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Magnetic-field-induced electronic phase transitions in semimetals in high magnetic fields

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Abstract. We report an experimental study of the magnetic-field-induced electronic phase transitions in semimetals (graphite and Bi) in pulsed high magnetic fields up to several hundred teslas. Far-infrared and millimetre-wave spectroscopies were employed to investigate the electric conductivity and the optical transitions between the Landau levels. In graphite a phase transition was observed in the mm-wave transmission through a specially designed strip-line system with a sample on one of the walls. Evidence of a density-wave phase transition was found in the infrared transmission at a transition field of about 30–40 T. Cyclotron resonance in very high field revealed that the $n = 0$ spin-up level is depopulated above a field of $B = 54$ T, while the $n = 0$ spin-down level persists under the Fermi level at least up to 200 T. In Bi, anomalous structures were observed in the strip-line mm-wave transmission spectra as well as the infrared transmission spectra, indicating a semimetal-to-semiconductor transition at around 85 T.

1. Introduction

In very high magnetic fields, the quantization of the electronic energy levels of conduction electrons becomes so prominent that various kinds of electronic phase transition are expected to take place. Recent advances in ultra-high-magnetic-field techniques have enabled us to perform experiments in a magnetic field of several megagauss (∼500–600 T) [1]. It is believed that very high magnetic fields of the order of 10^8–10^{15} T exist in astronomical matter such as neutron stars. Electronic states in such high magnetic fields have been theoretically argued for, and the existence of unusual states has been predicted. The relative importance of the magnetic field effect against the Coulomb energy is represented by the so-called γ-factor, which is the ratio of the cyclotron quantization energy of the ground state to the Rydberg energy. The energy states of interacting electrons in high magnetic fields are characterized by this factor. The magnetic field where γ becomes unity is $B = 2.3 \times 10^5$ T for hydrogen atoms, but in solids it is reduced to a much smaller value by a factor of $(m^*/\kappa)^2$, where m^* is the reduced mass of the positive and negative charges and κ is the dielectric constant.

Graphite and bismuth are semimetals whose electronic states are sensitively altered by the application of high magnetic fields due to the small effective masses. In the extreme quantization limit of the electronic states under sufficiently high magnetic fields, a field-induced semimetal–semiconductor transition or various types of electronic phase transition

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have been predicted. We have actually observed these transitions in both types of crystal by infrared, far-infrared, and mm-wave spectroscopy in very high magnetic fields up to 500 T. The high magnetic fields were produced either by electromagnetic flux compression (up to 500 T), the single-turn coil technique (up to 200 T), or non-destructive long-pulse magnets (up to 50 T) [1].

Graphite is known to exhibit a striking jump of the magneto-resistance at around 30–40 T [2, 3]. As the critical field depends on temperature, it has been explained as a charge-density-wave (CDW) phase transition by Yoshioka and Fukuyama (YF) [4]. They predicted a re-entrant nature of the phase transition. In a previous experiment on the magneto-resistance up to 52 T, a signature of the re-entrant nature of the phase transition was found [5]. Moreover, fine structures were observed in the magneto-resistance in the high-resistance state, suggesting other transitions with different nesting vectors. Theoretically, we can expect many more transitions in even higher fields. Therefore, it is important to measure the magneto-resistance in very high magnetic fields. Very recently, Yaguchi and Singleton observed a re-entrant phase transition by measuring the magneto-resistance up to 55 T [6]. A question arises from the fact that the YF theory assumed a nesting vector along the c-axis, whereas the measurements were performed in a plane perpendicular to the c-axis. Iye and Dresselhaus found non-linear transport phenomena typical of the pinned charge-density wave [7]. It is questioned whether the density wave with a wave-vector along the c-axis would give rise to a large change in the perpendicular direction. Yaguchi *et al* measured the conductivity in the c-direction, and found a different type of phase transition from that in the perpendicular direction including non-Ohmic conductivity [8]. It is important to obtain further evidence of the charge-density wave by means other than magneto-resistance. Takamasu and Miura found a photoconductivity in the mm-wave range and ascribed it to the excitation across the gap formed at the Fermi level [9].

Bi is another type of semimetal that has a conduction band bottom at the L point and the valence band top at the T point of the Brillouin zone. When we apply very high magnetic fields along the binary axis, a magnetic-field-induced semimetal-to-semiconductor transition takes place [10, 11]. Near the magnetic field where the band gap crosses zero, the field-induced excitonic phase transition may occur at low temperatures [12]. Moreover, the magneto-resistance exhibits an anomalous decrease under the quantum limit in high fields [11, 13]. Therefore, more detailed measurement of the magneto-resistance should be investigated to clarify the high-field properties of Bi.

In the present work, we investigated the magneto-absorption spectra for single-crystal graphite and Bi in high magnetic fields up to 400 T in the infrared, far-infrared, and mm-wave ranges. In the infrared range, we have observed various excitation spectra. In the mm-wave range, the measured the AC conductivity should be very close to the DC conductivity. The DC transport measurement is not an easy task in short ultra-high magnetic fields, because of the large induced voltage across the lead wires to the sample. Therefore, the mm-wave conductivity measured by means of the strip-line system should provide valuable information concerning the magneto-resistance of both materials.

2. Experimental procedure

In pulsed high fields, the transmission of infrared and far-infrared radiation was measured using molecular gas lasers. The radiation in the wavelength range $\lambda = 9$–$11\ \mu$m was obtained from a CO_2 laser, that in the range 5.3–5.7 μm from a CO laser, that at 3.39 μm from a He–Ne laser, lines at 16.9 and 28 μm from a pulsed H_2O laser, and that in the range 37–305 μm from a CO_2-laser-pumped far-infrared laser. For detecting the rapid change of the radiation, a photovoltaic detector of HgCdTe was used for 3.39 μm $< \lambda <$ 11 μm, an extrinsic photoconductivity of Ga-doped Ge (cooled to liquid He temperature) for 37 μm $< \lambda <$ 119 μm, and an extrinsic photoconductivity of GaAs for 200 μm $< \lambda <$ 305 μm.

The high-frequency conductivities in the mm-wave range at $\lambda = 2$ mm and 3 mm were also measured. The radiation was obtained from Gunn diodes and from a Carcinotron tube. The detector for the mm-wave radiation was a Ge diode. In order to obtain a sufficient coupling between the sample and the mm-wave radiation, we developed a strip-line transmission line [14] as shown in figure 1 [15].

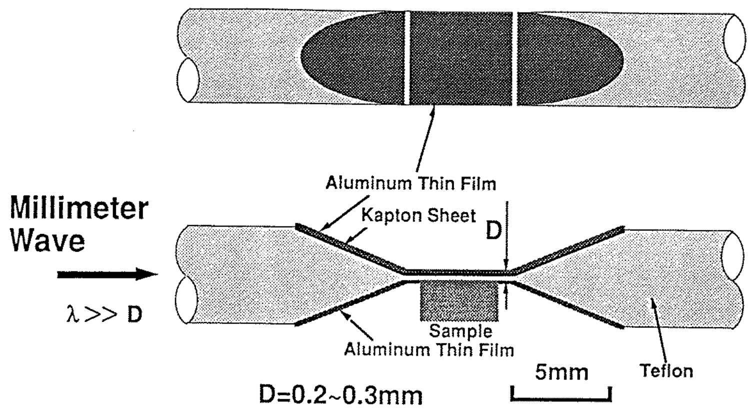


Figure 1. The strip-line system for the short-pulse magnetic field.

The guide of the radiation with a round cross-section and the part that was a thin flat plate were made from Teflon. On one side of the flat plate, the sample was mounted. The other side was coated with a copper foil. The thickness of the flat plate was about 200–300 μm, which was sufficiently smaller than the wavelength of the radiation (2–3 mm). In such a configuration, the transmitted intensity of the radiation reflects the conductivity along the direction of propagation of the radiation.

The samples of graphite were Kish graphite (single-crystal graphite). We cleaved the crystal to obtain a suitable thickness of the order of about 100 μm. For the strip-line transmission experiments, we used 'super-graphite' which has enough thickness to mount orienting the c-axis in the direction of the magnetic field. The samples of Bi were single crystals grown by the Czochralski method. The samples were cut by an etching cutter and polished with etching solution.

Pulsed high magnetic fields were produced by three different techniques: electromagnetic flux compression up to 500 T, the single-turn-coil technique up to 150 T, and non-destructive long-pulse magnets up to 50 T. The rise time of the first one was about 5 μs from 100 T to 500 T. The durations for the second and the third systems, which are nearly sinusoidal, were 7 μs and 15 ms, respectively, from zero to zero through the maximum. Liquid-He-flow-type cryostats were employed to refrigerate the sample.

3. Results and discussion

3.1. Graphite

3.1.1. Millimetre-wave strip-line spectra. First we discuss the mm-wave strip-line spectroscopy in the mm-wave range. The AC conductivity was measured by the strip-line transmission system as described in section 2. The magnetic field was applied parallel to the *c*-axis of the super-graphite. Figure 2 shows the traces of the transmitted signal measured in long-pulse fields up to 42 T at a frequency of 146 GHz ($\lambda = 2.05$ mm).

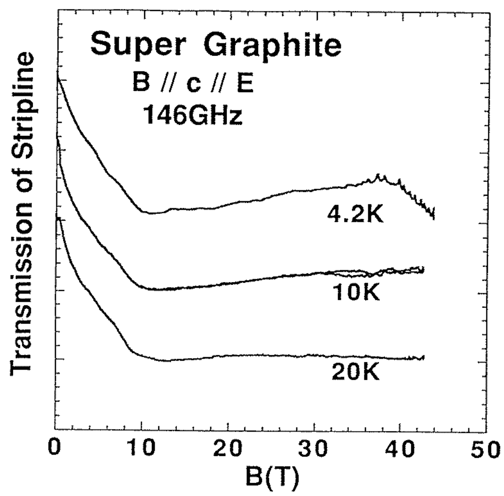


Figure 2. Strip-line transmission spectra of super-graphite at a frequency of 146 GHz at different temperatures.

The signal is considered to correspond to the conductivity along the *c*-axis. The transmission first decreases abruptly with increasing field, corresponding to the increase of the magneto-resistance up to about 10 T. At a temperature of 4.2 K, the transmission shows a drastic decrease at 38 T. This is considered to be due to the phase transition at this temperature. The curves for 10 K and 20 K showed no such sharp decrease up to 42 T. This result clearly demonstrates that the measuring system works well as regards detecting the magneto-resistance and observing the phase transition. The measurement was extended to the higher field produced by the single-turn-coil technique. Figure 3 shows the transmission spectra at 100 GHz ($\lambda = 3.0$ mm) up to 100 T.

The spectra are consistent with those shown in figure 2 up to $\sim$40 T. At temperatures of 5.1 K and 9.8 K, sharp structures are observed at 42 T and 50 T, respectively. These correspond to the first phase transition which we have observed in the long-pulse fields. Any subsequent phase transitions which might occur in higher fields should be visible in the spectra. In the higher fields, however, we can see no significant structure. The second transition associated with the re-entrant feature is not discernible, either. The reason for the absence of the re-entrant transition may be the temperature rise of the sample caused by the Joule heating. The temperature rise in the short-pulse fields is estimated to be about 5 K. At higher temperatures a broader structure is observed at around 55 T. This is probably due to the Shubnikov–de Haas peak at a field where the (0+) level crosses the Fermi level. To observe phase transitions which might take place in the megagauss range, we would have to make measurements on a smaller sample to suppress the Joule heating effect.

3.1.2. Infrared magneto-plasma effect. Next, we present the results for the infrared absorption due to the magneto-plasma effect in single-crystal graphite. The reflection and absorption show a large degree of structure when the dielectric constant crosses zero due to the magneto-plasma effect. Generally, the infrared reflection spectrum takes a minimum

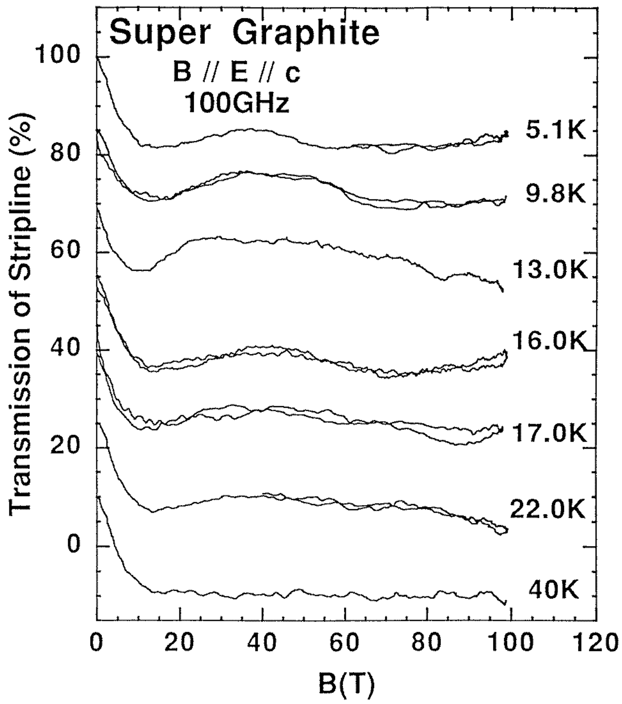


Figure 3. Strip-line transmission spectra of super-graphite at a frequency of 100 GHz observed in short-pulse fields up to 100 T.

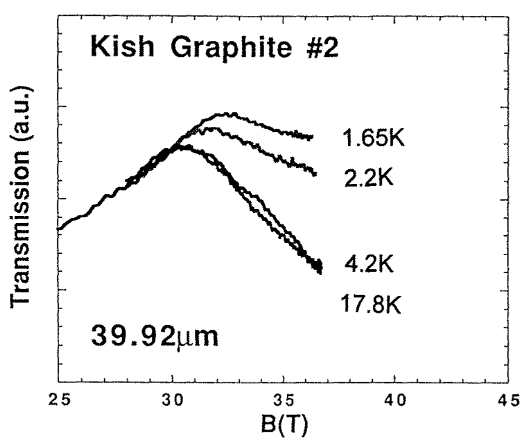


Figure 4. Infrared transmission spectra of single-crystal graphite at a wavelength of 39.92 μm at different temperatures.

at the zero-crossing point, while the transmission takes a maximum. Nakamura *et al* found in graphite an electron–hole coupled-magneto-plasma mode whose energy increases with increasing magnetic field [16]. It was found that the electronic transition between the $(-1\pm)$ and $(0\pm)$ subbands gives the dominant contribution to this plasma mode [17]. We measured the magneto-transmission spectra at photon energies for which the magneto-plasma peak appears at magnetic fields near the phase transition. Figure 4 shows the transmission spectra at a wavelength of 39.92 μm.

A large peak at 30–33 T corresponds to the zero crossing of the dielectric constant. The peak clearly shows a shift to higher fields and broadening as the temperature is lowered across the phase transition temperature. At 37.15 μm, the temperature dependence was also observed at low temperatures where the phase transition takes place. However, at a wavelength of 57 μm for which the plasma peak appears at a much lower field of 25 T, no temperature dependence was observed. That is, the plasma peak shifts to higher fields by a few teslas when the temperature is varied only in the field range where the phase transition occurs.

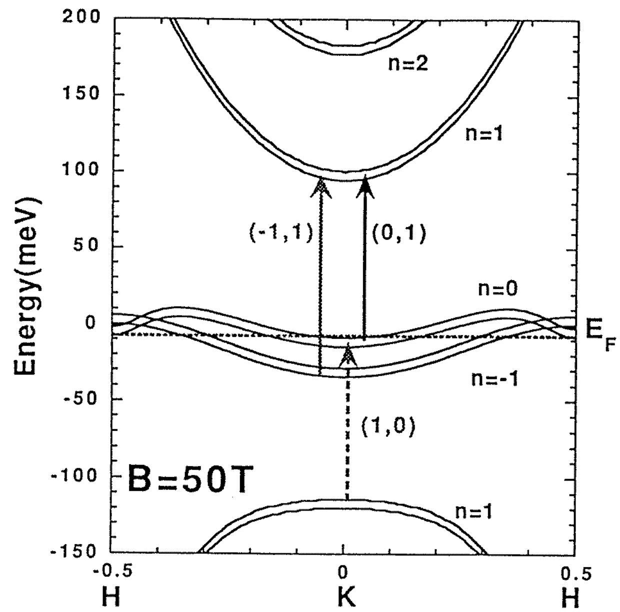


Figure 5. The energy of the Landau subbands for magnetic fields of 50 T applied parallel to the c-axis. The solid and broken lines indicate the electronic transitions for electron-active and hole-active polarizations, respectively.

The above results are interpreted in terms of the suppression of the electronic transition between the (0±) and (−1±) subbands in the CDW phase. It is reasonable if we consider that the density-wave gap opens up in one of the Landau subbands where the nesting occurs. The density-wave gap is estimated to be of the order of

$$2\Delta = 3.5 k_B T_c \sim 1 \text{ meV} \tag{1}$$

assuming a BCS-type relation, where T_c is the transition temperature. The coherent state in the density-wave phase comprises electronic states in the vicinity of the Fermi level with a width δk in the k-space:

$$\delta k \sim \frac{1}{\xi} \tag{2}$$

where ξ is the coherence length of the density wave, represented by

$$\xi \sim \frac{2\Delta}{\hbar v_F} \tag{3}$$

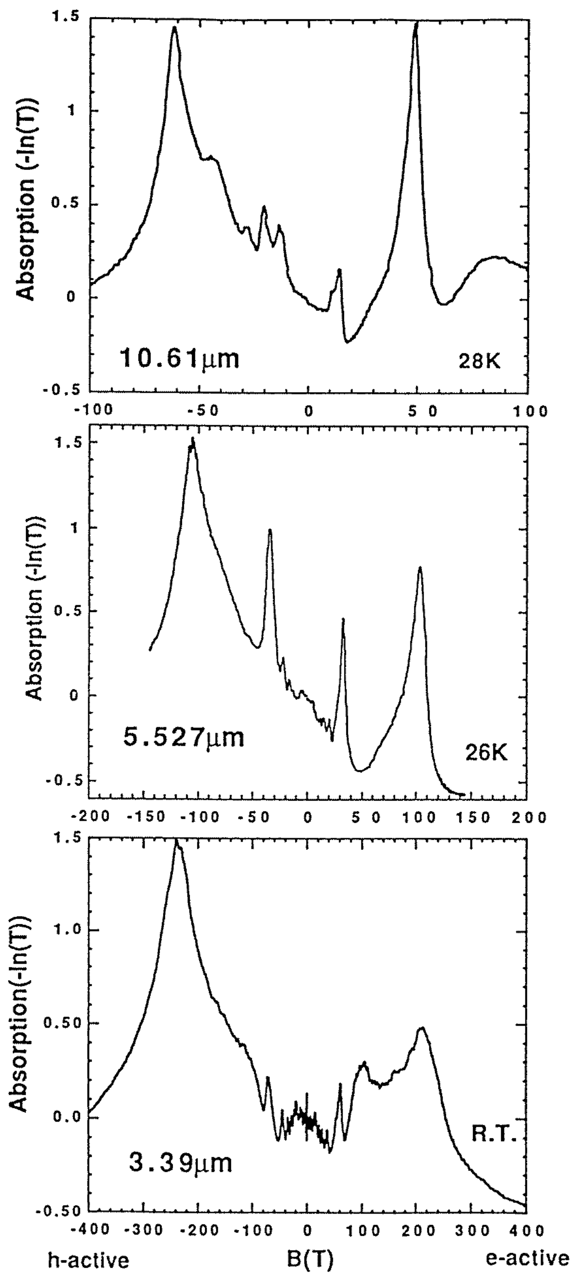


Figure 6. Cyclotron resonance spectra for single-crystal graphite. The abscissa of each panel is adjusted so that the same transition is seen at nearly the same position.

using the Fermi velocity v_F. The energy band dispersion of the $n = 0$ and $n = -1$ subbands is represented by a linear approximation:

$$E = \hbar v_F (k - k_F). \tag{4}$$

The range of the k-vector, δk, where the $n = -1$ to $n = 0$ transition occurs is given by

$$\delta K \sim \frac{\delta E}{\hbar v_F} \tag{5}$$

where δE denotes the interval between the $n = 0$ and $n = -1$ subbands, which is about 12 meV at around 30 T. Thus the $n = -1$ to $n = 0$ transition would be suppressed by the factor

$$\frac{\delta k}{\delta K} \sim \frac{\hbar v_F}{2\Delta\, \delta K} \sim \frac{\delta E}{2\Delta} \sim 10\%. \tag{6}$$

It can be shown by a quantum mechanical calculation of the optical density that if the $n = -1$ to $n = 0$ transition probability decreased by 10%, in association with the phase transition, the plasma peak would shift to a higher field by a few teslas. Thus we can explain the anomalous temperature effect of the magneto-plasma peak on the basis of the density-wave model.

3.1.3. Cyclotron resonance. Finally, we discuss the cyclotron resonance in graphite at very high magnetic fields. Figure 5 shows the Landau levels of graphite at $B = 50$ T, calculated using the Slonczewski–Weiss–McClure (SWM) model [18, 19], and the allowed optical transitions with circular polarization [20].

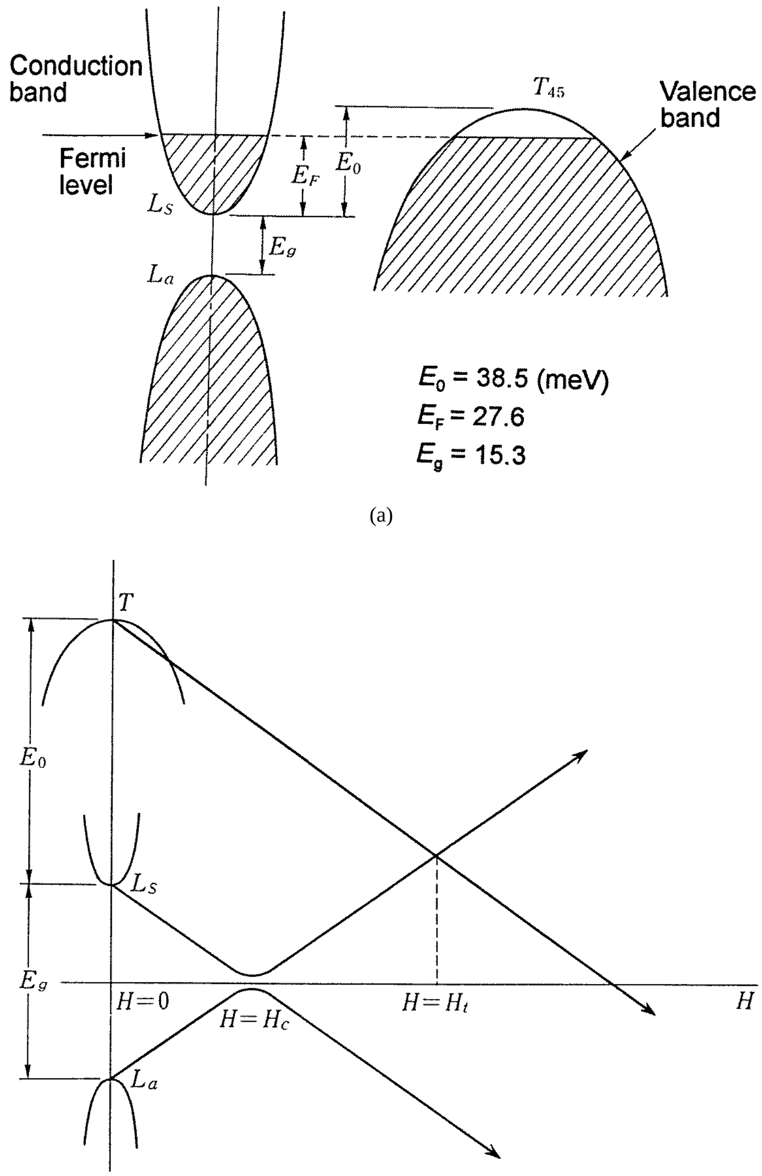
In the infrared range, such transitions are actually observed. Figure 6 shows the infrared absorption spectra at wavelengths of 10.61 μm, 5.527 μm, and 3.39 μm.

Here the spectra for both right and left circular polarizations are shown. In other words, ‘electron-active’ and ‘hole-active’ denote the former and the latter polarizations, respectively. Clear cyclotron resonance lines were observed for both of the polarizations. Comparing the photon energy with the Landau levels calculated using the SWM model and with the low-field data, we can assign the peaks to the electronic transitions as listed in table 1.

Table 1. Resonance fields and their assignment for the principal resonance absorption lines in graphite at various wavelengths.

Polarization	Wavelength (μm)	Photon energy (meV)	Resonant field (T)	Transition
Electron-active	10.61	116.9	48	$(0\pm) \to (1\pm)$
Hole-active	10.61	116.9	43	$(-1\pm) \to (1\pm)$
Hole-active	10.61	116.9	62	$(1+) \to (0+)$
Electron-active	5.527	224	103.5	$(0-) \to (1-)$
Hole-active	5.527	224	105.8	$(1+) \to (0+)$
Electron-active	3.39	336	208	$(0-) \to (1-)$
Hole-active	3.39	336	240	$(1+) \to (0+)$

A remarkable feature is seen for the transitions with which the $0\pm$ levels are associated. Namely, the $(1+) \to (0+)$ transition is observed at 62 T for the hole-active polarization. This fact indicates that the $(0+)$ level is already depopulated below 62 T. On the other hand, the observation of the $(1+) \to (0+)$ transition at 208 T for the other polarization gives strong evidence that the $(0-)$ level is still populated up to 208 T. In other words, the $(0-)$ level should cross the Fermi level at a much higher field. This is a striking result at first sight if we consider that the spin splitting should be small, as expected from $g \sim 2$. If one of the 0 levels $(0+)$ crosses the Fermi level, below 62 T, the spin counterpart of the



(b)

Figure 7. (a) The energy band structure of Bi. (b) The shift of the band extrema for the magnetic field applied parallel to the binary axis.

same level (0−) should cross the Fermi level at a field not so much higher than this field. In addition, this casts doubt on the validity of the YF theory which assumes nesting in the (0+) level, because the critical field of the phase transition should be of comparable order with 62 T at low temperatures.

Very recently, Takada and Goto calculated the renormalized Landau levels taking account of the self-energy corrections based on the SWM model [21]. According to their theory, the spin splitting is enhanced and the (0+) level crosses the Fermi level at about 53 T,

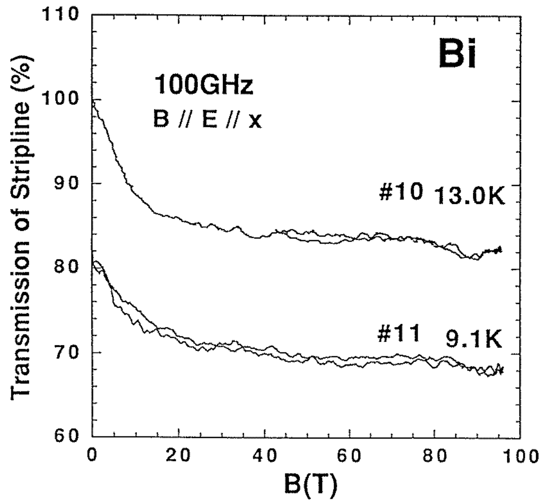
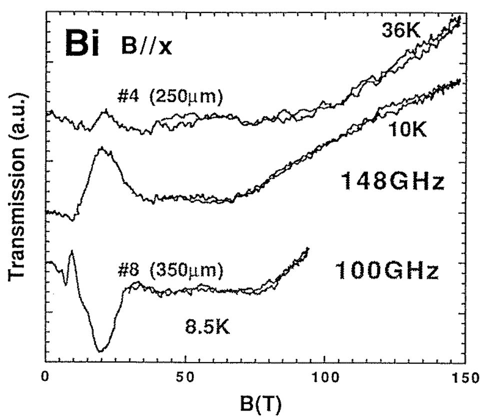


Figure 8. Strip-line transmission spectra of Bi for $\boldsymbol{B}$ ∥ binary axis for two different frequencies.



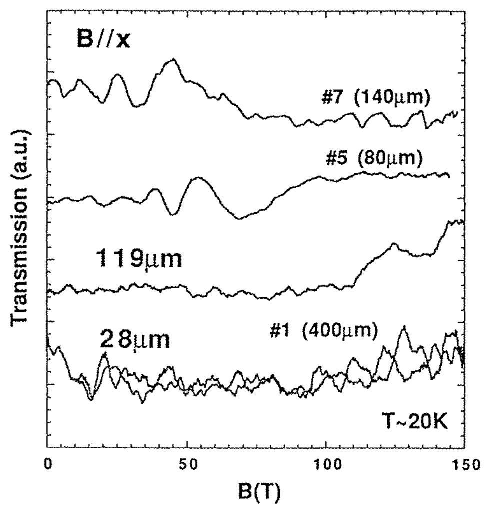


Figure 9. Millimetre-wave transmission spectra of Bi for $\boldsymbol{B}$ ∥ binary axis. The frequency is 100 GHz.

Figure 10. Transmission spectra of Bi for wavelengths of 28 μm and 119 μm.

whilst the (0−) level remains below the Fermi level up to a much higher field. In addition to the large splitting between the two spin-split zero levels, they also predicted that the Fermi energy measured from the bottom of the (0+) level collapses rapidly when the bottom approaches the Fermi level. These calculated features explain the present observations.

3.2. Bismuth

3.2.1. Millimetre-wave strip-line spectra. For Bi, it is known that the overlap between the conduction band at the L point and the valence band at the T point is $E_0 = 38.5$ meV as shown in figure 7(a).

When we apply magnetic fields along the binary axis, the energy of the conduction band (L_s) first decreases because the spin mass is smaller than the orbital mass, but anti-crossing occurs at $H = H_s$ above which the L_s band starts increasing. In a high enough field ($H = H_t$), the L_s band crosses the valence band, and the semimetal-to-semiconductor transition should occur, as shown in figure 7(b) [11]. The non-resonant high-frequency conductivity in Bi was measured by means of the strip-line transmission in magnetic fields applied parallel to the binary axis. Figure 8 shows the transmission as a function of magnetic field up to 100 T at 13 K and 9.1 K.

The transmission decreases with increasing field below about 20 T. This corresponds to the increase of the magneto-resistance. A change corresponding to the decrease of the magneto-resistance from about 30 T is only vaguely seen in the figure. A small but clear kink is observed at around 85 T. This kink corresponds to the semimetal-to-semiconductor transition observed in the previous far-infrared transmission experiments [10].

The anomaly at around 85 T was also observed in the direct transmission of mm waves at wavelengths of 2 and 3 mm, as shown in figure 9.

The transmission of the far-infrared and mm waves shows different spectra for different samples, depending on the thickness, temperature, and sample mounting. This is due to the interference effect. Figure 10 shows the transmission spectra at wavelengths of 119 μm and 28 μm. An oscillatory change due to the Alfvén-wave interference was observed up to high magnetic field at a wavelength of 119 μm. The oscillation was found to disappear at around 80 T [15], which is further indirect evidence of a semimetal-to-semiconductor transition. Evidence of the excitonic insulator phase has not been observed yet. However, the high-field state in Bi in ultra-high magnetic fields is still an interesting subject.

4. Conclusions

We have measured magneto-absorption and strip-line transmission spectra of infrared, far-infrared, and mm waves for graphite and Bi under high magnetic fields in the megagauss range. It was confirmed that a newly constructed strip-line system can measure the AC magneto-resistance in short-pulse fields. We observed the phase transition in graphite by using the strip-line system. We found that the plasma peak position is changed by the phase transition, consistently with the density-wave model. For cyclotron resonance in very high fields, it was found that the (0+) level crosses the Fermi level below 64 T, while the (0−) level remains below the Fermi level even at 208 T. In Bi, the field-induced semimetal-to-semiconductor transition was observed at about 85 T both in the magneto-absorption and in the strip-line transmission.

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