

# Physics of Conductive Conjugated Polymers\*

## A Primer

*Hemanth K Bilihalli*

**In this expository article, the importance of electron-phonon coupling to charge transport in conductive polymers—as a result of a vanishing *dynamic* bandgap—is outlined. The author’s interpretation of physical models for this process: models are presented for predicting the conduction modes activated during charge transport.**

### 1. Foreword

The well-known Hückel rule of physical chemistry states that organic rings with  $(4n + 2)$  electrons in  $\Pi$ -bonded orbitals exhibit ‘resonance’ or ‘delocalization’ of electrons. A similar condition holds in linear conjugated oligomers with half this number of  $\Pi$ -bonded electrons, with facility for charge transport, in the presence of ‘dopant’. Thus, it came as a surprise in 1977 that long conjugated organic polymers were found to conduct electricity when polyacetylene was prepared by accidental addition of  $\approx 1000$  times the required amount of the polymerization catalyst (acting as a dopant). The practical applications of conductive polymers are—to quote the public announcement of the 2000 Chemistry Nobel Prize [1],

“...in low-cost manufacturing using solution-processing of film-forming polymers. Light displays and integrated circuits, for example, could theoretically be manufactured using simple inkjet printer techniques.”

Some examples of commercially employed conductive polymers



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### Keywords

Phonon-electron interactions, localization effects, hopping transport, Le polymers.

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are: *polyaniline*, *poly(ethylenedioxythiophene)*, *poly(phenylene vinylidene)*, *poly(dialkylfluorene)*, *poly(thiophene)*, *poly(pyrrole)*, etc. These are primarily employed in anti-static coatings for electronic circuits, and sometimes in transistors and capacitors. More recently, electroluminescent polymers have been discovered [*poly(p-phenylene vinylene)* in 1990] that can form the active layers in light emitting diodes (LEDs).

Apart from their technological impact, research on these materials has proven to be fertile ground for testing novel theories on conduction-insulation transition.

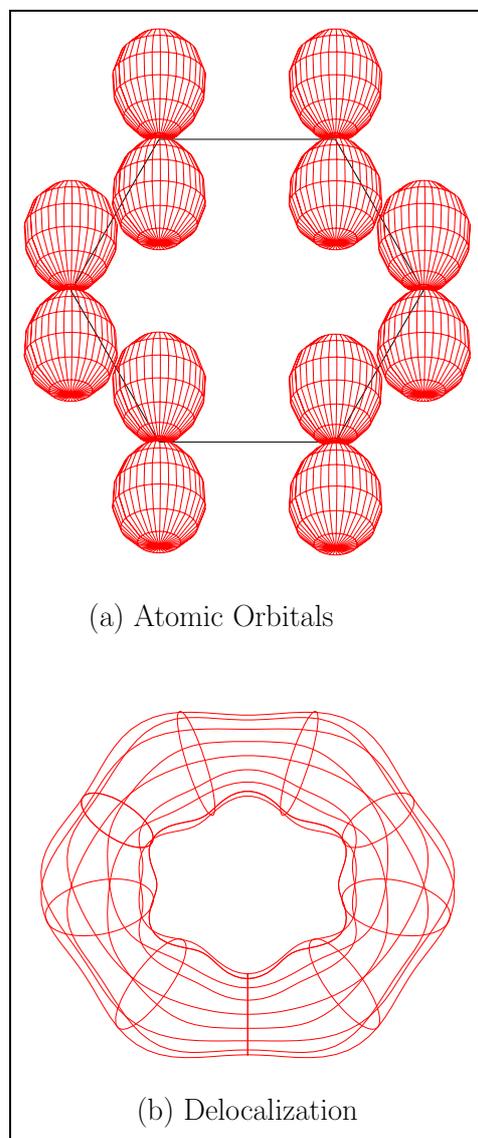
A cursory overview of conducting polymer structure via Raman spectrum analysis reveals a small (but electrically significant) alternation in bond length along the polymer chain ( $\approx 0.08\text{\AA}$  in the case of polyacetylene) [2], contrary to the “resonance school of thought” about such conjugated systems.

One of the themes of this article, is the physics of coupling between the electron and the crystal lattice, in addition to the pedagogical aim of explaining charge conduction in terms of ‘a band-gap eliminating midgap state’. This refers to an electronic state forming in the middle of the forbidden region between the conduction and the valence bands of the semiconductor structure of a polymer obtained by standard band structure calculation (see *Figure 8*, also cf. *Figure 3*).

Although the theory of Su, Schrieffer, and Heeger [2] was worked out soon (1980) after the discovery (1977) of these uncharacteristic materials, full justice with further treatment than provided here demands a foray into the quantum mechanics of two-electron interactions, suitably wrapped in the language of ‘second quantization’, for which advanced material, e.g. Altland and Simons’ book *Condensed Matter Field Theory* [3], must be consulted.

Instead, this article will verbalize the physical arguments that signal the chemical changes occurring in conjugated polymers and eventually facilitate the curious phenomenon of charge conduction. Section 2 is a first look at the electronic structure of electron-rich periodic lattices, and the only mathematical proof



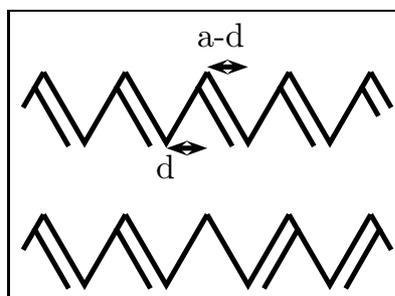


**Figure 1.** Delocalization of singly-filled  $p_z$  (atomic) orbitals on carbons of benzene to form an undulating molecule-wide band. For  $ka = 0$  the ground state for  $\frac{m_e A a}{\hbar^2} = 1$  has energy corresponding to  $\alpha a = 1.312$  (see Eq. 9).

in this work, of the phenomenon of ‘resonance’, familiar to all high-school students. Section 3 introduces the classical mechanics of vibrations in such lattices, and the two types possible when the lattice consists of a repeating positive-negative-ion unit cell.

Such vibrations are quantized for the ideal scenario we consider

**Figure 2.** *trans*-polyacetylene in insulating (top) and nearly-conducting (bottom) forms. A neutral ‘soliton’ (‘kink’ in center, *S* in Figure 8) forms in the latter case, due to a ‘Peierls instability’ [3].

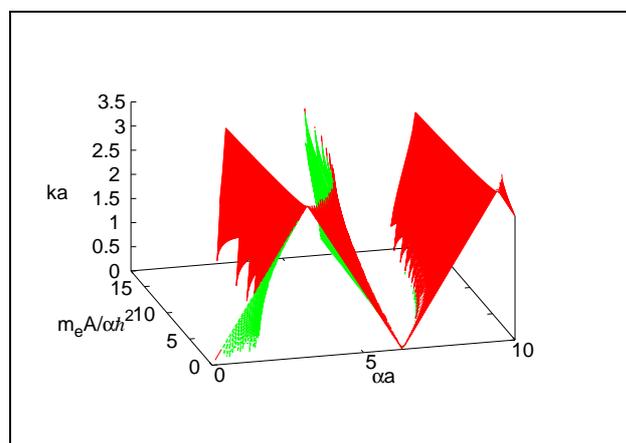


here—even the smallest degree of vibration has a ‘zero-point energy’ [4]—and are then known by the quantum mechanical term ‘phonons’. Section 4 is a syncretism of Sections 2 and 3 and specifies the ‘rules’ that these quantized vibrations must follow, including the ‘common-sensical rule’ of energy conservation. In Section 5, we introduce the first signatures of a current-producing non-equilibrium state: ‘localization’ and ‘soliton’. In analogy with ‘semiclassical dynamical theories’ [5] of solid-state (*e.g.* silicon-based) semiconductors, any realization of a finite current generating non-equilibrium state is possible when quantum mechanical dynamics is also included in the governing model(s). Such topics are deemed too advanced for our overview, so we draw analogies for these concepts from classical counterparts. Finally, Section 6 hints at the soliton pair (‘polaron’) production process induced by the addition of an external charge/application of external potential. Here we sketch an argument due to Sethna [6], supported by figures, for the charge transport mechanism of pairwise electron hopping.

## 2. Periodicity, Tunneling/Resonance [7]

$\Pi$ -electrons in a conjugated (i.e. alternating double-single bond) molecule such as benzene undergoes ‘hybridization’ and subsequently, resonance.

We start with a familiar high school principle which teaches us that the  $\Pi$ -electrons in a conjugated (i.e. alternating double-single bond) molecule such as benzene undergoes ‘hybridization’ and subsequently, resonance. The  $p_z$  orbital on each carbon atom has a free electron which chooses to delocalize over the entire



**Figure 3.** Band structure of a lattice ring of point ( $\delta$ -potential) ions.

molecule and form, along with the other five  $p_z$  electrons, a tubular orbital ('energy band'), spread over the entire benzene ring. The root cause of this is the *quantum mechanical* process of 'tunneling'. In this section, we present an elementary discussion of the process of energy band formation, starting from the Schrödinger equation.

At a first level of approximation, the benzene molecule is a lattice ring of point ( $\delta$  potential) ions.

Being periodic, one may expand the wave function on this lattice in a fourier series

$$\psi_k(x) = \sum_K \hat{u}_k(K) e^{i(k+K)x}, \quad (1)$$

where  $K$  is restricted to integer multiples of  $\frac{2\pi}{a}$ ,  $a$  being the 'lattice constant' (or ion-ion separation).

The Schrödinger equation for the wave function of the electron

$$H\psi_k = E_k\psi_k, \quad (2)$$

thus reduces to

$$\begin{aligned} -\frac{\hbar^2}{2m_e} \frac{d^2\psi_k}{dx^2} + V(x)\psi_k(x) &= E_k\psi_k \quad (3) \\ \Rightarrow \left[ \frac{\hbar^2(k+K)^2}{2m_e} - E_k \right] \hat{u}_k(K) + \sum_{K'} \hat{V}(K-K') \hat{u}_k(K') &= 0. \end{aligned}$$

Substituting the fourier transform of  $V(x) = A \sum_{n=-\infty}^{\infty} \delta(x - na)$ ,

$$\hat{V}(K) = \frac{1}{a} \int_{-a/2}^{a/2} dx V(x) e^{-iKx} = \frac{A}{a}, \quad (4)$$

into the above yields

$$\left[ \frac{\hbar^2(k + K)^2}{2m_e} - E_k \right] \hat{u}_k(K) + \frac{A}{a} \sum_{K'} \hat{u}_k(K') = 0. \quad (5)$$

Solving for  $\hat{u}_k(K)$

$$\frac{\hat{u}_k(K)}{\sum_{K'} \hat{u}_k(K')} = \frac{2\pi A / (a\hbar^2)}{2\pi m_e E_k / \hbar^2 - (k + K)^2}, \quad (6)$$

and summing over  $K$  yields an identity (LHS=1) and a relation between  $E_k$  and  $k$  ('band structure'). Defining  $\alpha^2 \equiv 2m_e E_k / \hbar^2$  and replacing the sum over  $K$  to one over index  $n$ , the above identity simplifies to

$$\begin{aligned} \frac{\hbar^2 a}{2m_e A} &= \sum_{n=-\infty}^{\infty} \frac{1}{\alpha^2 - \left(k + \frac{2\pi n}{a}\right)^2} \\ &= -\frac{a}{4\alpha} \sum_{n=-\infty}^{\infty} \left[ \frac{1}{\pi n + \frac{ka}{2} - \frac{\alpha a}{2}} - \frac{1}{\pi n + \frac{ka}{2} + \frac{\alpha a}{2}} \right]. \end{aligned} \quad (7)$$

Here we use a mathematical trick

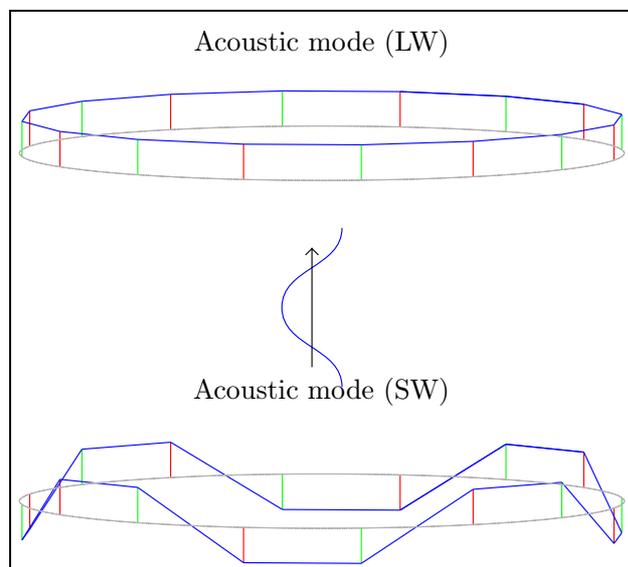
$$\cot x = \sum_{n=-\infty}^{\infty} \frac{1}{n\pi + x}, \quad (8)$$

and with a little trigonometry arrive at

$$\boxed{\cos(ka) = \cos(\alpha a) + \frac{m_e A}{\alpha \hbar^2} \sin(\alpha a)}, \quad (9)$$

the final result of our analysis, a transcendental relation between  $\alpha$  and  $k$ .

The function  $\cos(ka)$  being limited to the range  $[-1, 1]$ , has a support of  $ka$  in the range  $[0, 2\pi)$ . Higher wavenumbers  $ka$  are folded back into this range as  $ka \bmod 2\pi$ . Correspondingly, the surface represented by Eq. 9—see *Figure 3* for a plot—exhibits gaps



**Figure 4.** Time-lapse pictures of long ( $k = 0$ ) and short ( $k = \frac{\pi}{a}$ ) acoustic modes of a 1D crystal lattice, comprising two equispaced sublattices (of  $N_c$  particles each), vibrating in-phase. Displacement vectors of sublattice particles (in red and green) are to be understood as being tangential to the lattice ring, and not perpendicular, as shown.

where  $\cos^{-1}$  can't be defined. The visible surface spans the permissible regions for the energy of the electronic wave function. The lowest energy permitted for non-zero  $A$  is strictly positive and can be assumed to be the ground state for when the electron delocalizes [say, over a benzene ring (see *Figure 1* for the electronic density {wave function squared} of the ground state for  $\frac{m_e A a}{\hbar^2} = 1$ )].

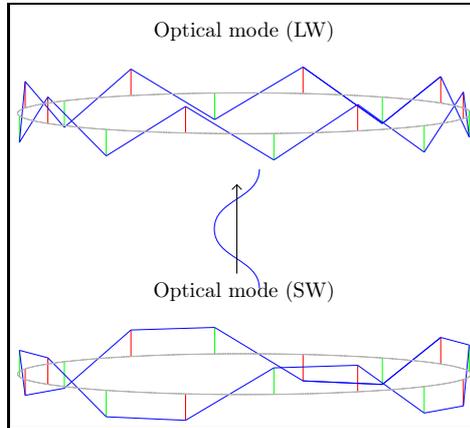
### 3. Lattice Vibrations, Normal Modes [8]

Atoms are not fixed on a lattice and at high temperatures oscillate about equilibrium positions. Interaction of light or electricity with solids necessarily involves coupling to vibrations of the crystal lattice. In this regard, the following is a quick survey of the theory of lattice vibrations, for the 1D case of a conductive polymer backbone with a non-identical ion or atom basis [5].

We consider a diatomic lattice, as is suitable for a conjugated polymer, with bond length alternating between single and double bonds. Each 'unit cell' then consists of a basis of two dissimilar particles (atoms or ions), and its position is specified in units of

Atoms are not fixed on a lattice and at high temperatures oscillate about equilibrium positions.

**Figure 5.** Time-lapse pictures of long ( $k = 0$ ) and short ( $k = \frac{\pi}{a}$ ) optic modes of a 1D crystal lattice, comprising two equispaced sublattices (of  $N_c$  particles each), vibrating out-of-phase.



the equilibrium lattice spacing  $a$ . Within each cell, the dissimilar particles are separated by the equilibrium distance (‘sublattice spacing’)  $d$  (or  $a - d$ ) (see *Figure 2*).

In a dynamic situation, each particle interacts with its neighbor through a harmonic (‘spring’) potential with ‘stiffness’ varying with equilibrium spacing ( $d$  or  $a - d$ ).

In a dynamic situation, each particle interacts with its neighbor through a harmonic (‘spring’) potential with ‘stiffness’ varying with equilibrium spacing ( $d$  or  $a - d$ ). Denote the dynamic position of particle  $c$  in the unit cell numbered  $P$  as

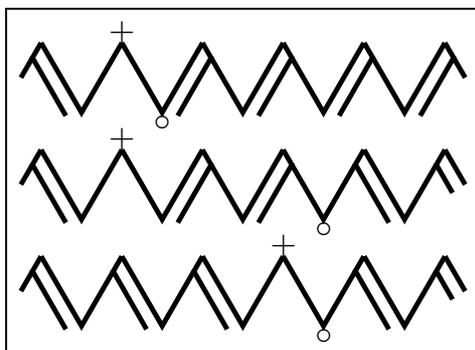
$$\begin{aligned} x_1^P &= Pa + d + u_1^P \quad (c = 1), \\ x_2^P &= (P + 1)a - d + u_2^P \quad (c = 2), \end{aligned} \quad (10)$$

where  $u_c^P$  is the displacement from equilibrium. Recognizing that these polymer lattices are nearly infinite (bulk condition), the recourse is to assume that all similar particles (within the  $c$ -type sublattice) move in phase, and form a periodic boundary, in an approximation due to von Karman and Born [5] (see *Figures 4* and *5* for illustration).

The sublattice vibrations are coupled, i.e.  $u_1$  in any cell couples to  $u_2(s)$  in the same cell. But one may diagonalize these coupled linear second-order ODEs, and solve for independent eigenmodes, i.e. one may picture each sublattice to be vibrating independently, so that

$$u_c^P(k) = e_c(k) \exp[i(k.Pa - \omega t)], \text{ for } c = 1, 2. \quad (11)$$





**Figure 6.** Migration of radical cation (‘polaron’) formed by the removal of one electron ( $S^+$  in *Figure 8*) [1].

Note the ‘polarization vector’  $e_c(k)$  differs only for different atoms in the same cell, but not for equivalent atoms in different cells. For a periodic lattice, the two sublattices move either in-phase (‘acoustic’, see *Figure 4*) or out of phase (‘optic’, see *Figure 5*) for the shortest and longest wavevector vibrations)<sup>1</sup>.

#### Short Wavelength (SW) Limit of Normal Modes

For the specific ‘short-wavelength’ (SW) case ( $k = \frac{\pi}{a}$ ) illustrated in *Figures 4* and *5*, motion changes by  $180^\circ$  from cell to cell. Because the spacing and spring strengths are the same for both sublattices, the acoustic and optic modes appear identical but for a phase-shift. Both modes travel at the same speed, for this special case.

The polarization vector (for the direction of oscillation in the wave amplitude) is tangent to the circular lattice, moving in the direction as indicated by the winding corkscrew in the figure’s center (see *Figures 4* and *5*).

#### 4. Simple Models of Electron-phonon Coupling

A feature of electron-phonon coupling is that an electron couples to lattice motion, and so to atomic velocities, so this interaction energy is to leading order

$$\mathcal{H}_{ep} = \sum_{P,c} \mathbf{x}_{P,c} \cdot \left[ \nabla_{\mathbf{x}_{P,c}} V(\mathbf{r}_e) \right]_{\text{all } \mathbf{x}_{P,c}=0}, \quad (12)$$

<sup>1</sup>One may even picture each of these modes as in an electromagnetic wave:  $\mathbf{E}$  and  $\mathbf{B}$  vibrating in phase in a linearly polarized wave, and out of phase in a circularly polarized wave.

where  $\mathbf{r}_e$  are electronic coordinates,  $V(\mathbf{r}_e)$  is the electron-ion interaction referred to in Section 2. Now, as the *lattice is periodic*, and only nearest neighbor atoms/ions interact, an electron of wavevector  $\mathbf{k}$  is transformed into wavevector  $\mathbf{k}'$  subject to conservation of wavevector addition or subtraction to a wavevector  $\mathbf{b}$  in the ‘reciprocal lattice’,

$$\mathbf{k}' - \mathbf{k} - \mathbf{q} = \mathbf{b}. \quad (13)$$

With the additional assumption that the electrons respond instantaneously to ionic movement, one may also derive the condition that optic phonons do not affect electrons while acoustic phonons do (see Appendix N of [5] or [9] for proof). Patterson and Bailey [9] offer the qualitative explanation:

“...in optic modes the adjacent atoms tend to vibrate in opposite directions, and so the net effect of the vibrations tends to be very small due to cancellation [cf. with *Figure 5*]”.

Finally, in all cases, the energy conservation rule

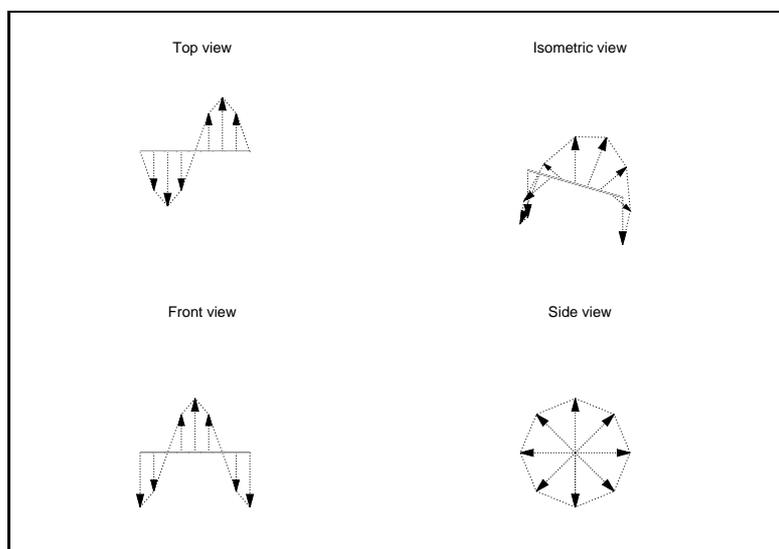
$$E_{\mathbf{k}'} = E_{\mathbf{k}} + \hbar\omega(\mathbf{q}) \quad (14)$$

must be satisfied [9].

## 5. Non-equilibrium States and Solitons

In the non-equilibrium state, the picture of particle dynamics presented in *Figures 4* and *5* deviates from the actual dynamics presented in *Figure 6* primarily due to quantum mechanical reasons of ‘localization’ [10]. This means that the soliton shape of particle motion is an attenuated beat pattern—quite like the emblem of *Resonance!*—best characterized as classical shuttling of a particle between two wells separated by a metastable barrier. The classical ‘sine-Gordon soliton’ is the continuum representation of coupled pendula (refer to *Figure 7*) in a vertical gravitational field with (cone-shaped) neighboring bobs strung together with





**Figure 7.** A topological soliton in classical mechanics. The conical pendulum bobs are in dynamic equilibrium as the locus of inextensible cords stringing them together winds over the suspension rod and swings between positions of lowest overall energy in a gravitational field.

inextensible cords. However, sine-Gordon dynamics fluctuate between angular positions ( $180^\circ$  apart) of energy minima in the gravitational field, whereas a soliton fluctuates between minima of the classical ‘action’—the equivalent of the energy to be minimized in (position, momentum)—space.

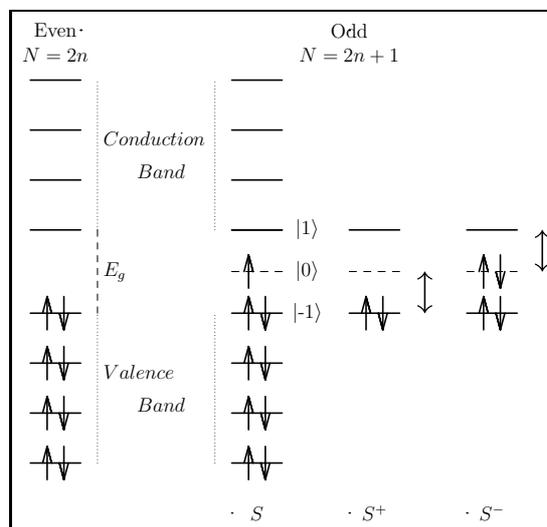
This does not mean that *Figure 4* is completely irrelevant. Solitons of somewhat smaller sizes [ $L \approx 6$  bond lengths for polyacetylene [2]  $\psi \sim \text{sech}\left(\frac{x}{L}\right) \cos\left(\frac{\pi x}{a}\right)$ ] form the backbone of electronic structure studies of conductive polymers [11]. These are modulated at the band edges  $k = \frac{\pi}{a}$  to preserve the periodic symmetry of the excitation. Furthermore these may be of the ‘topological type’, exhibiting a ‘winding around’—just like the coupled pendula in *Figure 7*—in state space, of an ‘action angle’ parametrizing the tunneling process.

## 6. Charge Transport

We stop at the penultimate topic in the process of formulation of the successful model of Su–Schrieffer–Heeger and reiterate that this is outside our purview. Instead, a layman’s description of the



**Figure 8.** Electronic configuration of acetylene oligomers. The odd bond-length chains are susceptible to Peierls instability, and form conducting diamagnetic ('magnetic-field-hating') structures on addition ( $S^-$ )/removal ( $S^+$ ) of an electron [12].



semi-classical model equivalent that also leads to the mechanism of charge transport will be provided.

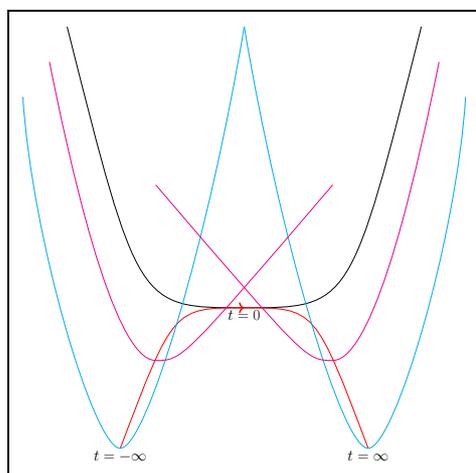
The electronic configuration of linear conjugated oligomers is shown in *Figure 8*, and is said to be half-filled, as the number of electrons is the same as the total number of (bonding and anti-bonding) orbitals. Before pairing up, a conjugated arrangement forms, with bond length alternation. This is known as 'Peierls instability' [3].

For an odd bond-length polymer, a 'midgap state' forms which develops further into two such mid-gap states on addition or removal of an electron.

For an odd bond-length polymer (see *Figure 2* for the 'kink' or 'soliton' configuration), a 'midgap state' forms (see  $S$ ,  $S^+$ ,  $S^-$  in *Figure 8*) which develops further into two such mid-gap states ('polaron' configuration) on addition or removal of an electron [12].

It is the latter which is freely conducting, the mechanism [1] for which is sketched out in *Figure 6*. This picture requires an external force (such as applied electric field) to move either the radical or charge on the polymer backbone, the justification for which may be found in numerical simulation literature [13].

In the following, we speculate on a more accurate quantitative explanation for phonon-mediated charge transport due to Sethna



**Figure 9.** Polymer lattice relaxation as habiting electron pair transitions between neighboring potential wells.

[6]. Without delving into the physics of this model, we present the reader with a (lay) description and a picture (*Figure 9*). The actual mode of charge transport is considerably complex as recently worked out computationally by Lin et al. [11].

Electrons under the influence of the phonon-coupling tend to ‘pair up’ and move together in concert. Sethna offers a vivid ‘rubber-sheet analogy’ for the description of ‘self-trapping’ [6]:

“...the second electron see[s] the potential hole sunk by the first, and together [makes] an even deeper hole.”

Further, as the electron pair attempts to ‘hop’ from one (nearly quadratic) potential well to another, the polymer lattice adapts to yield a joint (nearly quartic) potential with a broader well at the middle of the transport process, so that the phonon relaxation facilitates initially but traps finally (*Figure 9*). The reader may wonder why the two electrons do not repel in their pair-like configuration; one possible solution to this riddle is:

“The paired states are energetically favored, and electrons go in and out of those states preferentially.”

quoting the *Wikipedia* entry [14] on similarly-paired electrons in superconductivity.

Electrons under the influence of the phonon-coupling tend to ‘pair up’ and move together in concert.

## 7. Afterword

This article does not address the necessity of doping to induce charge transport through soliton propagation. Nor does it explain how midgap states induce the spontaneous formation of electron-hole pairs (excitons) that can tunnel through for current transport.

As long as the conducting localized units (solitons or polarons or bipolaron pairs) remain small compared to the polymer chain length, the physics doesn't change much, and models may be retained by adjusting parameters [15] to fit experimental data and/or simulation results.

The interested reader thrown off by jargon used here is encouraged to follow, with a little effort in matrix algebra and calculus, the online exposition of Zhu [16], complete with illuminating figures, for details on second quantization, Peierls instability, and topological solitons.

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## Suggested Reading

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## GENERAL ARTICLE

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