Three-body scattering problem and two-electron tunneling in molecular wires

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We solve the Lippmann-Schwinger equation describing elastic scattering of preformed pairs (e.g. bipolarons) off a short-range scattering center and find the two-particle transmission through a thin potential barrier. While the pair transmission is smaller than the single-electron transmission in the strong-coupling limit, it is remarkably larger in the weak coupling limit. We also calculate current-voltage characteristics of a molecule - barrier - molecule junction. They show unusual temperature and voltage behavior which are experimentally verifiable at low temperatures.

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Molecular-scale electronics is currently a very active area of research \textsuperscript{1}. It is envisaged that linear conjugated molecules would be used as the “transmission lines” in molecular circuitry \textsuperscript{2} in addition to active molecular elements discussed in the literature \textsuperscript{3,4}. When a so-called “molecular wire” is short, the dominant mechanism of transport is most likely a resonant tunneling through electronic molecular states (see \textsuperscript{3,4} and references therein). With increasing size of the wires one has to take into account strong interaction between carriers and vibronic excitations of the molecule, leading to self-trapping of electrons in polaronic states. The formation of polarons (and charged solitons) in polyacetylene (PA) was discussed theoretically in Refs. \textsuperscript{5,6} and formation of bipolarons (bound states of two polarons) in Ref. \textsuperscript{7}. Polarons in PA were detected optically in Ref. \textsuperscript{8} and since then studied in great detail. There is also an exceeding amount of evidence of the polaron and bipolaron formation in conjugated polymers such as polyphenylene, polypyrrole, polythiophene, polyphenylene sulfide \textsuperscript{10}, Cs-doped biphenyl \textsuperscript{11}, n-doped bithiophene \textsuperscript{12}, and other molecular systems. In many cases the doped polymers have bipolaron-like charge states to yield, in particular, the enhanced nonlinear optical properties \textsuperscript{13}.

Many experimental data provide evidence for hopping transport of (bi)polaronic carriers. However, at sufficiently low temperatures there should be a crossover to the band motion of polarons, as suggested long ago \textsuperscript{13,15}, and bipolarons \textsuperscript{13,16}. Indeed, due to recent extraordinary improvements in preparation of “plastic” molecular conductors, it became possible to measure their conductivity in a wide interval of temperatures and observe the crossover in two-dimensional films of organic conjugated molecules \textsuperscript{19}. In one-dimensional (1D) wires the band motion is expected to be strongly hindered by imperfections, and those imperfections are likely to be intentionally introduced in the system as functionalizing units \textsuperscript{21}. Moreover, the polarons in extended molecular wires/units are expected to be bound into real space bipolarons with lowering temperature. As it is known in the context of oxide semiconductors the bipolaron formation may strongly affect transport properties \textsuperscript{22,23}.

In this Letter we study elastic scattering of carriers bound into real-space pairs in one-dimensional organic and other conductors. We present an exact analytical solution in the limit of slow pairs. We also find an unusual temperature and voltage dependence of the tunnel conductance which may be experimentally verified at low temperatures.

In mathematical terms, the scattering of pairs is a three-body problem with the mass of the third particle taken to infinity. Let $\hat{U}(x_1 - x_2)$ be an attractive potential between the two moving particles and $\hat{V}(x_1, x_2)$ the repulsive external potential representing the barrier. The starting point is the Lippmann-Schwinger equation for the two-particle wave function $\hat{\Psi}(k_1, k_2)$ in momentum representation, which explicitly takes into account a boundary condition of the three-body scattering problem. It can be written as

$$\hat{\Psi} = -i\gamma \hat{G}(E + i\gamma)\Phi,$$

where $\hat{G}(E+i\gamma)$ is the exact two-particle Green’s function (GF) in the external potential, $\hat{\Phi}(k_1, k_2)$ is the wave function of a free ($\hat{V} = 0$) real-space pair in momentum representation, $\hat{\Phi}(k_1, k_2) = 2\pi\delta(q - Q)\phi(k)$. Here $q = k_1 + k_2$ is the center-of-mass momentum, $k = (k_1 - k_2)/2$ is the relative momentum, $E = -\epsilon + Q^2/4 < 0$ is the pair total energy in the absence of the external potential, and $\epsilon$ is its binding energy. The wave function $\phi(k)$ describes the internal structure of the pair. Hereafter we choose $h = k_B = m_1 = m_2 = 1, \gamma = +0$, and define $\hat{G}(E+i\gamma)\Phi$ as

$$\hat{G}\Phi = \iint\frac{dk_1'dk_2'}{(2\pi)^2} \hat{G}(k_1, k_2|k_1', k_2'; E)\Phi(k_1', k_2'),$$

for any $\hat{G}$ and $\Phi$. Using the two identities, $\hat{G} = \hat{G}_1 - \hat{G}_2 U \hat{G}$ and $\hat{G} = \hat{G}_12 - \hat{G}_12 V \hat{G}$ and the Lippmann-Schwinger equation one readily derives the equation for the Fourier component $T(k_1, k_2)$ of the product $\hat{U}\hat{\Psi}$. 

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\[ T(k_1, k_2) = (E - k^2 - q^2/4)\Phi - \hat{T}_{12}\Delta\hat{G}_3T, \quad (3) \]

where \( \Delta\hat{G}_3 = \hat{G}_3 - \hat{G}_0 \), \( \hat{G}_0 \) is the two-particle GF in the absence of any interaction \((\hat{U} = \hat{V} = 0)\), \( \hat{G}_3 \) is the GF of noninteracting particles in the external field \( \hat{V} \) (for \( \hat{U} = 0 \)), and \( \hat{G}_{12} \) is GF of two interacting particles with no external field, \( \hat{V} = 0 \). Here the scattering operator \( \hat{T}_{12}, \) defined by the relation \( \hat{T}_{12}\hat{G}_0 = \hat{U}\hat{G}_{12} \), is expressed via the particle-particle scattering t-matrix as

\[ T_{12}(k_1, k_2|k'_1, k'_2; E) = 2\pi t(k, k'; E - q^2/4)\delta(q - q'). \quad (4) \]

The t-matrix satisfies the equation

\[ t(k, k'; E) = u(k - k') - \int \frac{dp}{2\pi} u(k - p)\frac{tp(p, k'; E)}{p^2 - E - i\gamma}, \quad (5) \]

where \( u(p) \) is the Fourier component of the attractive potential \( \hat{U}(x_1 - x_2) \).

In many (in)organic semiconductors, the long-range Coulomb repulsion is usually significantly reduced by the strong Fröhlich interaction with optical phonons \([2,3]\), so that a net (attractive) potential between carriers is a short-range one, \( \hat{U}(x_1 - x_2) = -\alpha\delta(x_1 - x_2), \alpha > 0 \). Then Eq. (5) is readily solved resulting in the momenta-independent t-matrix

\[ t(k, k', E) = -\frac{\alpha\sqrt{-E}}{\sqrt{-E} - \alpha/2}, \quad (6) \]

which is valid for all energies provided that the square root is understood as its principal value. The binding energy is \( \epsilon = \alpha^2/4 \), and the normalized ground state wave function is \( \phi(k) = 2^{-1/2}\alpha^{3/2} / (k^2 + \epsilon) \). It is known that for a short-range inter-particle interaction \( T(k_1, k_2) \), Eq. (6), is proportional to the Fourier component of the center-of-mass wave function \( \Omega(q) \). \( T(k_1, k_2) = -2^{-1/2}\alpha^{3/2}\Omega(q) \).

Then the problem of elastic pair scattering is reduced to a single integral equation for the center-of-mass scattering amplitude \( \Upsilon(q) \). Substituting \( \Phi \) and \( \hat{T}_{12} \) in Eq. (3) one obtains

\[ \Omega(q) = 2\pi\delta(q - Q) - \frac{\Upsilon(q)}{q^2/4 - Q^2/4 - i\gamma}, \quad (7) \]

where \( \Upsilon(q) \) satisfies

\[ \Upsilon(q) = W(q, Q) - \int \frac{dq'}{2\pi} \frac{W(q, q')\Upsilon(q')}{q'^2/4 - Q^2/4 - i\gamma}. \quad (8) \]

The effective center-of-mass scattering potential \( W(q, q') \) is determined using GF of two noninteracting particles in the external potential \( (\hat{U} = 0 \text{ but } \hat{V} \neq 0) \) as

\[ W(q, q') = \alpha\chi(q) \int \frac{dk_2dk'_2}{(2\pi)^2} \Delta\hat{G}_3(q - k_2, k_2|q' - k'_2, k'_2; E), \quad (9) \]

where \( \chi(q) = E - q^2/4 + (\alpha/4)(q^2 - 4E)^{1/2} \). In the following we restrict our consideration to the scattering of slow pairs with \( Q^2 \ll 4\epsilon \). This condition allows us to replace \( W(q, q') \) with \( W(0, 0) \equiv W \) in all equations because the characteristic momenta \( q, q' \approx Q \) are much smaller than \( \sqrt{-E} \). Then the solution of Eq. (8) is given by \( \Upsilon(q) = WQ/(Q + 2iW) \) so that the pair transmission probability is

\[ T_2(Q) = 1 - \frac{2T(-Q)}{Q} = \frac{Q^2}{Q^2 + 4W^2}. \quad (10) \]

In general, \( G_3, W, \) and \( T_2 \) can be found only numerically. However, in many applications the scattering potential is also a short-range one, \( \hat{V}(x_1, x_2) = \beta[\delta(x_1) + \delta(x_2)] \), so that the full Hamiltonian takes the form

\[ H = -\frac{1}{2} \frac{\partial^2}{\partial x_1^2} - \frac{1}{2} \frac{\partial^2}{\partial x_2^2} - \alpha\delta(x_1 - x_2) + \beta[\delta(x_1) + \delta(x_2)]. \quad (11) \]

This three-body problem was considered before in \([25,26]\) but no general analytical solution was found. Here we present the analytical solution in the limit of slow pairs.

Consider the equation for the two-particle \( G_3 \)

\[ (k_1^2/2 + k_2^2/2 - E)G_3(k_1, k_2|k'_1, k'_2; E) + \beta \int \frac{dp_1}{2\pi} G_3(p_1, k_2|k'_1, k'_2; E) + \beta \int \frac{dp_2}{2\pi} G_3(k_1, p_2|k'_1, k'_2; E) = (2\pi)^2\delta(k_1 - k'_1)\delta(k_2 - k'_2), \quad (12) \]

which has a formal solution

\[ G_3(k_1, k_2|k'_1, k'_2; E) = G_0(k_1, k_2|k'_1, k'_2; E) \]

\[ \frac{D(k_2|k'_1, k'_2; E) + D(k_1|k'_2, k'_1; E)}{k_1^2/2 + k_2^2/2 - E}. \quad (13) \]

Here \( G_0(k_1, k_2|k'_1, k'_2; E) = (2\pi)^2\delta(k_1 - k'_1)\delta(k_2 - k'_2)(k_1^2/2 + k_2^2/2 - E)^{-1} \), and \( D(k_1|k'_2, k'_1; E) = (2\pi)^{-1}\beta \int dk_2 G_3(k_1, k_2|k'_1, k'_2; E) \) satisfies the integral equation

\[ D(k_1|k'_2, k'_1; E) \left[ 1 + \frac{\beta}{(k_1^2 - 2E)^{1/2}} \right] = \frac{2\pi\beta\delta(k_1 - k'_1)}{k_1^2/2 + k_2^2/2 - E} - \beta \int \frac{dk_2}{2\pi} D(k_2|k'_1, k'_2; E) \left[ \frac{D(k_2|k'_1, k'_2; E)}{k_2^2/2 + k_2^2/2 - E}. \quad (14) \]

We are interested in \( W = \alpha^3(2\pi)^{-2} \int \int dk_2dkD(k - p, p; E)(k^2 - E)^{-1} \). Integrating Eq. (14) with respect to \( k'_2 = -k'_1 \equiv -p \) one obtains for \( B(k; E) \equiv (2\pi)^{-1}\beta \int dpD(k - p, p; E)(k^2 - E)^{-1} \) the following equation:

\[ B(k; E) \left[ 1 + \frac{\beta}{(k^2 - 2E)^{1/2}} \right] = \frac{\beta}{2\pi k^2/2 + k^2/2 - E}. \quad (15) \]
It has the solution
\[ B(k; E) = \frac{\beta}{(k^2 - E) (1 + \beta/\sqrt{-E})}, \]
which is verified by direct substitution into Eq. (15). Finally we obtain
\[ W = \alpha^3 \int \frac{dk B(k; E)}{2\pi k^2 - E} = \frac{2\alpha\beta}{\alpha + 2\beta}. \]

This result together with Eq. (10) solves the problem of the elastic scattering of slow bound pairs for any strength of the short-range attractive and scattering potentials. It is instructive to compare the pair transmission \( T_2(Q), \) Eq. (17), with the single electron transmission \( T_1(p) = p^2/(p^2 + \beta^2) \) for equal kinetic energies \( p^2/2 = Q^2/4 \equiv K. \) If the binding potential is strong compared with the scattering potential \( (\alpha \gg 2\beta) \) the pair transmission is just the single-particle transmission of a particle with a double mass and double barrier strength, \( T_2(Q) = (Q^2/(Q^2 + 16\beta^2)), \) in accordance with a naive expectation. In the general case the ratio is
\[ \frac{T_2(Q)}{T_1(p)} = \frac{K + \beta^2}{K + 4\beta^2(1 + 2\beta/\alpha \alpha^2)^2}. \]

When the binding potential is weaker than the scattering potential \( (\alpha \ll \beta) \) the ratio is
\[ \frac{T_2(Q)}{T_1(p)} = \left( \frac{\beta}{\alpha} \right)^2 \gg 1. \]

Quite remarkably, a weak attraction between carriers helps the first transmitted particle to “pull” its partner through a strong potential barrier.

Another important difference between pair and single-electron tunnelling occurs due to their different statistics. While electrons are fermions, preformed pairs are bosons, so that their center-of-mass motion obeys the Bose-Einstein statistics. Hence, tunnelling conductance should be temperature dependent even at low temperatures \( T \) as has been already established in the bipolaron tunnelling to a normal metal with a decay of the \( \delta \)-function barrier. In the coordinate representation

\[ \Omega \]
\[ \text{FIG. 1. Zero-voltage conductance of MBM as a function of temperature (in units of } T_F \text{ for different relative strength of the barrier } 4\beta^2/T_F. \; G_0 = (2e^2)/h. \]

one has \( \Omega_l(X < 0) = e^{iQX} + Re^{-iQX}, \) and \( \Omega_r(X > 0) = Ce^{iP X} \) with \( 1 + R = C, \; CP_+ - (1 - R)Q = 8i\beta(1 + R), \) and \( P_+ = (Q^2 + 8eV)^{1/2}. \) The transmission is given by
\[ T_2(Q, P_+) \equiv 1 - |R|^2 = \frac{4QP_+}{(Q + P_+)^2 + 64\beta^2}, \]
for real \( P_+, \) and is zero otherwise. Multiplying the transmission by \( eQ \) and integrating with the Bose-Einstein distribution function \( f(Q) = \left[ \exp(Q^2/4T) - \mu/T - 1 \right]^{-1} \) yields the current as
\[ I(V) = e \int_0^{\infty} \frac{dQ}{2\pi} Qf(Q)[T_2(Q, P_+) - T_2(Q, P_-)], \]
where \( P_- = (Q^2 - 8eV)^{1/2} \) and \( \mu \) is the chemical potential determined by the number of \( \text{pairs } n \) using \( \int_0^{\infty} \frac{dQ}{2\pi} f(Q) = n. \) It is easy to calculate the integrals in the linear voltage classical limit, \( 2eV, T_F \ll T \) by expanding the transmission in powers of \( eV \) and replacing the Bose-Einstein distribution with the Boltzmann one, \( f(Q) \approx (2T_F/\pi T)^{1/2} \exp(-Q^2/4T) \) \( (T_F \equiv \pi^2 n^2/2 \) is the Fermi temperature of single carriers). The result for the conductance, \( \sigma \equiv (dI/dV)|_{V=0} \) is
\[ \sigma = \frac{2e^2}{\pi} \sqrt{\frac{2T_F}{\pi T}} \left[ 1 + \frac{4\beta^2}{T} e^{4\beta^2/T} \text{Ei}(-4\beta^2/T) \right], \]
where \( \text{Ei}(x) \) is the exponential integral function. The conductance behaves as \( \sigma = \frac{e^2}{\pi\beta} \sqrt{\frac{T_F}{2\pi T}} \) at \( T \ll 4\beta^2, \) and as \( \sigma = \frac{2e^2}{\pi} \sqrt{\frac{2T_F}{\pi T}} \) at \( T \gg 4\beta^2. \) In the last case it has a universal magnitude independent of the barrier strength. Apart from numerical coefficients, conductance of tightly
bound pairs is, of course, the same as conductance of single electrons in the classical limit. It is not the case, however, in a degenerate system, when $T \leq T_F$. Numerical integration of Eq. (21) at fixed density $n$ reveals a temperature dependence in this limit, Fig. 1 in comparison with the temperature independent conductance of fermionic noninteracting carriers at low temperatures. This remarkable difference is entirely due to the bosonic nature of pairs. The conductance is proportional to the mean velocity of carriers which in the case of bosons grows as $\sqrt{T}$ (while it is temperature-independent for fermions). This explains the low-temperature behavior of the conductance. Interpair correlations may reduce the difference in 1D wires. However, higher-dimension corrections readily restore it. There is also a breakdown of Ohm’s law when $2eV \geq T$, as shown in Fig. 2 for low temperatures, again in contrast with the Fermi statistics, where a non-linearity appears only at $eV \geq T_F \gg T$. We suggest that the most appropriate materials for experimental observation of the unusual current-voltage characteristics (Figs.1,2) are doped molecular semiconductors such as Cs-doped biphenyl [1], where bipolarons were explicitly detected by photoelectron and electron-energy-loss spectroscopies, and single crystals of pentacene, tetracene, rubrene, quaterthiophene ($\alpha$-4T), sexithiophene ($\alpha$-8T), where the coherent (bi)polaron tunnelling has been recently observed below room temperature [13].

In conclusion, we have solved the Lippmann-Schwinger equation in the effective mass approximation for single carriers and pairs in 1D conductors (molecular wires), which is valid for (bi)polarons if their size is larger than the lattice constant. While mapping of this problem onto discrete lattices is straightforward with a negative Hubbard $U$ model, the model itself can be applied to bipolarons only in the extreme nonadiabatic limit when the characteristic phonon frequency is larger than the binding energy $\hbar \omega_0$. In this limit a discrete Lippmann-Schwinger equation also has the analytical solution for slow pairs, which shows that the continuous model remains qualitatively correct even for lattice size (small) nonadiabatic bipolarons. In the opposite adiabatic regime bipolaron tunnelling is not a three-body problem because of the emission and absorption of (virtual) phonons [17]. We have found the scattering amplitude of elastic scattering of slow bipolarons, and conductance of the molecular junction (MBM) with preformed pairs. While the pair transmission is smaller than the single-electron transmission in the strong-coupling regime, it is surprisingly larger in the weak coupling regime. The current-voltage characteristics of MBM junction show unusual temperature and non-linear voltage behavior, Figs.1,2.

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