# The effect of dephasing on thermoelectric properties of polythiophene molecular junction

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**Abstract**

*The thermoelectric properties of disordered polythiophene molecular junction are investigated using Büttiker method within Green function formalism in linear response regime. An extended Su-Schrieffer-Heeger model is used to describe the Hamiltonian of the molecule. The decrease of height of transmission probability peaks and thermoelectric coefficients are observed in presence of Büttiker probes.*

Study of charge and spin-dependent thermoelectric properties of the molecular junction has become a hot topic in recent years for conversion of heat to electricity [1,2]. The thermal efficiency depends on a dimensionless quantity as a figure of merit, $ZT = \frac{G_e S^2 T}{\kappa}$ which is a function of thermopower (S), operating temperature (T), electrical and thermal conductances, $G_e$ and κ ,respectively. In present work, we study the effect of dephasing on thermoelectric properties of of polythiophene (PT) molecule coupled to three dimensional ferromagnetic (FM) electrodes using Green function formalism in linear response regime that has not been reported so far.

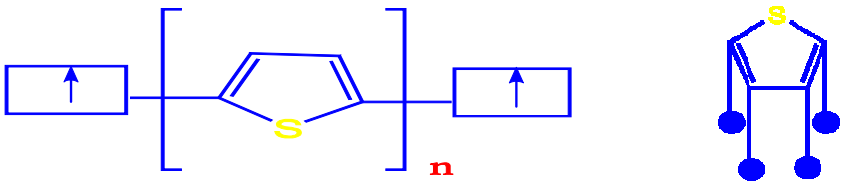


Fig.1. The FM/polythiophene/FM junction. All carbon atoms are connected to the factious probes.

We consider a PT chain connected to semi-infinite FM electrodes (Fig. 1) The Hamiltonian of isolated molecule can be described by an extended SSH model [3]:

$$H_{mol} = \sum_{j,\sigma}\{(\varepsilon\, d^{+}_{j,\sigma} d_{j,\sigma} - t_{j,j+1}(d^{+}_{j+1,\sigma} d_{j,\sigma} + h.c)\}) - \sum_{j,\sigma}\{\varepsilon_s (d^{+}_{4j-3,\sigma} d_{4j-3,\sigma} + d^{+}_{4j,\sigma} d_{4j,\sigma}) +$$

$$t_s (d^{+}_{4j-3,\sigma} d_{4j,\sigma} + h.c)\} + \frac{K_0}{2}\sum_j (u_{j+1} - u_j)^2, \quad (1) \qquad t_{j,j+1} = t_0 - t_1 \cos(j\,\frac{\pi}{2}) - \alpha(u_{j+1} - u_j) \quad (2)$$

Here, $d^{+}_{j,\sigma}(d_{j,\sigma})$ creates (annihilates) a π-electron with spin σ at the lattice site j. The index j runs over the π-orbitals of the carbon atom along the PT molecule. $t_{j,j+1}$ denotes the hopping integrals between two near neighbor electrons, $\varepsilon = \varepsilon_j + eV_G$ that $\varepsilon_j$ is the on-site energy and $V_G$ denotes gate voltage used to control the molecular energy levels. $\varepsilon_s$ and $t_s$ show the effects of sulfur atom on Hamiltonian of PT molecular chain. In Eq.2, $t_0, t_1, \alpha, K_0$ and $u_j$ are zero displacement hopping integral, non-degeneracy parameter, electron-lattice coupling constant, the elastic constant and the lattice displacement at site j, respectively. The Schrödinger equation can be solved for the electronic part of Hamiltonian Eq. 1 to find the electronic eigenstate $|\psi_{\mu,\sigma}\rangle = \sum_i \varphi_{i,\mu,\sigma} d^{+}_{i,\sigma}|0\rangle$ and eigenvalues $\varepsilon_{\mu,\sigma}$ [4].

The Hamiltonian of the left (right) FM electrode is written as:

$$H_{\alpha}=\sum_{i_{\alpha},\sigma}(\varepsilon_0-\sigma.J_{\alpha})c^{+}_{i_{\alpha},\sigma}c_{i_{\alpha},\sigma}-t(c^{+}_{i_{\alpha},\sigma}c_{i_{\alpha}+1,\sigma}+c^{+}_{i_{\alpha}+1,\sigma}c_{i_{\alpha},\sigma}),\quad (3)$$

where, $c^{+}_{j,\sigma}(c_{j,\sigma})$ creates (annihilates) an electron with spin σ at site i in electrode $\alpha$ (=L or R). $\varepsilon_0$ and $t$ are the on-site energy and the hopping matrix element in the electrode, respectively. $J_{\alpha}$ is the integral exchange energy and σ denotes the Pauli spin operator. The coupling between the molecule and electrodes is given by:

$$H_T=\sum_{j}\sum_{i}t_{c(j,\sigma,i)}(c^{+}_{i,\sigma}d_{j,\sigma}+hc),\quad (4)$$

where, $t_{c(j,\sigma,i)}=t_c$ stands for the coupling strength between PT molecule and FM electrodes. The spin-flip effect is neglected in our work. We introduce the dephasing effect in our system using D'Amato calculations [4]. The retarded Green function of the coupling is:

$$G^{r}_{\sigma}(\varepsilon,V_G)=[(\varepsilon+i0^{+})I-H_{mol}-\Sigma_{L,\sigma}(\varepsilon)-\Sigma_{R,\sigma}(\varepsilon)-\Sigma_{D}]^{-1},\quad (5)$$

where, $0^{+}$ is the infinitesimal value. $\Sigma_{\alpha,\sigma}(\varepsilon)$ is the self-energy function that describes the effect of semi-infinite FM electrodes on the molecular chain and is:

$\sum_{\alpha,\sigma}=\tau^{+}_{c,L}g_{\alpha,\sigma}\tau_{c,L}$, $g_{\alpha,\sigma}$ is surface Green function of FM electrodes that has been obtained in [5]. $\Sigma_D=\sum_{l=1}^{N}-i\gamma_l c_l^{\dagger}c_l$ is the self-energy of the factious probes. In order to compute the thermoelectric properties of the junction, we calculate the charge and heat current as follows,

$$I=\frac{e}{h}\sum_{\sigma}\int d\varepsilon[f_L(\varepsilon)-f_R(\varepsilon)]T_{eff\,\sigma}(\varepsilon),\quad (7a)\qquad Q=\frac{1}{h}\sum_{\sigma}\int d\varepsilon(\varepsilon-\mu)[f_L(\varepsilon)-f_R(\varepsilon)]T_{eff\,\sigma}(\varepsilon),\quad (7b)$$

$T_{eff\,\sigma}=Tr[\Gamma_{L,\sigma}(\varepsilon)G^{r}_{\sigma}(\varepsilon)\Gamma_{R,\sigma}(\varepsilon)G^{a}_{\sigma}(\varepsilon)]+\sum_{i,j=1}^{N}T_{R,i}W^{-1}_{ij}T_{j,L}$ is the effective transmission coefficient, where $W$ is the symmetric matrix[6]. In linear response regime the charge and heat currents are given as: $I=e^2\sum_{\sigma}L_{0\sigma}\Delta V+\frac{e}{T}\sum_{\sigma}L_{1\sigma}\Delta T,\quad (8a)\qquad Q=e\sum_{\sigma}L_{1\sigma}\Delta V+\frac{1}{T}\sum_{\sigma}L_{2\sigma}\Delta T,\quad (8b)$

$$L_{n\sigma}=-\frac{1}{h}\int d\varepsilon(\varepsilon-\mu)^n\frac{\partial f}{\partial\varepsilon}T_{eff\,\sigma}(\varepsilon).\quad (9)$$

The thermoelectric coefficients are as follows:

$$S=-\frac{1}{2eT}\left(\frac{L_{1\uparrow}}{L_{0\uparrow}}+\frac{L_{1\downarrow}}{L_{0\downarrow}}\right),\ (10a)\qquad G_e=e^2\sum_{\sigma}L_{0\sigma}\quad (10b)\quad and\quad \kappa=\frac{1}{T}\sum_{\sigma}\left[L_{2\sigma}-\frac{L_{1\sigma}^2}{L_{0\sigma}}\right],\ (10c)$$

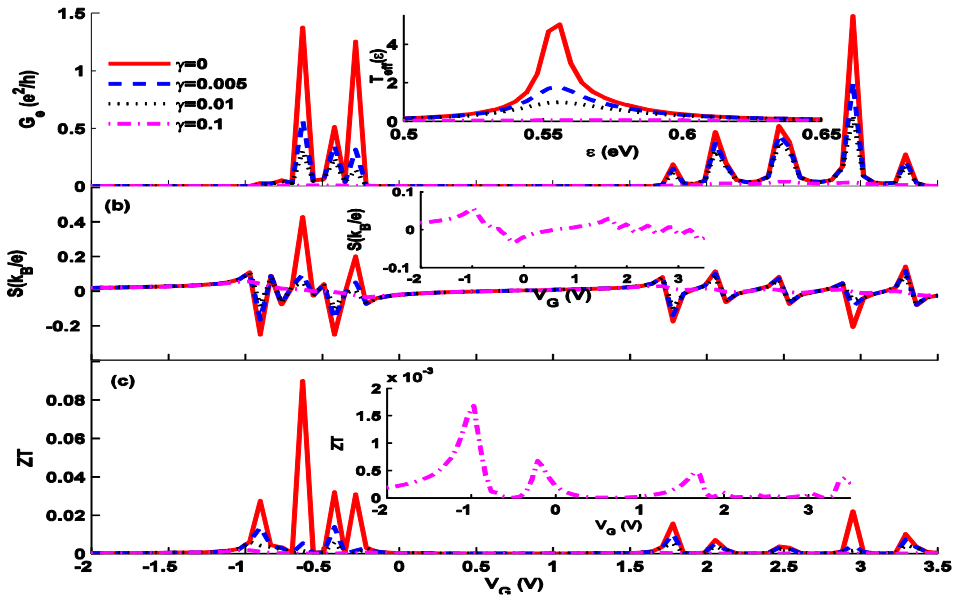


Fig.2. The effect of dephasing on thermoelectric properties of the FM/PT/FM junctions.

Fig.2 shows the dependence of the charge thermoelectric properties on gate voltage for typical PT molecule with five rings in presence and absence of fictitious probes in parallel configuration. It is obvious from Fig.2a with increasing of γ the electrical conductance decreases. It can be found from the effective transmission coefficient of the junction. One can see from the inset of Fig.2a that the height of the effective transmission coefficient decreases in presence of dephasing due to increasing of rate of the scattering in the fictitious probes. It is also observed from the inset of Fig .2a that the peak of transmission probability becomes wider in the presence of additional probes (γ) in comparison with γ=0. This broadening is risen from dominance of randomizing effect over backscattering due to loss of phase coherence in dephasing process [7]. We indicate only one peak of effective transmission coefficient for more clarity. The peak becomes wider and its height decreases by increase of γ due to more scattering rate. The charge thermopower and figure of merit are indicated in Figs.2b and 2c. The thermopower of PT has oscillating behavior denoting the change in the number of electrons in the molecular energy levels [8] and decreases in the presence of dephasing. The value of charge thermopower decreases more with γ because of the changes in effective transmission coefficient peaks due to more scattering in atomic sites of molecular chain. In strong coupling regime the number of oscillation of the peaks decreases because the effective transmission coefficient peaks almost disappear in greater value of γ. The main factor for the shape of ZT is charge thermopower. The ZT is zero in points that S=0 and the extremum points of S are the extremum of ZT. Therefore, the value of the ZT decreases with increasing of γ.

## Conclusion

In this work, we have studied the thermoelectric properties of polythiophene (PT) molecular junction with ferromagnetic electrodes in presence of additional probes. Our method is based on a tight-binding model within Green function formalism that can help to find of the thermoelectric coefficients of disordered PT chain in the linear response regime. Results show the decrease of thermoelectric coefficients due to decrease of height and increase of wide of effective transmission coefficient peaks in presence of additional probes.

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