

## Contribution to the Theory of Impurity Band Conduction. II

—Hall Effect—

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(Received June 10, 1968)

In this paper we discuss the Hall coefficient of heavily doped semiconductors at very low temperatures by making use of the general method developed in previous two papers (Matsubara-Toyozawa and the first part of this series). The effect of weak magnetic field on the impurity band conduction is taken into account on the basis of the Kubo formalism and the diagram method. An expression for the Hall coefficient is obtained in terms of the Green's function and it is proved that this expression is reduced in a special case to the usual one derived from the Boltzmann equation method. Some discussion of the general feature of Hall effect in a random system is given.

### § 1. Introduction

The concept of the impurity band conduction was originally introduced to interpret the low temperature anomaly of the Hall effect in heavily doped semiconductors:<sup>1)</sup> the Hall coefficient  $R$  has a maximum at a certain temperature and tends to a constant value

$$R_0 = \frac{1}{nec} \quad (1.1)$$

at very low temperatures. Here  $n$  is regarded as the number density of the carriers in the "impurity band". Although there are few experiments in which the Hall coefficient at very low temperature is measured as a function of donor concentration, such experiments so far carried out seem to show that the relation (1.1) does hold. Figure 1 is one example of the experimental curve<sup>2)</sup> for n-type Ge doped with Sb, In and As, which clearly shows that in the so-called metallic region ( $N_D > 10^{17} \text{ cm}^{-3}$ )  $R_0^{-1}$  is proportional to  $N_D$  as is expected from (1.1).

One might think this is quite a natural result. From a purely theoretical point of view, however, (1.1) is by no means a straightforward conclusion. For an electron moving in random lattice such as the electron in "impurity band", many of the conventional concepts used in the nearly free electron model lose their meaning: thus wave vector, effective mass, the Fermi surface and even the

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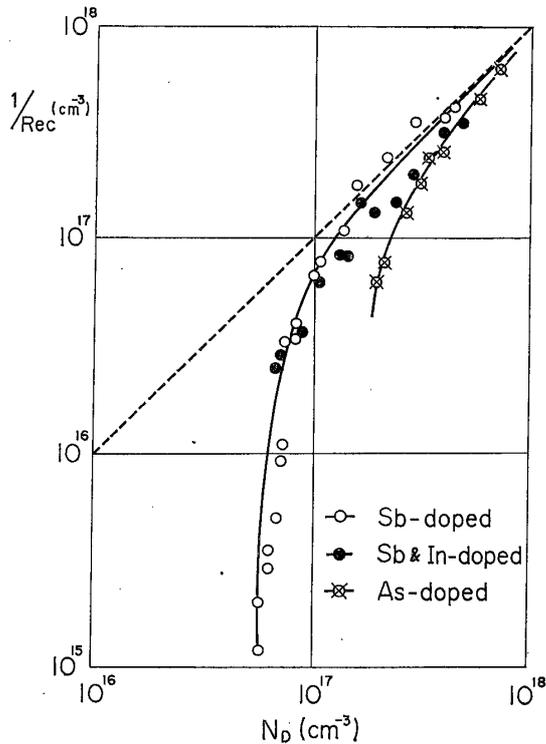


Fig. 1. Relation between the Hall density  $1/|e|R|c$  at  $4.2^\circ\text{K}$  and the donor concentration  $N_D$  for n-type Ge.

concept of positive hole cannot be defined distinctly. Equation (1.1) is derived essentially on the basis of the nearly free electron model and the Boltzmann transport equation. Just as many authors have discussed recently,<sup>8)</sup> when the mean free path of electron becomes comparable with or shorter than lattice spacing, there is no reason to believe that the length of the mean free path or the relaxation time does not appear in the final expression for the Hall coefficient as in (1.1). It is also not clear that what factor determines the sign of the Hall coefficient for the electrons in the impurity band where the concepts of effective mass and positive hole are somewhat obscured.

The main purpose of the present paper is to apply the general method of the impurity band conduction developed in previous two papers (which are referred to as M-T<sup>4)</sup> and M-K<sup>5)</sup>) to the Hall effect in order to clarify some points

of the Hall conduction in random systems which are not yet explored theoretically. In § 2 the formulation given in M-T and M-K is extended to the case where a weak magnetic field is applied. In the scheme of the extended formulation, a diagram expansion method is developed and the effect of the magnetic field on the Green's function is carefully examined in § 3. A closed expression for the Hall conductivity is given in terms of the averaged Green's function. In § 4 the results obtained are checked by considering a limiting case of a regular lattice and it is shown that the new expression for the Hall coefficient is reduced to the conventional one derived on the basis of the Boltzmann equation in a limiting case. In the last two sections some analysis of the Hall coefficient for the impurity band electron is given.

## § 2. Formulation

In dealing with the impurity band conduction, we have adopted in the previous papers (M-T and M-K) a simplified Hamiltonian of the following form:

$$\mathcal{H} = \sum_{\{m\}} \sum_{\{n\}} V_{mn} a_m^* a_n, \quad (2.1)$$

$$V_{mn} = \int \phi_0(\mathbf{r} - \mathbf{R}_m) V(\mathbf{r} - \mathbf{R}_m) \phi_0(\mathbf{r} - \mathbf{R}_n) d\mathbf{r}. \quad (2.2)$$

In these expressions,  $\phi_0(\mathbf{r}-\mathbf{R}_m)$  represents the Hydrogen-like 1s wave function of an electron trapped by impurity potential  $V(\mathbf{r}-\mathbf{R}_m)$  at  $\mathbf{R}_m$ .  $a_m^*$  and  $a_m$  are respectively the creation and destruction operators for the electron localized at  $\mathbf{R}_m$ , and  $\sum_{\{m\}}$  means that we are to take sum over the lattice points occupied by impurity atoms. The Hamiltonian (2.1) tells us that the integrals  $V_{mn}$  cause electron transfer from one impurity site to another, and owing to a random distribution of the impurities the set of random variables  $\{V_{mn}\}$  gives rise to the formation of both broad impurity energy band and decaying random electron wave.

Now we try to extend (2.1) to the case where a weak magnetic field  $H$  is applied to the  $z$  direction. For an electron trapped by the potential  $V(\mathbf{r})$  at the origin, Schrödinger equation, in the presence of the magnetic field, becomes

$$\Delta\phi + \frac{2m}{\hbar^2} \left[ E - V(\mathbf{r}) - \frac{ieH}{2mc} \left( x \frac{\partial}{\partial y} - y \frac{\partial}{\partial x} \right) + \frac{e^2 H^2}{8mc^2} (x^2 + y^2) \right] \phi = 0. \quad (2.3)$$

We assume that we know all the solutions of (2.3) and let  $\phi_0(\mathbf{r})$  be the wave function of the ground state. Then, as Peierls has proved,<sup>6)</sup> the solution for a similar problem in which an impurity atom is located at  $\mathbf{R}$  in place of the origin is given by

$$\phi_{\mathbf{R}}(\mathbf{r}) = \exp \left[ -i \frac{e}{2\hbar c} \mathbf{H} \cdot (\mathbf{R} \times \mathbf{r}) \right] \phi_0(\mathbf{r}-\mathbf{R}). \quad (2.4)$$

Therefore one method to include the effect of a magnetic field into the Hamiltonian (2.1) is to replace  $\phi_0(\mathbf{r}-\mathbf{R})$  in (2.2) by  $\phi_{\mathbf{R}}(\mathbf{r})$  given above, that is, to use instead of  $V_{mn}$  the following integral:

$$\tilde{V}_{mn} = \int \phi_0(\mathbf{r}-\mathbf{R}_m) V(\mathbf{r}-\mathbf{R}_m) \phi_0(\mathbf{r}-\mathbf{R}_n) \exp \left[ -i \frac{e}{2\hbar c} \mathbf{H} \times (\mathbf{R}_n - \mathbf{R}_m) \cdot \mathbf{r} \right] d\mathbf{r}. \quad (2.5)$$

Since hereafter we are mainly interested in the weak-field Hall effect, it is enough to consider the quantities up to the term linear in  $H$ . In this approximation we may ignore the  $H$ -dependence of the function  $\phi_0(\mathbf{r}-\mathbf{R})$  because it depends on  $H$  quadratically for a weak field. Thus  $\tilde{V}_{mn}$  can be simplified as

$$\tilde{V}_{mn} = V_{mn} \exp(-if_{mn}), \quad (2.7)$$

where  $V_{mn}$  is given by (2.2) and  $f_{mn}$  by

$$f_{mn} = \frac{e}{2\hbar c} \mathbf{H} \cdot [\mathbf{R}_n \times \mathbf{R}_m], \quad (2.8)$$

and we have a model Hamiltonian to describe the weak-field Hall effect

$$\tilde{\mathcal{H}} = \sum_{\{m\}} \sum_{\{n\}} \tilde{V}_{mn} a_m^* a_n. \quad (2.9)$$

According to the general theory of the transport phenomena, the Hall coefficient in a weak field is given by

$$R = \frac{\sigma_{xy}}{\sigma_{xx}^2 H}. \quad (2.10)$$

$\sigma_{xx}$  is the longitudinal component of the electrical conductivity in zero magnetic field, which has already been computed in M-T.  $\sigma_{xy}$  designates the transverse component of the electrical conductivity. To be more precise we need the anti-symmetric part of  $\sigma_{xy}$  which is proportional to  $H$ :

$$\sigma_{xy}^a = \frac{1}{2}(\sigma_{xy} - \sigma_{yx}). \quad (2.11)$$

A general expression for  $\sigma_{xy}$  based on the Kubo formalism<sup>7)</sup> is

$$\sigma_{xy} = \lim_{s \rightarrow +0} \int_0^\infty dt \int_0^\beta d\lambda \exp(-st) \langle J_x(-i\hbar\lambda) J_y(t) \rangle. \quad (2.12)$$

$J_\alpha(t)$  is the Heisenberg representation of the current operator

$$J_\alpha = \frac{ie}{\hbar} [\tilde{\mathcal{H}}, \sum_{\{m\}} R_m^\alpha a_m^* a_m] = \frac{ie}{\hbar} \sum_{\{m\} \neq \{n\}} R_{mn}^\alpha \tilde{V}_{mn} a_m^* a_n. \quad (2.13)$$

As in the case of M-T, it is convenient to introduce the bases such as diagonalize (2.9):

$$\tilde{\mathcal{H}} = \sum_\mu E_\mu A_\mu^* A_\mu, \quad a_m^* = \sum_\mu C_{m\mu}^* A_\mu^*, \quad a_n = \sum_\nu C_{n\nu} A_\nu. \quad (2.14)$$

In this representation the current operator is expressed as

$$J_\alpha = \sum_\mu \sum_\nu J_{\mu\nu}^\alpha A_\mu^* A_\nu, \quad (\alpha = x, y, z) \\ J_{\mu\nu}^\alpha = \frac{ie}{\hbar} \sum_{\{m\}} \sum_{\{n\}} R_{mn}^\alpha \tilde{V}_{mn} C_{n\mu}^* C_{n\nu}. \quad (2.15)$$

The procedure of reduction of  $\sigma_{xy}$  on the basis of this representation is quite similar to that in M-T, and we are easily led to the result

$$\sigma_{xy} = \lim_{s \rightarrow +0} \sum_\mu \sum_\nu J_{\mu\nu}^\alpha J_{\nu\mu}^\beta \frac{f(E_\mu) - f(E_\nu)}{E_\mu - E_\nu} \frac{i\hbar}{(E_\nu - E_\mu) + i\hbar s} \quad (2.16)$$

with the Fermi distribution function  $f(E) = [\exp \beta(E - E_F) + 1]^{-1}$ . This can be further transcribed into a more compact form in terms of the Green's function which is defined by

$$\tilde{G}_{mn}(E) = \frac{i}{2\pi} [\tilde{G}_{mn}^{(+)}(E) - \tilde{G}_{mn}^{(-)}(E)], \quad (2.17)$$

$$\tilde{G}_{mn}^{(\pm)}(E) = \sum_\mu C_{m\mu} C_{n\mu}^* \frac{1}{E \pm i s - E_\mu} = \langle 0 | a_m \frac{1}{E \pm i s - \tilde{\mathcal{H}}} a_n^* | 0 \rangle.$$

The final expression is summarized in the following form:

$$\sigma_{xy}^a = \left( \frac{e^2}{\hbar} \right) \lim_{s \rightarrow +0} \iint dE_1 dE_2 \Xi^{xy}(E_1 E_2) \frac{f(E_1) - f(E_2)}{(E_1 - E_2)^2 + (s\hbar)^2}, \quad (2.18)$$

$$\mathcal{E}^{xy}(E_1 E_2) = (-i) \sum_{\{m\}} \sum_{\{n\}} \sum_{\{l\}} \sum_{\{l\}} R_{mn}^x \tilde{V}_{mn} \tilde{G}_{nk}(E_2) R_{kl}^y \tilde{V}_{kl} \tilde{G}_{lm}(E_1). \quad (2.19)$$

Hence our problem is reduced to the following three steps; evaluating (2.19), taking the average over the random distributions of impurity atoms and determining  $\sigma_{xy}^a$  from (2.18). The Hall coefficient  $R$  is obtained from (2.9) by substituting  $\sigma_{xy}^a$  thus calculated together with  $\sigma_{xx}$  which has been calculated in M-T. In the next section we present a method to obtain the average of  $\mathcal{E}^{xy}(E_1 E_2)$ .

§ 3. Effect of magnetic field on the Green's function and  $\mathcal{E}^{xy}(E_1 E_2)$

Within the same approximation as in M-T, we examine the effect of magnetic field on the Green's function and  $\mathcal{E}^{xy}(E_1 E_2)$  up to the term linear in  $H$ . In the first place we expand  $\tilde{G}_{mn}$  by iteration as

$$Z\tilde{G}_{mn} = \delta_{mn} + \frac{1}{Z} \tilde{V}_{mn} + \sum_{\nu=1}^{\infty} \frac{1}{Z^{\nu+1}} \sum_{\{l_1\}} \dots \sum_{\{l_\nu\}} \tilde{V}_{ml_1} \tilde{V}_{l_1 l_2} \dots \tilde{V}_{l_\nu n}, \quad (3.1)$$

where complex variable  $Z$  is used in place of  $E \pm is$ . Each term in the above expansion can be represented by diagrams exactly the same way as in M-T, only difference being the appearance of  $\tilde{V}_{mn}$  instead of  $V_{mn}$ . Thus we have the diagonal and non-diagonal component of the Green's functions

$$\tilde{\zeta}_{mn} = Z\tilde{G}_{mn}, \quad (3.2)$$

$$\tilde{\xi}_m = Z\tilde{G}_{mm} \quad (3.3)$$

as the sums of diagrams as shown in Fig. 2. It is readily proved that we can discard  $H$ -dependence of  $\tilde{\xi}_m$  in the linear approximation. To see this it is enough to write down the first few terms in the expansion for  $\tilde{\xi}_m$ :

$$\tilde{\xi}_m = 1 + \frac{1}{Z^2} \sum_{\{l\}} \tilde{V}_{ml} \tilde{V}_{lm} + \frac{1}{Z^3} \sum_{\{l\}} \sum_{\{n\}} \tilde{V}_{ml} \tilde{V}_{ln} \tilde{V}_{nm} + \dots \quad (3.4)$$

In the second term the phase factor obviously cancels and hence  $H$ -dependence does not appear. The summand of the third term is given by

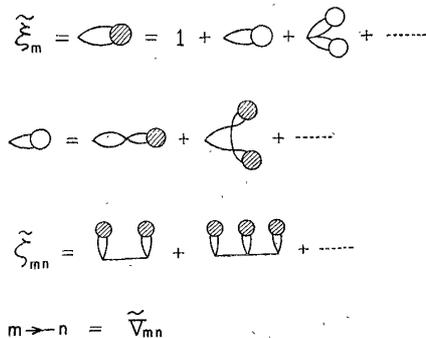


Fig. 2. Diagram representation of  $\tilde{\xi}_m$  and  $\tilde{\zeta}_{mn}$ .

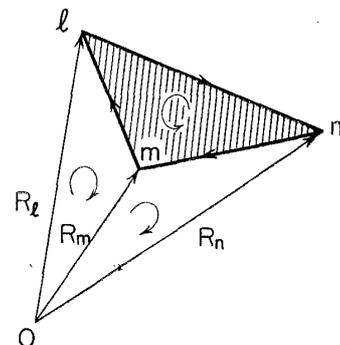


Fig. 3. Geometry of the phase factor  $H \cdot (R_l \times R_m + R_n \times R_l + R_m \times R_n)$ .

$$V_{ml} V_{ln} V_{nm} \exp \left[ -\frac{ie}{2\hbar c} \mathbf{H} \cdot (\mathbf{R}_l \times \mathbf{R}_m + \mathbf{R}_n \times \mathbf{R}_l + \mathbf{R}_m \times \mathbf{R}_n) \right]$$

whose phase factor may be put in a form

$$\exp \left[ -\frac{ie}{2\hbar c} \mathbf{H} \cdot (\mathbf{R}_l \times \mathbf{R}_m + \mathbf{R}_n \times \mathbf{R}_l + \mathbf{R}_m \times \mathbf{R}_n) \right] = \exp \left[ \frac{ie}{\hbar c} \mathbf{H} \cdot \mathbf{S}_{mln} \right], \quad (3.5)$$

where  $\mathbf{S}_{mln}$  is a vector representing an area of the triangle formed by closed three transfers  $m \rightarrow l \rightarrow n \rightarrow m$  (see Fig. 3). Therefore  $\mathbf{H} \cdot \mathbf{S}_{mln}$  takes either sign according to the sense of the transfer  $m \rightarrow l \rightarrow n \rightarrow m$ . This means that the linear term in  $H$  should vanish on average in the third term. The same argument can be applied to terms of any order in (3.4) and thus all the loop diagrams are shown to have no term linear in  $H$ . We can push further this argument to show that  $\tilde{G}_{mn}(Z)$  is approximated as

$$\tilde{G}_{nm}(Z) \simeq \exp(-if_{nm}) G_{nm}(Z). \quad (3.6)$$

On making use of (3.6),  $\mathcal{E}^{xy}(E_1 E_2)$  is expressed as

$$\begin{aligned} \mathcal{E}^{xy}(E_1 E_2) &\simeq (-i) \sum_{\{m\}} \sum_{\{n\}} \sum_{\{k\}} \sum_{\{l\}} R_{mn}^x V_{mn} G_{nk}(E_2) R_{kl}^y V_{kl} G_{lm}(E_1) \\ &\times \exp[-i(f_{mn} + f_{nk} + f_{kl} + f_{lm})]. \end{aligned} \quad (3.7)$$

What we need is the term linear in  $H$  and hence upon expanding the exponent and retaining only the linear term, we finally arrive at the result

$$\begin{aligned} \mathcal{E}^{xy}(E_1 E_2) &= \sum_{\{k\}} \sum_{\{l\}} \sum_{\{m\}} \sum_{\{n\}} \frac{e}{2\hbar c} \mathbf{H} \cdot \{(\mathbf{R}_{kl} \times \mathbf{R}_{lm}) + (\mathbf{R}_{mn} \times \mathbf{R}_{nk})\} \\ &\times \{R_{mn}^x V_{mn} G_{nk}(E_2) R_{kl}^y V_{kl} G_{lm}(E_1)\}. \end{aligned} \quad (3.8)$$

To express more explicitly, we take

$$H = (0, 0, H), \quad R_{mu} = (X_{mu}, Y_{mu}, Z_{mu}).$$

Then (3.8) becomes

$$\begin{aligned} \mathcal{E}^{xy}(E_1 E_2) &= \left( \frac{e}{2\hbar c} \right) H \sum_{\{m\}} \sum_{\{n\}} \sum_{\{k\}} \sum_{\{l\}} [X_{mn}^2 V_{mn} Y_{nk} G_{nk}(E_2) Y_{kl} V_{kl} G_{lm}(E_1) \\ &- X_{mn} Y_{mn} V_{mn} X_{nk} G_{nk}(E_2) Y_{kl} V_{kl} G_{lm}(E_1) \\ &+ X_{mn} V_{mn} G_{nk}(E_2) X_{kl} Y_{kl} V_{kl} Y_{lm} G_{lm}(E_1) \\ &- X_{mn} V_{mn} G_{nk}(E_2) Y_{kl}^2 V_{kl} X_{lm} G_{lm}(E_1)]. \end{aligned} \quad (3.9)$$

$V_{mn} = V(\mathbf{R}_{mn})$  is always expanded in the Fourier series:

$$V(\mathbf{R}_{mn}) = \frac{1}{N} \sum_{\mathbf{k}} \exp(i\mathbf{k} \cdot \mathbf{R}_{mn}) V_{\mathbf{k}}. \quad (3.10)$$

Similarly we define the "Fourier component" of the Green's function

$$G_{\mathbf{k}\mathbf{k}'}(Z) = \frac{1}{N} \sum_{\{m\}} \sum_{\{n\}} \exp(-i\mathbf{k} \cdot \mathbf{R}_m) \exp(i\mathbf{k}' \cdot \mathbf{R}_n) G_{mn}(Z). \quad (3.11)$$

In terms of these Fourier components, (3.9) is written as

$$\begin{aligned} \Xi^{xy}(E_1 E_2) = & \frac{eH}{2\hbar c} \sum_{\mathbf{k}} \sum_{\mathbf{k}'} \left[ \frac{\partial^2 V_{\mathbf{k}}}{\partial k_x^2} \frac{\partial V_{\mathbf{k}'}}{\partial k_y'} G_{\mathbf{k}'\mathbf{k}}(E_1) \left( \frac{\partial}{\partial k_y} + \frac{\partial}{\partial k_y'} \right) G_{\mathbf{k}\mathbf{k}'}(E_2) \right. \\ & - \frac{\partial^2 V_{\mathbf{k}}}{\partial k_x \partial k_y} \frac{\partial V_{\mathbf{k}'}}{\partial k_y'} G_{\mathbf{k}'\mathbf{k}}(E_1) \left( \frac{\partial}{\partial k_x} + \frac{\partial}{\partial k_x'} \right) G_{\mathbf{k}\mathbf{k}'}(E_2) \\ & + \frac{\partial V_{\mathbf{k}}}{\partial k_x} \frac{\partial^2 V_{\mathbf{k}'}}{\partial k_x' \partial k_y'} G_{\mathbf{k}'\mathbf{k}}(E_2) \left( \frac{\partial}{\partial k_y'} + \frac{\partial}{\partial k_y} \right) G_{\mathbf{k}'\mathbf{k}}(E_1) \\ & \left. - \frac{\partial V_{\mathbf{k}}}{\partial k_x} \frac{\partial^2 V_{\mathbf{k}'}}{\partial k_y'^2} G_{\mathbf{k}\mathbf{k}'}(E_2) \left( \frac{\partial}{\partial k_x'} + \frac{\partial}{\partial k_x} \right) G_{\mathbf{k}'\mathbf{k}}(E_1) \right]. \quad (3.12) \end{aligned}$$

At this stage it is necessary to take average of  $G_{\mathbf{k}\mathbf{k}'}$   $G_{\mathbf{k}'\mathbf{k}}$  over the random distributions of impurity sites. In the first paper referred to as M-T this average was simplified by making use of the approximation

$$\langle G_{\mathbf{k}\mathbf{k}'} G_{\mathbf{k}'\mathbf{k}} \rangle \simeq \langle G_{\mathbf{k}\mathbf{k}'} \rangle \langle G_{\mathbf{k}'\mathbf{k}} \rangle \quad (3.13)$$

which is not always justified. A more accurate method of averaging was presented in M-K. In this paper, however, we content ourselves with the approximation (3.13) as was used in M-T, because we are going to utilize the numerical result for  $\sigma_{xx}$  given in M-T in the course of computing the Hall coefficient  $R$ . It is hoped that this simplification is enough to keep the essential feature of Hall effect in random system. Within the approximation (3.13)  $\langle G_{\mathbf{k}\mathbf{k}'}(E) \rangle$  becomes diagonal with respect to  $\mathbf{k}$  and  $\mathbf{k}'$ , and hence (3.12) is further simplified to

$$\begin{aligned} \Xi^{xy}(E_1 E_2) = & \frac{eH}{2\hbar c} \sum_{\mathbf{k}} \left[ \frac{\partial^2 V_{\mathbf{k}}}{\partial k_x^2} \frac{\partial V_{\mathbf{k}}}{\partial k_y} G_{\mathbf{k}}(E_1) \frac{\partial}{\partial k_y} G_{\mathbf{k}}(E_2) - \frac{\partial^2 V_{\mathbf{k}}}{\partial k_x \partial k_y} \frac{\partial V_{\mathbf{k}}}{\partial k_y} \right. \\ & \left. \times G_{\mathbf{k}}(E_1) \frac{\partial}{\partial k_x} G_{\mathbf{k}}(E_2) \right. \\ & \left. + \frac{\partial V_{\mathbf{k}}}{\partial k_x} \frac{\partial^2 V_{\mathbf{k}}}{\partial k_x \partial k_y} G_{\mathbf{k}}(E_2) \frac{\partial}{\partial k_y} G_{\mathbf{k}}(E_1) - \frac{\partial V_{\mathbf{k}}}{\partial k_x} \frac{\partial^2 V_{\mathbf{k}}}{\partial k_y^2} G_{\mathbf{k}}(E_2) \frac{\partial}{\partial k_x} G_{\mathbf{k}}(E_1) \right]. \quad (3.14) \end{aligned}$$

So far explicit forms for  $V_{\mathbf{k}}$  or  $G_{\mathbf{k}}(E)$  are not necessary. For further reduction, however, we need to take a definite model and to give explicit forms for  $V_{\mathbf{k}}$  and  $G_{\mathbf{k}}(E)$ . As in M-T we shall adopt the hydrogenic model of impurity levels under the effective mass and continuum approximation. Then  $V_{\mathbf{k}}$  is a function of only  $|\mathbf{k}|$  and  $G_{\mathbf{k}}(E)$  has a form

$$G_{\mathbf{k}}(E) = \frac{i}{2\pi} \left[ \frac{\alpha^{(+)}}{1 - \alpha^{(+)} V_{\mathbf{k}}} - \frac{\alpha^{(-)}}{1 - \alpha^{(-)} V_{\mathbf{k}}} \right], \quad (3.15)$$

where  $\alpha^{(\pm)}$  depends only on  $E \pm is$  and the concentration of impurities. A detailed discussion of the method of determining  $\alpha$  has been given in M-K (see Eqs. (3.2) and (3.12) in M-K). By making use of  $V_k = V(|\mathbf{k}|)$  and (3.15), the differentiation of  $V_k$  and  $G_k$  with respect to  $k_\alpha$  can readily be performed as

$$\frac{\partial V_k}{\partial k_x} = \frac{dV_k}{dk} \frac{k_x}{k},$$

$$\frac{\partial^2 V_k}{\partial k_x^2} = \left\{ \frac{d^2 V_k}{dk^2} \right\} \frac{k_x^2}{k^2} + \left\{ \frac{dV_k}{dk} \right\} \left( \frac{1}{k} - \frac{k_x^2}{k^3} \right), \quad (3.16)$$

$$\frac{\partial^2 V_k}{\partial k_x \partial k_y} = \left\{ \frac{d^2 V_k}{dk^2} \right\} \frac{k_x k_y}{k^2} - \left\{ \frac{dV_k}{dk} \right\} \frac{k_x k_y}{k^3},$$

$$\frac{\partial}{\partial k_x} G_k(E) = \left\{ \frac{dV_k}{dk} \right\} \frac{k_x}{k} G_k(E) G_k^0(E), \quad (3.17)$$

where

$$G_k^0(E) = \frac{1}{2} [G_k^{(+)}(E) + G_k^{(-)}(E)]. \quad (3.18)$$

Inserting all these results into (3.14), and noting that  $x$ - and  $y$ -directions are equivalent on average, we have

$$\Xi^{xy}(E_1 E_2) = \frac{eH}{\hbar c} I^{xy}(E_1 E_2) \quad (3.19)$$

with

$$I^{xy}(E_1 E_2) = \frac{1}{3} \sum_{\mathbf{k}} \left\{ \frac{dV_k}{dk} \right\}^3 \frac{1}{k} G_k(E_1) G_k(E_2) \{G_k^0(E_2) - G_k^0(E_1)\} \quad (3.20)$$

and from (2.17), (3.19) the expression for the Hall conductivity  $\sigma_{xy}^a$  is written as

$$\sigma_{xy}^a = \frac{e^3 H}{\hbar^2 c} \iint dE_1 dE_2 I^{xy}(E_1 E_2) \frac{f(E_1) - f(E_2)}{(E_1 - E_2)^2}. \quad (3.21)$$

This is the basic formula from which we shall start for the discussion in the following sections.

#### § 4. Reduction to the conventional formula

In order to check the validity of the result obtained in the previous section, let us examine in detail a limiting case where impurity atoms are distributed in a regular lattice.

For this purpose it will be in due order to review briefly the conventional way to derive the Hall coefficient on the basis of the Boltzmann equation. For simplicity we confine ourselves to an isotropic system. Longitudinal conductivity  $\sigma_{xx}$  and the Hall conductivity  $\sigma_H = \sigma_{xy}^a / H$  are respectively given in terms of the

distribution function  $f_{\mathbf{k}}$  for the electron with wave vector  $\mathbf{k}$  as<sup>3)</sup>

$$\sigma_{xx} = \sigma = -\frac{1}{3} e^2 \int v_{\mathbf{k}}^2 \tau(\mathbf{k}) \frac{\partial f_{\mathbf{k}}}{\partial \varepsilon_{\mathbf{k}}} d\mathbf{k}, \tag{4.1}$$

$$\sigma_H = \sigma_{xy} / H = -\frac{1}{3} e^2 \int v_{\mathbf{k}}^2 \frac{\tau^2(\mathbf{k})}{m_{\mathbf{k}}^* c} \frac{\partial f_{\mathbf{k}}}{\partial \varepsilon_{\mathbf{k}}} d\mathbf{k}. \tag{4.2}$$

In these expressions,  $\varepsilon_{\mathbf{k}}$  is the energy of the electron with wave vector  $\mathbf{k}$ ,  $v_{\mathbf{k}} = (1/\hbar) (\partial \varepsilon_{\mathbf{k}} / \partial \mathbf{k})$  is the velocity of the electron,  $m_{\mathbf{k}}^*$  effective mass and  $\tau(\mathbf{k})$  the relaxation time of the electron. When both  $\tau(\mathbf{k})$  and  $m_{\mathbf{k}}^*$  are constants independent of  $\mathbf{k}$ , it follows from (4.1) and (4.2) that

$$\sigma_H = \frac{\tau}{m^* c} \sigma \tag{4.3}$$

and the Hall coefficient  $R$  is given by

$$R = \frac{\sigma_H}{\sigma^2} = \frac{\tau}{m^* c \sigma}. \tag{4.4}$$

On substituting into (4.4) the usual expression for  $\sigma$

$$\sigma = \frac{ne^2}{m^*} \tau, \tag{4.5}$$

$R$  becomes

$$R = \frac{1}{nec} = R_0 \tag{4.6}$$

which is the same as (1.1).

Now we turn to the case of impurity band conduction. In the notation of this paper,  $\sigma_{xx}$  is written as

$$\sigma_{xx} = \frac{\pi e^2}{3\hbar} \int dE \sum_{\mathbf{k}} \left\{ \frac{dV_{\mathbf{k}}}{dk} \right\}^2 \left\{ G_{\mathbf{k}}(E) \right\}^2 \left\{ -\frac{\partial f(E)}{\partial E} \right\}. \tag{4.7}$$

As has been proved in M-K, in the limit of a regular lattice the Green's function becomes

$$G_{\mathbf{k}}^{(\pm)} = \frac{1}{E \pm is - V_{\mathbf{k}}}. \tag{4.8}$$

This is the Green's function of an electron with energy  $V_{\mathbf{k}}$ . Therefore, to make the correspondence more obvious, we take

$$V_{\mathbf{k}} = \varepsilon_{\mathbf{k}}$$

and add a small damping term  $\Gamma$  in the denominator of (4.8). Then

$$G_{\mathbf{k}}(E) = \frac{1}{\pi} \frac{\Gamma}{(E - \varepsilon_{\mathbf{k}})^2 + \Gamma^2}. \tag{4.9}$$

By noting that

$$\frac{1}{\hbar} \frac{dV_k}{dk} = \frac{1}{\hbar} \frac{\partial \varepsilon_k}{\partial k} = v_k,$$

$$\{G_k(E)\}^2 \approx \delta(E - \varepsilon_k) \frac{1}{\pi\Gamma}, \quad (4.10)$$

we may rewrite (4.7) as

$$\sigma = -\frac{e^2}{3} \int dE \int dk \frac{\pi}{\hbar} \left( \frac{\partial \varepsilon_k}{\partial k} \right)^2 \delta(E - \varepsilon_k) \frac{1}{\pi\Gamma} \frac{\partial f(E)}{\partial E}$$

$$= -\frac{e^2}{3} \int dk (v_k)^2 \frac{\hbar}{\Gamma} \frac{\partial f(\varepsilon_k)}{\partial \varepsilon_k}. \quad (4.11)$$

This is nothing but the formula (4.2) if  $\hbar/\Gamma \equiv \tau_k$ . In the next place we consider  $\sigma_H = \sigma_{xy}^a/H$  which is given by (3.20) and (3.21):

$$\sigma_H = \frac{e^3}{3\hbar^2 c} \iint dE_1 dE_2 \sum_{\mathbf{k}} \left\{ \frac{dV_k}{dk} \right\}^3 \frac{1}{k} G_k(E_1) G_k(E_2) \{G_k^0(E_2) - G_k^0(E_1)\}$$

$$\times \frac{f(E_1) - f(E_2)}{(E_1 - E_2)^2}. \quad (4.12)$$

The real part of the Green's function, in the limit of a regular lattice, is

$$G_k^0(E) = \frac{E - \varepsilon_k}{(E - \varepsilon_k)^2 + \Gamma^2} \quad (4.13)$$

and hence

$$G_k^0(E_1) - G_k^0(E_2) = \frac{(E_1 - E_2) \{ \Gamma^2 - (E_1 - \varepsilon_k)(E_2 - \varepsilon_k) \}}{\{(E_1 - \varepsilon_k)^2 + \Gamma^2\} \{(E_2 - \varepsilon_k)^2 + \Gamma^2\}}. \quad (4.14)$$

By substituting (4.9), (4.10), (4.14) into (4.12) and taking a limit of a very small  $\Gamma$ , we have

$$\sigma_H = \frac{e^3}{3\hbar^2 c} \iint dE_1 dE_2 \int \frac{d\mathbf{k}}{k} \left( \frac{\partial \varepsilon_k}{\partial k} \right)^3 \delta(E_1 - \varepsilon_k) \delta(E_2 - \varepsilon_k) \frac{f(E_1) - f(E_2)}{(E_1 - E_2)}$$

$$\times \frac{\Gamma^2 - (E_1 - \varepsilon_k)(E_2 - \varepsilon_k)}{\{(E_1 - \varepsilon_k)^2 + \Gamma^2\} \{(E_2 - \varepsilon_k)^2 + \Gamma^2\}}$$

$$= -\frac{e^3}{3c} \int d\mathbf{k} (v_k)^2 \left( \frac{1}{k} \frac{\partial \varepsilon_k}{\partial k} \right) \frac{\partial f(\varepsilon_k)}{\partial \varepsilon_k} \frac{1}{\Gamma^2}. \quad (4.15)$$

If we identify

$$\frac{1}{\hbar^2 k} \frac{\partial \varepsilon_k}{\partial k} = \frac{v_k}{\hbar k} = \frac{1}{m^*},$$

$$\frac{\hbar}{\Gamma} = \tau, \quad (4.16)$$

then (4.15) is precisely the same as (4.2). In this way we can recover all the conventional formulae for the Hall effect as a limiting case of the general expressions expressed in terms of the Green's functions.

The formal correspondence between two different approaches to the Hall effect becomes more complete when we consider an anisotropic system. For instance, from (3.15) we note

$$\frac{\partial}{\partial k_x} G_k(E) = \frac{\partial V_k}{\partial k_x} G_k(E) G_k^0(E) \tag{4.17}$$

and using this relation we can rewrite (3.14) as

$$\begin{aligned} \Xi^{xy}(E_1 E_2) = \frac{eH}{2\hbar c} \sum_k \left[ \left\{ \frac{\partial^2 V_k}{\partial k_x^2} \left( \frac{\partial V_k}{\partial k_y} \right)^2 - \frac{\partial^2 V_k}{\partial k_x \partial k_y} \frac{\partial V_k}{\partial k_y} \frac{\partial V_k}{\partial k_x} \right\} G_k(E_1) G_k(E_2) G_k^0(E_2) \right. \\ \left. + \left\{ \frac{\partial^2 V_k}{\partial k_x \partial k_y} \frac{\partial V_k}{\partial k_x} \frac{\partial V_k}{\partial k_y} - \frac{\partial^2 V_k}{\partial k_y^2} \left( \frac{\partial V_k}{\partial k_x} \right)^2 \right\} G_k(E_1) G_k(E_2) G_k^0(E_1) \right]. \tag{4.18} \end{aligned}$$

Assuming equivalence of  $x$ - and  $y$ -directions, we introduce anisotropic effective mass through the well-known relations

$$\begin{aligned} \frac{1}{\hbar^2} \frac{\partial^2 V_k}{\partial k_x^2} = \frac{1}{M_{xx}} = \frac{1}{\hbar^2} \frac{\partial^2 V_k}{\partial k_y^2}, \\ \frac{1}{\hbar^2} \frac{\partial^2 V_k}{\partial k_x \partial k_y} = \frac{1}{M_{xy}}. \end{aligned} \tag{4.19}$$

Then (4.18) becomes

$$\begin{aligned} \Xi^{xy}(E_1 E_2) = \frac{eH\hbar^3}{2c} \sum_k \left\{ \frac{1}{M_{xx}} v_y^2 - \frac{1}{M_{xy}} v_x v_y \right\} \\ \times G_k(E_1) G_k(E_2) \{G_k^0(E_2) - G_k^0(E_1)\}, \end{aligned} \tag{4.20}$$

and from (2.18) the Hall conductivity is given by

$$\begin{aligned} \sigma_H = \frac{e^3}{2c} \iint dE_1 dE_2 \sum_k \left\{ \frac{1}{M_{xx}} v_y^2 - \frac{1}{M_{xy}} v_x v_y \right\} G_k(E_1) G_k(E_2) \{G_k^0(E_2) - G_k^0(E_1)\} \\ \times \frac{f(E_1) - f(E_2)}{(E_1 - E_2)^2}. \end{aligned} \tag{4.21}$$

Hereafter we can follow the same step to the limiting process as for an isotropic system, and it is a straightforward task to derive

$$\sigma_H = -\frac{e^3}{c} \int d\mathbf{k} \left\{ \frac{1}{M_{xx}} v_y^2 - \frac{1}{M_{xy}} v_x v_y \right\} \tau_k^2 \left\{ \frac{\partial f(\epsilon_k)}{\partial \epsilon_k} \right\}. \tag{4.22}$$

This is exactly the generalized version of (4.2) obtained by Jones and Zener for an anisotropic system.<sup>9)</sup>

In the course of the above reduction to the conventional formula, we notice

in the Hall effect an important difference between the nearly free electron case and the impurity band conduction. The longitudinal conductivity is mainly contributed from the integral over the Fermi surface. This is also the case for the Hall conductivity of the nearly free electron as seen from (4.2). In the case of impurity band conduction, however, the general expression (4.12) shows that  $\sigma_H$  is not determined by the contribution from the Fermi surface alone, but rather by electrons with the whole range of energies. The manner of contribution to  $\sigma_H$  from each electron is not simple, depending on the energy shift and damping due to the randomness and also on the  $k$ -dependence of impurity potential.

### § 5. Weakly disordered case

We would gain more deep insight into the characteristic feature of the Hall effect in a random system by extending the discussion of the previous section to a case where the randomness in the distribution of impurity atoms is not large. We start as before with two expressions

$$\sigma = \frac{\pi e^2}{3\hbar} \int dE \sum_{\mathbf{k}} \left\{ \frac{dV_{\mathbf{k}}}{dk} \right\}^2 \{G_{\mathbf{k}}(E)\}^2 \left\{ -\frac{\partial f(E)}{\partial E} \right\} \quad (5.1)$$

and

$$\sigma_H = \frac{e^3}{3\hbar^2 c} \iint dE_1 dE_2 \sum_{\mathbf{k}} \left\{ \frac{dV_{\mathbf{k}}}{dk} \right\}^3 \frac{1}{k} G_{\mathbf{k}}(E_1) G_{\mathbf{k}}(E_2) \{G_{\mathbf{k}}^0(E_2) - G_{\mathbf{k}}^0(E_1)\} \frac{f(E_1) - f(E_2)}{(E_1 - E_2)^2}. \quad (5.2)$$

According to the previous paper M-K, the Green's function is approximately given by the solution of the following set of self-consistent equations:

$$G_{\mathbf{k}}^{(\pm)}(E) = \frac{\alpha^{(\pm)}}{1 - \alpha^{(\pm)} V_{\mathbf{k}}}, \quad (5.3)$$

$$\alpha^{(\pm)} = \frac{\bar{c}}{E \pm is - V_{\pm}^*}, \quad (5.4)$$

$$V_{\pm}^* = \frac{1}{N} \sum_{\mathbf{k}} \frac{V_{\mathbf{k}}}{1 - \alpha^{(\pm)} V_{\mathbf{k}}}. \quad (5.5)$$

$\bar{c}$  is the concentration of impurity atoms. From (5.4) and (5.5)  $V_{\pm}^*$  is generally a complex number dependent on only  $E$ . Therefore we may put

$$V_{\pm}^* = \Delta \pm i\Gamma. \quad (5.6)$$

Then, after substituting (5.4) and (5.6) into (5.3), we have

$$G_{\mathbf{k}}^{(\pm)}(E) = \frac{\bar{c}}{(E - \bar{c} V_{\mathbf{k}} - \Delta) \pm i(\Gamma + s)} \quad (5.7)$$

from which the imaginary part and real part are separated (see the definitions

(3.15) and (3.18)) as

$$G_k(E) = \frac{1}{\pi} \frac{\bar{c}\Gamma}{(E - \bar{c}V_k - \Delta)^2 + \Gamma^2}, \quad (5.8)$$

$$G_k^0(E) = \frac{\bar{c}\{E - \bar{c}V_k - \Delta\}}{(E - \bar{c}V_k - \Delta)^2 + \Gamma^2}. \quad (5.9)$$

$\Delta$  and  $\Gamma$  are the functions of  $E$ ; in order to know the  $E$ -dependence of these quantities we have to carry out somewhat laborious numerical calculations to solve Eqs. (5.4) and (5.5). This task is undertaken in the next paper, but before going into the details of the numerical results we content ourselves in this section with a qualitative discussion assuming the smallness of  $\Gamma$ .

As a function of  $E$ ,  $G_k(E)$  has generally a maximum at the energy  $E_k$  which is a root of an equation:

$$E_k = \bar{c}V_k + \Delta(E_k). \quad (5.10)$$

By expanding the denominator of (5.8) in a Taylor series around  $E_k$

$$\begin{aligned} E - \bar{c}V_k - \Delta(E) &= E - E_k - \left(\frac{\partial\Delta}{\partial E}\right)_{E=E_k} (E - E_k) + \dots \\ &\simeq \left\{1 - \left(\frac{\partial\Delta}{\partial E}\right)_{E=E_k}\right\} (E - E_k) \end{aligned} \quad (5.11)$$

and by putting

$$\Gamma_k = \frac{\Gamma(E_k)}{1 - (\partial\Delta/\partial E)_{E=E_k}}, \quad (5.12)$$

$G_k(E)$  can be approximated through a Lorentzian curve:

$$G_k(E) \simeq \frac{\bar{c}}{\{1 - (\partial\Delta/\partial E)_{E=E_k}\}} \frac{1}{\pi} \frac{\Gamma_k}{(E - E_k)^2 + \Gamma_k^2}. \quad (5.13)$$

In the same approximation

$$G_k^0(E) \simeq \frac{\bar{c}}{\{1 - (\partial\Delta/\partial E)_{E=E_k}\}} \frac{E - E_k}{(E - E_k)^2 + \Gamma_k^2}. \quad (5.14)$$

Therefore, within this approximation, except for a multiplication constant  $\bar{c}\{1 - (\partial\Delta/\partial E)_{E=E_k}\}^{-1}$ , the behaviour of the Green's function as a function of energy  $E$  is similar to the case discussed in § 4 in the limit of small  $\Gamma_k$ , and hence the evaluation of  $\sigma$  and  $\sigma_H$  can be carried out in almost the same manner as in the previous section. Thus we have finally

$$\sigma = -\frac{e^2}{3} \int d\mathbf{k} (v_k)^2 \frac{\hbar}{\Gamma(E_k)} \frac{\partial f(E_k)}{\partial E_k} \frac{\bar{c}^2}{\{1 - (\partial\Delta/\partial E)_{E=E_k}\}}, \quad (5.15)$$

$$\sigma_H = -\frac{e^3}{3c} \int d\mathbf{k} \frac{(v_k)^2}{m_k^*} \frac{\hbar^2}{\Gamma(E_k)^2} \frac{\partial f(E_k)}{\partial E_k} \frac{\bar{c}^3}{\{1 - (\partial\Delta/\partial E)_{E=E_k}\}}, \quad (5.16)$$

where use has been made of the following notation:

$$\frac{1}{\hbar} \frac{dV_k}{dk} = v_k, \quad \frac{v_k}{\hbar k} = \frac{1}{m_k^*}. \quad (5.17)$$

It turns out from these results that the Hall coefficient is given by

$$R = R_0 \left\{ \frac{\bar{c}}{1 - (\partial \Delta / \partial E)_{E_k = E_F}} \right\}, \quad (5.18)$$

where  $R_0$  is the Hall coefficient which would be derived from the Boltzmann equation for quasi-particles with energy  $E_k$ , velocity  $v_k$ , effective mass  $m_k^*$  and relaxation time  $\tau_k = \hbar / \Gamma(E_k)$ . On the other hand, if we calculate the state density by making use of the Green's function, we would have

$$D(E) = \sum_{\mathbf{k}} G_{\mathbf{k}}(E) \simeq \sum_{\mathbf{k}} \frac{\bar{c}}{\{1 - (\partial \Delta / \partial E)_{E_k = E_k}\}} \delta(E - E_k). \quad (5.19)$$

Let us define

$$D_0(E) = \sum_{\mathbf{k}} \delta(E - E_k) \quad (5.20)$$

and

$$g = \frac{D(E_F)}{D_0(E_F)} = \frac{\bar{c}}{\{1 - (\partial \Delta / \partial E)_{E_k = E_F}\}}. \quad (5.21)$$

Then from (5.18), it follows that

$$R/R_0 = 1/g. \quad (5.22)$$

This is the form similar to a relation conjectured by Mott in connection with the Hall coefficient of liquid metals.<sup>10)</sup>

We have not clearly indicated the condition to justify the above argument. In the hydrogenic model in Ge or Si discussed in the next paper, we shall find that the assumption of "weakly disordered case" is completely broken down.

## § 6. Sign of the Hall coefficient

In this section we briefly discuss some qualitative aspect of the results obtained for the case of hydrogenic model. More detailed account of numerical calculation and of comparison with experiments will be shortly reported in a forthcoming paper.

Taking the simplest model for the impurity state in semiconductors, we assume hydrogen-like 1s-state wave function

$$\phi_0(\mathbf{r} - \mathbf{R}) = \left( \frac{a^3}{\pi} \right)^{1/2} \exp(-a|\mathbf{r} - \mathbf{R}|). \quad (6.1)$$

The transfer energy is given by

$$V_{mn} = -V_0(1 + aR_{mn})\exp(-aR_{mn}) \tag{6.2}$$

with

$$V_0 = \frac{e^2 a}{\kappa_0} = 2\epsilon_0 \tag{6.3}$$

where  $\epsilon_0$  is the ionization energy of the 1s-state,  $\kappa_0$  dielectric constant and  $a^{-1}$  the radius of the 1s-orbit. Hereafter we adopt the continuum approximation and ignore the Brillouin zone effect, replacing the sum over wave vector  $\mathbf{k}$  by an integral over whole  $\mathbf{k}$ -space. Then from (3.21)  $\sigma_H = \sigma_{xy}^a/H$  is written as

$$\sigma_H = -\frac{e^3}{\hbar^2 c} \iint dE_1 dE_2 \frac{f(E_1) - f(E_2)}{(E_1 - E_2)^2} \frac{4\pi}{3} \int d\mathbf{k} k \left(\frac{dV_k}{dk}\right)^3 \times G_k(E_1) G_k(E_2) \{G_k^0(E_1) - G_k^0(E_2)\}, \tag{6.4}$$

where  $V_k$  is the Fourier transform of (6.2) given by

$$V_k = -V_0 \frac{32\pi a^3}{(a^2 + k^2)^3}. \tag{6.5}$$

When this  $V_k$  is inserted in (6.4), and the numerical results for the Green's function computed with the approximation same as in M-T are used, it turns out that  $\sigma_H$  has definitely negative sign irrespective of the number of electrons filling the impurity band. To see this we first notice that, for given  $k$ , the imaginary part of the Green's function  $G_k(E)$  is positive definite in a certain energy range, say  $E_b \leq E \leq E_t$ , and vanishes otherwise. On the other hand, according to the numerical calculation, the real part  $G_k^0(E)$  is given as a monotonously varying function of  $E$  within the same energy range. Figure 4 shows an example of the graphs for  $G_k(E)$  and  $G_k^0(E)$  calculated on the basis of the hydrogenic model.\*) Since the Fermi distribution function  $f(E)$  is also a monotonously decreasing function of  $E$ , the integrand of (6.4)

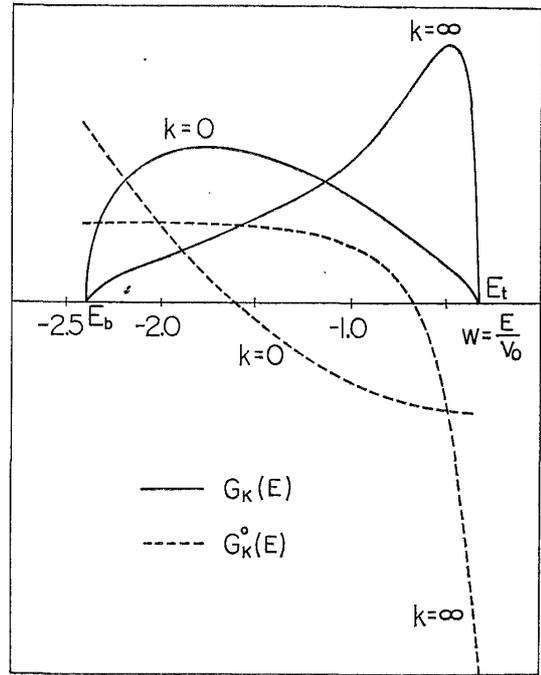


Fig. 4.  $G_k(E)$  and  $G_k^0(E)$  are shown as functions of  $w = E/V_0$  for two particular values of  $k$ ;  $k=0$  and  $k=\infty$ . The concentration of impurity atoms is taken as  $32\pi N_i/a^3 = 0.5$ .

\*) These figures are the results of our new calculation carried out by making use of the M-T approximation.<sup>4)</sup>

$$k \left( \frac{dV_k}{dk} \right)^3 \frac{\{f(E_1) - f(E_2)\} \{G_k^0(E_1) - G_k^0(E_2)\}}{(E_1 - E_2)^2} G_k(E_1) G_k(E_2)$$

is always positive, and hence

$$\sigma_H \leq 0. \quad (6.6)$$

This means that the sign of the Hall coefficient in the impurity band conduction for  $n$ -type semiconductors is always negative as has been predicted by Mott.<sup>11)</sup> This conclusion is in remarkable contrast to the case of the usual band electrons, where the sign of the Hall coefficient is either negative or positive according as the Fermi surface is either electron-like or hole-like.

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