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NON-OHMIC HOPPING CONDUCTION - A TREATMENT BY DIRECTED PERCOLATION THEORY

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Abstract.- The hopping current induced by phonons and a strong electric field is discussed on the basis of the rate equation using the directed percolation method. Essential informations about the non-ohmic behaviour can be obtained from the analysis of the vertex star.

Since the famous paper of N. F. Mott /1/ the problem of hopping conduction in disordered semiconductors is under discussion until now. In performing the hopping transport theory an important point has been the application of the percolation analysis for describing the stochastic behaviour of localized electrons interacting with phonons in the presence of an external electric field /2, 3/. When the field is sufficiently low there works the linear theory. The conductivity is field independent and may be expressed by the two particle equilibrium Greens function of the disordered system. In the non-ohmic region the charge transport has to be discussed under conditions of a non-equilibrium state and directed percolation. The macroscopic density being the self averaging quantity in our problem can be written as follows /4/

\[ \mathbf{J}(t) = \lim_{\alpha \to \infty} \frac{1}{2\alpha} \sum_{i,j} (R_i - R_j) \mathbf{I}[i,j] \]  

(1)

with

\[ \mathbf{I}[i,j] = \frac{1}{\Omega} \frac{e}{2} \left[ \tilde{\mathbf{W}}_{ij} f_i (1-f_j) - \tilde{\mathbf{W}}_{ij} f_j (1-f_i) \right] \]  

(2)

as the net pair current between the centres \( i \) and \( j \) in the four-dimensional energy-coordinate space, \( i = (E_i, \mathbf{R}_i) \). The expression (2) results from the well-known rate equation for the non-equilibrium distribution function \( f_i(t) \) of the localized electrons. It should be mentioned that tunnel transitions between the localized states due to the action of the electric field alone without participation of phonons are neglected and the Markovian approximation has been used. In our treatment

\[ \tilde{\mathbf{W}}_{ij} = \frac{2\pi}{\hbar} \sum_q |\beta_q| \left| \left[ N_q \left( \tilde{E}_i - \tilde{E}_j + \hbar \omega_q \right) + (N_q + 1) \right] \right|^2 \]  

(3)

is the quantum mechanical transition probability for processes with assistance of one phonon. The phonon ensemble is assumed to be in the equilibrium state, \( N_q = (\exp(\beta \hbar \omega_q) - 1)^{-1}, \beta = (KT)^{-1} \). The tilde used in the formulae (2) and (3) denotes the electron energy renormalization due to the interaction with the local electric field \( \mathbf{E}_i \), \( \tilde{\mathbf{E}}_i = E_i + V_i, V_i = -e \mathbf{F} \cdot \mathbf{R}_i \). Using the dimensionless denotations \( (\beta E_i, \mathbf{R}_i) = (\tilde{E}_i, \tilde{\mathbf{R}}_i) \), the localization radius, we assume the stationary
non-equilibrium distribution function to be of the form

\[ f_i = \left[ \exp \left( E_i - \gamma_i \right) + 1 \right]^{-1} \tag{4} \]

with the total electrochemical potential \( \gamma_i = \beta (\mu - V_i + \delta \mu_i) \). Here \( \delta \mu_i (F_i) \) represents the current induced local change of the chemical potential \( \mu \).

Since the pair currents flow from sites with higher electrochemical potentials to such with lower ones they form a network with directed bonds. In a graph theoretic formulation \( /5/ \) we are concerned with an irregular four-dimensional directed graph \( G = (V, A) \) with the arc set \( A = \{ I(i,j) \} \) and the set \( V \) of the vertices \( x_i \) being incident with \( A \). The arcs are defined by

\[ I[i,j] = \Theta (\eta_{ij}) I(i,j) - \Theta (\eta_{ji}) I(j,i) \quad , \tag{5} \]

where the ordered pair \( (i,j) \) means the direction from \( i \) to \( j \). Using (2) - (5) we find

\[ \sinh Y_2(\eta_{ij}) \exp(-\delta \mu_{ij}) \cosh Y_2(E_i - \delta \mu_j) \cosh Y_2(E_j - \delta \mu_j) \sinh Y_2(E_{ij}) \]

with \( \eta_{ij} = -\eta_{ji} > 0 \) and \( E_{ij} = E_i - E_j > 0 \). Here we point out that a neglect of the bond orientation is only possible in the ohmic region where the percolation model can be related to a network with field-independent conductivities. Evidently the charge transfer in our system is realized along self-avoiding paths \( r(e, e') \) between the vertices \( e \) and \( e' \) at the limiting contacts, \( (e, e') \in E = E \times E' \). Since the electrochemical potential decreases along such a path the graph \( G \) is coverable by the union of self-avoiding paths and does not contain circuits in the stationary case.

The problem of the percolation on directed graphs may be discussed in two ways. Either we ask for the most effective path (first - passage percolation) or for the critical pair current \( I_c \) characterizing the macroscopic current density \( J \).

In the following we sketch the general procedure for finding \( I_c \). Let us denote the operation of deletion of the arc set \( A_I = \{ I(i,j) \} \)

\[ I(i,j) \in A \setminus I(i,j) < I \] by \( D^{AIG} = (V, A - A) \) the critical current may be expressed as

\[ I_c = \inf \{ I : I \in D \} \]

Thus, \( I_c \) is that value of \( I \) for which the percolation cluster \( C_x \) appears. Here a cluster means a maximal current-conducting subgraph always defined with respect to a source vertex \( q \). In general, a cluster \( C_{ab} \) will be a part of a graph component \( K_{a} = U_{b} C_{ab} \). Therefore, the generalization of the cluster binding criterion to the problem of directed percolation has the form

\[ I(i,j) \in C_{ab} \iff I(i,j) \supset I(i,j) \cap r_{ab} (q_{ab}, t_{ab}) \]

\[ \tag{8} \]

Here \( t_{ab} \) is the terminal vertex of a self-avoiding path \( r_{ab} \) rooted in \( q_{ab} \) with \( C_{ab} = U_{t_{ab}} r_{ab} (q_{ab}, t_{ab}), \{ t_{ab} \} = T_{ab}, T_{ab} = \{ q_{ab} \} x T_{ab} \).

Essential information about the path formation and the growth and the unification of clusters former being isolated one obtains by considering the vertex star for a fixed \( i \)

\[ S_{i}^{1} = S_{out}^{1} I U S_{in}^{1} I ^{1} \cap S_{out}^{1} I = ( \{ x_i \}, \emptyset ) \]

\[ \tag{9a} \]
where \( i.e. \)

\[
\mathbf{s}_{\text{out},I}^{i} = (a_{\text{out},I}^{i}, a_{\text{out},I}^{i}), \quad a_{\text{out},I}^{i} = \{ I(i,j) \mid I(i,j) \in \mathbf{A} / I(i,j) \geq I \}.
\]  

(9b)

Our primary interest is in the energetic and spatial distribution of the out- and in-valences depending on the parameters \( I, F \) and \( \Delta \mu_{ij} \). One example of such an investigation found by computer analysis is shown in Fig. 1.

Fig. 1. Vertex star for \( (E_{i} - \mu) / kT = -5, \)

\( eF/2kT = 0.5, \quad \Delta \mu_{ij} / kT = 0.2 \)

Summarizing these results we emphasize that
- for \( E_{i} < \beta \mu \) the out- and in-valences are maximal and they vanish for \( E_{i} < \beta \mu, \quad E_{i} < \beta \mu \),
- if the electric field strength \( F \) is increased, the out- and in-valences are also increased and the critical pair current \( I_{c}(F) \) increases monotonously and non-linearly, furthermore, the spatial projections of the out- and in-valences are preferentially oriented in the direction of the electric field, the connectedness of the current network is maximal in this direction, the paths are smoothed and their mean length decreases,
- for increasing field strength the charge transfer in the pair currents is predominantly assisted by the emission of phonons,
- for \( \Delta \mu_{ij} > 0 \) there arise diffusion currents opposite to the electric field, complicating the path topology and lengthening the paths.

The critical cluster \( C_{p} \) is essentially characterized by the averaged out- and in-valences of its vertices,

\[
\mathcal{V}_{\text{out}}^{\text{dir}} = \langle |a_{\text{out},I_{c}}^{i}| \rangle_{C_{p}}, \quad \mathcal{V}_{\text{in}}^{\text{dir}} = \langle |a_{\text{in},I_{c}}^{i}| \rangle_{C_{p}}.
\]

(10)

Here the brackets denote the average over all vertices of the critical cluster. The calculation of these parameters of the macroscopic
system requires the solution of the problem of anisotropic, directed percolation on an irregular four-dimensional lattice. This problem has not been solved as yet and there exist only few informations concerning directed percolation. In particular, it holds

$$\gamma_{\text{dir c, out}} + \gamma_{\text{dir c, in}} \geq \gamma_{\text{c}}, \quad (11)$$

where $\gamma_{\text{c}}$ is the mean valence of a vertex in the critical cluster of the corresponding problem of undirected percolation. Furthermore, one has to note that the critical behaviour for $I \rightarrow I_c$ here belongs to an other universality class as for the corresponding model of undirected percolation /6/. It is to expect that the critical exponents of such model parameters as the percolation probability, the mean cluster size and others yield a non-analytical behaviour of thermodynamic parameters weaker than in the undirected case. Taking into account the critical parameter (11) the non-ohmic dependence of the hopping current density (1) on the electric field is estimated to

$$J(F) \sim \exp \left[ -a_{\text{c}}^{\text{dir}} \left( \alpha^{-3} kT N_0 \right)^{\gamma_4} + \frac{e^2 f_2}{b_{\text{c}}^{\text{dir}}} \frac{\left( \alpha^{-3} kT N_0 \right)^{\gamma_4}}{e^2 kT N_0} \right] \frac{e F}{2 kT} \ll 1, \quad (12a)$$

$$J(F) \sim \exp \left[ -c_{\text{c}}^{\text{dir}} \left( N_0 eF \right)^{-\gamma_4} \right], \quad \frac{e F}{2 kT} \ll 1, \quad (12b)$$

$$J(F) \sim \exp \left[ -d_{\text{c}}^{\text{dir}} \left( N_0 eF \right)^{\gamma_3} \right], \quad \frac{e F}{2 kT} \gg 1, \quad (12c)$$

in the cases of (a) weak, (b) intermediate and (c) strong electric fields.

The expressions (12a-c) show both the exponential $F^2$- and $-F^{-\gamma_4}$-behaviour for stronger fields /7, 8/. The latter is connected with the mechanism of spontaneous phonon emission. The saturation effect in the case of very strong electric fields results from the assumption of a band of localized states of the width $\Delta E$. The density of states $N_0$ is assumed to be constant in the vicinity of the Fermi level. The parameters resulting from the percolation analysis are of the order $a_{\text{c}}^{\text{dir}}, c_{\text{c}}^{\text{dir}}, d_{\text{c}}^{\text{dir}} \sim 1$ and $b_{\text{c}}^{\text{dir}} \sim 10$.

Obviously the field dependence of the current density depends sensitively both on the chosen model for the current conducting hopping system and the approximations for the non-equilibrium distribution $f_i$ and the transition probabilities $W_i$. Correlations in the site distribution and population may essentially influence the non-ohmic behaviour and have to be also discussed in more detail /3/.

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