



Effect of Electron-Phonon Interaction on the Quantum Transport through La@C₈₂ Endohedral Complex

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Abstract

We have investigated the effect of electron-phonon interaction on the spin-dependent transport through a La@C₈₂ endohedral fullerene based on the Green's function method and Landauer-Büttiker formalism. Effect of electron-phonon (e-ph) interaction is studied using Büttiker formalism. We have studied the influence of e-ph interaction on the spin dependent currents, and tunnel magnetoresistance (TMR) of the junction. Our results indicate that in the presence of e-ph interaction, the spin dependent currents increase but the TMR reduces. Furthermore, we have investigated the effect of the strength of coupling between electrode and molecule on the current-voltage characteristics of the junction. It is shown that the transport properties of molecule are obviously affected by the change of electrode/molecule coupling strength.

Keywords: Spin-dependent transport, green's function, dephasing, endohedral fullerene.

Introduction

Great advances have been gained in spinelectronic technology in the recent years. In the typical magnetic tunnel junction, an organic molecule is sandwiched between two magnetic electrodes. Spin dependent transport through organic molecules has been investigated both experimentally and theoretically in the recent years¹⁻⁵. Among many types of molecules, endohedral metallofullerenes are interesting candidate as a bridge in molecular junctions because of their unique structure in the field of nano-electronic devices^{6,7}. Carbon atoms in these molecules form a cage enclosing an atom or ion. Since the discovery of the first stable endohedral metallofullerene, La@C₈₂, by Chai et al in 1991, many theories and experiments on the structural and electronic⁸⁻¹⁰ properties of this molecule have been presented. In the endohedral fullerene species such as La@C₈₂, the encapsulated La atom loses its valence electrons to the C₈₂ cage due to the higher electron affinity of the cage^{11,12}. Due to existence of the strong La-fullerene bonds and a large dipole moment, centre of the cage is the unstable position and the La ion moves towards the side of the cage⁸. La@C₈₂ fullerene have nine isomers (C_{2v}, C_{3v}(1), C_{3v}(2), C₂(1), C₂(2), C₂(3), C_s(1), C_s(2), C_s(3)) that satisfy the isolated pentagon rule¹³.

We used isomer 3 with C₂ symmetry of C₈₂, whereas, a tight binding molecular dynamics (TBMD) study of C₈₂ showed isomer 3 as the most stable isomer having the lowest energy and largest HOMO-LUMO gap. It accords with the results of local density functional (LDA) method¹⁴. It is shown that spin-polarized transport through molecules can be influenced by changing the spin alignment of electrodes. For instance, a theoretical study of quantum transport through a Ni-bezenedithiol (BDT)-Ni molecular junction based on density

functional theory, showed that the current in the parallel alignment is higher than antiparallel alignment¹⁵. In coherent regime, in which the inelastic process is neglected quantum transport through molecules can be explained in the framework of Landauer formalism¹⁶. A main source of phase-breaking in the transport through the molecular junctions is electron-phonon (e-ph) interaction. A theoretical method for investigating the e-ph interaction is based on density functional theory (DFT) and non-equilibrium Green's function (NEGF) method^{17,18}. But this computational method even for small structures is very time consuming. Another method to consider dephasing in mesoscopic systems was offered by Büttiker^{19,20}. The main idea of this method is to introduce some virtual electronic reservoirs into the coherent system^{19,21,22}. When electrons tunnel through the molecule, they get scattered into virtual reservoirs, and they re-inject into device with a random phase. To carry out this idea each scatterer can be considered as an electron reservoir coupled to the device.

In this study, we consider a La@C₈₂ molecule which is sandwiched between two ferromagnetic (FM) electrodes, to investigate the possibility of a single-molecule spintronics device. Using Green's function method based on the Landauer-Büttiker formalism, we investigate the effect of dephasing on the current-voltage characteristics and TMR of the junction. The rest of the paper is arranged as follows. In section 2, we present our calculation method for investigating the e-ph interaction on the spin dependent transport through the molecule. In section 3, the results and discussions are presented. Followed by conclusions in section 4.

Methodology

Model and Formalism: The Hamiltonian of the system in the absence of dephasing reservoirs is expressed as:

$$\hat{H} = \hat{H}_L + \hat{H}_R + \hat{H}_C + \hat{H}_E \quad (1)$$

where \hat{H}_L and \hat{H}_R describe the Hamiltonians of the left (L) and right (R) ferromagnetic electrodes, \hat{H}_E describes the Hamiltonian of central endohedral molecule and \hat{H}_C represents the coupling between the electrodes and the molecule. Hamiltonian of the electrodes can be written as:

$$\hat{H}_\alpha = \sum_{i_\alpha, s} (\epsilon_{i_\alpha} - \sigma \cdot E_\alpha) a_{i_\alpha, s}^+ a_{i_\alpha, s} - \sum_{\langle i_\alpha, j_\alpha \rangle, s} t_{i_\alpha, j_\alpha} a_{i_\alpha, s}^+ a_{j_\alpha, s} \quad (2)$$

where $a_{i_\alpha, s}^+$ ($a_{i_\alpha, s}$) denotes the creation (annihilation) operator of an electron with spin s at site i in the left and right electrodes. ϵ_{i_α} and t_{i_α, j_α} are the on-site energy and the nearest neighbor hopping integral. $-\sigma \cdot E_\alpha$ is the internal exchange energy with E_α denoting the molecular field at site i_α , and σ is the Pauli spin operator. The coupling Hamiltonian between the electrodes and carbon atoms of the central part is described as:

$$\hat{H}_C = \sum_{\alpha \in \{L, R\}} \sum_{\langle i_\alpha, j \rangle, s} -t_{i_\alpha, j} (a_{i_\alpha, s}^+ b_{j, s} + h.c.) \quad (3)$$

where the matrix elements $t_{i_\alpha, j}$ is the coupling strength between the molecule and electrodes.

We extended SSH model for the Hamiltonian of the central part as:

$$\hat{H}_M = \sum_{i, s} \epsilon_i b_{is}^+ b_{is} + \sum_{\langle i, j \rangle, s} (-t_{ij} + \alpha y_{ij}) (b_{is}^+ b_{js} + h.c.) + \frac{1}{2} \sum_{\langle i, j \rangle, s} K (y_{ij})^2 + V_{\circ i} \quad (4)$$

Here b_{is}^+ (b_{is}) are the usual creation (annihilation) operators of the π electron at the i -th carbon atom; t_{ij} is the hopping integral, α is the coupling constant between electron and phonon, K is spring constant and y_{ij} is the change of the bond length between i -th and j -th atoms and the sum is taken over nearest neighbor $\langle i, j \rangle$ pair sites. ϵ_i is the orbital energy of π electrons,

$$\epsilon_i = -\frac{e}{r_{\circ i}} \quad (5)$$

where $r_{\circ i}$ is the distance between the La ion and carbon atom at the site i . The forth term in the Hamiltonian is the interaction between the La ion and the carbon atoms on the cage and can be taken as:

$$V_{\circ i} = A \frac{e^{-\beta r_{\circ i}}}{r_{\circ i}^2} \quad (6)$$

Here, A is a strength factor and $1/\beta$ is screening length. Parameters A and $1/\beta$ can be specified by other results of first-principle calculations²³.

For calculating the Hamiltonian we must solve the Schrödinger equation for π electrons to find eigenvalue and eigenstates of the system.

$$\epsilon_{\kappa} \phi_{\kappa, \sigma}(i) = \sum_{ij} (-t_{ij} - \alpha y_{ij}) \phi_{\kappa, \sigma}(j) \quad (7)$$

We can determine the change of bond length by minimizing the total energy of the central molecule.

$$y_{ij} = \frac{2\alpha}{K} \sum_{\kappa, \sigma} \phi_{\kappa, \sigma}(i) \phi_{\kappa, \sigma}(j) - \frac{2\alpha}{K} \frac{1}{N_b} \sum_{\langle kl \rangle} \sum_{\kappa, \sigma} \phi_{\kappa, \sigma}(k) \phi_{\kappa, \sigma}(l) \quad (8)$$

where the prime means the sum over the occupied states, and N_b is the number of π -bonds of C_{82} cage.

Equations (7) and (8) are solved iteratively in the following way:

First equation (7) is solved for the initial set of the bond variables. Employing the electronic eigenstates obtained above, we calculate the next set from the equation (8). The iteration is repeated until the difference between values of y_{ij} (in unit of A^0) from two consecutive iterations is less than 10^{-5} . We have added the dephasing effects in the Hamiltonian of the system simply through a scattering selfenergy term as follows:

$$\Sigma_D = \sum_{l=1}^N -i\gamma_l d_l^+ d_l \quad (9)$$

The e-ph interaction strength is characterized by γ_l parameter.

The Green's function of the system in the presence of e-ph interaction can be expressed as:

$$\hat{G}(\epsilon) = [\epsilon \hat{I} - H_C - \Sigma_L(\epsilon - eV_a/2) - \Sigma_R(\epsilon + eV_a/2) - \Sigma_D]^{-1} \quad (10)$$

Here $\Sigma_{\alpha, s} = \tau_{m, \alpha} g_{\alpha, s} \tau_{\alpha, m}$ is the self-energy matrix which is the outcome of coupling of molecule to the electrodes. τ_{α} is the hopping matrix and $g_{\alpha, s}$ is the surface Green's function of the semi-infinite isolated electrodes, and can be given as²⁶:

$$g_{\alpha, s}(\epsilon) = \sum_k \frac{\psi_k(r_n) \psi_k^*(r_m)}{z - \epsilon + \sigma \cdot E_\alpha - \epsilon(k)} \quad (11)$$

Where $r_n \equiv (x_n, y_n, z_n)$, $k \equiv (l_x, l_y, k_z)$, $z = \epsilon + i0^+$.

$$\epsilon(k) = \epsilon_{\circ} - 2t [\cos(\frac{l_x \pi}{(N_x + 1)}) + \cos(\frac{l_y \pi}{(N_y + 1)}) + \cos(k_z a)]$$

$$\psi_k(r_n) = \frac{2\sqrt{2}}{\sqrt{(N_x+1)(N_y+1)N_z}} \sin\left(\frac{l_x x_n \pi}{(N_x+1)}\right) \sin\left(\frac{l_y y_n \pi}{(N_y+1)}\right) \sin(k_z z) \quad (12)$$

Here, N_x and N_y are the number of atoms at the surface of electrodes. For simplicity we suppose the electrodes have simple cubic structure and only the central atom at the cross section of electrodes is connected to the end of the molecule. The total effective transmission probability of an electron from the left electrode to the right in the presence of e-ph interaction can be expressed as²⁷:

$$T_{eff}(\mathcal{E}) = T_{L,R} + \sum_{i,j=1}^N T_{R,i} W_{i,j}^{-1} T_{j,L} \quad (13)$$

The first term of the above expression refers to the electrons that propagate coherently through the system, whereas the second term refers to the electrons scattering due to e-ph interaction and propagate incoherently.

W matrix is defined as follows:

$$W_{i,j} = [(1 - R_{i,j})\delta_{i,j} - T_{i,j}(1 - \delta_{i,j})] \quad (14)$$

Where

$$R_{i,i} = 1 - \sum_{i \neq j} T_{i,j} \quad (15)$$

$R_{i,i}$ is the reflection probability of an electron from i -th reservoir and $T_{k,m}$ is the transmission probability between any pair of reservoirs and can be given as:

$$T_{k,m}(\mathcal{E}) = Tr(\Gamma_k G_\sigma \Gamma_m G_\sigma^+) \quad (16)$$

Γ_k is the broadening matrix and can be defined as:

$$\Gamma_k = -2 \text{Im}(\Sigma_k) \quad (17)$$

We assumed that the electron's spin alignment doesn't change in the transport through the molecule. Therefore we can divide spin-dependent current into two parts; one for spin-up and the other for spin-down. In the parallel (P) alignment, the spin-up (spin-down) electrons are scattering from the left electrode only into empty spin-up (spin-down) states in the right electrode therefore the total transmission probability for the P alignment can be express as:

$$T_{eff}(\mathcal{E}) = T_{eff \uparrow \uparrow}(\mathcal{E}) + T_{eff \downarrow \downarrow}(\mathcal{E}) \quad (18)$$

In the anti-parallel (AP) alignment, the spin-up (spin-down) electrons are scattering from the left electrode into empty spin-down (spin-up) states in the right electrode and the total transmission probability for the AP alignment can be written as:

$$T_{eff}(\mathcal{E}) = T_{eff \uparrow \downarrow}(\mathcal{E}) + T_{eff \downarrow \uparrow}(\mathcal{E}) \quad (19)$$

The current-voltage (I - V) characteristic is calculated by the Landauer-Bütikker formula¹⁶:

$$I(V_a) = \frac{e}{h} \int_{-\infty}^{+\infty} T_{eff}(\mathcal{E}) [f(\mathcal{E} - \mu_R) - f(\mathcal{E} - \mu_L)] d\mathcal{E} \quad (20)$$

Here f is the Fermi distribution function with electrochemical potential $\mu_{L,R}$ of the left and right electrodes. The tunnel magnetoresistance (TMR) can be defined as²⁸:

$$TMR \equiv \frac{I_p - I_{AP}}{I_p} \quad (21)$$

Here I_p and I_{AP} are the total currents in parallel and anti-parallel alignments respectively.

Results and Discussion

Here we use the methods described above to study a La@C₈₂ molecule sandwiched between two electrodes. For numerical calculations, we consider the electrons spin alignments in the left electrode in the +y direction, while in the right electrode it can be flipped into the +y or -y direction. In our numerical calculations, we take $t_0 = 2.50\text{eV}$, $\alpha = 5.65\text{eV/\AA}^\circ$, $K = 33.1\text{eV/\AA}^2$ ²⁹ and $t = 1\text{eV}$, $|E_{el}| = 1.5\text{eV}$. The band gap is calculated for C₈₂ molecule with theses parameters is in accordance with experimental results³⁰. The electrode/molecule coupling strength and the dephasing strength are set to $t_c = 0.6t_0$ and $\gamma = 0.005\text{eV}$ respectively.

From equation (20) it is seen that the current consists of two factors which are the transmission function and the difference between the Fermi functions of the left and right electrodes. Therefore only electrons with energies within the energy region, $[\mu_L, \mu_R]$, chip in the total current, which is called the bias window³¹. In figure 1, the current-voltage (I - V) characteristics of the molecule for P alignment are indicated with and without dephasing. It is seen that when we consider the effect of e-ph interaction, the current reaches a higher value compared to coherent regime. We are plotted the transmission probability as a function of energy in figure 4. In the absence of interactions, the electron wave function propagates coherently across the whole system. Accordingly, when the electron energy is equal to the molecular levels, the electron resonantly broadcast through the molecule and a sharp peak appears in the transmission function. It is seen that in the presence of e-ph interaction, the widths of the peaks become broadened compared to the coherent transmission. In the presence of e-ph interaction the phase coherence of electrons is broken therefore destructive interference of electrons is reduced and it can enhance transmission function. Therefore in the presence of dephasing, the current is increased due to increase of the area under the transmission curve according to equation 20.

In figure 2, the current-voltage (I-V) characteristics of the molecule for AP alignment, are indicated with and without dephasing. By comparing figure 1 and figure 2, it can be seen that the current for P alignment reaches a higher value in comparison with the current for AP alignment. This is due to the difference between the surface density of states of FM electrodes for spin-up and spin-down electrons. In the parallel alignment, the electrons with spin-up in the left electrode move into the empty spin-up states in the right electrode and there is no asymmetry in the SDOS. For AP alignment the electrons with spin-up in the left electrode move into empty states with spin-down in the right electrode and the electrons with spin-down move into empty states with spin-up. In fact, in the AP alignment, the spin-up and spin-down currents are asymmetric. Consequently, the total current through the molecule for parallel alignment, is larger than for antiparallel arrangement. Also in figure 3 we showed the TMR which is calculated by the results of the P and AP currents. It is seen that TMR have their maximum value at the lower bias voltages. With increasing the

applied voltage, it is seen that the TMR decreases. it is similar to the conventional magnetic tunnel junctions³². When the bias voltage increases further, the TMR ratio increases and then, reduces again. Also, when we consider the dephasing effect, it is seen that TMR decreases with respect to coherent regime. Our result is in accordance with the experiment results^{33,34}.

In figure 5 and figure 6 we have represented variations of the currents for the P and AP alignments and TMR as a function of dephasing strength (γ) in the bias voltage $V=2$ V respectively. Our calculations indicate that when γ increases, the currents increase too. Figure 6 indicates that the TMR decreases as we consider the dephasing effects which resulting of increasing the currents in this situation. When the molecule is coupled to the electrodes depends on the strength of the coupling its discrete states broaden. Accordingly we have investigated the effect of the molecule/electrode coupling strength t_c on the spin currents and TMR, as illustrated in figures 7-9, respectively.

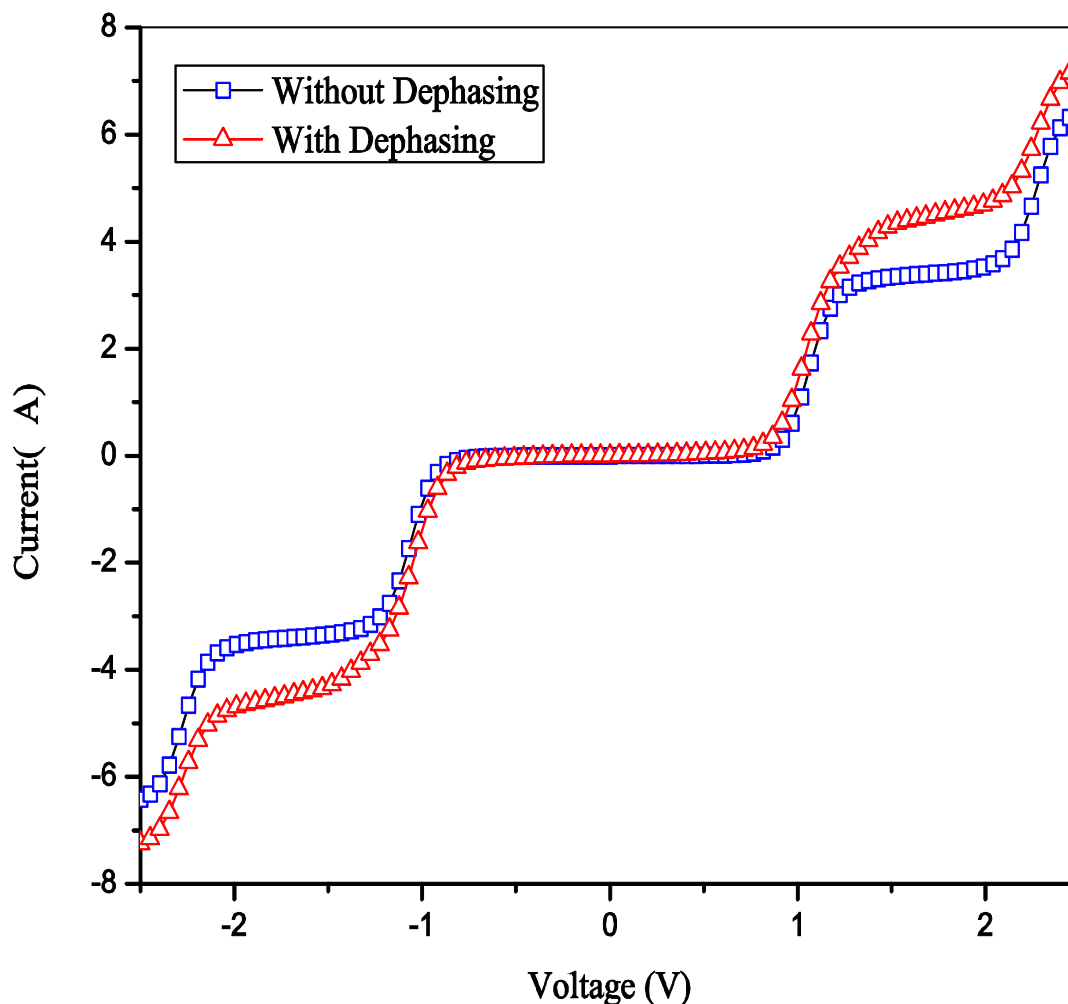


Figure-1
The current-voltage characteristics for the P spin alignment with and without dephasing

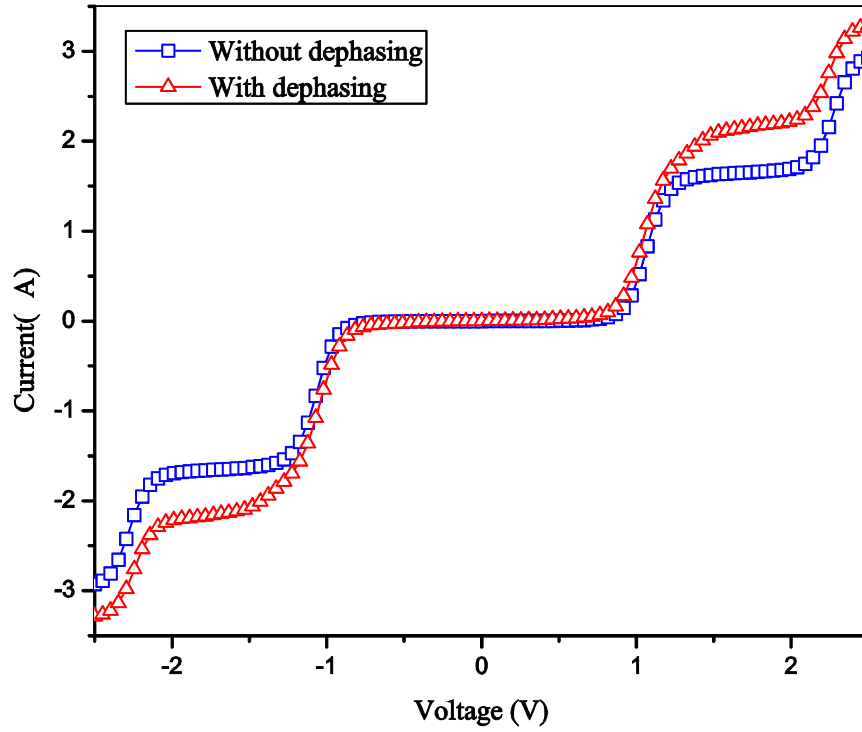


Figure-2
The current-voltage characteristics for the AP spin alignment with and without dephasing

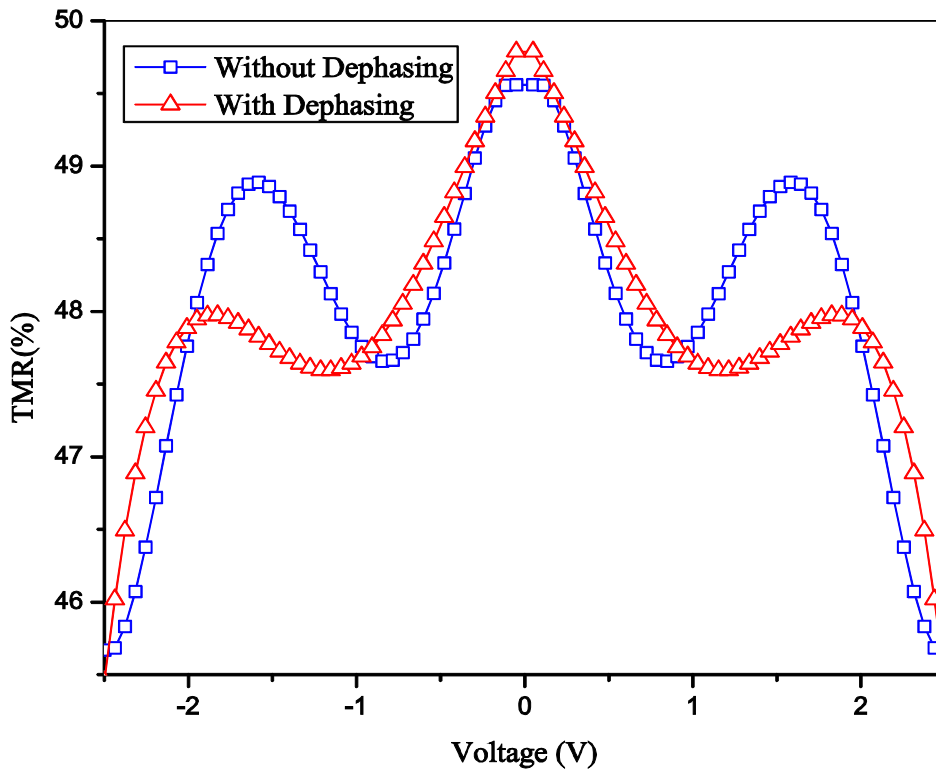


Figure-3
TMR versus applied voltage with and without dephasing

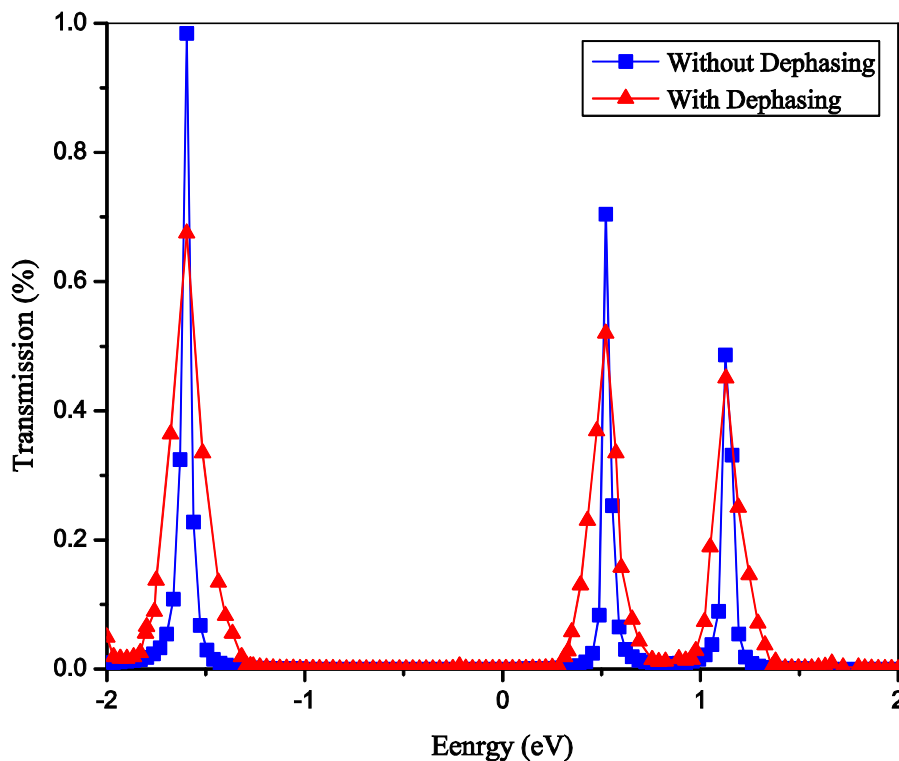


Figure-4

Transmission probability as a function of versus energy characteristics with and without dephasing

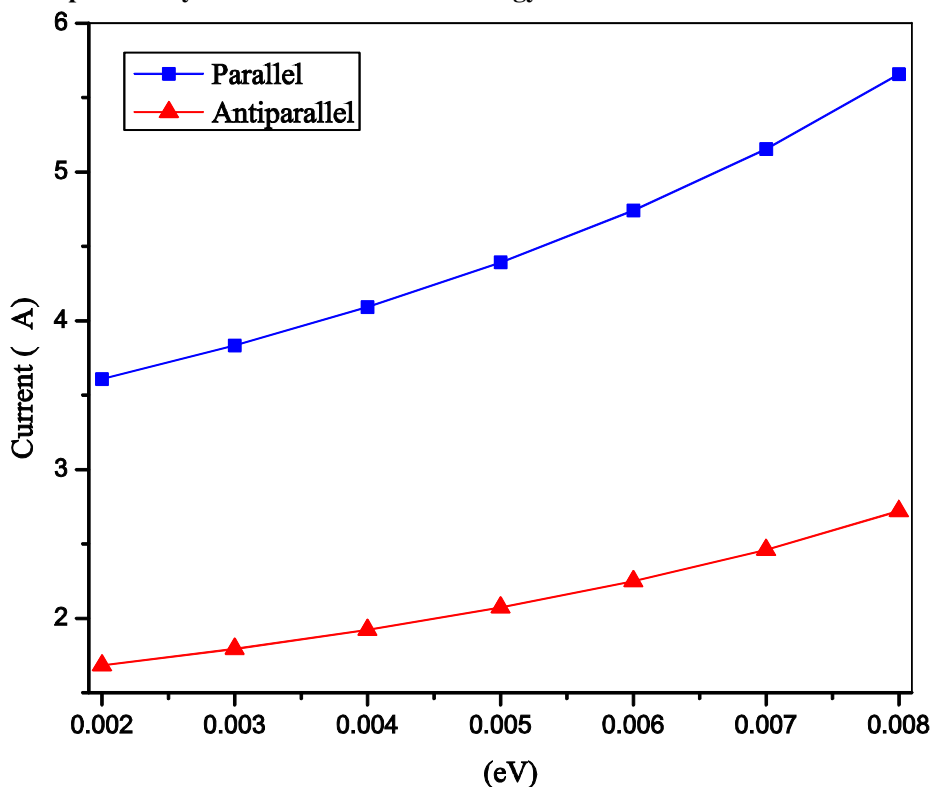


Figure-5

The currents for the P and AP spin alignments as a function of the dephasing strength (γ)

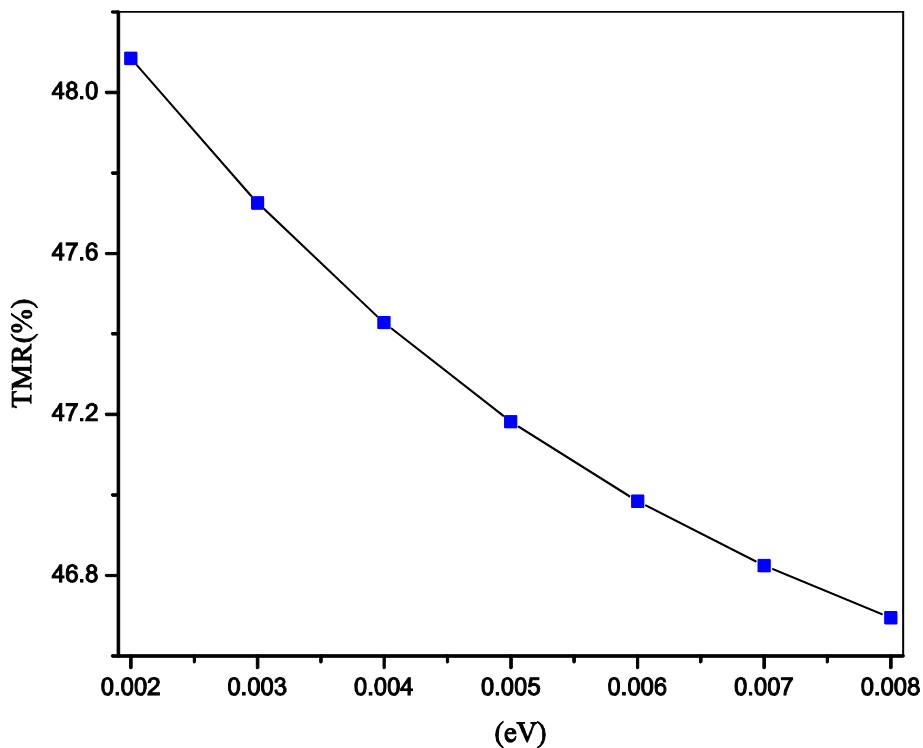


Figure-6
TMR as a function of the dephasing strength (γ)

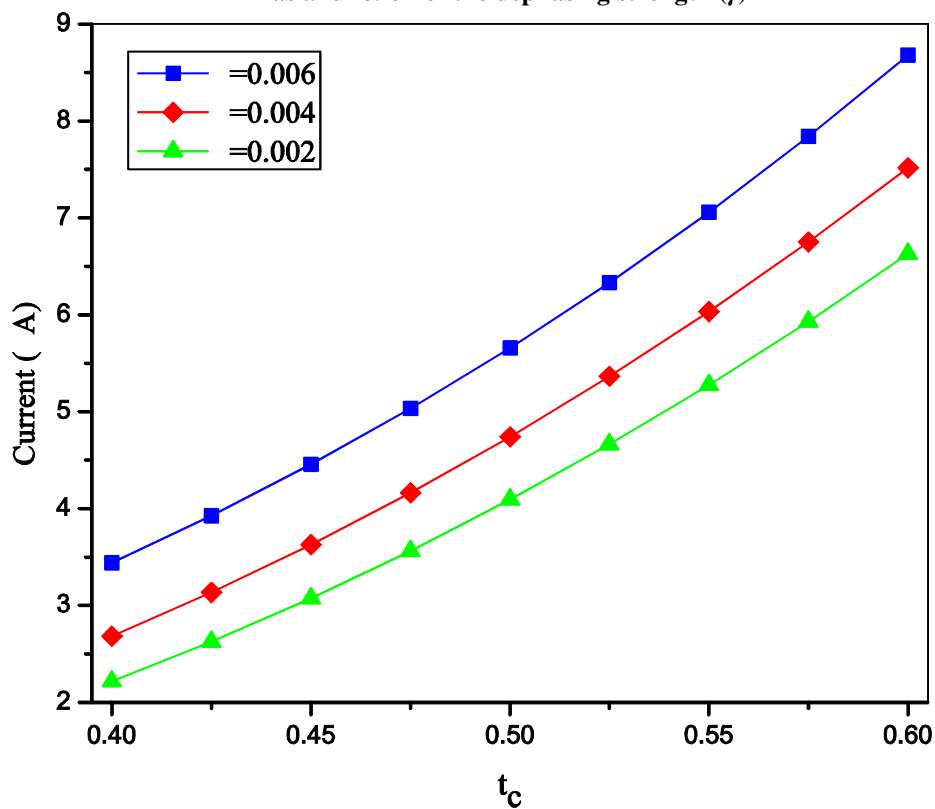


Figure-7
The current for the P spin alignment versus t_c with $\gamma=0,002, 0,004$ and $0,006$

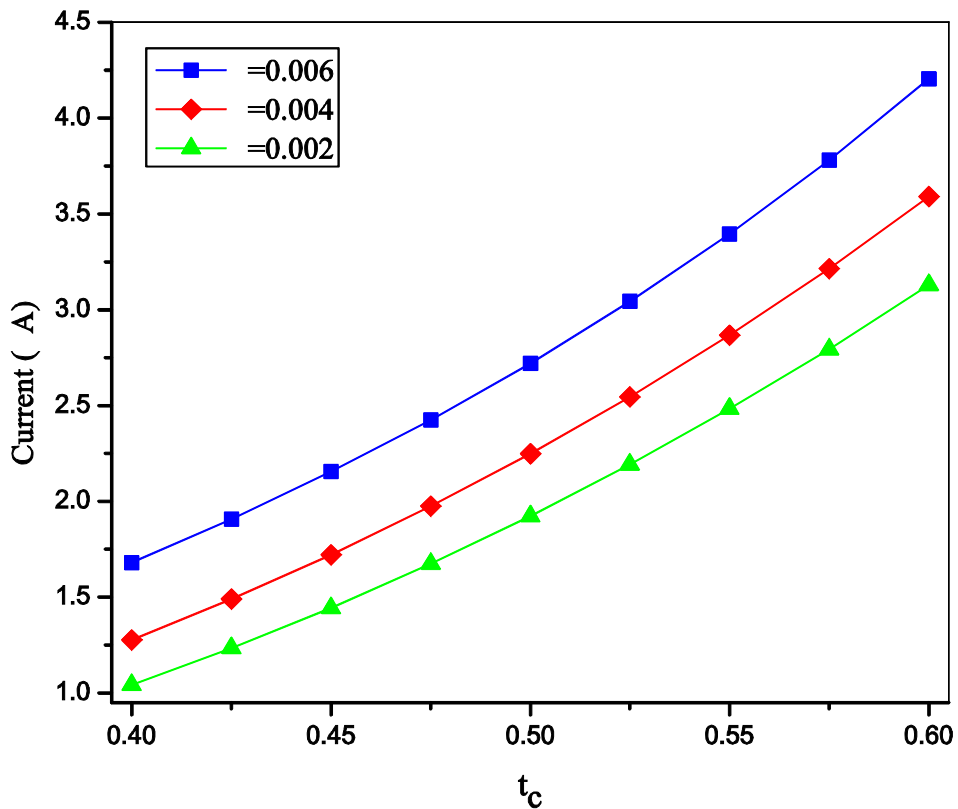


Figure-8

The current for the AP spin alignment versus t_c with $\gamma=0,002, 0,004$ and $0,006$

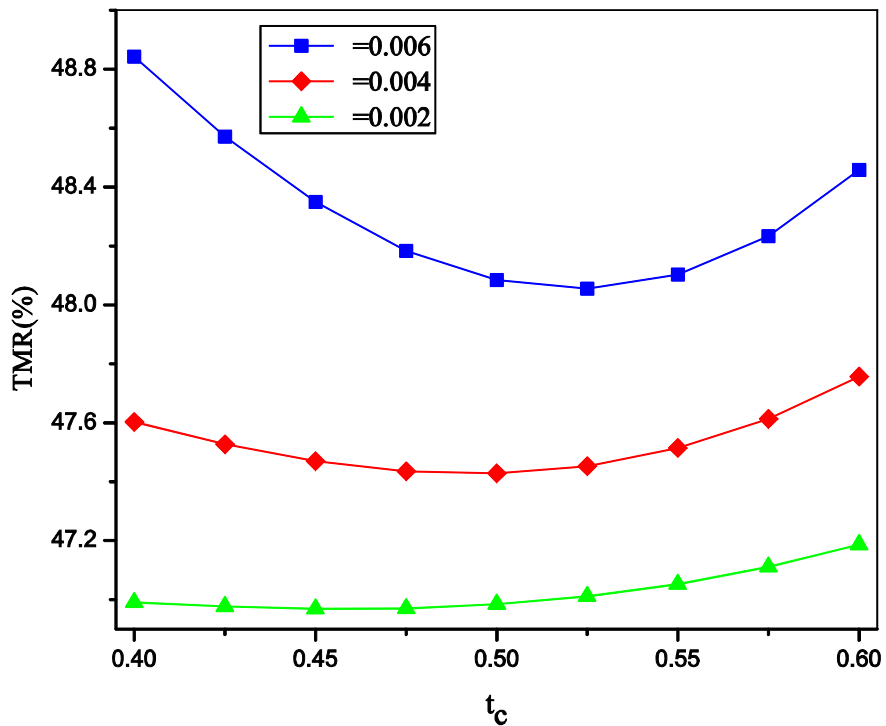


Figure-9

TMR versus t_c with $\gamma=0,002, 0,004$ and $0,006$

Our calculations have been done for a constant applied bias voltage, $V=2.0$ V, and three different values of γ .

With increasing of t_c , the self-energies of the right and left electrodes increase too and the current gains much bigger values. Fig.9, indicates that with increasing of t_c , TMR decreases and for $t_c =0.5$ eV it reaches a minimum value and when t_c increases further it does too.

Conclusion

We have investigated the effects of coupling strength and the electron-phonon interaction on the transport through La@C₈₂ molecule sandwiched between two FM metallic electrodes. The dephasing effect is investigated using the Büttiker dephasing model. We have studied current-voltage characteristics and tunnel magnetoresistance of the system. We have observed that in the presence of dephasing reservoirs, the currents amplitude in the P and AP alignments increase but the TMR decreases. Furthermore TMR maximum ratio is obtained at a lower applied voltage. With increasing of coupling strength, the currents increase too. In this study, we have ignored the effect of electron-electron interaction which we wish to investigate in our future studies.

References

1. Hai P.N., Sakata Y., Yokoyama M., Ohya S. and Tanaka M., *Phys. Rev.*, B **77**, 214435 (2008)
2. Petta J.R., Slater S.K., Ralph D.C., *Phys. Rev. Lett.*, **93**, 136601 (2004)
3. Sothmann B. and König J., *Phys. Rev. B* **82**, 245319 (2010)
4. Ouyang M. and Awschalom D.D., *Science*, **301**, 1074 (2003)
5. Wang Z.C., Su G., Gao S., *Phys. Rev.*, B **63**, 224419 (2001)
6. Shinohara H., *Rep. Prog. Phys.*, **63**, 843 (2000)
7. Liu S. and Sun S., *J. Organomet. Chem.*, **599**, 74 (2000)
8. Poirier D.M., Knapfer M. and Weaver J.H., *Phys. Rev. B*, **49**, 17403 (1994)
9. Hino S., Takahashi H., Iwasaki K. and Matsumoto K., *Phys. Rev. Lett.*, **71**, 4261 (1993)
10. Ton-That C., Dowd A., Shard A.G., Dhanak V.R., Taniaka, A., Shinohara H., Welland M.E., *Phys. Rev.*, B **76**, 165429 (2007)
11. Yang S., Yoon M., Hicke, C., Zhang, Z., Wang, E.: *Phys. Rev. B* **78** 115435 (2008)
12. Warner J.H. et al., *Nano Lett.*, **8**, 1005 (2008)
13. Fong-Ping O., Hui X., *Chin. Phys. Lett.*, **24** 2369 (2007)
14. Sun G. and Kertesz M., *J. Phys. Chem.* **105** 5468 (2001)
15. Waldron D., Haney P., Larade B., MacDonald A. and Guo H., *Phys. Rev. Lett.*, **96** 166804 (2006)
16. Landauer R., *IBM J. Res. Dev.*, **1** 223 (1957)
17. Sergueev N., Roubtsov D. and Guo H., *Phys. Rev. Lett.*, **95** 146803 (2005)
18. Sergueev N., Demkov A.A., Guo H., *Phys. Rev. B*, **75** 233418 (2007)
19. Büttiker M., *Phys. Rev.*, B **33** 3020 (1986)
20. Büttiker M., *IBM J. Res. Dev.* **32** 63 (1988)
21. Brouwer P.W. and Beenakker C.W.J., *Phys. Rev.*, B **55** 4695 (1997)
22. Beenakker C.W.J., *Rev. Mod. Phys.*, **69**, 731 (1997)
23. Liu J. and Iwata S., *Phys. Rev. B*, **50** 5522 (1994)
24. Dong J., Jiang J., Wang Z.D, Xing, D.Y., *Phys. Rev.*, B **51** 1977 (1995)
25. Harigaya K., *Phys. Rev.*, B **45** 13676 (1992)
26. Asai Y. and Fukuyama H., *Phys. Rev.*, B **72**, 085431 (2005)
27. D'Amato J.L., Pastawski H.M., *Phys. Rev.*, B **41**, 7411 (1990)
28. Wang B., Zhu Y., Ren W., Wang J. and Guo H., *Phys. Rev.*, B **75** 235415 (2007)
29. Sun G. and Kertesz M., *J. Phys. Chem.*, A **105** 5468 (2001)
30. Ton-That C., Shard A.G., Egger S., Taninaka A., Shinohara H., Welland M.E., *Surface Science*, **522**, L15 (2008)
31. Taylor J., Brandbyge M. and Stokbro K., *Phys. Rev.*, B **68**, 121101 (2003)
32. Moodera, J.S. et al., *Phys. Rev. Lett.* **80**, 2941 (1998)
33. Moodera J.S., et al., *Phys. Rev. Lett.* **74**, 3273 (1995)
34. Moodera J.S. and Kinder L.R., *J. Appl. Phys.*, **79**, 4724 (1996)