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Motion of localized carriers in a strong electric field

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Abstract. Localized carriers can contribute to charge transport in a weak electric field through thermally activated hopping. If the electric field is sufficiently strong, the potential barrier vanishes and the transport acquires band-like character. In the case of small polarons this corresponds to their dissociation. We propose a phenomenological model which describes this transition. This model is based on the assumption that charged classical particles, which are localized in a potential well, can be thermally excited by phonons, and dissipate their acquired kinetic energy. Taking the particles' inertia into account we find a decrease in the strength of the above-mentioned transition field and a hysteresis in the current–voltage relation for certain parameter ranges.

1. Introduction

During the last two decades, the investigation of the localization behaviour of electrons in strong electric fields has received a great deal of attention, both theoretically and experimentally. In the theory of weak localization, self-consistent equations of the Vollhardt–Wölfle type are derived [1–8] as well as exact solutions for one-dimensional (1D) systems [9]. The transport in systems with strong localization is dominated by hopping mechanisms. This includes Anderson-localized carriers in disordered systems [10–16] as well as small polarons in crystals [17, 18], which is the focus of our paper.

The influence of the electric field F on the current density j in the hopping regime is generally taken into account by decreasing the potential barrier by a value of the order of $eF\bar{R}$, where $\bar{R}$ denotes the hopping distance. In this case the hopping probability is proportional to $\exp(ceF\bar{R}/kT)$ (c is a numerical coefficient). The main difficulty for disordered systems consists in applying an averaging procedure (for details see [19]).

For small polarons in crystals, one obtains as the current–voltage characteristic in a simple two-site approximation [17]

$$j \propto \sinh\left(\frac{eF\lambda}{2kT}\right)\exp\left(-\frac{E_a}{kT}-\frac{(eF\lambda)^2}{16E_akT}\right) \tag{1}$$

where λ is the lattice constant and E_a the activation energy for hopping of a small polaron. Equation (1) is valid only for fields that are not too high, namely for $F \leqslant F_c'$, where

$$F_c' = 4E_a/e\lambda. \tag{2}$$

In a larger field, the hopping barrier vanishes and band-like transport sets in, superseding the hopping transport. Strictly speaking, at $F = F_c'$ the polaronic state will dissociate, i.e. the electron will lose its cloud of phonons. The question of the dissociation of small polarons

in an electric field was experimentally investigated in [20]. The materials $Co_{1+x}Cr_{2-x}O$ and $Co_{1-x}Li_xO$ were found to possess a discontinuity (breakdown) in the current–voltage relation at a threshold field strength $F_c' = 10^4$–10^5 V cm^{-1}, i.e. a transition from a state of low conductivity to a state of high conductivity. If the field is lowered again, the low-conductivity state is regained only after a field $F_c < F_c'$ is reached, i.e. the transition between the two conductivity states shows a hysteretic behaviour. A similar effect has been observed in the amorphous oxides Ta_2O_5 and Nb_2O_5 [21].

The attempt to create a rigorous microscopic theory of charge transport in the model of small polarons including their dissociation proved to be complicated [18], since one has to describe the transition between localized electronic states and plane waves. In [18] it was noted, that, as the hopping distance of the polaron increases with the field, the task changes from a two-site to a multi-site one. In this context one has also to note the series of studies (see, e.g., [22–25]) concerning the instability of the large polaron in an electric field. If the field is increased in a system of large polarons so that the drift velocity reaches a critical value (of the order of the sound velocity), then the interaction of a classical particle with the phonons becomes impossible. This effect can be interpreted as the dissociation of the polaron state.

In this paper we propose a simple phenomenological model which allows us to describe the crossover between hopping and band-like transport in an electrical field. The model is based on the picture of dissipative motion of a charged classical particle in a lattice potential, comprised of potential barriers between neighbouring sites, under the influence of an electric field. In a sufficiently weak field, the presence of these potential barriers leads to the localization of the particles on the lattice sites. They can be spatially displaced only by a random force (interaction with phonons). This transport mechanism has hopping-like character. At $F = F_c'$, the potential barrier vanishes, and the motion of the particle becomes unbounded. The velocity does not diverge due to energy dissipation and the transport acquires band-like character, described by the classical Drude formula. This model, despite its extreme simplicity, allows us to describe both limiting cases—the hopping transport and the band-like motion—and the crossover between these two cases. This model reminds us of the method used to describe the dynamics of vortices in Josephson-junction arrays of type II superconductors [26, 27]. Although the main concern of this paper is the problem of the dissociation of the small polaron in an electric field, the results obtained are probably applicable to other systems with strong localization and activated transport as well (e.g. disordered media with Anderson localization).

The primary result of this paper is the description of the transition between the low- and high-conductivity states at a certain critical field F_c, which for dissipation that is not too large and a large mass of the localized state can be considerably weaker than the critical field F_c', given by (2). This effect is due to the fact that at non-zero temperature the dissociation of the polaron state takes place at a field for which there are still potential barriers between the sites, but they are overcome by the particle due to its inertia. As in the case for Josephson vortices, this crossover is accompanied by hysteretic effects, which are especially pronounced at very low temperatures.

2. The phenomenological equation of motion

The principal idea of the method set forth below consists in the introduction of a time-dependent collective coordinate for the centre-of-mass motion of the localized particle $\boldsymbol{R}(t)$. This approach is also used to describe the motion of large polarons in strong electric fields [24, 25]. In this case, the collective coordinate is usually introduced by taking the

classical limit of the equation for the Wigner distribution function. We will use the classical description right from the beginning and determine $\boldsymbol{R}(t)$ in the following way. We consider the Hamiltonian H of an electron–phonon system, and restrict ourselves to the adiabatic approximation (i.e. we neglect the kinetic energy of the lattice subsystem). Furthermore, we only consider the one-electron and one-band approximation. This means, in particular, that the occupation numbers n_m of the sites m obey the relation

$$\sum_m n_m = 1. \tag{3}$$

Now one has to minimize the total energy corresponding to H for a given value of the centre-of-mass coordinate

$$\boldsymbol{R} = \sum_m \boldsymbol{R}_m n_m \tag{4}$$

where $\boldsymbol{R}_m$ denotes the radius vector of site m. Technically, the constraints (3) and (4) can be taken into account, e.g., by introducing Lagrange factors—the chemical potential μ and the 'electric field' $\boldsymbol{\Phi}$. We denote the quantity obtained by this procedure by $\tilde{U}(\mu, \boldsymbol{\Phi})$:

$$\tilde{U}(\mu, \boldsymbol{\Phi}) = \langle H \rangle - \sum_m (\mu + e\boldsymbol{\Phi}\boldsymbol{R}_m)\langle n_m \rangle. \tag{5}$$

Using the relations

$$\frac{\partial \tilde{U}(\mu, \boldsymbol{\Phi})}{\partial \mu} = 1 \qquad \frac{\partial \tilde{U}(\mu, \boldsymbol{\Phi})}{\partial \boldsymbol{\Phi}} = e\boldsymbol{R}$$

we obtain the quantities μ and $\boldsymbol{\Phi}$ as functions of $\boldsymbol{R}$. This allows us to express the adiabatic term $\tilde{U}(\boldsymbol{R})$ of our electron–phonon system as a functional of the centre-of-mass coordinate:

$$\tilde{U}(\boldsymbol{R}) = \tilde{U}\,[\mu(\boldsymbol{R}), \boldsymbol{\Phi}(\boldsymbol{R})] + \mu(\boldsymbol{R}) + e\boldsymbol{R}\boldsymbol{\Phi}(\boldsymbol{R}).$$

This programme, in particular, is performed in [28] for a two-site model of small polarons. Finally, the quantity obtained, $\tilde{U}(\boldsymbol{R})$, has to be minimized with respect to lattice displacements. We denote this minimized energy by $U(\boldsymbol{R})$. This quantity plays the central role in the calculational method proposed below. It describes the potential energy for the motion of a localized charge carrier and is minimal at the points $\boldsymbol{R} = \boldsymbol{R}_0$, where $\boldsymbol{R}_0$ denotes the coordinates of the localization centres.

The implementation of the proposed procedure to determine the effective potential energy is very intricate, as it requires one to solve the Schrödinger equation for an electron in an electrical 'field' $\boldsymbol{\Phi}$ in the presence of arbitrarily large lattice displacements. The appendix gives an example of such a calculation for small polarons for a vanishingly small value of the resonance integral. In [27] a similar calculation was carried out for pinned Josephson vortices. It is more feasible to introduce the potential $U(\boldsymbol{R})$ phenomenologically, provided that certain statistical properties of it are known. For small polarons in crystals, as we will see below, it is sufficient to know two of its properties: owing to the translational symmetry $U(\boldsymbol{R} + \boldsymbol{G}) = U(\boldsymbol{R})$, where $\boldsymbol{G}$ denotes a translation vector of the crystal, the (site-invariant) height of the potential barrier E_a can be introduced. The minima of the function $U(\boldsymbol{R})$ coincide with the lattice sites.

The introduction of the potential $U(\boldsymbol{R})$ permits one to phenomenologically formulate the equation of motion of a localized particle in an external electric field $\boldsymbol{F}$, which with the introduction of a random force gives the form of a Langevin equation:

$$m^\star \ddot{\boldsymbol{R}} + \boldsymbol{f}(\dot{\boldsymbol{R}}) + \frac{\mathrm{d}U(\boldsymbol{R})}{\mathrm{d}\boldsymbol{R}} - e\boldsymbol{F} = \xi(t) \tag{6}$$

where $m^\star$ denotes the effective mass of the particle. The term $\boldsymbol{f}(\dot{\boldsymbol{R}})$ describes the friction for moving in a dissipative medium and its functional form deserves further consideration. We start with the case of a strong electric field:

$$eF \gg \left| \frac{\mathrm{d}U(\boldsymbol{R})}{\mathrm{d}\boldsymbol{R}} \right|$$

i.e. the moving electron does not 'feel' the spatial variation of the potential. In this case the problem considered is equivalent to the problem of large polarons and the corresponding Langevin equation has been investigated in numerous papers [23–25]. If the dissipative bath manifests itself in essentially elastic scattering on impurities, then, to a good degree of accuracy, the friction function is linear in the velocity:

$$\boldsymbol{f}(\dot{\boldsymbol{R}}) \approx \eta \dot{\boldsymbol{R}} \qquad \eta = \frac{m^\star}{\tau} \tag{7}$$

where τ is the elastic relaxation time. Scattering on phonons is more complicated. Here, the utilization of the adiabatic approximation leads to divergencies [29, 30]. The term $\boldsymbol{f}(\dot{\boldsymbol{R}})$ generally has a non-Markovian character, which avoids the divergencies. In classical calculations the Markovian limit is usually taken, if one restricts the considerations to appropriate timescales.

The linear approximation (7) is employed in the investigation of large polarons, which interact with optical phonons of frequency ω_0, in the limit of a small drift velocity $\dot{R} < w$ ($w = \omega_0/k_{max}$, k_{max} being the largest wave vector in the Brillouin zone). For $\dot{R} > w$ the classical limit of the Fröhlich model yields $\boldsymbol{f}(\dot{\boldsymbol{R}}) \propto \ln(\dot{R}/w)$ (see, e.g., [25]). The random forces do not play a significant role in this range of the electric field.

The situation is very different for small polarons in the range of electric fields that are not too strong, when the potential landscape has the determining influence and leads to a hopping mechanism for the transport. But, in the model of small polarons, there is no unambiguous recipe for dividing the force into random (fast) and friction (slow) forces and then obtaining the Langevin equation. The role of 'friction' is played here by thermal relaxation processes of the electron at a site [28]. After all, with the elucidation of this problem, a consequent construction of a theory of small polarons would be within reach. These dissipative processes are caused by the dispersion of the optical phonons $\Delta\omega$. As we will see below, the current–voltage characteristic considered by us is not too sensitive to the actual form of the friction function $\boldsymbol{f}(\dot{\boldsymbol{R}})$. But if one uses the linear approximation, equation (7), then the relaxation time

$$\tau_p \approx \frac{1}{\Delta\omega}\left(\frac{E_a}{kT}\right)^{2/3}$$

has to be invoked [31].

Now we consider the limits of applicability of the adiabatic approximation which lie in the nature of the effective potential obtained, $U(\boldsymbol{R})$. Firstly, the applicability of this approximation is strongly related to the value of the dimensionless parameter

$$\eta_2 = J^2/[\hbar\omega_0(E_a kT)^{1/2}]$$

(J denotes the resonance integral, and ω_0 the frequency of the optical phonons) [28]. The state of adiabatic polarons is realized for $\eta_2 \gg 1$. Secondly, equation (6) has adiabatic character, i.e. the equation assumes that the polaron responds instantaneously to an electron displacement. The response of the phonon subsystem to the electron displacement is determined by the characteristic time [32]

$$\bar{t} = \hbar/[(E_a kT)^{1/4}(\hbar\omega_0)^{1/2}].$$

Thus, the adiabatic approximation is valid only in the limit of a small polaron drift velocity, i.e. for $\dot{R} \ll \lambda\bar{t}^{-1}$. The formation of the potential landscape $U(\boldsymbol{R})$ is more and more delayed for increasing velocity of the polaron, and does not happen at all in the limit $\dot{R} > \lambda\bar{t}^{-1}$. Basically, this corresponds to the dissociation of the polaron state in an electric field.

The central approximation of our paper is the assumption of an adiabatic motion of small polarons. We assume that non-adiabatic level transitions are unimportant, not only in weak electric fields (corresponding to $\eta_2 \ll 1$), but also in strong electric fields. Of course, a possible alternative model of polaron dissociation would be the increase of the transition probabilities between the adiabatic levels [28] with increasing field due to the increment of the polaron velocity near the position of the avoided level crossing. This causes the behaviour of the polaron to become non-adiabatic at a certain value of the electric field. This transition could be responsible for the hysteretic phenomenon at the polaron dissociation (see also section 5).

3. Hopping transport and band-like motion

In the following, we restrict ourselves to the motion of a small polaron in an ordered one-dimensional (1D) chain, where $U(R + \lambda) = U(R)$. According to (6), if the particle is localized at any site before the electric field is applied, then the particle will not move to another site as the electric field is increased in the absence of random forces, as long as the potential barrier is present, i.e.

$$eF \leqslant \max \frac{\mathrm{d}U}{\mathrm{d}R} \approx AE_a/\lambda. \tag{8}$$

Here A is a numerical coefficient, which depends on the actual form of the potential $U(R)$. It is assumed that there is only one characteristic length, namely the lattice constant λ, and only one characteristic energy, the barrier height E_a. The effective potential (A4), which is derived in the appendix, yields $A = 4$. The inequality (8) determines the critical field F_c', equation (2), below which the current vanishes in the framework of (6) (without random forces).

The random force $\xi(t)$ in (6), which is connected with scattering of polarons on phonons, will be described in the following way. The particle absorbs a phonon of energy ϵ_0 at the time $t = 0$ and obtains an initial velocity

$$v = \pm\sqrt{2\epsilon_0/m_p}$$

where m_p denotes the effective mass $m^\star$ of the polaron state, '+' corresponds to motion parallel and '−' to motion anti-parallel with respect to the external field. This can lead to three types of motion.

(i) If the phonon energy is sufficiently small, $\epsilon_0 < E_a - (eF\lambda)/2$, the particle cannot escape from the potential well, starts a damped oscillation and finally thermalizes at the initial lattice site.

(ii) If $\epsilon_0 > E_a - (eF\lambda)/2$, the particle overcomes the potential barrier and moves through the crystal. During the motion it gradually loses energy due to the friction and finally localizes at a site (which is, say, the site numbered l, where the initial site is site $l = 0$). This happens if the field is not so strong that it compensates for the energy loss. Obviously, the distance covered by the motion depends on the initial velocity, $l = l(v)$. For sufficiently small friction and a strong field, a particle which initially moved against the field ($v < 0$) may change its direction and one might even find that $l(-v) > 0$. But this possibility will not be considered in the following.

(iii) Finally, the excited particle will never again localize, but assumes a state of stable stationary drift motion in the field. In this case, band-like transport supersedes the hopping transport.

We start with the case of sufficiently small fields, where the motion is hopping-like. In this situation, the current arising from collisions with phonons can be determined from

$$j = en\nu[\rho(v) - \rho(-v)] \tag{9}$$

where n is the electron concentration, ν is the frequency of the collisions with phonons, and $\rho(\pm v) = \pm\lambda l(\pm v)$ is the distance covered by the motion following a collision with a phonon (with and against the field). Here it was assumed that the collisions with phonons are sufficiently rare that $\nu\,\delta t < 1$, where δt denotes the characteristic thermalization time after a collision (see below). With increasing field, the distance ρ becomes larger, and correspondingly the characteristic time δt increases, so the above-proposed scheme of Markovian transitions breaks down. The polaron will not come to rest between two subsequent collisions with phonons and the jumps will now assume a non-Markovian character. In this case a specific transport type occurs, in which the electron is displaced due to the impact of random forces, but it does not thermalize at a site in the time between collisions. The smaller the friction, the lower the fields at which the transition to non-Markovian processes takes place. At very small friction, non-Markovian hops occur even in the limit of very small F (in linear response theory). Non-Markovian hops of small polarons in the Ohmic regime have been investigated in [31].

In the following we restrict ourselves to Markovian transitions, i.e. our calculation is applicable only far away from the critical field at which the length of the jump diverges, and the transition to a band-like motion takes place. Below we give a quantitative criterion for the applicability of the model of Markovian transitions (see (37)). In passing, we note that in this transition region a rigorous calculation is burdened not only by the non-Markovian character of the processes. Additionally, the mechanism of friction, i.e. the functional form of $\boldsymbol{f}(\dot{\boldsymbol{R}})$ in (6), changes and the effective mass changes from $m^\star = m_p$ (polaronic) to $m^\star = m_e$ (electronic).

Averaging (9) over the initial velocities, we obtain

$$\begin{aligned} j &= en\nu\frac{m_p}{2kT}\int_0^\infty \mathrm{d}v\ v\exp\left(-\frac{m_p v^2}{2kT}\right)[\rho(v) - \rho(-v)] \\ &= en\nu\frac{1}{2kT}\int_0^\infty \mathrm{d}\epsilon_0\ \exp\left(-\frac{\epsilon_0}{kT}\right)[\rho_+(\epsilon_0) - \rho_-(\epsilon_0)] \end{aligned} \tag{10}$$

where $\rho_\pm(\epsilon_0) = \rho(\pm v)$.

Thus, the problem is reduced to the determination of the hopping length $\rho_\pm(\epsilon_0)$ using equation (6), where the influence of the random force $\xi(t)$ is replaced by the initial conditions

$$R\big|_{t=0} = R_0 \qquad \dot{R}\big|_{t=0} = \pm\sqrt{\frac{2\epsilon_0}{m_p}} \tag{11}$$

with R_0 the equilibrium position in the zeroth potential well determined by

$$\mathrm{d}U/\mathrm{d}R\big|_{R=R_0} = eF.$$

4. Current–voltage characteristics in the hopping regime

The situation is very simple for weak electric fields and sufficiently strong damping, in which case only hopping between nearest neighbours occurs. Then only $\rho_\pm = 0, \pm\lambda$ is

possible and according to (10)

$$j = \frac{en\lambda\nu}{2kT}\left[\int_{E_a-eF\lambda/2}^{\infty} \mathrm{d}\epsilon_0 - \int_{E_a+eF\lambda/2}^{\infty} \mathrm{d}\epsilon_0\right] \exp\left(-\frac{\epsilon_0}{kT}\right) = en\lambda\nu \exp\left(-\frac{E_a}{kT}\right) \sinh\left(\frac{eF\lambda}{2kT}\right). \tag{12}$$

Comparing this expression with the microscopically derived mobility of adiabatic polarons [19, 28, 32], one finds $\nu = \omega_0/2\pi$ for the frequency of the collisions with phonons, where ω_0 is the optical phonon frequency. The relation (12) is identical to the current–voltage characteristic (1), except for the anomalous factor $\exp[-(eF\lambda)^2/16E_akT]$, which is absent from (12).

Now we turn to the case of hopping to distant sites, which occurs if the damping is weak or the electric field is strong. Using the initial conditions (11), the equation of motion (6) can be integrated:

$$\frac{m_p\dot{R}^2}{2} + U(R) - U(R_0) = \epsilon_0 + eF(R - R_0) - \int_{R_0}^{R} \mathrm{d}R'\ f\left[\dot{R}(R')\right]. \tag{13}$$

We introduce the notion of the particle energy at the equilibrium position at site l, $R_{0l} = R_0 + \lambda l$:

$$\epsilon_l = \frac{1}{2}m_p\left[\dot{R}(R_0 + \lambda l)\right]^2. \tag{14}$$

The energy at the site $l = 0$ is identical to the energy of the absorbed phonon ϵ_0. If we set $R = R_{0l}$ and $R = R_{0l+1}$ in (13) and subtract one of the equations obtained from the other one, we get a recurrence relation for the energy loss of the polaron between neighbouring lattice sites:

$$\epsilon_{l+1} - \epsilon_l = \lambda[eF - L(\epsilon_l)] \tag{15}$$

where

$$L(\epsilon_l) = \frac{1}{\lambda}\int_{R_l}^{R_{l+1}} \mathrm{d}R\ f[\dot{R}(R, \epsilon_0)] = \frac{1}{\lambda}\int_{R_0}^{R_0+\lambda} \mathrm{d}R\ f[\dot{R}(R, \epsilon_l)]. \tag{16}$$

The expression $\dot{R}(R, \epsilon)$ denotes the velocity of the particle at the point R for an initial energy of ϵ, and the relation $\dot{R}(R + R_{0n}, \epsilon_0) = \dot{R}(R, \epsilon_n)$ was used in deriving (16).

In the case considered, of hopping to distant sites, the energy change between neighbouring sites is small, and, applying the substitution $\epsilon_l \to \epsilon(R_{0l}) \to \epsilon(R)$, the recurrence relation (15) can be replaced by a differential equation:

$$\frac{\mathrm{d}\epsilon(R)}{\mathrm{d}R} = eF - L(\epsilon) \qquad \epsilon(0) = \epsilon_0. \tag{17}$$

This equation is solved by

$$R(\epsilon) = \int_{\epsilon}^{\epsilon_0} \frac{\mathrm{d}\epsilon'}{L(\epsilon') - eF}. \tag{18}$$

Relation (18) holds for movement in the direction of the field. For a movement against the field one has to substitute $-F$ for F.

The energy of the particle which it possesses at the potential minimum of a well decreases as it moves from site to site due to the friction, and finally reaches the minimal value $E_a \mp (eF\lambda)/2$ ('$-$' and '$+$' for the motion with and against the field, respectively). Thereafter, it thermalizes in the corresponding well. Thus, we obtain the hopping distance

$$\rho_{\pm}(\epsilon_0) = \int_{E_a\mp eF\lambda/2}^{\epsilon_0} \frac{\mathrm{d}\epsilon}{L_{\pm}(\epsilon) \mp eF}. \tag{19}$$

The quantities $L_\pm$ differ in the sign of the electric field. Putting (19) into (10), integrating partially, and taking the equality $\rho_\pm(E_a \mp \frac{1}{2}eF\lambda) = 0$ into account, we obtain the following expression for the current:

$$j = en\nu \left\{ \int_{E_a - eF\lambda/2}^{\infty} \mathrm{d}\epsilon \ \frac{\exp(-\epsilon/kT)}{L_+(\epsilon) - eF} - \int_{E_a + eF\lambda/2}^{\infty} \mathrm{d}\epsilon \ \frac{\exp(-\epsilon/kT)}{L_-(\epsilon) + eF} \right\}. \quad (20)$$

Now it remains to determine the damping function $L_\pm(\epsilon)$, equation (16). In the limit of weak friction, it can be obtained by integrating (13):

$$L_\pm(\epsilon) = \frac{1}{\lambda} \int_0^\lambda \mathrm{d}R \ f\left[\sqrt{\frac{2}{m_p}\epsilon - U(R + R_0) + U(R_0) \pm eFR} \right]. \quad (21)$$

Below we will consider the case of electric fields that are not too strong, when $eF\lambda \ll E_a$ and, thus, $L_+(\epsilon) = L_-(\epsilon) \equiv L(\epsilon)$. Only in this case does hopping motion actually take place, as we will see later. Furthermore, we can assume $R_0 = 0$, i.e. we neglect the displacement of the potential minimum due to the electric field. With these assumptions the following quantity can be introduced:

$$K(\epsilon) \equiv L(\epsilon + E_a) = \frac{1}{\lambda} \int_{-\lambda/2}^{\lambda/2} \mathrm{d}R \ f\left(\sqrt{\frac{2}{m_p}[\epsilon - U(R)]} \right). \quad (22)$$

Here, the origin of the position coordinate is moved to the position of the maximum of the potential $U(R)$ ($R \to R + \lambda/2$), and the energy origin to the value at the maximum of the potential ($U(0) = 0$, $U(R) \leqslant 0$). The expression (20) for the current now takes the form

$$j = en\nu \exp\left(-\frac{E_a}{kT}\right) \int_0^\infty \mathrm{d}\epsilon \ \exp\left(-\frac{\epsilon}{kT}\right) \left\{ \frac{\exp[eF\lambda/(2kT)]}{K(\epsilon) - eF} - \frac{\exp[-eF\lambda/(2kT)]}{K(\epsilon) + eF} \right\}. \quad (23)$$

The damping function $K(\epsilon)$ increases with increasing energy, and so the condition for the applicability of the approach considered reads $K(0) \geqslant eF$. Consequently, there is a critical value of the electric field

$$eF_c = K(0) \quad (24)$$

whose attainment leads to a diverging hopping length, and finally to the transition of the hopping transport to band-like transport. If the potential $U(R)$ is a smooth function and there is only one characteristic energy, namely the height of the barrier E_a, and the friction function can be used in the linear approximation, equation (7), then

$$eF_c \approx B\eta_p \sqrt{2E_a/m_p} \quad (25)$$

where B is a numerical coefficient and η_p denotes the friction coefficient in the range of hopping transport. For weak damping (large τ) and a large mass of the polaron, $eF_c\lambda \ll E_a$, which justifies the assumptions used in calculating $L_\pm(\epsilon)$. Additionally, for this critical field, $F_c \ll F_c'$ (see (2)).

Since we are interested in the case where $E_a \gg kT$, the largest contribution to the integral over ϵ in (23) comes from small values of ϵ. In the linear approximation of the function $f(\dot{R})$ we find

$$K(\epsilon) \cong eF_c \left(1 + \frac{\epsilon}{E_a B\sqrt{2C}} \left[\ln \frac{CE_a}{\epsilon} + 1 \right] \right) \quad (26)$$

where the number C is defined by

$$\mathrm{d}^2U/\mathrm{d}R^2\big|_{R=0} = -C\lambda^{-2}E_a.$$

Taking into account the condition $E_a \gg kT$ and (23), the following expression for the current–voltage relation in the hopping regime is obtained:

$$j \cong \frac{n\nu kT}{2F_0}\exp\left(-\frac{E_a}{kT}\right)\left\{\exp\left(-\frac{eF\lambda}{2kT}+\frac{F+F_c}{F_0}\right)\mathrm{Ei}\left(-\frac{F+F_c}{F_0}\right)\right.$$
$$\left. - \exp\left(\frac{eF\lambda}{2kT}+\frac{F_c-F}{F_0}\right)\mathrm{Ei}\left(-\frac{F_c-F}{F_0}\right)\right\}. \tag{27}$$

Here, $\mathrm{Ei}(x)$ is the exponential integral function and F_0 is a characteristic electric field:

$$F_0 = F_c\frac{kT}{E_aB\sqrt{2C}}\left[\ln\frac{CE_a}{kT}+1\right]^{-1} \ll F_c. \tag{28}$$

Far away from the transition, when $(F_c - F) \gg F_0$,

$$j \cong \frac{1}{2}n\nu kT\exp\left(-\frac{E_a}{kT}\right)\left\{\frac{\exp(eF\lambda/2kT)}{F_c-F}-\frac{\exp(-eF\lambda/2kT)}{F_c+F}\right\}. \tag{29}$$

We note that the expressions (12) and (29) for the current do not coincide in the linear approximation in the electric field. The reason for this is that the traditional expression (12) for small polarons corresponds to the case of nearest-neighbour hopping at large friction, whereas (29) corresponds to hopping to distant sites, which takes place for small friction.

Near the transition, according to (27),

$$j \cong \frac{n\nu kT}{2F_0}\exp\left(-\frac{E_a}{kT}+\frac{eF\lambda}{2kT}\right)\ln\frac{F_0}{F_c-F}. \tag{30}$$

5. Current–voltage characteristics in the region of band-like transport

If the electric field is larger than F_c, then the electronic motion is unlimited even in the absence of random forces. In this region of the field, a stationary asymptotic drift motion is established, in which the energy loss due to friction over a distance of one lattice spacing is exactly compensated by the energy gained over this distance due to the electric field. If Θ denotes the time needed to move one lattice period, then the stationary current has the form

$$j = en\frac{\lambda}{\Theta}. \tag{31}$$

The period Θ can be determined from the following considerations. The condition of energy balance reads (see (13), (16), (22) and (26))

$$eF = K(\epsilon) \cong eF_c\left(1+\frac{\epsilon}{E_aB\sqrt{2C}}\left[\ln\frac{CE_a}{\epsilon}+1\right]\right). \tag{32}$$

Here, we assumed that the friction function admits the linear approximation (7), and ϵ denotes the kinetic energy of the electron at the potential minimum in the stationary regime of motion. Equation (32) relates F and ϵ in the range of electric fields larger than F_c and for $(F - F_c) \ll F_c$. On the other hand, the relation between Θ and ϵ reads

$$\Theta = \sqrt{\frac{m_e}{2}}\int_{-\lambda/2}^{\lambda/2}\frac{\mathrm{d}R}{\sqrt{\epsilon-U(R)}} \cong \sqrt{\frac{m_e}{CE_a}}\ln\frac{CE_a}{\epsilon}. \tag{33}$$

The equations (31)–(33) determine the current–voltage characteristics in the region of the field immediately above the threshold. Up to double-logarithmic precision, we obtain

$$j \cong en\sqrt{\frac{CE_a}{m_e}}\ln^{-1}\left(\frac{BC\sqrt{2C}F_c}{F-F_c}\right). \tag{34}$$

For $F \geqslant F_c'$ (see (2)), the electron can get over the barrier regardless of its inertia. In this regime not only can the random force be omitted from (6) but so also can the second time derivative. Then,

$$j \propto \sqrt{F - F_c'} \qquad F - F_c' \ll F_c'. \tag{35}$$

For an even further increased field, the term $\mathrm{d}U/\mathrm{d}R$ in (6) can be omitted, as well. This results in the transition to the linear regime, $j = e^2 n \tau_e F/m_e$, if the drift velocity is still smaller than the critical value w, which is determined by the instability for large polarons (see the discussion following (6)). Furthermore, we assumed that a renormalization of the particle mass and the relaxation time takes place ($m_p \to m_e$, $\tau_p \to \tau_e$).

The above-stated considerations lead for small temperatures (in the absence of random forces) to a hysteretic current–voltage relation, similar to the phenomenon which occurs in the theory of the motion of Josephson vortices [26]. In an increasing field, the current is zero in the range $0 < F < F_c'$. Thereafter, the current–voltage relation either follows (35) under neglect of the kinetic energy term, or, if the inertia is taken into account, the current leaps from zero to the value determined by (34). If, on the other hand, the field is decreased, the current is not equal to zero in the interval $F_c < F < F_c'$ if the inertia is taken into account. However, the model of classical motion for small polarons considered here is not applicable to the region of low temperatures (see [32]). But a hysteresis will nevertheless occur in small-polaron models at higher temperatures due to a different mechanism.

When the field is lowered from above F_c, the drift velocity of the charge carrier at $F = F_c$ is still so large that the crystal lattice is unable to form a polaron well (see section 2). Thus, the current stays on the band-like branch and has Ohmic character. This is the case until the drift velocity reaches a critical value $\lambda/\bar{t}$, corresponding to a field strength $F_c'' \approx \lambda m_e/(e\tau_e \bar{t}) < F_c$. Then, the polaron well starts to be formed, which further lowers the drift velocity, and the current decreases in an avalanche-like fashion to the hopping branch. This yields a hysteresis between F_c'' and F_c, which is of a fundamentally different nature to the above-described hysteresis between F_c and F_c', which occurs only for low temperatures.

6. Results and discussion

The proposed model allows us to describe the transition from hopping conduction to band transport. This transition takes place at a critical field F_c, which, for low friction and large particle mass, is lower than the field F_c' that is required to compensate the potential barrier between neighbouring sites. The physical reason for the occurrence of the unbounded motion while there is still a barrier lies in the inertia of the particle, which can enable the electron to cross the subsequent potential barriers after having once obtained enough energy to leave the initial site, if the friction is sufficiently weak. The critical field can even be so small that the relation $eF_c\lambda \ll kT$ is valid. Then, in the hopping regime, i.e. for $F < F_c$, the current–voltage characteristic reads

$$j = nvkT \exp\left(-\frac{E_a}{kT}\right)\frac{F}{F_c^2 - F^2}. \tag{36}$$

Here, the factor $\exp(eF\lambda/2kT)$, which is characteristic for hopping, is absent.

Due to the non-Markovian crossover, the current–voltage relation (30) in the hopping region and (34) in the band-like transport region do not coincide at the critical field $F = F_c$. Because the regime of non-Markovian processes secures a continuous transition between hopping and band-like transport, we estimate the width of this transition region. To this

end it is necessary to find the thermalization time δt of the charge carrier after it has been excited by a phonon. In the continuum approximation, the velocity of the particle at the point R can be described by

$$\dot{R}^{-1}(R) = \frac{\sqrt{m_p}}{\lambda\sqrt{2}} \int_{-\lambda/2}^{\lambda/2} \frac{\mathrm{d}R'}{\sqrt{\epsilon(R) - U(R')}}$$

where $\epsilon(R)$ denotes the energy of the particle, relative to the potential maximum. Then,

$$\delta t \cong \int_0^{R_+} \frac{\mathrm{d}R}{\dot{R}(R)} = \int_0^{\epsilon_0 - E_a} \frac{\mathrm{d}\epsilon\, \dot{R}^{-1}(\epsilon)}{\eta_p K(\epsilon) - eF}$$
$$\cong \frac{m_p}{\eta_p} \ln\left\{1 + \frac{F_c}{F_c - F} \frac{\epsilon_0 - E_a}{E_a B\sqrt{2C}} \left[1 + \ln \frac{C E_a}{\epsilon_0 - E_a}\right]\right\}.$$

Here we used (17), (22) and (26). We are interested in the collisions with phonons with an energy larger than E_a. The averaging of the inequality $\nu\, \delta t \ll 1$ over these phonons yields the condition of applicability of the Markovian description of transport:

$$\frac{F_c - F}{F_c} \gg \frac{kT}{E_a} \ln\left(\frac{E_a}{kT}\right) \exp\left[-\frac{1}{\omega_0 \tau_p} \exp\left(\frac{E_a}{kT}\right)\right]. \tag{37}$$

The numerical coefficients are omitted here, the relations $\eta_p = m_p/\tau_p$ and $\nu = \omega_0/2\pi$ are used, and the polaron relaxation time τ_p is defined in section 2. Thus, the transition region is very narrow for sufficiently large values of the parameter E_a/kT. Precisely this circumstance also leads to the fact that the transition from hopping to band-like motion has the character of a sudden (discontinuous) increase of the current, which reminds one of an avalanche breakdown.

We numerically calculated the current–voltage relation using (6) and the potential $U(R)$ of the form (A4). For this potential the above-introduced numerical coefficients are $A = 4$, $B = 1/2$ and $C = 8$. The primed critical field becomes $F_c' = 4E_a/(e\lambda)$.

The friction coefficient is approximated as $\eta = m/\tau$, where m is the electron or polaron mass, respectively, and τ is of the order of 10^{-11} s. For an excitation by optical phonons,

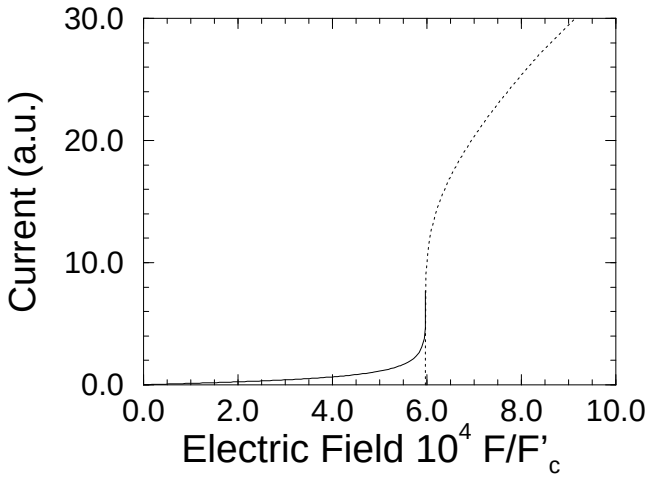

Figure 1. The numerically computed current (in arbitrary units) showing the transition from hopping transport (full curve) to band-like transport (dotted curve) at $F = F_c$. Note that F_c is about three orders of magnitude smaller than F_c' (experiments suggest two orders of magnitude).

ν is of the order of 10^{13} s^{-1}. The parameters for the numerical computation are chosen as $\lambda = 5$ Å, $F_c' = 10^7$ V cm^{-1}, $kT = 0.2E_a$, $m_p = 100m_e$ and $\tau_p = 10^{-11}$ s. This yields $F_c = 6 \times 10^3$ V cm^{-1}, $E_a = 1.25$ meV and $T = 290$ K. The remaining parameters are $\tau_e = 10^{-12}$ s and $\nu = 10^{13}$ s^{-1} for figure 1, whereas figure 2 is obtained by using $\tau_e = \sqrt{10} \times 10^{-12}$ s and $\nu = 10^{14}$ s^{-1}.

The full curves correspond to the low-field (hopping) solution, and are calculated through integration of the ODE and computing the integral (10) numerically. Clearly, the current diverges near F_c. The dotted lines show the band-like solution. They are calculated using the parameters m_e and τ_e instead of m_p and τ_p, and represent the iteratively determined steady-state solution of (6). The dashed line in figure 2 shows the approximation according to (27), which is reasonably close to the exact solution.

The reversible breakdown in the current–voltage characteristic, which is described here, was experimentally observed for the oxides $Co_{1+x}Cr_{2-x}O_4$ and $Co_{1-x}Li_xO$ at fields $F_c = 10^4$–10^5 V cm^{-1} in [20], and was interpreted by the authors as a delocalization phenomenon for small polarons. We suppose that the observed hysteretic current–voltage relation corresponds to our proposed hysteresis with $F_c/F_c'' \cong 1.15$ in this case. An important fact, which favours the applicability of our considerations to this experiment, consists in the relatively small value of the critical field, which is experimentally found to be two orders of magnitude lower than the field $F_c' \approx 10^7$ V cm^{-1} at which the potential barrier vanishes.

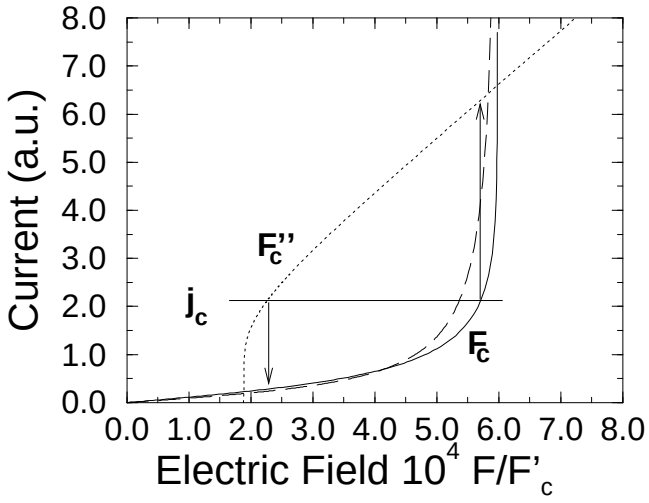

Figure 2. The numerically computed current for a different parameter set to that of figure 1 (see the text). The full curve represents the hopping current, and the dotted curve is for band-like transport. Additional to the breakdown at F_c (equal to $6 \times 10^{-4}F_c'$), there is now a hysteresis in the current–voltage characteristic. The broken curve represents the approximation (27) for the hopping regime. The hysteresis is caused by the non-adiabatic behaviour of the polaron near the dissociation point. The polaron velocity reaches the critical value $\dot{R}_c = \lambda\bar{t}^{-1} = j_c/en$ in an increasing field at $F = F_c$, the behaviour becomes non-adiabatic, the polaron well vanishes and the current–voltage characteristic jumps to the Ohmic branch. The velocity is still larger than $\dot{R}_c$ at $F = F_c$ in a decreasing field, and the current remains Ohmic for $F < F_c$. Finally, at $F = F_c''$, the velocity becomes $\dot{R}_c$, a polaron well forms and the current–voltage characteristic jumps to the hopping branch. The arrows indicate these transitions schematically. The horizontal line represents the (here arbitrarily chosen) value of $\dot{R}_c$.

The calculated band-like current shown in figure 2 does not become zero at $F = F_c$, which contradicts (34). This is caused by the mass and relaxation time renormalization around the transition region. For a fixed relaxation time, the critical field is $F_c \propto \sqrt{m}$ according to (25). Thus, under the assumption that the relaxation time renormalization is rather small compared to the effective-mass renormalization, F_c is formally smaller for band-like motion than for hopping motion. This can also cause the current to stay on the upper branch of the current–voltage relation when the electric field is lowered from above F_c. In a narrow transition region around F_c, complex processes take place, e.g. non-Markovian hops, mass and relaxation time renormalization, and non-adiabaticity of the motion, which cannot be described within our proposed model. However, all of these are processes with positive feedback, so the transition between hopping and band-like transport is very fast and resembles an electrical breakdown. For band-like motion, when the term $\mathrm{d}U/\mathrm{d}R$ can be omitted in (6), the transport can be described by the methods of large-polaron theory [22–25].

The calculations in this paper have been carried out for small polarons in an ordered 1D chain. For three-dimensional (3D) crystals, in which the current path forms a straight line, the current–voltage relation is qualitatively equivalent to that in the 1D case. The basic qualitative results of this paper (the decrease of the threshold field due to inertia, hysteresis, transition from an activated temperature dependence to a non-activated one and sudden increase of the current with increasing field near the threshold) should be valid even for disordered 3D systems which show activated transport. However, in disordered systems, where the potential energy $U(R)$ is a random relief, the current path is strongly bent as shown by percolation theory. Therefore, the 3D (and 2D) system is essentially different from the 1D system. In disordered media with current percolation in a strong electric field, the carriers are pooled before the highest (critical) barrier, which forms a 'bottle-neck'. In this situation the Fermi statistics starts to become essential even in the case in which the electron concentration is low.

Finally, a quite different area of physics might benefit from the approach proposed in this paper. The resistively shunted junction model [26] of a Josephson junction is described by an equation which is formally equivalent to (6). (The effective potential U would be a sine function in this case.) There are only few analytic results for this model, so a quantitative study based on our approach would be interesting. At very least, the qualitative results of this paper (apart from the parameter renormalization at the transition) fully correspond to findings for Josephson junctions.

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Appendix. The effective potential for small polarons

In the adiabatic approximation, the Hamiltonian of small polarons with short-range electron–phonon interaction and with dispersionless optical phonons reads [19, 28, 32]

$$H = \sum_m \left(\frac{1}{2} M \omega_0^2 x_m^2 - \gamma x_m n_m \right) \tag{A1}$$

where γ is the electron–phonon coupling constant, M the reduced mass of the unit cell of the crystal and x_m the displacement of the nuclei from their equilibrium position (in the

optical oscillation mode). Here, we obtain the effective potential without exploitation of Legendre factors, by directly solving (3) and (4). We introduce the quantity $\langle n_m \rangle$, which denotes the occupation number of site m under the condition that the system energy is minimal and the constraints (3) and (4) are fulfilled. Minimizing (A1) with respect to the x_m, we obtain

$$U(R) = -2E_a \sum_m \langle n_m \rangle^2 \tag{A2}$$

where

$$E_a = \frac{\gamma^2}{4M\omega_0^2} \tag{A3}$$

denotes, as we will see below, the barrier height between neighbouring sites, and $2E_a$ is the polaron shift.

The simplest way to obtain the minimum (A2) is to employ the two-site model, where $n_m = 0$ at all sites, except at $m = 0$ and 1. This approximation yields the following set of equations for $U(R)$:

$$U(R) = -2E_a(n_0^2 + n_1^2) \qquad R = \lambda n_1 \qquad n_0 + n_1 = 1.$$

We finally obtain the expression for the effective potential:

$$U(R) = -2E_a + 4E_a \frac{R}{\lambda}\left(1 - \frac{R}{\lambda}\right) \qquad 0 < R < \lambda. \tag{A4}$$

We note that, by applying the two-site model, the occupation numbers n_0 and n_1 of the sites can in general be obtained without minimization, because the number of variables (n_0, n_1) is equal to the number of constraints. Minimization is required for models in which three or more sites are considered.

The potential obtained, equation (A4), does not contain the resonance integral J, the square of which is usually proportional to the hopping probability. The absence of the resonance integral is due to the restriction to adiabatic transitions [28], where the gap arising from level crossing of the adiabatic states, which is proportional to J, is sufficiently large. According to the theory of small polarons, adiabatic transitions occur if

$$J^2/(\hbar\omega_0\sqrt{E_a kT}) \gg 1.$$

The pre-exponential factor $J^2/\hbar\sqrt{E_a kT}$ for the transition probability of non-adiabatic polarons is replaced by ω_0 for adiabatic polarons.

The approach used to obtain $U(R)$ is very simple, because the deformation of the localized electronic wave packet due to its displacement relative to the localization centre is not taken into account. The consequent procedure using Legendre multipliers and solving the corresponding Schrödinger equation in the adiabatic approximation is too complicated to be practically feasible. Here, one only has to note that in the case where the localization radius is smaller than the lattice constant (as is the case for small polarons), this effect can have a significant influence on $U(R)$. Specifically, an additional small dimensionless parameter may be introduced: the ratio between the localization length and the lattice constant (this equals J/E_a for small polarons). However, this will lead not to drastic changes of the results presented in this paper, but merely to the numerical coefficients A, B, and C possibly becoming dependent on this small parameter.

The problem is easier in the case of a state for which the radius is larger than the lattice constant. Then the deformation of the electronic wave packet due to its displacement relative to the localization centre is small and the potential energy can be represented as a Fourier

series with fast-decreasing terms, so that $U(R) = E_a \cos(2\pi R/\lambda)$, and the barrier height E_a is exponentially small compared to a large parameter, i.e. the ratio between the localization length and the lattice constant. In this case there are actually only two parameters present, the lattice constant λ and the activation energy E_a, and the problem is formally equivalent to the problem of the dynamics of Josephson vortices [26, 27].

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