ON HOPPING TRANSPORT IN A DISORDERED LATTICE\

PHYSICS

1968

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Abstract

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UDC 539.2

PHYSICS

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ON HOPPING TRANSPORT IN A DISORDERED LATTICE*

(Presented by Academician S. V. Vonsovskii, 31 X 1967)

§ 1. **Statement of the problem.** In this paper a quantum theory is developed (the basic relations) for the even (with respect to the magnetic field H) coefficients $\sigma_{AB}^{(s)}$ of hopping transport in certain disordered lattices. For definiteness, the theory is set forth for the electrical conductivity $\sigma_{xx} \equiv \sigma_p^{(l)}$ in the ohmic (and in a certain non-ohmic) region of electric fields E, and for the thermopower $\gamma_p^{(l)}$. The basic relations describe, at least for $l = 1$, the transport of nonadiabatic small polarons (below, s.p.) in a doped host lattice of a semiconductor; for $l = 2$, impurity conduction (in a more general approach, cf. ([3,4])), in particular, for strong-coupling polarons of both large and small radius.

A. Retaining essentially the model (and notation) of ([1,2]), we consider a quantum system of interacting second-quantized electrons (holes) and phonons in the "random" field of a disordered lattice of the proper "ions" (host impurity centers—for $l = 2$); the corresponding states $|s\rangle$ of Wannier type are basic for the electrons. For brevity, the discussion concerns mainly the case of strong electron-phonon coupling, when its parameter $\Phi \equiv \Phi^{(l)}(T) \geq \Phi_0 \equiv \Phi^{(l)}(0) \gg 1$. Taking into account here the smallness of Bloch electronic overlaps $\Delta_e(s_{12})$ (formulas (2), (3)) and the pair repulsion I of polarons on an individual "ion," the Hamiltonian here (see also ([14])) may be taken in the form ($\hbar \equiv 1$)**:

$$\hat{\mathcal{H}} \equiv \hat{\mathcal{H}}^{(l)} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_1; \qquad \hat{\mathcal{H}}_0 = \sum_{s\sigma} \varepsilon(s)\hat{n}_{s\sigma} + \sum_{\Lambda} \omega_\Lambda c_\Lambda^+ c_\Lambda + I\sum_s \hat{n}_{s,1/2}\hat{n}_{s,-1/2};$$

$$\hat{\mathcal{H}}_1 = \sum_{s_1 s_2 \sigma} \sum_{\Lambda} \Delta_e(s_{12}) \left(\hat{T}_\Lambda^{s_1} + \hat{T}_\Lambda^{s_2}\right) a_{s_1\sigma}^+ a_{s_2\sigma}, \tag{1}$$

where

$$\hat{n}_{s\sigma} \equiv a_{s\sigma}^+ a_{s\sigma}; \qquad c_\Lambda \equiv \hat{T}_\Lambda^+ b_\Lambda \hat{T}; \qquad \sigma = \pm\tfrac{1}{2}; \qquad \Lambda \equiv \{fj\} \text{ or } \{\lambda\};$$

$j = 1, 2, ...,$ and $\hat{T}_\Lambda \equiv \prod \hat{T}^s_\Lambda$ is a renormalizing unitary transformation ([2]); λ describes possible local phonons ([5]). Below the actual case meant is $I \gg I_0 \equiv kT + D_{cl}$. In calculating $N_c(\zeta)$ and other

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* The main results (§ 2 A, B and § 3) were contained in the author's review report at the International Conference on Magnetic Oxides (Czechoslovakia, 1966) ([16]); see also ([1a]).

** In what follows the following notation is also used: $c \equiv N_{iN}0^{-1} < 1$, where $N_0 \equiv N^{(i=1)}$ and $N_i \equiv N^{(i=2)}$; $N \equiv N^{(l)} = 3/4\pi r_0^3$ and $N_c \equiv N_c^{(l)}$ are the concentrations of "ions" and current carriers, respectively, with $r_0 \equiv r_0^{(l)}$; K is the degree of compensation of the host impurity, $0 \leq K \leq 1$.

macroscopic quantities ([5,1]) require an adequate averaging $\langle ... \rangle_{AV}$ over "random" configurations $\{s\}$ of "ions," with allowance for concentration broadening due to the fluctuations $\{\Delta_e(s)\}$ and $\{u\}$, $u \equiv \varepsilon(s) - \bar{\varepsilon} \equiv u_s$ (energy measured from the bottom of the conduction band at $c = 0$); it turns out that the chemical potential of the charge carriers (hereafter called polarons) $\bar{\zeta} \equiv \zeta - \bar{\varepsilon} \ll I/2$ for $p \equiv p^{(l)} \equiv N_{cN}^{-1} \ll 1$, with $p^{(1)} \ll 1$ for $c(1-K) \ll 1 - K_0$ and $p^{(2)} \ll 1$ for $K \gg K_0 \sim \exp(-I/2kT) \ll 1$;

B. We apply Kubo' s general formulas for $\sigma_{AB}^{(s)}$, which also include $\langle ... \rangle_{AV}$ (see above), and the special perturbation theory from ([2,1]) in the present case of classical concentration broadening (D_{cl}), with effective width $D_{cl} \ll (2\mathcal{E}\omega_p)^{1/2} \simeq \Phi_0^{1/2}\omega_p$ (and $D_{cl} \ll \delta_0 \equiv |\varepsilon_{c=0}^{(2)}|$). From an analysis of the series of terms in the expansion of σ_p (in Δ_e) it follows that the principal contribution $\sigma_p = {\sigma_p^{(hop)}}_0$ is due to uncorrelated two-site hopping involving incoherent (multi-)phonon processes ([2]). For $p \ll 1$, the inequalities ([1]) were adopted as criteria of the theory (for $\omega_\Lambda \simeq \text{const} \equiv \omega_p$): $\Delta_0 \ll (\mathcal{E}kT)^{1/2} (< \mathcal{E})$ for $\mathcal{E}/k > T > T_0 \equiv \hbar\omega_p/2k$, or $\Delta_0 \ll \xi D_{cl}$ for $T < T_0(\operatorname{Ar\,sh} 2\Phi_0)^{-1}$ for m.p.; moreover $\mathcal{E} \equiv \mathcal{E}^{(l)} \simeq \frac{1}{2}\omega_p\Phi_0$ and $\xi \equiv \xi^{(l)}$, $\xi^{(1)} \approx 1$ and $\xi^{(2)} > 1$; $\Delta_0^{(2)} \ll \xi^{(2)} D_{cl}^{(2)}$ for $\xi^{(2)} \gtrsim 1$ for continuum polarons of strong coupling. Here $\Delta_0 \equiv \Delta_0^{(l)}(r_0) \equiv \Delta_e[r_0 a_l(r_0)]$, with $a_l(r_0) \approx 1$, while $\Delta_0^2(r_0)$ can be estimated from the values $\sigma_{\ell 0} \propto \Delta_0^2$ (see (2) and below). The criterion $\Delta_0^{(2)} \ll \xi^{(2)} D_{cl}^{(2)}$ can be satisfied for small $N_i < N_{cr} \equiv 3/4\pi r_{cr}^3$ and, tentatively, apparently $r_{cr}^3 (r_B^{imp})^{-3} \sim (10^2 \div 10^3)$ (cf. ([3,7])).

§ 2. Electrical conductivity. Optical absorption.

A. The explicit formula obtained for $\bar{\sigma}_{p0}(\omega) \equiv \operatorname{Re} {\sigma_p^{(l)}}_0(\omega)$ (in a weak field E) takes into account the "self-consistent" character of the hops of charge carriers under their mutual repulsion I and the Pauli correlation in Fermi degeneracy. In particular, for $\eta \equiv \mu_{BH}\sigma \ll kT$ and $p \ll 1$ (cf. $\bar{\sigma}_{xx}^h(\omega)$ in ([2]))

$$\bar{\sigma}_{p0}(\omega) \equiv |e| N_c \mu_p(\omega) = \frac{2e^2}{\omega} \operatorname{th} \frac{\beta\omega}{2} \cdot \varphi_p^{(0)}(\omega), \tag{2}$$

where

$$\varphi_p^{(q)}(\omega) \equiv \sum_{s_1 s_2} \sum_{u_1 u_2} \left\langle \sigma_p(s_{12}; u_1, u_2; \omega) \left(\frac{u_1 + u_2}{2} \right)^q \right\rangle_{AV};$$

$$\sigma_p(s_{12}; u_1, u_2; \omega) \simeq (s_{12})_x^2 W_h^{12}(\omega) Z_{cc}^{(12)};$$

$$W_h^{(12)}(\omega) = \frac{1}{2} |\Delta_e(s_{12})|^2 \sum_{\pm} \operatorname{Re} \int_0^\infty dt \, \left(e^{\Psi(t)} - \right.$$

$$\left. -1\right) \exp[-2\Phi + it(\eta_1 - \eta_2) + i(t - i\beta/2)(u_{12} \pm \omega)];$$

$$Z_{cc}^{(12)} \equiv f_\infty(u_1)(1 - f_\infty(u_2));$$

$$f_\infty(u) \equiv f(u; \beta I \gg 1) = \{1 + \tfrac{1}{2} \exp(\beta u - \beta\bar{\zeta})\}^{-1};$$

$$\beta \equiv 1/kT; \qquad s_{12} = s_1 - s_2; \qquad u_{12} \equiv u_1 - u_2;$$

$$\Psi(t) = \sum_{\Lambda} |\gamma_\Lambda^{s_1} - \gamma_\Lambda^{s_2}|^2 \omega_\Lambda^{-2} \cos \omega_\Lambda t \left(\operatorname{sh} \frac{\beta\omega_\Lambda}{2} \right)^{-1}.$$

W_h is the "one-particle" (without allowing for I) hopping probability (per 1 sec.), with $\mu_p \ll \mu_0 \equiv |e| r_0^2 \hbar^{-1}$. For sufficiently small $p < p_0 (\ll 1)$, when $\zeta(p) < \zeta(p_0) < 0$ (absence of degeneracy), the contribution of I is insignificant and $\sigma_{p0} \propto p \simeq 2Z_1 \exp(-\beta|\bar{\zeta}|)$ for $Z_1 \equiv \langle \exp(-\beta u) \rangle_{AV}$. For $p \ll 1$ and $T > T_0$

$$\sigma_p^{(l)}(\omega = 0) \sim \mu_0 \cdot \beta z \Delta_0^2 / (\mathcal{E} kT)^{1/2} \cdot \exp[-\beta \mathcal{E}^{(l)} - \beta W^{(l)}],$$

with $W^{(l)} = W^{(l)}(c, K; T)$; here

$$N_c^{(l)} \propto \exp(-\beta W^{(l)}).$$

And, in general,

$$\sigma_p^{(2)} (N_c^{(2)} W_h^{(2)}(0))^{-1} \propto \exp(-\beta W^{(2)}).$$

B. In general, the characteristic shape of the absorption band associated with $\bar{\sigma}_p(\omega)$ by a standard specimen (for $\omega > 2kT$ and $\omega \gg D_{cl}$, both for $T > T_0$ and for $T < T_0$) is obtained from the expansion of the ω- "distribution" $W_h(\omega)$ in its semi-invariants λ_ν, $\nu = 1, 2, \dots$ (for $D_{cl} \ll \{(\mathcal{E}kT_0)^{1/2}\}$)[13]

$$\bar{\sigma}_p(\omega) \propto \omega^{-1}\chi(x), \quad \chi(x) \simeq G - \frac{\lambda_3}{6\lambda_2^{3/2}}\frac{d^2G}{dx^2} + \frac{\lambda_4}{24\lambda_2^2}\frac{d^4G}{dx^4} + \frac{\lambda_3^2}{72\lambda_2^3}\frac{d^6G}{dx^6}, \qquad (3)$$

where $G \equiv G(x) = e^{-x^2/2}$ and $x \equiv (\omega - \bar{\omega}_0)D^{-1}$, with

$$\bar{\omega}_0 \equiv \omega_0 + \chi D_{cl}\big|_{\chi\sim 1} \simeq \omega_0;$$

$$\lambda_3\lambda_2^{-3/2} \simeq (2\Phi_0)^{-1/2}(\operatorname{th} T_0/T)^{3/2}, \quad \lambda_4\lambda_2^{-2} \simeq (2\Phi_0)^{-1}\operatorname{th} T_0/T;$$

$$\lambda_2 = D^2 \simeq 4\mathcal{E}\omega_p \operatorname{cth} T_0/T,$$

and for $\Phi_0 \gg 1$ formula (3) is adequate, at least in the region of width $2D$ of the band peak for $|x| \lesssim 1$—at $T \leq \mathcal{E}/2k$ (cf. [1,8]) $\bar{\sigma}_p(\omega)$ has a broad ($2D > kT + \omega_p + D_{cl}$), but distinct ($2D < \omega_0$), peak at $\omega \simeq \omega_0 \equiv 4\mathcal{E}$ with a shape the closer to Gaussian the higher T is (similarly for m.p., see [1,2,8,9,17]). Owing to concentration broadening, for $l = 1$ the peak frequency (and effectively its width) may depend on c, while

$$\bar{\sigma}_p^{(2)}(\bar{\omega}_0) = \bar{\sigma}_p^{(2)}(\bar{\omega}_0; c, K) \propto (\Delta_0^{(2)}(c))^2.$$

C. It turns out that a strong (constant) electric field E may here (for $\Phi_0 \gg 1$) play a role analogous to that of the absorbed quantum ω, which activates (for $T < \mathcal{E}/2k$ and $\omega \simeq \omega_0$) the jumps. This is connected with the fact that, when the interaction $\hat{V}$ of local polarons with the field $\bar{E} \equiv E_x$ is taken into account, $\hat{V} = eE\hat{x}$ and $\hat{\mathcal{H}} \to \hat{\mathcal{H}} + \hat{V}$, the Hamiltonian retains its structure, with

$$\varepsilon(s) \to \varepsilon(s) + eEs_x, \quad \Delta_e(s_{12}) \to \Delta_{12}(E) \equiv \Delta_e(s_{12}) + eEx_{12}$$

for $x_{12} = \langle s_1|x|s_2\rangle(1 - \delta_{s_1 s_2})$. In this case one can in an analogous way (see § 1 and [1,2]) explicitly calculate $\sigma_p(E)$ also in a certain range of non-ohmic fields

$$E \equiv \nabla\varphi(\mathbf{r}) \quad (E < E_m \equiv E_m^{(l)}),$$

if it is assumed that $\sigma_p(E)$ is determined by the Kubo formula for σ_{xx}, in which the current correlator $\operatorname{Re}\langle \hat{j}_x \hat{j}_x(t)\rangle$ is modified in the sense that

$$\hat{j}_x(t) = \exp[it(\hat{\mathcal{H}} + \hat{V})]\hat{j}_x \times \exp[-it(\hat{\mathcal{H}} + \hat{V})]$$

with the contribution of $\hat{V}(\propto E)$ taken into account. This apparently follows if, in considering

$$\sigma_p(E) \equiv \sigma_p^{(l)}(E),$$

one applies method [15] for not too strong "non-ohmic" fields

$$E < E_m \equiv E_m^{(l)} \ (\equiv \delta\varphi/r_m),$$

for which the characteristic length

$$\Lambda_0(\sim |\Delta\varphi/\varphi|) > r_m > r_0.$$

In this case (allowing for the replacements mentioned in (1)) the inequalities from § 1,B are still adopted as criteria of the theory (for $E < E_m$), and in the formula for $\sigma_p(E)$, analogous to (2), $eE(s_{12})_x$ plays the role of the "external" frequency ω.

The region of non-ohmic behavior occurs for

$$E > E_0 \equiv E_0^{(l)} = 2kT/|e|r_0,$$

where $E_0^{(1)} \gg E_0^{(2)}$ for $c \ll 1$, and one may assume that for $kT \ll \mathcal{E}$ usually $E_m \gg E_0$. The principal result is that, for all the T considered and for $E_0 < E(< E_m)$, at least for

$$|x_E| \equiv |x_E^{(l)}| \lesssim 1,$$

$$\sigma_p(E) \equiv \sigma_p^{(l)}(E) \propto \Delta_0^2(E)\chi(x_E)E^{-1}, \tag{4}$$

and for $T > T_0$ and $0 < E \lesssim \omega_0 T(|e|r_0 T_0)^{-1}(< E_m)$

$$\sigma_p^{(1)}(E) \simeq \sigma_p^{(1)}(E=0)\,\frac{\operatorname{sh}\left(\frac{1}{2}\beta|e|Er_0^{(1)}\right)}{\frac{1}{2}\beta|e|r_0^{(1)}E}\exp\left[-\frac{(eEr_0^{(1)})^2}{16\mathcal{E}kT}\right],$$

$$\sigma_p^{(1)}(E)\Big|_{E>E_0^{(1)}} \propto \frac{\Delta_0^2}{E}\,G(x_E), \tag{5}$$

where $x_E \equiv (|e|Er_0 - \bar{\omega}_0)D^{-1}$, and for real $\omega_0 \equiv 4\mathcal{E}$ one may put $E_m > \omega_0/|e|r_0$. As follows for $T < \mathcal{E}/2k$, $\sigma_p(E)$, from (4)–(5), has a characteristic "resonance" peak at $eEr_0 \approx \omega_0$, with a shape close to Gaussian; moreover, as for $\sigma_p(\omega)$, this peak is broad (and it is easier to observe it for $T \lesssim T_0$). In particular, what has been said, together with (4), (5), for $l = 1$ describes $\sigma_p^{(1)}(E)$ for s.p. in an ideal lattice (for $kT \gg \Delta_e e^{-\Phi_0}$ and $D_{cl}^{(1)} = 0$), also for $E > E_0^{(1)}$; formula (5), for $D_{cl}^{(1)} = 0$, is analogous to the formula obtained in ([16]) for $T > T_0$ (for $p^{(1)} < p_0^{(1)} \ll 1$) in Holstein' s quasiclassical scheme ([6]), and the "quasiclassical" region E, apparently (see above), is applicable also outside this region, in particular for $|e|Er_0^{(1)} \approx \omega_0 \equiv 4\mathcal{E}$ for $kT \lesssim \mathcal{E}$.

§ 3. For $p \ll 1$, to calculate the thermoelectric power γ_p (neglecting phonon drag) from the Kubo formulas, as in ([10,2,1,12]), the canonical energy-current operator of the system is used in the form $\hat{I}_x = \hat{I}_x^{(el)} + \hat{I}_x^{(ph)}$, where its phonon part $\hat{I}_x^{(ph)}$ has the standard form (and is not written explicitly in [10,2]); I_x is $\hat{I}_x^{(el)}$, while

$$\hat{I}_x^{(el)} = (2e)^{-1}\{j_x, \mathcal{H}_c + \mathcal{H}_{int}\}$$

for $\Phi_0 \gg 1$; $\{a, b\} \equiv \frac{1}{2}(ab + ba)$. Similarly, energy transfer has mainly the character of convection (without the kinetic contribution), so that γ_p is isotropic in the general case, i.e. $(\gamma_p)_{\mu\nu} \simeq \gamma_p \delta_{\mu\nu}$, and [1a]

$$\gamma_p \simeq (eT)^{-1}(U_0 - \bar{\zeta}), \qquad U_0 = (u) + x_0 I_p\big|_{x_0 \sim 1}, \qquad (u) \equiv \varphi_p^{(1)}(0)/\varphi_p^{(0)}(0). \quad (6)$$

For $D_{cl}^{(1)} \ll \{\Delta_e^{(1)}, kT; \omega_p\}$, formulas (2)–(6) actually describe $\bar{\sigma}_p(\omega)$, $\sigma_p(E)$, and γ_p for s.p. (including degenerate ones) in an ideal lattice; for $p < p_0^{(1)}$, i.e. in the absence of degeneracy,

$$\gamma_{0p}^{(1)} \simeq -\frac{\bar{\zeta}}{eT} = \frac{k}{e} \ln \frac{2N_0}{N_i^{(1)}}$$

cf. [2,1,10–12,16,17]*.

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Received
9 X 1968

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* See also § 11.1 of ([1a]) and footnote 5 in ([17]).

** These contain the relevant references on the theory of transport by small polarons in an ideal lattice.

Note: Figure translations are in progress. See original paper for figures.