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High-magnetic-field electronic phase transition in graphite observed by magnetoresistance anomaly

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The magnetoresistance of pure graphite has been measured at low temperatures down to 0.48 K, under a steady magnetic field up to 28.5 T with the use of a hybrid magnet. A striking anomaly in the magnetoresistance was found at fields above 22 T. The fact that the critical field of the anomaly has a strong temperature dependence suggests the onset of an electronic phase transition, and makes an explanation of this phenomenon in terms of known one-electron properties of graphite unlikely.

I. INTRODUCTION

The electronic properties of semimetals at high magnetic fields have been the subject of a number of investigations since the early years of solid-state physics.[1] Because of the small carrier masses, high mobilities, and low carrier concentrations, even a moderate magnetic field can have a profound effect on the electronic motion in these materials, and the quantum limit can be attained with accessible static field strengths.

Along with the group-V semimetals (Bi, Sb, As), graphite has been studied by many investigators. The carriers of graphite occupy a narrow region along the *H-K-H* edge of the hexagonal Brillouin zone. The electronic band structure in the vicinity of the Fermi level is most successfully described by the Slonczewski-Weiss-McClure (SWMcC) model.[2,3] This model gives the energy dispersion of the four relevant π bands near the *H-K-H* edge in terms of seven parameters $\gamma_0, \gamma_1, \ldots, \gamma_5$ and Δ. Figure 1 shows the profile of the π bands in the case of $\gamma_3 = 0$ (no trigonal warping). The E_3 band is doubly degenerate along the *H-K-H* edge. Each of the π bands has a twofold spin degeneracy in addition to the twofold degeneracy associated with two nonequivalent Brillouin-zone edges *H-K-H* and *H'-K'-H'*.

Based on the SWMcC model, the Landau-level scheme of graphite in a magnetic field applied parallel to the *c* axis was calculated by McClure,[4] Inoue,[5] Dresselhaus and Dresselhaus,[6] and Nakao.[7] A unique feature of the Landau levels of graphite (see Fig. 2) is that the lowest two spin-split Landau levels, near the Fermi level indexed by $n=-2$, $\sigma=-$; $n=-1$, $\sigma=+$; $n=-1$, $\sigma=-$;

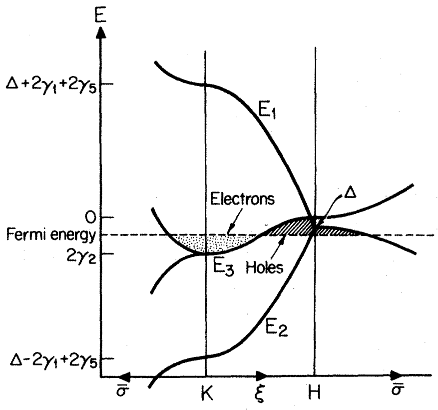

FIG. 1. Electronic energy bands of graphite in the vicinity of the *H-K-H* axis. Along the *H-K-H* axis, the E_3 band is doubly degenerate, and ξ is a dimensionless wavevector. The dimensionless wave vector perpendicular to the *H-K-H* axis is denoted by $\bar{\sigma}$, and the double degeneracy of the E_3 band is lifted away from the zone edge except on the zone boundary away from *H*. The Brillouin zone of graphite is a hexagonal prism with electron and hole Fermi surfaces located along the *H-K-H* zone edges.

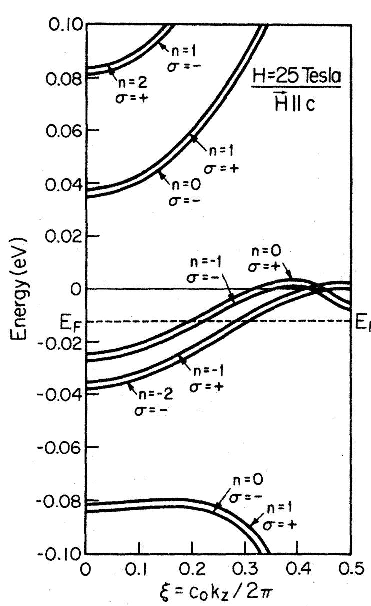

FIG. 2. The calculated Landau-level contours for graphite in a magnetic field of 25 T applied parallel to the *c* axis, using the values of the Slonczewski-Weiss-McClure band parameters $\gamma_0=3.16$ eV, $\gamma_1=0.39$ eV, $\gamma_2=-0.019$ eV, $\gamma_3=0.0$ eV, $\gamma_4=0.044$ eV, $\gamma_5=0.038$ eV, and $\Delta=-0.008$ eV, following Ref. 8. The spin-orbit parameters (see Ref. 6) are all assumed to vanish: $\lambda_{12}^z=\lambda_{33}^z=\lambda_{13}=\lambda_{23}=0$.

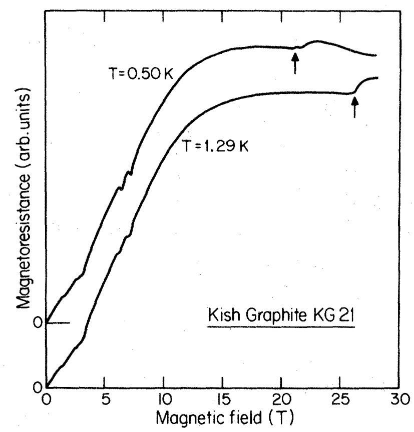

FIG. 3. Magnetoresistance of Kish graphite (KG21) at temperatures 1.29 and 0.50 K, covering a broad magnetic field range. Below 7.3 T, the oscillatory features are associated with the Shubnikov—de Haas effect. The onset points of the high-field anomaly are indicated by arrows.

and $n=0$, $\sigma=+$ (using the notation of Ref. 6), are almost insensitive to the magnetic field. The higher Landau levels $n=1$, $\sigma=+$ and $n=0$, $\sigma=-$ are found in the present experiment to pass through the Fermi level at 6.6 and 7.3 T, respectively; above this field value the magnetic Hamiltonian[8] is in the quantum limit. The Landau-level scheme at $H=25$ T (typical of the quantum limit) is shown in Fig. 2. At extremely high field, the $n=-1$, $\sigma=-$ and $n=0$, $\sigma=+$ levels at the *K* point rise above the energy of the $n=-1$, $\sigma=+$ level at the *H* point, and the extreme quantum limit is attained. However, this occurs only at fields as high as 70 T, far beyond the present magnetic field range.

The electron transport properties of graphite at high magnetic fields have been studied by several experimentalists.[9–11] Particular attention has been given to the electron transport in the layer plane, with the magnetic field parallel to the *c* axis; unless otherwise specified, this geometry is assumed in the following discussion. Figure 3 shows recorder traces of the magnetoresistance of a single-crystal graphite sample up to 28.5 T at temperatures of 1.29 and 0.50 K. The behavior of the magnetoresistance below 22 T is consistent with previous studies and is summarized as follows. In the low-field region ($0 \leq H \leq 7.3$ T), Shubnikov—de Haas oscillations are observed[12,13] with two distinct frequencies, at 6.6 T (electron pocket) and 4.8 T (majority hole pocket), corresponding to the Fermi-surface extremal cross sections.

In the quantum limit, where only the lowest two Landau levels are occupied [above 7.3 T (Ref. 14)], a linear field dependence of the magnetoresistance (first reported by McClure and Spry[9]) is observed. This linear magnetoresistance is explained by McClure and Spry in terms of a magnetic-field-dependent scattering range for the ionized impurity scattering mechanism, the dominant scattering mechanism at low temperatures.[9]

Above about 12 T, the magnetoresistance starts to saturate, as shown in Fig. 3. The saturation of the magnetoresistance was observed by Brandt *et al.*[10] who carried out pulsed-field measurements up to 52 T for temperatures as low as 2.1 K, and by Woollam *et al.*[11] who investigated a high-quality single-crystal sample in static fields up to 22 T at a temperature of 4.2 K. To explain the saturation of the magnetoresistance, Brandt *et al.*[10] and Sugihara and Woollam[15] invoked the magnetic freeze-out effect of the ionized impurity scattering centers.

The discussion above summarizes the general behavior of the magnetoresistance of graphite up to about 20 T. Recently, three of the present authors (Iye, Furukawa, and Tanuma) carried out measurements up to 40 T using a pulsed magnet and found a high-field anomaly in the magnetoresistance.[16,17] In order to elucidate this phenomenon further, we refined the measurement in the work reported in this paper by using a static field supplied by a hybrid magnet,[18] newly constructed at the Francis Bitter National Magnet Laboratory, MIT. We also extended the measurements down to the ^{3}He temperature range. The experimental setup is described in Sec. II. Discussion about the graphite samples used in this work is given in Sec. III. The results are presented and discussed in Sec. IV, and the conclusions are summarized in Sec. V.

II. EXPERIMENTAL DETAILS

The measurements of resistivity and magnetoresistance were made using the usual four-terminal dc method. Electrical contacts were achieved using silver paint. In the magnetoresistance measurement, we used a constant current of 1 mA and detected the voltage signal by a Keithley 140 dc amplifier. A magnetic field up to 28.5 T was applied in the direction parallel to the c axis of the crystal by using a hybrid magnet.[18] The hybrid magnet consists of an outer superconducting magnet generating a field up to 7 T, which is superimposed on a field up to 21.5 T generated by an inner water-cooled Bitter magnet. The diameter of the magnet bore is 33 mm.

A ^{3}He evaporation refrigerator was used to attain low temperatures down to 0.48 K. Samples were directly immersed in ^{3}He liquid. The temperature was determined by measuring the saturated vapor pressure of ^{3}He with a diaphragm-type pressure gauge (MKS Baratron).

III. SAMPLES

Three different kinds of graphite crystals were investigated: Kish graphite (KG), Madagascar graphite (MD), and highly oriented pyrolytic graphite (HOPG). Kish graphite is a single-crystal flake obtained by crystallization from molten steel.[19] The Kish-graphite samples used in the present ex-

TABLE I. Dimensions, resistivities (ρ), residual resistivity ratios (RRR), and whether the anomaly was observed for the various graphite samples measured in this work. The dimensions length, width, and thickness are denoted by l, w, and t, respectively; PG, MD, and KG denote HOPG, Madagascar, and Kish graphite, respectively. The residual resistivity ratio RRR is defined as $\rho(300\text{ K})/\rho(4.2\text{ K})$.

Sample	l (mm) ±0.5 mm	w (mm) ±0.5 mm	t (mm) ±0.01 mm	ρ ($\mu\Omega$ cm) (300 K)	RRR	Anomaly observed
PG1	3.0	2.0	0.20	34	1.7	yes
PG10	3.0	2.0	0.18	13	1.5	no
MD7	2.0	0.5	0.04	33	18	no
KG3	2.0	2.0	0.23	16	19	yes
KG8	2.0	2.0	0.15	14	38	yes
KG9	1.0	1.0	0.31	23	24	yes
KG21	1.5	1.0	0.22	10.0	23	yes

periment (supplied by Toshiba Ceramics Co., Japan) were purified by annealing in a fluorine-gas flow. A mass spectrographic analysis of the Kish samples identified impurity levels of Fe (below 1 ppm), Al, Be, Ca, Mg (no greater than 10 ppm), and Si (no greater than 100 ppm). The Kish-graphite samples clearly exhibited single-crystal Laue x-ray diffraction patterns. The Madagascar graphite is a natural single crystal, and was found to contain significant amounts of Al, Ca, Cu, Fe, Mg, Mn, and Si impurities. HOPG is a quasi-single-crystal material prepared by compression annealing of pyrolytic graphite, which is initially produced by pyrolysis of hydrocarbons. The HOPG is well oriented with respect to the c axis, but the in-plane crystallite size is of the order of 1 μm.[20] An assay of the impurity concentration in similar HOPG samples showed Al, Cu, Mn (<10 ppm), and Si ($<$ 100 ppm) impurities.

A measure of the quality of the crystals used is given by residual resistivity ratio (RRR) defined by $\rho(300\ \mathrm{K})/\rho(4.2\ \mathrm{K})$, where the numerator and the denominator denote the in-plane resistivities at room temperature and 4.2 K, respectively. The values of RRR are listed in Table I for the samples used in the present work. In the case of Kish graphite and Madagascar graphite, the value of RRR was significantly different from flake to flake. We found RRR ranges from 10 to 38 in our Kish-graphite batch, and from 2 to 18 in our Madagascar-graphite batch. Samples with relatively high RRR values were chosen for use in the magnetoresistance measurements. We did not trim the irregular shapes of the flakes, to avoid possible damage to the sample quality. In the case of the HOPG samples, the RRR value was typically between 2 and 5. The HOPG samples were cut into a suitable size by a wire abrasive saw and cleaved to a suitable thickness. The dimensions and room-temperature resisitivities of the samples used in the magnetoresistance measurements are listed in Table I.

IV. RESULTS AND DISCUSSION

The present results for the magnetoresistance below 22 T are consistent with the previously reported data.[9–14] At higher fields, we found several new and novel features. The most striking feature is an abrupt increase of the magnetoresistance as indicated in Fig. 3 by an arrow on the 1.29-K curve. This phenomenon is the same as that first discovered in the pulsed-field experiments.[16,17]

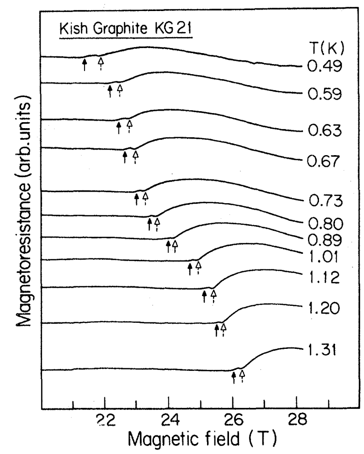

FIG. 4. High-field magnetoresistance of Kish graphite (KG21), the same sample as for Fig. 3, in the field range from 20 to 28.5 T, at several temperatures. The onset point of the anomaly is indicated by a solid arrow at each temperature, and the higher-field feature of the two-step structure is indicated by a dashed arrow. The anomaly moves towards lower field as the temperature is lowered.

The use of a static field in the present work enabled us to make a detailed study of this anomalous behavior of the magnetoresistance. As we show in the following, the anomaly was observed in many different samples, and although the detailed behavior was somewhat sample dependent, the onset point of the anomaly, indicated by the solid arrows[21] in the figure, was found to be sample independent and reversible upon sweeping the field up and down. This sample independence and

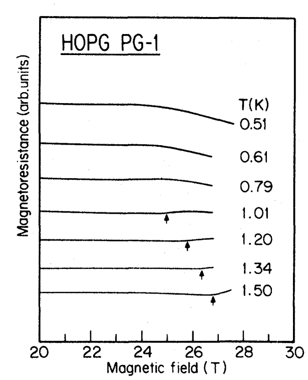

FIG. 5. Magnetoresistance of HOPG (PG1) in the field range $20<H<27.7$ T. At higher temperatures ($T \gtrsim 0.8$ K) an anomaly similar to that observed in Kish graphite (see Fig. 4) is found. At lower temperatures ($T \lesssim 0.8$ K), no anomaly in the magnetoresistance can be identified. The negative magnetoresistance is more pronounced in this temperature range for HOPG than that observed for the Kish samples.

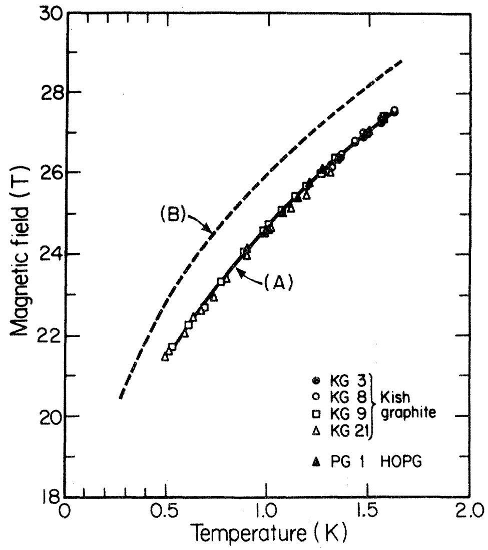

FIG. 6. Summary of the critical points of the magnetoresistance anomaly on a temperature-magnetic field plot. The experimental results for four Kish-graphite samples and one HOPG are shown. Curve (B) shows the "phase boundary" of the CDW state calculated by Yoshioka and Fukuyama (Ref. 22), while curve (A) represents our fit to the data using a phenomenological equation [Eq. (2)] of a form suggested by the Yoshioka-Fukuyama theory.

reversibility indicates that the magnetoresistance anomaly is an intrinsic property of graphite.

The field value at the onset of the anomaly was found to be strongly temperature dependent. The temperature dependence of the critical field is seen in Figs. 4 and 5, which show the recorder traces of the magnetoresistance in the field range from 20 to 28 T for the temperatures indicated on the figures; the traces are for one Kish-graphite sample[21] (Fig. 4) and one HOPG sample (Fig. 5). It should be noted that the traces in Fig. 3 (broad-field sweep) and in Fig. 4 (high-field range) are for the same Kish sample (KG 21). Results obtained on other Kish samples are very similar to those reported in Fig. 4 (see Table I for a comparison of the samples). It is seen that as the temperature is decreased, the magnetoresistance anomaly occurs at a lower field. Figure 6 is a plot of the onset points (indicated by solid arrows[21] in Figs. 3–5) of the magnetoresistance anomaly in the temperature–magnetic-field plane. The results for four Kish-graphite samples and one HOPG sample are shown in Fig. 6, and indicated by different symbols for each sample. The universality of the dependence of the onset point on temperature and magnetic field is clearly seen in Fig. 6.

An interpretation of this anomalous change in the magnetoresistance within the framework of the one-electron (or -band) properties, such as a Shubnikov–de Hass-type oscillation, is precluded because of the strong temperature dependence of the critical field. Furthermore, as the temperature is decreased, the sharpness of the anomaly decreases, in contrast with typical one-electron-type phenomena (e.g., Shubnikov–de Haas effect). Therefore, it is concluded that a many-body effect is responsible for this anomaly. Moreover, the abrupt change in the magnetoresistance strongly suggests that the electron system undergoes some kind of electronic phase transition. It should, however, be pointed out that a calculation of the magnetoresistance above 20 T based on a one-electron model including spin-orbit coupling and trigonal warping has not yet been carried out; such a calculation is necessary for the quantitative understanding of the present experiments.

It is of course difficult to deduce experimentally the dimensionality and order of the phase transition from the magnetoresistance data alone. Yoshioka and Fukuyama (YF) at the University of Tokyo have developed a theory for this phase transition based on a cosine-band Landau-level scheme and on a possible charge-density-wave (CDW) instability of the electronic system of graphite in high magnetic fields (YF theory).[22] At high magnetic fields along the c axis, the electronic motion within the layer plane of graphite becomes localized, so that the electronic band structure becomes one dimensional. Yoshioka and Fukuyama investigated various types of electronic instabilities associated with these one-dimensional bands [e.g., charge-density waves (CDW), spin-density waves (SDW), and an excitonic phase]. They found that in a given magnetic field, the highest transition temperature pertains to the Coulomb-interaction-derived CDW instability corresponding to the wave number spanning the two Fermi wave vectors of the $n=0$, $\sigma=+$ subband (see Fig. 2).

According to YF theory such a CDW instability will open up a gap in the $n=0$, $\sigma=+$ subband at the Fermi level, and thus increase the resistivity. In the YF theory the critical temperature T_c is calculated as a function of magnetic field according to the relation

$$T_c=\frac{8\gamma}{\pi}E_F(H)\frac{\cos^2(\pi\xi)}{\cos(2\pi\xi)} \times\exp\left[\left[\frac{2\pi U(\epsilon)}{c_0(2\pi l)^2}\frac{d\xi}{dE_{0,+}}\right]^{-1}\right], \quad (1)$$

where ξ is the dimensionless Fermi wave vector along k_z corresponding to the $n=0$, $\sigma=+$ magnetic subband. The effective electron-electron interaction $U(\epsilon)$ is a function of the dielectric constant ϵ and the field. The Fermi energy E_F is related to ξ through $E_F=-2\gamma_2\sin^2(\pi\xi)$. The Larmor radius l is given by $l^2=c/eH$, the constant $\gamma=1.78$, and the c axis lattice constant $c_0=6.74$ Å. The units are defined such that $k_B=\hbar=1$, where k_B is the Boltzmann constant. Assuming a value for ϵ of 10, YF evaluated Eq. (1) explicitly. The result is shown by the dashed curve (B) in Fig. 6. The YF critical temperature has a dependence on magnetic field that is qualitatively similar to the experimental data.

The YF theory suggests a functional form of the BCS type for the transition temperature

$$T_c=\alpha(H)\exp[-1/\lambda(H)]\ , \qquad (2)$$

where the prefactor $\alpha(H)$ may, generally speaking, be field dependent, and λ is given by the product of the density of states at the Fermi level and the effective interaction. A fit of the experimental points to Eq. (2) is consistent with taking α as a constant and λ with a linear dependence on magnetic field. That is, we assume $\lambda=\zeta H$. (The data preclude λ independent of magnetic field, but, at the same time, cannot be made to differentiate between a linear or quadratic field dependence.) We find that $\alpha=94.9$ K and $\zeta=8.93\times10^{-3}$ T^{-1} represents the best fit of Eq. (2) to our data in a least-squares sense. This fit is depicted by curve (A) in Fig. 6.

The YF theory is a one-parameter model predicting a CDW instability for a one-dimensional metal. The one free parameter is ϵ, a measure of the strength of the effective interaction. If we assume that the effective interaction in the YF theory scales with the inverse of the dielectric constant and then use ϵ as a parameter to fit the YF function over the field range from 20 to 30 T with $\zeta=8.93\times10^{-3}$ T^{-1}, we find $\epsilon=5.9$. The value of ϵ determined in this way is in satisfactory agreement with energy-loss measurements[23] ($\epsilon=5.4$) and a-face reflectivity measurements by Nemanich *et al.*[24] ($\epsilon=5.8$), but not with the a-face reflectivity measurements by Zanini *et al.*[25] ($\epsilon=3.4$). In view of the critical dependence of the evaluation of ϵ on the functional form assumed for T_c in Eq. (1), we do not consider the present numerical determination of ϵ to be quantitative nor the exact functional dependence on H to be firmly established. (The a face contains both an a axis and a c axis, and the a-face reflectivity refers to the polarization $\vec{E}\,||\,c$ axis. Even for polycrystalline HOPG, the c axis is well defined.)

Now we discuss the detailed behavior of the magnetoresistance at and above the transition. The general features can be summarized as follows: (1) the magnetoresistance shows a sharp increase at the transition point (indicated in Figs. 3–5 by arrows); (2) at fields above the transition point, the magnetoresistance first increases, then goes through a broad maximum and finally decreases, and (3) no hysteresis is observed with respect to the up and down sweep of the magnetic field.

Certain aspects of feature (1) are particularly sample dependent though the transition point (Fig. 6) is not sample dependent. The magnitude of the increase in the magnetoresistance at the transition point differs from sample to sample. It should be mentioned that we could not observe the anomaly in all samples that we investigated, i.e., the Madagascar-graphite sample, one of the HOPG samples and one Kish-graphite sample did not show the anomaly. The same situation was experienced in the previous experiments done with the pulsed magnet,[16,17] in which nine Kish-graphite samples were investigated and the anomaly was observed in six of them. It should also be recalled that Brandt *et al.*[10] studied the magnetoresistance of Kish graphite up to 52 T at 2.1 K, but reported no such anomaly. Therefore, with regard to those samples which do not show the magnetoresistance anomaly, it is not clear at present whether the electronic phase transition does not occur at all or the the phase transition does occur but its effect on the resistivity is so small that we cannot detect the phenomenon.

Feature (2) is a commonly observed behavior of the magnetoresistance above the transition. The detailed behavior was found, however, to be different between the Kish-graphite samples and the HOPG sample. In the case of the Kish-graphite sample (Fig. 4), the magnetoresistance was found to increase above the transition and then to decrease at higher fields. The magnetoresistance of the HOPG sample (Fig. 5) showed a similar behavior at higher temperatures ($T>0.8$ K), but at lower temperatures the increase of the magnetoresistance was no longer observed, though the decrease of the magnetoresistance was found to be more pronounced than for the Kish-graphite samples.

The absence of hysteresis on going up and down in field [feature (3)] suggests that the electronic phase transition is of second or higher order.

In two of the Kish graphite samples an additional feature is observed near the transition; namely, the magnetoresistance anomaly has a two-step structure (see Fig. 4). This structure is not observed in the other samples probably as a result of the poor signal-to-noise ratio peculiar to that set of measurements, the dc amplifier being the origin of the problem. The temperature dependence of the magnetic field separation between the first and the second step is found to be sample dependent. The origin of the higher-field component of the two-step structure is not understood, nor is the sample-dependent temperature dependence.

Besides the sharp anomaly discussed above, we observed a few novel features of the magnetoresistance below the transition. One is the negative slope of the magnetoresistance in the field region between about 17 T and the transition (e.g., see the trace at $T=0.50$ K in Fig. 3 just below 20 T). Previous studies[10,11,15] reported only the saturation of the magnetoresistance and not any decrease in resistance at yet higher fields. Previous workers explained the saturation in terms of the magnetic freeze-out effect of the ionized impurity scattering centers.[10,15] However, it seems that this mechanism alone cannot explain the presently observed negative field dependence of the resistivity just below and above the transition. On the other hand, a recent magnetostriction experiment[26] showed that in the quantum limit, the carrier concentration of graphite increases linearly with increasing field. Such an increase in the carrier concentration could explain the observed negative magnetoresistance at high fields. This effect of increased carrier concentration should also be important in the interpretation of the linear field dependence and the saturation of the magnetoresistance in the field range above the passage of the $n=1$, $\sigma=+$ level through the Fermi level (7.3 T) but below the onset of the high field transition.

V. CONCLUSIONS

We have investigated the magnetoresistance of pure graphite at high fields up to 28.5 T and have found an anomaly above 22 T at low temperatures. The critical-field value at the onset of the anomaly is the same for several different samples and has a strong temperature dependence. The strong temperature dependence of the critical field precludes an interpretation of this phenomenon in terms of known one-electron properties of graphite, and suggests the occurrence of some kind of electronic phase transition. One plausible interpretation is the CDW instability of one-dimensional Landau subbands, proposed by Yoshioka and Fukuyama (Ref. 22). The "phase boundary" curve given by the YF theory explains qualitatively the temperature and magnetic field dependence of the experimental transition points, as shown in Fig. 6. The detailed behavior of the magnetoresistance, however, remains to be discussed theoretically. For instance, such features as the decrease in the resistance at fields much higher than the transition, and the differences in behavior between the Kish-graphite and HOPG samples do not seem to be derived in such a straightforward fashion from the CDW gap model alone. A detailed transport calculation is necessary for a direct comparison with the experimental results.

On the experimental side, it is expected that the electronic phase transition can be observed by a variety of experimental techniques, e.g., measurements of magnetic susceptibility, magnetostriction, and Alfven wave propagation. Such experiments will give complementary information necessary to elucidate the nature of the electronic phase transition. In carrying out these experiments relevant to the phase transition, the static high field generated by the hybrid magnet should be very convenient.

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