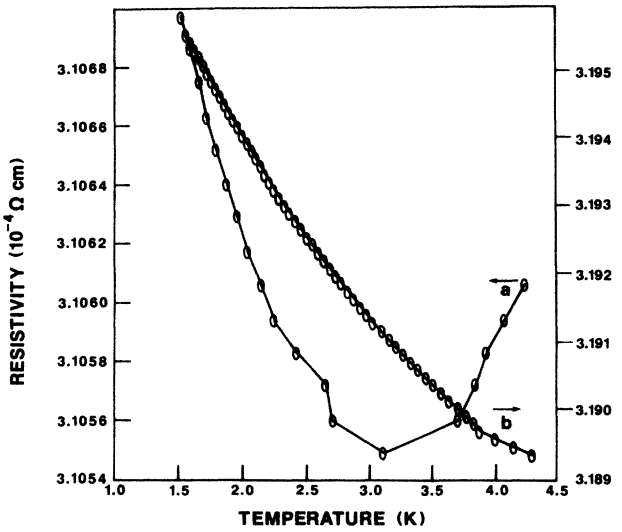

FIG. 8. Electrical resistivity vs temperature. Curve *a*, film thickness is 150 Å, zero magnetic field; and curve *b*, 150 Å, 20 kG.

ness in the vicinities of 500 and 250 Å, and the much smaller resistivity and magnetoresistance in the ultrathin films where the charge carriers perform two-dimensional motions. It should be emphasized that such results are not due to the electron localization effect in thin films. The anomalous temperature dependence of the resistance, which serves as a measure of the electron localizations, was consistently observed for all the films ranging from 101 to 4504 Å. To reinforce this support, the numerical calculations in Sec. III also indicated that a film with carriers performing two-dimensional motion in zero magnetic field may undergo a transition to performing three-dimensional motion in a sufficiently high magnetic field. In this respect, note the line shape of the 254-Å film in Fig. 6, where the low-field behavior resembles that of the group having two-dimensional motion and the high-field behavior resembles that of the group of three-dimensional motion. Figure 7 shows typical first-order derivatives of the magnetoresistance for both the thicker and the thinner groups. Also shown in Fig. 7 is the interesting result of the first-order derivative of the 254-Å film which consists of the feature of the thinner group in the low-field region and the feature of the thicker group in the high-field region. Again, the fairly good numerical coincidence between the calculations in Sec. III and our experimental results indicates not only that the electronic energy-band structures in bismuth remain somewhat similar even in films as thin as 100 Å, provided that they preserve good single crystallinity through careful fabrication, but also that the numerical values of the band parameters (effective masses) may not undergo appreciable variations while reducing the thickness of the film.

In the temperature range of 1.5 to 4.2 K, the electric resistivity in films of thickness greater than 250 Å typically decreases with an increasing temperature. This is the case either in zero magnetic field or in a magnetic field up to 80 kG. For films of thickness less than 250 Å, the temperature dependence shows a similar anomaly except that in zero magnetic field the resistivity minima are located somewhere between 3 and 4 K. The minima obviously shift to higher temperatures beyond 4.2 K when a magnetic field is applied. The resistivity versus temperature for a 150-Å film in 0 and 20 kG fields are shown in Fig. 8. The thermal energy needed to liberate a localized electron seems to increase slightly when a magnetic field is applied perpendicular to a thin film.

V. CONCLUSION

It is concluded, based on the experimental results (Sec. IV) and the calculations (Sec. III), that bismuth thin films preserve reasonably well an energy-band structure similar to that in the bulk crystal. The band parameters do not vary appreciably in thin films with the decreasing thickness. An extreme quantum size effect can be observed in films of thickness below 500 Å. No semimetallic-semiconducting state transition has been observed.[7,8] It is inappropriate, therefore, to apply the boundary condition of vanishing wave function in the study of the size quantizations in bismuth thin films. With the application of the vanishing gradient boundary condition, the quantum size effect makes it possible to divide the bismuth thin films into a "thicker" group in which the charge carriers perform three-dimensional motion and a "thinner" group of two-dimensional motion. Experimental study has observed the division of the two groups from a thickness of about 250 Å.

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