ISSN 2347-3487

Electron transport in double bridges system

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ABSTRACT

In this paper we investigate theoretically the effect of introducing wide band, and Narrow band approximations for the bridge energy band on the electron transport process (ET) through the donor-bridges-acceptor (DBA) system. We using one electron model, for which the Hamiltonian of the system consists of a single-level for both Donor and Accepter (i.e. QD) both coupled to a band bridge as a tight binding interaction. The time dependent Schrödinger equation give us a formula for the occupation probabilities for donor and acceptor levels. The probability of (ET) to the donor and accepter is small because the electron accumulated on the bridge units. Also, the current of acceptor and the effect of different system parameters are studied.

Indexing terms/Keywords

Electron transport; Charge transport; donor-bridge-accepror; wied band and narrow band.

Academic Discipline And Sub-Disciplines

Solid state physics, Nanotechnology;

SUBJECT CLASSIFICATION

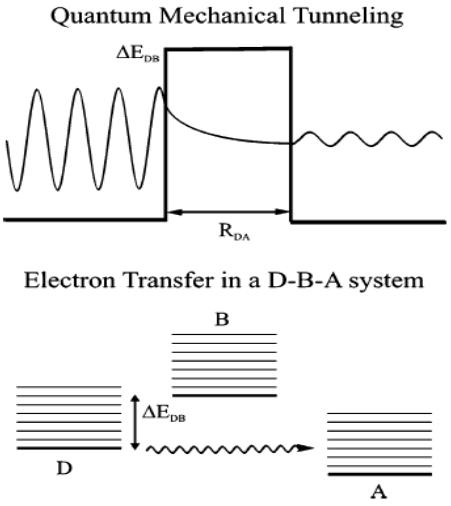
73.21.Fg; 73.21.La; 73.22.Dj; 73.20.At

Council for Innovative Research
Peer Review Research Publishing System
Journal: JOURNAL OF ADVANCES IN PHYSICS
Vol . 9, No. 2
www.cirjap.com, japeditor@gmail.com

ISSN 2347-3487

INTRODUCTION

Electron transport(ET) [1,2] between a donor (D) and an acceptor (A) mediated by a bridge (B) has a fundamental role in a wide set of areas, physical, chemical and biological processes [3]. Recently, D–B–A systems have gained much interest due to the emergence of important potential applications [4] where molecular nano-electronics has now come to fore with potential applications in problems such as sensors, photonics, electro-catalysis and solar photo-conversion [5,6], charge transfer systems in DNA and large biomolecules [7,8] . Bridge mediated ET reactions can occur via different mechanisms [9,10]: incoherent sequential or coherent super exchange [11,12]. Changing a building block of the complex [13] or changing the environment [11,14] can modify which mechanism is mainly at work. The interested parameter to compute is the occupation probability $P_A(t)$ which define the probability of finding the system, that is prepared in the donor state, is in acceptor state. The electron tunneling rate in DBA system is determine by the barrier height (the energy gap between D and B) and width (spacer length between D and A) (see fig.(1)).



Figs.(1): Diagram shows the quantum mechanical tunneling and electron transfer in DBA system [15].

Recently, we have formulated and discuss the electron transfer through single unit bridge in DBA system [16]. The purpose of this study is to investigate the effect of the approximations for the bridge energy band on the occupation probabilities of the donor and acceptor, on the transport of the electron. For this purpose we give a simple, analytical solvable model based on the one electron Hamiltonian model formalism which depends on the evolutions of the wave function amplitude of the relevant DBA system component using time dependent perturbation. Our interest in the electron transport through bridge systems in which both donor and accepter has a discrete levels, same as the quantum dot have, where we chose a single discrete level for each of them that responsible for the process. While, the density of bridge band level is taken in the first approximation as a constant, same as the quantum well density of the states have, and in the second approximation is taken as a nearly single level the one may corresponding to the quantum dot density of states.

THEORTICAL METHOD

In this paper we assumed that the electron transport between the donor (D) and acceptor (A) mediated by a two bridge units ($B_{1,2}$) (see fig.(2))and the electronic coupling between sites is modeled as a tight binding interaction, in terms of nearest neighbor hopping matrix elements, as in McConnell's superexchange model [17], the corresponding Hamiltonian is:

$$\hat{H} = \hat{\mathrm{H}}_{\circ} + \hat{V} \quad (1)$$

where $\hat{\mathrm{H}}_{\circ}$ contains the local Hamiltonian for the donor, acceptor, and bridge units, and $\hat{V}$ is the electronic coupling interaction responsible for the ET process in the isolated system,

$$\hat{\mathrm{H}}_{\circ} = E_D|D\rangle\langle D| + E_A|A\rangle\langle A| + \sum_{j}^{2}\sum_{k}^{m} E_{B_{jk}}|B_{jk}\rangle\langle B_{jk}| \quad (2)$$

$$\hat{V} = \sum_{k}^{m}\left(V_{DB_{1k}}|D\rangle\langle B_{1k}| + V_{B_{1k}D}|B_{1k}\rangle\langle D|\right) + \sum_{k}^{m}\left(V_{AB_{2k}}|A\rangle\langle B_{2k}| + V_{B_{2k}A}|B_{2k}\rangle\langle A|\right) + \sum_{j,l\neq j}^{2}\sum_{k,h}^{m} V_{B_{jk}B_{lh}}|B_{jk}\rangle\langle B_{lh}| \quad (3)$$

Where $V_{DB_{1k}}$, $V_{AB_{2k}}$, and $V_{B_{jk}B_{lh}}$ are the matrix elements which represented the overlap between the wave functions for the components of the (D-B-A) system through a potential regions, and k is the number of the energy level of bridge B_j. The

ISSN 2347-3487

wave function of this system is taken to be a linear combination of bridge units as well as the donor and acceptor wave functions as (in Dirac notations) :

$$\psi(t) = C_D(t)|D\rangle + C_A(t)|A\rangle + \sum_{j}^{2}\sum_{k}^{m} C_{B_{jk}}(t)|B_{jk}\rangle \quad (4)$$

The ET process from the donor to the acceptor through the bridge units is simulated according to the time-dependent Schrodinger equation, $\hat{H}\psi(t) = i\hbar\frac{\partial\psi(t)}{\partial t}$, to obtain the equations of motion for this system as :

$$i\dot{C}_D(t) = E_D C_D(t) + \sum_{k}^{m} V_{DB_{1k}} C_{B_{1k}}(t) \quad (5)$$

$$i\dot{C}_A(t) = E_A C_A(t) + \sum_{k}^{m} V_{AB_{2k}} C_{B_{2k}}(t) \quad (6)$$

$$i\dot{C}_{B_{1k}}(t) = E_{B_{1k}} C_{B_{1k}}(t) + V_{B_{1k}D} C_D(t) + \sum_{k,h}^{m} V_{B_{1k}B_{2h}} C_{B_{2h}}(t) \quad (7)$$

$$i\dot{C}_{B_{2k}}(t) = E_{B_{2k}} C_{B_{2k}}(t) + V_{B_{2k}D} C_D(t) + \sum_{k,h}^{m} V_{B_{2k}B_{1h}} C_{B_{1h}}(t) \quad (8)$$

We taking $\hbar = 1$, assume the following separation:

$$V_{DB_{1k}} = \nu_{B_{1k}} V^{DB_1}, V_{AB_{2k}} = \nu_{B_{2k}} V^{AB_2}, V_{B_{jk}B_{1h}} = \nu_{B_{jk}}\nu_{B_{1h}} V^{B_{jk}B_{1h}}, C_{B_{jk}}(t) = \nu_{B_{jk}}\bar{C}_{B_{jk}}(t), and\ C_{B_{1h}}(t) = \nu_{B_{1h}}\bar{C}_{B_{1h}}(t)$$

Where, $\nu_{B_{1,2k}}$ related with an electronic density of state [18] of bridge as follow:

$$\rho_{B_j}(E_{B_j}) = \sum_{k} \left|\nu_{B_{jk}}\right|^2 \delta(E_{B_{jk}} - E_{B_j})$$

Then the set of equations (5-8) are taking the following forms:

$$\dot{C}_D(t) = -iEC_D(t) - iV^B \int \rho_{B_1}(E_{B_1}) dE_{B_1} \bar{C}_{B_{1k}}(t) \quad (9)$$

$$\dot{C}_A(t) = -iEC_A(t) - iV^B \int \rho_{B_2}(E_{B_2}) dE_{B_2} \bar{C}_{B_{2k}}(t) \quad (10)$$

$$\dot{\bar{C}}_{B_{1k}}(t) = -iE_B\bar{C}_{B_{1k}}(t) - iV^B C_D(t) - iV^{BB} \int \rho_{B_2}(E_{B_2}) dE_{B_2} \bar{C}_{B_2}(t) \quad (11)$$

$$\dot{\bar{C}}_{B_{2k}}(t) = -iE_B\bar{C}_{B_{2k}}(t) - iV^B C_A(t) - iV^{BB} \int \rho_{B_1}(E_{B_1}) dE_{B_1} \bar{C}_{B_1}(t) \quad (12)$$

The approximations for bridge energy band

In this section we discuss two approximation for ET process in (D-B-A) system in which both donor and accepter has a discrete levels, quantum dot, where we chose a single discrete level for each of them that responsible for the process. While, the width of the bridge energy band which have a connection with that of quantum well, wide band approximation, or with that of quantum dot, narrow band approximations [19].

In this paper we refer to a symmetric uniform bridge model, for which $E_{B_1} = E_{B_2} = E_B$and the hopping matrix element between the bridge and the donor/acceptor sites is denoted by V^B and the internal coupling between the bridge units is denoted by V^{BB}. The electron is assumed to be initially localized on a donor so that $C_D(0) = 1, C_A(0) = 0, and\ C_{B_{1,2}}(0) = 0$.

Wide band approximation

In this approximation the density of bridge band level, $\rho_{B_{1,2}}(E)$, is taken as a constant by taking its average over the energy, $\bar{\rho}_{B_{1,2}} = 1/4\beta_{B_{1,2}}$, with $4\beta_{B_{1,2}}$ is the band width of the bridges 1 and 2. Consequently, the (D-B-A) system which are the Quantum Dot-Quantum well-Quantum Dot (QD-QW-QD) system. The set of equations (9-12) becomes:

$$\dot{C}_D(t) = -iEC_D(t) - iV^B\bar{\rho}_{B_1}\int \bar{C}_{B_{1k}}(t) dE_{B_1} \quad (13)$$

$$\dot{C}_A(t) = -iEC_A(t) - iV^B\bar{\rho}_{B_2}\int \bar{C}_{B_{1k}}(t)dE_{B_2} \quad (14)$$

$$\dot{\bar{C}}_{B_{1k}}(t) = -iE_B\bar{C}_{B_{1k}}(t) - iV^B C_D(t) - iV^{BB}\bar{\rho}_{B_2}\int \bar{C}_{B_2}(t)dE_{B_2} \quad (15)$$

$$\dot{\bar{C}}_{B_{2k}}(t) = -iE_B\bar{C}_{B_{2k}}(t) - iV^B C_A(t) - iV^{BB}\bar{\rho}_{B_1}\int \bar{C}_{B_1}(t)dE_{B_1} \quad (16)$$

By using Green's function [20] to solve eqs.(15) and (16), and utilizing the integral $\delta(t-\acute{t}) = \frac{1}{\pi}\int e^{-iE_B(t-\acute{t})}dE_B$, $\delta(t-\acute{t})$ represent Dirac Delta function[21], and since $\int_a^b \delta(t-\acute{t})f(\acute{t})d\acute{t} = f(t)$ we get:

$$\int \bar{C}_{B_1}(t)dE_{B_1} = \frac{-i\pi V^{B_1 D}C_D(t) - \pi^2 V^{B_2 A}V^{B_1 B_2}\bar{\rho}_{B_2}C_A(t)}{1+\pi^2|V^{B_1\bar{B}_2}|^2\bar{\rho}_{B_1}\bar{\rho}_{B_2}} \quad (17)$$

$$\int \bar{C}_{B_2}(t)dE_{B_2} = \frac{-i\pi V^{B_2 A}C_A(t) - \pi^2 V^{B_1 D}V^{B_2 B_1}\bar{\rho}_{B_1}C_D(t)}{1+\pi^2|V^{\bar{B}_1\bar{B}_2}|^2\bar{\rho}_{B_1}\bar{\rho}_{B_2}} \quad (18)$$

Using eqs.(17) in (13) and (18) in (14). We get:

$$\dot{C}_D(t) = -i(E - i\Delta_{D2})C_D(t) + iV_{d2}C_A(t) \quad (19)$$

$$\dot{C}_A(t) = -i(E - i\Delta_{A2})C_A(t) + iV_{d2}C_D(t) \quad (20)$$

Where:

$\Delta_D = \frac{\pi|V^B|^2\rho_{B_1}}{1+\pi^2|V^{BB}|^2\bar{\rho}_{B_1}\bar{\rho}_{B_2}}$, $V_d = \frac{\pi^2|V^B|^2V^{BB}\rho_{B_1}\bar{\rho}_{B_2}}{1+\pi^2|V^{BB}|^2\bar{\rho}_{B_1}\bar{\rho}_{B_2}}$, and $\Delta_A = \frac{\pi|V^B|^2\rho_{B_2}}{1+\pi^2|V^{BB}|^2\bar{\rho}_{B_1}\bar{\rho}_{B_2}}$

Now by using Laplace transform to solve the eqs.(19) and (20), and then applying on the results its inverse Laplace transform [22] one gets,

$$C_D(t) = \frac{1}{2}e^{-\frac{a_1}{2}t}\left[\frac{2c_1\cos(c_1t)+(2iE-a_1+2\Delta_A)\sin(c_1t)}{c_1}\right] \quad (21)$$

$$C_A(t) = i\frac{V_d}{c_1}e^{-\frac{a_1}{2}t}\sin(c_1t) \quad (22)$$

Where,

$a_1 = i\big(2E - i(\Delta_D + \Delta_A)\big), b_1 = -E^2 + iE(\Delta_D + \Delta_A) + \Delta_D\Delta_A + V_d^2$, and $c_1^2 = b_1 - \frac{a_1^2}{4}$

Narrow band approximation

In this approximation the density of bridge band level, $\rho_{B_{1,2}}(E)$, is taken as a nearly single level the one may corresponding to the quantum dot density of states. Consequently, the (D-B-A) system which are the Quantum Dot-Quantum Dot-Quantum Dot (QD-QD-QD) system. The set of equations (9-12) becomes:

$$\dot{C}_D(t) = -iEC_D(t) - iV^B\bar{C}_{B_{1k}}(t) \quad (23)$$

$$\dot{C}_A(t) = -iEC_A(t) - iV^B\bar{C}_{B_{1k}}(t) \quad (24)$$

$$\dot{\bar{C}}_{B_{1k}}(t) = -iE_B\bar{C}_{B_{1k}}(t) - iV^B C_D(t) - iV^{BB}\bar{C}_{B_2}(t) \quad (25)$$

$$\dot{\bar{C}}_{B_{2k}}(t) = -iE_B\bar{C}_{B_{2k}}(t) - iV^B C_A(t) - iV^{BB}\bar{C}_{B_1}(t) \quad (26)$$

By using Laplace transform to solve above equations:

$$SC_D(S) - 1 = -iEC_D(S) - iV^B\bar{C}_{B_1}(S) \quad (27)$$

$$SC_A(S) = -iEC_A(S) - iV^B\bar{C}_{B_2}(S) \quad (28)$$

$$S\bar{C}_{B_1}(S) = -iE_B\bar{C}_{B_1}(S) - iV^B C_D(S) - iV^{BB}\bar{C}_{B_2}(S) \quad (29)$$

$$S\bar{C}_{B_2}(S) = -iE_B\bar{C}_{B_2}(S) - iV^B C_A(S) - iV^{BB}\bar{C}_{B_1}(S) \quad (30)$$

Using eq.(29) in eq.(27) and (30) in eq.(28), we get:

$$C_D(S) = \frac{G_D[(S+iE_B)^2 - i^2|V^{BB}|^2]}{[G_D^{-1} - i^2G_A^2]} \quad (31)$$

$$C_A(S) = -i\frac{G_A\left[(S+iE_B)^2 - i^2|V^{BB}|^2\right]}{\left[G_D^2 - i^2G_A^2\right]} \quad (32)$$

Where:

$G_D = (S+iE)[(S+iE_B)^2 - i^2|V^{BB}|^2] - i^2|V^B|^2(S+iE_B)$, and $G_A = i^2V^{BB}|V^B|^2$

Then we can rewrite the eqs.(31) and (32) as the following forms:

$$C_D(S) = \frac{AS+B}{\left[(S+\frac{a_2}{2})^2+c_2^2\right]} + \frac{CS+D}{\left[(S+\frac{\acute{a}_2}{2})^2+\acute{c}_2^2\right]} \quad (33)$$

$$C_A(S) = -i\left\{\frac{\acute{A}S+\acute{B}}{\left[\left(S+\frac{a_2}{2}\right)^2+c_2^2\right]} + \frac{\acute{C}S+\acute{D}}{\left[\left(S+\frac{\acute{a}_2}{2}\right)^2+\acute{c}_2^2\right]}\right\} \quad (34)$$

Where:

$a_2 = i(E+E_B-V^{BB}), \acute{a}_2 = i(E+E_B+V^{BB}), a_3 = i(E+2E_B),\ b_2 = -E(E_B-V^{BB}) + |V^B|^2,$

$\acute{b}_2 = -E(E_B+V^{BB}) + |V^B|^2, b_3 = -\left(E_B^2 + 2EE_B - \left|V_s^{BB}\right|^2 - \left|V_s^B\right|^2\right), c_2 = \sqrt{b_2 - \frac{a_2^2}{4}}, \acute{c}_2 = \sqrt{\acute{b}_2 - \frac{\acute{a}_2^2}{4}},$

$c_3 = -i(EE_B^2 - E|V^{BB}|^2 - E_B|V^B|^2), G = a_3\left(b_2\acute{b}_2 - \acute{b}_2^2\right) - \acute{b}_2(b_3(a_2-\acute{a}_2) + \acute{a}_2b_2) + a_2\acute{b}_2^2 - c_3\left(\acute{a}_2^2 - a_2\acute{a}_2 + b_2 - \acute{b}_2\right), \acute{G} = G_A\left(a_2\acute{a}_2 - \acute{a}_2^2 + \acute{b}_2 - b_2\right), F = \left(2b_2\acute{b}_2 - \acute{b}_2^2 - a_2^2\acute{b}_2 + a_2\acute{a}_2\acute{b}_2 - b_2^2 + a_2\acute{a}_2b_2 - \acute{a}_2^2b_2\right)$

$A = 1 - \frac{(a_1-\acute{a}_2)\acute{b}_2F - Fc_1 + G(b_2-\acute{b}_2)}{\acute{b}_2F(a_2-\acute{a}_2)}, \acute{A} = \frac{H_DF - \acute{G}(b_2-\acute{b}_2)}{\acute{b}_2F(a_2-\acute{a}_2)}, B = \frac{Fc_1 - b_2G}{\acute{b}_2F}, \acute{B} = \frac{H_DF - b_2\acute{G}}{\acute{b}_2F}, D = \frac{G}{F}, \acute{D} = \frac{\acute{G}}{F},$

$C = \frac{(a_2-\acute{a}_2)\acute{b}_2F - Fc_1 + G(b_2-\acute{b}_2)}{\acute{b}_2F(a_2-\acute{a}_2)}, and\ \acute{C} = \frac{-H_DF + \acute{G}(b_2-\acute{b}_2)}{\acute{b}_2F(a_2-\acute{a}_2)}$

Now by applying on the eqs.(33) and (34) its inverse Laplace transform one gets,

$$C_D(t) = \frac{e^{-\frac{a_2}{2}t}[-a_2A\sin(c_2t) + 2Ac_2\cos(c_2t) + 2B\sin(c_2t)]}{2c_2} + \frac{e^{-\frac{\acute{a}_2}{2}t}[-\acute{a}_2C\sin(\acute{c}_2t) + 2C\acute{c}_2\cos(\acute{c}_2t) + 2D\sin(\acute{c}_2t)]}{2\acute{c}_2} \quad (35)$$

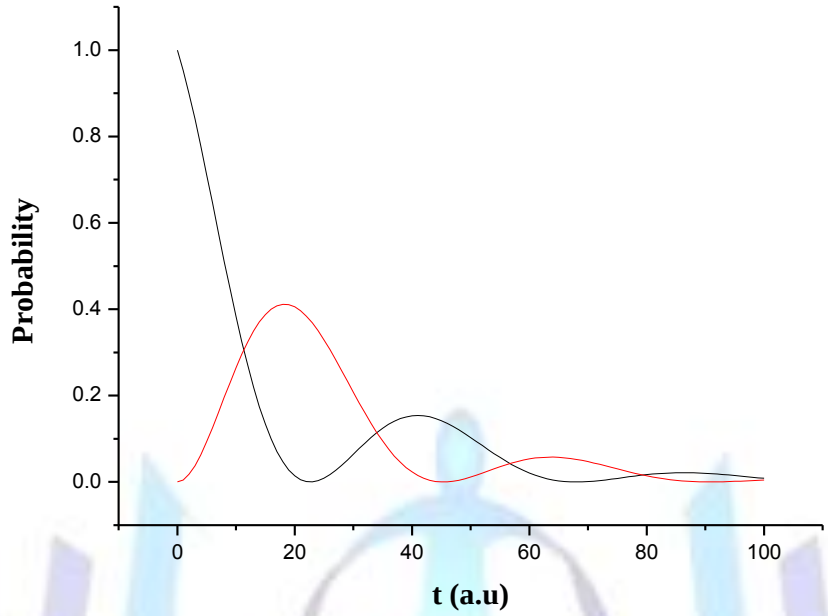
$$C_A(t) = -i\left\{\frac{e^{-\frac{a_2}{2}t}[-a_2\acute{A}\sin(c_2t) + 2\acute{A}c_2\cos(c_2t) + 2\acute{B}\sin(c_2t)]}{2c_2} + \frac{e^{-\frac{\acute{a}_2}{2}t}[-\acute{a}_2\acute{C}\sin(\acute{c}_2t) + 2\acute{C}\acute{c}_2\cos(\acute{c}_2t) + 2\acute{D}\sin(\acute{c}_2t)]}{2\acute{c}_2}\right\} \quad (36)$$

RESULTS AND DISCUSSIONS

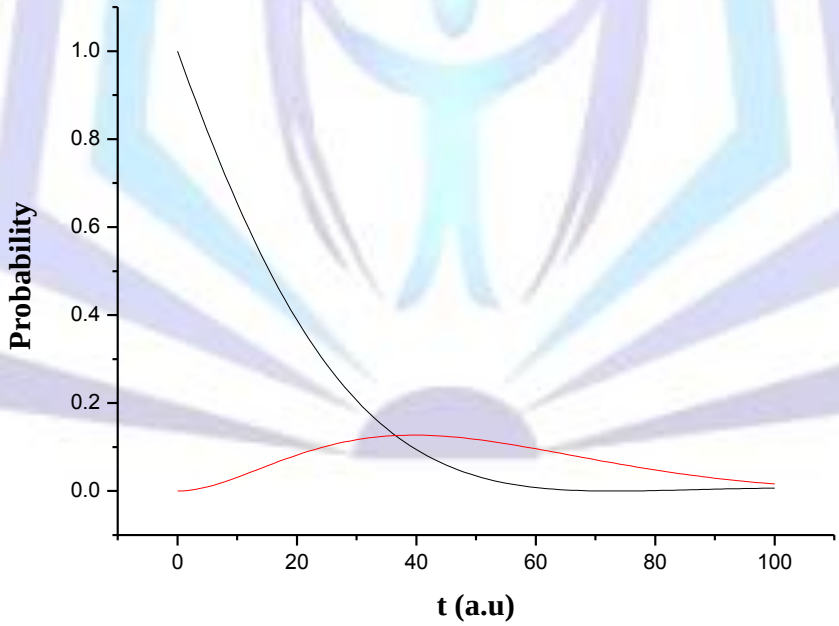
There are many system parameters characterize the DBA system which affect the electron transport process and consequently affect the occupation probabilities($P_D(t) = |C_D(t)|^2$ and $P_A(t) = |C_A(t)|^2$). Such parameters are the time t variation, spacer length between donor and acceptor, or may be bridge length, as well as other parameter that characterize the bridge system such as coupling matrix elements (V^B between donor/acceptor and bridge units, V^{BB} between bridge units) which are depend on the spatial variation of the corresponding wave functions.

Results of electron transport simulations performed according to the method described in theoretical method Section, where we are arranging the bridge units in a parallel configuration along the straight line between donor and acceptor, are shown in the following Figures: In the case of wide bridge band, the charge decay from the donor level or its evolve on the acceptor level then it's the occupation probabilities saturated at small value and the charge accumulation on the bridge units because each level (of donor or acceptor) has broadened by an amount $\Delta_{(D,A)}$ due to their interactions with the levels of bridge units, and it is a collective effect from all bridge units, which make the amplitude of the oscillation less and overall acceptor occupation probability is less also the interaction V_d resulted from the interactions of the bridges levels among them self and with both the donor and the acceptor levels, which leading to less electron transport because the electron accumulated on the bridges, When the interaction strength between donor / acceptor and bridge units is smaller than in between the bridge units interactions leads to increases the number of oscillations that appears in the occupation probabilities (see fig.(2)). However, these oscillations are vanished when the interaction strength in between donor/ acceptor and bridge units becomes larger than in between the bridge units (see fig.(3)).

ISSN 2347-3487



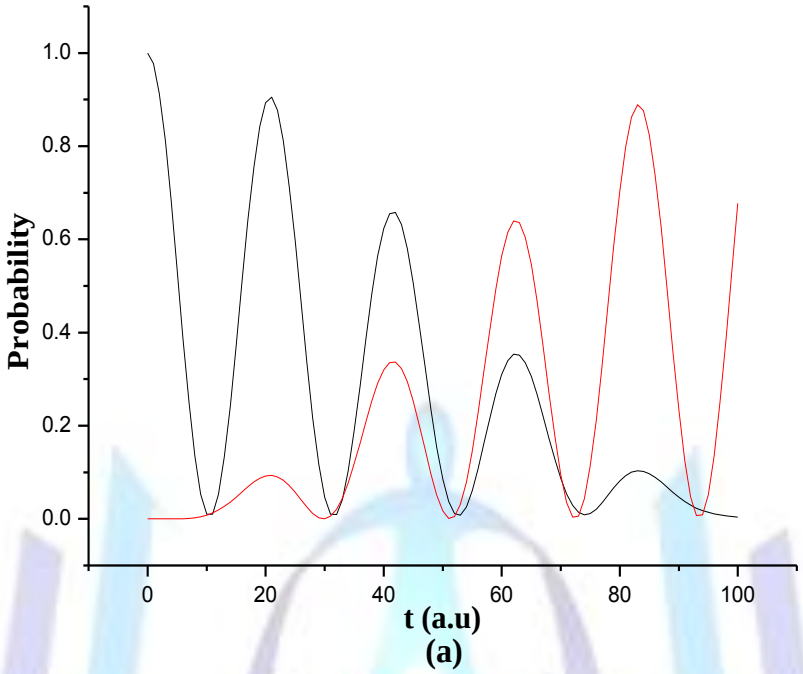
Figs.(2): Time-dependent of survival probability of charge on a donor(black color) and acceptor (red color) as function of time, using the following values: $E = -0.15, E_B = -0.17, V_s^B = 0.1, V_s^{BB} = 0.15$.



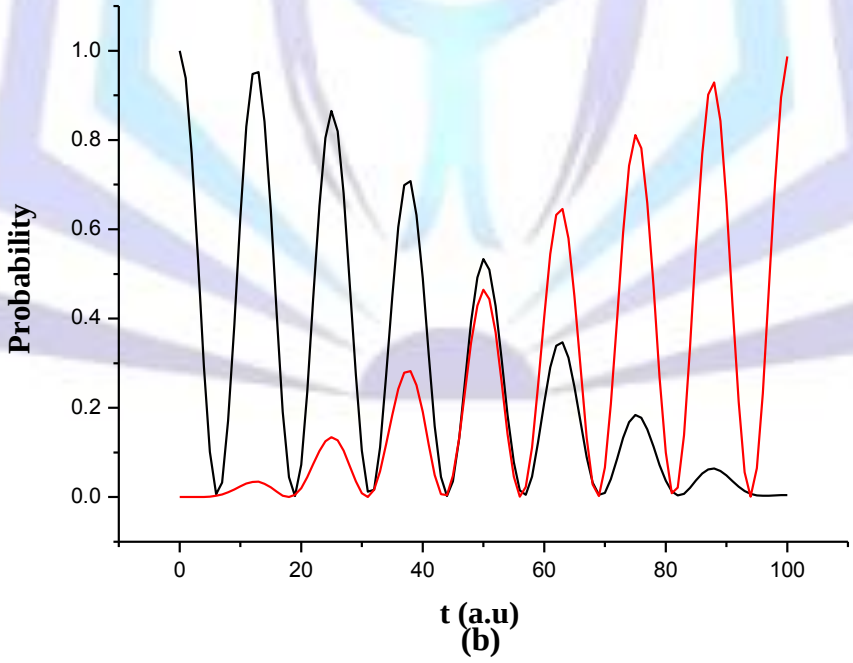
Figs.(3): Time-dependent of survival probability of charge on a donor(black color) and acceptor (red color) as function of time, using the following values: $E = -0.15, E_B = -0.17, V_s^B = 0.1, V_s^{BB} = 0.05$.

In the case of narrow bridge band, we notes the oscillatory behavior and the number of oscillations within any time range it depends on the strength of the coupling between donor/acceptor and bridge units (see fig.(4)), However, these oscillations are increase when the interaction strength in between donor/ acceptor and bridge units becomes smaller than in between the bridge units (see fig.(5)).

ISSN 2347-3487

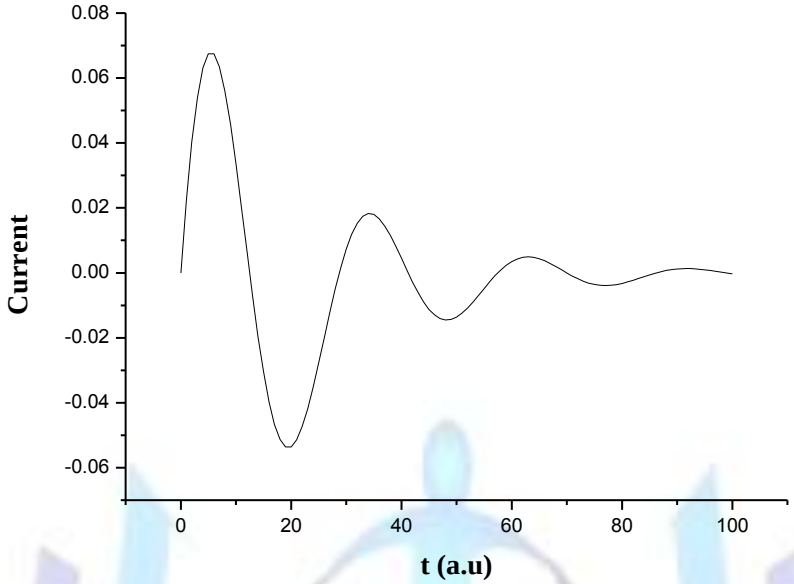


Figs.(4): Time-dependent of survival probability of charge on a donor(black color) and acceptor (red color) as function of time, using the following values: $E = -0.15, E_B = -0.17, V_?^B = 0.15, V_?^{BB} = 0.03$.

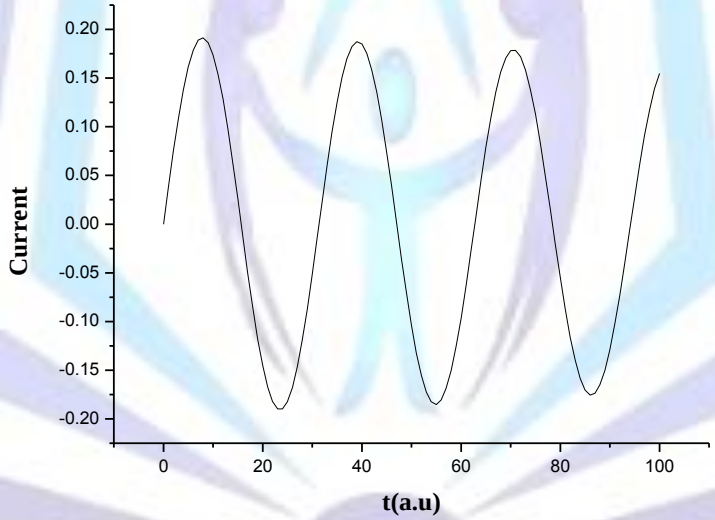


Figs.(5): Time-dependent of survival probability of charge on a donor(black color) and acceptor (red color) as function of time, using the following values: $E = -0.15, E_B = -0.17, V_?^B = 0.25, V_?^{BB} = 0.03$.

In the wide bridge band we notes growth the current of acceptor, $j(t) = dP_A(t)/dt$, (see fig.(6)) and we note the oscillatory behavior in the case of narrow bridge band (see fig.(7)).



Figs.(6): The current of acceptor as a function of time, the case of wide bridge band.



Figs.(7): The current of acceptor as a function of time, the case of narrow bridge band.

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