

On the temperature dependence of fast electron transport in crystal lattices

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Received 22 June 2015 / Received in final form 25 June 2015

Published online 10 August 2015 – © EDP Sciences, Società Italiana di Fisica, Springer-Verlag 2015

Abstract. Building upon the findings of Muto et al. [Phys. Lett. A **136**, 33 (1989)] and Marchesoni and Lucheroni [Phys. Rev. E **44**, 5303 (1991)] about the growth of the number of (anharmonic) lattice solitons with increasing temperature and using a recent transport theory developed by the present authors [A.P. Chetverikov, W. Ebeling, G. Röpke, M.G. Velarde, Eur. Phys. J. B **87**, 153 (2014)] here we provide the fractional power law of the temperature dependence of resistivity in a rather general model for one-dimensional crystal lattices as, e.g., conducting polymers. We also show that the determining factor for the transport is the possibility of forming electron-soliton bound states (in short solectrons) with a most significant contribution arising from the (bosonic) bound state of two electrons to a soliton (in short bisolectrons).

1 Introduction

Long ago, Mott [1] considered materials showing a minimum of resistivity as a function of temperature with the possibility of being metallic and non-metallic, respectively, above and below the temperature of such a minimum. Mott also argued that the non-metallic region may exhibit a fractional power law with temperature, a prediction found correct in quite a number of experiments, in particular for conducting polymers [2–7]. Mott’s approach builds upon the thermally-activated or otherwise variable-range hopping approximation [1]. Then for solid state materials he proposed that the probability, P , of hopping between states of spatial separation R and energy separation ΔE , at temperature T , can be set as:

$$P(R, \Delta E) \approx \exp[-2\alpha R - \Delta E/kT],$$

where the range parameter α accounts for wave function exponential decay length of an electron placed at a lattice site; k is Boltzmann’s constant. The hopping may very well be beyond nearest-neighbors. Then from such an assumption, Mott established that at low enough temperatures the resistivity, ρ , obeys the following fractional power law

$$\rho(T) \approx \rho_0 \exp\left[-2(T_0/T)^{1/(d+1)}\right],$$

a law, in principle, valid in three-dimensions ($d = 3$) for any disordered system at temperatures below the resistivity minimum and presumably strictly valid at only very

low temperatures. The quantities ρ_0 and T_0 are constants whose value is not needed here. Accordingly, the predicted exponent is between (1/2) and (1/4) if we allow d to account for $d = 1, 2$ or 3 dimensions.

Here we provide a different line of thought by considering a model based on a quasi one-dimensional system consisting on a nonlinear/anharmonic lattice with added, excess, free electrons. Generally, such a dynamical system has been shown to offer the possibility of long range, fast transport in the form of, e.g., soliton-assisted “electron surfing”. We shall focus on only one of its significant features, i.e., the above mentioned temperature dependence in the so-called non-metallic regime at low enough temperatures. Our assumptions are:

- The quasi 1D-lattice consists of N units, called molecules or atoms, with Morse interactions between them. Moreover, the lattice, of length L , is able to sustain soliton-like excitations if appropriate momentum is given to one or a few lattice units. We consider this a mechanical excitation at say zero K .
- Upon heating the lattice, i.e., thermally activating it lattice solitons can also be excited. Results are known about the density of such “thermally excited” lattice solitons [8–10].
- Both mechanically excited and thermally excited lattice solitons are able to trap added, excess, free electrons thus forming bound states denoted in short solectrons. If a soliton binds two electrons the compound is denoted bisolectron in short (with due account done of Coulomb repulsion and Pauli’s exclusion principle) [11–15].

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- The electron transport can be described using the lattice Morse Hamiltonian coupled to an electron Hamiltonian taken in the standard tight binding approximation (TBA) [16,17].

2 The model and the low temperature behavior of soliton-assisted conductivity

The model Hamiltonian we shall be using is a direct generalization of the Hamiltonian earlier used by several authors [16–20] for various crystalline enough albeit originally non-metallic materials.

Let n, m denote the internal quantum numbers of the states of electrons bound to the corresponding atoms at sites r_n and r_m , respectively. We will assume, for simplicity, that there is only one quantum state per atom with Gaussian shape, which can be occupied by the added electron. The internal state that characterizes the orbit as well as spin, can be included in the quantum number n . We set the electron wave function and Hamiltonian as follows [16–20]

$$\begin{aligned}\psi(r, t) &= \sum_n c_n(t) \psi_n(r); \\ H_e &= \sum_n E_n c_n^\dagger c_n + \sum_{n,m} t_{m,n} c_m^\dagger c_n.\end{aligned}\quad (1)$$

The energy levels E_n will be approximated by constant values $E_n = E_0$. The quantities $c_n^\dagger(c_n)$ denote creation (annihilation) operators in an appropriate Fock space eventually leading to electron probability density at site n . The coupling to the lattice is given by the transition matrix elements $t_{n,m}$, which depend on the atomic distances, $t_{n,m} = t(r_n - r_m)$. Following Slater [21], Launay and Verdaguer [22], we take an exponential dependence

$$t_{n,m} = V_0 \exp[-\alpha(r_{mn} - r_0)], \quad (2)$$

with $r_{mn} = |r_n - r_m|$. The characteristic equilibrium inter-tunneling distance, r_0 , is used as unit of length.

As noted above, the range parameter α can be related to the tunneling probability. For the atomic lattice part, the Hamiltonian with Morse interactions reads

$$\begin{aligned}H_a &= \frac{M}{2} \sum_n v_n^2 + \frac{D}{2} \sum_{m,n} \exp[-2b(r_{mn} - r_0)] \\ &\quad - 2 \exp[-b(r_{mn} - r_0)].\end{aligned}\quad (3)$$

The subscripts locate the atoms all with equal mass, M , at lattice sites and the summations run from 1 to N . In the Morse potential the lattice units repel each other with exponentially repulsive forces of range $1/b$ (b -stiffness) and binding energy strength D while attracting each other with weak dispersion forces. Time is measured in units of the reciprocal frequency of the linearized Morse spring (around the minimum of its potential well depth).

Note that the Davydov model [18] is obtained, if the energies E_n are considered as linear functions of

the shifts $u_n \sim r_n - r_{n0}$ of units from equilibrium position r_{n0} , with $\alpha = 0$. On the other hand we get the simplest SSH-model for undimerized chains, if the E_n are constant and if the Morse potential and the Slater exponential in equation (2) are linearized. The key elements in our model are the lattice anharmonicity and the (exponential) nonlinearity in the coupling of electrons to the lattice.

Disregarding unnecessary details we assume that added, excess or otherwise free electrons are able to form (localized) bound states with local compressions of the nonlinear lattice. They may be considered as natural generalization of the nowadays textbook concept of polaron quasiparticle/dressed charge carrier of Landau [23] and Pekar [24]. The transport is based on the hopping of the charges. According to Muto et al. [8], and Marchesoni and Lucheroni [9], the density of thermally excited solitons in an one-dimensional lattice increases with a fractional power law $T^{1/3}$. This result was originally obtained for a Toda potential with parameter values corresponding to DNA and related biomolecules. They showed that thermal solitons may exist in such systems up to physiological or otherwise room temperature (ca. 300 K). For the Morse potential, computer simulations exhibit similar soliton-like excitations with small deviations from the exact, analytical solutions of the Toda case. Hence we shall consider this as a negligible error and will use the Morse potential suitably adapted up to third derivative to the Toda potential. Accordingly, in our Morse lattice the number of solitons per site increases also like $T^{1/3}$. Thus with a well depth D (0.1–0.3 eV) the fraction of solitons per site, x_s , at low enough T is [25]

$$x_s(\tilde{T}) = 0.75\tilde{T}^{1/3}, \quad (4)$$

with $\tilde{T} \equiv T/2D$ (save k , the Boltzmann constant). Starting from zero- K we consider as the first significant value of the temperature the one corresponding to a level when about ten percent of all lattice sites are involved in solitons. In view of the above we have

$$0.75\tilde{T}_{s0}^{1/3} \sim 0.1 \longrightarrow \tilde{T}_{s0} \sim 10^{-3}. \quad (5)$$

Assuming that the energy unit, taken as twice the depth of the Morse well, is about 0.5 eV, the soliton temperature T_{s0} will be in the range of 5 K or more generally in the bracket (1–10 K). This is indeed the range where the fractional power law is observed. Besides the soliton temperature, there are several other significant albeit higher temperatures which are connected with the formation of solectrons, $s + e \rightarrow (se)$, with binding energy E_{se} (s, e and se denote, respectively, lattice soliton/deformation, electron and solectron). We assume that at low enough T each of the existing solitons binds an electron, and hence some of the added, excess electrons are left free, i.e., unbound. Then the fraction of solectrons per site at low enough temperature is approximately

$$\begin{aligned}x_{se}(\tilde{T}) &\approx x_s(\tilde{T}) = 0.75\tilde{T}^{1/3}, \\ \log \rho(\tilde{T}) &\approx (1/x_{se}) = C - (1/3) \log(\tilde{T}).\end{aligned}\quad (6)$$

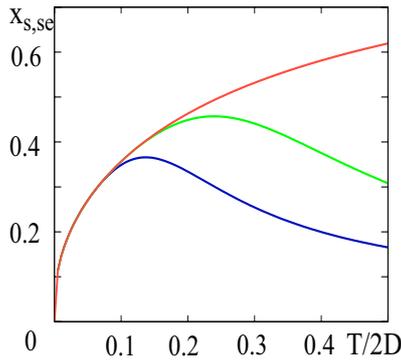


Fig. 1. Lattice solitons and solectrons vs. temperature. The upper/red curve shows the density of thermal solitons, i.e., the fraction of lattice sites occupied by solitons. The bottom/blue and the middle/light green curves show the corresponding fraction of electron-soliton bound states (sollectrons) for doping fraction 0.3 and for two binding energies D (blue) and $2D$ (green). The larger the binding energy is, the larger is the range of validity of the $1/3$ power law (upper/red curve).

This straightforwardly provides a fractional power ($1/3$) as a direct consequence of the soliton-assisted binding process. As upon increasing T not all solitons would be able to catch an electron, the sollectron “chemical reaction” $s + e \rightarrow (se)$ demands careful consideration.

3 Why a minimum in the resistivity vs. temperature is expected?

Let us consider the mass action law for the formation of bound states (sollectrons) $s + e \rightarrow (se)$ according to the statistical theory for transport developed in reference [25]. At chemical equilibrium we have

$$\frac{x_{se}}{x_s x_e} = \left(\frac{T_{de}}{T}\right)^{1/2} \exp\left(-\frac{E_{se}}{kT}\right), \quad (7)$$

where T_{de} is a kind of “degeneration” temperature of the free electrons as it appears in the standard mass action laws, at least in one-dimensional systems, those of our interest here.

Assuming as binding energies either $E_{se} \sim -D$ or $E_{se} \sim -2D$ we get for the sollectron density dependence on T curves like those presented in Figure 1. We see that at low enough temperatures, $\tilde{T} < 0.1$, nearly all solitons are charged, i.e., the number of sollectrons follows the $T^{1/3}$ law. Thus the maximum in the (se) -formation can be estimated as:

$$kT_{se} \sim 0.1 |E_{se}|. \quad (8)$$

This leads to $kT_{se} \sim 0.1D$, i.e., around 200 K or, in dimensionless units, $\tilde{T}_{se} \sim 0.05$.

Above the expected maximum determined by the sollectron binding energy, the $T^{1/3}$ power law ought to cease being valid. We may guess that a maximum of

sollectron density is related to a minimum of resistivity. Thus assuming that the conductivity is proportional to the density of sollectrons which have the mobility μ_{se} this means that the conductivity will show a maximum and the resistivity a corresponding minimum. This provides a first qualitative explanation of such a minimum. A better description has been offered by the transport theory of sollectron-assisted conductivity, developed in reference [25]. The sollectron contributes to the current and hence to the conductivity as a most significant factor. In the presence of an electric field, E , the sollectron current, j_{se} , and conductivity, σ_{se} , are

$$j_{se}(T) = n_{se} e \mu_{se} E, \quad (9)$$

$$\sigma_{se} \approx n x_{se}(T) e \mu_{se} = 0.75 \tilde{T}^{1/3} n e \mu_{se},$$

with e denoting electron charge. This gives for low enough temperatures, where all solitons bind electrons, thus all forming sollectrons, the fractional power laws for the contribution of sollectrons to conductivity/resistivity

$$\log \sigma_{se}(\tilde{T}) \approx \frac{1}{3} \log(\tilde{T}) + \text{const.} \quad (10)$$

So far we have followed the simple Drude theory. More complete results follow from the computation of the sollectron contribution to the Kubo formula expressed using mean square displacements. Indeed we have

$$\sigma_{se}(T) = n_{se} \frac{e^2}{kT_q \nu_{se}} \langle v^2 \rangle_T, \quad (11)$$

where $n_{se} = x_{se} N/L$ and ν_{se} is a collision frequency. In the basis of bistable distributions for the sollectrons [25,26] we found that

$$\langle v^2 \rangle_T = \frac{kT_q}{m_{se}} + v_{se}^2(T),$$

$$kT_q = \frac{\hbar\omega}{2} \coth \frac{\hbar\omega}{2kT},$$

$$v_{se}^2(T) = v_s^2(T) - 1.8\alpha V_0 v_0^2. \quad (12)$$

T_q is a quantum temperature associated to the oscillations of the electron in the soliton well. The second expression provides the decrease of the sollectron velocity relative to the (initial) soliton velocity, $v_s(T)$, as function of α and V , equation (2), as shown in reference [25]. There we also studied the enhancement factor and found a relatively sharp maximum in T . The electron bound to a moving soliton is not just carried by the soliton but oscillates around the minimum of a quantum well with a frequency ω_{se} . The depth of the well is of the order of the depth of the Morse well D . This leads to the quantum temperature of the sollectron oscillator $T_q = \hbar\omega_{se}/2k$ and, in dimensionless units, $\tilde{T}_q \sim \hbar\omega_{se}/4D \sim 10^{-2}$.

Let us only sketch the idea how to derive the full current and the total conductivity according to the theory in reference [25] which was developed in the spirit of the

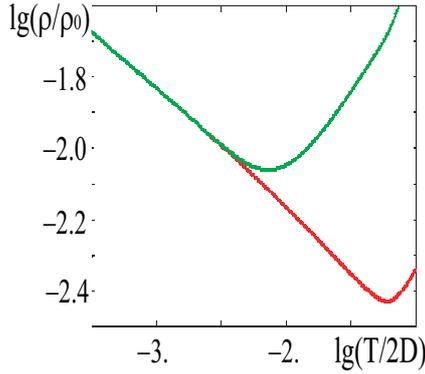


Fig. 2. Log-log plot of resistivity ρ/ρ_0 vs. temperature \tilde{T} (T in units $2D$) incorporating contributions from solectrons for two values of the set of free parameters estimated in [25]: $a = 10^{-1}$, $a = 10^{-2}$ and $b = 10^3$ (lower/red and upper/light green curves, respectively). The former corresponds to a too low temperature whereas the latter corresponds to a too wide potential well with a too low oscillation frequency of electron motions.

Drude relaxation time heuristic approach. The conductivity demands the contribution of both free and bound electrons (solectrons)

$$\sigma(T) = \frac{n_e e^2}{\rho_e(T)} + \frac{n_{se} e^2}{\rho_{se}(T)} r_{se}(T),$$

$$r_{se}(T) = 1 + \frac{m_{se} v_{se}^2(T)}{kT_q} \coth\left(\frac{T_q}{T}\right), \quad (13)$$

where $\rho_e = m_e \nu_e(T)$ and $\rho_{se}(T) = m_{se} \nu_{se}(T)$ correspond to free electrons and solectrons, respectively. Further we introduced a temperature-dependent enhancement factor $r_{se}(T)$ through internal parameters of the solectron a and b (here a is proportional to the frequency of oscillations in the soliton well and b is proportional to the solectron velocity [25]). Figure 2 offers a log-log plot of resistivity $\rho = 1/\sigma$ vs. \tilde{T} using values for two sets of the parameters $a = kT_q[2D]$ and $b = m_{se} v_{se}^2(T)/kT_q$. We see clearly the universality of the $T^{1/3}$ fractional power law followed by a minimum, whose actual location depends indeed on the parameter values chosen, an arbitrariness yet to be discussed. The minimum appears to be at too low temperatures, here about $10^{-2}[2D]$, i.e., around 50 K except for choices of the values of the parameters which are unrealistic. This is the case of the lower/red curve in Figure 2, corresponding to an unrealistic value of parameter a which is proportional to the internal oscillation frequency of the solectron.

To summarize the significant results so far obtained we can safely say that the fractional power law rests on the validity of following assumptions:

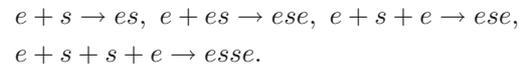
- The main contribution to conductivity is given by solectrons.
- At low enough temperature the solectron density is determined by the thermal soliton density.
- At some higher enough temperature a minimum is reached which is due to a maximum of solectron

density and to quantum effects due to solectron oscillations around the minimum of the binding energy between the electron and the soliton.

In order to go a step forward in our theory, in the next section we shall consider the contribution to conductivity of bisolectrons. We expect that bisolectrons will lead to higher values of the temperature where the minimum of resistivity appears. In order to do this we shall study the chemical equilibrium of the triplet electrons-solectrons-bisolectrons [3,4,13–15].

4 Going beyond solectrons. Contribution of bisolectrons to conductivity (a first chemical approach)

The “chemical reactions” leading to solectrons and bisolectrons hence binding one or two electrons are



To simplify we shall restrict our study to the case when only the bound species “ se ” and “ $esse$ ” are present. Indeed, the bisolectron “ ese ” can be disregarded due to its lower binding energy in comparison to that of the “ $esse$ ” and that there is full chemical equilibrium. The “ $esse$ ” bisolectrons were numerically found by Zolotaryuk et al. [27]. We set

$$\frac{x_{esse}}{x_{se}^2} = \left(\frac{T_{dse}}{T}\right)^{1/2} \exp\left(-\frac{E_{esse}}{kT}\right). \quad (14)$$

For the first reaction we assume, as above, the binding energy D whereas we take $3D$ for the second (the last) reaction. Recall that D is around 0.1–0.3 eV, and we use, for illustration, $2D \sim 0.5$ eV. In the case that the prevailing bound species are those with the highest energy, the bisolectrons “ $esse$ ” containing two solitons, the temperature dependence can be approximated as:

$$x_{esse} \approx [x_s(T)]^2 = 0.563 \tilde{T}^{2/3},$$

$$\log(1/x_{esse}) = \text{const.} - (2/3) \log \tilde{T}. \quad (15)$$

It seems reasonable to expect that including bisolectrons, and hence the additional equilibrium solectrons-bisolectrons, to the earlier given theory with only solectrons the latter would end up by providing a change in the slope thus deviating the behavior from the already established $T^{1/3}$ law. Bisolelectrons of type “ $esse$ ” (two solitons binding two electrons like H_2 molecules) have a rather high binding energy, up to $3D$, and a strongly temperature-dependent abundance in the system as depicted in Figure 3. We can see that blue curve (fraction of solectrons) starts only at higher T , since the binding energy is 3 times lower. To complete the picture the green curve depicts the fraction of free electrons (hence unbound to solitons). At $T \sim 0.1$ (in units $2D$) there is a minimum of free electrons

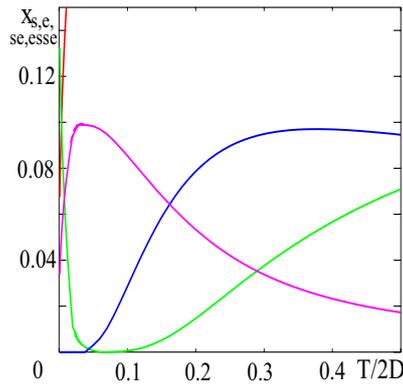


Fig. 3. Fraction of solitons “*se*” (binding energy D , middle/blue curve) and bisolotrons “*esse*” (binding energy “ $3D$ ”, upper/magenta curve) for a doping fraction $d = 0.2$. The lower/light green and the extreme left/red curves account, respectively, for the fraction of free, unbound, electrons and the soliton density (as shown in Fig. 1).

corresponding to a maximum of the bound species “*se*” and “*esse*” (recall that “*ese*” has been discarded). We see that around $\tilde{T} \sim 0.1$ nearly all electrons are bound and most of them are in the bisoloteron state.

To a first approximation the “*esse*” bisolotrons contribute to the current density (μ denote mobilities)

$$\begin{aligned} j_{esse}(T) &= n_{esse}(2e)\mu_{esse}E, \\ \sigma_{esse} &\approx nx_{esse}(T)(2e)\mu_{esse} \\ &\approx 0.563(\tilde{T})^{2/3}n(2e)\mu_{esse}. \end{aligned} \quad (16)$$

The mass action law analysis yields the range where the $T^{1/3}$ power law works which is small and the T -dependence is influenced by the chemical composition. We expect the slope of $\alpha(T)$ changing with temperature so that:

$$\log \rho_{esse}(\tilde{T}) \approx \text{const.} - \frac{\alpha(\tilde{T})}{3} \log \tilde{T}. \quad (17)$$

Including bisolotrons leads to the following general expressions for the conductivity, assuming that the contributions of solitons and bisolotrons are additive and do not interfere with each other:

$$\begin{aligned} \sigma(T) &= \frac{n_e e^2}{\rho_e(T)} + \frac{n_{se} e^2}{\rho_{se}(T)} r_{se}(T) + \frac{n_{esse} (2e)^2}{\rho_{esse}(T)} r_{esse}(T), \\ r_{se} &= 1 + \frac{m_{se} v_{se}^2}{kT_q}, \\ r_{esse} &= 1 + \frac{m_{esse} v_{esse}^2}{kT_q}. \end{aligned} \quad (18)$$

Thus, relative to the case of solely having solitons, the formation of bisolotrons not only shifts the location of the minimum to higher temperatures but also influences the power law, as depicted in Figure 4. For illustration we have used the parameter values $a_1 = 0.02$, $a_2 = 1$, $b_1 = 1000$, $b_2 = 2000$, and two doping fractions $d = 0.2$

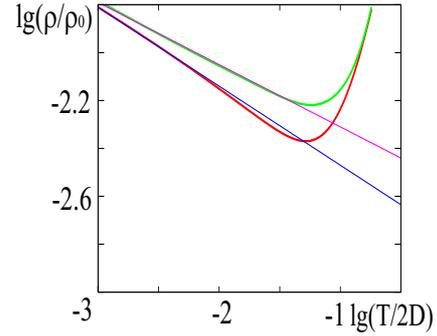


Fig. 4. Log-log plot of resistivity ρ/ρ_0 versus temperature (in $2D$ units) when both (*se*) solitons and (*esse*) bisolotrons are simultaneously contributing, and assuming as binding energies $E = -D$ and $E = -3D$, respectively. Two doping fractions have been considered ($d = 0.3$, lower/red curve, blue slope: 0.33; $d = 0.2$, upper/light green curve, magenta slope/red curve, blue slope: 0.26). It appears a weak dependence of the power law with the increase in the value of the doping fraction.

(green) and $d = 0.3$ (red). Needless to say our theory involves several “free” parameters whose values are not well known and which we had to fix based on plausibility, e.g., we assumed binding energies $E = -D$ and $E = -3D$, and for the frictions $\rho_{esse}(T) \sim \rho_{se}(T) \sim \rho_e(T)$. The resistivity was given in the units for free electrons $\rho_0 = \rho_e(T)/e^2$. Yet it appears clear that the inclusion of the “chemical equilibrium” solitons-bisolotrons significantly shifts the maximum as well as it produces a change in the shape of the curves including slowly varying slopes at high enough temperatures, with a small increase of the slope in the range $\tilde{T} \sim 0.01-0.05$. According to Figure 4, this is the region where bisolotrons are replaced by solitons in the range of $\tilde{T} \sim 0.05$.

5 Comparison with some experimental findings

Without pretending to offer a clear-cut explanation of experiments where a fractional power law and a minimum of resistivity exist in the so-called non-metallic regime of a material, we would like to point out that our predictions may underlie significant features of some experimental results. For instance, one of the experimental facts about dopable conducting polymers is the fractional dependence of their resistivity on temperature at “low enough” temperatures that may reach the range $1-10^2$ K [4,5]:

$$\rho(T) = \text{const.} T^{-\gamma}; \quad \ln[\rho(T)] = \text{const.} - \gamma \ln(T).$$

The fractional dependence with γ in the range $1/4 - 2/3$ is most often explained using Mott’s variable-range hopping theory which leads in $d = 2$ to $\gamma \sim 1/3$ and in $d = 3$ to $\gamma \sim 1/4$. However, the data do not really permit to distinguish between $1/3$ and $1/4$. This led Roth and Bleier [4] to propose the choice $1/3$ as a compromise (see Fig. 5). In all cases the value $1/3$ seems to be nearest to the data. The minimum is usually between 100 and

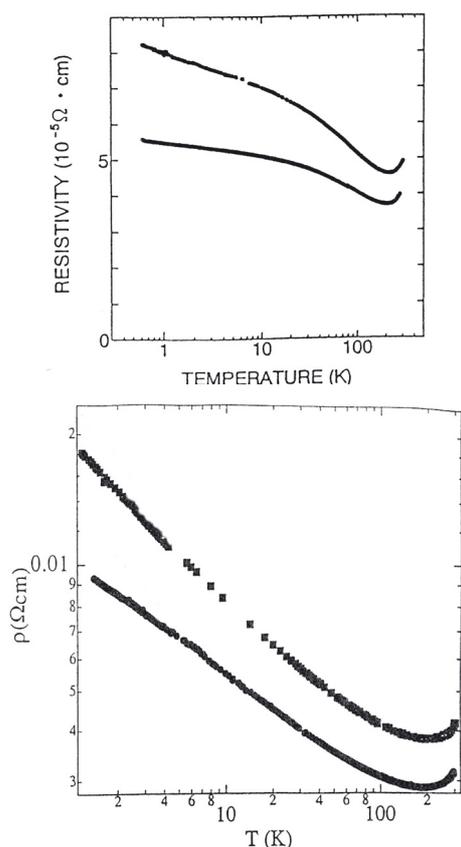


Fig. 5. Log-log plot of resistivity vs. temperature of conducting polymers in the non-metallic regime. Typically there is a range with a fractional power law in the temperature dependence and there is also a clear minimum of resistivity in a temperature range around 100 K. Upper panel is adapted from Figure 7 of Kaneko and Ishiguro (1994) for fully FeCl_3 -doped polyacetylene fresh samples [4,7]. Lower panel is adapted from Menon et al. ([5], Fig. 2.6) for two polyaniline (PANI-CSA) samples.

200 K. A close inspection shows two types of experimental situations indicated by the change of slope tending finally to the resistivity minimum. There is also the argument about a percolation phase transition of a conductive phase in an insulating matrix [28] and the idea that the electrons are transferred by crosslinks between metallic fibrils [29]. Curiously enough, data on Bechgaard salts also exhibit fractional power law with T and the existence of a minimum [30–32].

6 Concluding remarks

Our soliton-assisted transport theory which in its most complete version corresponds to transport via bisolelectrons (bosons) may be useful to explain some experimental data in, e.g., conducting polymers [3–7]. We assume a standard Hamiltonian albeit including non-standard nonlinear/anharmonic lattice dynamics using the experimentally based Morse potential rather than a harmonic potential. Further we assume that conduction is based on hopping and that an essential part of the current is driven by

charged thermal solitons which may carry with the lattice deformation one (solelectrons) or two electrons (bisolelectrons) the latter being bosons. Our theory predicts both a fractional power law $T^{1/3}$ at low enough temperatures and the existence of a minimum of resistivity upon increasing temperature. Further we predict a weak dependence of the scaling exponent on the doping fraction. We also predict curves with slowly changing slope which seem to be more realistic, in particular, with respect to the position of the minimum. The formation of the minimum follows from several effects such as, e.g., the chemical equilibria between solitons, free electrons, solelectrons and bisolelectrons and the oscillations in the soliton wells leading to bound states. In future work on two-dimensional lattices we plan to discuss percolation-type transitions as described in references [28,29,33,34].

The authors wish to express their gratitude to Harald Böttger, Leonor Cruzeiro, Jean-Pierre Launay, Bernhard Wessling and E. Guy Wilson for enlightening discussions and valuable advice. This research was supported by the Ministry of Education and Science of the Russian Federation under Grant number 1008.

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