Effects of Hole Doping by Neutron Irradiation on Magnetic-Field Induced Electronic Phase Transitions in Graphite

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Abstract.
We have investigated effects of hole doping by fast-neutron irradiation on the magnetic-field induced phase transitions in graphite using specimens irradiated with fast neutrons. Resistance measurements have been done in magnetic fields of up to above 50 T and at temperatures down to about 1.5 K. The neutron irradiation creates lattice defects acting as acceptors, affecting the imbalance of the electron and hole densities and the Fermi level. We have found that the reentrant transition field shifts towards lower fields with hole doping, suggestive of the participation of the electron subband in the magnetic-field induced state.

1. Introduction
Whilst the realisation of graphene, perhaps the best two-dimensional carrier system to study, has attracted great research interest recently [1], graphite, which may be regarded as multi-layered graphene, also has been known to exhibit very interesting phenomena at high magnetic fields and low temperatures. The electron-hole system in the compensated semimetal graphite undergoes a magnetic-field induced electronic phase transition and a reentrant transition back to the normal state at a higher magnetic field. These transitions manifest themselves as abrupt changes in magnetoresistance [2, 3].

Whereas the real nature of this phenomenon still remains unknown, the magnetic-field induced state has been often discussed in terms of the formation of a density wave involving $2k_F$-type nesting. As graphite is a semimetal, its carrier density is much smaller than those of usual metals. Consequently, the carrier system reaches its quantum limit in moderately strong magnetic fields parallel to the c-axis, such as $B > 20$ T. In the quantum limit, only the lowest electron ($n = 0$) and hole ($n = -1$) Landau subbands (each of them spin-split) are occupied and all the other subbands are very far from the Fermi level in energy. Under such circumstances, several nestings across two Fermi points are possible at sufficiently low temperatures (Fig. 1); this theoretical scenario was originally proposed by Yoshioka and Fukuyama[4]. However, which Landau subband is the most favourable remains unclear on theoretical grounds [4, 5, 6].
In order to gain insight into which Landau subband is responsible for the magnetic-field induced density-wave state, we have investigated effects of hole doping by fast-neutron irradiation. The fast-neutron irradiation introduces lattice defects in graphite, some of which act as acceptors. Since graphite is a semimetal, such hole doping greatly affects the Fermi level and the Fermi wave numbers of both the electron and hole Landau subbands.

![Figure 1](image_url) Landau levels of graphite in a magnetic field of 30 T parallel to the c-axis. The nesting vector for the charge density wave in the $n = 0$, spin-up subband is shown as an example.

### 2. Experimental

The samples used in the present study were flakes of single crystal graphite (Kish graphite). One of them was pristine, and the rest were irradiated with fast neutrons of a flux ($E > 1$ MeV) of $5.5 \times 10^{12}/\text{cm}^2$ at $\sim 50$ $\text{C}^\circ$ at JAERI JRR-4 for 1, 2 and 4 hours. (The imbalance of the electron and hole densities $p - n$ are estimated to be $0.7, 1, 2 \times 10^{18}/\text{cm}^3$ from Hall measurements, respectively.) Typical dimensions of the samples were $\sim 3$ mm in length, $\sim 1$ mm in width and a few tens of $\mu\text{m}$ in thickness. The electrical contacts were made with silver paste. The resistance as a function of magnetic field was measured by a conventional four-contact method. A phase sensitive detection technique was employed with an alternating current of typically $200 \mu\text{A}$ at $200 \text{kHz}$. High magnetic fields of up to above $50$ T with a pulse duration of $\sim 7 \text{ ms}$ were generated using a pulsed magnet at the pulsed field facility in the University of Oxford. Low temperatures down to $\sim 1.5$ K were obtained by means of $^4\text{He}$ inserts.

### 3. Results and Discussion

We have measured the transverse magnetoresistance of graphite samples with different neutron dosages in magnetic fields of up to above $50$ T at several temperatures between $1.5$ K and $4.2$ K. Figure 2 compares resistance traces taken at $4.2$ K for samples with different dosages. All of the traces in fig. 2 commonly exhibit the onset transition (labelled $\alpha$) and the reentrant transition (labelled $\alpha'$) back to the normal phase.

Figure 3 displays the phase diagram of graphite in the temperature-magnetic-field plane. With increasing neutron dosage, the onset transition field $\alpha$ shifts towards higher fields whilst
Figure 2. Tranverse magnetoresistance traces of neutron-irradiated specimens and a pristine one at 4.2 K. Both the onset transition (labelled $\alpha$) and the reentrant transition (labelled $\alpha'$) are observed.

The reentrant transition back to the normal state may be understood as a consequence of Fermi-level crossing (and depopulation) of the Landau subband(s) responsible for the field-induced density-wave state [3]. Takada and Goto [8] calculated the renormalised band structure based on the Slonczewski-Weiss-McClure model [9, 10] by taking into account self-energy corrections. They concluded that the $n = 0$, spin-up and $n = -1$, spin-down Landau subbands cross the Fermi level, upwards and downwards respectively, almost simultaneously at $\sim 53$ T. This result suggests that the higher-field phase boundary or the reentrant field should tend to zero temperature at the same magnetic field. (In fig. 3, the reentrant transition field extrapolates to $B \approx 52.5$ T at zero temperature, showing an excellent agreement with Takada and Goto’s calculations.) Therefore, as far as pristine graphite is concerned, the reentrant transition could be associated with the $n = 0$, spin-up and/or $n = -1$, spin-down subband, and the nature of the magnetic-field induced state between the $\alpha$ and $\alpha'$ transitions remains somewhat uncertain.

For neutron-irradiated specimens as well as the pristine one, the extrapolation of the reentrant field to zero temperature probably gives a good approximation to the field at which the relevant Landau sub-band crosses the Fermi level. Such an extrapolated field shifts towards lower fields with neutron irradiation (or hole doping). This provides strong evidence for the involvement of the electron ($n = 0$) subband with the magnetic-field induced state, because each electron (hole) Landau subband’s crossing field should move to a lower (higher) field with hole doping. However, it should be noted that the possibility of the participation of the hole ($n = -1$) subband in the field induced state is not necessarily excluded.
Figure 3. Phase diagram of graphite in the magnetic-field-temperature plane. The crosses represent data points of the onset ($\alpha$) and reentrant ($\alpha'$) fields of a pristine graphite sample (after Ref. [3]). The solid curve through the data points is a guide to the eye and provides an approximate phase boundary for pristine graphite. Open symbols represent data points of graphite doped with holes by neutron irradiation (for 1, 2 and 4 hours).

4. Summary
We have investigated effects of hole doping by fast-neutron irradiation on the magnetic-field induced transitions in graphite. We have found that the reentrant transition field shifts towards lower fields with neutron irradiation, suggestive of the involvements of the electron subband with the magnetic-field induced state.

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