QUANTUM THEORY OF THE NONSTATIONARY CONDUCTIVITY OF POLARONS IN SOME DISORDERED SYSTEMS\

PHYSICS

1970

SovietRxiv

Abstract

Full Text

UDC 539.2

PHYSICS

M. I. KLINGER

QUANTUM THEORY OF THE NONSTATIONARY CONDUCTIVITY OF POLARONS IN SOME DISORDERED SYSTEMS*

(Presented by Academician S. V. Vonsovskii, 29 XII 1969)

In the present communication a theory is given of the nonstationary conductivity $\sigma(\omega)$ of polarons in a homogeneous (weak) electric field $E \equiv E_x \propto e^{i\omega t}$ (polaron absorption of electromagnetic waves) in a three-dimensional, spatially homogeneous disordered system consisting of spatially ($\mathbf{R}_i$) and energetically (ε_i) inequivalent localization centers for polarons (in states $|i\rangle$). We have in mind such systems, for example, at comparatively low concentrations N of centers and temperatures T (and appropriate other parameters; see ([1])), in which $\sigma \equiv \sigma(0)$ is determined by nonadiabatic jumps of polarons over distances $R \equiv R_{ij} \equiv |\mathbf{R}_i - \mathbf{R}_j|$ between nearest centers. At the mean distances under consideration, $r \equiv (3/4\pi N)^{1/3} \gg r_B$ (r_B is the characteristic Bohr radius of the electron at the center), the initial "small" quantity is the electronic resonance integral $\Delta_e(R) \propto \exp(-R/r_B)$, while the parameters of the (electron-phonon) coupling $\Phi_0 \equiv \Phi(0)$ and $\Phi \equiv \Phi(T)$ are practically independent of r (of R). As in ([1,2]), a polaron is understood in the generalized sense as a quasiparticle of electron (hole) type plus lattice deformation (i.e., an electron plus the phonons (bosons) of any branch bound to it), both for weak ($\Phi_0 \equiv \Phi(T = 0) \ll 1$) and for strong ($\Phi_0 \gg 1$) coupling, including small polarons for $\Phi_0 \gg 1$ and $r_B < a$ (a is the lattice constant of the regular lattice). An example of such processes is impurity conduction—the theory and model of similar systems and processes were discussed in ([1,4,5]). The criteria for $\sigma \equiv \sigma(\omega = 0)$ are given in ([1,5]) in the form $\Delta_{av} \ll A$. For $\omega \gg A$, it is possible that for $\sigma(\omega)$ they have the form $\Delta_{av} \ll \omega$, broadening the region (so that, possibly, N_{cr} increases with ω).

1. The basic considerations are as follows. Processes of two principal types determine $\sigma(\omega)$: 1) jumps along "conducting chains" through the entire system—the "normal" conductivity $\Sigma(\omega)$—is determined (in the Kubo formula) by the Fourier transform $K_{vv}(\omega)$ of the time (t) correlator $K_{vv}(t)$ of polaron velocities (v_x): $\Sigma(0) = \sigma(0) \equiv \sigma$; 2) local polarization currents in separate clusters $z(\geq 2)$ of centers—the "polarization" conductivity $S(\omega)$ is determined by the Fourier transform $K_{pp}(\omega)$ of the correlator of dipole moments ($\hat{p}_x$): $S(0) = 0$. The contribution $S(\omega)$ is substantial if, in the

states $|k\rangle$ of a polaron in a cluster with energies $U^{(k)}$ $(k = 1, 2, ...)$, there exist "large" dipole moments $p_{kk'} \equiv \langle k|\hat{p}_x|k'\rangle \sim eR$ (of zeroth order in the "small" $\Delta_e(R)$). In the systems under consideration with hopping conductivity, when the width D of the band (D) of fluctuations of the levels ε_i and $\omega_{ij} \equiv \varepsilon_i - \varepsilon_j$ is sufficiently large, i.e. $|\omega_{ij}| \gg \Delta_e(R_{ij})$ and $D \gg \Delta_e(r)$, $P_d^2 \sim (eR)^2 \gg P_{nd}^2$; P_d and P_{nd} are the characteristic diagonal (in k) and off-diagonal dipoles.

---

* Some of the principal results were briefly presented in a review report delivered by the author in September 1969 at the International Conference on Amorphous and Liquid Semiconductors in Cambridge (England).

The theory is based on an adequate decoding of the Kubo formula for $\sigma(\omega) = \Sigma(\omega) + S(\omega)$, following the approach and methods of [1] (§§ 3, 10). The "normal" conductivity $\Sigma(\omega) \simeq \Sigma^h(\omega) \simeq |e|N_c\mu_0\beta\widehat{W}_h(\omega, T)$ $(|e|N_c\mu_0)^*$ is determined by the effective probability $\widehat{W}_h(\omega, T) = W_h(\omega)E_\beta^{-1}(\omega) \equiv z\Delta_{AV}^2 V_h(\omega)E_\beta^{-1}(\omega)$ of uncorrelated 2-site hops (per 1 sec.); here $\beta \equiv 1/T$; $E_\beta(\omega) = \frac{1}{2}\omega \operatorname{cth} \beta\omega/2$; $\Delta_{AV} \equiv \Delta_{AV}(r) = \Delta_e(R_0)$ and $R_0 \sim r$; $V_h(\omega) \propto \exp(-\beta W_D)$, and the activation energy $W_D(\lesssim D)$ is due to fluctuations $(\bar{\varepsilon}_i)$ (in [1,5] the case $T > D$, $\exp(-\beta W_D)(\simeq 1)$, is not written explicitly). The characteristic features of $\Sigma(\omega; T)$ (activated behavior with T and nonmonotonicity with ω) and their meaning are discussed in [1,2]. In particular, for $\Phi_0 \gg 1$, $T < \omega/2$ and $T < \mathcal{E}/2$, $\Sigma(\omega)$ (for $\omega \sim \omega_0^{ij}$) is determined by the average $\langle ... \rangle_{AV}$ decomposition of $V_h(\omega)$ in its semi-invariants and is a superposition (overlapping for $|\omega_{ij}| \ll \omega_0$) of nearly Gaussian peaks at $\omega = \omega_0^{ij} \equiv \omega_0 + \omega_{ij}$ and $\omega_0 = 4\mathcal{E} \simeq 2\omega_{ph}\Phi_0$: approximately $\Sigma(\omega) \sim |e|N_c\mu_0\Delta_{AV}^2(\omega\delta_0)^{-1}\langle\exp[-(\omega - \omega_0^{ij})^2/2\delta_0^2]\rangle_{AV}$.

2. Generally speaking, $S(\omega) = S^I(\omega) + S^{II}(\omega)$: $S^I(\omega)$ and $S^{II}(\omega)$ correspond, respectively, to polaron motion near a defect (over the nearest $z(\geq 2)$ lattice sites at $R = \text{const}$) and over clusters of $z(\geq 2)$ centers; for $\Omega \equiv |\omega_{ij}/\Delta_e(R_{ij})| \ll 1$, $S^I(\omega)$ is discussed in detail in [2]. (In [2] one may have $P_d^2 \sim (eR)^2$ for $z = 2$, if $\Omega \gg 1$, or for $z > 2$, if $\Omega \ll 1$ and the main level U^1 is degenerate.) Like $S^I(\omega)$, $S^{II}(\omega) = S_h^{II}(\omega) + S_b^{II}(\omega)$ contains contributions $S_h^{II}(\omega)$ of activated hops and $S_b^{II}(\omega)$ of nonactivated "tunneling" (zonal-type) transfer under scattering of polarons by phonons with characteristic relaxation time τ_D (one –when $z = 2$)**.

In the case considered for $S^{II}(\omega)$ $(\Omega \gg 1)$, the estimates and $(\omega; T)$-dependences of $S^{II}(\omega; T)$ are in fact determined by the contribution of individual characteristic pairs $(z = 2)$ of centers, at least if their characteristic sizes $r_{ef} \lesssim r$ or are qualitatively similar (see below). In such a "pair" approximation

$S_b^{II}(\omega) \simeq S_D^{II}(\omega)$ (cf. [2]); $S_D^{II}(\omega)$ corresponds to "tunneling" (not hopping) motion (of "large" dipoles P_d) of polarons in the band of "relaxational" broadening ($\sim \tau_D^{-1}$) of the main level ($U^{(1)}$), so that
$S_D^{II}(\omega) \propto P_d^2$, but
$S_b^{II}(\omega) - S_D^{II}(\omega) \propto P_{nd}^2$ and $P_{nd}^2 \ll P_d^2$. The concentration of such pairs (each containing one of the N_c polarons) $n \propto N_c$: apparently, approximately $n \simeq NN_c$. For $\beta\omega/2 \ll 1$, $S_b^{II}(\omega) \simeq S_D^{II}(\omega)$ is described by a Debye-type formula, which follows from

$$S_h^{II}(\omega) \simeq nE_\beta^{-1}(\omega)\left\langle \sum_2 \sum_{\varepsilon_1\varepsilon_2} \chi_D^{(12)} P_d^2(\omega_{12}) \left(\mathrm{ch}\,\frac{\beta U^{12}}{2}\right)^{-2}\right\rangle_{AV}, \qquad \chi_D^{(12)}(\omega) \equiv \frac{\omega^2\tau_D^2(\omega)}{1+\omega^2\tau_D^2(\omega)}; \tag{1}$$

$$S_b^{II}(\omega) \simeq e^2 nE_\beta^{-1}(\omega)\left\langle \sum_2 \bar{\chi}_D^{(12)}(\omega)\, G(|\omega_{12}| < D_0;\, R_{12}) R_{12}^2\right\rangle_{AV},$$

$$\bar{\chi}_D^{(12)}(\omega) \equiv \{\chi_D^{(12)}(\omega) \quad \text{for} \quad |\omega_{12}| = D_0 < \{2T;\, D\}\}, \tag{2}$$

* We denote: N_c is the concentration of current carriers; ω_{ph} and ω_M are the characteristic and maximum phonon frequencies (of the essential branch); (ac) and (opt) are acoustic and optical phonons;
$\omega_{ph}^{(ac)} = \pi s_0(\max\{r_B; a\})^{-1}$,
$\omega_M^{(ac)} \equiv \omega_D = \pi s_0/a$ (s_0 is the speed of sound) in the long-wavelength approximation, quantitatively adequate for $r_B \gg a$;
$\omega_M^{(opt)} \simeq \omega_{ph}^{(opt)} \equiv \omega_{opt}$. The polaron gas is not strongly degenerate and
$T > \Delta_{AV}\exp(-\Phi)$; $\hbar \equiv 1$; $k \equiv 1$.

** If $\Phi_0 \gg 1$, then $S^{II}(\omega) = S_h^{II}(\omega)$ for $T > T_{xx}$ or (and) $\omega > \omega_{xx}$, but $S^{II}(\omega) = S_b^{II}(\omega)$ for $T < T_{xx}$ and $\omega < \omega_{xx}$, where
$T_{xx} < T_x$, $\omega_{xx} \sim \omega_M/2$, and $\tau_D^{-1} \ll \omega_{xx} < \omega_{ph}$;
$T_0^{(opt)} = T_1^{(opt)}(\mathrm{Arsh}\,2\Phi_0)^{-1}$;
$T_1^{(ac)} \simeq T_0^{(ac)}(2\pi\Phi_0^{1/4})^{-1}$. If $\Phi_0 \ll 1$, then $S^{II}(\omega) = S_b^{II}(\omega)$ at least for $T < T_0$ and $\omega < \omega_{ph}$.

where $U^{12} \equiv U^1 - U^2$; $P_d(\omega_{12}) \equiv |p^{(1)} - p^{(2)}| \sim eR$ for $\Omega \gg 1$; $G(|\omega_{12}| < x;\ R_{12}) = \int_{-x}^{x} d\omega_{12} g_2(\omega_{12}; R_{12})$; g_2 is the distribution of "pairs" (R_{12}) over ω_{12}, so that $G(|\omega_{12}| \le x;\ R_{12}) \approx 1$ for $x > D$; for $x < D$, evidently, for estimates one may take $G(|\omega_{12}| < x) \approx G_0(|\omega_{12}| < x) \equiv x/D$. In (2) it is taken into account that the main contribution to (1) is made by $|\omega_{12}| < \{2T;\ D\}$, since the expression $\chi_D^{(12)}(\omega)P_d^2(\omega_{12})(\mathrm{ch}\,\beta U^{12}/2)^{-2}$ is maximal at $|\omega_{12}| = D_0 < \{2T;\ D\}$.

Estimates of $\tau_D(\omega)$, decreasing with T, give the following (these estimates are valid both for $S_b^I(\omega)$ and for $\sigma_{xx}^b(\omega)$ of small polarons [(1)] in an ideal lattice): 1)

$\tau_D^{-1}(\omega) = W_{sc}(\omega;\ |\omega_{12}| < D_0) \ll \{\omega_{ph}; T\}$ in the region of applicability of the theory (W_{sc} is the probability of polaron scattering per 1 sec.); $\tau_D(\omega \ll 2T) \simeq \tau_D \equiv \tau_D(\omega = 0)$, but $\tau_D(\omega)$ may increase with ω for $\omega > 2T$ (for $\Phi_0 > 1$ this is substantial only if also $\omega_{xx} > 2T$); 2) for $\Phi_0 \ll 1$, $\tau_D^{-1}(\omega) \approx W_h(\omega;\ R = R_{12})/z \propto \Delta_e^2(R_{12})$, but for $\Phi_0 \gg 1$ usually $\tau_D^{-1}(\omega) \gg z^{-1} W_h(\omega;\ R = R_{12})$ and $\tau_D^{-1}(\omega) \propto \Delta_e^4(R_{12})$; 3) for $\Phi_0 \ll 1$ and $T < T_0^{(ac)}$, for (ac) $\tau_D^{-1} \propto T$ (or $\tau_D^{-1} \propto T^\alpha,\ 0 < \alpha \leq 1$) for 1-phonon scattering (not too small ω_{ph}/D), but $\tau_D^{-1} \propto T^3$ for 2-phonon scattering; 4) for $\Phi_0 \gg 1$ and $T < T_1^{(ac)}$, for (ac): $\tau_D^{-1} \propto T$ (or $\tau_D^{-1} \propto T^\alpha,\ 0 < \alpha \leq 1$) for 1-phonon and $\tau_D^{-1} \propto T^7$ for 2-phonon scattering; 5) for (opt), in general, $\tau_D \propto (\mathrm{sh}\, \beta\omega_{opt}/2)^2$ (for $\omega_M^{(opt)} \simeq \omega_{ph}^{(opt)} \equiv \omega_{opt}$).

Because of the sharp dependence of τ_D on R_{12}, in (2) one may approximate (cf. (3)) $(\omega\tau_D(\omega, R_{12}) + 1/\omega\tau_D(\omega; R_{12}))^{-1} \approx \pi r_R \delta(R_{12} - \rho_0)$ for $\rho_0 \lesssim r$, where $\rho_0 \equiv \rho_0(\omega, T)$ is determined from $\omega\tau_D(\omega, \rho_0) = 1$. This means that (for $\rho_0 \lesssim r$) only selective pairs with $R_{12} = \rho_0(\omega, T)$ determine $S_i^{II}(\omega)$; for example, for $\beta\omega/2 \ll 1$ we have for (ac): $\rho_0/r_B \propto \ln(T/\omega)$ for 1-phonon scattering both for $\Phi_0 \gg 1$ and for $\Phi_0 \ll 1$ (or $\rho_0/r_B \propto \ln(T^\alpha/\omega),\ 0 < \alpha \leq 1$), while for 2-phonon scattering $\rho_0/r_B \propto \ln(T^3/\omega T_{ph}^2)$ for $\Phi_0 \ll 1$, or $\rho_0/r_B \propto \ln(T^7/\omega T_{ph}^6)$ for $\Phi_0 \gg 1$; for (opt), $\rho_0/r_B \propto A - \beta\omega_{opt},\ A = \mathrm{const};\ T_{ph} \equiv \omega_{ph}/2$. However, the contribution of jumps $S_h^{II}(\omega)$ is determined rather by pairs $R_{12} \lesssim r$ and is not so sensitive to the value of R_{12} (since pairs $R_{12} \sim r$ give "large" dipoles $P_d \sim er$), for it contains no sharp R-dependences of the type $\Delta_e(R)$.

3. As a result one can write the following estimates for $S^{II}(\omega)$. In the region $(G_h) \equiv \{T > T_{xx}$ or (and) $\omega > \omega_{xx}$ for $\Phi_0 \gg 1\}$

$$S^{II}(\omega) = S_h^{II}(\omega) \sim e^2 n \omega^2 r_{ef}^5 E_\beta^{-1}(\omega) V_h(\omega, T) \quad \text{for } r (\gtrless) r_{ef} (\gg r_B), \tag{3}$$

so that $S_h^{II}(\omega, T) \propto \omega^2 E_\beta^{-1}(\omega) V_h(\omega, T)$: the behavior of $S_h^{II}(\omega, T)$ with varying ω and T is analogous to the behavior of $\omega^2 \Sigma(\omega, T)$ (see above for $\Sigma(\omega, T)$) and is characteristic of jumps, including an almost Gaussian peak at $\omega \sim \omega_0 = 4\mathcal{E}_\sigma$. In the region $(G_b) \equiv \{\omega < \omega_{xx}$ and $T < T_{xx}$ for $\Phi_0 \gg 1,$ or $\omega < \omega_{ph}$ and $T < T_0$ for $\Phi_0 \ll 1\}$

$$S^{II}(\omega) = S_b^{II}(\omega) \sim \pi e^2 r_B n \omega \rho_0^4 E_\beta^{-1}(\omega) G(|\omega_{12}| < D_0; \rho_0) \quad \text{for } r \gtrsim \rho_0 (\gg r_B). \tag{4}$$

Hence, taking into account the estimates for $\rho_0(\omega, T)$ and G (for simplicity $G \approx G_0$), the ω- and T-dependences for $S^{II}(\omega, T)$ in (G_b) follow, for $\omega_{\min} < \omega \ll \omega_{\max}$ and $T'_{\min} \ll T < T_{\max}$*, when $r \gtrsim \rho_0 \gg r_B$. (For $\Phi_0 \gg 1$, the conditions $T < T_{\max}$ and $\omega < \omega_{\max}$ are usually inessential, since $\omega_{xx} < \omega_{\max}$ and $T_{xx} < T_{\max}$–the region of such ω and T that $r \gtrsim \rho_0 \gg r_B$ is relevant experimentally.) Thus in (G_b): 1) $S^{II}(\omega, T)$ grows with T: for $D \gg T$, $S^{II}(\omega, T) \propto \rho_0^4(\beta E_\beta(\omega))^{-1}$; in particular, this growth is weak (almost logarithmic, weaker than $\sim T$) for

* A preliminary analysis of $S_b^{II}(\omega)$ for $\rho_0 \gtrsim r$ suggests that, probably, also in this case the essential clusters of centers can be treated as a set of effective pair clusters with $R_{12} \sim r$. If this is so, the estimate (4) for $\rho_0 \gtrsim r$ is modified only by the replacement $\pi r_B \rho_0^4 \to \omega r^5$ and ($\rho_0 \to r$ in G).

$\omega < 2T$ and under scattering (especially one-phonon scattering) by acoustic phonons (ac); 2) $S^{\text{II}}(\omega, T)$, generally speaking (at least for $\omega \ll 2T$), increases with ω, $S^{\text{II}}(\omega) \propto \omega\rho_0^4 E_\beta^{-1}(\omega)$, or, approximately, $S^{\text{II}}(\omega) \propto \omega^s$ for $0 < s \equiv s(\omega, T) < 1$, where s decreases (logarithmically) with ω/T, but possibly also $s \approx 0$ (a plateau) for $\omega \gtrsim 2T$ (and even a decrease with ω, $s < 0$, for $\omega \gg \omega_1 > 2T$ and $\Phi_0 \ll 1$, with $\omega_1 \gg \tau_D^{-1}$, but apparently $\omega_1 < \omega_{ph}$).

In general, taking account of $S^{\text{I}}(\omega)$ from [2], we have: 1) for $\Phi_0 \gg 1$, $S(\omega)$ increases with ω up to an almost Gaussian peak at $\omega \sim \omega_0$: as ω^s in (G_b) (with $0 \le s < 1$, or $s = 2$, and (for $(\omega\tau_D) > 1$) $s = 0$ for the cases and frequencies for which $S(\omega) = S^{\text{II}}(\omega)$ or $S(\omega) = S^{\text{I}}(\omega)$, respectively), but more strongly (see above and [1,2]) in the region (G_h); 2) for $\Phi_0 \ll 1$ and $\omega < \omega_1$, $S(\omega)$ does not decrease with ω and increases as ω^s ($0 \le s < 1$ or $s = 2$) for $\omega < 2T$. In contrast to $\Sigma(\omega)$, $S(\omega)$ does not contain a strong dependence $\sim \Delta_A \nu^2(r)$, and in (G_b) it likewise does not contain a temperature activation factor: $S(\omega) \propto T^\alpha \to 0$ as $T \to 0$, $\alpha \sim 1$ (cf. [3,4]). Qualitatively, the graph of $S(\omega)$ is analogous to Fig. 1 in [2] (to Fig. 4 in [2] and Fig. 2 in [1] for $T < T_{xx}$, there may be either $\sigma \equiv \sigma(0) < \sigma(\omega_0)$, or $\sigma > \sigma(\omega_0)$, depending on T and other parameters).

In view of the similarity of the behavior of $S(\omega, T)$ in (G_h) and $\Sigma(\omega, T)$ (more precisely, $\omega^2\Sigma(\omega, T)$), the behavior of $\sigma(\omega, T)$ for $\Phi_0 \gg 1$ is also qualitatively analogous, independently of the magnitude of $\Sigma(\omega)/S(\omega)$ ($\gtrless 1$), although in (G_b), for $\omega < 2T$, the parameter s may vary as indicated. In fact, $\sigma(\omega) \simeq S(\omega)$ for $\omega > \Delta_A \nu$, at least, but in (G_b) in reality also at lower $\omega > \omega_2$, where $\omega_2 < \{\Delta_A \nu; \tau_D^{-1}\}$, with $\Sigma(\omega_2) = S(\omega_2)$. For $\omega < \omega_2$ (in (G_b)) or $\omega < \Delta_A \nu$, $\sigma(\omega) \simeq \Sigma(\omega)$, and for $\omega < 2T$, $\sigma(\omega) - \sigma \propto \omega^{s=2}$ [1]. For $\Phi_0 \ll 1$, $\sigma(\omega) \simeq S(\omega)$, and $S(\omega)$ determines the behavior of $\sigma(\omega)$ for $\omega > \omega_2$.

For $\Phi_0 \ll 1$ and $\omega \ll 2T$, the formulas following from the theory, $S^{\text{II}}(\omega) = S_D^{\text{II}}(\omega)$, are analogous (in justification and range of applicability) to those proposed in [3] for $S_D^{\text{II}}(\omega)$ (on the basis of the Debye formula for dielectric losses), but with the interpretation of $S_D^{\text{II}}(\omega)$ as the conductivity of the tunneling (not hopping) type in the sense that the correlator $K_{pp}^b(t) \propto \exp(-t/\tau_D)$ for $t \sim \tau_D \gg \beta$, whereas the behavior of the hopping correlator $K_{pp}^h(t)$ at substantially small $t \ll \beta$ is essentially different [1]. This distinction, and the distinction between $S_h^{\text{II}}(\omega)$ and $S_b^{\text{II}}(\omega)$ for $\Phi_0 \gg 1$, is essential for a correct calculation of $S(\omega)$ for $\Phi_0 \gg 1$ (and, in general, of other kinetic coefficients for $\omega \neq 0$).

Institute of Semiconductors
Academy of Sciences of the USSR
Leningrad

Received
20 XII 1969

REFERENCES

[1] M. I. Klinger, *Reports Progr. Phys.*, **31**, 225 (1968).
[2] M. I. Klinger, E. V. Blakher, *Phys. Stat. Solid.*, **31**, 515 (1969).
[3] M. Pollak, *Phys. Rev.*, **A133**, 564 (1964).
[4] I. G. Austin, N. F. Mott, *Adv. Phys.*, **18**, No. 71, 41 (1969).
[5] M. I. Klinger, DAN, **183**, 311 (1968).

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