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Interchain tunnelling of large polarons and bipolarons in quasi-1D solids

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Abstract. The formation of interchain bands for continual polarons and bipolarons in quasi-1D conducting polymers and molecular systems is considered at zero temperature. For the case of non-adiabatic t_{\perp} hopping, a qualitatively strong dependence is found of the interchain tunnelling of polarons/bipolarons on their intrachain motion and on the type of 3D ordering. Relevant experiments are proposed.

1. Introduction

It is now well established that charge carriers in quasi-one-dimensional (Q1D) solids frequently self-trap into highly anisotropic states having a large (or intermediate) size along the chains and a small one across them [1–4]. A good example is given by kink solitons, polarons and bipolarons in some conducting polymers which are fairly well described by continuum models for the intrachain (longitudinal) direction (see e.g. [2, 3]).

The transport properties of such states are clearly of interest. Therefore detailed theoretical studies have been made of their quasi-free intrachain motion (see reviews in [4]). On the other hand, only a few papers have dealt with the interchain (transverse) transfer of continual polarons and bipolarons. The Franck–Condon factor for the transverse tunnelling of a polaron in trans-polyacetylene has been computed in [5] and interchain bipolaron hops have been described in a phenomenological way in [6]. However, the natural question of what qualitative features of interchain transfer are inherent for the large-radius states has not been discussed. Meanwhile, results of experiments with oriented conducting polymers apparently imply the importance of the interchain migration in long-lived charge photoproduction and relaxation, in the conductivity, etc [7]. That is why this problem is discussed in the present paper.

Compared with small-radius polarons in conventional molecular systems [4], two specific new properties could be expected here. The first one is a connection of inter- and intrachain motions arising owing to the spatial extent of the self-trapped electron wavefunctions. The second one is a dependence of the transverse transfer on 3D ordering in the dimerisation pattern of conducting polymers inferred from the already found differences of 3D electronic spectra [8, 9], polarisational optical properties [10, 11] and polaron/bipolaron stability [9] for in-phase (*AA*) and out-of-phase (*AB*) orders. In order to demonstrate these features, we consider the extremely simplified models of the

transverse band formation for polarons/bipolarons due to interchain tunnelling at zero temperature (more details are available in [12]). Continual polarons and bipolarons will be characterised by their gravity centres (or ‘centroids’ [1]) and the above features reveal themselves through the properties of the generalised ‘interchain transfer integrals’.

2. Polaron tunnelling

Bare interchain coupling is provided, as usual, by ‘at-point’ free electron hopping t_{\perp} :

$$\mathcal{H}_{\perp} = \sum_n t_{\perp} \int dx \psi_{n+1}^{\dagger}(x) \psi_n(x) + \text{HC.} \quad (1)$$

For the interchain transfer of large polarons, however, one could propose a phenomenological description with use of continuous functions $\tau(\xi)$:

$$H_{\perp} = \sum_n \int d\xi_1 d\xi_2 \tau(\xi_2 - \xi_1) c_{n+1}^{\dagger}(\xi_2) c_n(\xi_1) + \text{HC.} \quad (2)$$

The operators $\psi_n^{\dagger}(x)$ and $c_n^{\dagger}(\xi)$ in (1, 2) create the electron or polaron respectively on the n th chain[†] with the longitudinal coordinates x or ξ (i.e. the centroid at ξ). With account of (2), the band energy of polaron is of the form

$$\mathcal{E}_{qk}^{\pm} = \mathcal{E}_P(q) \pm 2\tau_q \cos(k\alpha_{\perp}) \quad (3)$$

(q is used for the intrachain momenta and k for the interchain ones). Here $\mathcal{E}_P(q) = E_P + q^2/2M_P$ is the one-chain zero-phonon energy ($\hbar = 1$) [1], E_P and M_P being the energy at rest and effective mass correspondingly; α_{\perp} is the interchain distance and $|k| \leq \pi/2\alpha_{\perp}$. We restrict ourselves to the cases when the Fourier transform $\tau_q = \int d\xi \tau(\xi) \exp(-iq\xi)$ can be chosen real, $\tau_q = \tau_q^*$, that is $\tau(\xi) = \tau^*(-\xi)$.

Of course, the problem is to determine $\tau(\xi)$. Let us do it for non-adiabatic t_{\perp} hopping which means that, while the lattice motion is an adiabatic perturbation for the intrachain movement of electrons, the lattice itself adiabatically follows interchain electron hops. Then one can use Holstein’s approach to the small polaron motion [13] considering \mathcal{H}_{\perp} in (1) within first-order perturbation theory. The unperturbed states in our problem are

$$|mq\rangle = \delta_{nm} \varphi_m(x - \xi_m, \mathbf{Q}^{(m)}) L^{-1/2} \exp(iq\xi_m) |m\rangle \prod_{n \neq m} |n\rangle. \quad (4)$$

In the state $|mq\rangle$, the polaron with the longitudinal momentum q is on the m th chain of length L , the other chains are empty. The first two factors in (4) are the wavefunction of the self-trapped electron related to the polaron centroid, $|m\rangle$ and $|m\rangle$ are the phonon vacua of the m th chain without and with a polaron on it correspondingly. The $|m\rangle$ vacuum is defined with respect to the set of normal vibrational coordinates $\mathbf{Q}^{(m)} = \{Q_{\nu}^{(m)}\}$ where the (discrete or quasi-continuous) index $\nu \neq 0$: the zero-frequency translational mode $Q_0^{(m)}$ (see [1, 14] for 1D molecular systems with optical phonons and [15] for trans-polyacetylene) is separated and replaced by the collective coordinate ξ_m [1], as it is usually performed at the solitons’ quantisation [16]. Then the one-chain potential for electrons reads [1, 16]

$$\Delta(x - \xi, \mathbf{Q}) = \Delta_P(x - \xi) + \sum_{\nu \neq 0} Q_{\nu} \eta_{\nu}(x - \xi) \quad (5)$$

with $\Delta_P(x)$ being the static polaron deformation and $\eta_{\nu}(x)$ the orthonormalised oscillations around it. The $|m\rangle$ vacuum is defined with respect to other vibrational coordinates

[†] For simplicity, a layered 2D structure of chains (each equivalent for polarons) is considered.

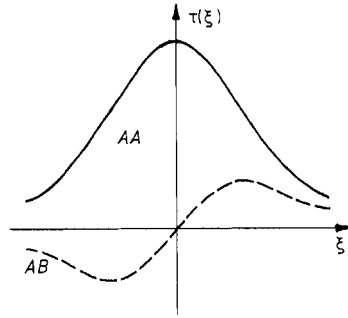


Figure 1. The qualitative shape of the function $\tau(\xi)$ for AA and AB orderings as drawn according to $t(\xi)$ in equation (8).

$U^{(m)}$ so that, when integrating over $dQ d\xi$ in the matrix elements, the Jacobian $J = \partial(U)/\partial(Q, \xi)$ should appear. Being explicitly independent of ξ , $J(Q)$ is a smooth function of Q alternating from $J \sim (\mu/\alpha_{\parallel})^{1/2}$ at $Q = 0$ to $J = 0$ at such Q that $\partial\Delta(x)/\partial x = 0$ in (5). The change of scale of the function $\Delta_P(x)$, $1/\mu$, here is $\sim l_P$, l_P being the polaron length and α_{\parallel} the longitudinal lattice spacing [12]. Thus we obtain $\langle mq | \mathcal{H}_{\perp} | m+1, q' \rangle = \tau_q \delta_{qq'}$ corresponding to equation (2) with

$$\tau(\xi) = t_{\perp} \int dx \Phi_m^*(x) \Phi_{m+1}(x + \xi) \tag{6}$$

$$\Phi_m(x - \xi_m) = \langle m | J^{1/2}(Q^{(m)}) \varphi_m(x - \xi_m, Q^{(m)}) | m \rangle. \tag{7}$$

Equations (6) and (7) allow one to study the symmetric properties of $\tau(\xi)$ as follows. In the Peierls model of conducting polymers, the one-chain electronic Hamiltonian $h(x) = -iv_F(\partial/\partial x)\sigma_3 + \Delta(x)\sigma_1$ (σ are the Pauli matrices) so that, for the potentials (5), the relationship $\Phi_m(-x) = \sigma_1 \Phi_m(x)$ is valid with accuracy to the sign [12]. On the other hand, the wavefunctions on the neighbouring chains are related as $\Phi_{m+1}(x) = \hat{A} \Phi_m(x)$, the operator \hat{A} is determined by the type of interchain ordering. Here we consider AA orders with $\Delta_m = \Delta_{m+1}$ and AB orders with $\Delta_m = -\Delta_{m+1}$ known in the polyacetylene theories [8, 9]. For coupling with the constant t_{\perp} (1), one then has $\hat{A} = 1$ for AA and $\hat{A} = \sigma_3$ for AB. Another limiting case is the complete alternation of t_{\perp} over even and odd sites $(t_{\perp})_e = -(t_{\perp})_o$. Such orders denoted in [17] and $\tilde{A}\tilde{A}$ and $\tilde{A}\tilde{B}$ correspond to $\hat{A} = \sigma_2$ and σ_1 respectively (see also [18]). Thus quite different symmetries are possible: $\tau(-\xi) = +\tau(\xi)$ in AA, $\tilde{A}\tilde{B}$ but $\tau(-\xi) = -\tau(\xi)$ in AB, $\tilde{A}\tilde{A}$, moreover $\tau(\xi) \equiv 0$ in the $\tilde{A}\tilde{A}$ phase.

The exact shape of $\tau(\xi)$ has to be found numerically but it is instructive to consider the electronic overlap

$$t(\xi) = \int dx \varphi_m^*(x) \varphi_{m+1}(x + \xi) \tag{8}$$

of φ functions at $Q = 0$ for the well known potential [2, 3] $\Delta(x; \alpha)/\Delta_0 = 1 - \mu\xi_0(t_+ - t_-)$,

$$t_{\pm} = \tanh(\mu x \pm \alpha/2) \quad \mu\xi_0 = \tanh(\alpha). \tag{9}$$

At different α values, $\Delta(x; \alpha)$ corresponds to the ground state Δ_0 , to static polaronic $\Delta_P(x)$ or bipolaronic $\Delta_{BP}(x)$. One then has $t(\xi) = \mu\xi/\sinh(\mu\xi)$ for AA order and also for molecular systems; $t(\xi) = i(f_- - f_+)$ for AB where $f_{\pm} = (\mu\xi \pm \alpha)/2 \sinh(\mu\xi \pm \alpha)$; $t(\xi) = 0$ for $\tilde{A}\tilde{A}$ and $t(\xi) = (f_+ + f_-)$ for $\tilde{A}\tilde{B}$. Probably, the ξ dependence of $\tau(\xi)$ is qualitatively similar to that of $t(\xi)$, see figure 1, but with some ‘average’ (according to (7)) values of the parameters μ , α between polaronic μ_P , α_P and the ground-state value $\alpha = 0$. The same is valid for the average value of J . τ_q in (3) behaves analogously to t_q :

$$t_q = f_0(q) = \pi^2/2\mu \cosh^2(\pi q/2\mu) \quad \text{in AA} \tag{10}$$

$$t_q = f_0(q) \sin(q\alpha/\mu) \quad \text{in AB} \tag{11}$$

$$t_q = f_0(q) \cos(q\alpha/\mu) \quad \text{in } \widetilde{AB}. \quad (12)$$

Thus polaronic $\tau(\xi)$ is expected to have a fall scale over ξ comparable with l_p ; the fall momentum for τ_q , $q_c \sim 1/l_p$, and the period of oscillations for AB , $\Delta q \sim q_c$.

The remaining factor in (7) from the overlap of truly vibrational states, denoted by $\exp(-S/2)^\dagger$, determines the magnitude of τ : the polaronic band is renormalised as $t_\perp \rightarrow \tilde{t}_\perp = t_\perp \exp(-S)$. For an optical-phonon-polaron, S can be estimated as $S \sim \varepsilon_p/\omega$, ε_p and ω being the polaron binding energy and phonon frequency respectively. However, S values become much larger for nearly topological self-trapped states, say for an acoustic-phonon-polaron or for a ‘long’ bipolaron (in analogy to the results of [19]).

3. Bipolaron tunnelling

The interchain transfer of a bipolaron with two fermions should proceed through intermediate states which, in our perturbation approach, are the one-chain polarons. The reactions of bipolaron fission (or fusion) into two polarons on the neighbouring chains, $BP_n \rightleftharpoons P_n + P_{n\pm 1}$, could phenomenologically be described by the amplitude $\Theta(\xi, \xi')$:

$$H'_\perp = \sum_n \int d\xi_1 d\xi_2 d\xi_3 (\Theta(\xi_1 - \xi_3, \xi_2 - \xi_3) (c_{n+1}^+(\xi_1) + c_{n-1}^+(\xi_1)) c_n^+(\xi_2) b_n(\xi_3) + \text{HC}) \quad (13)$$

$b_n^+(\xi)$ creates the bipolaron with the centroid at ξ . In the case of a large difference between the energies of the bipolaron and two polarons, the correction to the one-chain bipolaron energy $\mathcal{E}_{BP}(q) = E_{BP} + q^2/2M_{BP}$ due to (13) is easily calculated; its k -dependent part is

$$\mathcal{E}_\perp(q, k) = Y_q \cos(k\alpha_\perp) \quad (14)$$

where [12]

$$Y_{2q} = \frac{1}{\pi} \int_{-\infty}^{\infty} dq' \frac{\Theta^*(q + q', q - q') \Theta(q - q', q + q') + \text{HC}}{\mathcal{E}_P(q + q') + \mathcal{E}_P(q - q') - \mathcal{E}_{BP}(2q)} \quad (15)$$

and the Fourier transform $\Theta(q, q') = \int d\xi d\xi' \Theta(\xi, \xi') \exp(-iq\xi - iq'\xi')$. To determine the Θ function, the unperturbed states (4) have to be added to the analogously constructed bipolaronic states [12]. The result is

$$\Theta(\xi, \xi') = t_\perp \int dx \Phi_m^*(x) \Psi_{m+1}(x + \xi, \xi') \quad (16)$$

$$\Psi_m(x, \xi') = (m| J^{1/2}(Q^{(m)}) J_1^{1/2}(\mathbf{R}^{(m)}) \varphi_m(x - \xi', \mathbf{Q}^{(m)}) |m\rangle. \quad (17)$$

Here $|m\rangle$ is the phonon vacuum of the m th chain with bipolaron, the corresponding vibrational coordinates $\mathbf{R}^{(m)}$ are related to $\mathbf{Q}^{(m)}$ by the equation

$$\Delta_{BP}(x) - \Delta_P(x - \xi') = \sum_{\nu \neq 0} (Q_\nu \eta_\nu(x - \xi') - R_\nu \tilde{\eta}_\nu(x))$$

$\tilde{\eta}_\nu(x)$ being the normal oscillations around $\Delta_{BP}(x)$. $J_1(\mathbf{R}) = \partial(U)/\partial(\mathbf{R}, \xi)$ is analogous to J (in addition, $\mu_P \sim \mu_{BP}$). The symmetric properties of (16) can also be readily obtained: $\Theta(-\xi, -\xi') = \pm \Theta(\xi, \xi')$ with the sign ‘+’ for AA, \widetilde{AB} and ‘-’ for AB, \widetilde{AA} orders. The renormalisation of t_\perp occurs in accordance with (7, 17).

[†] Defined in such a way, the Franck–Condon factor $\exp(-S)$ deviates somewhat from that computed in [5] where the zero-frequency mode has been not separated.

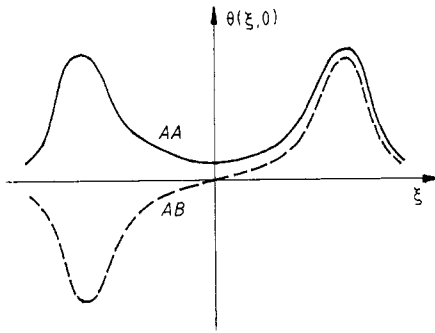


Figure 2. A sketch of the $\Theta(\xi, \xi' = 0)$ amplitude behaviour for 'long' bipolarons in the *AA* and *AB* phases.

The calculation of $\Theta(\xi, \xi')$ is obviously difficult. Let us however discuss the case of a 'long' bipolaron when $\alpha_{BP} \gg 1$ and the distance between composing kinks, $l_{BP} \sim \alpha_{BP}\xi_0$, is much larger than the kink width $\sim \xi_0 \sim l_p$. Then the distribution of self-trapped electron density resembles a dumbbell suggesting the assumption that even after 'averaging' in equations (7, 17) the function $\Theta(\xi, \xi' = 0)$ retains a double peaked structure (sketched in figure 2), the peak width comparable with l_p and interpeak distance with l_{BP} . Besides the relative changes in the peak positions and heights, there is a general tendency for $|\Theta(\xi, \xi')|$ to fall with increasing $|\xi'|$. Since at the length $\sim l_{BP}$ the vibrational overlap in (17) should decrease by the factor $\sim \exp(-C\varepsilon_p/\omega)$, $C \sim 1$, the characteristic fall scale in the ξ' direction seems to be $l' \sim l_{BP}(\omega/\varepsilon_p)$. In the ξ direction, the $|\Theta|$ falls with the scale $l \sim l_p$ and, hence, the convolution function Y_q in (15) has the characteristic fall momentum $q_c \sim 1/\max(l, l')$. On the other hand, the oscillations of Y_q are expected to have the period $\Delta q \sim 1/l_{BP} < q_c$.

4. Discussion

We have demonstrated above how the interchain tunnelling of large (i.e. extended along the chains) polarons/bipolarons is related to their intrachain motion at various 3D orderings. In terms of the group velocity v_g , the transverse components $v_{g\perp}$ become explicitly dependent on the longitudinal momenta q (3, 14). Interpreting these results one may think of some (but not complete) analogy to the optical diffraction from a finite-width slit or double slit (more strictly, one needs smooth slits with continuously decreasing transparencies [20]). For an infinitely narrow slit we would have the constant intensity diffraction picture (an analogue of small polaron tunnelling with no q dependence) but in the case of interest we have falling intensities and interference effects. So the possible oscillation-like dependence of $v_{g\perp}$ on q is caused by the interference of different tunnelling paths for polarons/bipolarons. For long bipolarons, such oscillations would occur in both *AA* and *AB* phases. For polarons in *AA* they would be absent (compare the corresponding double and single peaked amplitude structures in figures 1 and 2). The period of oscillations may be close to the characteristic fall momentum (polarons in *AB*) or smaller than it (long bipolarons). It should be noted that the approach used of a 'rigid polaron' remains valid at the values of $q \sim \mu_p$: the dependence of l_p and M_p on q becomes essential at larger $q \gg (\varepsilon_p/\omega)\mu_p$ (in symbols).

It is reasonable to define the interchain bandwidth w_\perp for q such that v_g is directed across the chains. For *AA* and *AB* orders with even τ_q (10, 12) it is the region of $q = 0$ and $w_\perp \propto \tilde{t}_\perp$. However, for the *AB* order with odd τ_q (11), the minima of the spectrum

(3) are located at finite q , at small \tilde{t}_\perp these are $\mp q_0$ with $q_0 \propto M_P \tilde{t}_\perp / \mu$ ($q_0 < \mu$ is assumed) and $w_\perp \propto M_P (\tilde{t}_\perp / \mu)^2$ (that is $w_\perp \propto \tilde{t}_\perp^2 \Delta_0 / \omega^2$ in symbols). In any case the interchain polaron mobility in the AB phase is strongly suppressed compared with AA order with the same t_\perp . As for the transverse bipolaron bandwidth in equation (15), $w_\perp \propto \tilde{t}_\perp^2$ for both AA and AB at $q = 0$ but with different signs and values of Y (the 'shorter' bipolaron the smaller w_\perp in the AB phase).

It is difficult at the present stage to make definite predictions for experiments. Tentatively, it seems that the described features could reveal themselves at low temperatures in high-quality samples of conducting polymers with large polarons/bipolarons through (non-trivial) dependences of the transverse conductivity on a longitudinally applied electric field, $\sigma_\perp = \sigma_\perp(E_\parallel)$, or as an unusual temperature behaviour of σ_\perp . Let us refer e.g. to the photoconductivity experiments in oriented trans-(CH)_x [21–23] and poly(phenylene–vinylene) [24] performed at different light polarisations and different directions of applied electric field E . We propose to perform similar experiments but with cross-imposed electric fields in order to measure the transverse current as a function of E_\parallel at fixed E_\perp . Using the data of [25] for the intrachain mobility of polarons in trans-(CH)_x, the fields $E_\parallel \sim 10^4$ – 10^5 V cm⁻¹ correspond to $v_\parallel \sim 10^6$ cm s⁻¹ or, setting $M_P \sim m_e$ [3, 25], to a length ~ 100 Å. A large quantitative uncertainty being accounted for, such a value is comparable with the 'averaged' (7) size of polaron so that the supposed dependence of σ_\perp on E_\parallel could be observed†. However, to substantiate such an assumption, the present qualitative model has to be extended in many respects including the effects of scattering by phonons and defects.

In this connection let us list some problems (partly known in the theory of small polarons [13]) for future discussion. At increasing temperatures thermally activated interchain hops of large polarons would dominate over band-type transport. Besides non-adiabatic t_\perp transfer considered in this paper, the adiabatic t_\perp regime is possible. In this case large enough t_\perp could even prevent the formation of continual polarons/bipolarons in crystals [9, 26] but we emphasise here that there may be a natural origin of one-chain polaron/bipolaron stabilisation leading also to hopping interchain transport. This is the disorder in the one-chain polaron energies (caused e.g. by the different chain lengths) that may strengthen the self-trapping effect more than additively [27]. It would also be interesting to clarify the relevance of the questions considered here for bipolaronic superconductivity [28].

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† The effects of the E influence on the exciton destruction [11, 21] and the recombination escape probability [23] should be separated properly for this, e.g. comparing the transverse and longitudinal currents.

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