DRIFT MOBILITY OF LARGE POLARONS IN THE INTERMEDIATE COUPLING REGION

A. WEIJLAND

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CHAPTER I

THE POLARON-PHONON SYSTEM AND THE KUBO FORMALISM

Synopsis

Starting from Kubo's expression for the frequency dependent electrical conductivity, the low-temperature drift mobility of large polarons is evaluated in the intermediate coupling region

The first part of this study is devoted to a detailed discussion of the method, which is based on the fact that the Kubo formula can be adapted to the intermediate coupling region by using a unitary transformation and expanding the conductivity in a power series expansion in the polaron-phonon coupling

The unitary transformation which has been used generates a modified L L P. polaron model, maintaining the many polaron character of the theory An expression for the effective mass of drifting polarons is derived, which in general depends on the temperature, the frequency of the applied field and the coupling constant.

In the second part the effective mass, the d.c drift mobility and the corresponding correction terms will be calculated explicitly.

1. *Introduction.* Much work has already been devoted to the mobility problem of a slow electron in the conduction band of a polar crystal. The large number of expressions, derived by various authors, ref. 1 to ref. 9, differ considerably in the experimentally interesting situation, ref. 10. The main reason for these large differences is that the interaction between the electron and the field of longitudinal optical phonons is rather strong. If one expresses the strength of the electron-phonon interaction by a dimensionless parameter α, it turns out that for many polar crystals α is about 3. As the electron moves through the crystal, it distorts the surrounding lattice and the distortion in turn acts back on the electron. In the language of field theory this means that the electron is dressed with a cloud of virtual phonons. The quasi-particle consisting of an electron plus a virtual phonon cloud, is called a polaron. The determination of the properties of the polaron in the intermediate coupling region, where α is about 3, constitutes the main problem. Up till now, an exact solution of this problem cannot be obtained, not even for small polaron velocities. However, the coupling strengths one

is interested in, are too weak for an adiabatic method to be applicable and too strong to use traditional perturbation theory.

As we are mainly interested in the transport properties of polarons, we only mention two methods attacking the polaron problem. The first one is the intermediate coupling theory of Lee, Low and Pines[2]), who used two canonical transformations to convert the electron into a polaron. They obtained reasonable results in the intermediate coupling region. The second method is that of Feynman[11]), who approximates the effect of the lattice field by a fictitious particle, bound to the electron with a spring. For the polaron problem the last method is probably the best one, but for the transport problem this is not so clear (See ref. 10).

Depending on the accuracy of the polaron model, the polaron will emit and reabsorb virtual phonons more or less intensively. At nonzero temperatures, however, when there are real phonons in the field, we have in addition to consider the effect of polaron-thermal phonon scattering processes.

These thermal phonons induce transitions between the quasi-particle states of the polaron. The energy of these quasi-particle states must be below the phonon emission threshold, that is the energy of the polaron must be less than the phonon frequency, otherwise the polaron is able to emit a phonon, a proces that is not described by the quasi-particle states. Then the polaron concept breaks down. When the temperature is low enough one expects that this condition is satisfied. At the other hand the temperature must be sufficiently high to neglect all but the scattering by the longitudinal optical phonons.

If these conditions are satisfied one may consider the response of the system to a small external electric field. In principle the philosophy of most authors, attacking the mobility problem, is simple. The starting point is the assumption that the polaron-phonon system is governed by the Hamiltonian:

$$H = E(\boldsymbol{p}) + \sum_{\boldsymbol{q}} \omega b_{\boldsymbol{q}}^{\dagger} b_{\boldsymbol{q}} + \sum_{\boldsymbol{q}} \lambda_{\boldsymbol{q}} Z_{\boldsymbol{q}}^{\frac{1}{2}} (e^{i\boldsymbol{q}\cdot\boldsymbol{r}} b_{\boldsymbol{q}} + e^{-i\boldsymbol{q}\boldsymbol{r}} b_{\boldsymbol{q}}^{\dagger}). \tag{1.1}$$

Here $E(\boldsymbol{p})$ is the energy of the polaron being a function of the polaron momentum in the lowest quasi-particle state, $b_{\boldsymbol{q}}^{\dagger}$, $b_{\boldsymbol{q}}$ are phonon creation and destruction operators for a phonon in the mode $\boldsymbol{q}$, while ω is the corresponding phonon frequency, being independent of $\boldsymbol{q}$. The coupling of the lattice mode $\boldsymbol{q}$ with the Fourier component of the polaron density, $e^{i\boldsymbol{q}\cdot\boldsymbol{r}}$, is given by $\lambda_{\boldsymbol{q}} Z_{\boldsymbol{q}}^{\frac{1}{2}} \cdot Z_{\boldsymbol{q}}$ represents the renormalization of the electron-phonon coupling $\lambda_{\boldsymbol{q}}$, due to the fact that the polaron has an internal structure. The charge of the electron is smeared out over a certain region in a similar way as is indicated by the adiabatic and Feynman descriptions of the polaron. Now it is not clear that the actual polaron-phonon system is governed by the Hamiltonian (1.1). For instance the L.L.P. polaron, a reasonable model in the intermediate coupling region, has a quite different interaction with

thermal phonons. But even if we accept (1.1), the question arises how to choose the polaron energy $E(\boldsymbol{p})$. One often assumes a parabolic approximation $E(\boldsymbol{p}) = E_0 + p^2/2m^*$, where m^* is the effective polaron mass. This means that in a polaron-phonon scattering process, the polaron, after absorbing a phonon, is still a simple particle with the same effective mass m^*. The next assumption is the validity of the Boltzmann equation to describe the transport process. For a d.c. field the appropriate Boltzmann equation reads:

$$\frac{e\boldsymbol{E}}{m^*} V_{\boldsymbol{p}} f_{\boldsymbol{p}} = \left(\frac{\partial f_{\boldsymbol{p}}}{\partial t}\right)_{\text{coll.}}. \tag{1.2}$$

Here $f_{\boldsymbol{p}}$ is the occupation probability density for the quasi-particle state of momentum $\boldsymbol{p}$. The quantity $e\boldsymbol{E}/m^*$ represents the average acceleration of the quasiparticle between the collisions, due to the applied electric field $\boldsymbol{E}$. The use of the equations (1.2) and (1.1) implies a number of assumptions, which are well discussed in the literature (Schultz, thesis[3]). Most important is the assumption that it is meaningful to speak of a quasi-particle and the lattice field as if they were physically separable, the former possessing well defined velocities being characterized by the probability density function $f_{\boldsymbol{p}}$, the lattice field as an infinite sea of phonons at constant temperature that somehow maintains the equilibrium distribution. The interaction between the two must be weak and is described in the form of collision processes, occurring independently of each other. At low temperatures, when $kT \ll \hbar\omega$, these assumptions may be reasonable. But actually, the interaction between the polaron and the lattice remains intermediate or strong. This means that the probability of higher order processes, characterizing the creation and annihilation of virtual phonons during collision processes, are not a priori negligible.

Another objection against the Boltzmann equation approach must be raised. Because of the highly inelastic polaron-phonon collisions the solution of the Boltzmann equation is extremely difficult for all temperatures.

Howarth and Sondheimer[8]), using variational methods, obtained approximate solutions when all polaron effects may be neglected. Up till now we have mentioned some of the serious drawbacks of the Boltzmann equation approach. It suggests that one must avoid the use of the Boltzmann equation as an a priori starting point. We want to do this for coupling constants in the intermediate region, and in principle for finite frequencies of the applied electric field. In fact, we shall use a density matrix approach. A density matrix approach for the polaron mobility problem has already been given by Platzmann, Feynman, Hellswarth and Iddings[7]) using the Feynman model of the polaron. But in the low-temperature region there are serious doubts to their results, as has been pointed out by Kadanoff[9]). In principle a density matrix approach for the transport problem of an electron in the conduction band of a polar crystal can be

formulated without using a polaron model explicitly. Such a procedure is satisfying because the mobility expression will not depend on the specific properties of some hypothetical polaron model. But since the exact density matrix expressions cannot be solved exactly we have to expand the expressions in a perturbation series that must converge rapidly.

It is for that reason that we start with an exact canonical transformation of the Fröhlich Hamiltonian much like Lee, Low and Pines[2]) did in their polaron model. Since the expression for the frequency dependent electrical conductivity, as given among others by Bonch-Bruevich[15]), can exactly be transformed by the unitary transformation, we only have to fulfil the demand of rapid convergence of the perturbation expansion without introducing the assumptions of the ordinary Boltzmann equation approach. We will call the transformed operators of the electrons, polaron operators, though the polaron will radiate virtual phonons during collision processes with thermal phonons. In fact the transformation is choosen in such a way that a part of the lattice deformation is rigidly attached to the electron while the interaction that remains is one with thermal phonons only.

It is for that reason that we shall not find a renormalization of the coupling constant. The perturbation series of the linear deviation from the polaron equilibrium distribution has in the limit of zero frequency a certain class of contributions that are predominant. In summing this class of contributions we shall show that the result gives the lowest order contribution to the polaron conductivity. The formalism will give us immediately an expression for the effective mass of a slow drifting polaron which in general depends on the temperature, the frequency of the applied field, the coupling constant and the phonon frequency.

In the second part of our investigation we apply the theory to the L.L.P. polaron model and calculate mobility and effective mass quantitatively. Comparing our results with those obtained by others, shows reasonable agreement especially with Osaka's intermediate coupling calculations[5]).

2. *The polaron-phonon system.* In general we may write the Hamiltonian of an electron interacting with longitudinal optical phonons as:

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{V} \tag{2.1}$$

where in the occupation number representation:

$$\mathscr{H}_0 = \sum_{\boldsymbol{k}} \frac{k^2}{2m} a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} + \sum_{\boldsymbol{q}} \omega b_{\boldsymbol{q}}^\dagger b_{\boldsymbol{q}} \tag{2.2}$$

$$\mathscr{V} = \sum_{\boldsymbol{kq}} a_{\boldsymbol{k}+\boldsymbol{q}}^\dagger a_{\boldsymbol{k}} (\lambda_{\boldsymbol{q}} b_{\boldsymbol{q}} + \lambda_{\boldsymbol{q}}^* b_{-\boldsymbol{q}}^\dagger) \tag{2.3}$$

$$\lambda_{\boldsymbol{q}} = -\frac{i\omega}{q} \cdot \left(\frac{1}{2m\omega}\right)^{\frac{1}{4}} \cdot \left(\frac{4\pi\alpha}{\Omega}\right)^{\frac{1}{2}}. \tag{2.4}$$

We take Planck's constant $\hbar = h/2\pi = 1$.

$a_k^\dagger$, a_k are creation and annihilation operators for an electron in the state $|k\rangle$, with energy $k^2/2m$. They obey the anti-commutation relations:

$$[a_{k_1}, a_{k_2}^\dagger]_+ = \delta_{k_1, k_2}, [a_{k_1}, a_{k_2}]_+ = 0 \text{ etc.}$$

$b_q^\dagger$, b_q are the corresponding quantities for the phonons, obeying the commutation relations:

$$[b_{q_1}, b_{q_2}^\dagger]_- = \delta_{q_1, q_2}, [b_{q_1}, b_{q_2}]_- = 0 \text{ etc.}$$

The wave vectors k and q run over the points of an infinite cubic lattice in momentum space with a lattice spacing $2\pi\Omega^{-\frac{1}{3}}$; Ω is the volume of the cube in which the system is enclosed with the usual periodic boundary conditions. The Hamiltonian (2.1) has been derived by Fröhlich[13]). The essential assumption is that he replaces the lattice by a dielectric: a macroscopic continuum, whose properties are determined by a dielectric constant ε.

The dielectric constant enters in (2.4) by means of the coupling constant

$$\alpha = e^2 \cdot \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon}\right) \sqrt{\frac{m}{2\omega}}.$$

Fröhlich supposed that the dependence of ε and the phonon frequency ω on the wave vector q may be neglected. This assumption is quite good provided that the wave lengths with which we have to deal are long compared with the atomic distances. The kinetic energy of the electron is $k^2/2m$ where m is determined by the band structure of the crystal, α measures the strength of the interaction between the electron and the polarization field and is in the experimental interesting range about 3.

The most general one-electron state of (2.1) reads:

$$|\Psi_K^{(1)}\rangle = \sum_{n=0}^{\infty} \frac{1}{\sqrt{Z_K}} \frac{1}{n!} \sum_{kq_1 \ldots q_n} \delta(K - \sum_{i=1}^{n} q_i - k)\, C_k^{(n)}(k; q_1 \ldots q_n) \times \\ \times b_{q_1}^\dagger \ldots b_{q_n}^\dagger a_k^\dagger |0\rangle \qquad (2.5)$$

where $|0\rangle$ means the unperturbed, normalized vacuum state for electrons and phonons, and Z_K is the normalizing constant given by:

$$\langle \Psi_{K_1}^{(1)} | \Psi_{K_2}^{(1)} \rangle = \delta_{K_1, K_2} \qquad (2.6)$$

The delta function enters in (2.5) because the system (2.1) is invariant for translation; that is the total momentum operator:

$$\mathcal{P} = \sum_k k a_k^\dagger a_k + \sum q b_q^\dagger b_q \qquad (2.7)$$

commutes with $\mathcal{H}$, K is an eigenvalue of $\mathcal{P}$:

$$\mathcal{P} |\Psi_K^{(1)}\rangle = K |\Psi_K^{(1)}\rangle.$$

Because the probability amplitudes $C_K^{(n)}(k; q_1 \ldots q_n)$ cannot be found

exactly, L.L.P. approximate $C_K^{(n)}(\boldsymbol{k};\, \boldsymbol{q}_1 \ldots \boldsymbol{q}_n)$ as:

$$C_K^{(n)}(\boldsymbol{k};\, \boldsymbol{q}_1 \ldots \boldsymbol{q}_n) = \prod_{j=1}^{n} F(\boldsymbol{q}_j) \tag{2.8}$$

or

$$|\Psi_K^{(1)}\rangle' = \sum_{n=0}^{\infty} \frac{1}{\sqrt{Z_K}} \int \mathrm{d}\boldsymbol{r} \exp\{i(\boldsymbol{K}-\boldsymbol{k})\,\boldsymbol{r} \exp \sum_{\boldsymbol{q}} \cdot F_{\boldsymbol{q}} \exp(-i\boldsymbol{q}\cdot\boldsymbol{r})\, b_{\boldsymbol{q}}^{\dagger}\} a_{\boldsymbol{k}}^{\dagger} |0\rangle \quad (2\,8a)$$

This assumption means that all virtual phonons in the polaron cloud are emitted into the same state, characterized by a momentum distribution $F_{\boldsymbol{q}}$. Thus (2.8) neglects all correlation effects between the phonons. In fact (2.8*a*) is the exact solution of (2.1) if the electron has infinite mass.

Another aspect of this trial function is that the lattice deformation follows the electron instantaneously. This can been shown by writing (2.5) and (2.8) in the coordinate representation of the electron, maintaining the occupation number representation of the phonons.

Let $|\boldsymbol{r}'\rangle$ be an eigenvector of the electron position operator $\boldsymbol{r} : \boldsymbol{r}\,|\boldsymbol{r}'\rangle = \boldsymbol{r}'\,|\boldsymbol{r}'\rangle$. Then (2.5) together with (2.8) reads:

$$\langle \boldsymbol{r}'|\Psi_K\rangle' = \frac{1}{\sqrt{Z_K}} \sum_{\boldsymbol{k}} \int \mathrm{d}\boldsymbol{r} \exp\{i(\boldsymbol{K}-\boldsymbol{k})\,\boldsymbol{r} + \sum_{\boldsymbol{q}} F_{\boldsymbol{q}} \exp(-i\boldsymbol{q}\cdot\boldsymbol{r}) b_{\boldsymbol{q}}^{\dagger}\} \langle \boldsymbol{r}'\,|a_{\boldsymbol{k}}^{\dagger}|\,0\rangle \tag{2.9}$$

Because $\langle \boldsymbol{r}'\,|a_{\boldsymbol{k}}^{\dagger}|\,0\rangle$ is equal to the free electron wave function $\mathrm{e}^{i\boldsymbol{k}\boldsymbol{r}'}$ and $\boldsymbol{r}$ is the position coordinate of the lattice deformation, the summation over $\boldsymbol{k}$ gives the delta function: $\delta(\boldsymbol{r}' - \boldsymbol{r})$. This shows the statement above.

This aspect of the Lee, Low and Pines model is responsible for the fact that it is not easy to renormalize the polaron-phonon coupling as is done for instance in the Feynman theory of the polaron (see equation (1.1). The trial function (2.9) can simply be generated by the unitary operator:

$$U = \exp -\sum_{\boldsymbol{k}\boldsymbol{q}} a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}+\boldsymbol{q}} (F_{-\boldsymbol{q}}^{*} b_{-\boldsymbol{q}} - F_{\boldsymbol{q}} b_{\boldsymbol{q}}^{\dagger}). \tag{2.10}$$

Applying a unitary transformation to the electron operators, we define the polaron operators:

$$\alpha_{\boldsymbol{k}}^{\dagger} = U a_{\boldsymbol{k}}^{\dagger} U^{-1}, \quad \text{etc.} \quad \text{with} \quad \alpha_{\boldsymbol{k}}^{\dagger}\,|0\rangle = |\Psi_{\boldsymbol{k}}^{(1)}\rangle' \tag{2.11}$$

$\alpha_{\boldsymbol{k}}^{\dagger}$ and $\alpha_{\boldsymbol{k}}$ obey of course the same anti-commutation relations as do the operators $a_{\boldsymbol{k}}^{\dagger}$ and $a_{\boldsymbol{k}}$.

The new phonon operators are

$$\beta_{\boldsymbol{q}}^{\dagger} = U b_{\boldsymbol{q}}^{\dagger} U^{-1}, \text{ etc.} \tag{2.12}$$

or

$$\beta_{\boldsymbol{q}}^{\dagger} = b_{\boldsymbol{q}}^{\dagger} - F_{\boldsymbol{q}}^{*} \sum_{\boldsymbol{k}} a_{\boldsymbol{k}}^{\dagger} a_{\boldsymbol{k}+\boldsymbol{q}} \quad \text{while} \quad [\beta_{\boldsymbol{q}_1}, \beta_{\boldsymbol{q}_2}^{\dagger}] = \delta_{\boldsymbol{q}_1, \boldsymbol{q}_2} \text{ etc.}$$

The vacuum state $|0\rangle$ remains invariant under the transformation.

Rewriting the Hamiltonian in terms of the new operators we find: $\mathscr{H} = H_0 + V_1 + V_2$, where:

$$H_0 = \sum_k E_k \alpha_k^\dagger \alpha_k + \sum_q \omega_q \beta_q^\dagger \beta_q. \tag{2.13}$$

$$V_1 = \sum_{kq} \alpha_k^\dagger \alpha_{k+q} (V_{-q}^*(k)\, \beta_{-q} + V_q(k+q)\, \beta_q^\dagger) \tag{2.14}$$

$$V_2 = \sum_{kq_1q_2} \alpha_k^\dagger \alpha_{k+q_1+q_2} \frac{q_1 q_2}{2m} (F_{-q_1}^* F_{-q_2}^* \beta_{-q_1}, \beta_{-q_2} + \text{c.c.}) - \\ - \sum_{\substack{kq_1q_2 \\ q_1 \neq -q_2}} \alpha_k^\dagger \alpha_{k+q_1+q_2} \frac{q_1 q_2}{2m} 2F_{q_1} F_{-q_2}^* \beta_{q_1}^\dagger \beta_{-q_2} \tag{2.15}$$

and

$$E_k = \frac{1}{2m} (k - \sum_q q|F_q|^2)^2 + \sum_q (\lambda_q F_q + \lambda_q^* F_q^*) + \sum_q |F_q|^2 \left(\omega + \frac{q^2}{2m}\right) \tag{2.16}$$

$$V_{-q}^*(k) = \lambda_q + F_{-q}^*(E_{k+q} - E_k + \omega) \tag{2.17}$$

$$V_q(k+q) = \lambda_q^* - F_q(E_{k+q} - E_k - \omega), \tag{2.18}$$

while

$$\omega_q = \omega + \frac{q^2}{m} |F_q|^2 \cdot N, \tag{2.19}$$

N is the total number of polarons. In deriving (2.13), (2.14) and (2.15) we neglected terms of order of the square of the polaron density. One recovers the results of Lee, Low and Pines if we minimize E_k in (2.16) with respect to F_q.
Then

$$\lambda_q + F_q^* \left(\omega + \frac{q^2}{2m} - \frac{q}{m} (k - \sum_{q_1} q_1 (|F_{q_1}|^2))\right) = 0 \tag{2.20}$$

and

$$E_k = -\alpha\omega + \frac{k^2}{2m'}, \quad m' = m(1 + \alpha/6) \tag{2.21}$$

The second order correction to E_k is then, using ordinary perturbation theory

$$\Delta E_k = -0.0159\alpha^2\omega - 0.02 \frac{\alpha^2}{(1+\alpha/6)^2} \cdot \frac{k^2}{2m}. \tag{2.22}$$

Thus for $\alpha = 3$ the correction to the ground state energy is 6% while the effective mass is changed by about 5%.

This result indicates that the L.L.P. polaron model is good enough to describe the transport problem of polarons in the experimentally interesting range of coupling constants.

Note, that the total momentum operator is left invariant. Thus

$$\mathscr{P} = \sum_k \boldsymbol{k}\alpha_k^\dagger\alpha_k + \sum_q \boldsymbol{q}\beta_q^\dagger\beta_q. \tag{2.23}$$

We have to remark that in their one-electron description L.L.P. use a second unitary transformation to transform the centre of the coordinate system to the actual position of the electron. As a consequence their transformed Hamiltonian does not contain the polaron operators any longer, but is more complicated. We prefer the description, given above, though the results are completely the same. Note that the theory can trivially be extended to the case of high electron density, taking into account the electron-electron interaction.

3. *The correlation functions.* Let the system, described by the Hamiltonian (2.1), be initially in thermodynamic equilibrium. At time $t = -\infty$ we adiabatically switch on an electric field of frequency ω.

The interaction between the system and the field is given by:

$$\Lambda H = \boldsymbol{E}\cdot\mathscr{D}\, e^{i\omega t - \varepsilon|t|} \tag{3.1}$$

where $\mathscr{D}$ stands for the total dipole moment of the system:

$$\mathscr{D} = -e\int \mathrm{d}\boldsymbol{r}\cdot\boldsymbol{r}\psi_r^\dagger\psi_r. \tag{3.2}$$

In this expression $\psi_r^\dagger\psi_r$ is the electron density operator, ψ_r is related to a_k by:

$$\psi_r = \sum_k a_k \frac{e^{ikr}}{\sqrt{\Omega}}.$$

Following among others Bonch-Bruevich[15]) we may write the frequency dependent electrical conductivity as:

$$\sigma_{\mu\nu}(z) = i\int_0^\infty e^{izt}\,\mathrm{d}t\langle[\mathscr{J}_\mu(t), \mathscr{D}_\nu(0)]\rangle \tag{3.3}$$

where $z = \omega + i\varepsilon$. $\mathscr{J}_\mu(t)$ is the Heisenberg operator of the μ-th component of the current operator:

$$\mathscr{J} = -\frac{e}{\Omega}\sum_k \nabla_k T_k a_k^\dagger a_k. \tag{3.4}$$

T_k is the kinetic energy of the electron, given by $k^2/2m$.

The expression $\langle[\mathscr{J}_\mu(t), \mathscr{D}_\nu]\rangle$ has the meaning of a grand canonical ensemble average of the commutator of the operators $\mathscr{J}_\mu(t)$ and $\mathscr{D}_\nu$ over systems, described by the Hamiltonian (2.1); Thus

$\langle[A, B]\rangle$ = Trace $\{\rho[A, B]\}$ where the density matrix ρ is given by

$$e^{-\beta(\mathscr{H}-\mu N)}/\mathrm{Tr}\{e^{-\beta(\mathscr{H}-\mu N)}\},\ \beta = \frac{1}{kT}.$$

The definition of the drift mobility follows immediately from $\sigma_{\mu\nu}(z)$ by the relation:

$$\sigma_{\mu\nu}(z) = e\bar{n}\mu_{\mu\nu}(z). \tag{3.5}$$

$\bar{n}$ is the average electron density in the system, N/Ω.

From (3.3) we see that we have to evaluate the correlation function: $\langle\langle \mathscr{J}_\mu; \mathscr{D}_\nu \rangle\rangle_z$ defined by:

$$\langle\langle \mathscr{J}_\mu; \mathscr{D}_\nu \rangle\rangle_z = -\frac{ie}{\Omega} \sum_{\boldsymbol{k}} (V_{\boldsymbol{k}} \cdot T_{\boldsymbol{k}})_\mu \int_0^\infty e^{izt}\, dt \langle [e^{i\mathscr{H}t} a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}}\, e^{-i\mathscr{H}t}, \mathscr{D}_\nu] \rangle \tag{3.6}$$

Then the definition of $\langle\langle a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}}; \mathscr{D} \rangle\rangle_z$ is:

$$\langle\langle a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}}; \mathscr{D} \rangle\rangle_z = i \int_0^\infty e^{izt}\, dt \langle [e^{i\mathscr{H}t}\, a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}}\, e^{-i\mathscr{H}}, \mathscr{D}] \rangle. \tag{3.7}$$

As we will express the correlation functions in terms of polaron operators we use the unitary transformation (2.10) to obtain:

$$\mathscr{J} = \frac{-e}{m\Omega} \sum_{\boldsymbol{k}} \boldsymbol{k} a_{\boldsymbol{k}}^\dagger a_{\boldsymbol{k}} = \frac{-e}{m\Omega} \sum_{\boldsymbol{k}} \boldsymbol{k}\, U^{-1} \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}}\, U =$$

$$= \frac{-e}{m\Omega} \{ \sum_{\boldsymbol{k}} (\boldsymbol{k} - \sum_{\boldsymbol{q}} \boldsymbol{q} |F_{\boldsymbol{q}}|^2)\, \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}} - \sum_{\boldsymbol{kq}} \boldsymbol{q} \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}+\boldsymbol{q}} (F_{-\boldsymbol{q}}^* \beta_{-\boldsymbol{q}} - F_{\boldsymbol{q}} \beta_{\boldsymbol{q}}^\dagger) \}$$

$$\mathscr{J} = \boldsymbol{J} + \boldsymbol{J}' \tag{3.8}$$

where we have neglected terms of order $\bar{n}^2$. Noting that the total dipole moment $\mathscr{D}$ is invariant under the transformation we may write the electrical conductivity as:

$$\sigma_{\mu\nu}(z) = \langle\langle J_\mu; \mathscr{D}_\nu \rangle\rangle_z + \langle\langle J'_\mu; \mathscr{D}_\nu \rangle\rangle_z \tag{3.9}$$

where

$$\langle\langle J_\mu; \mathscr{D}_\nu \rangle\rangle_z = -\frac{e}{m\Omega} \sum_{\boldsymbol{k}} (k_\mu - \sum_{\boldsymbol{q}} q_\mu |F_{\boldsymbol{q}}|^2)\, \langle\langle \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}}; \mathscr{D}_\nu \rangle\rangle_z \tag{3.10}$$

and

$$\langle\langle J'_\mu; \mathscr{D}_\nu \rangle\rangle_z = \frac{e}{m\Omega} \sum_{\boldsymbol{kq}} q_\mu \langle\langle \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}+\boldsymbol{q}} (F_{-\boldsymbol{q}}^* \beta_{-\boldsymbol{q}} - F_{\boldsymbol{q}} \beta_{\boldsymbol{q}}^\dagger); \mathscr{D}_\nu \rangle\rangle_z \tag{3.11}$$

As is pointed out by L.L.P., ref. 2, the vector-function $\sum_{\boldsymbol{q}} \boldsymbol{q} |F_{\boldsymbol{q}}|^2$ may be written as $\eta_{\boldsymbol{k}} \boldsymbol{k}$. Using this expression, (3.10) can be simplified in the low-temperature region. Then the correlation function $\langle\langle \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}}; \mathscr{D}_\nu \rangle\rangle_z$ falls of rapidly with increasing $\boldsymbol{k}$ and $\eta_{\boldsymbol{k}}$ can, in a good approximation, be replaced by its value for $\boldsymbol{k} = 0$. Thus, for low temperatures, we may write instead of (3.10)

$$\langle\langle J_\mu; \mathscr{D}_\nu \rangle\rangle_z = \frac{-e}{m'\Omega} \sum_{\boldsymbol{k}} k_\mu \langle\langle \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}}; \mathscr{D}_\nu \rangle\rangle_z \tag{3.10a}$$

where the effective mass $m' = m/(1 - \eta_0) = m(1 + \alpha/6)$ in the case of the L.L.P. polaron model.

As an illustration of the use of the Kubo formula we shall give a simple example. The expression for the conductivity, (3.3), takes on a very obvious form in the special case when the current correlation function is simply exponentially damped.

$$\langle[\mathscr{J}_\mu(t), \mathscr{D}_\nu]\rangle = \langle[\mathscr{J}_\mu, \mathscr{D}_\nu]\rangle \, \mathrm{e}^{-|t|/\tau} \tag{3.12}$$

Since

$$\langle[\mathscr{J}_\mu, \mathscr{D}_\nu]\rangle = \frac{e^2}{m\Omega} \langle[\sum_k k_\mu a_k^\dagger a_k, \mathscr{D}_\nu]\rangle = -\frac{ie^2\bar{n}}{m} \delta_{\mu\nu}$$

we find

$$\sigma_{\mu\nu}(z) = \frac{\bar{n}e^2}{m} \delta_{\mu\nu} \frac{\tau}{(z^2\tau^2 + 1)} (1 + iz\tau) \tag{3.13}$$

This frequency dependence is well known from the usual transport theory. As we have pointed out, the situation is more complicated in the case of polarons. The assumption (3.12) is identical with the use of a simplified Boltzmann equation but the introduction of a relaxation time τ, at least in this simple form, seems to be unlikely. But, as we shall show, the expression (3.13) is justified provided that we replace m by an effective mass m^* keeping the temperature low enough.

Before going into a detailed investigation we discuss some useful conservation laws. The conserved quantities are: The polaron number operator; $N = \sum_k \alpha_k^\dagger \alpha_k$. The total momentum $\mathscr{P} = \sum_k \boldsymbol{k} \alpha_k^\dagger \alpha_k + \sum \boldsymbol{q} \beta_q^\dagger \beta_q$, the energy $\mathscr{H}$ and the total dipole moment $\mathscr{D}$. From the conservation of total number of polarons follows

$$\langle[N, \mathscr{D}]\rangle = 0 \quad \text{or} \quad \sum_k \langle\langle \alpha_k^\dagger \alpha_k ; \mathscr{D}_\nu \rangle\rangle_z = 0 \ . \tag{3.14}$$

The conservation of total momentum leads to

$$\langle[\mathscr{P}_\mu, \mathscr{D}_\nu]\rangle = -ieN\delta_{\mu\nu}$$

or

$$\sum_k k_\mu \langle\langle \alpha_k^\dagger \alpha_k ; \mathscr{D}_\nu \rangle\rangle_z + \sum_q q_\mu \langle\langle \beta_q^\dagger \beta_q ; \mathscr{D}_\nu \rangle\rangle_z = \frac{ieN}{z} \delta_{\mu\nu}. \tag{3.15}$$

As Michel and Van Leeuwen[17]) showed, the correlation functions $\langle\langle \alpha_k^\dagger \alpha_k ; \mathscr{D}_\nu \rangle\rangle_z$ and $\langle\langle \beta_q^\dagger \beta_q ; \mathscr{D}_\nu \rangle\rangle_z$ may be interpreted as the linear deviation from the polaron equilibrium distribution, n_k, and the linear deviation from the phonon equilibrium distribution, ν_q, respectively. Equation (3.15) expresses that in the limit $z \to +i0$, either the polaron current or the phonon current or both must tend to infinity. To avoid such a physically uninteresting situation we make use of the fact that the polaron density is

vanishingly small with respect to the phonon density. The consequence is then that the polarons in the system cannot change the distribution of the infinite number of phonons. Because the phonon distribution is in equilibrium at $t = -\infty$, it will remain so and ν_q is always equal to

$$\nu_q = (e^{\beta\omega} - 1)^{-1}. \tag{3.16}$$

In order to evaluate the polaron current correlation function, $\langle\langle J_\mu; \mathscr{D}_\nu \rangle\rangle_z$, we use the well-known series expansion of the time evolution operator:

$$e^{i\mathscr{H}t} = \{1 + \sum_{n=1}^{\infty} i^n \int_0^t dt_n \dots \int_0^{t_2} dt_1 V_{t_1} V_{t_2} \dots V_{t_n}\} e^{iH_0 t} \tag{3.17}$$

and

$$e^{-i\mathscr{H}t} = e^{-iH_0 t}\{1 + \sum_{n=1}^{\infty} (-i)^n \int_0^t dt_n \dots \int_0^{t_2} dt_1 V_{t_n} \dots V_{t_2} V_{t_1}\} \tag{3.18}$$

where

$$V_{t_j} = e^{iH_0 t_j} V e^{-iH_0 t_j} \quad \text{and} \quad V = V_1 + V_2.$$

Inserting (3.17) and (3.18) into (3.10) yields

$$\langle\langle J_\mu; \mathscr{D}_\nu \rangle\rangle_z = i \int_0^\infty e^{izt}\, dt \cdot \langle [J_{\mu,t} + i \int_0^t dt_1 [V_{t_1}, J_{\mu t}] + \\ + \dots + i^n \int_0^t dt_n \dots \int_0^{t_2} dt [V_{t_1}, [V_{t_2}, \dots [V_{t_n}, J_{\mu,t}] \dots]]\, \mathscr{D}_\nu] \rangle \tag{3.19}$$

We rewrite this expression by using the convolution integral theorem. Then (3.19) becomes

$$\langle\langle J_\mu; \mathscr{D}_\nu \rangle\rangle_z = \sum_{n=1}^{\infty} i^n \int_0^\infty e^{izt_1}\, dt_1 \dots \int_0^\infty e^{izt_n}\, dt_n$$

$$\langle [e^{iH_0 t_1} [V, e^{iH_0 t_2} [V, \dots e^{iH_0 t_{n-1}} [V, e^{iH_0 t_n} J_\mu e^{-iH_0 t_n}] e^{-iH_0 t_{n-1}} \dots] e^{iH_0 t_2}] e^{-iH_0 t_1}, \mathscr{D}_\nu] \rangle \tag{3.20}$$

This may be expressed in the more condensed form:

$$\langle\langle \boldsymbol{J}; \mathscr{D} \rangle\rangle_z = \sum_{m=0}^{\infty} \langle [\boldsymbol{J}_m(z), \mathscr{D}] \rangle, \tag{3.20a}$$

where $\boldsymbol{J}_m(z)$ is defined by

$$\boldsymbol{J}_m(z) = i \int_0^\infty e^{izt}\, dt [V, \boldsymbol{J}_{m-1}(z)]_t \tag{3.21}$$

and $[A, B]_t$ stands for $e^{iH_0 t} [A, B] e^{-iH_0 t}$ while $\boldsymbol{J}_0(z)$ is given by

$$\boldsymbol{J}_0(z) = i \int_0^\infty e^{izt}\, dt (\boldsymbol{J})_t = -\frac{1}{z} \boldsymbol{J}. \tag{3.22}$$

Notice that the series expansion (3.20) is valid for any arbitrary operator instead of J. For instance,

$$\langle\langle [V, J_n(z)]; \mathscr{D}\rangle\rangle_z = \sum_{m=n+1}^{\infty} \langle [J_m(z), \mathscr{D}]\rangle.$$

This equation combined with (3.20) yields:

$$\langle\langle J; \mathscr{D}\rangle\rangle_z = \sum_{m=0}^{n} \langle [J_m(z), \mathscr{D}]\rangle + \langle\langle [V, J_n(z)]; \mathscr{D}\rangle\rangle_z. \tag{3.23}$$

In the case $n = 0$ expression (3.23) can easily be recognized as the fourier transform of the equation of motion. Then

$$\langle\langle J; \mathscr{D}\rangle\rangle_z = -\frac{1}{z} \langle [J, \mathscr{D}]\rangle - \frac{1}{z} \langle\langle [\mathscr{H}, J]; \mathscr{D}\rangle\rangle_z. \tag{3.24}$$

As it stands, (3.24) is not useful, because we are not able to express the correlation function $\langle\langle [\mathscr{H}, J]; \mathscr{D}\rangle\rangle_z$ as a known function of $\langle\langle J; \mathscr{D}\rangle\rangle_z$. This, however, is approximately possible for correlation functions of the type $\langle\langle [\mathscr{H}, \alpha_k^\dagger \alpha_k]; \mathscr{D}\rangle\rangle_z$. For that reason we shall evaluate the series expansion (3.20) directly, instead of looking for an integral equation of $\langle\langle J; \mathscr{D}\rangle\rangle_z$.

As it stands (3.20) is not a convergent series expansion. We will meet terms that are of order z^{-n} and thus highly singular in the limit $z \to i0$. Besides this aspect, the interaction part of the Hamiltonian, V, possesses the delta singularity property, introduced by Van Hove[16]). Due to this fact only a certain class of contributions are of importance in the limit of an infinite volume. In the present situation this means that some terms will possess an extra summation over the internal states, which, in the limit of an infinite volume, gives an extra factor of order of the volume. Taking these features into account we first perform a partial summation in expression (3.20) in order to assure a rapid convergence of the series expansion in the polaron-phonon interaction.

We separate from (3.20) the terms

$$\langle\langle J; \mathscr{D}\rangle\rangle_z^{(0)} = \sum_{n=0}^{\infty} \langle [J_{2n,d}^{(0)}(z), \mathscr{D}]\rangle_0 \tag{3.25}$$

$$J_{2n+2,d}^{(0)}(z) = i^2 \int_0^\infty e^{izt_1}\, dt_1 \int_0^\infty e^{izt_2}\, dt_2 [V, [V, J_{2n,d}^{(0)}(z)]_{t_2}]_{t,d} \tag{3.26}$$

The grand canonical ensemble average has to be taken over systems, described by the unperturbed Hamiltonian H_0.

The subscript d means the diagonal part of the corresponding operator in the α, β representation. We have to evaluate (3.25) explicitly. One may call (3.25) the lowest order contribution to the conductivity, because, for small coupling constants α, it is of order α^{-1}.

For $n = 1$ $\boldsymbol{J}^{(0)}_{2,d}$ reads

$$\boldsymbol{J}^{(0)}_{2,d}(z) = i^2 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \{V\cdot\{V\boldsymbol{J}_0(z) - \boldsymbol{J}_0(z)\,V\}_{t_2} - \\ - \{V\cdot\boldsymbol{J}_0(z) - \boldsymbol{J}_0(z)\cdot V\}_{t_2}\cdot V\}_{t_1,d},$$

Or

$$\boldsymbol{J}^{(0)}_{2,d}(z) = i^2 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \{(V\cdot\{V\}_d\boldsymbol{J}_0(z)\}_{t_2} + \\ + \{\boldsymbol{J}_0(z)\cdot(V\}_{t_2}\cdot V)_d\}_{t_1} - i^2 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \{(V\cdot\{\boldsymbol{J}_0(z)\cdot V\}_{t_2})_d\cdot + \\ + (\{V\cdot\boldsymbol{J}_0(z)\}_{t_2}\cdot V)_d\}_{t_1} = \boldsymbol{A}^{(0)}_{2,d}(z) - \boldsymbol{B}^{(0)}_{2,d}(z). \tag{3.27}$$

Evaluating $\boldsymbol{A}^{(0)}_{2,d}(z)$ explicitly yields:

$$\boldsymbol{A}^{(0)}_{2,d}(z) = \\ = i^2 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \{(V\{V\}_d\boldsymbol{J}_0(z)\}_{t2} + \{\boldsymbol{J}_0(z)\cdot(V\}_{t_2}\cdot V)_d\}_{t_1} = \\ = \frac{1}{z^2}\,\boldsymbol{J}\cdot\sum_{\boldsymbol{k}\boldsymbol{q}_1\boldsymbol{q}_2}\alpha^\dagger_{\boldsymbol{k}}\alpha_{\boldsymbol{k}}\cdot\Big\{|V_{\boldsymbol{q}}(\boldsymbol{k}+\boldsymbol{q})|^2\,\beta^\dagger_{\boldsymbol{q}}\beta_{\boldsymbol{q}}\cdot\Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega; z) + \\ + |V_{-\boldsymbol{q}}(\boldsymbol{k})|^2\,\beta_{-\boldsymbol{q}}\,\beta^\dagger_{-\boldsymbol{q}}\cdot\Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega; z) + \\ + \frac{q_1^2 q_2^2}{(2m)^2}\cdot|F_{-\boldsymbol{q}_1}|^2\cdot|F_{-\boldsymbol{q}_2}|^2\cdot(\beta^\dagger_{-\boldsymbol{q}_1}\beta_{-\boldsymbol{q}_1} + \beta_{-\boldsymbol{q}_1}\beta^\dagger_{-\boldsymbol{q}_1} + \beta^\dagger_{-\boldsymbol{q}_2}\,\beta_{-\boldsymbol{q}_2} + \beta_{-\boldsymbol{q}_2}\beta^\dagger_{-\boldsymbol{q}_2})\times \\ \times\,\Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}_2} - 2\omega; z) + \\ + \frac{2q_1^2 q_2^2}{(2m)^2}\cdot(|F_{-\boldsymbol{q}_1}|^2\cdot|F_{\boldsymbol{q}_2}|^2\cdot\beta^\dagger_{\boldsymbol{q}_1}\beta_{\boldsymbol{q}_1} + |F_{-\boldsymbol{q}_2}|^2\cdot|F_{\boldsymbol{q}_1}|^2\cdot\beta^\dagger_{\boldsymbol{q}_2}\beta_{\boldsymbol{q}_2})\times \\ \times\,\Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}_2}; z)\Big\} = \\ = \frac{1}{z^2}\,\boldsymbol{J}\sum_{\boldsymbol{k}}\alpha^\dagger_{\boldsymbol{k}}\alpha_{\boldsymbol{k}}\cdot\{\sum_{\boldsymbol{q}}\tilde{W}^{(1)}_{\boldsymbol{q}}(\boldsymbol{k};\boldsymbol{k}+\boldsymbol{q}) + \sum_{\boldsymbol{q}_1\boldsymbol{q}_2}\tilde{W}^{(2)}_{\boldsymbol{q}_1,\boldsymbol{q}_2}(\boldsymbol{k};\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}_2)\}. \tag{3.28}$$

We introduced the definition:

$$\Delta(x; z) = \frac{1}{x - z} - \frac{1}{x + z}.$$

This expression reduces to $2\pi i\delta(x)$ in the limit $z = +i0$.

Since we are only interested in the low-temperature and low electron density behaviour we neglect contributions of order $\nu_{\boldsymbol{q}}^2$ with respect to $\nu_{\boldsymbol{q}}$ and $n_{\boldsymbol{k}}$ with respect to $\nu_{\boldsymbol{q}}$.

The evaluation of $\langle[\boldsymbol{A}^{(0)}_{2,d}(z), \mathscr{D}]\rangle_0$ will give no difficulties at this stage.

Keeping the low electron density approximation there is only one possible way to decouple the correlation functions:

$$\langle[\alpha_k^\dagger \alpha_k \beta_q^\dagger \beta_q, \mathscr{D}]\rangle_0 \quad \text{and} \quad \langle[\alpha_k^\dagger \alpha_k \beta_q \beta_q^\dagger, \mathscr{D}]\rangle_0.$$

That is replacing these expressions by

$$v_q \langle[\alpha_k^\dagger \alpha_k, \mathscr{D}]\rangle_0 \quad \text{and} \quad (1 + v_q)\langle[\alpha_k^\dagger \alpha_k, \mathscr{D}]\rangle_0$$

respectively.

Then the operator expressions

$$\tilde{W}_q^{(1)}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}) \quad \text{and} \quad \tilde{W}_{q_1 q_2}^{(2)}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}_1 + \boldsymbol{q}_2)$$

go over into the transition probabilities

$$W_q^{(1)}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}) \quad \text{and} \quad W_{q_1 q_2}^{(2)}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}_1 + \boldsymbol{q}_2)$$

where we have substituted $\beta_q^\dagger \beta_q$ by v_q and $\beta_q \beta_q^\dagger$ by $(1 + v_q)$, respectively. Taking this into account we write $\langle[\boldsymbol{A}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0$ as

$$\begin{aligned}\langle[\boldsymbol{A}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0 = \frac{-e}{m'\Omega z^2} \sum_{kq} \boldsymbol{k} \cdot \{W_q^{(1)}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}) + \\ + \sum_{q_1} W_{qq_1}^{(2)}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q} + \boldsymbol{q}_1)\}\langle[\alpha_k^\dagger \alpha_k, \mathscr{D}]\rangle_0 = \\ = \frac{-e}{m'\Omega z^2} \sum_{kq} W_q(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}) \langle[\alpha_k^\dagger \alpha_k, \mathscr{D}]\rangle_0 \end{aligned} \tag{3.29}$$

We can use the same argument for the terms accounting for $\langle[\boldsymbol{B}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0$. The result is

$$\langle[\boldsymbol{B}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0 = \frac{-e}{m'\Omega z^2} \sum_{kq} W_q(\boldsymbol{k} + \boldsymbol{q}; \boldsymbol{k}) \langle[\alpha_{k+q}^\dagger \alpha_{k+q}, \mathscr{D}]\rangle_0. \tag{3.30}$$

Since $\langle[\alpha_k^\dagger \alpha_k, \mathscr{D}]\rangle_0$ is given by $-ie\nabla_k n_k$, the tensor $\langle[\boldsymbol{J}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0$ reads finally

$$\langle[\boldsymbol{J}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0 = \frac{ie^2}{m'\Omega z^2} \sum_{kk_1} \boldsymbol{k}\{W_{k_1-k}(\boldsymbol{k}; \boldsymbol{k}_1) \nabla_k n_k - W_{k-k_1}(\boldsymbol{k}_1; \boldsymbol{k}) \nabla_{k_1} n_{k_1}\} \tag{3.31}$$

The detailed calculation of $\langle[\boldsymbol{J}_{2,d}^{(0)}(z), \mathscr{D}]\rangle_0$ together with

$$\langle[\boldsymbol{J}_0(z), \mathscr{D}]\rangle_0 = \frac{-ie^2}{m'\Omega z} \sum_k \boldsymbol{k} \nabla_k n_k = \frac{ie^2}{m'z} \bar{n} \cdot \boldsymbol{I} \cdot \tag{3.32}$$

gives us in fact all the information we need to evaluate the lowest order contribution to the conductivity.

Looking at the equations (3.32) and (3.31) shows that simply replacing $\nabla_k n_k$ in $\langle[\boldsymbol{J}_0(z), \mathscr{D}]\rangle_0$ by

$$\frac{-1}{z} \sum_{k_1} \{W_{k_1-k}(\boldsymbol{k}; \boldsymbol{k}_1) \nabla_k n_k - W_{k-k_1}(\boldsymbol{k}_1; \boldsymbol{k}) \nabla_{k_1} n_{k_1}\},$$

will give us $\langle[J^{(0)}_{2,d}(z), \mathscr{D}]\rangle_0$. It suggests that this rule will hold in general. To show that this is indeed the case we consider the operator expression

$$J^{(0)}_{2n+2,d}(z) = i^2 \int_0^\infty e^{izt_1}\, dt_1 \int_0^\infty e^{izt_2}\, dt_2 [V,[V,\, J^{(0)}_{2n,d}(z)]_{t_2}]_{t_1,d} \tag{3.26}$$

and remember that $J^{(0)}_{2n,d}(z)$ contains a product of phonon operators of the type

$$(\beta^\dagger_{q_1}\beta_{q_2})_d\,(\beta_{q_3}\beta^\dagger_{q_4})_d \cdots (\beta^\dagger_{q_{2n-1}}\beta_{q_{2n}})_d$$

where all interchanges of the phonon operators between the brackets are allowed. Then $J^{(0)}_{2n+2,d}(z)$ must contain at least two phonon operators more, for instance in the form:

$$(\beta_{q_{2n+1}}(\beta^\dagger_{q_1}\beta_{q_2})_d\,(\beta_{q_3}\beta^\dagger_{q_4})_d \cdots (\beta^\dagger_{q_{2n-1}}\beta_{q_{2n}})_d\,\beta^\dagger_{q_{2n+2}})_d \tag{3.33}$$

Now in order to calculate $\langle[J^{(0)}_{2n+2,d}(z), \mathscr{D}]\rangle_0$, we have to contract (3.33) and find that the contraction:

$$(1 + \nu_{q_{2n+1}})\,\delta_{q_{2n+1},q_{2n+2}}\nu_{q_1}\delta_{q_1,q_2} \cdots \nu_{q_{2n-1}}\cdot\delta_{q_{2n-1},q_{2n}} \tag{3.34}$$

yields one summation over the wave vectors q more than all other possible contractions. Thus in the limit of an infinite volume, only the contributions to $\langle[J^{(0)}_{2n+2,d}(z), \mathscr{D}]\rangle_0$ might be considered that contain contractions like (3.34). But the contractions of the type of (3.34) can be obtained by contracting the phonon operators before commuting $J^{(0)}_{2n,d}(z)$ with the interaction V. Hence, in the recursion formula (3.26), we may replace the operator $\alpha^\dagger_k\alpha_k$, appearing in $J^{(0)}_{2n,d}(z)$, by $-1/z\,\sum_{k_1}\{W_{k_1-k}(k;\, k_1)\,\alpha^\dagger_k\alpha_k - W_{k-k_1}(k_1;\, k_1)\,\alpha^\dagger_{k_1}\alpha_{k_1}\}$ in order to find $J^{(0)}_{2n+2,d}(z)$. This is just the simple rule given above.

As an illustration we evaluate the contributions of $\langle[J^{(0)}_{4,d}(z), \mathscr{D}]\rangle_0$.

Using (3.26) one finds:

$$J^{(0)}_{4,d}(z) = i^2 \int_0^\infty e^{izt_1}\, dt_1 \int_0^\infty e^{izt_2}\, dt_2 [V,\, [V,\, J^{(0)}_{2,d}(z)]_{t_2}]_{t_1} \tag{3.35}$$

Or

$$J^{(0)}_{4,d}(z) = i^2 \int_0^\infty e^{izt_1}\, dt_1 \int_0^\infty e^{izt_2}\, dt_2 \{J^{(0)}_{2,d}(z)\cdot(V\cdot)_{t_2}\cdot V)_d -$$

$$- (V\cdot\{J^{(0)}_{2,d}(z)\cdot V\}_{t_2})_d - (\{V\cdot J^{(0)}_{2,d}(z)\}_{t_2}\cdot V)_d + (V\cdot\{V)_d J^{(0)}_{2,d}(z)\}_{t_2}\}_{t_1}$$

where

$$J^{(0)}_{2,d}(z) = A^{(0)}_{2,d}(z) - B^{(0)}_{2,d}(z).$$

To prevent lengthly formulas we combine a contribution from the first

and last term in expression (3.35) and find:

$$i^2 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \langle[\{\{A^{(0)}_{2,d}(z)\cdot(V\}_{t_2}\cdot V)_d + (V\cdot\{V)_d\cdot A^{(0)}_{2,d}(z)\}_{t_2}\}_{t_1}, \mathscr{D}]\rangle_0 =$$

$$= +\frac{1}{z^3}\,\frac{e}{m'\Omega}\sum_{\boldsymbol{k}\boldsymbol{q}_1\boldsymbol{q}_2} \boldsymbol{k}\langle[\tilde{W}_{q_1}(\boldsymbol{k};\boldsymbol{k}+\boldsymbol{q}_1)\cdot\tilde{W}_{q_2}(\boldsymbol{k};\boldsymbol{k}+\boldsymbol{q}_2)\cdot\alpha^\dagger_k\alpha_k, \mathscr{D}]\rangle_0.$$

Besides the contraction

$$\frac{1}{z_3}\,\frac{e}{m'\Omega}\sum_{\boldsymbol{k}\boldsymbol{q}_1\boldsymbol{q}_2} \boldsymbol{k}W_{q_1}(\boldsymbol{k};\boldsymbol{k}+\boldsymbol{q}_1)\,W_{q2}(\boldsymbol{k};\boldsymbol{k}+\boldsymbol{q}_2)\,\langle[\alpha^\dagger_k\alpha_k, \mathscr{D}]\rangle_0$$

there is the possibility to contract the operator product $\beta^\dagger_{q_1}\beta_{q_1}\cdot\beta^\dagger_{q_2}\beta_{q_2}$ as $\beta^\dagger_{q_1}\beta_{q_1}\ \beta^\dagger_{q_2}\beta_{q_2}$, leading to $\nu_{q_1}\cdot(1+\nu_{q_1})$.

In the limit of an infinite volume, the last contribution will be vanishly small with respect to the first one because it contains one summation over the wave vectors $\boldsymbol{q}$ less.

Calculating the other terms in (3.35) we finally arrive at

$$\langle[J^{(0)}_{4,d}(z), \mathscr{D}]\rangle_0 = \frac{-ie^2}{m'\Omega z^3}\sum_{\boldsymbol{k}\boldsymbol{k}_1\boldsymbol{k}_2} \boldsymbol{k}\cdot$$

$$\cdot\{W_{k_1-k}(\boldsymbol{k};\boldsymbol{k}_1)\cdot(W_{k_2-k}(\boldsymbol{k};\boldsymbol{k}_2)\,\nabla_k n_k - W_{k-k_2}(\boldsymbol{k}_2;\boldsymbol{k})\,\nabla_{k_2} n_{k_2}) -$$

$$- W_{k-k_1}(\boldsymbol{k}_1;\boldsymbol{k})\cdot(W_{k_2-k_1}(\boldsymbol{k}_1;\boldsymbol{k}_2)\,\nabla_{k_1} n_{k_1} - W_{k_1-k_2}(\boldsymbol{k}_2;\boldsymbol{k}_1)\,\nabla_{k_2} n_{k_2})\}.$$

Knowing the effect of the recursion formula (3.26) we can represent the different terms in the series expansion (3.25) by diagrams. The contribution of (3.32) is represented by fig. 1.

We read the diagram from the left to the right. The solid lines are polaron creation and annihilation lines:

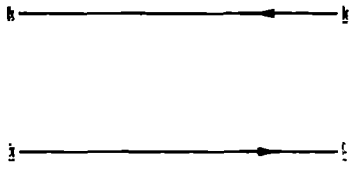

Fig. 1
The contribution of $\langle[\mathscr{J}_0(z), \mathscr{D}]\rangle_0$

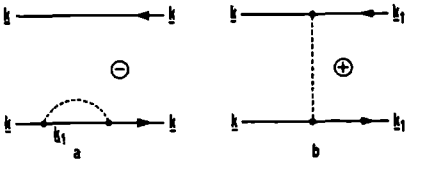

Fig. 2.
The contribution of $W_{k_1-k}(\boldsymbol{k};\boldsymbol{k}_1)\,\nabla_k n_k$ The contribution of $W_{k-k_1}(\boldsymbol{k}_1;\boldsymbol{k})\,\nabla_{k_1} n_{k_1}$

The contributions of (3.31) is then represented by fig. 2. The dotted line represents the connection of the transition probability $W_{k_1-k}(\boldsymbol{k};\boldsymbol{k}_1)$

We note that connecting the dotted line to different polaron lines gives a plus sign, otherwise we have a minus sign. The contributions of $\langle[J^{(0)}_{4,d}(z), \mathscr{D}]\rangle_0$ consist of all possible combinations of two diagrams of the

type of fig. 2*a* or fig. 2*b*. The correlation effect contained in fig. 2*b* will bring us into trouble if we want to sum

$$\langle\langle \boldsymbol{J}; \mathscr{D}\rangle\rangle_z^{(0)} = \sum_{n=0}^{\infty} \langle [\boldsymbol{J}_{2n}^{(0)}(z), \mathscr{D}]\rangle_0 \tag{3.25}$$

explicitly. This is just the source of the difficulties one encounters if one wants to solve the Boltzmann equation. Happily in the case of polarons the temperature must be much smaller than the Debye temperature in order to make the polaron concept meaningful. Then only diagrams of the type of fig. 2*a* are of importance, as we shall show.

Besides the contributions of (3.25) we have to consider the lowest order contributions of (3.11). Thus

$$\langle\langle J'_\mu; \mathscr{D}_\nu\rangle\rangle_z^{(0)} = \frac{e}{m\Omega} \sum_{\boldsymbol{q}} \boldsymbol{q}_\mu \langle \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}+\boldsymbol{q}} (F_{-\boldsymbol{q}}^* \beta_{-\boldsymbol{q}} - F_{\boldsymbol{q}} \beta_{\boldsymbol{q}}^\dagger); \mathscr{D}_\nu\rangle\rangle_z^{(0)}. \tag{3.36}$$

Or

$$\langle\langle \boldsymbol{J}'; \mathscr{D}\rangle\rangle_z^{(0)} = \sum_{n-0}^{\infty} \langle [\boldsymbol{J}'^{(0)}_{2n}(z), \mathscr{D}]\rangle_0 \tag{3.37}$$

where $\boldsymbol{J}'^{(0)}_{2n}(z)$ is defined by the recursion relations (3.26) but $\boldsymbol{J}'_0(z)$ is given by

$$\boldsymbol{J}'^{(0)}_0(z) = i^2 \int_0^\infty \mathrm{e}^{izt_1}\, \mathrm{d}t_1 \int_0^\infty \mathrm{e}^{izt_2}\, \mathrm{d}t_2 [V, \boldsymbol{J}'_{t_2}]_{t_1, d} =$$

$$= \frac{e}{m\Omega z} \sum_{\boldsymbol{k}\boldsymbol{q}} \boldsymbol{q} \left\{ (\alpha_{\boldsymbol{k}+\boldsymbol{q}}^\dagger \alpha_{\boldsymbol{k}+\boldsymbol{q}} - \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}}) \cdot \left(\nu_{-\boldsymbol{q}} \frac{F_{-\boldsymbol{q}}^* V_{-\boldsymbol{q}}(\boldsymbol{k})}{(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega + z)} - \right.\right.$$

$$(1 + \nu_{\boldsymbol{q}}) \frac{F_{\boldsymbol{q}} V_{\boldsymbol{q}}^*(\boldsymbol{k}+\boldsymbol{q})}{(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega + z)} \Big) - \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}} \Big(\frac{F_{-\boldsymbol{q}}^* V_{-\boldsymbol{q}}(k)}{E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega + z} +$$

$$\left. + \frac{F_{\boldsymbol{q}} V_{\boldsymbol{q}}^*(\boldsymbol{k}+\boldsymbol{q})}{(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega + z)} \Big) \right\}. \tag{3.38}$$

Together with $\langle\langle \boldsymbol{J}; \mathscr{D}_\nu\rangle\rangle_z^{(0)}$ expression (3.37) yields for the electrical conductivity:

$$\sigma^{(0)}(z) = \frac{-e}{m\Omega} \sum_{\boldsymbol{k}} \left(\boldsymbol{k} - \sum_{\boldsymbol{q}} \boldsymbol{q} |F_{\boldsymbol{q}}|^2 + \sum_{\boldsymbol{q}} \boldsymbol{q} \chi_z^{(0)}(\boldsymbol{k}; \boldsymbol{q}) \right) \cdot \langle\langle \alpha_{\boldsymbol{k}}^\dagger \alpha_{\boldsymbol{k}}; \mathscr{D}\rangle\rangle_z^{(0)} \tag{3.39}$$

where

$$\sum_{\boldsymbol{q}} \boldsymbol{q} \chi_z^{(0)}(\boldsymbol{k}; \boldsymbol{q}) = \sum_{\boldsymbol{q}} \boldsymbol{q} \left\{ (1 + \nu_{-\boldsymbol{q}}) \cdot \left(\frac{F_{-\boldsymbol{q}}^* V_{-\boldsymbol{q}}(\boldsymbol{k})}{E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega + z} + \right.\right.$$

$$\left. + \frac{F_{-\boldsymbol{q}} V_{-\boldsymbol{q}}^*(\boldsymbol{k})}{E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega - z} \right) -$$

$$\left. - \nu_{\boldsymbol{q}} \cdot \left(\frac{F_{\boldsymbol{q}} V_{\boldsymbol{q}}^*(\boldsymbol{k}+\boldsymbol{q})}{E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega + z} + \frac{F_{\boldsymbol{q}}^* V_{\boldsymbol{q}}(\boldsymbol{k}+\boldsymbol{q})}{E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega - z} \right) \right\}. \tag{3.40}$$

Because $\langle\langle \alpha_k^\dagger \alpha_k ; \mathcal{D} \rangle\rangle_z^{(0)}$ is proportional to $n_k \equiv \exp.\{-\beta E_k\}$ the main contribution in (3.39) arises from small values of k, if the temperature is low enough.

This means that we may linearize (3.40) in k and interprete $\sum_q q\chi_z^{(0)}(k; q)$ as a contribution to the effective mass of the polaron. Thus in the case of low temperatures (3.39) may be approximated by

$$\sigma^{(0)}(z) = \frac{-e}{m^*(z)\cdot\Omega} \sum_k k \langle\langle \alpha_k^\dagger \alpha_k ; \mathcal{D} \rangle\rangle_z^{(0)} =$$

$$= \frac{m}{m^*(z)} \langle\langle J ; \mathcal{D} \rangle\rangle_z^{(0)} = \frac{m}{m^*(z)} \sum_{n=0}^{\infty} \langle [J_{2n,d}^{(0)}(z), \mathcal{D}] \rangle_0 \qquad (3.41)$$

where the effective mass, $m^*(z)$ depends on the temperature, the frequency of the applied field and the coupling constant.

The summation of the terms in (3.41) can be done following the procedure of fig. 3:

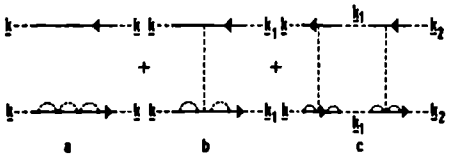

Fig. 3. Representation of the first three terms in $\sigma_{\mu\nu}^{(0)}(z)$.

The first term (fig. 3*a*) then reads:

$$\sigma_{(z)}^{(00)} = \frac{-ie^2}{m^*(z)\,\Omega z} \sum_k k \cdot \nabla_k n_k \cdot \left\{ 1 - \frac{\Gamma_k(z)}{z} + \frac{\Gamma_k^2(z)}{z^2} - \ldots \right\} =$$

$$= \frac{-ie^2}{m^*(z)\,\Omega} \sum_k \frac{k \cdot \nabla_k n_k}{z + \Gamma_k(z)}, \qquad (3.42)$$

where we defined

$$\Gamma_k(z) = \frac{i}{\tau_k(z)} = \sum_{k_1} W_{k_1 - k}(k; k_1) \qquad (3.43)$$

Thus (3.41) goes over into

$$\sigma_{\mu\nu}^{(0)}(z) = \frac{-ie^2}{m^*(z)\,\Omega} \sum_k k_\mu \left\{ \frac{1}{(z + \Gamma_k(z))} \frac{\partial}{\partial k_\nu} n_k + \right.$$

$$+ \sum_{k_1} \frac{1}{z + \Gamma_k(z)} W_{k-k_1}(k_1; k) \frac{1}{z + \Gamma_{k_1}(z)} \frac{\partial}{\partial k_{1\nu}} n_{k_1} +$$

$$+ \sum_{k_1 k_2} \frac{1}{z + \Gamma_k(z)} W_{k-k_1}(k_1; k) \frac{1}{z + \Gamma_{k_1}(z)} \cdot$$

$$\left. \cdot W_{k_1 - k_2}(k_2; k_1) \frac{1}{z + \Gamma_{k_2}(z)} \frac{\partial}{\partial k_{2\nu}} n_{k_2} + \ldots \right\} = \sigma_{\mu\nu}^{(00)}(z) + \sigma_{\mu\nu}^{(01)}(z). \qquad (3.44)$$

The convergence of the series expansion (3.44) is not a priori obvious. It depends strongly on the specific behaviour of the functions $W_{k-k_1}(k_1; k)$ and Γ_k. In the case of polarons, however, a rapid convergence can be obtained in the low temperature region. To show this, it is necessary to perform the integrations in (3.44) explicitly.

Because we devote the second part of this investigation to numerical results we delay this question to the second part where we shall calculate the correction term, $\sigma_{\mu\nu}^{(01)}(z)$, and

$$\sigma_{\mu\nu}^{(00)}(z) = \frac{-ie^2}{m^*(z)\,\Omega} \sum_k k_\mu \frac{1}{z + \Gamma_k(z)} \frac{\partial}{\partial k_\nu} n_k \tag{3.45}$$

explicitly.

Only under the condition that $\sigma_{\mu\nu}^{(01)}(z)$ may be neglected it is possible to calculate the first order contribution to the conductivity, $\sigma_{\mu\nu}^{(10)}(z)$. This first order contribution, $\sigma_{\mu\nu}^{(10)}(z)$, results by considering the terms in the series expansion (3.20), that are of order α^0 in the weak coupling limit.

As will be clear from the foregoing, the neglection of $\sigma_{\mu\nu}^{(01)}(z)$ is identical with neglecting contributions of the type of fig. 2*b*. This kind of contributions complicates the already rather involved expressions for $\sigma_{\mu\nu}^{(10)}(z)$ extremely and makes the evaluation of $\sigma_{\mu\nu}^{(10)}(z)$ rather hopeless. But the explicit evaluation of $\sigma_{\mu\nu}^{(10)}(z)$ is necessary in order to conclude if the present treatment is really an intermediate coupling theory. As we shall see in the second part we solve this problem under the condition that $\beta\omega \gg 1$.

4. *Conclusion.* The effects of the electron-phonon interaction on the system are twofold. In the first place we have to take into account the cloud effects of the interaction. We did this by using a canonical transformation. As a result we found that the effective mass of a drifting polaron in general depends on the frequency of the applied field, the temperature and the coupling constant. Further it turned out that the concept of effective mass is only useful in the low temperature region, when $\beta\omega \gg 1$. This is in agreement with the results obtained by Langreth and Kadanoff[18]).

In the second place we have to deal with the dissipative effects of the interaction. Using Kubo's formula we develop a method to calculate the lowest order contribution to the conductivity and the several correction terms, $\sigma_{\mu\nu}^{(01)}(z)$ and $\sigma_{\mu\nu}^{(10)}$, directly, passing by the special assumptions needed in the Boltzmann equation approach.

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CHAPTER II

APPLICATION OF THE KUBO FORMULA TO THE MODIFIED L.L.P. POLARON MODEL

Synopsis

An intermediate coupling calculation of the drift mobility of polarons has been carried out, using Kubo's formula for the linear response coefficient, together with a modified L L P polaron model At low temperatures the drift mobility turns out to be

$$\mu = \frac{e}{m^* 2\alpha\omega\bar{\nu}(1 + 0\,06\alpha)}$$

where α is the electron-phonon coupling constant and m^* the effective mass of the polaron, given by

$$m^* = m \frac{(1 + \alpha/6)}{(1 + 0\,2\alpha\bar{\nu})}$$

A comparison, made with other intermediate coupling theories, shows that the obtained mobility expression agrees closely with Osaka's formula in the coupling range $\alpha \leqslant 3$ The calculated correction term being

$$\mu \frac{0\,06\,\alpha}{(1 + 0\,06\,\alpha)},$$

ustifies the validity of th e obtained results in the intermediate coupling range

1. *Introduction.* In a previous paper, hereafter to be referred to as I, we developed a method to evaluate the drift mobility of polarons avoiding the special assumptions needed in a Boltzmann equation approach. Starting from Kubo's formula for the frequency dependent electrical conductivity and using the Frohlich Hamiltonian to describe the electron-phonon system, we showed that the low-temperature electrical conductivity could be written as a power series expansion in the polaron-phonon coupling. The essential assumption, entering in the treatment, was the requirement of convergence of the perturbation series in the intermediate coupling region.

It is the aim of this paper to calculate explicitly the first two terms of the series expansion in order to show that this assumption is reasonable.

The properties of polarons are taken into account by describing the polaron-phonon system with the Hamiltonian.

$$\mathscr{H} = H_0 + V,\ V = V_1 + V_2, \tag{1.1}$$

where

$$H_0 = \sum_{\boldsymbol{k}} E_{\boldsymbol{k}} \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}} + \sum_{\boldsymbol{q}} \omega \beta_{\boldsymbol{q}}^{\dagger} \beta_{\boldsymbol{q}} \tag{1.2}$$

$$V_1 = \sum_{\boldsymbol{kq}} \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}+\boldsymbol{q}} \{V_{-\boldsymbol{q}}^{*}(\boldsymbol{k}) \beta_{-\boldsymbol{q}} + V_{\boldsymbol{q}}(\boldsymbol{k}+\boldsymbol{q})\, \beta_{\boldsymbol{q}}^{\dagger}\}. \tag{1.3}$$

$$V_2 = \sum_{\boldsymbol{kq_1q_2}} \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}_2} \frac{\boldsymbol{q}_1 \boldsymbol{q}_2}{2m} \{F_{-\boldsymbol{q}_1}^{*} F_{-\boldsymbol{q}_2}^{*} \beta_{-\boldsymbol{q}_1} \beta_{-\boldsymbol{q}_2} + \text{c.c.}\} -$$

$$- \sum_{\substack{\boldsymbol{kq_1q_2} \\ \boldsymbol{q}_1 \neq -\boldsymbol{q}_2}} \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}_2} \frac{\boldsymbol{q}_1 \boldsymbol{q}_2}{2m} 2F_{\boldsymbol{q}_1} F_{-\boldsymbol{q}_2}^{*} \beta_{\boldsymbol{q}_1}^{\dagger} \beta_{-\boldsymbol{q}_2} \tag{1.4}$$

and

$$E_{\boldsymbol{k}} = \frac{1}{2m} (\boldsymbol{k} - \sum_{\boldsymbol{q}} \boldsymbol{q}\, |F_{\boldsymbol{q}}|^2) + \sum_{\boldsymbol{q}} (\lambda_{\boldsymbol{q}} F_{\boldsymbol{q}} + \lambda_{\boldsymbol{q}}^{*} F_{\boldsymbol{q}}^{*}) + \sum_{\boldsymbol{q}} |F_{\boldsymbol{q}}|^2 \left(\omega + \frac{q^2}{2m}\right) \tag{1.5}$$

$$\omega_{\boldsymbol{q}} = \omega + \frac{q^2}{m} |F_{\boldsymbol{q}}|^2 \cdot N. \tag{1.6}$$

$$V_{-\boldsymbol{q}}^{*}(\boldsymbol{k}) = \lambda_{\boldsymbol{q}} - F_{-\boldsymbol{q}}^{*}(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega) \tag{1.7}$$

$$V_{\boldsymbol{q}}(\boldsymbol{k}+\boldsymbol{q}) = \lambda_{\boldsymbol{q}}^{*} + F_{\boldsymbol{q}}(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega) \tag{1.8}$$

$$\lambda_{\boldsymbol{q}} = \frac{-i\omega}{q} \left(\frac{1}{2m\omega}\right)^{\frac{1}{4}} \left(\frac{4\pi\alpha}{\Omega}\right)^{\frac{1}{2}}. \tag{1.9}$$

The Hamiltonian (1.1) is generated by applying a unitary transformation to the Fröhlich Hamiltonian and neglecting terms of second and higher order in the polaron density. The meaning of the symbols is the same as used in I. The transformed current operator has been given as

$$\mathscr{J} = \boldsymbol{J} + \boldsymbol{J}',$$

where

$$\boldsymbol{J} = \frac{-e}{m\Omega} \sum_{\boldsymbol{k}} (\boldsymbol{k} - \sum_{\boldsymbol{q}} \boldsymbol{q}\, |F_{\boldsymbol{q}}|^2)\, \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}} \tag{1.10}$$

$$\boldsymbol{J}' = \frac{e}{m\Omega} \sum_{\boldsymbol{kq}} \boldsymbol{q} \alpha_{\boldsymbol{k}}^{\dagger} \alpha_{\boldsymbol{k}+\boldsymbol{q}} (F_{-\boldsymbol{q}}^{*} \beta_{-\boldsymbol{q}} - F_{\boldsymbol{q}} \beta_{\boldsymbol{q}}^{\dagger}) \tag{1.11}$$

Using the Kubo formula, the electrical conductivity could be written as

$$\sigma_{\mu\nu}(z) = \sum_{n=0}^{\infty} \langle [\mathscr{J}_{\mu,\, n}(z), \mathscr{D}_{\nu}] \rangle \tag{1.12}$$

with

$$\mathscr{J}_n(z) = i \int_0^{\infty} \mathrm{e}^{izt}\, \mathrm{d}t [V, \mathscr{J}_{n-1}(z)]_t \tag{1.13}$$

while

$$\mathscr{I}_0(z) = i\int_0^\infty e^{izt}\,dt\; e^{iHot}\, J\, e^{-iHot}. \tag{1.14}$$

The lowest order contribution to the conductivity is defined by

$$\sigma_{\mu\nu}^{(0)}(z) = \frac{-e}{m\Omega}\sum_k \{k_\mu - \sum q_\mu |F_q|^2 - \sum_q q_\mu \chi_z^{(0)}(\boldsymbol{k};\boldsymbol{q})\}\langle\langle \alpha_k^\dagger \alpha_k; \mathscr{D}_\nu\rangle\rangle_z^{(0)} \tag{1.15}$$

where $\sum_q \boldsymbol{q}\chi_z^{(0)}(\boldsymbol{k};\boldsymbol{q})$ is given by

$$\sum_q \boldsymbol{q}\chi_z^{(0)}(\boldsymbol{k};\boldsymbol{q}) = \sum_q \boldsymbol{q}\left\{(1+\nu_{-q})\left(\frac{F_{-q}^*(\boldsymbol{k})}{E_k - E_{k+q} - \omega_q + z} + \right.\right.$$
$$+ \frac{F_{-q}V_{-q}^*(\boldsymbol{k})}{E_k - E_{k+q} - \omega_q - z}\Big) - \nu_q\left(\frac{F_q V_q^*(\boldsymbol{k}+\boldsymbol{q})}{E_k - E_{k+q} + \omega_q + z} + \right.$$
$$\left.\left. + \frac{F_q^* V_q(\boldsymbol{k}+\boldsymbol{q})}{E_k - E_{k+q} + \omega_q - z}\right)\right\}. \tag{1.16}$$

The terms between brackets in (1.15) constitute in fact the effective mass of the polaron, being frequency and temperature dependent.

The function F_q, defining the average number of phonons in the polaron cloud, was obtained by minimizing E_k with respect to F_q.

Hence

$$F_q^* = \frac{-\lambda_q^*}{\omega + \dfrac{q^2}{2m} - \dfrac{\boldsymbol{q}}{m}(\boldsymbol{k} - \sum \boldsymbol{q}_1 |F_{q_1}|^2)}. \tag{1.17}$$

This choice of F_q^* leads immediately to the polaron results of Lee, Low and Pines[2]). They showed that it is convenient to put $\sum_q \boldsymbol{q}\,|F_q|^2$ equal to $\eta_k \boldsymbol{k}$. Then in the limit $\boldsymbol{k} \to 0$ we may write

$$\eta_0 = \frac{\alpha/6}{1 + \alpha/6}. \tag{1.18}$$

2. *The effective mass of the polaron.* In order to evaluate, in a lowest order approximation, the effective mass of a drifting polaron we return to expression (1.15)

$$\sigma^{(0)}(z) = \frac{-e}{m\Omega}\sum_k \{\boldsymbol{k} - \sum_q \boldsymbol{q}\,|F_q|^2 - \sum_q \boldsymbol{q}\chi_z^{(0)}(\boldsymbol{k};\boldsymbol{q})\}\langle\langle \alpha_k^\dagger \alpha_k; \mathscr{D}\rangle\rangle_z^{(0)} \tag{1.15}$$

and introduce

$$F_q = \frac{-\lambda_q}{\omega + \dfrac{q^2}{2m} - \dfrac{\boldsymbol{qk}}{m}(1 - \eta_k)} \tag{1.17}$$

into the term between brackets.

The result is

$$\sigma^{(0)}(z) = \frac{-e}{m\Omega} \sum_{k} \left\{ k(1-\eta_k) + \bar{\nu} \sum_{q} q \frac{q^2}{2m} \frac{|\lambda_q|^2}{\left(\omega + \frac{q^2}{2m} - \frac{qk}{m}(1-\eta_k)\right)^2} \times \right.$$

$$\left. \times \left(\frac{1}{\omega - \frac{q^2}{2m} - \frac{qk}{m}(1-\eta_k) + z} + \frac{1}{\omega - \frac{q^2}{2m} - \frac{qk}{m}(1-\eta_k) - z} \right) \right\} \cdot$$

$$\cdot \langle\langle \alpha_k^\dagger \alpha_k ; \mathscr{D} \rangle\rangle_z^{(0)} \tag{2.1}$$

We note that this expression is only useful, if the denominator:

$$\omega + \frac{q^2}{2m} - \frac{qk}{m}(1-\eta_k)$$

cannot vanish. This means that $k < \sqrt{2m\omega}\,(1 + \alpha/6)$, a condition which restricts us to low temperatures. This restriction is inherent to the L.L.P. polaron model and can only be avoided with another choice of F_q. The a priori choice of F_q as in (1.17) is in fact the reason that we restrict the theory from the beginning to low temperatures, which simplifies the calculations considerably. For example it enables one to expand the terms between brackets in (2.1) in a power series in k. Then the first contribution comes from terms linear in k. These terms will give us the effective mass of the polaron. The contribution of the next term is of order $\bar{\nu}(T/\theta)$ provided we take a Maxwell-Boltzmann distribution for the polaron equilibrium density n_k. When $T \ll \theta$, we may neglect this kind of contributions. Hence, changing the summation into an integration and performing the integrals, we arrive, in the limit $z = i0$ at

$$\sigma^{(0)}(0) = \frac{-e}{m\Omega} \sum_{k} \{k(1-\eta_0) + 0{,}2\alpha\bar{\nu}k(1-\eta_0)\} \langle\langle \alpha_k^\dagger \alpha_k ; \mathscr{D} \rangle\rangle_{i0}^{(0)} =$$

$$= \frac{-e}{m_0^* \Omega} \sum_{k} k \langle\langle \alpha_k^\dagger \alpha_k ; \mathscr{D} \rangle\rangle_{i0}^{(0)} \tag{2.2}$$

where the effective mass m_0^* in a lowest order approximation, reads

$$m_0^* = m \frac{(1 + \alpha/6)}{(1 + 0.2\alpha\bar{\nu})}. \tag{2.3}$$

In terms of the infinite series (see I equation (3.41) we find

$$\sigma^{(0)}(0) = \sum_{n=0}^{\infty} \langle [J_{2n,d}^{(0)}(z), \mathscr{D}] \rangle_0, \tag{2.4}$$

where

$$J_0^{(0)}(z) = \frac{e}{m^* \Omega z} \sum_{k} k \alpha_k^\dagger \alpha_k. \tag{2.5}$$

The expression for the effective mass, (2.3), agrees fairly well with the results obtained by Yokota[4])

$$m_Y^* = m\left\{1 + \frac{\alpha/6}{(2\bar{\nu}+1)^{\frac{1}{2}}}\right\}. \tag{2.6}$$

But the temperature dependence of (2.3) and (2.6) is in contradiction with Fulton's expression for the effective mass[3])

$$m_F^* = m\{1 + (1+\bar{\nu})\,\alpha/6\}. \tag{2.7}$$

In fig. 4, we compare our results with those of Yokota, Fulton and Osaka[15]), in the special case of $\alpha = 3$

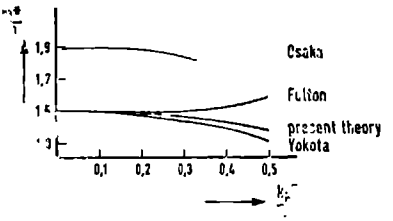

Fig. 4. Low-temperature effective mass for $\alpha = 3$.

We note that the temperature dependence of Osaka's effective mass expression agrees qualitatively with ours. The constant difference of about 0.4 is due to the use of different polaron models. Osaka's calculation is based on the Feynman polaron theory while Fulton and Yokota uses the L.L.P. polaron model. As we have seen the temperature dependent effective mass, m_0^*, follows straightforward from Kubo's formula in the intermediate coupling region. This in contrast with the method followed by the other authors. Yokota calculated the free energy of the polaron state using the Hartree approximation. Variational parameters, appearing in his treatment, were determined in such a way that the free energy is minimum. Osaka followed in fact the same way, but he made use of Feynman's variational principle to calculate the free energy. We remember that an extension of the theory to the intermediate temperature region (T comparable with θ) is, from a practical point of view, a rather difficult problem. For that reason one often uses the low-temperature mobility results in a temperature region, where it is, strictly speaking, not allowed. For instance, Van Heyningen[10]) obtained experimentally the drift mobility of polarons between 70°K and 350°K in AgCl ($\theta = 280$°K). Following Brown[5]) a considerable improvement of the fit between these experimental data and the calculated values is obtained, provided that one introduces a temperature dependent effective mass in the low-temperature mobility expression.

3. *The drift mobility at low temperatures.* As we have shown in I, the lowest order contribution to the conductivity follows from a partial summation

of the infinite series expansion (see I eq. (3.41). Introducing the expression for the effective mass, the result has been given by equation I (3.44)

$$\sigma_{\mu\nu}^{(0)}(z) = \frac{-ie^2}{m_z^*\Omega} \sum_{\boldsymbol{k}} k_\mu \Big\{ \frac{1}{z + \Gamma_{\boldsymbol{k}}(z)} \frac{\partial}{\partial k_\nu} n_{\boldsymbol{k}} +$$

$$+ \sum_{\boldsymbol{k}_1} \frac{1}{z + \Gamma_{\boldsymbol{k}}(z)} W_{\boldsymbol{k}-\boldsymbol{k}_1}(\boldsymbol{k}_1; \boldsymbol{k}) \frac{1}{z + \Gamma_{\boldsymbol{k}_1}(z)} \frac{\partial}{\partial k_{1\nu}} n_{\boldsymbol{k}_1} +$$

$$+ \sum_{\boldsymbol{k}_1, \boldsymbol{k}_2} \frac{1}{z + \Gamma_{\boldsymbol{k}}(z)} W_{\boldsymbol{k}-\boldsymbol{k}_1}(\boldsymbol{k}_1; \boldsymbol{k}) \frac{1}{z + \Gamma_{\boldsymbol{k}_1}(z)} \cdot$$

$$\cdot W_{\boldsymbol{k}_1-\boldsymbol{k}_2}(\boldsymbol{k}_2; \boldsymbol{k}_1) \frac{1}{z + \Gamma_{\boldsymbol{k}_2}(z)} \frac{\partial}{\partial k_{2\nu}} n_{\boldsymbol{k}_2} + \ldots \tag{3.1}$$

where the reciprocal relaxation time $\Gamma_{\boldsymbol{k}}(z)$ is defined by

$$\Gamma_{\boldsymbol{k}}(z) = \frac{i}{\tau_{\boldsymbol{k}}(z)} = \sum_{\boldsymbol{k}_1} W_{\boldsymbol{k}_1-\boldsymbol{k}}(\boldsymbol{k}; \boldsymbol{k}_1) \tag{3.2}$$

and

$$W_{\boldsymbol{q}}(\boldsymbol{k}; \boldsymbol{k} + \boldsymbol{q}) = |V_{-\boldsymbol{q}}(\boldsymbol{k})|^2 (1 + \bar{\nu}) \, \Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} - \omega; z) +$$

$$+ |V_{\boldsymbol{q}}(\boldsymbol{k} + \boldsymbol{q})|^2 \cdot \bar{\nu} \cdot \Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}} + \omega; z) +$$

$$+ \sum_{\boldsymbol{q}_1} \frac{q^2 q_1^2}{m^2} \{ |F_{-\boldsymbol{q}_1}|^2 \, |F_{-\boldsymbol{q}}|^2 \, (1 + 2\bar{\nu}) \, \Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}} - 2\omega; z) +$$

$$+ \bar{\nu} \sum_{\boldsymbol{q}_1} (|F_{-\boldsymbol{q}}|^2 \, |F_{\boldsymbol{q}_1}|^2 + |F_{-\boldsymbol{q}_1}|^2 \, |F_{\boldsymbol{q}}|^2) \, \Delta(E_{\boldsymbol{k}} - E_{\boldsymbol{k}+\boldsymbol{q}_1+\boldsymbol{q}}; z) \tag{3.3}$$

with the definitions

$$\Delta(x; z) = \frac{1}{x - z} - \frac{1}{x + z} \quad \text{and} \quad \bar{\nu} = \frac{1}{(e^{\beta\omega} - 1)}. \tag{3.4}$$

Note that $\Delta(x; z)$ reduces to $2\pi i \delta(x)$ in the limit $z = i0$.

Let us further put

$$\sigma_{\mu\nu}^{(0)}(z) = \sigma_{\mu\nu}^{(00)}(z) + \sigma_{\mu\nu}^{(01)}(z) \tag{3.5}$$

where

$$\sigma_{\mu\nu}^{(00)}(z) = \frac{-ie^2}{m^*\Omega} \sum_{\boldsymbol{k}} k_\mu \frac{1}{z + \Gamma_{\boldsymbol{k}}(z)} \frac{\partial}{\partial k_\nu} n_{\boldsymbol{k}} \tag{3.6}$$

and

$\sigma_{\mu\nu}^{(01)}(z)$ stands for the second and following terms in the series expansion (3.1). We want to show that $\sigma_{\mu\nu}^{(00)}(z)$ will give us the low-temperature contribution to the mobility, while $\sigma_{\mu\nu}^{(01)}(z)$ is at least of order $(T/\theta)(\theta = \omega/k)$ and hence may be neglected.

Introducing (3.3) into (3.2) we find in the limit $z = i0$

$$\Gamma_k(0) = 2\pi i \sum_q |\lambda_q|^2 \{(1+\bar{\nu})\,\delta(E_k - E_{k+q} - \omega) + \\ + \bar{\nu}\delta(E_k - E_{k+q} + \omega)\} + \sum_{q_1 q_2} \frac{q_1^2 q_2^2}{m^2} |F_{-q_1}|^2 |F_{-q_2}|^2 \cdot \\ \cdot \{(1+2\bar{\nu})\,\delta(E_k - E_{k+q_1+q_2} - 2\omega) + \bar{\nu}\,\delta(E_k - E_{k+q_1+q_2})\}. \tag{3.6}$$

Then, using (1.17), changing the summation into an integration and performing the integrals we arrive, in the limit $\boldsymbol{k} \to 0$, at

$$\Gamma_0 = i2\alpha\omega\bar{\nu}(1 + 0.06\alpha). \tag{3.7}$$

Hence $\sigma_{\mu\nu}^{(00)}(0)$ becomes

$$\sigma_{\mu\nu}^{(00)}(0) = \frac{e^2\bar{n}}{m_0^* 2\alpha\omega\bar{\nu}(1 + 0.06\alpha)}\,\delta_{\mu\nu}. \tag{3.8}$$

To estimate the order of magnitude of $\sigma_{\mu\nu}^{(01)}(z)$, it is convenient to introduce some simplifying assumptions, which avoid unnecessary involved integral expressions. Because our system is isotropic $\sigma_{\mu\nu}^{(0)}(z)$ reduces to a scalar ($\mu = \nu$) and we may replace k_μ^2 by $\frac{1}{3}k^2$. Further we put $z = i0$ and $\beta\omega \gg 1$, or $\bar{\nu} \ll 1$. For the polaron distribution, n_k, we take the Maxwell-Boltzmann distribution. Furthermore we restrict ourselves to small coupling constants α only assuming that the result is valid in the intermediate coupling region if we replace the band mass of the electron, m, by the effective mass of the polaron m^*. Because we are mainly interested in the temperature dependence of $\sigma_{\mu\nu}^{(01)}(z)$ these assumptions are reasonable.
Using these simplifications, and expressing Γ_k in terms of dimensionless variables, we obtain

$$\Gamma_k = 2i\alpha\omega \frac{1}{k} \{(1+\bar{\nu}) \cosh^{-1} k + \bar{\nu} \sinh^{-1} k\} + 0(\alpha^2) \tag{3.10}$$

while the transition probability is

$$W_{k-k_1}(\boldsymbol{k}_1; \boldsymbol{k}) = \frac{i\alpha\omega}{\pi} \frac{1}{|\boldsymbol{k} - \boldsymbol{k}_1|^2} \{(1+\bar{\nu})\,\delta(k_1^2 - k^2 - 1) + \bar{\nu}\,\delta(k_1^2 - k^2 + 1)\} \tag{3.11}$$

Note that the function $\cosh^{-1} k = \sinh^{-1}\sqrt{k^2 - 1}$ is only defined for $k \geqslant 1$ because of the delta function entering in (3.11).

Then introducing the normalized Maxwell distribution

$$n_k \equiv N \left(\frac{\beta\omega}{\pi}\right)^{\frac{3}{2}} e^{-\beta\omega k^2} \tag{3.12}$$

into (3.1) and changing the summation into an integration we arrive at

$$\sigma_0^{(01)} = \frac{ie^2\bar{n}}{m}\left(\frac{\beta\omega}{\pi}\right)^{\frac{1}{2}} 2\beta\omega \left\{ \int\int_{k_1 k_2} \frac{W_{k_1-k_2}(\boldsymbol{k}_2; \boldsymbol{k}_1)}{\Gamma_{k_1}} \frac{e^{-\beta\omega k_2^2}}{\Gamma_{k_2}} k_{1\mu} k_{2\mu} + \right.$$

$$\left. + \int\int\int_{k_1 k_2 k_3} \frac{W_{k_1-k_2}(\boldsymbol{k}_2; \boldsymbol{k}_1)}{\Gamma_{k_1}} \frac{W_{k_2-k_3}(\boldsymbol{k}_3; \boldsymbol{k}_2)}{\Gamma_{k_2}} \frac{e^{-\beta\omega k_3^2}}{\Gamma_{k_3}} k_{1\mu} k_{3\mu} + \ldots \right\} \quad (3.13)$$

From (3.13) it is clear that we have to estimate the order of magnitude of

$$A(\boldsymbol{k}_1; \boldsymbol{k}_2) = \frac{W_{k_1-k_2}(\boldsymbol{k}_2; \boldsymbol{k}_1)}{\Gamma_{k_1}}. \quad (3.14)$$

Written out explicitly it is

$$A(\boldsymbol{k}_1; \boldsymbol{k}_2) = \frac{1}{2\pi} \frac{1}{|\boldsymbol{k}_1 - \boldsymbol{k}_2|^2} k_1 \frac{(1+\bar{\nu})\,\delta(k_2^2 - k_1^2 - 1) + \bar{\nu}\delta(k_2^2 - k_1^2 + 1)}{((1+\bar{\nu})\cosh^{-1}k_1 + \bar{\nu}\sinh^{-1}k_1)} \quad (3.15)$$

We shall investigate in detail the behaviour of $A(\boldsymbol{k}_1; \boldsymbol{k}_2)$ for different values of $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$.

If $k_2 < 1$, the first term in (3.15) is zero because of the delta function. Then $A(\boldsymbol{k}_1; \boldsymbol{k}_2)$ can be approximated as

$$A(\boldsymbol{k}_1; \boldsymbol{k}_2) \approx \frac{1}{2\pi} \frac{k_1}{|\boldsymbol{k}_1 - \boldsymbol{k}_2|^2} \frac{\bar{\nu}\delta(k_2^2 - k_1^2 + 1)}{\cosh^{-1}k_1}, \quad 1 \leqslant k_1 < 2 \quad (3.16)$$

which is of order $\bar{\nu}$. In general $A(\boldsymbol{k}_1; \boldsymbol{k}_2)$ is equal to (3.16) if

$$m - 1 \leqslant k_2 < m \quad \text{and} \quad m \leqslant k_1 < m + 1$$

where m is a positive integer.

In order to illustrate what happens, we picture this behaviour in fig. 5.

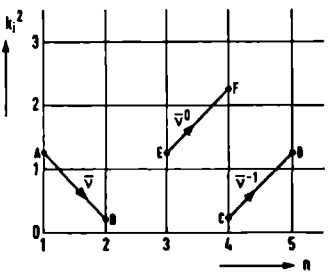

Fig. 5.

As abscissa we choose the number of variables in a certain term in (3.13). As ordinate we take the possible values of k_i^2. Then (3.16) is represented by the arrow AB, the order of magnitude is $\bar{\nu}$. The integration variable k_1^2

moves between the values 1 and 2 if the integration variable k_2^2 moves between the values 0 and 1.

It is then immediately clear that all arrows pointing downwards are of order $\bar{\nu}$. But if we assume that $k_4^2 < 1$ and $1 \leqslant k_5^2 < 2$ the order of magnitude of $A(\boldsymbol{k}_4; \boldsymbol{k}_5)$ is

$$A(\boldsymbol{k}_4; \boldsymbol{k}_5) \approx \frac{1}{2\pi} \frac{k_4}{|\boldsymbol{k}_4 - \boldsymbol{k}_5|^2} \frac{\delta(k_5^2 - k_4^2 - 1)}{\bar{\nu} \sinh^{-1} k_4}, \tag{3.17}$$

which is of order $\bar{\nu}^{-1}$.

We represent this by the arrow CD. All arrows pointing upwards, crossing the horizontal line $k^2 = 1$, are of order $\bar{\nu}^{-1}$. This is not the case for instance with the arrow EF. Because $1 \leqslant k_3^2 < 2$ and $2 \leqslant k_4^2 < 3$, the order of magnitude of $A(\boldsymbol{k}_3; \boldsymbol{k}_4)$ is

$$A(\boldsymbol{k}_3; \boldsymbol{k}_4) \approx \frac{1}{2\pi} \frac{k_3}{|\boldsymbol{k}_3 - \boldsymbol{k}_4|^2} \frac{\delta(k_4^2 - k_3^2 - 1)}{\cosh^{-1} k_3} \tag{3.18}$$

which is of order $\bar{\nu}^0$.

This means that all arrows pointing upwards above the line $k^2 = 1$ are of order $\bar{\nu}^0$. With these illustrations it is not difficult to estimate the order of magnitude of the n^{th} term in the expansion (3.13). But let us first discuss the first term. Here we have only two integration variables $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$. Because of the exponential dependence on k_2 in $e^{\beta\omega k_2^2}$, small values of k_2 contribute most to the integral, leading to a Γ_{k_2} of order $\bar{\nu}$. Hence $A(\boldsymbol{k}_1; \boldsymbol{k}_2)$ may be represented by the arrow AB in fig. 5, which is of order $\bar{\nu}$. As a result the first term in the expansion (3.13) is of order $\bar{\nu}^0$ and may be neglected.

The second term contains three integration variables, $\boldsymbol{k}_1$, $\boldsymbol{k}_2$ and $\boldsymbol{k}_3$. The possible contributions are represented in fig. 6 by the possible paths AEB, AEC, DEB and DEC

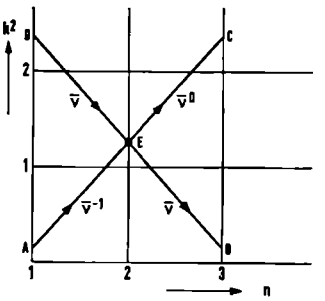

Fig. 6

The paths AEC and AEB are of order $\bar{\nu}^1$ and $\bar{\nu}^0$, respectively. Multiplying both contributions by the factor $e^{-\beta\omega k_3^2}/\Gamma_{k_3}$ yields for the order of magnitude

of the contribution AEC $\bar{\nu}^0$, while for the contribution along AEB we get $\bar{\nu}^{-1}$. Hence the contribution along the path AEB reads explicitly

$$\frac{ie^2\bar{n}}{m}\left(\frac{\beta\omega}{\pi}\right)^{\frac{1}{2}}2\beta\omega\int\!\!\int\!\!\int_{k_1\,k_2\,k_3}\frac{1}{2\pi}\;\frac{1}{|\boldsymbol{k}_1-\boldsymbol{k}_2|^2}\;\frac{k_1}{\bar{\nu}\sinh^{-1}k_1}\,\delta(k_2^2-k_1^2-1)\times$$

$$\times\frac{1}{2\pi}\;\frac{1}{|\boldsymbol{k}_2-\boldsymbol{k}_3|^2}\;\frac{k_2}{\cosh^{-1}k_2}\,\bar{\nu}\,\delta(k_3^2-k_2^2+1)\,\frac{k_3}{2i\alpha\omega\bar{\nu}\sinh^{-1}k_3}\,e^{-\beta\omega k_3^2}\,k_{1_\mu}k_{3_\mu} \tag{3.19}$$

which is equal to

$$\frac{e^2\bar{n}}{m2\alpha\omega\bar{\nu}}\left(\frac{\beta\omega}{\pi}\right)^{\frac{1}{2}}2\beta\omega\,\frac{4\pi}{3}\int_0^\infty k_1^4\,dk_1\left\{\frac{1}{2}\;\frac{k_1\sqrt{k_1^2+1}}{\sinh^{-1}k_1}\cdot\right.$$

$$\left.\cdot\int_{-1}^{+1}\frac{x\,dx}{(2k_1^2+1-2k_1x\sqrt{k_1^2+1})}\right\}^2\frac{k_1}{\sinh^{-1}k_1}\,e^{-\beta\omega k_1^2}\approx\frac{e^2\bar{n}}{m\,2\alpha\omega\bar{\nu}\cdot\beta\omega}. \tag{3.20}$$

It is now straightforward to make an estimation of the general term in (3.13). The n^{th} term reads

$$\frac{ie^2\bar{n}}{n}\left(\frac{\beta\omega}{\pi}\right)^{\frac{1}{2}}2\beta\omega\int\!\!\int\!\cdots\!\int_{k_1\qquad k_{n+1}}\frac{W_{k_1-k_2}(\boldsymbol{k}_2;\,\boldsymbol{k}_1)}{\Gamma_{k_1}}\cdots$$

$$\cdots\frac{W_{k_n-k_{n+1}}(\boldsymbol{k}_{n+1};\,\boldsymbol{k}_n)}{\Gamma_{k_n}}\;\frac{e^{-\beta\omega k^2_{n+1}}}{\Gamma_{k_{n+1}}}\cdot\boldsymbol{k}_1\boldsymbol{k}_{n+1}. \tag{3.21}$$

There are 2^n possible contributions that may be represented by 2^n possible paths in a graph. Drawing these it is clear that only the broken line of fig. 7 gives a contribution of order ν^{-1}.

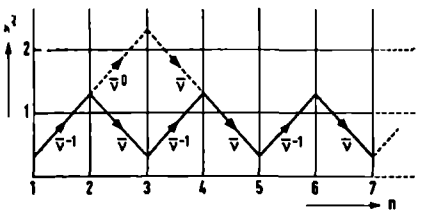

Fig. 7

We can see this by noting that only below the line $k^2 = 1$ arrows, pointing upwards, will be of order $\bar{\nu}^{-1}$, while as soon as we are beyond the line $k^2 = 2$ a factor $\bar{\nu}^{-1}$ is lost. (See for instance the dotted line in fig. 7).

Hence we may approximate (3.21) by the contribution of fig. 7 or ex-

plicitly

$$\frac{ie^2\bar{n}}{m}\left(\frac{\beta\omega}{\pi}\right)^{\frac{1}{2}}2\beta\omega\int_{k_1}\cdots\int_{k_n}\frac{1}{2\pi}\frac{k_1}{|\boldsymbol{k}_1-\boldsymbol{k}_2|^2}\frac{\delta(k_2^2-k_1^2-1)}{\sinh^{-1}k_1}\times$$

$$\times\frac{1}{2\pi}\frac{k_2}{|\boldsymbol{k}_2-\boldsymbol{k}_3|^2}\frac{\delta(k_3^2-k_2^2+1)}{\cosh^{-1}k_2}\cdots\frac{1}{2\pi}\frac{k_n}{|\boldsymbol{k}_n-\boldsymbol{k}_{n+1}|^2}\frac{\delta(k_{n+1}^2-k_n^2+1)}{\cosh^{-1}k_n}\cdot$$

$$\cdot\frac{e^{-\beta\omega k_{n+1}^2}\boldsymbol{k}_1\boldsymbol{k}_{n+1}}{2i\alpha\omega\bar{\nu}\dfrac{1}{k_{n+1}}-\sinh^{-1}k_{n+1}} \tag{3.22}$$

if n is even, thus an even number of factors $A(\boldsymbol{k}_i;\boldsymbol{k}_{i+1})$.

If n is odd, or if we have an odd number of $A(\boldsymbol{k}_i;\boldsymbol{k}_{i+1})$, the contribution is always of order $\bar{\nu}^0$, as in the first term of (3.13).

The expression (3.22) can be written as

$$\frac{e^2\bar{n}}{m}\frac{1}{2\alpha\omega\bar{\nu}}\left(\frac{\beta\omega}{\pi}\right)^{\frac{1}{2}}2\beta\omega\cdot\frac{4\pi}{3}\int_0^\infty k_1^4\,\mathrm{d}k_1\times$$

$$\times\left\{\frac{1}{2}\frac{k_1\sqrt{k_1^2+1}}{\sinh^{-1}k_1}\int_{-1}^{+1}\frac{x\,\mathrm{d}x}{(2k_1^2+1-2k_1x\sqrt{k_1^2+1})}\right\}^n\frac{k_1}{\sinh^{-1}k_1}-\mathrm{e}^{-\beta\omega k_1^2} \tag{3.23}$$

which is at least of order $(\beta\omega)^{-n/2}$ in the case that n is even.

Hence the low-temperature mobility is given by the first term in the series expansion (3.1) and turned out to be

$$\mu^{(00)}=\frac{e}{m_0^*2\alpha\omega\bar{\nu}(1+0.06\alpha)}. \tag{3.24}$$

At this stage, it is instructive to compare the result (3.24) with other mobility theories. Howarth and Sondheimer[6]) investigated the mobility problem with the aid of the Boltzmann equation approach neglecting the polaron properties of the electrons. Their result is exact in the limit $\alpha\to 0$ and $\beta\omega\to\infty$. They obtained

$$\mu_0=\frac{e}{m2\alpha\omega\bar{\nu}},\quad \alpha\ll 1,\quad \beta\omega\gg 1. \tag{3.25}$$

Low and Pines[7]) used Low's scattering formula together with the L.L.P. wave functions for the polaron low energy states. Their basic approximation is a one phonon cut-off procedure.

The result is

$$\mu_{\mathrm{LP}}=\mu_0\left(\frac{m}{m'}\right)^3 f(\alpha)\quad\text{where}\quad m'=m(1+\alpha/6),\ \beta\omega\gg 1. \tag{3.26}$$

The function $f(\alpha)$ is a slowly varying quantity of order 5/4 for $3 < \alpha < 6$. Using the optical theorem, Langreth[13]) modified the L.P. mobility result by

$$\mu_L = \mu_0 \frac{m}{m'} f(\alpha) \tag{3.27}$$

Perhaps the best results in the intermediate coupling region are found by Osaka[8]). He evaluated a mobility expression, using the Feynman polaron model and Feynman's path integral method. We represent his results in fig. 8, where the mobility as a function of the coupling constant has been plotted. For small coupling constants one finds

$$\mu_{0s} = \mu_0(1 - 0{,}173\alpha + \ldots) \quad \alpha \ll 1, \beta\omega \gg 1 \tag{3.28}$$

Besides these formulas the Feynman model calculations of Schultz and Morita[14]) are plotted in fig. 8 in order to show that widely different values of the polaron mobility are obtained by using different theories.

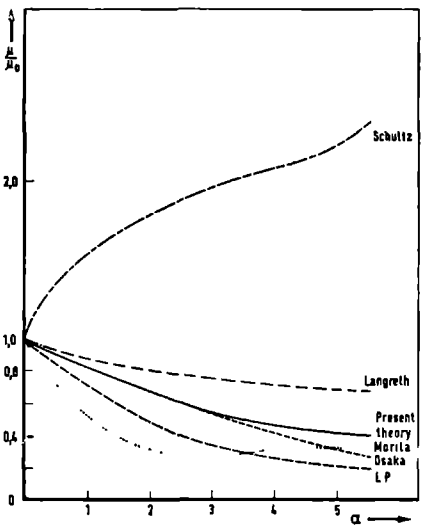

Fig. 8. Comparison of various mobility theories

Therefore, it is remarkable that the present results (3.24), are in complete accordance with Osaka's results up till α about 3, though the methods used in both cases are completely different. Beyond α about 3 the difference between Osaka's results and ours increases but for $\alpha > 7$ the Osaka mobility rises steeply. This increase of the mobility at large coupling constant cannot be expected in the present treatment.

The close correspondence between Osaka mobility and (3.24) suggests that in the intermediate coupling region the Osaka formula or (3.24) should be used in preference to the other ones. It is also noted that the obtained

mobility expression seems to be reasonably insensitive to a special polaron model in the intermediate coupling region. Besides the argument just mentioned, we can justify this by calculating the first order correction term to the mobility. It will be done in the next paragraph. A next argument has been given by Langreth and Kadanoff.

In their paper[9]) these authors stated that a possible criterion for reliability of a weak and intermediate coupling theory is, that its expansion in powers of α corresponds quite closely with the exact power series expansion. They calculated the first two terms of the exact series expansion and found

$$\mu = \mu_0(1 - \alpha/6 + \ldots) \tag{3.29}$$

Now the first nontrivial term in Osaka's expansion is $-0.173\mu_0\alpha$. It is easy to see that such a lowest term in our mobility expression (3.24) gives $-0.227\mu_0\alpha$, which is too small. But we shall show in § 4 that there are more contributions to the mobility of order $\mu_0\alpha$ by inspecting the higher order correction terms. It will been shown that the first order correction will exhaust together with (3.24), all possible contributions to the mobility of order μ_0 and $\mu_0\alpha$. These contributions will rectify the value $-0.227\ \mu_0\alpha$.

§ 4. *The correction terms.* Besides the correction $\sigma_0^{(01)}$, which is at least of order (T/θ) we have to evaluate the correction term $\sigma_0^{(10)}$, which does not vanish in the limit $(T/\theta) \to 0$. In the weak coupling limit its contributions are of order α^0. The power series expansion of the conductivity then becomes

$$\sigma_0 = \frac{e^2\bar{n}}{m2\alpha\omega\bar{\nu}}(1 + c_1\alpha + c_2\alpha^2 + \ldots) \tag{4.1}$$

c_1, c_2 etc. are numerical constants, independent of α and T/θ.

The explicit value of c_1, as calculated by Langreth and Kadanoff, turned out to be $-1/6$, indicating that the series expansion (4.1) converges rather badly in the intermediate coupling region.

In our treatment we obtain a series expansion analogous to (4.1). The first term has been given as

$$\sigma_0^{(00)} = \frac{e^2\bar{n}}{m(1 + \alpha/6)\ 2\alpha\omega\bar{\nu}(1 + 0.06\alpha)} \tag{4.2}$$

The next term, $\sigma_0^{(10)}$, may be calculated by considering the first order contributions to

$$\sigma_{\mu\nu}(z) = \sum_{n-0}^{\infty} \langle[\mathscr{I}_{\mu n}(z), \mathscr{D}_\nu]\rangle. \tag{1.12}$$

Three classes of first order contributions must be distinguished. The first one, $H_{\mu\nu}^{(1)}(z)$, consists of correction terms of pure statistical nature. The second class, $H_{\mu\nu}^{(2)}(z)$, contains statistical as well as dynamical effects, while

the last one, $H_{\mu\nu}^{(3)}(z)$, is a pure dynamical correction. $H_{\mu\nu}^{(1)}(z)$ is obtained by considering again the contribution

$$\frac{e}{m^*\Omega\Gamma_0}\sum_k k_\mu\langle[\alpha_k^\dagger\alpha_k, \mathscr{D}_\nu]\rangle. \tag{4.3}$$

The lowest order contribution was obtained by replacing in (4.3) the full statistical operator by the unperturbed one.

Hence a correction term of statistical character is obtained by expanding the statistical operator as

$$\mathrm{e}^{-\beta(H'_0+V)} = \mathrm{e}^{-\beta H_0'}\{1 - \int_0^\beta \mathrm{d}\beta_1\, \mathrm{e}^{\beta_1 H_0'}\, V\, \mathrm{e}^{-\beta_1 H_0'} + \\ + \int_0^\beta \mathrm{d}\beta_2 \int_0^{\beta_2} \mathrm{d}\beta_1\, \mathrm{e}^{\beta_2 H_0'}\, V\, \mathrm{e}^{-\beta_2 H_0'}\, \mathrm{e}^{\beta_1 H_0'}\, V\, \mathrm{e}^{-\beta_1 H_0'} + \ldots\} \tag{4.4}$$

We define H_0' as $H_0 - \mu N$. Taking into account the third term in (4.4) together with (4.3) we have to evaluate

$$H_{\mu\nu}^{(1)}(0) = \\ = \frac{e}{m^*\Omega\Gamma_0}\sum_k k_\mu \int_0^\beta \mathrm{d}\beta_2 \int_0^{\beta_2} \mathrm{d}\beta_1 \langle \mathrm{e}^{\beta_2 H_0'}\, V\, \mathrm{e}^{-\beta_2 H_0'}\, \mathrm{e}^{\beta_1 H_0'}\, V\, \mathrm{e}^{-\beta_1 H_0'}\, [\alpha_k^\dagger\alpha_k, \mathscr{D}_\nu]\rangle_0 = \\ = \frac{-ie^2}{m^*\Omega\Gamma_0}\sum_k k_\mu \frac{\partial}{\partial k_\nu}\Big\{\int_0^\beta \mathrm{d}\beta_2 \int_0^{\beta_2} \mathrm{d}\beta_1 \cdot \\ \cdot(|V_{-q}(k)|^2(1+\bar\nu)\exp\{\beta_2(E_k - E_{k+q} - \omega) + \beta_1(E_{k+q} - E_k + \omega)\} + \\ + |V_q(k+q)|^2\bar\nu \exp\{\beta_2(E_k - E_{k-q} + \omega) + \beta_1(E_{k+q} - E_k - \omega)\})\langle\alpha_k^\dagger\alpha_k\rangle_0 + \\ + \text{terms of order } \alpha^2\Big\}. \tag{4.5}$$

which in the low-temperature limit may be written as

$$H^{(1)}(0) = -\frac{ie^2\bar n}{m^*}\,\frac{1}{\Gamma_0}\,\alpha^2\psi^{(1)}(\bar\nu)\,\frac{1}{\Gamma_0}\cdot. \tag{4.6}$$

Note that $\psi^{(1)}(\bar\nu)$ is proportional to $i\omega\bar\nu$, and independent of α. The class of contributions of the mixed type, $H_{\mu\nu}^{(2)}(z)$, arises by considering.

$$H_{\mu\nu}^{(2)}(z) = \sum_{n=0}^{\infty}\langle[J_{k,2n+1,\mu}^{(1)}(z), \mathscr{D}_\nu]\rangle \tag{4.7}$$

Here we define

$$J_{2n+1}^{(1)}(z) = i\int_0^\infty \mathrm{e}^{izt}\,\mathrm{d}t[V, J_{2n,d}^{(0)}(z)]_t, \tag{4.8}$$

where

$$J^{(0)}_{2n,d}(z) = i^2 \int_0^\infty e^{izt_1}\, dt_1 \int_0^\infty e^{izt_2}\, dt_2 [V,[V,J^{(0)}_{2n-2}(z)]_{t_2}]_{t_1,d} \qquad \text{(I,3.26)}$$

and

$$J^{(0)}_0(z) = \frac{e}{m^*\Omega z} \sum_k k \alpha^\dagger_k \alpha_k. \qquad (2.5)$$

Written out explicitly we arrive at

$$H^{(2)}_{\mu\nu}(z) = \sum_{n=0}^\infty \left(\frac{-\Gamma_0(z)}{z}\right)^n \cdot -i \int_0^\infty e^{izt}\, dt \langle [[V,\, J^{(0)}_{\mu,0}(z)]_t,\, \mathscr{D}_\nu]\rangle =$$

$$= \frac{-1}{1 + \dfrac{\Gamma_0(z)}{z}} - i \int_0^\infty e^{izt}\, dt \langle [[V,\, J^{(0)}_{\mu,0}(z)]_t,\, \mathscr{D}_\nu]\rangle \qquad (4.9)$$

Or

$$H^{(2)}_{\mu\nu}(z) = \frac{1}{(z + \Gamma_0(z))} \frac{e}{m^*\Omega} \left\{ \left\langle \left[\sum_{kq} q_\mu \alpha^\dagger_k \alpha_{k+q} \left(\frac{V^*_{-q}(k)\, \beta_{-q}}{E_k - E_{k+q} - \omega + z} + \right.\right.\right.$$

$$\left.\left.\left. + \frac{V_q(k+q)\, \beta^\dagger_q}{(E_k - E_{k+q} + \omega + z)} \right), \mathscr{D}_\nu \right] \right\rangle + \text{terms of order } \alpha^2 \right\}. \qquad (4.10)$$

Because

$$\langle [\alpha^\dagger_k \alpha_{k+q} \beta_{-q},\, \mathscr{D}_\nu]\rangle \text{ is equal to } -ie \frac{\partial}{\partial k_\nu} \langle \alpha^\dagger_k \alpha_{k+q} \beta_{-q}\rangle$$

and

$$\langle [\alpha^\dagger_k \alpha_{k+q} \beta^\dagger_q,\, \mathscr{D}_\nu]\rangle = -ie \frac{\partial}{\partial k_\nu} \langle \alpha^\dagger_k \alpha_{k+q} \beta^\dagger_q\rangle$$

we find

$$H^{(2)}_{\mu\nu}(z) = \frac{-ie^2}{m^*\Omega(z + \Gamma_0(z))} \sum_{kq} q_u \left\{ \frac{V^*_{-q}(k)}{E_k - E_{k+q} - \omega + z} \frac{\partial}{\partial k_\nu} \langle \alpha^+ \dagger_k \alpha_{k+q} \beta_{-q}\rangle + \right.$$

$$\left. + \frac{V_q(k+q)}{E_k - E_{k+q} + \omega + z} \frac{\partial}{\partial k_\nu} \langle \alpha^\dagger_k \alpha_{k+q} \beta^\dagger_q\rangle \right\}. \qquad (4.11)$$

Correlating this expression with the second term of (4.4) the second class of contributions is

$$H^{(2)}_{\mu\nu}(z) = \frac{ie^2}{m^*\Omega(z + \Gamma_0(z))} \int_0^\beta d\beta_1 \sum_{kq} q_\mu \times$$

$$\times \left\{ \frac{V^*_{-q}(k)}{E_k - E_{k+q} - \omega + z} \frac{\partial}{\partial k_\nu} \langle e^{\beta_1 H_0'} V e^{-\beta_1 H_0'} \alpha^\dagger_k \alpha_{k+q} \beta_{-q}\rangle_0 + \right.$$

$$\left. + \frac{V_q(k+q)}{E_k - E_{k+q} + \omega + z} \frac{\partial}{\partial k_\nu} \langle e^{\beta_1 H_0'} V e^{-\beta_1 H_0'} \alpha^\dagger_k \alpha_{k+q} \beta^\dagger_q\rangle_0 \right\}. \qquad (4.12)$$

Taking the low-temperature limit and putting z equal to $+i0$, the result may be expressed by

$$H^{(2)}(0) = \frac{ie^2\bar{n}}{m^*} \frac{1}{\Gamma_0} \alpha^2 \psi^{(2)}(\bar{\nu}) \frac{1}{\Gamma_0} \tag{4.13}$$

Note that $\psi^{(2)}(\bar{\nu})$ is proportional to $i\omega\bar{\nu}$ and independent of α.

The last class of contributions is purely dynamical in character and may be written as (4.6) or (4.13) by summing all possible diagrams of the type of fig. 9.

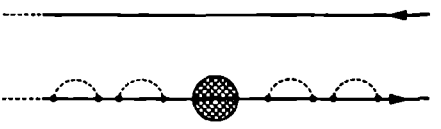

Fig. 9. Contribution to $H_{\mu\nu}^{(3)}(z)$

The cross-hatched circle represents a contribution of order α^2. We indicate it by $\alpha^2\psi_z^{(3)}(\bar{\nu})$, where the function $\psi_z^{(3)}(\bar{\nu})$ is independent of α.
The simplest example of this kind of contributions arises from the contributions of

$$\begin{aligned} i^3 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \int_0^\infty e^{izt_3}\,dt_3 \langle [\{[V_2,[V_1,\,[V_1,\,\boldsymbol{J}_0(z)]_{t_3}]_{t_2,\,nd}]_{t_1,\,d} + \\ + [V_1,[V_2,\,[V_1,\,\boldsymbol{J}_0(z)]_{t_3}]_{t_2,\,nd}]_{t_1,\,d} + [V_1,[V_1,[V_2,\,\boldsymbol{J}_0(z)]_{t_3}]_{t_2,\,nd}]_{t_1,\,d}\}, \\ + i^4 \int_0^\infty e^{izt_1}\,dt_1 \int_0^\infty e^{izt_2}\,dt_2 \int_0^\infty e^{izt_3}\,dt_3 \int_0^\infty e^{izt_4}\,dt_4 \cdot \\ \cdot \langle [[V_1,\,[V_1,\,[V_1,\,[V_1,\,J_0(z)]_{t_4}]_{t_3,\,nd}]_{t_2}]_{t_1,\,d},\,\mathscr{D}] \rangle_0 \end{aligned} \tag{4.14}$$

where $[A,\,B]_{t_2,\,nd}$ means the non-diagonal part of the operator $[A,\,B]$ at time t_2.
(4.14) is of order α^2. Again, in the low-temperature limit we may replace (4.14) by

$$\alpha^2\psi_z^{(3)}(\bar{\nu}) \langle [\boldsymbol{J}_0^{(0)}(z),\,\mathscr{D}] \rangle_0 \tag{4.15}$$

which defines the function $\psi_z^{(3)}(\bar{\nu})$.

Summing all possible contributions of this type (see fig. 9), we finally obtain in the limit $z = i0$

$$H^{(3)}(0) = \frac{ie^2\bar{n}}{m^*} \frac{1}{\Gamma_0} \alpha^2 \psi^{(3)}(\bar{\nu}) \frac{1}{\Gamma_0}. \tag{4.16}$$

Putting:

$$\psi(\bar{\nu}) = \psi^{(1)}(\bar{\nu}) + \psi^{(2)}(\bar{\nu}) + \psi^{(3)}(\bar{\nu}) \tag{4.17}$$

the first order contribution to the conductivity turned out to be of the form

$$\sigma_0^{(10)} = \frac{ie^2\bar{n}}{m^*} \frac{1}{\Gamma_0} \alpha^2 \psi(\bar{\nu}) \frac{1}{\Gamma_0} \tag{4.18}$$

Hence

$$\sigma_0^{(10)} = \frac{e^2\bar{n}}{m(1+\alpha/6)} \cdot \frac{1}{2\alpha\omega\bar{\nu}(1+0{,}06\alpha)} \alpha^2\psi(\bar{\nu}) \frac{1}{2i\alpha\omega\bar{\nu}(1+0.06\alpha)} \tag{4.19}$$

which is of order α^0 in the weak coupling limit. It is clear that in order to find the correct result, $\psi(\bar{\nu})$ must be proportional to $i\omega\bar{\nu}$.

Taking this into account the first correction term to the mobility may be written as

$$\mu_0^{(10)} = \frac{e}{2m^*\alpha\omega\bar{\nu}(1+0.06\alpha)} \frac{c\alpha}{2(1+0.06\alpha)} \tag{4.20}$$

The evaluation of the numerical constant c might be done directly, using the formula (4.5), (4.12) and (4.14).
On the other hand it is easier to obtain the explicit value of c by using the fact that the mobility does not depend on the special form of the function F_q. Hence, using different functions F_q, two different power series expansions of the mobility in a coupling parameter are obtained, leading to exactly the same power series expansion, provided that one expands both series expansions in the coupling constant α. Using this fact we compare the series expansion

$$\sigma_0 = \frac{e^2\bar{n}}{2m(1+\alpha/6)} \frac{1}{\alpha\omega\bar{\nu}(1+0.06\alpha)} \left(1 + \frac{c}{2(1+0.06\alpha)} + \ldots\right) \tag{4.21}$$

with the series expansion obtained by taking $F_q = 0$. It will be clear that the last one is much easier to evaluate.

Using to above developed method we arrive, in the case $F_q = 0$, at

$$\sigma_0 = \frac{e^2\bar{n}}{2m\alpha\omega\bar{\nu}} (1 - \alpha/6 \ldots). \tag{4.22}$$

which is just the result of Langreth and Kadanoff.

Hence expanding (4.21) up till terms of order α^0 and comparing the coefficients with (4.22) we obtain $c = 0.12$.

As a result the first correction term to the low-temperature mobility is

$$\mu_0^{(10)} = \mu_0^{(00)} \frac{0.06\alpha}{(1+0.06\alpha)} \tag{4.23}$$

This correction term changes the mobility $\mu_0^{(00)}$ by some 15% when α is 3 and by 26% when α is 6. This is in accordance with our assertion that the present treatment leads to a satisfactory convergence of (4.21) in the intermediate coupling region.

5. *Concluding Remarks.* As we have shown the introduction of a unitary transformation into the Kubo formalism acts to increase the convergence

of the power series expansion in the coupling constant of the low-temperature mobility of polarons.

The first term in this series expansion is sufficient if we choose for the unitary transformation the L.L.P. transformation and if we restrict ourselves to the intermediate coupling region. The close correspondence with the mobility expression of Osaka shows in addition, that the obtained results are reasonably insensitive to the properties of the used polaron model.

The L.L.P. polaron model as well as the Feynman description will lead to essentially the same mobility behaviour in the experimentally interesting range of coupling constants.

Nevertheless, the fit of the theoretical formula with the experimental results, though slightly improved, remains rather unsatisfactory. The reason for this is not obvious. Of course other than polaron-phonon scattering processes might be important. But besides these questions it is worth-while to note that most experiments are performed in a temperature region where the requirement $T \ll \theta$ is not very well satisfied. As a consequence the correction term $\mu_0^{(01)}$, starting with a contribution of order $T/\theta\ \mu_0^{(00)}$, may not be neglected any longer.

At the other hand higher temperatures reduce the effective mass of the polaron, tending to increase the fit between theory and experiment. In principle, the analysis given here can be extended to the case of intermediate temperatures. Calculations of the drift mobility at intermeditate temperatures are in progress.

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MOTIVERING EN SAMENVATTING

Het polaronprobleem, waarin de eigenschappen van elektronen in de geleidingsband van polaire kristallen onderzocht worden, is sinds de jaren 30 om verschillende redenen actueel gebleven.

In de eerste plaats wegens de toepassingsmogelijkheden in de halfgeleiderfysica. Een tweede belangrijke reden is de fundamentele rol die het probleem in de ontwikkeling van de vaste stof fysica heeft gespeeld. Het heeft deze rol te danken aan het feit dat het polaron in principe een eenvoudig model voorstelt van een deeltje in wisselwerking met een gekwantiseerd veld, echter zonder dat hierbij de divergenties optreden die b.v. in de kwantum elektrodynamica zo'n belangrijke rol spelen.

Dit leidde ertoe dat het polaronprobleem als testobject werd gebruikt om nieuwe methoden en concepties te toetsen. Al in 1933 voerde Landau, in een poging de eigenschappen van F-centra te verklaren, het polaron in als quasi-deeltje. Een elektron, aangebracht in de geleidingsband van een ionenkristal, produceert, door zijn Coulomb-interactie met de ionen, een roosterdeformatie en bijgevolg een polarisatieveld. Landau vermoedde dat verschillende eigenschappen van ionenkristallen verklaard konden worden door het elektron plus de roosterdeformatie als eenheid op te vatten en zijn beweging door het kristal te bestuderen.

Naast het werk van Landau gaf Fröhlich in 1937 een kwantitatieve behandeling van elektronenverstrooiing in ionenkristallen. Door de wisselwerking van de elektronen met de roostertrillingen in de behandeling te betrekken, voerde hij voor het eerst het veld van de roostertrillingen in als nieuw begrip. 11 Jaar later paste Fröhlich de kwantum veldentheorie op het probleem toe, in, zoals hij zelf zegt: "A preliminary investigation to approach the problem of superconductivity".

Het belang dat Landau en Fröhlich, op grond van het polaron probleem, aan de elektron-phonon wisselwerking toekende, is de basis geweest van de snelle ontwikkeling in de vaste stof fysica in de jaren 50. Reeds in 1950 kon

Fröhlich de elektron-phonon wisselwerking aanwijzen als oorzaak van het bestaan van supergeleiding en in 1953 konden Böhm en Pines het collectief gedrag van elektronen in metalen (plasmonen) als fysische oorzaak noemen van de divergenties die bij de mathematische problemen optraden. Böhm en Pines konden dit uitvoeren dank zij een unitaire transformatie techniek die Pines, in samenwerking met Lee en Low, juist te voren met zoveel succes op het polaron probleem had toegepast.

Hiermee is aan enkele voorbeelden geïllustreerd hoe het polaron probleem als testobject voor verschillende technieken en concepties heeft gediend.

Dit alles leidde echter tot de ontwikkeling van vele polaronmodellen, die allen verschillende resultaten opleverden, zowel wat betreft de zelfenergie en effective massa als wat betreft de mobiliteit (Zie fig. 8, hoofdstuk II). Uit de verschillende resultaten een juiste keuze te doen scheen een probleem.

Een oplossing leverde het Kubo formalisme, dat Kadanoff en Langreth in 1964 de mogelijkheid gaf het mobiliteitsprobleem op exacte wijze te formuleren. D.w.z. zij gaven een formele reeks ontwikkeling aan van de mobiliteit in machten van de elektron-phonon koppelingsconstante en berekenden de eerste twee termen expliciet.

Iedere mobiliteitstheorie moet nu tot deze reeksontwikkeling herleidbaar zijn, onafhankelijk van het toegepaste polaron model. Aangezien deze reeks ontwikkeling slechts voor kleine waarden van de koppelingsconstante voldoende snel convergeert, kan deze theorie alleen een criterium vastleggen waaraan andere theorieën moeten voldoen zonder zelf een uitspraak te kunnen geven omtrent de waarde van de mobiliteit in het intermediaire koppelingsgebied.

Daar echter de koppelingsconstante van de meeste polaire kristallen in het intermediaire gebied liggen, is dit een belangrijke tekortkoming van deze theorie.

In dit proefschrift wordt een exacte reeksontwikkeling van de mobiliteit in machten van de polaron-phonon wisselwerking voorgesteld, die voldoende snel convergeert in het intermediaire gebied om met de eerste term van de reeks genoegen te kunnen nemen.

In het eerste hoofdstuk worden de zelfenergie en effectieve massa bestudeerd van een polaronmodel dat als een vereenvoudiging van het L.L.P. model opgevat kan worden. Er wordt aangetoond dat de eigenschappen van dit model, voor intermediaire koppelingssterkten, dezelfde zijn als die van het L.L.P. model. Verder wordt met behulp van de Kubo formule het elektrisch geleidingsvermogen en dus de driftmobiliteit uitgedrukt in de eigenschappen van het vereenvoudigd L.L.P. model. Deze methode levert behalve een uitdrukking voor de mobiliteit tevens een uitdrukking op voor de effectieve massa, die nu, in het algemeen, afhangt van de temperatuur, de frequentie van het aangelegde veld en de koppelingsconstante.

In het tweede hoofdstuk worden de effectieve massa en de mobiliteit, in

een lage temperatuur benadering, expliciet berekend. Twee typen correctie termen worden bestudeerd. De eerste is een temperatuurcorrectie terwijl de tweede bepaald wordt door de waarde van de koppelingsparameter.

De temperatuurcorrectie blijkt, in eerste instantie, van de orde T/θ te zijn, waarbij T de absolute temperatuur en θ de Debye-temperatuur van de optische phononen voorstelt. De tweede correctie wordt pas van belang wanneer de koppelingsconstante groter is dan 3. De exacte formulering van het probleem heeft nu de eigenschap dat, in de lage temperatuur limiet en in het intermediaire koppelingsgebied, de resultaten voor de mobiliteit onafhankelijk moeten zijn van het toegepaste polaronmodel. Een vergelijking van de in hoofdstuk II verkregen resultaten met die van andere mobiliteitstheorieën leert dan dat er in het intermediaire koppelingsgebied een nauwkeurige overeenstemming bestaat met de resultaten van Osaka. Opgemerkt dient te worden dat Osaka gebruik maakte van het Feynman polaronmodel.

STELLINGEN

I

De berekening van de diffusiecoëfficiënt van het ééndimensionale harde bollengas m.b.v. de Boltzmannvergelijking levert voor lage dichtheden een waarde op die 13% afwijkt van de exacte waarde.

Jepsen, D. W., Jour. of Math. Phys. **6** (1965) 405.

II

De hogere orde dichtheidscorrecties van de transportcoëfficienten van het ééndimensionale harde bollengas, berekend m.b.v. de methode van Bogolyubov, zijn allen singulier.

III

De triple-botsingsterm van het tweedimensionale onzuiverhedenmodel bezit een singulariteit van het logarithmische type. Bij de berekening van transportcoëfficiënten geeft dit aanleiding tot een correctieterm van het type $n \ln n$ (n = dichtheid).

Kawasi, K., Oppenheim, I., Phys. Rev. **139** (1965) A 1763.

IV

De coëfficiënt van de logarithmische term in de dichtheidsontwikkeling van transportcoëfficiënten wordt, voor klassieke gassen met repulsieve korte afstandswisselwerking, alleen bepaald door de triple-botsingsterm. De hogere orde botsingsprocessen hebben op de waarde van deze coëfficiënt geen invloed.

V

De door Ernst toegepaste correlatiefunctie-methode om de transportcoëfficiënten van het harde bollengas te berekenen, is aan bedenkingen onderhevig.

Ernst, M.H., Transport coëfficiënts in dense gases IV. Wordt gepubliceerd in Physica.

VI

De interferentietermen, gedefiniëerd door Michel en Verboven en volgend uit een generalisatie van de Boltzmannvergelijking, zijn verschillend van nul in het geval van het harde bollengas.

Michel, K. H., Verboven, E., Physics Letters **8** (1964) 176.

VII

De theorie van Singwi voor coherente neutronverstrooiing bevat enkele ernstige tekortkomingen.

Singwi, K. S., Phys. Rev. **136** (1964) A 969.

VIII

De differentiaal-integraal vergelijking, die het klassieke Feynman-polaron beheerst, is invariant onder de canonische transformatie die het klassieke equivalent voorstelt van de transformatie, die gedefiniëerd wordt in hoofdstuk I, formule 2 : 18 van dit proefschrift.

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IX

De Ornstein-Zernike theorie voor kritische verschijnselen wordt in 6 of meer dimensies reeds exact.

X

Er bestaat een ongegronde neiging om alles in "Letters" te publiceren.

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