THE EFFECTS OF FAST NEUTRON IRRADIATION ON THE GALVANOMAGNETIC PROPERTIES OF GRAPHITE

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1. Introduction

Highly oriented pyrolytic graphite was irradiated with fast neutrons at ~90°C in the NBS reactor (Gaithersburg, Md) to doses 0.18-2.9x10^{17}nvt(E>1MeV) or 0.29-4.6x10^{17}nvt(E>.1MeV). Galvanomagnetic effects (Hall resistivity, ρyx, and magnetoresistance, ρxx) were measured between 1.2 and 4.2K in the water-cooled solenoids at NRL (Washington, D.C.) in fields up to 9Tesla (90kg). Description of starting specimens used in the present work and detailed measurements of galvanomagnetic measurements are given elsewhere(1). From the resistivity ratio (ρ298/ρ4.2K = 14.2) the present specimens are of high quality.

2. Results for σxx

From these data the conductivity components σxx and σxy were obtained. Classical Theory (2) predicts that $\sigma xx\ B^2$ should saturate in high fields, $B \gg B_{1/2}$, where $B_{1/2}$ is the field at which the resistivity is doubled. As seen in figure 1 this condition is satisfied for all specimens at 9Tesla but it is $\sigma xx B$ that saturates for all specimens. This behavior has been observed in pristine graphite by a number of workers in this temperature and field range (See ref 1 for discussion).

3. Results for σxy

Classical theory also predicts that σxyB should saturate and this behavior is observed (fig. 2). The limiting value of σxyB allows the number of acceptors NA to be calculated from the formulae:

$$(\sigma xyB)_{B\gg B_{1/2}} = (P-N)|e| \qquad [1]$$

$$NA = (P-N) \qquad [2]$$

where P,N are the densities of holes and electrons respectively.

It is noted that values of (P-N) should also be given from the limiting value of the Hall Parameter (RH→$(\sigma xyB)^{-1}$ for $B\gg B_{1/2}$, according to theory). However, RH does not saturate at high fields because the limiting value of RH is predicted on the basis that $\rho xx \ll \rho xy$ or $\sigma xx \ll \sigma xy$ in high fields. Neither of these conditions is satisfied at high fields, and RH did not saturate. Earlier workers (3) had estimated (P-N) from Hall data at ambient temperature at relatively low fields (eg B~0.5T). Their estimates for the number of acceptors per unit dose level would be several times higher than that given here for the same data. For instance, using the value of RH at 9T, and comparing with data for σxyB, we obtain:

No. of acceptors = per dose	(14±2)ppm/10^{17}nvt(E>1MeV)-σxy data
	(30±10)ppm/10^{17}nvt(E>1MeV)-RH data.

If Hall data were used at lower field the calculated value of NA would be even higher.

4. Shift of Shubnikov de Haas Oscillations in the Quantum Limit

In figs 1 and 2 an oscillatory component of the conductivity components can be seen-the Shubnikov de Haas oscillations. As the number of acceptors increases the Fermi level should fall so that the extremal positions of these oscillations should shift. Cooper et al(4) measured these shifts in the field region up to ~25T for specimens irradiated to 1.2x10^{17}nvt(no criterion given for neutron energy). Since (i) the Shubnikov de Haas frequencies for hole and electrons in graphite differ only by a factor of ~1.5 (ii) irradiation increases the scattering of carriers and (iii) shifts of frequencies are small in this dose region, the present work concentrated on the shift of the quantum limit oscillations which can be observed more readily without recourse to complicated curve-fitting proceedures. (An analysis showing the difficulties inherent in Cooper et al's work is given in ref 5).

Data showing the quantum-limit oscillations for specimens used in the present work are given in fig. 3. At each of the extrema, the Fermi level is in coincidence with a magnetic energy level (Landau level). Since the c-axis lattice spacing is changed by less than 0.02% for doses less than 10^{18}nvt it is reasonable to assume initially that band overlap parameters do not change with irradiation. A calculation was made of the shift of the n=1,2 (electron) and n=1 (hole) extrema with acceptor level and comparison is made with the experimental data in fig. 4 using values of NA calculated from [1] and [2].

5. Conclusions

The following conclusions follow from the present work.

(1) The shift of the coincidence fields for electron levels generally follows the predictions of the rigid band model, but the shift of the hole level is much smaller than predicted. Agreement cannot be obtained by employing a scaling factor for the acceptor levels calculated using [1] and [2]. Thus a rigid band model does not appear to be appropriate.

(2) A calculation was made to assess the change of coincicence fields with band overlap parameters. The data could not be fitted with a reasonable set of parameter changes.

(3) The shifts of the coincidence levels are generally in agreement with a band model in which the parameter $\gamma 2$ is negative.(6) This is in agreement with Cooper et al's results(4) but in disagreement with Soule's results on boron-doped graphite(7).

(4) In the low dose region the abrupt shift of the n=1 electron level with dose is difficult to explain unless changes occur in phase-shift parameters.

(5) The absolute increase in the amplitude of the n=1,2 (electron) oscillation for sample #2 is difficult to explain. Since the introduction of acceptors should increase the scattering probabilities absolutely, as shown by changes in conductivity, an absolute decrease in the amplitude of the Shubnikov de Haas oscillations should be observed.

Acknowledgements

This work was supported by a grant from the Atomic Energy Commission to the University of Maryland. Thanks are due to the staff of the Magnet Facility, Naval Research Laboratory and to the staff of the Reactor Facility, National Bureau of Standards. Dr. A.W. Moore kindly supplied the graphite samples.

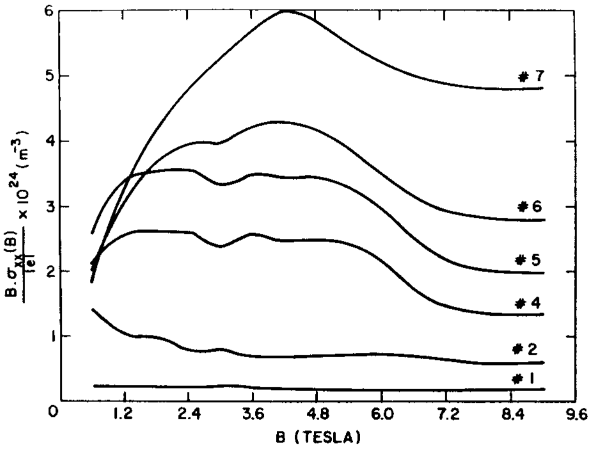

Fig. 1. σxx.B versus field plot. Hours in the reactor for these specimens are given in figure 3. Doses can be interpolated directly from exposure time and dose of specimen irradiated to 32 hours (#7) (2.9×10^{17}nvt(E>1MeV); 4.6×10^{17}nvt(E>.1MeV)).

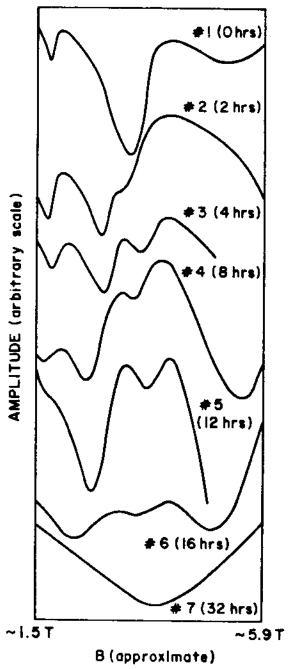

Fig. 3. Oscillatory components of ρxx after midline drift subtracted with a bucking signal. In these data, small magnet calibration errors, which differ from sample to sample, are not corrected for. Values in brackets refer to hours in the reactor.

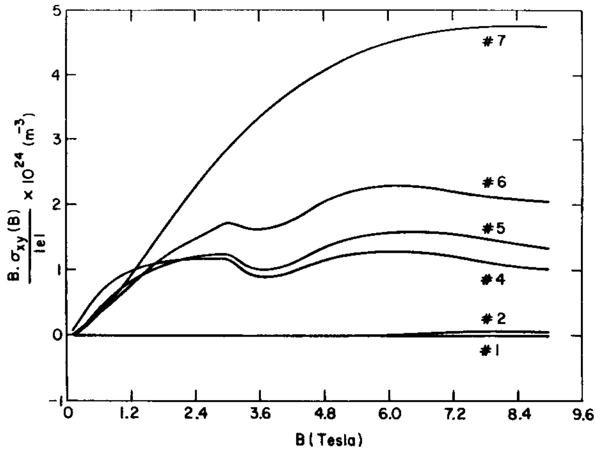

Fig. 2. σxy.B versus field plot.

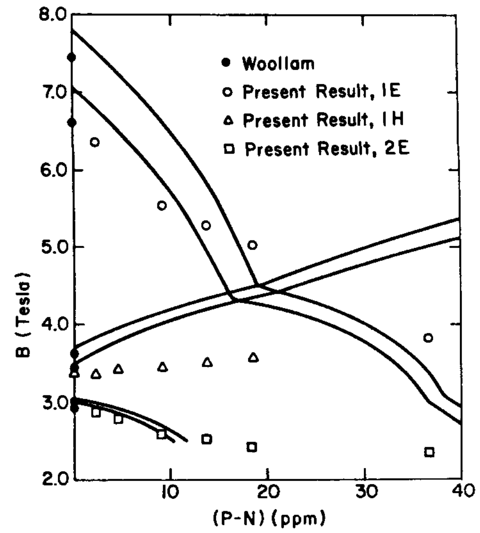

Fig. 4. Summary plot of the shift in the coincidence fields 1E, 2E, 2H (E and H refer to electron and hole) with (P-N) obtained from σxyB data. Theoretical curves based on the rigid band model are shown. Double curves refer to spin-splitting which can only be resolved experimentally for the pristine sample.

References

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