Shubnikov–de Haas Effect in Dilute Bismuth-Antimony Alloys. II. Quantum Oscillations in High Magnetic Fields

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The transverse magnetoresistance of a dilute Bi-Sb system was measured at 4.2°K with a magnetic field (up to 80 kG and above) in the trigonal plane. The Shubnikov–de Haas oscillations due to holes were observed systematically. The electron Fermi energy for the alloys was found to decrease with the magnetic field in a similar fashion to that for pure Bi. The hole Fermi energies were found to increase with the magnetic field. Through numerical analyses, we suggest that the energy overlap of the conduction band and the valence band should decrease with the magnetic field for both pure Bi and the alloys. Spin splitting effect for holes was observed for the Bi-Sb alloy of 0.95-at.% Sb concentration. For this alloy, the spin effective mass M_s was found to be approximately 2.6 times larger than the orbital effective mass M_c [$M_s \simeq (2.6\pm0.4)M_c$] in the trigonal plane.

I. INTRODUCTION

In spite of the fact that a large amount of research[1–7] has been devoted to the study of the band structure and the involved parameters of pure Bi and Bi-Sb alloys, there have been no observations on the quantum oscillations in the semimetallic Bi-Sb alloys in high magnetic fields.

The Bi-Sb system has extremely small effective masses of the conduction electrons and low chemical potentials. The extreme quantum limit with Landau level spacings comparable to the Fermi energy can be reached in a moderately high magnetic field. In high fields, the Fermi energy will vary with the field strength due to the extreme quantum effect. Smith *et al.*[8] have reported this effect in pure Bi, and later Pelikh and Eremenko[9] made a direct measurement of the displacement of the Fermi level for pure Bi. They found that the Fermi energy of pure Bi decreased with the magnetic field H for H along the bisectrix and the binary axes. These results clearly indicated that the variation of the Fermi energy in high fields plays an important role in the high-field effects.

The band structure of Bi-Sb alloys was experimentally[4–7] verified to be similar to that of pure Bi. To date, all the experiments on these alloys were performed in low magnetic field regions ($H<20$ kG). It is expected that the resemblance in the band structure would remain valid in high magnetic fields. The effect of the variation of the Fermi energy with magnetic field in Bi-Sb alloys should also be analogous to that in pure Bi.

Brandt and co-workers[10] reported in their recent paper the relative motion of the valence band with respect to the conduction band such that the semiconducting Bi-Sb alloys (Sb concentration$>$8 at.%) may be turned into the semimetallic state in a sufficiently high magnetic field. The valence band mentioned above would be the hole band when the semimetallic state had been reached. It is of interest to inquire to what extent can the band motion of this kind be observed in a Bi-Sb system of low Sb concentrations.

Smith *et al.*[8] also found spin splitting due to holes in pure Bi and tried to assign a spin effective mass M_s for holes that was approximately seven times larger than the orbital cyclotron mass M_c($M_s \simeq 7M_c$) when the magnetic field was in the trigonal plane.

In the present high-field experiment, the Shubnikov–de Haas (SdH) effect due to holes was observed systematically for both pure Bi and dilute Bi-Sb alloys with Sb concentration ranging 0–1 at.%. We found that the oscillations were not periodic in $1/H$, and the Fermi energy of every alloy was found to decrease with the magnetic field. In the 0.95% sample, the spin splittings due to holes were observed. The increase of the hole Fermi energy was found to be not equal to the decrease of the electron Fermi energy. It therefore suggests that the energy overlap should not remain constant in the high magnetic fields.

According to the two-band model, which has been investigated in some detail in Ref. 7 (hereafter referred to as I), it is easy[7,8] to calculate the number of states per unit volume in a magnetic field with energy less than the Fermi energy.

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Electrons (for one of the three ellipsoids):

$$n_{(E\leq E_F)}=\frac{2^{3/2}eH}{h^2c}(m_z)^{1/2}\times\sum_{n,S}\left[E_F\left(1+\frac{E_F}{E_g}\right)-(n+\tfrac{1}{2})\frac{eh}{m_cc}H+\tfrac{1}{2}S\frac{eh}{m_sc}H\right]^{1/2};\quad(1)$$

holes:

$$N_{E\leq(E_0-E_F)}=\frac{2^{3/2}eH}{h^2c}(M_z)^{1/2}\times\sum_{N,S}\left((E_0-E_F)-(N+\tfrac{1}{2})\frac{eh}{M_cc}H+\tfrac{1}{2}S\frac{eh}{M_sc}H\right)^{1/2},\quad(2)$$

where E_F is the electron Fermi energy, E_0 is the energy overlap, E_g is the energy gap, m_z and M_z are the longitudinal effective masses along the magnetic field, and the sums are over those values of n and N, and of $S=\pm1$ such that the radicands are non-negative.

The charge neutrality condition is that

$$\sum_i n=N,\quad(3)$$

where the summation is over the three electron ellipsoids.

The spin effective mass m_s and the orbital cyclotron mass m_c of electrons should be equal (except for H along directions very close to the binary direction) if the two-band model is assumed to be valid:

$$m_s=m_c.\quad(4)$$

The derivatives of (1) and (2) with respect to the Fermi energy give the density of states at the Fermi level.

Electrons (for one of the three ellipsoids):

$$n(E_F)=\frac{2^{3/2}eH}{h^2c}(m_z)^{1/2}\sum_{n,S}\tfrac{1}{2}\left(1+\frac{2E_F}{E_g}\right)\times\left[E_F\left(1+\frac{E_F}{E_g}\right)-(n+\tfrac{1}{2})\frac{eh}{m_cc}H+\tfrac{1}{2}S\frac{eh}{m_sc}H\right]^{-1/2};\quad(5)$$

holes:

$$N(E_0-E_F)=\frac{2^{3/2}eH}{h^2c}(M_z)^{1/2}\times\sum_{N,S}\tfrac{1}{2}\left((E_0-E_F)-(N+\tfrac{1}{2})\frac{eh}{M_cc}H+\tfrac{1}{2}S\frac{eh}{M_sc}H\right)^{-1/2}.\quad(6)$$

When the magnetic field strength increases continuously, the quantized Landau levels will cross over the Fermi level one by one. Whenever H reaches such a value that the radicand in (5) or (6) for some n or N becomes zero, the density of states due to electrons or holes becomes infinite. Such a singularity can be removed by adding, for example, the nonzero temperature effect into the calculation. However, it remains true that the density of states assumes a maximum whenever the radicand becomes zero.

For transverse magnetoresistance, the maximum in the density of states at the Fermi level corresponds to a minimum in the magnetoresistance. It is very complicated to derive a complete formula for the magnetoresistance of such complicated materials as Bi or Bi-Sb; actually, there has yet been no such formula available (cf. Ref. 11, for example). However, the condition for having a minimum in the magnetoresistance can be obtained through the condition for the maximum in the density of states at the Fermi level (cf. Refs. 12 and 8).

Therefore, the conditions for having a minimum in the magnetoresistance are the following.

Due to electrons:

$$E_F\left(1+\frac{E_F}{E_g}\right)=(n+\tfrac{1}{2})\frac{eh}{m_cc}H\pm\tfrac{1}{2}\frac{eh}{m_sc}H=n(eh/m_cc)H\quad(\text{if } m_c=m_s),\quad n=0,1,2,\ldots;\quad(7)$$

due to holes:

$$E_0-E_F=(N+\tfrac{1}{2})\frac{eh}{M_cc}H\pm\frac{1}{2}\frac{eh}{M_sc}H\cong(N+\tfrac{1}{2})(eh/M_cc)H\quad(\text{if } M_s\gg M_c),\quad N=0,1,2,\ldots.\quad(8)$$

The factors $\frac{1}{2}$ in Eq. (7), that originally appeared in the expression for the energy levels, have been theoretically[13,14] and experimentally (for references, see I) verified. The first factor $\frac{1}{2}$ in Eq. (8) comes from the model for the energy surface of holes in Bi and has been used by authors so far. However, it could be some other constant, for example, $0<\gamma<1$. Since there will not be qualitative interference in our results and conclusions except for a slight change in the values of the hole Fermi energy if the factor $\frac{1}{2}$ is replaced by $\gamma\neq\frac{1}{2}$, we will not consider any alteration of this factor. The factor $\frac{1}{2}$ for the spin term in Eq. (8) can be considered simply for convenience, since M_s is to be defined in this way.

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II. EXPERIMENTAL ASPECTS

We described in I some details of the sample preparation and the dc circuit for measuring the SdH effect.

In this experiment, the high magnetic fields (up to 80 kG and higher) were obtained by using a superconducting solenoid made by Magnion, Inc. The magnetic field was vertical. In order to measure the transverse magnetoresistance along different crystallographic orientations, a gear system was installed in the sample holder so that we could rotate the sample about a horizontal axis.

To figure out the exact orientation of the bisectrix axis, we followed the same procedure as that in our previous experiment.[7] Once the bisectrix orientation had been found, we could adjust the micrometer head attached to the gear system to get any desired orientation in the trigonal plane.

No bucking technique for the magnetoresistance was applied for observing the SdH effect in high magnetic fields. The oscillations were large enough to be observed in the high-field region. The magnetic field strength was read by using a magnetic probe which was calibrated at the Brookhaven National Laboratory.

III. RESULTS AND DISCUSSION

The magnetoresistance of pure Bi and Bi-Sb alloys (Sb concentrations: 0.23, 0.48, 0.72, and 0.95 at. %) in a high magnetic field region (up to 80 kG and above) was measured at 4.2°K for different orientations in the trigonal plane. Figures 1(a)–1(c) show typical curves of ΔV (the potential drop across the two potential leads of the sample which was proportional to the resistance as the current was kept constant, and was read by the Y terminal of the XY recorder) versus magnetic field. Actually, the X terminal of the XY recorder read the potential drop across the potential leads of the magnetic probe (current = 32 mA, $T=4.2$°K). This in turn gave the field strength according to the calibration mentioned previously. To determine the positions of the minima, we drew two envelopes above and below the measured curve. The mean of these two envelopes gave the monotonically increasing part of the magnetoresistance. The positions at which the measured signals were minimal with respect to this curve were the minima for the SdH effect (cf. Babiskin[15]).

Note that the oscillations in the high magnetic field region are clear enough to be observed, even though the bucking technique has not been applied. These oscillations have been attributed to holes (cf. Ref. 8) for two reasons. First, the field values at the minima in the magnetoresistance for holes do not depend very much on the orientations in the trigonal plane because of the rotational symmetry of the hole ellipsoid about the trigonal axis; this is not the case for electrons. Second, we can identify the quantum numbers of the Landau levels for each minimum by using the known data for the effective masses of pure Bi. We have found that it is satisfactory to identify them as due to holes (cf. subsequent discussions).

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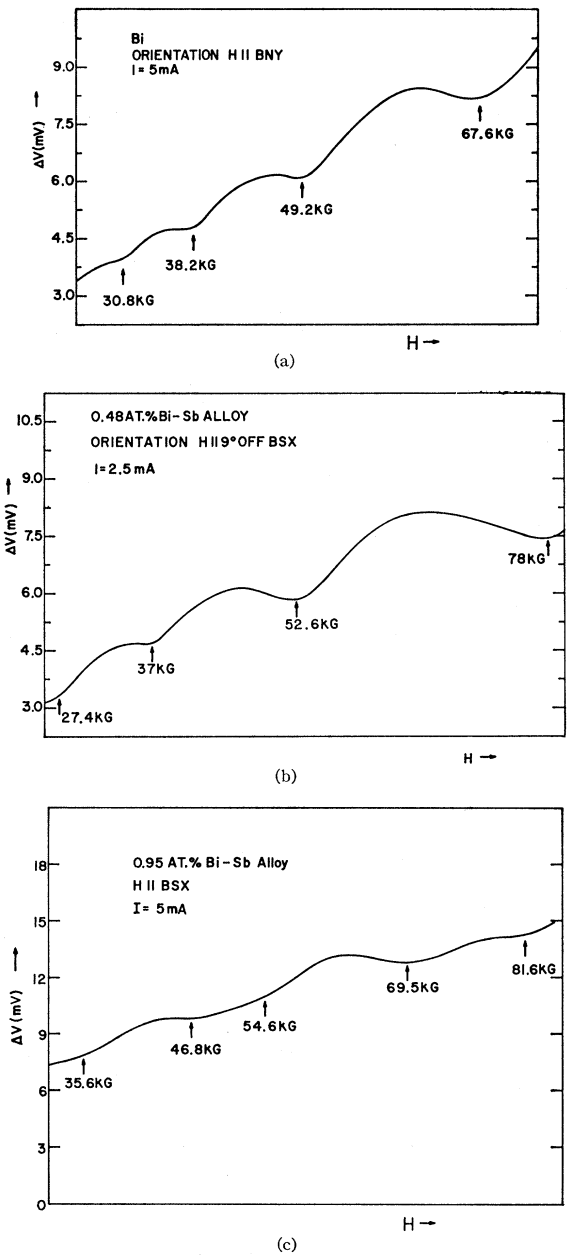

FIG. 1. (a) ΔV versus magnetic field showing SdH oscillations due to holes. (b) ΔV versus magnetic field showing SdH oscillations due to holes. (c) ΔV versus magnetic field showing SdH oscillations due to holes. Spin splittings are shown (minima at 46.8 and 54.6 kG, and at 69.5 and 81.6 kG).

We measured the magnetoresistance for every 3°–5° rotation of H in the trigonal plane for the Bi-Sb system. Some of the measured field values at which the minima in the magnetoresistance due to holes occur are listed in Tables I–IV.

TABLE I. Field values in kG for the minima in the magnetoresistance due to holes with the magnetic field parallel to the bisectrix axis.[a]

	Pure Bi	0.23 at.%	0.48 at.%	0.72 at.%	0.95 at.%
$N=3$			79.6±2	62.4±2	50.7[b]±2
$N=4$	71±1	56(1.27)±1	53.8(1.32)±1	44(1.61)±1.5	35.6(1.99)±1.5
$N=5$	50.2±1	39.8(1.26)±1	38 (1.32)±1	32(1.57)±1	
$N=6$	36.6±1	29.8(1.23)±1	28.4(1.29)±1		
$N=7$	28.4±1				

[a] The figures in parentheses are the ratios of the field values for pure Bi and the corresponding (in the same row in the table) field values for the alloys. Errors in the field values due to the probe calibration have not been included.
[b] Averaged value from the two values split by spin.

Equation (8) states the condition for having a minimum in the magnetoresistance due to holes. When the magnetic fields were oriented in the trigonal plane, we did not find apparent spin splittings in the quantum oscillations due to holes for pure Bi and Bi-Sb alloys of 0.23-, 0.48-, and 0.72-at.% Sb concentrations. Thus we have to assign either $M_s \gg M_c$ or $M_s \simeq M_c$ to these crystals.

Smith *et al.*[8] proposed $M_s \simeq 7M_c$ for pure Bi. Besides, it also seems to be more acceptable to assign $M_s \gg M_c$ to those crystals since the condition for $M_s \simeq M_c$ is closely related to the two-band model in the case of electrons (no other band lies close to the hole band).

We see that $M_s \gtrsim 7M_c$ is in agreement with our experiment, since the small spin splitting associated with the value of M_s will not give rise to observable spin effect in our magnetic field region. From our experiment, we can set a limit for the hole spin mass for pure Bi and those alloys mentioned above, i.e., $M_s/M_c > 4$. We will discuss this point later along with the discussion of the observed spin splitting of the 0.95-at.% alloy. The data listed in Tables I–IV cover those for alloys of Sb concentration ≤ 0.72 at.%, whose M_s are large enough, and a part (in lower fields) of that for the alloy of 0.95-at.% Sb. Thus, in the following discussion based on the data listed, we can ignore the spin effect without affecting our results.

Thus, the condition for having a minimum in the magnetoresistance due to holes can be simplified to the form

$$E_0 - E_F = (N+\tfrac{1}{2})\hbar eH/M_c c, \quad N=0,1,2,\ldots. \quad (9)$$

To identify the quantum number N for each minimum, we first make the identifications for pure Bi. The identification for Bi-Sb alloys will then follow without too much difficulty.

The measured field values at the minima for pure Bi when the magnetic field is parallel to the bisectrix axis are 71, 50.2, 36.6 and 28.4 kG. If we try to assign $N=4$ to 71 kG, $N=5$ to 50.2 kG, . . ., etc., then we have

$$H=71\text{ kG}:\quad E_F = E_0 - (4+\tfrac{1}{2})e\hbar/M_c c(71\text{ kG}) = 20.86\text{ meV};$$

$$H=50.2\text{ kG}:\quad E_F = E_0 - (5+\tfrac{1}{2})e\hbar/M_c c(50.2\text{ kG}) = 23.26\text{ meV};$$

$$H=36.6\text{ kG}:\quad E_F = E_0 - (6+\tfrac{1}{2})e\hbar/M_c c(36.6\text{ kG}) = 25.37\text{ meV};$$

$$H=28.4\text{ kG}:\quad E_F = E_0 - (7+\tfrac{1}{2})e\hbar/M_c c(28.4\text{ kG}) = 26.74\text{ meV},$$

where $E_0 = 38.5$ meV and $M_c = 0.21m_0$.[8] To know whether these assignments are correct or not, the satisfaction of the neutrality condition is now to be examined. The neutrality condition is that the number of electrons be equal to the number of holes.

When the magnetic field is along the bisectrix axis, all the energy levels except the lowest one for each of the three electron ellipsoids have crossed over the Fermi level as long as the field strength is larger than about 20 kG. Therefore, according to (1) and (2), the neutrality condition will have the simple form

$$(m_{z1}^{1/2} + 2m_{z2}^{1/2})[E_F(1+E_F/E_g)]^{1/2} = 2M_z^{1/2}\sum_N [(E_0 - E_F) - (N+\tfrac{1}{2})\hbar eH/M_c c]^{1/2}, \quad (10)$$

where[8] $m_{z1} = 0.26m_0$ is the longitudinal mass of the electrons in the ellipsoid whose bisectrix axis is parallel to the magnetic field, $m_{z2} = 0.06585m_0$ is the longitudinal effective mass of the electrons in the other two ellipsoids whose bisectrix axes are at an angle of 60° (120°) to the field direction, $M_z = 0.064m_0$ is the longitudinal effective mass of holes, and $E_g = 15.3$ meV is the energy gap. The factor of 2 on the right-hand side comes from the spin degeneracy for holes. In (10) we have already used the two-band model for electrons.

TABLE II. Field values in kG for the minima in the magnetorestistance due to holes with the magnetic field in the trigonal plane 9° off the bisectrix axis.

	Pure Bi	0.23 at.%	0.48 at.%	0.72 at.%	0.95 at.%
$N=3$			78±1.5	62±1.5	
$N=4$	71.6±1.5	55.2(1.29)±1	52.6(1.36)±1	42.8(1.67)±1	35.6(2.0)±1.5
$N=5$	49.6±1	39.8(1.25)±1	37(1.34)±1		24(2.0)±1
$N=6$	36.6±1	29.2(12.5)±1	27.4(1.34)±1		
$N=7$	29.8±1				

TABLE III. Field values in kG for the minima in the magnetoresistance due to holes with the magnetic field in the trigonal plane 18° off the bisectrix axis.

	Pure Bi	0.23 at.%	0.48 at.%	0.72 at.%	0.95 at.%
$N=3$				59.4±1.5	
$N=4$	69±1	54(1.27)±1	51.6(1.34)±1	40.4(1.70)±1	35.6(1.94)±1.5
$N=5$	48.4±1	40.5(1.20)±1	37.8(1.28)±1		
$N=6$	38.2±1	31.6(1.21)±1	30.3(1.26)±1		
$N=7$	30.4±1				

Equation (10) becomes numerically

$$4.04[E_F(1+E_F/15.3)]^{1/2} = 2\sum_N [(38.5-E_F)-(N+\tfrac{1}{2})5.52\times10^{-2}H]^{1/2}, \quad (11)$$

where H is the magnetic field in kG.

For $H=71$ kG, the summation is over $N=0$, 1, 2, 3 and $E_F=20.86$; for $H=50.2$ kG, the summation is over $N=0$, 1, 2, 3, 4 and $E_F=23.26$, etc. The calculated results on both sides of Eq. (11) are

	Left-hand side	Right-hand side
$H=71$ kG:	28.37(31.27)	24.34
$H=50.2$ kG:	30.93(32.87)	27.90
$H=36.6$ kG:	33.18(34.52)	30.79
$H=28.4$ kG:	34.63(35.65)	33.76.

The agreement is fairly good and much better than any other assignment. Therefore, the assignment of the quantum numbers $N=4$, 5, 6, 7 to the observed minima has been proven to be correct. In practice the optimal choice of quantum numbers was obtained by trial and error. In this way we obtained the quantum numbers (N) shown in Tables I–IV.

We also calculated the number of electrons taking into consideration the possible deviations from the two-band model. The orbital and spin effective masses were those used in Ref. 8. The results have been shown in the parentheses in the above table. We can see that our data again favor the conclusion we made in Ref. 7, i.e., the two-band model is an excellent description of the electron band, especially in the bisectrix direction.

Although the identification of the quantum numbers N is successful, the satisfaction of the neutrality condition is beyond the errors in the above calculation. The errors on the right-hand side are less than 5% (due to errors in the field values and the possible existence of hole spin splittings).

We propose that the main cause of the discrepancy in the satisfaction of the neutrality condition is the assumption of a constant energy overlap E_0. Brandt *et al.*[10] in their recent work studied such relative motion between the conduction band and the valence band. They found that the valence band in semiconducting (Sb concentration > 8 at.%) Bi-Sb alloys moves upwards in magnetic fields.

We now make an alternative calculation. First, calculate the hole Fermi energy using the data in Tables I–IV, and then calculate the electron Fermi energy through the neutrality condition. The energy overlap can then be obtained by adding together these two Fermi energies. The following is such a calculation for an alloy of 0.48-at.% Sb concentration and for the orientation of the magnetic field along the bisectrix axis.

From Eq. (9) we have

$$H=79.6:\quad E_0-E_F=(3+\tfrac{1}{2})(5.52\times10^{-2})(79.6) = 15.38 \text{ (meV)};$$

$$H=53.8:\quad E_0-E_F=(4+\tfrac{1}{2})(5.52\times10^{-2})(53.8) = 13.36 \text{ (meV)};$$

$$H=38:\quad E_0-E_F=(5+\tfrac{1}{2})(5.52\times10^{-2})(38) = 11.54 \text{ (meV)};$$

$$H=28.4:\quad E_0-E_F=(6+\tfrac{1}{2})(5.52\times10^{-2})(29.6) = 10.19 \text{ (meV)}.$$

Here we have assumed that M_c has the same value in dilute Bi-Sb alloys that it has in pure Bi.

Using Eq. (10), we have

$$H=79.6:\quad 4.04[E_F(1+E_F/15.3)]^{1/2} = 2\sum_{N=0}^{2}[15.38-(N+\tfrac{1}{2})(5.52\times10^{-2})(79.6)]^{1/2} = 17.38;$$

$$E_F=10.84 \text{ (meV)},\quad E_0=10.84+15.38=26.22 \text{ (meV)};$$

TABLE IV. Field values in kG for the minima in the magnetoresistance due to holes with the magnetic field parallel to the binary axis.

	Pure Bi	0.23 at.%	0.48 at.%	0.72 at.%	0.95 at.%
$N=3$			73±1.5	59.2±1.5	
$N=4$	67.6±1	53(1.27)±1	51(1.33)±1	43.2(1.56)±1	36(1.88)±1.5
$N=5$	49.2±1	39.6(1.24)±1	38(1.29)±1		
$N=6$	38.2±1	31.8(1.20)±1	30.8(1.24)±1		
$N=7$	30.8±1				

$$H=53.8:\quad 4.04[E_F(1+E_F/15.3)]^{1/2}$$
$$=2\sum_{N=0}^{3}[13.36-(N+\tfrac{1}{2})(5.52\times10^{-2})(53.8)]^{1/2}$$
$$=21.18,$$
$$E_F=14.23,\quad E_0=27.59;$$

$$H=38:\quad 4.04[E_F(1+E_F/15.3)]^{1/2}$$
$$=2\sum_{N=0}^{4}[11.54-(N+\tfrac{1}{2})(5.52\times10^{-2})(38)]^{1/2}$$
$$=24.29,$$
$$E_F=17.08,\quad E_0=28.62;$$

$$H=28.4:\quad 4.04[E_F(1+E_F/15.3)]^{1/2}$$
$$=2\sum_{N=0}^{5}[10.19-(N+\tfrac{1}{2})(5.52\times10^{-2})(28.4)]^{1/2}$$
$$=27.12,$$
$$E_F=19.70,\quad E_0=29.89.$$

We may apply this calculation to the other Bi-Sb alloys as well as to pure Bi. The results are listed in Tables V–IX.

From Tables V–IX, we see that the electron Fermi energy E_F decreases with the magnetic field strength, and the hole Fermi energy (E_0-E_F) increases with the field strength. We realize that if the magnetic field is parallel to the bisectrix axis, all the electron Landau levels except the 0^- level will have crossed over the Fermi level provided the field strength is greater than about 20 kG. According to the two-band model, this level remains unchanged in magnetic field. However, the degeneracy of this level or the population of electrons at this level will increase with the field strength. This might be one of the reasons why the Fermi energy should fall down. On the other hand, the spin splitting is small for holes, so that every Landau level of holes will move in a changing magnetic field. The motion of the hole levels tends to push down the Fermi level.

We also see from the Tables V–IX that the energy overlap E_0 decreases with the field strength. This is a very interesting result. Even if E_0 is a constant, the downward motion of the hole levels will finally cross the 0^- level of electrons, if the 0^- level is fixed. In other words, a magnetic-field-induced "transition" from a semimetallic state to a semiconducting state is feasible, provided the two-band model remains valid. The decrease of E_0 will certainly expedite the transition to happen at a less strong magnetic field. For example, a Bi-Sb alloy of 0.48-at.% Sb having an energy overlap equal to roughly 35 meV at zero magnetic field will have an energy overlap equal to about 26 meV at the field strength of about 80 kG.

TABLE V. Values of hole Fermi energy (E_0-E_F), electron Fermi energy (E_F), energy overlap (E_0), and the spatial density of the charge carriers (N) for pure Bi. The magnetic field is parallel to the bisectrix axis.

H(kG)	N	E_0-E_F(meV)	E_F(meV)	E_0(meV)	$N(10^{17}/\text{cm}^3)$
71	4	17.64	17.13	34.77	5.45
50.2	5	15.24	20.41	35.65	4.42
36.6	6	13.13	23.18	36.31	3.55
28.4	7	11.76	25.91	37.66	3.02

TABLE VI. Values of hole Fermi energy (E_0-E_F), electron Fermi energy (E_F), energy overlap (E_0), and the spatial density of the charge carriers (N) for the Bi-Sb alloy of 0.23-at.% Sb. The magnetic field is parallel to the bisectrix axis.

H(kG)	N	E_0-E_F(meV)	E_F(meV)	E_0(meV)	$N(10^{17}/\text{cm}^3)$
56	4	13.91	14.63	28.54	3.82
39.8	5	12.08	17.59	29.67	3.12
29.8	6	10.69	20.31	31.00	2.61

TABLE VII. Values of hole Fermi energy (E_0-E_F), electron Fermi energy (E_F), energy overlap (E_0), and the spatial density of the charge carriers (N) for the Bi-Sb alloy of 0.48-at.% Sb. The magnetic field is parallel to the bisectrix axis.

H(kG)	N	E_0-E_F(meV)	E_F(meV)	E_0(meV)	$N(10^{17}/\text{cm}^3)$
79.6	3	15.38	10.84	26.22	4.36
53.8	4	13.36	14.23	27.59	3.59
38	5	11.54	17.08	28.62	2.91
28.4	6	10.19	19.70	29.89	2.43

TABLE VIII. Values of hole Fermi energy (E_0-E_F), electron Fermi energy (E_F), energy overlap (E_0), and the spatial density of the charge carriers (N) for the Bi-Sb alloy of 0.72-at.% Sb. The magnetic field is parallel to the bisectrix axis.

H(kG)	N	E_0-E_F(meV)	E_F(meV)	E_0(meV)	$N(10^{17}/\text{cm}^3)$
62.4	3	12.06	9.11	21.17	3.30
44	4	10.93	12.42	23.35	2.66
32	5	9.72	15.25	24.97	2.25

TABLE IX. Values of hole Fermi energy (E_0-E_F), electron Fermi energy (E_F), energy overlap (E_0), and the spatial density of the charge carriers (N) for the Bi-Sb alloy of 0.95-at.% Sb. The magnetic field is parallel to the bisectrix axis.

H(kG)	N	E_0-E_F(meV)	E_F(meV)	E_0(meV)	$N(10^{17}/\text{cm}^3)$
50.7	3	9.80	7.81	17.61	2.22
35.6	4	8.84	10.70	19.54	1.94

If such a transition happens, we expect that the magnetoresistance will undergo some changes through the transition. Immediately before the transition, the Fermi level is roughly coincident with the 0^- level of electrons and the 0 level (or 0^- level, if a spin effect exists) of holes, the density of states at the Fermi level is then large so that the magnetoresistance is small. After the transition, the alloy is a semiconductor, and the resistance will suddenly go up. We were pleased to note very recently that Brandt and co-workers[16] observed such semimetal-semiconductor transition at some strong magnetic field in Bi-Sb alloys of 4.9- and 6.5-at.% Sb.

In Tables I–IV, the ratios of the corresponding field values at the minima in the magnetoresistance have been shown in the parenthesis. For example, the value 1.61 (in the parentheses under 0.72 at.% and in the row of $N=4$ in Table I) from 71/44 is the ratio of the field value 71 kG at the minimum of $N=4$ for pure Bi when H is parallel to the bisectrix axis and the field value 44 kG at the minimum of the corresponding $N(=4)$ for the 0.72-at.% alloy when H is parallel to the same orientation. We see that the ratios for a given Sb concentration are roughly constant in the field region from about 30 kG to about 70 kG (the field region in which most of our observations were performed) at different orientations. Figure 2 shows the dependence of such a ratio on the Sb concentration, where for a given Sb concentration the averaged value of the ratios in the field region mentioned above and for different orientations is being used. Let us denote this function by $a(x)$, where $a(0)=1$, $a(0.23)=1.24$, $a(0.48)=1.3$, $a(0.72)=1.6$, and $a(0.95)=1.95$. A straight line can approximately fit these data, which give $a(x)\cong 1+0.85x$ (x in at. %). We can make some further analyses.

Let us assume, in general, that the hole Fermi energy (denoted by E_h) is a function of the Sb concentration x, the field strength H, and the orientation θ of the field with respect to the crystallographic axes system, i.e.,

$$E_h=E_h(x,H,\theta)\,, \tag{12}$$

where $x=0$ stands for pure Bi.

The experimental data give a relation between $E_h(x)$ and $E_h(0)$ at those field values at the minima in the magnetoresistance, i.e.,

$$\frac{E_h(0,aH,\theta)}{E_h(x,H,\theta)}=\frac{[H]_{\mathrm{Bi}}}{[H]_{\mathrm{alloy}}}=a(x)\cong 1+0.85x\,, \quad (x \text{ in at.\%}) \tag{13}$$

provided the effective mass M_c does not change appreciably in the dilute Bi-Sb system. So we have

$$E_h(x,H,\theta)=[1/a(x)]E_h(0,aH,\theta)\,. \tag{14}$$

Equation (14) is valid only at those field values at the minima in the magnetoresistance. However, it is reasonable to assume its validity for continuously varying field values if we are only interested in *the general shape of this functional relation* (possible oscillations may be superimposed on it).

We can rewrite Eq. (14) in an alternative form

$$a(x)E_h(x,H/a(x),\theta)=E_h(0,H,\theta)\,. \tag{15}$$

[16] N. B. Brandt, E. A. Svistova, and Yu. G. Kashivskii, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu 9, 232 (1969) [English transl.: Soviet Phys.—JETP Letters 9, 136 (1969)].

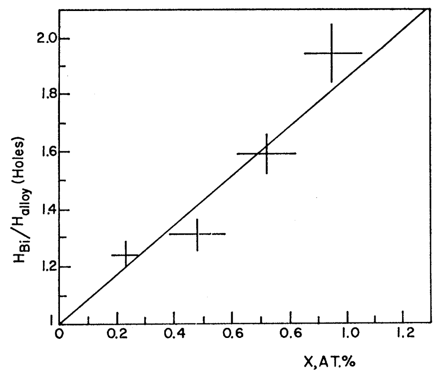

FIG. 2. Ratio of the corresponding (with same N) field values at minima $H_{\mathrm{Bi}}/H_{\mathrm{alloy}}$ (holes) versus Sb concentration.

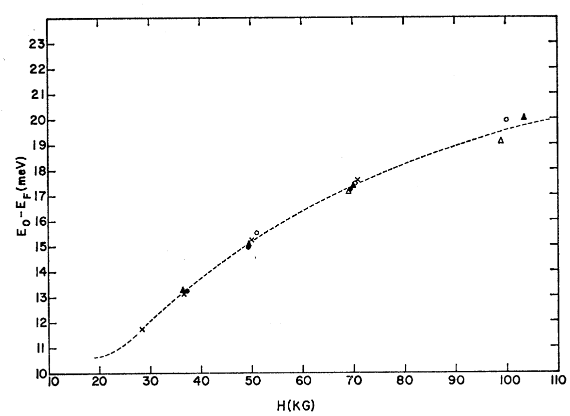

FIG. 3. Hole Fermi energy E_0-E_F (or E_h) versus magnetic field. ×: pure Bi, in units of meV and kG; •: Bi-Sb alloy with 0.23-at.% Sb, E in unit 1/1.24 meV, H in 1/1.24 kG; ▲: Bi-Sb alloy with 0.48-at.% Sb, E in unit 1/1.3 meV, H in 1/1.3 kG; ○: Bi-Sb alloy with 0.72-at.% Sb, E in unit 1/1.6 meV, H in 1/1.6 kG; △: Bi-Sb alloy with 0.95-at.% Sb, E in unit 1/1.95 meV, H in 1/1.95 kG; H: along bisectrix axis.

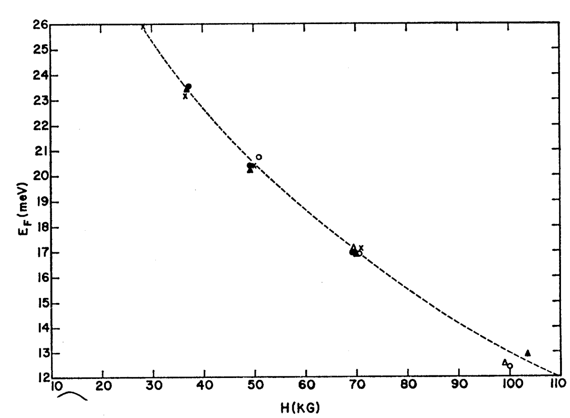

FIG. 4. Electron Fermi energy E_F versus magnetic field. ×: pure Bi, in units of meV and kG; •: Bi-Sb alloy with 0.23-at.% Sb. E_F in unit 1/1.16 meV, H in 1/1.24 kG; ▲: Bi-Sb alloy with 0.48-at.% Sb, E_F in unit 1/1.19 meV, H in 1/1.3 kG; ○: Bi-Sb alloy with 0.72-at.% Sb, E_F in unit 1/1.36 meV, H in 1/1.6 kG; △: Bi-Sb alloy with 0.95-at.% Sb, E_F in unit 1/1.6 meV, H in 1/1.95 kG; H along bisectrix axis.

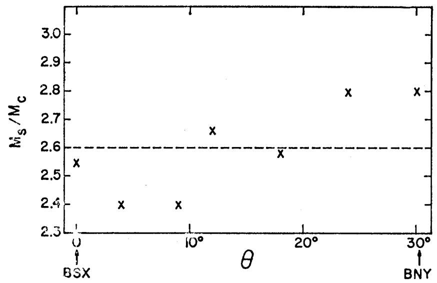

FIG. 5. Angular dependence of M_s/M_c in the trigonal plane for the Bi-Sb alloy with 0.95-at.% Sb.

This means that if we adjust the scales (or units) of the energy and the field strength for every Bi-Sb alloy, the dependence of E_h on H for every alloy will coincide with that for pure Bi. Figure 3 shows graphically this statement, where data in Tables V–IX are used.

Equation (14) or (15) is valid for the whole trigonal plane. We see that the variation of the hole Fermi energy with the magnetic field takes the same form for the entire dilute Bi-Sb system; one can get a general knowledge of this variation if he knows the complete variation for any alloy or pure Bi in the system.

Figure 4 shows the variation of the electron Fermi energy with the magnetic field. Again we used the data given in Tables V–IX. The scales or units for the energy and field have been also adjusted for alloys.

It is clear that the variation of the electron Fermi energy also has the same form for the entire dilute Bi-Sb system. For H along the bisectrix direction, we have, similar to (15), the following:

$$b(x)E_F(x, H/a(x), \theta) = E_F(0,H,\theta), \qquad (16)$$

where $b(0)=1$, $b(0.23)=1.16$, $b(0.48)=1.19$, $b(0.72)=1.36$, and $b(0.95)=1.6$; $a(x)$ is the same function as that used in Eq. (14) or (15).

Equations (15) and (16) strongly suggest that the resemblance in the band structure of a Bi-Sb system remains valid even in high magnetic fields.

From Fig. 3, we see that the variation of the hole Fermi energy slows down in higher magnetic fields. To estimate the spin splitting effect for the Bi-Sb alloy with 0.95-at. % Sb, let us assume that the hole Fermi energies at the two minima (due to spin splitting) associated with one Landau level are roughly equal. Therefore,

$$E_0 - E_F = (N+\tfrac{1}{2})\frac{e\hbar}{M_c c}H_1 + \frac{1}{2}\frac{e\hbar}{M_s c}H_1$$

and

$$E_0 - E_F = (N+\tfrac{1}{2})\frac{e\hbar}{M_c c}H_2 - \frac{1}{2}\frac{e\hbar}{M_s c}H_2,$$

where $H_2 > H_1$ are the field values at the two minima. We have

$$(N+\tfrac{1}{2})\frac{e\hbar}{M_c c}(H_2 - H_1) - \frac{1}{2}\frac{e\hbar}{M_s c}(H_2 + H_1) \cong 0$$

or

$$\frac{M_s}{M_c} \cong \frac{1}{2N+1}\frac{H_1+H_2}{H_2-H_1}. \qquad (17)$$

Since we have identified the quantum numbers N, we can easily calculate the ratio M_s/M_c through (17) from the measured data of H_1 and H_2. We found that M_s/M_c for the 0.95-at.% alloy was approximately constant in the trigonal plane and the value was about 2.6 ± 0.4. Figure 5 shows the plot of M_s/M_c against the orientation in the trigonal plane for this alloy.

For larger M_s/M_c and for lower magnetic fields (or N larger), $H_2 - H_1$ should be small according to the approximate estimation from (17). If $H_2 - H_1$ is smaller than 4 kG, for instance, the splitting would not be observable in our experiment. This explains why we did not find apparent spin splittings in our experiment (H in the trigonal plane, $H \lesssim 80$ kG) for the other alloys and pure Bi.

IV. CONCLUSION

From the observation of the SdH effect in high magnetic fields, the variation of the Fermi energy (both electron Fermi energy and hole Fermi energy) with the field strength has been confirmed for both pure Bi and dilute Bi-Sb alloys. The resemblance in shape of such variation for the Bi-Sb system has been found, assuming no appreciable variation of the effective masses for holes in the dilute alloys.

It has been suggested that the energy overlap of the conduction band and the valence band decreases with the magnetic field for the Bi-Sb system, assuming the validity of the two-band model.

An alternative explanation might be possible. For instance, that the effective masses (at the band edges) might change (i.e., the curvature of the band changes) in magnetic fields could account for the experimental data we obtained and satisfy the neutrality condition. We need then a theory to explain the change of the effective masses with the field strength.

The spin splitting effect due to holes has been observed. However, we have not found this effect in the trigonal plane for pure Bi and Bi-Sb alloys with Sb concentration less than or equal to 0.72 at.%.

ACKNOWLEDGMENTS

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