

SUPPORTING INFORMATION

Reversed Conductance Decay of 1D Topological Insulators by Tight-Binding Analysis

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1. Su-Schrieffer-Heeger Model

The Su-Schrieffer-Heeger model on isolated polyacetylene chain has been well-described elsewhere¹⁻³ and we included it here for completeness. The single-particle Hamiltonian is defined as

$$\mathbf{H} = -t_1 \sum_{m=1}^M (|m, B\rangle\langle m, A| + H. c.) - t_2 \sum_{m=1}^{M-1} (|m+1, A\rangle\langle m, B| + H. c.) \quad (1)$$

where M is the number of unit cells, A and B are the two sublattices (see Figure 1A), and t_1, t_2 are the intracell and intercell hopping constants. The property of the bulk states should not depend on the two edges. Here we apply periodic boundary conditions to close the bulk part of the chain into a ring. The corresponding bulk Hamiltonian is written as

$$\mathbf{H}_{\text{bulk}} = \sum_{m=1}^M (-t_1 |m, B\rangle\langle m, A| - t_2 |(m \bmod M) + 1, A\rangle\langle m, B|) + H. c. \quad (2)$$

Since the bulk has translation invariance, we apply Bloch's theorem and look for eigenstates in a plane wave form. The plane wave basis states transformed from the site basis are

$$|k\rangle = \frac{1}{\sqrt{M}} \sum_{m=1}^M e^{imk} |m\rangle \quad (3)$$

for $k \in \{\delta_k, 2\delta_k, \dots, M\delta_k\}$ with $\delta_k = 2\pi/M$. Therefore, the bulk momentum-space Hamiltonian is defined as

$$\mathbf{H}(k) = \langle k | \mathbf{H}_{\text{bulk}} | k \rangle = \sum_{\alpha, \beta \in \{A, B\}} \langle k, \alpha | H_{\text{bulk}} | k, \beta \rangle \cdot |\alpha\rangle\langle\beta| \quad (4)$$

The eigenvector $|u_n(k)\rangle$ is defined as a linear combination of sublattice states $|A\rangle$ and $|B\rangle$.

$$|u_n(k)\rangle = a_n(k)|A\rangle + b_n(k)|B\rangle \quad (5)$$

With matrix representation, the Hamiltonian and the eigenvector becomes

$$\mathbf{H}(k) = - \begin{pmatrix} 0 & t_1 + t_2 e^{-ik} \\ t_1 + t_2 e^{ik} & 0 \end{pmatrix} = -\hat{\mathbf{o}}_x (t_1 + t_2 \cos k) - \hat{\mathbf{o}}_y t_2 \sin k \quad (6)$$

and

$$|u_n(k)\rangle = \begin{pmatrix} a(k) \\ b(k) \end{pmatrix} \quad (7)$$

For equation (6), in the dimerization limits, if $t_1 = 1, t_2 = 0$, which means that all the intercell hopping is forbidden, $\mathbf{H}(k) = -\hat{\mathbf{c}}_x$. Otherwise, if $t_1 = 0, t_2 = 1$, which means that each dimer is made by sublattices of two adjacent unit cells, $\mathbf{H}(k) = -\hat{\mathbf{c}}_x \cos k - \hat{\mathbf{c}}_y \sin k$. In both cases, the eigenvalues are $E(k) = \pm 1$, independent of k , which indicates zero-group velocity along the chain.

However, at the long chain limit (the thermodynamic limit), the edge states are at zero-energy. To solve for the edge states, one needs to investigate the zero-energy eigenstates of the single particle Hamiltonian (equation (1)),

$$\mathbf{H} \sum_{m=1}^M (a_m |m, A\rangle + b_m |m, B\rangle) = 0 \quad (8)$$

where a_m and b_m are the coefficients dependent on the unit cell index. By substituting equation (1) to equation (8), we obtain $2M$ separate recursive equations

$$\begin{cases} t_1 a_m + t_2 a_{m+1} = 0 \\ t_2 b_m + t_1 b_{m+1} = 0 \end{cases} \quad (9)$$

with $m = 1, \dots, M - 1$. At boundaries, we have $t_1 a_M = 0$ (when $m = M$), and $t_1 b_1 = 0$ (when $m = 0$). The solutions are easily obtained as below.

$$\begin{cases} a_m = \left(\frac{-t_1}{t_2}\right)^{m-1} a_1 & (m = 2, \dots, M) \\ b_m = \left(\frac{-t_1}{t_2}\right)^{M-m} b_M & (m = 1, \dots, M - 1) \end{cases} \quad (10)$$

We also need $b_1 = a_M = 0$ to fulfill the boundary conditions. Moreover, we can translate the above solutions to

$$\begin{cases} |a_M| = |a_1| e^{\frac{M-1}{\xi}} \\ |b_1| = |b_M| e^{\frac{M-1}{\xi}} \end{cases} \quad (11)$$

In equation (11) we define $\xi \equiv 1/|\ln(t_1/t_2)|$ as the delocalization length. Because of the nature of the exponential relations, we can never achieve a zero-energy eigenstate. However, if $M \rightarrow \infty$, there could be two approximate solutions.

$$\begin{cases} |L\rangle = \sum_{m=1}^M a_m |m, A\rangle \\ |R\rangle = \sum_{m=1}^M b_m |m, B\rangle \end{cases} \quad (12)$$

where a_m and b_m values are indicated by the solutions above. a_1 and b_M are used to fix the norm of $|L\rangle$ and $|R\rangle$, respectively.

In the short chain limit, the two edge states, $|L\rangle$ and $|R\rangle$, could couple and generate a small energy splitting. The overlap is approximated as

$$\langle R | \mathbf{H} | L \rangle = \left| a_1 e^{-\frac{M-1}{\xi}} t_1 b_M \right| e^{i\phi} \quad (13)$$

where $\phi \in [0, 2\pi)$ marks the phase difference. The new eigenstates are

$$\begin{cases} |0_+\rangle = \frac{1}{\sqrt{2}} \left(e^{-\frac{i\phi}{2}} |L\rangle + e^{\frac{i\phi}{2}} |R\rangle \right) \\ |0_-\rangle = \frac{1}{\sqrt{2}} \left(e^{-\frac{i\phi}{2}} |L\rangle - e^{\frac{i\phi}{2}} |R\rangle \right) \end{cases} \quad (14)$$

and the corresponding energies are

$$E_{\pm} = \pm \left| a_1 e^{-\frac{M-1}{\xi}} t_1 b_M \right| \quad (15)$$

2. Calculating $T(E)$ and $T(E_F)$

This derivation follows closely the discussion found in these books^{4, 5}. In the molecular junction model, \mathbf{H}_0 describes the Hamiltonian of the isolated single molecule. When two electrodes are appended to the molecule, it becomes an open system. The Hamiltonian of this system includes the interaction between the molecule and electrodes,

$$\tilde{\mathbf{H}} = \mathbf{H}_0 + \boldsymbol{\Sigma} = \mathbf{H}_0 + \boldsymbol{\Sigma}_L + \boldsymbol{\Sigma}_R \quad (16)$$

where $\boldsymbol{\Sigma}_L = \Sigma_L|1\rangle\langle 1|$ and $\boldsymbol{\Sigma}_R = \Sigma_R|N\rangle\langle N|$ are the self-energies of the left and right electrodes (assuming the molecular system consists of N sites). These self-energy terms are non-Hermitian, which are described by Γ_L and Γ_R , the scattering rates at molecule-electrode interface on each side.

$$\Gamma_L = i(\Sigma_L - \Sigma_L^\dagger) = -2\text{Im}[\Sigma_L] \quad (17)$$

$$\Gamma_R = i(\Sigma_R - \Sigma_R^\dagger) = -2\text{Im}[\Sigma_R] \quad (18)$$

The shift of molecular orbital (MO) energy due to the self-energies are:

$$\Delta_L = \Sigma_L + \Sigma_L^\dagger = 2\text{Re}[\Sigma_L] \quad (19)$$

$$\Delta_R = \Sigma_R + \Sigma_R^\dagger = 2\text{Re}[\Sigma_R] \quad (20)$$

For the tight-binding simulations, we assumed the *wide-band limit* (the coupling between molecule and electrodes is energy-independent). By neglecting Δ_L and Δ_R in the weak coupling regime, the self-energies become,

$$\begin{cases} \Sigma_L = -\frac{i}{2}\Gamma_L \\ \Sigma_R = -\frac{i}{2}\Gamma_R \end{cases} \quad (21)$$

and only affect the terminal sites of the molecule.

Since $\tilde{\mathbf{H}}$ is non-Hermitian, its eigenstates and eigenvalues are:

$$\tilde{\mathbf{H}}|\psi_i\rangle = (\mathbf{H}_0 + \boldsymbol{\Sigma})|\psi_i\rangle = z_i|\psi_i\rangle \quad (22)$$

Note that the eigenvalues z_i are complex and the eigenstates $|\psi_i\rangle$ do not form an orthogonal set. The Hermitian adjoint of equation (22) is:

$$\tilde{\mathbf{H}}^\dagger |\bar{\psi}_i\rangle = (\mathbf{H}_0 + \boldsymbol{\Sigma}^\dagger) |\bar{\psi}_i\rangle = z_i^* |\bar{\psi}_i\rangle \quad (23)$$

$|\psi_i\rangle$ and $|\bar{\psi}_i\rangle$ form a bi-orthonormal set in which $\langle \bar{\psi}_i | \psi_j \rangle = \delta_{ij}$. The Green's function matrix $\mathbf{G}(E)$ can be written in terms of these eigenstates and eigenvalues as:

$$\mathbf{G}(E) = [E\mathbf{I} - \tilde{\mathbf{H}}]^{-1} = \sum_i \frac{|\psi_i\rangle\langle\bar{\psi}_i|}{E - z_i} \quad (24)$$

where \mathbf{I} is the identity matrix and E is the energy of the transmitting electron. The energy-dependent transmission is:

$$T(E) = \text{Tr}[\boldsymbol{\Gamma}_L \mathbf{G} \boldsymbol{\Gamma}_R \mathbf{G}^\dagger] \quad (25)$$

where \mathbf{G} and \mathbf{G}^\dagger are the retarded and advanced Green's functions respectively, and $\boldsymbol{\Gamma}_{L/R}$ are the coupling matrices ($\boldsymbol{\Gamma}_L = \Gamma_L |1\rangle\langle 1|$ and $\boldsymbol{\Gamma}_R = \Gamma_R |N\rangle\langle N|$). For transmission at Fermi energy (E_F , with $E_F = 0$), the Green's function can be simplified to

$$\mathbf{G}(E_F) = [\tilde{\mathbf{H}}]^{-1} = \sum_i \frac{|\psi_i\rangle\langle\bar{\psi}_i|}{-z_i} \quad (26)$$

For an electron propagating across the junction, $T(E)$ is dictated by the $(1,N)$ element of the Green's function in its matrix representation⁶,

$$T(E) = \Gamma_L \Gamma_R |\mathbf{G}_{1,N}|^2 \quad (27)$$

where 1 and N are indices for the terminal sites. With Cramer's rule, $T(E)$ at E_F can be written as⁷

$$T(E_F) = \Gamma_L \Gamma_R \left| \frac{\Delta_{N,1}}{\det(\tilde{\mathbf{H}})} \right|^2 \quad (28)$$

in which $\Delta_{N,1}$ is the minor determinant of $\tilde{\mathbf{H}}$.

3. Quantum Interference in 1D Topological Chain

To evaluate quantum interference, we seek to express the transmission function as a sum of complex transmission coefficients for each resonance (i),

$$T(E) = \left| \sum_i t_i(E) \right|^2 \quad (29)$$

We can expand the Green's function (equation (27)) about its eigenbasis, yielding

$$t_i(E) \approx \sqrt{\Gamma_L \Gamma_R} \frac{C_{i,L} C_{i,R}}{E - z_i} \quad (30)$$

where $C_{i,L/R}$ are the coefficient of the molecular conductance orbital (MCO) index i at the left and right molecule-electrode interface. We make the approximation that the MCO coefficients are similar to the MO coefficients of the isolated Hamiltonian (as this is in the weak-coupling limit). In equation (22), the eigenvalues z_i take the form,

$$z_i = \varepsilon_i - i\gamma_i \quad (31)$$

where γ_i represents the coupling to the electrodes,

$$\gamma_i = \frac{1}{2}(\gamma_i^L + \gamma_i^R) \quad (32)$$

and

$$\begin{cases} \gamma_i^L = \Gamma_L C_{i,L}^2 \\ \gamma_i^R = \Gamma_R C_{i,R}^2 \end{cases} \quad (33)$$

We can express the transmission coefficient in polar form,

$$t_i(E) = \pm \sqrt{T_i^{\max}} \sin(\varphi_i) e^{i\varphi_i} \quad (34)$$

where \pm is given by $\text{sgn}[C_{i,L} C_{i,R}]$ and we have defined,

$$\varphi_i(E) = \arg(E - z_i^*) \quad (35)$$

and

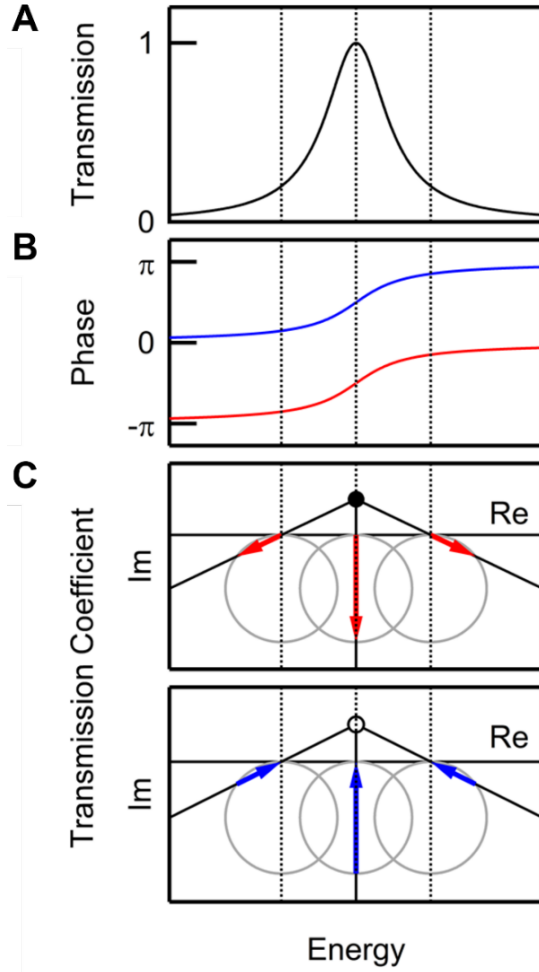


Figure S1. (A) Transmission probability vs energy for a single level (B) Phase of transmission coefficient vs energy for in-phase (red) and out-of-phase (blue) molecular orbital (C) Geometric construction of transmission coefficient from the complex eigenvalue for the in-phase (red) and out-of-phase (blue) orbitals. The centers of the grey circles are located at different energies. The diameters of the circles are equal to T^{\max} . The chord vectors are generated by connecting the resonance dot (black or white) to the tangent point of each circle to the horizontal line in which $\text{Im} = 0$. The magnitude of the transmission coefficient as a function of energy is obtained by the ratio of the magnitude of the vectors to the diameter of the circle.

$$T_i^{\max} = \frac{4\gamma_i^L \gamma_i^R}{(\gamma_i^L + \gamma_i^R)^2} \quad (36)$$

This leads to a geometric construction of the transmission coefficient (Figure S1). We will use this formalism to understand the conductance of the 1D topological chain.

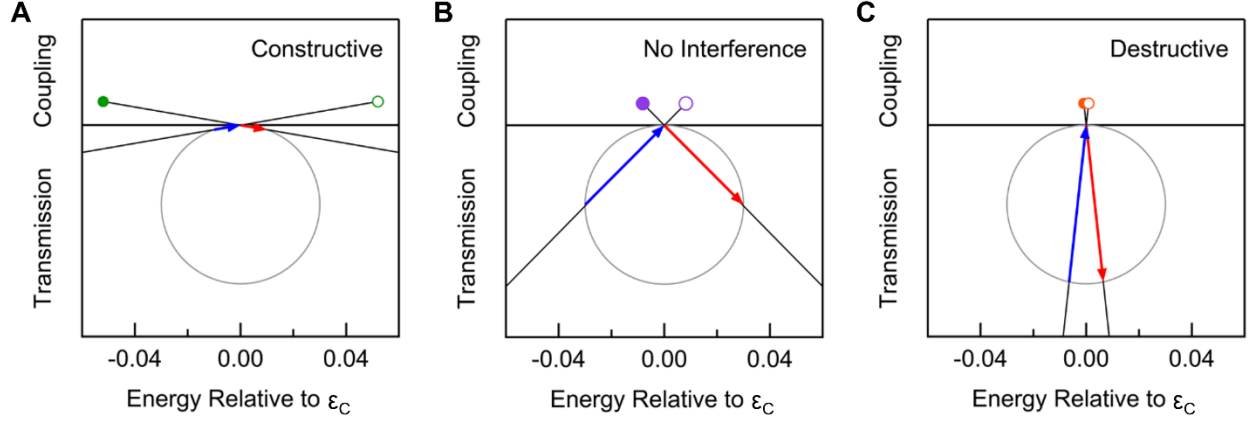


Figure S2. Energy coupling diagrams when two edge-state derived resonances approach to the center as chain length increases. ϵ_C is the on-site energy of carbon. The geometric construction clearly demonstrates how the transmission coefficients evolve from constructively interfering (A, when the angle between the two vectors is smaller than 90°) to destructively interfering (C, when the angle between the two vectors is larger than 90°). The maximum transmission should occur when the transmission coefficients are orthogonal.

In a symmetrically coupled junction,

$$\Gamma \equiv \Gamma_L = \Gamma_R \quad (37)$$

Based on this we see that peak transmission is obtained when the two eigenvalues for the edge states produce secant lines at ϵ_C that are perpendicular (Figure S2). This is given by the condition,

$$\gamma_0 = \frac{E_+ - E_-}{2} \quad (38)$$

where we have defined $\gamma_0 = \gamma_{\text{LUMO}} = \gamma_{\text{HOMO}}$, and E_\pm is defined by equation (15). This condition simply states that the vertical and horizontal components of each eigenvalue in the complex plane must be equal. Assuming the phase difference of the two edge states is $\phi = 0$, the HOMO-LUMO gap is

$$E_+ - E_- = 2 \left| a_1 e^{-\frac{M-1}{\xi}} t_1 b_M \right| \quad (39)$$

4. Transmission through Polyacetylene System

From here on, we use m to represent the number of unit cells in molecular systems. For polyacetylene system in molecular junction (Figure 2A), as shown in the main text, the Hamiltonian is

$$\tilde{\mathbf{H}}_m = -\frac{i\Gamma_L}{2} |1\rangle\langle 1| - \frac{i\Gamma_R}{2} |2m\rangle\langle 2m| - \sum_{k=1}^{2m-1} t_0 e^{(-1)^k \delta} |k\rangle\langle k+1| + H.c. \quad (40)$$

t_1 and t_2 are defined as $t_0 e^{-\delta}$ and $t_0 e^{\delta}$, respectively. Thus,

$$\delta \equiv \ln\left(\frac{t_0}{t_1}\right) = \ln\left(\frac{t_2}{t_0}\right) = \frac{1}{2} \ln\left(\frac{t_2}{t_1}\right) \quad (41)$$

and for $t_2 > t_1$ (diradical case),

$$\delta = \frac{\xi}{2} \quad (42)$$

We next derive $\Delta_{N,1}(\tilde{\mathbf{H}}_m)$ and $\det(\tilde{\mathbf{H}}_m)$ analytically.

$$\begin{cases} \Delta_{2m,1}(\mathbf{H}_{2m}) = -t_1^m t_2^{m-1} \\ \det(\mathbf{H}_{2M}) = (-1)^m \left(t_1^{2m} + t_2^{2(m-1)} \frac{\Gamma_L \Gamma_R}{4} \right) \end{cases} \quad (43)$$

For polyacetylene, the chain has both quinone and diradical features. In a symmetric junction, $\Gamma_L = \Gamma_R \equiv \Gamma$. The corresponding $T_m(E_F)$ with respect to m is derived as

$$T_m(E_F) = \left| \frac{4t_0^{2m-1} e^{-\delta} \Gamma}{4t_0^{2m} e^{-2m\delta} + t_0^{2m-2} e^{(2m-2)\delta} \Gamma^2} \right|^2 \quad (44)$$

Equation (44) can be rewritten as hyperbolic secant functions.

$$T_m(E_F) = \text{sech}^2 \left[(2m-1)\delta + \ln\left(\frac{\Gamma}{2t_0}\right) \right] \quad (45)$$

The two-level expression of $T_m(E_F)$ in the main text is directly derived from equation (44). If both the numerator and the denominator are divided by $4t_0^{2m-2} e^{(2m-2)\delta}$, we get:

$$T_m(E_F) = \left| \frac{t_0 e^{-(2m-1)\delta} \Gamma}{t_0^2 e^{-2(2m-1)\delta} + \frac{\Gamma^2}{4}} \right|^2 \quad (46)$$

Equation (46) can be rewritten as

$$T_m(E_F) = \left| \frac{\Gamma/2 \left((t_0 e^{-(2m-1)\delta} + i\Gamma/2) - (-t_0 e^{-(2m-1)\delta} + i\Gamma/2) \right)}{(t_0 e^{-(2m-1)\delta} + i\Gamma/2)(-t_0 e^{-(2m-1)\delta} + i\Gamma/2)} \right|^2 \quad (47)$$

Finally, we obtain:

$$T_m^{(2)}(E_F) = \left| \frac{(-1)^{m-1} \Gamma/2}{t_0 e^{-(2m-1)\delta} + i\Gamma/2} + \frac{(-1)^m \Gamma/2}{-t_0 e^{-(2m-1)\delta} + i\Gamma/2} \right|^2 \quad (48)$$

where $\frac{(-1)^{m-1} \Gamma/2}{t_0 e^{-(2m-1)\delta} + i\Gamma/2} = t_{\text{HOMO}}$, and $\frac{(-1)^m \Gamma/2}{-t_0 e^{-(2m-1)\delta} + i\Gamma/2} = t_{\text{LUMO}}$.

The maximum transmission occurs when the argument of the hyperbolic secant function (equation (45)) is zero.

$$(2m_{\text{max}} - 1)\delta + \ln\left(\frac{\Gamma}{2t_0}\right) = 0 \quad (49)$$

Therefore, the maximum transmission occurs at a chain length of m_{max} .

$$m_{\text{max}} = \frac{1}{2} \left(1 - \frac{1}{\delta} \ln\left(\frac{\Gamma}{2t_0}\right) \right) \quad (50)$$

The expression of β_m is obtained by taking first derivative of $\ln(T_m(E))$ with respect to m .

$$\beta_m \equiv \frac{\partial \ln(T_m(E))}{\partial m} = 4\delta \tanh \left[(2m - 1)\delta + \ln\left(\frac{\Gamma}{2t_0}\right) \right] \quad (51)$$

According to equation (51), at the long chain limit, $\lim_{m \rightarrow \infty} \beta_m = 4\delta$, and at short chain limit,

$$\lim_{m \rightarrow 0} \beta_m = -4\delta.$$

5. Transmission through X-Terminated Polyacetylene System

The X-terminate polyacetylene model is reasonably similar to the previous polyacetylene model, with a non-carbon atom at the two terminal sites (Figure 4A) with a non-zero on-site energy. The Hamiltonian for this molecule is:

$$\tilde{\mathbf{H}}'_m = \left(\varepsilon_X - \frac{i\Gamma_L}{2} \right) |1\rangle\langle 1| + \left(\varepsilon_X - \frac{i\Gamma_R}{2} \right) |2m\rangle\langle 2m| - \sum_{k=1}^{2m-1} t_0 e^{(-1)^k \delta} |k\rangle\langle k+1| + H.c. \quad (52)$$

The determinant values related to $\tilde{\mathbf{H}}'_m$ are obtained below by substituting $i\Gamma_{L/R}/2$ to $i\Gamma_{L/R}/2 - \varepsilon_X$.

$$\begin{cases} \Delta_{2m,1}(\tilde{\mathbf{H}}'_m) = -t_1^m t_2^{m-1} \\ \det(\tilde{\mathbf{H}}'_m) = (-1)^m \left(t_1^{2m} - t_2^{2(m-1)} \left(\varepsilon_X - \frac{i\Gamma_L}{2} \right) \left(\varepsilon_X - \frac{i\Gamma_R}{2} \right) \right) \end{cases} \quad (53)$$

We then obtain the corresponding transmission at E_F in symmetric junction:

$$T'_m(E_F) = \frac{\Gamma^2}{\left| \varepsilon_X - \frac{i\Gamma}{2} \right|^2} \left| \frac{\varepsilon_X - \frac{i\Gamma}{2}}{t_0 e^{-(2m-1)\delta} - t_0^{-1} e^{(2m-1)\delta} \left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2} \right|^2 \quad (54)$$

which can be rewritten as

$$T'_m(E_F) = \frac{\Gamma^2}{4\varepsilon_X^2 + \Gamma^2} \left| \operatorname{csch} \left[(2m-1)\delta + \ln \left(\frac{\varepsilon_X - \frac{i\Gamma}{2}}{t_0} \right) \right] \right|^2 \quad (55)$$

The hyperbolic functions are multiplied by a coefficient $\Gamma^2/(4\varepsilon_X^2 + \Gamma^2)$.

The two-level expression for $T'_m(E_F)$ in the main text is directly derived from equation (54) by simple manipulations to yield:

$$T'_m(E_F) = \left| \frac{\Gamma t_0 e^{-(2m-1)\delta}}{t_0^2 e^{-2(2m-1)\delta} - \left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2} \right|^2 \quad (56)$$

Equation (56) can be rewritten as:

$$T'_m(E_F) = \left| \frac{\Gamma/2 \left((-\varepsilon_X + t_0 e^{-(2m-1)\delta} + i\Gamma/2) - (-\varepsilon_X - t_0 e^{-(2m-1)\delta} + i\Gamma/2) \right)}{(-\varepsilon_X + t_0 e^{-(2m-1)\delta} + i\Gamma/2)(-\varepsilon_X - t_0 e^{-(2m-1)\delta} + i\Gamma/2)} \right|^2 \quad (57)$$

Finally, we obtain:

$$T'_m{}^{(2)}(E_F) = \left| \frac{(-1)^{m-1} \Gamma/2}{-\varepsilon_X + t_0 e^{(N-1)\delta} + i\Gamma/2} + \frac{(-1)^m \Gamma/2}{-\varepsilon_X - t_0 e^{(N-1)\delta} + i\Gamma/2} \right|^2 \quad (58)$$

where $\frac{(-1)^{m-1} \Gamma/2}{-\varepsilon_X + t_0 e^{(N-1)\delta} + i\Gamma/2} = t_{\text{HOMO}}$, and $\frac{(-1)^m \Gamma/2}{-\varepsilon_X - t_0 e^{(N-1)\delta} + i\Gamma/2} = t_{\text{LUMO}}$.

For X-terminated polyacetylene, it is non-trivial to obtain the analytical solution of β'_m as a function of δ . The solution of β'_N requires comparing real and imaginary parts of the argument of the hyperbolic cosecant function in $T'_N(E_F)$ (equation (55)), which are

$$\text{Re} = (N - 1)\delta - \ln \left(\sqrt{\varepsilon_X^2 + (\Gamma/2)^2} / t_0 \right) \quad (59)$$

and

$$\text{Im} = \tan^{-1}(\Gamma/2\varepsilon_X) \quad (60)$$

When $|\text{Re}| > \text{Im}$, β'_N can be approximated by

$$\beta'_N \approx \begin{cases} 4\delta \left| \coth \left[(N - 1)\delta + \ln \left(\frac{t_0}{\varepsilon_X - \frac{i\Gamma}{2}} \right) \right] \right| & (\text{Re} > \text{Im}) \\ -4\delta \left| \coth \left[(N - 1)\delta + \ln \left(\frac{t_0}{\varepsilon_X - \frac{i\Gamma}{2}} \right) \right] \right| & (-\text{Re} < \text{Im}) \end{cases} \quad (61)$$

When $|\text{Re}| \leq \text{Im}$, there is no trivial analytical solution.

6. Transmission through X-Terminated Polyphenylene System

The structures are shown in Figure 5A and 5B, where m represents the number of phenylene units in the chain.

1) Quinone

For the quinone structure, the Hamiltonian is

$$\begin{aligned} \tilde{\mathbf{H}}_{m,q} = & \left(\varepsilon_X - \frac{i\Gamma_L}{2} \right) |1\rangle\langle 1| + \left(\varepsilon_X - \frac{i\Gamma_R}{2} \right) |6m+2\rangle\langle 6m+2| \\ & - t_1 \sum_{j=0}^{m-1} \left(|6j+2\rangle\langle 6j+3| + |6j+2\rangle\langle 6j+4| \right) \\ & \left(|6j+5\rangle\langle 6j+7| + |6j+6\rangle\langle 6j+7| \right) \\ & - t_2 \left(\sum_{j=0}^m |6j+1\rangle\langle 6j+2| + \sum_{j=0}^{m-1} (|6j+3\rangle\langle 6j+5| + |6j+4\rangle\langle 6j+6|) \right) + H.c. \end{aligned} \quad (62)$$

where t_1 and t_2 are the hopping parameters through single bond and double bond, respectively.

With $\Gamma_L = \Gamma_R \equiv \Gamma$, the determinant values are

$$\begin{cases} \Delta_{6m+2,1}(\tilde{\mathbf{H}}_{m,q}) = (-1)^{m-1} 2^m t_1^{2m} t_2^{4m+1} \\ \det(\tilde{\mathbf{H}}_{m,q}) = (-1)^m \left((2t_1^2 t_2)^{2m} \left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2 - t_2^{6m+2} \right) \end{cases} \quad (63)$$

The transmission at E_F can be derived as

$$T_{m,q}(E_F) = \Gamma^2 \left| \frac{2^m t_1^{2m} t_2^{2m+1}}{(2t_1^2)^{2m} \left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2 - t_2^{4m+2}} \right|^2 \quad (64)$$

Finally, $T_{m,q}(E_F)$ has the form

$$T_{m,q}(E_F) = \frac{\Gamma^2}{4\varepsilon_X^2 + \Gamma^2} \left| \operatorname{csch} \left[m \ln 2 + 2m \ln \left(\frac{t_1}{t_2} \right) + \ln \left(\frac{\varepsilon_X - \frac{i\Gamma}{2}}{t_2} \right) \right] \right|^2 \quad (65)$$

2) Diradical

For the diradical structure, the Hamiltonian is

$$\begin{aligned} \tilde{\mathbf{H}}_{m,r} = & \left(\varepsilon_X - \frac{i\Gamma_L}{2} \right) |1\rangle\langle 1| + \left(\varepsilon_X - \frac{i\Gamma_R}{2} \right) |6m+2\rangle\langle 6m+2| \\ & -t \sum_{j=0}^{m-1} \left(|6j+2\rangle\langle 6j+3| + |6j+2\rangle\langle 6j+4| + |6j+3\rangle\langle 6j+5| \right) \\ & \quad \left(+|6j+4\rangle\langle 6j+6| + |6j+5\rangle\langle 6j+7| + |6j+6\rangle\langle 6j+7| \right) \\ & -t_1 \sum_{j=0}^m |6j+1\rangle\langle 6j+2| + H.c. \end{aligned} \quad (66)$$

where t and t_1 are the hopping parameters through intra-phenylene bond and inter-phenylene bond, respectively. Next, we obtain

$$\begin{cases} \Delta_{6m+2,1}(\tilde{\mathbf{H}}_{m,r}) = (-1)^{m-1} 2^m t^{5m} t_1^{m+1} \\ \det(\tilde{\mathbf{H}}_{m,r}) = (-1)^m \left((2t^3)^{2m} \left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2 - t^{4m} t_1^{2m+2} \right) \end{cases} \quad (67)$$

and the transmission at E_F is

$$T_{m,r}(E_F) = \Gamma^2 \left| \frac{(2t)^m t_1^{m+1}}{(2t)^{2m} \left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2 - t_1^{2m+2}} \right|^2 \quad (68)$$

As always, we rewrite $T_{m,r}(E_F)$ as a hyperbolic function.

$$T_{m,r}(E_F) = \frac{\Gamma^2}{4\varepsilon_X^2 + \Gamma^2} \left| \operatorname{csch} \left[m \ln 2 + m \ln \left(\frac{t}{t_1} \right) + \ln \left(\frac{\varepsilon_X - \frac{i\Gamma}{2}}{t_1} \right) \right] \right|^2 \quad (69)$$

The two-level expression of $T'_m(E_F)$ in the main text is directly derived from equation (68).

We divide the numerator and the denominator by $(2t)^{2m}$ to get:

$$T_{m,r}(E_F) = \Gamma^2 \left| \frac{\tau}{\left(\varepsilon_X - \frac{i\Gamma}{2} \right)^2 - \tau^2} \right|^2 \quad (70)$$

where $\tau = t_1^{m+1}/(2t)^m$. We can rewrite equation (70) as:

$$T_{m,r}(E_F) = \left| \frac{\Gamma/2 \left((-\varepsilon_X + \tau + i\Gamma/2) - (-\varepsilon_X + \tau + i\Gamma/2) \right)}{(-\varepsilon_X + \tau + i\Gamma/2)(-\varepsilon_X - \tau + i\Gamma/2)} \right|^2 \quad (71)$$

And finally, we obtain the two-level expression:

$$T_{m,r}^{(2)}(E_F) = \left| \frac{(-1)^{m-1} \Gamma/2}{-\varepsilon_X + \tau + i\Gamma/2} + \frac{(-1)^m \Gamma/2}{-\varepsilon_X - \tau + i\Gamma/2} \right|^2 \quad (72)$$

where $\frac{(-1)^{m-1}\Gamma/2}{-\varepsilon_X + \tau + i\Gamma/2} = t_{\text{HOMO}}$, and $\frac{(-1)^m\Gamma/2}{-\varepsilon_X - \tau + i\Gamma/2} = t_{\text{LUMO}}$.

7. Additional Figures

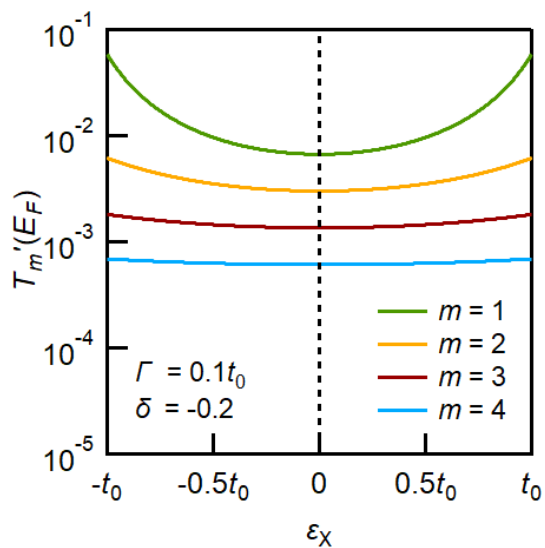


Figure S3. $T'_m(E_F)$ as an even function with respect to ϵ_X , with $m = 1-4$.

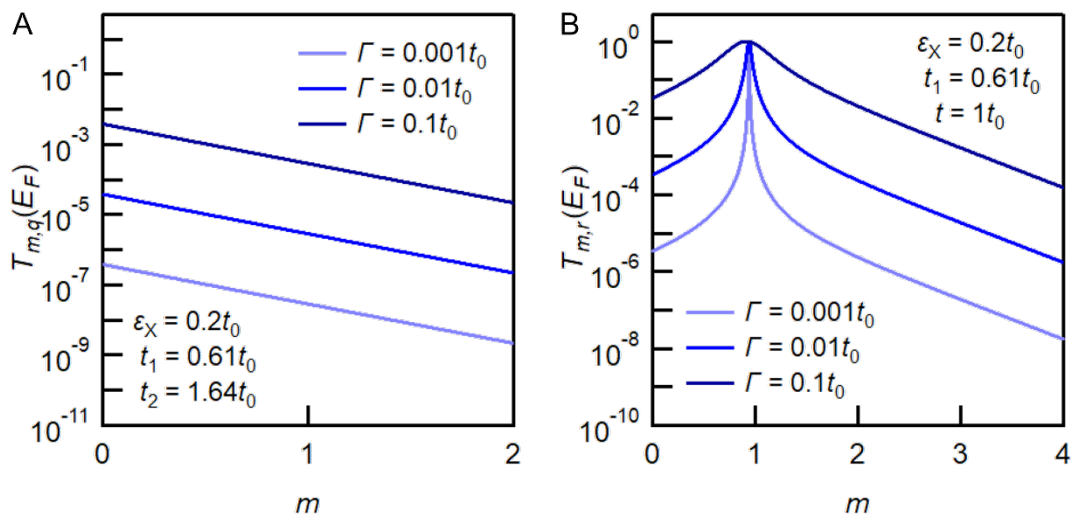


Figure S4. Transmission at E_F of X-terminated polyphenylene system at (A) quinone, and (B) radical structures, with $\Gamma = 0.1t_0, 0.01t_0,$ and $0.001t_0$.

8. Sample Mathematica Code

```
(* Taking Diradical Polyacetylene (m = 2) as example *)
(* Parameters *)
t0 = 1;  $\gamma$  = 0.1;  $\delta$  = 0.5;
t1 = t0 * Exp[-  $\delta$ ]; t2 = t0 * Exp[ $\delta$ ];

(* Hamiltonian and Gamma matrices *)
H = {{0, -t1, 0, 0}, {-t1, 0, -t2, 0}, {0, -t2, 0, -t1}, {0, 0, -t1, 0}};
 $\Gamma$ l = SparseArray[{1, 1} ->  $\gamma$ , {4, 4}];
 $\Gamma$ r = SparseArray[{4, 4} ->  $\gamma$ , {4, 4}];

(* Self - energy *)
 $\Sigma$  = - I / 2( $\Gamma$ l +  $\Gamma$ r);

(* Green's Function *)
G[E_] := Inverse[E IdentityMatrix[4] - (H +  $\Sigma$ )];

(* Transmission function *)
T[E_] := Tr[ $\Gamma$ l.G[E]. $\Gamma$ r.G[E][ConjugateTranspose]];

(* Plot transmission function *)
LogPlot[{T[E]}, {E, -1.5, 1.5}]
```

9. References

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