# TEMPERATURE DEPENDENCE OF THE MAGNETORESISTANCE ANISOTROPY OF ANTIMONY

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The anisotropy of the magnetoresistance of antimony is measured in transverse magnetic fields up to 80 kOe at  $77^{\circ}$ ,  $20.4^{\circ}$ , and  $4.2^{\circ}$ K. In high fields the anisotropy is found to be strongly temperature dependent. This effect is related to the temperature dependence of Fermi-surface anisotropy in antimony.

## INTRODUCTION

**A**NTIMONY is a semimetal with a rhombohedral crystal structure, <sup>[1]</sup> which contains  $\sim 10^{-3}$  charge carriers per atom. Investigations of the kind of energy spectrum that is common to semimetals of this type have shown that the Fermi surface regions containing electrons are located along the binary axes of the Brillouin zone, while the hole region is at the center of the zone. <sup>[2]</sup> The electron part of the Fermi surface of antimony is represented by a system of three (or six) ellipsoids, one of which is along a binary axis while another forms an angle  $\theta = 36^{\circ}$  with the trigonal axis. <sup>[3]</sup> The form of the hole surface has not yet been determined. Data on cyclotron resonance and oscillatory effects <sup>[4-8]</sup> can apparently be accounted for by the existence of several hole regions at the coordinate origin.

Several investigations <sup>[9-11]</sup> have indicated strong pressure dependence of the lattice parameters and energy spectrum of antimony. At 10,000 atm the number of carriers is decreased by 20-40%, and the inclination of the electron ellipsoids is also diminished. <sup>[10]</sup> At about 70,000 atm a primitive cubic structure is observed. <sup>[11]</sup>

In the present work we have studied the magnetoresistance anisotropy of antimony in high transverse magnetic fields. Our results appear to indicate that the anisotropy of the Fermi surface is temperature dependent.

#### EXPERIMENT

We measured effects in antimony single crystals having rational orientations and subjected to pulsed transverse magnetic fields up to 80 kOe along with static fields up to 20 kOe. The samples, with dimensions  $1-1.5 \times 1-1.5 \times 8-12$  mm obtained by spark cutting from ingots that had been grown from "Su-1000" antimony. Cutting of the sample and installation in its holder caused at most 3° deviation from its nominal orientation.

Our data pertain to the following samples: Sb-I  $(J \parallel C_3, H \text{ in the } C_1C_2 \text{ plane})$ , Sb-II and Sb-II'  $(J \parallel C_1, H \text{ in the } C_2C_3 \text{ plane})$ , and Sb-III and Sb-III'  $(J \parallel C_2, H \text{ in the } C_1C_3 \text{ plane})$ . J is the current through the sample; H is the magnetic field; C<sub>1</sub>, C<sub>2</sub>, and C<sub>3</sub> are the bisectrix, binary, and trigonal crystallographic axes. In the experimental work precautions were taken against extraneous effects resulting from incorrect positioning.<sup>[12]</sup> Measurements were performed at 77°, 20.4°, and 4.2°K. The values obtained for the ratio  $\rho(300^{\circ}\text{K})/\rho(\text{T}^{\circ}\text{K})$  were 6.4, 105, and 730, respectively, from sample Sb-II'. Here  $\rho(\text{T}^{\circ}\text{K})$  is the electrical resistance in zero field at the given temperature. We note that fields below 5 kOe were not used at hydrogen and helium temperatures.

#### EXPERIMENTAL RESULTS

Magnetoresistance anisotropy is usually characterized by the ratio of the resistances observed with different orientations of the transverse magnetic field. The principal crystal axes were taken as the basic directions of H.

1. The anisotropy of magnetoresistance,

$$\alpha_{C_{1}/C_{1}} = \left(\frac{\Delta\rho_{H}}{\rho_{0}}\right)_{\mathbf{H} \parallel C_{1}} : \left(\frac{\Delta\rho_{H}}{\rho_{0}}\right)_{\mathbf{H} \parallel C_{1}}$$
(1)

(where  $\Delta \rho_{\rm H} = \rho_{\rm H} - \rho_0$ ,  $\rho_{\rm H}$  is the electrical resistivity in a field H and  $\rho_0$  is the electrical resistivity in zero field) of sample Sb-I remains constant in fields higher than 15 kOe at 77°K or higher than 5 kOe at 20.4° and 4.2°K, but with quite different values at the different temperatures (Fig. 1a):

<i>T</i> °, K∶	77	20,4	4.2
$\alpha_{C_1/C_1}$ :	1,25	1,75	1,35
$\alpha_{C_1/C_2}$ :	0.8	1,95	0,9
α <sub>C2/C3</sub> :	3,2	2,3	0,8

The temperature dependence of  $\alpha_{C_1/C_2}$  is seen to be nonmonotonic.

2. The anisotropy  $\alpha_{C_2/C_3}$  of sample Sb-II exhibits qualitative as well as quantitative temperature depen-



FIG. 1. Rotation diagrams (arbitrary scale).  $a - \text{sample Sb-I} (\varphi = 0^{\circ} \text{ corresponds to } H \parallel C_2)$ ;  $b - \text{Sb-II} (\varphi = 0^{\circ} \text{ corresponds to } H \parallel C_3)$ .  $\Delta - T = 77^{\circ}\text{K}$ , H = 50 kOe;  $X - T = 20.4^{\circ}\text{K}$ , H = 70 kOe;  $\bigcirc -T = 4.2^{\circ}\text{K}$ , H = 70 kOe.



FIG. 2. Rotation diagram of Sb-II' in rectangular coordinates. H || C<sub>3</sub> corresponds to  $\varphi = 130^\circ$ ;  $\Theta - T = 77^\circ K$ , H = 18 k0e (left-hand ordinates);  $O - T = 4.2^\circ K$ , H = 18 k0e (right-hand ordinates).

dence. At 4.2°K the minimum of magnetoresistance for  $H \parallel C_3$  that is observed at 77° and 20.4°K is replaced by a maximum, while shallow minima appear for  $H \parallel C_2$  and field directions forming angles  $\varphi = \pm 30^\circ$  with C<sub>2</sub>. The weak minimum for  $H \parallel C_2$  appears at hydrogen temperature (Fig. 1b). At constant T,  $\alpha_{C_2/C_3}$  remains constant in fields above 20 kOe at nitrogen temperature and above 5 kOe at hydrogen and helium temperatures, with the values given above.

Several control experiments were performed on Sb-II samples; measurements for Sb-II' were obtained in a static field (Fig. 2). Good agreement is found between the experimental results obtained with different techniques for different samples. The discrepancy between our results and those of Steele in<sup>[7]</sup>, where the rotation diagrams of an Sb-II single crystal are qualitatively alike at nitrogen and helium temperatures, can evidently have resulted from the relatively large amount of impurities (0.052%) in the sample used by Steele.

3. Measurements on Sb-III samples also reveal a nonmonotonic temperature dependence of  $\alpha$ . Small deviations from a given orientation also strongly influence the shape of the diagram. A total deflection of  $2-3^{\circ}$  incurred in the cutting and mounting (in a holder) of the sample can seriously distort the symmetry of the diagram (Fig. 3a). A similar behavior is observed in bismuth single crystals at the same orientation.<sup>[13]</sup> Therefore the rotation diagrams are not alike for different samples, with the most appreciable changes occurring at 77°K (Fig. 3). At 4.2°K, as at hydrogen temperature, the angular diagram is rotated slightly without a change of shape. The values of  $\alpha_{C_1/C_3}$  for Sb-III are given above.

For all orientations the electrical resistance of the samples exhibits almost quadratic dependence on the magnetic field at helium, hydrogen, and nitrogen temperatures.

### DISCUSSION OF RESULTS

At the present time no experimental data are available for the mean free path of carriers in antimony. It is therefore impossible to obtain a sufficiently accurate value of  $H_0$ , the magnetic field in which the radius r of the carrier orbit is comparable with their mean free path *l*. We can state, however, that the criterion of a "high magnetic field" was satisfied in our measurements ( $r \ll l$ ). (We here refer to fields higher than 20 kOe at 77°K and higher than 5 kOe at hydrogen and helium temperatures.) Our affirmation is based on the following facts:



FIG. 3. Rotation diagrams (arbitrary scale). a - sB-III (0° corresponds to H || C<sub>1</sub>); b - Sb-III (0° corresponds to H || C<sub>1</sub>).  $\Delta - T = 77^{\circ}K$ , H = 50 k0e; X - T = 20.4°K, H = 70 k0e; O - T = 4.2°K, H = 70 k0e.

1. We used antimony single crystals of high purity. The ratio  $\rho(300^{\circ}\text{K})/\rho(20.4^{\circ}\text{K})$  for our samples exceeded the ratio for the samples used in<sup>[7]</sup>, where at 4.2°K in fields of the order of 10 kOe Shubnikov-de Haas oscillations were observed, thus showing that  $r \ll l$  was satisfied. Consequently, at 20.4°K our samples were obviously in a high magnetic field.

2. Calculations based on Eckstein's data<sup>[14]</sup> give  $2 \times 10^{-6}$  cm for the orbital radius r in fields  $\sim 5 \times 10^4$  Oe. This result agrees in order of magnitude with the mean free path of carriers in metals at room temperature. In other words, the condition  $r \sim l$  in fields of a few tens of kiloersteds is evidently satisfied for antimony at T  $\approx 300^{\circ}$ K.

3. The anisotropy of the magnetoresistance of antimony at a fixed temperature is independent of the magnetic field. We shall also assume that if the values of  $\Delta\rho_{\rm H}/\rho_0$  coincide at different temperatures the sample is located in an identical "effective" field. In the case of sample Sb-II, for example, when  ${\bf H} \parallel {\bf C}_2 (\varphi = 90^\circ)$ , we have

$$(\Delta \rho_H / \rho_0)_{T=20,4^{\circ} \text{ K}, H=70 \text{ kOe}} = (\Delta \rho_H / \rho_0)_{T=4.2^{\circ} \text{ K}, H=15 \text{ kOe}}$$

When  $H \parallel C_3$  the equality is fulfilled at 70 and 10 kOe, respectively. A comparison of these results with the values of  $\alpha_{C_2/C_3}$  shows that the anisotropy of magnetoresistance is independent of the "effective magnetic field." The foregoing discussion makes it obvious that the anomalous behavior of the anisotropy in the case of antimony is associated directly with temperature. It has been shown in<sup>[15,16]</sup> that in high magnetic fields

It has been shown in <sup>[15,16]</sup> that in high magnetic fields the anisotropy of magnetoresistance in a metal is not dependent on collision processes and is governed only by the geometry of the Fermi surface. On the other hand, the experimental results presented in the present work indicate that  $\alpha$  varies considerably both qualitatively and quantitatively as a function of temperature. Therefore within the framework of the theory presented in <sup>[15,16]</sup> the anisotropy of the antimony Fermi surface is temperature dependent. Measurements performed on Sb-III samples indicate that the Fermi surface exhibits its greatest anisotropy at 77°K. At the same time the surface topology and the electron/hole ratio appear to remain unaltered, as indicated by the character of the dependence of  $\Delta \rho_{\rm H}/\rho_0$  on H for different current and field directions.

Temperature dependence of the anisotropy of magnetoresistance was previously observed in the case of bismuth and was accounted for by a temperature-dependent change in the number of carriers in conjunction with nonquadratic carrier dispersion; [13] the same effect may be occurring in antimony. While, to be sure, variation of the carrier concentration of antimony was not observed in<sup>[10]</sup>, it has been shown recently that in the multivalent metals Pb, Sn, Al, and In the concentration of conduction electrons increases with the temperature. For these metals the concentration increases 5-20% as the temperature rises from  $4.2^{\circ}$  to  $300^{\circ}$ K; this is accounted for by the temperature dependence of the Fourier components of the pseudopotential.<sup>[17,18]</sup> The authors of <sup>[10]</sup> may have overlooked similar behavior in the case of antimony, since they performed no measurements below 77°K and determined the carrier concentration with  $\pm 10\%$  error. For these reasons they may have overlooked the temperature dependence of the number of carriers in antimony which results from the temperature dependence that is usually exhibited by the lattice anisotropy of metals.

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