# **VIII-2**.

# Effect of Carrier-Carrier Scattering on the Hall Effect in Graphite

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The importance of the carrier-carrier scattering on the Hall effect in graphite is pointed out. Possibility of removal of the disagreement between the experiment and the theory is shown in the calculation which takes account of the carrier-carrier scattering. In the theory disregarding this effect the contribution of the minority electrons whose effective masses are very small is overestimated. However, their contribution is considerably reduced due to the effect of the minority-majority carrier scattering.

#### §1. Introduction

Zero-field Hall coefficients of graphite calculated by the use of the electron-phonon coupling constants which fit the observed conductivity have opposite signs to the experimental results at 77°K and 300°K.<sup>1,2)</sup> At 77°K the calculation produces just the opposite dependence on field strength and its slope is steeper than the observed one.

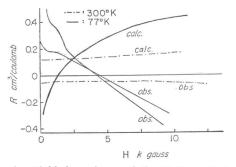


Fig. 1. Field dependence of the Hall coefficients at  $77^{\circ}$ K and  $300^{\circ}$ K.

In the room temperature complicated situations arise because of the low Fermi temperature ( $T_f = 260^{\circ}$ K), thus we shall confine our discussions to the degenerated region only.

Ratio of the contributions of the holes and electrons to the Hall effect at 77°K is about 1: 2. The major part of the electron contribution comes from the minority electrons which are located in the vicinity of the edge of the Fermi surface  $(k_z \cong (k_z)_{max})$ . These carriers are few in number but their effective masses being very small play an important role in making the Hall coefficient negative and its field dependence strong. If the mobilities of these carriers are reduced somehow, we can expect to remove the disagreement between the theory and the observed values.

In the following it will be shown that considerable reduction in the minority carrier mobility can be expected by the introduction of the minority-majority scattering effect. Importance of the carrier-carrier scattering on the mobilities of the carriers in semiconductor has been pointed out already by several investigators<sup>3,4)</sup>. Since there are no appreciable differences between the effective masses and the relaxation times of the majority holes and electrons, the drag effect is not important on the reduction of the minority carrier mobility and the majority carriers are affected only slightly due to the collisions between them.

### § 2. Effect of Carrier-Carrier Scattering on the Minority Carrier Mobility

Band structure of graphite is too complicated to perform calculation<sup>5)</sup> and we will propose a simplified three ellipsoid model as is shown in Fig. 3.

Since effective mass changes continuously over the Fermi surface in actual graphite, we cannot define the minority carriers correctly and

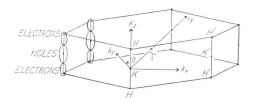


Fig. 2. Brillouin zone and the Fermi surface for graphite.

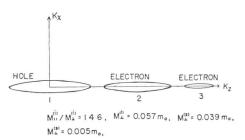


Fig. 3. Simplified ellipsoidal band model of graphite.

then in the simplified model we have assumed the minority carrier concentration  $N_3 = 0.15 \times 10^{18} \,\mathrm{cm}^{-3}$  which corresponds to the carrier concentration lying the region  $|(k_z)_{\max} - k_z| < 1/10(k_z)_{\max}$ . Anisotropy ratios of the effective masses of the majority carriers were determined due to the angular dependence of the Shubnikovde Haas effect<sup>6)</sup>.

Assumptions made in our calculation are as follows: i) As the shielding of the Coulomb interaction is inefficient (screening length  $1/q_D \cong 10^{-6}$  cm) interband transition may be negligible, ii) Born approximation is used in estimating the scattering cross-sections.

In the following we shall calculate the zerofield Hall coefficient by the use of the variation principle.

The Boltzmann equation describing the motion of the *i*-th carrier is written in the form

$$-eFv_{ix}\frac{\partial f_i^0}{\partial E_i} + \tau_i^{-1}\Phi_i\frac{\partial f_i^0}{\partial E_i} + \sum_j \Delta_j f_i = 0 \quad (e>0) ,$$

$$(2.1)$$

where F is an electric field applied in the xdirection and  $\tau_i$  denotes the relaxation time of the *i*-th carrier controlled by phonon scattering. The term of the form  $\Delta_j f_i$  represents the effect on carriers of type *i* of scattering by carriers of type *j*. The distribution function  $f_i$  is related to  $\Phi_i$ 

$$f_i = f_i^{\ 0} - \Phi_i \frac{\partial f_i^{\ 0}}{\partial E_i} . \tag{2.2}$$

Here  $\Phi_i$  is taken to be expressed by the following power series:

$$\Phi_{i} = eFv_{ix} \sum_{n=0}^{\infty} \chi_{i}^{(n)} [(E_{i} - \zeta)/k_{0}T]^{n}, \qquad (2.3)$$

where  $\zeta$  is the Fermi energy. Keeping only terms of first order in  $\Phi$ , we obtain

where

$$P_{k_{i}',k_{j}'}^{k_{i}',k_{j}'} = \frac{2\pi}{\hbar} \left(\frac{4\pi e^{2}}{\kappa}\right)^{2} [|k_{i}'-k_{i}|^{2}+q_{D}]^{-2} \delta(E_{i}'+E_{j}' - E_{i}-E_{j})f_{i}^{0}(E_{i})f_{j}^{0}(E_{j})\{1-f_{i}^{0}(E_{i}')\} \times \{1-f_{j}^{0}(E_{j}')\} .$$
(2.5)

Using the variation principle, the simultaneous equations which determine the first approximation terms are derived with a little algebra,

$$-\frac{\chi_{j}^{(0)}}{2k_{0}T}\left(\frac{\hbar^{2}}{M_{\perp}^{(1)}M_{\perp}^{(2)}}\right)_{\substack{k_{i}, k_{j} \\ k_{i}', k_{j}'}} P_{k_{i}', k_{j}'}^{k_{i}', k_{j}'}(k_{jx}'-k_{jx})^{2} +\chi_{i}^{(0)}\left\{\sum_{k_{i}} v_{ix}^{2}\tau_{i}^{-1}\frac{\partial f_{i}^{0}}{\partial E_{i}} -\frac{1}{2k_{0}T}\left(\frac{\hbar}{M_{\perp}^{(i)}}\right)^{2} \\\times\sum_{\substack{k_{i}, k_{j} \\ k_{i}', k_{j}'}} P_{k_{i}', k_{j}'}^{k_{i}', k_{j}'}(k_{ix}'-k_{ix})^{2}\right\} = \sum_{k_{i}} v_{ix}^{2}\frac{\partial f_{i}^{0}}{\partial E_{i}} (i, j = 1, 2) \quad (2.6)$$

$$-\sum_{i=1,2} \left(\frac{e_i}{e}\right) \frac{\chi_i^{(0)}}{2k_0 T} \left(\frac{\hbar^2}{M_{\perp}^{(i)} M_{\perp}^{(3)}}\right) \sum_{\substack{k_i, k_3 \\ k_i', k_{3'}}} P_{k_i', k_{3'}}^{k_i', k_{3'}} \\ \times (k_{3x'} - k_{3x})^2 + \chi_3^{(0)} \left\{ \sum_{k_3} v_{3x}^2 \tau_3^{-1} \frac{\partial f_3^0}{\partial E_3} - \frac{1}{2k_0 T} \right. \\ \left. \times \left(\frac{\hbar}{M_{\perp}^{(3)}}\right)^2 \sum_{i=1,2} \sum_{\substack{k_i, k_3 \\ k_i', k_{3'}}} P_{k_3', k_i'}^{k_3', k_i'} (k_{3x'} - k_{3x})^2 \right\} \\ = \sum_{k_3} v_{3x}^2 \frac{\partial f_3^0}{\partial E_3} . \quad (2.7)$$

where  $e_i$  denotes the charge of the *i*-th carrier. The first terms in (2.6) and (2.7) represent the drag effect<sup>4)</sup> which depends on the relative drift motion of the two types of carriers. In our case the drag effect does not play an important role in the reduction of the mobilities of the majority and the minority carriers.

Solving eqs. (2.6) and (2.7), we obtain the following expressions for the effective relaxation times  $\tau_i^* (= \chi_i^{(0)})$  including the carrier-carrier scattering effect:

$$\tau_{i}/\tau_{i}^{*} \cong 1 + 8A(k_{0}T)^{2}(M_{\perp}^{(1)}M_{\perp}^{(2)})^{2} \\ \times \left(\frac{\tau_{1}}{N_{1}M_{\perp}^{(1)}} + \frac{\tau_{2}}{N_{2}M_{\perp}^{(2)}}\right), \quad i = 1, 2 \quad (2.8)$$
  
$$\tau_{3}/\tau_{3}^{*} \cong 1 + 8A(k_{0}T)^{2}\frac{\tau_{3}M_{\perp}^{(3)}}{N_{3}} \\ \times \{(M_{\perp}^{(1)})^{2} + 2(M_{\perp}^{(2)})^{2}\}, \quad (2.9)$$

where  $A = \frac{\sqrt{3}}{24\pi^3 \hbar^7 q_D} \left(\frac{e^2}{\kappa}\right)^2$ .

Factor 2 appearing in the square bracket of (2.9) comes from the difference of the number of the hole- and the electron-ellipsoids.

Though 77°K is not a sufficiently low temperature as compared with the Fermi temperature  $T_f$ , it may be permissible to use the formulae (2.8) and (2.9) to get an approximate result. In our previous calculation<sup>2</sup> we have arrived at the result that the contribution of the electrons to the Hall effect is two times larger than that of the holes at 77°K. Using the value of  $\tau_i$  chosen so as to give the above mentioned ratio of the contributions to the Hall effect. This is the same order of magnitude of our previous calculation  $-0.42 \text{ cm}^3/\text{coul}$ .

Substituting the  $\tau_i^*$  obtained in (2.8) and (2.9) into the expression of the Hall coefficient, we have  $R=0.1 \text{ cm}^3/\text{coul}$  which agrees qualitatively with the observed value  $0.2 \text{ cm}^3/\text{coul}$  in spite of our crude model. Such a remarkable modification of the Hall effect results from the considerable reduction of the minority carrier mobility. Namely one obtains  $\tau_3/\tau_3^* \cong 3$ , while  $\tau_i/\tau_i^* \cong 1.3$  for i = 1, 2 at 77°K.

In the above discussion we have invoked the Born approximation to estimate the scattering cross-sections. This procedure might overestimate the cross-sections of the carrier-carrier scattering several times. However,  $3\rightarrow 2$  transition forbidden in our model will contribute to further reduction of the minority carrier mobility because of the continuous distribution of the effective mass over the Fermi surface in actual graphite and so this effect will compensate the over-estimation of the scattering cross-sections due to the Born approximation.

Present treatment of the majority carriers is oversimplified and the intraband transitions which are ineffective in our model will be important for the majority carriers in real graphite crystal.

#### References

- 1) D. E. Soule: Phys. Rev. 122 (1958) 698.
- S. Ono and K. Sugihara: J. Phys. Soc. Japan: 21 (1966) 861; J. W. McClure and L. B. Smith: *Proc. Fifth Carbon Conf.* (Pergamon Press, 1963) Vol. 11, p. 3.
- J. Appel: Phys. Rev. 125 (1962) 1815; J. Appel and R. Bray: Phys Rev. 127 (1962) 1603.
- T. P. McLean and E. G. S. Paige: J. Phys. Chem. Solids 16 (1960) 220.
- J. C. Slonczewski and P. R. Weiss: Phys. Rev. 109 (1958) 272.
- D. E. Soule, J. W. McClure and L. B. Smith: Phys. Rev. 98 (1964) 254.

#### DISCUSSION

**Glicksman, M.:** Is the reduction in minority carrier mobility you obtain sufficient to make the calculated Hall coefficient positive at low magnetic fields?

Sugihara, K.: Yes, it is possible. Inserting the approximate value of  $\tau_i$  into the expressions of  $\tau_i/\tau_i^*$ , we can get positive Hall effect at 77°K. In this case values of  $\tau_i$  (*i*=1, 2, 3) are chosen so as to fit the calculated values at 77°K (Fig. 1) based on the real band structure (S. Ono and K. Suginara: J. Phys. Soc. Japan 21 (1966) 861).

Herring, C.: Is it possible to sort out the effect of carrier-carrier scattering from those of other types of scattering by means of its temperature dependence, which I believe should be  $T^2$  in the range well below the degeneracy temperature? Also, is it possible to compare theory and experiment on the Hall coefficient in the range where the scattering is dominated by impurities?

Sugihara, K.: Yes, I think it may be possible to ascertain the carrier-carrier scattering effect on the Hall effect by studying the impurity scattering. However, we have no reliable data at these temperatures.