Observation of the Magnetic-Field-Induced Semimetal-Semiconductor Transition in Bi under Megagauss Fields

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The magnetic-field-induced semimetal-semiconductor transition has been observed in Bi under very high magnetic fields parallel to the binary axis. The transition was observed at 88 T as a change of the transmission of the far-infrared radiation at 337-μm wavelength at low temperature. The field position of the transition as well as the positions of absorption peaks arising from the Shubnikov-de Haas effect were analyzed in terms of newly determined band parameters.

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In this paper, we report the first observation of the magnetic-field-induced semimetal-semiconductor transition in Bi under very high magnetic fields in the megagauss range. When high magnetic fields are applied parallel to the binary or bisectrix axes in Bi, the spacing between the lowest Landau levels of the conduction band for light electrons and the valence band at the L point first decreases because of the large spin-orbit interaction. However, above about 10 T, it starts to increase because of the band repulsion.[1,2] Thus at high enough field, the overlap between the conduction band at the L point and the hole band at the T point vanishes, and so the magnetic-field-induced semimetal-semiconductor (SM-SC) transition is expected to occur.[3,4] For Bi-Sb alloys, in which the overlap between the conduction and the hole bands is smaller than in Bi, such a SM-SC transition has been actually observed in magnetic fields below 50 T.[4,5] In pure Bi, the transition field has been predicted as close to 150 T (1.5 mG) by Brandt, Svistova, and Kashirskii.[3] Near the SM-SC transition, the possibility of an electronic phase transition such as the excitonic phase transition[6-8] or the gas-liquid type phase transition[9] has been proposed. The recent progress of the megagauss field technique has enabled us actually to investigate the SM-SC transition in Bi.

The megagauss magnetic fields were generated by an electromagnetic flux-compression technique.[10] The magnetic fields rose from the level of about 20 T to the megagauss range in several microseconds. Because of the difficulty of measuring the dc magnetoresistance in the megagauss fields, we measured the far-infrared magnetotransmission which monitors the ac resistivity. An HCN laser was employed as a radiation source for a wavelength of 337 μm. The transmitted radiation was detected by a GaAs photoconductive detector cooled to liquid-helium temperature. The measurements were performed in the temperature range between 4.5 and 8.8 K. For refrigerating the samples in the megagauss fields, we employed a specially designed sample holder made of a double glass tube.[11] By the flowing of a large amount of liquid helium in the gap between the two glass tubes surrounding the sample, the temperature of the sample was lowered to near 4 K. The unpolarized far-infrared radiation was focused by mirrors on the sample with a spot size of about 3 mm. In order to obtain a good signal-to-noise ratio of the detected light signal, a large area of the sample was needed. In contrast, for obtaining a high magnetic field, it was necessary to use a sample holder as small as possible, so that the sample size was limited. As a result of compromise, measurement was performed on samples with a diameter of 3-4 mm, and the outermost diameter of the sample holder was 5-6 mm. The temperature of samples was monitored by using an iron-gold thermocouple. The increase of the sample temperature due to the eddy current during the measurement was theoretically calculated and estimated to be less than 4.7 K. The intensity of the magnetic fields was measured by using a pickup coil. The transient signals of the radiation and the magnetic field were recorded by a two-channel transient recorder. The magnetic fields were applied parallel to the binary axis.

Figure 1 shows a typical example of the recorder trace for the magnetotransmission and the field intensity. A remarkably sharp increase of the transmission of the far-infrared radiation was observed at 86 T for this sample. In trying a number of measurements, enough field intensity as well as enough transmitted radiation were obtained in only four shots. All of these four shots showed a similar sharp rise of the transmission,

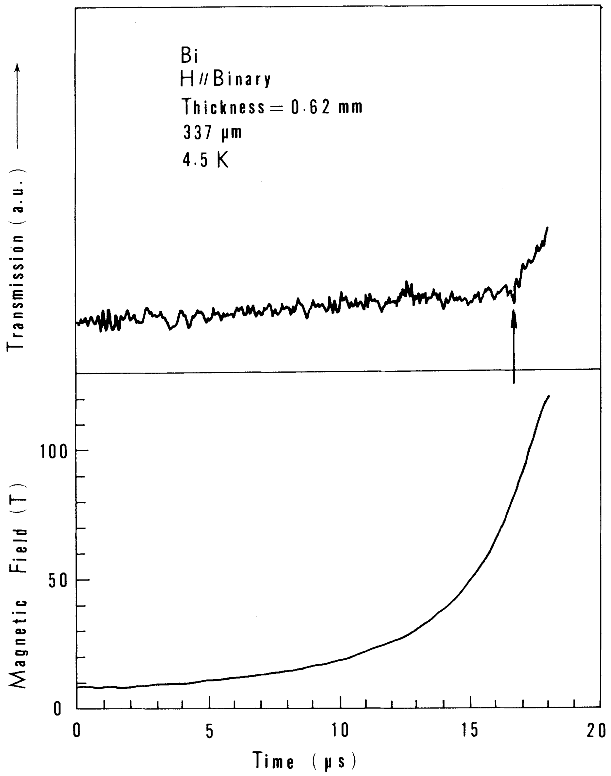

FIG. 1. Experimental recording exhibiting the magnetic-field–induced semimetal-semiconductor transition in Bi. At the position shown by an arrow, the transition occurs.

demonstrating good reproducibility. The field intensity where the sharp rise occurred varied within the range between 85 and 91 T, as listed in Table I.

The observed sharp rise of the far-infrared transmission at 88 T corresponds to the SM-SC transition, since the far-infrared magnetotransmission generally reflects the transverse magnetoresistance of the sample. In fact, in the study of the Alfvén-wave transmission in Bi under nondestructive magnetic fields below 50 T, we found that the envelope of the Fabry-Pérot interference pattern due to the Alfvén-wave transmission is modulated by a curve reflecting the Shubnikov–de Haas effect.[12] This is because both the high-field conductivity and the free-carrier absorption of the far-infrared radiation in the sample are proportional to the scattering rate of free carriers.[13] For further confirmation of the relation between the magnetotransmission and the magnetoresistance, we measured both quantities on a sample of $Bi_{95.1}Sb_{4.9}$ in a little lower nondestructive fields up to 42 T. In this sample, the SM-SC transition occurred at about 29 T. It was found that the magnetoresistance starts to increase rapidly at 29 T where the SM-SC transition occurs. Correspondingly, the magnetotransmission showed a sharp rise at nearly the same field.

TABLE I. List of the successful experiments to observe the semimetal-semiconductor transition in Bi ($H \parallel$ binary).

No.	Thickness of sample (mm)	Starting temperature[a] (K)	Transition field (T)
1	0.62	4.5	86
2	0.62	4.8	85
3	0.40	8.8	91
4	0.40	7.8	90
average			88

[a] Because of the eddy current, the sample temperature rises by a few degrees from the starting temperature during the measurement.

Besides the SM-SC transition, some structures were also observed in the magnetotransmission for Bi below 90 T. The extent of the transmission at lower fields varied considerably among the measurements, depending upon the quality of samples, the sample thickness, the sample temperature, the alignment of the optical path, etc. Figure 2 shows transmission versus magnetic field for the recordings when fairly large transmission was obtained. The scale of the ordinate was magnified considerably in comparison to Fig. 1. The top trace (*a*) was obtained in nondestructive fields in which measurement can be performed with a better signal to noise ratio. The Fabry-Pérot interferograms are seen on this line.[12] Modulating the interferograms, absorption peaks are seen. On the lines *b*–*d* for higher-field experiments, several absorption peaks can also be seen in spite of a rather large noise. The peak designated as CO2 is a peak arising from the combined resonance of holes. The peaks L0–L3 correspond to the Shubnikov–de Haas effect of holes: namely, the 0th to 3rd Landau levels for holes cross the Fermi level at these respective magnetic fields. The peak L0 corresponds to the SM-SC transition. The positions of the peaks observed below 40 T agreed well with the results of more accurate measurements of the oscillatory magnetoresistance[14,15] and the magnetotransmission[12] in a nondestructive field: i.e., the CO2, L3, L2, and L1 appeared at 11, 14, 25, and 42 T, respectively.

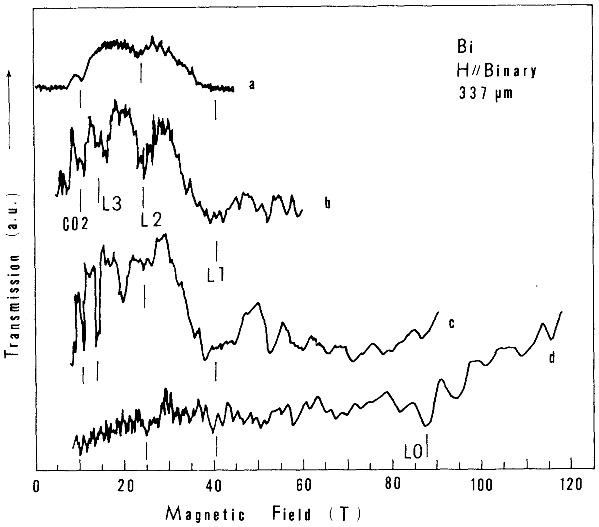

FIG. 2. Magnetotransmission as a function of magnetic field for samples of Bi which showed relatively good transmission. The top line (*a*) was obtained in a nondestructive pulsed field. The thickness and temperature of the samples are for curve *a*, 1.61 mm, 6.8 K; *b*, 0.60 mm, 4.2 K; *c*, 0.40 mm, 8.8 K; *d*, 0.62 mm, 4.5 K.

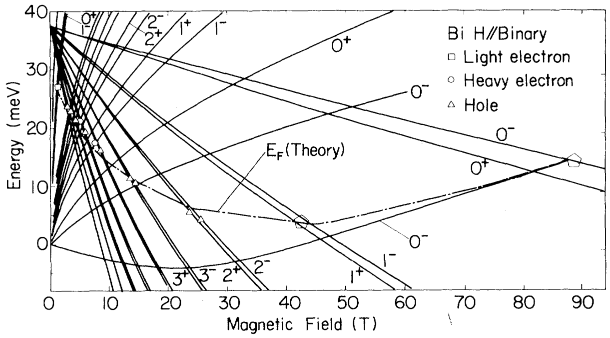

FIG. 3. Landau levels of electrons and holes in the Fermi level in Bi as functions of magnetic field. Theoretical lines were calculated on the basis of the model of Vecchi, Pereira, and Dresselhaus (Ref. 1) with newly determined parameters. The experimental points were obtained by the measurement of Shubnikov-de Haas effect. The points represented by pentagons were obtained by the present experiment.

The field positions of the SM-SC transition and the Shubnikov–de Haas peaks were compared with a calculation of the Landau levels and the Fermi level of Bi. For this purpose, the calculation of the lowest Landau level of light electrons is required. Vecchi, Pereira, and Dresselhaus calculated the dispersion of the lowest Landau level, and determined the band parameters in the dispersion by the measurement of the magnetoreflection up to 15 T.[1] According to their calculation, the lowest Landau levels for light electrons and holes should cross each other at about 40 T, and only a small amount of heavy electrons and holes should remain above this field. However, our Shubnikov–de Haas experiment at lower field showed that all heavy electron levels go above the Fermi level above about 15 T, and so the SM-SC transition should occur as soon as the lowest Landau levels of light electrons and holes cross each other. This indicates the necessity of a determination of band parameters which are relevant in the ultraquantum limit.

In our calculation, the band parameters for holes and heavy electrons were determined from the analysis of the Shubnikov–de Haas effect[14-16] and the magnetoreflection[17] on the assumption of the two-band model. The band parameters for light electrons were determined on the basis of the model of Vecchi, Pereira, and Dresselhaus.[1] The details of the calculation and the determined parameters will be reported elsewhere.[15] Figure 3 shows the calculated Landau levels and the Fermi level for the band parameters leading to the best fit with the experimental results. As is shown in Fig. 3, the experimentally observed field positions of the SM-SC transition and the Shubnikov–de Haas peaks agree well with the calculation.

As for the electronic phase transition such as excitonic phase transition or the gas-liquid type transition, we have not observed so far any sign of phase transitions. The small temperature dependence of the SM-SC transition as shown in Table I should be considered as a scatter of the experimental data because of the error involved in the field measurement (± 3%). The various types of electronic phase transitions are predicted to occur at very low temperatures, below 0.1 K, in a magnetic field of about 10 T, where the Landau levels of $N = 1$ play an important role. In much higher magnetic fields as attained in the present experiment, we can expect the possibility of such phase transitions at reasonably higher temperature. In fact, the binding energy of excitons in Bi becomes significantly large in the field range near 88 T. The dimensionless parameter γ for excitons representing the relative importance of the magnetic field is given by

$$\gamma = \hbar\omega_c/2R^* = \frac{H(T)}{2.35\times10^5}\left(\frac{m_0}{m^*}\kappa\right)^2, \qquad (1)$$

where R^* is the binding energy of excitons, κ is the dielectric constant, and m^* is the reduced mass of excitons. If we take $m^* = 0.036 m_0$ estimated from the cyclotron mass of electrons and holes at 88 T and $\kappa = 100$, we obtain $\gamma = 2890$ at 88 T. The binding energy of excitons is then estimated to be 4.6 meV.[18] Thus it is worthwhile to investigate in more detail this interesting problem of the phase transition which might occur near the gapless state in very high magnetic fields.

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Anisotropic Superconducting and Magnetic Properties of a Single Crystal of ErRh$_4$B$_4$

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The magnetic response of a single crystal of the tetragonal compound ErRh$_4$B$_4$ in the $\bar{c}$ (hard) and $\bar{a}$ (easy) directions is presented. Extreme anisotropy is found in the magnetic and superconducting behavior, allowing a natural separation of the bare superconducting properties from those affected by magnetism. Below T_{c2} the data reconcile the discrepancy between the ferromagnetic moment per erbium atom measured by neutron scattering and by Mössbauer effect.

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Reentrant superconductivity found in the stoichiometric compounds ErRh$_4$B$_4$ and HoMo$_6$S$_8$ provides a unique opportunity to study the interaction of superconductivity and ferromagnetism.[1,2] As the temperature is lowered, ErRh$_4$B$_4$ transforms through four distinct phases: paramagnetic for $T > T_{c1} = 8.6$ K, superconducting for $T_{c1} \gtrsim T \gtrsim 1.2$ K, coexistence for $1.2\ \mathrm{K} \gtrsim T > T_{c2} = 0.7$ K, and normal ferromagnetic for $T < T_{c2}$. Considerable work on polycrystalline specimens has yielded much information on the thermal,[3] paramagnetic,[1,4] superconducting,[5-8] and ferromagnetic behavior[9,10] of ErRh$_4$B$_4$. Recently, neutron scattering from a single crystal of ErRh$_4$B$_4$ has provided detailed information on the coexistence phase.[11] However, none of this work has examined the important role that anisotropy plays in determining the superconducting and magnetic properties of the various phases.

In this paper we report on the magnetic response of a single crystal of the tetragonal compound ErRh$_4$B$_4$ in the $\bar{c}$(hard) and $\bar{a}$(easy) directions. Both ac susceptibility and dynamic dc susceptibility and magnetization data were obtained in fields between 0 and 20 kOe and temperatures between 0.4 and 15 K. The ac measurements were obtained with a mutual inductance bridge operating at 79 Hz with an excitation field of 1 Oe. The dc measurements utilized the net signal from a balanced pair of opposing coils, one containing