Theoretical Evaluation of Flow Electronic Rate at Au /TFB Interface

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Abstract. This investigation presents a quantum model for electron transfer allows us to calculate the flow electronic transfer rate through metal/molecule devices. The flow electronic transfer rate at interfaces between Au metal contact with poly(9,9'-dioctylfluorene-co-bis-N,N'-(4-butylyphenyl)-diphenylamine) (TFB) and solvents media with polarity is ranging from 0.315 for Diethyl ether to 0.5361 for Methanol is investigated via quantum theory of donor acceptor model. The flow rate are large with Au energy level alignment with TFB levels has been found when the potential barrier at interface is 0.066 eV with Diethyl ether solvents. Flow electronic transfer rate for Au/TFB system with methanol solvent decreased cross interface when the potential barrier is as large as 0.122 eV. This lead to produce accumulate the charges on the both side of Au and TFB when the potential at interface is large. The resulting slow transfer of electrons cross interface. Furthermore, Au metal contact on TFB molecule interface frequently with different solvents media and have large flow rate with Diethylether solvents and low flow rate with Methanol.

Keywords: Flow electronic rate, Au metal, TFB molecule.

1. Introduction

One of the important active field research in recent years is electronic transfer in different science chemistry, physical, biophysical and electronic material [1]. It has basic processes for using different materials in promises technology, such as photo catalysis, light harvesting, molecular electronics and solar cell [2]. The electronic transition field has been extended in different branch of physical, technologic material and microelectronic devices [3]. The optoelectronic and organic electronic devices have been studiedly and investigated in recent years in much more technology for applications such as Organic Light Emission Diodes (OLEDs) [4]. Electronic transfer at the levels of molecules plays an important role in different areas of physics, chemistry, biology and variety materials science [5]. After the seminal work of Marcus in 1958, the electron transfer still a more active field in different research area. The distribution of charge in condensed matter and molecules belongs to the more an important event in all photo induced processes in biology and chemical systems and
technological materials [6]. However, the levels of energy for molecules relative to levels energy for metals are the fundamental rule for electronic transfer at molecule-metal system [7]. The molecular orbital levels alignment with the Fermi energy levels of metal play an important to determine the potential barriers [8], the energy levels for Au contact with TFB are shown in Fig.1 [4,9]. In this research, the theoretical flow electronic rate at Au/TFB device are evaluated and studied via quantum theory. For the system, the transition energy, activation, barrier and flow electronic rate will be present.

Fig.1. Illustrated of the energy levels of Au metal contact with TFB molecule [4,9].

2. Theory

The flow electronic transfer from metal to molecule cross interface is given by [10].

$$J_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \frac{F(\epsilon)}{(\mathcal{M}_{MM})^2} \delta(\epsilon_k - \epsilon) d\epsilon \quad \cdots \cdots \ (1)$$

Where $\hbar$, $\hbar = \frac{h}{2\pi}$, $h$ is the Planck constant, $F(\epsilon)$ is the Fermi-Dirac occupation $\mathcal{M}_{MM}$ is coupling parameters and $\delta(\epsilon_k - \epsilon)$ is the delta Dirac function for energy at $k$ state of metal and energy levels of molecule.

The effective density of molecule-metal states system $\rho_{ef}(\epsilon)$ is equivalent to [11].

$$\rho_{ef}(\epsilon) = \int \delta(\epsilon_k - \epsilon) dk \quad \cdots \cdots \ (2)$$

The Fermi-Dirac occupation for electrons in levels state may be written as [12].

$$F(\epsilon) = \left( e^{\frac{\epsilon}{k_B T}} + 1 \right)^{-1} \quad \cdots \cdots \ (3)$$

Substituting the Eqs. (2) and (3) in Eq. (1) to give:

$$J_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \left( e^{\frac{\epsilon}{k_B T}} + 1 \right)^{-1} \left( \mathcal{M}_{MM} \right)^2 \rho_{ef}(\epsilon) d\epsilon \quad \cdots \cdots \ (4)$$

We introduce the density of state for electrons $\hat{\rho}(E_i)$ in system, then Eq. (4) become.

$$J_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \left( e^{\frac{\epsilon}{k_B T}} + 1 \right)^{-1} \left( \mathcal{M}_{MM} \right)^2 \hat{\rho}(E_i) \rho_{ef}(\epsilon) d\epsilon \quad \cdots \cdots \ (5)$$

The density of state for metal/molecule system was given:

$$\hat{\rho}(E_i) = \sqrt{\frac{1}{4\pi\hbar\Lambda_{M}k_B} e^{-\frac{(\Lambda_{MM} + \Delta U_0)^2}{4\hbar\Lambda_{MM}k_B}}} \quad \cdots \cdots \ (6)$$

Inserting Eq. (6) in Eq. (5) produce:

$$J_{MM} = \frac{2\pi}{\hbar} \sqrt{\frac{1}{4\pi\hbar\Lambda_{M}k_B}} \int_{-\infty}^{\infty} \left( e^{\frac{\epsilon}{k_B T}} + 1 \right)^{-1} e^{-\frac{(\Lambda_{MM} + \Delta U)^2}{4\hbar\Lambda_{MM}k_B}} \left( \mathcal{M}_{MM} \right)^2 \rho_{ef}(\epsilon) d\epsilon \quad \cdots \cdots \ (7)$$

However, the electrons will be transfer above Fermi level with energy $\epsilon$, the activation energy $\Delta U^0$ for transfer reduced to [13].
\[ \Delta U = \Delta \epsilon - \epsilon \quad \text{ .......... (8)} \]

Substituting Eq.(8) in Eq.(7) reduce to:
\[ \mathbb{J}_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \frac{e^\epsilon}{4\lambda M_k \hbar^2} e^{\frac{\epsilon}{\hbar^2}} \left( \langle \mathcal{M}_m \rangle \right)^2 \rho_{eff}(\epsilon) d\epsilon \quad \text{ .......... (9)} \]

Furthermore, the exponent factor in the Eq. (9) simply to:
\[ e^{(\Delta \lambda M_k + \Delta \epsilon) / 4\lambda M_k \hbar^2} = e^{(\Delta \lambda M_k + \Delta \epsilon) / 4\lambda M_k \hbar^2} e^{\Delta \epsilon / 4\lambda M_k \hbar^2} e^{- \epsilon^2 / 4\lambda M_k \hbar^2} \quad \text{ .......... (10)} \]

Under assume \( A = (\lambda M_k + \Delta \epsilon) / 4\lambda M_k \hbar^2 \) and \( B = 1 / 4\lambda M_k \hbar^2 \) and inserting Eq.(10) in Eq.(9). results.
\[ \mathbb{J}_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \frac{e^\epsilon}{4\lambda M_k \hbar^2} e^{\frac{\epsilon}{(\lambda M_k + \epsilon - IE) / 4\lambda M_k \hbar^2}} \left( \langle \mathcal{M}_m \rangle \right)^2 \rho_{eff}(\epsilon) d\epsilon \quad \text{ .......... (11)} \]

The potential barrier is given by:
\[ \Delta V_{eff} = \frac{(\lambda M_k + \Delta \epsilon)^2}{4\lambda M_k \hbar^2} = \frac{(\lambda M_k + \epsilon - IE)^2}{4\lambda M_k \hbar^2} \quad \text{ .......... (12)} \]

Where \( \epsilon_F \) is Fermi energy and \( IE \) is ionization energy. Furthermore, the effective density \( \rho_{eff}(\epsilon) \) is given by [13].
\[ \rho_{eff}(\epsilon) = \rho_M(E) \frac{L_M}{d_{adm}^{2/3}(\frac{h}{\pi})^2} \quad \text{ .......... (13)} \]

Where \( \rho_M(E) \) is the metal density of states, \( L_{ecl} \) is effect of coupled length and \( d_{adm} \) is atomic density metal.

Inserting Eq.(13) and Eq.(12) with Eq.(11) to obtained:
\[ \mathbb{J}_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \frac{e^\epsilon}{4\lambda M_k \hbar^2} e^{\frac{\epsilon}{(\lambda M_k + \epsilon - IE) / 4\lambda M_k \hbar^2}} \left( \langle \mathcal{M}_m \rangle \right)^2 \rho_{eff}(\epsilon) \frac{L_M}{d_{adm}^{2/3}(\frac{h}{\pi})^2} \int_{-\infty}^{\infty} e^{\frac{\epsilon}{\hbar^2}} \left( \langle \mathcal{M}_m \rangle \right)^2 d\epsilon \quad \text{ .......... (14)} \]

Assume that
\[ I = \int_{-\infty}^{\infty} e^{\frac{\epsilon}{\hbar^2}} \left( \langle \mathcal{M}_m \rangle \right)^2 d\epsilon \quad \text{ .......... (15)} \]

Where
\[ e^{-\epsilon A} e^{-\epsilon B} = 1 - \epsilon A + \frac{(\epsilon A)^2}{2!} - \frac{(\epsilon A)^3}{3!} + \frac{(\epsilon A)^4}{4!} - \ldots \quad \text{ .......... (16)} \]

And
\[ e^{-\epsilon B} = 1 - \epsilon B + \frac{(\epsilon B)^2}{2!} - \frac{(\epsilon B)^3}{3!} + \frac{(\epsilon B)^4}{4!} - \ldots \quad \text{ .......... (17)} \]

The solution of integrals in Eq. (15) give:
\[ I = 2k_B T(T) \xi(n) \xi(n) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(3)(k_B T)^3 \xi \left( \frac{1}{n} \right) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(5)(k_B T)^5 \xi \left( \frac{1}{n} \right) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(7)(k_B T)^7 \xi \left( \frac{1}{n} \right) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(9)(k_B T)^9 \xi \left( \frac{1}{n} \right) + \ldots \quad \text{ .......... (18)} \]

Inserting Eq. (18) in Eq. (14) to results:
\[ \mathbb{J}_{MM} = \frac{2\pi}{\hbar} \int_{-\infty}^{\infty} \frac{e^\epsilon}{4\lambda M_k \hbar^2} e^{\frac{\epsilon}{(\lambda M_k + \epsilon - IE) / 4\lambda M_k \hbar^2}} \left( \langle \mathcal{M}_m \rangle \right)^2 \rho_{eff}(\epsilon) \frac{L_M}{d_{adm}^{2/3}(\frac{h}{\pi})^2} e^{-\epsilon A - \epsilon B} \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(3)(k_B T)^3 \xi \left( \frac{1}{n} \right) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(5)(k_B T)^5 \xi \left( \frac{1}{n} \right) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(7)(k_B T)^7 \xi \left( \frac{1}{n} \right) + 2 \left( \frac{\pi}{2!} - \frac{\pi}{4!} + \frac{\pi}{4!} \right) \Gamma(9)(k_B T)^9 \xi \left( \frac{1}{n} \right) + \ldots \quad \text{ .......... (19)} \]
The transition energy $\Lambda_{mM} eV$ for electron transfer is [14]:

$$\Lambda_{mM}(eV) = \frac{e^2}{8\pi \varepsilon_0 \varepsilon} \left( \frac{1}{n^2} - \frac{1}{\varepsilon} \right) \left( \frac{1}{r} - \frac{1}{2R} \right) \quad \ldots \ldots \quad (20)$$

where $e$ is electrons charge, $\varepsilon_0$ is permittivity of vacuum, $\varepsilon$ and $n$ are dielectric constant and refractive index of the solvents, $R$ is distance between the molecule and metal electrode, $r$ is molecule radius.

The radii of material could be estimation using an expression approach [15].

$$r = \left( \frac{3M}{4\pi ND} \right)^{\frac{1}{3}} \quad \ldots \ldots \quad (21)$$

here $M$ is the molecular weight, $D$ is the mass density and $N$ is Avogadro’s number.

### Results

Calculate and investigate here the flow electronic rate of metal/molecule interface and contact with each other. Use theoretical model due to quantum picture to examine the flow electronic rate of these devices, and evaluate the transition energy, driving energy and barrier for electrons. The Au/ poly(9,9'-dioctyfluorene-co-bis-N,N'-(4-butylphenyl) diphenylamine) (TFB) model is chosen to be investigated. The flow electronic transfer cross contacts that’s shown in Fig.1. The transition energy can be calculated using Eq.(20) and calculated radii of metal and TFB molecule using Eq.(21) with molecule weight and mass density from Table1. Results are shown in Table 1.

#### Table 1. The main properties Au and TFB molecule dye [16, 17].

| Properties                  | Gold (Au) [16] | TFB [17] |
|-----------------------------|----------------|----------|
| Element Formula             | Au             | C$_{10}$H$_{2}$F$_{12}$ |
| Atomic Metal Weight (g/mol) | 196.966569     | 350.104  |
| Density at room temperature (g/cm$^3$) | 19.30          | 1.567    |
| Crystal system              | Cubic          | Triclinic|
| Structure type              | FCC            | Triclinic|
| HOMO                        | 5.3 eV         |          |
| LUMO                        | 2.3 eV         |          |
| Fermi energy                | 5.53 eV        |          |
| Melting point (°C)          | 1064.18        |          |
| Boiling point (°C)          | 3825           |          |
| heat capacity (Cp) / J g$^{-1}$ K$^{-1}$ at 25 °C, 100 kPa | 0.129 |          |
| Ionization energy           | 5.5 eV         |          |
| Radius calculated(Å)        | 1.5934         | 4.4574   |

The transition energy of Au/TFB interface is strictly by dielectric constant and refractive index of solvents and radi of Au and TFB molecule, it is evaluated by Eq.(20) using characteristic of solvents from Table 2 and, $R$ is the distance between the Au electrode and TFB molecule $R = r_{Au} + r_{TFB}$ and taken the radius of Au and the TFB molecule from Table 1 and $\frac{e^2}{8\pi \varepsilon_0} = 14.4 eV$, results are listed in Table 2.
Table 2. Calculated results of transition energy $\Lambda_{mM}(eV)$ for Au/TFP

| Solvent                  | Chemical Formula | Refractive index (n) at 20° | Static dielectric constant ($\varepsilon$) at 20° | The transition energy (eV) Au/TFP |
|--------------------------|------------------|----------------------------|-----------------------------------------------|----------------------------------|
| 2,2,2-Trifluoroethanol   | C$_2$H$_3$F$_3$O | 1.2910                     | 8.55                                          | 0.4928                           |
| Methanol                 | CH$_4$O          | 1.3284                     | 32.7                                          | 0.5470                           |
| Acetonitrile             | C$_2$H$_3$N      | 1.3441                     | 37.5                                          | 0.5375                           |
| Diethyl ether            | C$_4$H$_9$O      | 1.3524                     | 4.33                                          | 0.3222                           |
| Acetone                  | C$_2$H$_6$O      | 1.3587                     | 21.01                                         | 0.5041                           |
| Ethanol                  | C$_2$H$_6$O      | 1.3614                     | 24.5                                          | 0.5088                           |
| Nitromethane             | CH$_3$NO$_2$     | 1.3817                     | 35.87                                         | 0.5060                           |
| Tetrahydrofuran(THF)     | C$_4$H$_8$O      | 1.4072                     | 7.58                                          | 0.3806                           |

Furthermore, the Au metal and TFB molecule contact, the Fermi energy of Au and ionization energy (IE) making the interface potential barrier. It evaluates using Eq.(12) by taking the transition energy in Table 2 and ionization energy of TFB is 5.5 eV and Fermi energy of Au metal is 7.25 eV, results have been shown in Table 3 knowing that highest orbitals molecule energy (HOMO) energy levels is 5.3 eV and the lowest orbitals molecule energy (LUMO) is 2.3 eV [18].

Table 3. Results of evaluation the potential $\Delta V_{ef}$ for Au/TFB system

| Solvent type       | Polarity function f(n,e) | Transition energy $\Lambda_{mM}(eV)$ | Potential energy $\Delta V_{ef}$ (eV) |
|--------------------|--------------------------|-------------------------------------|---------------------------------------|
| 2,2,2-Trifluoroethanol | 0.4830                   | 0.4928                              | 0.1087                                |
| Methanol           | 0.5361                   | 0.5470                              | 0.1222                                |
| Acetonitrile       | 0.5269                   | 0.5375                              | 0.1198                                |
| Diethyl ether      | 0.3158                   | 0.3222                              | 0.0663                                |
| Acetone            | 0.4941                   | 0.5041                              | 0.1115                                |
| Ethanol            | 0.4987                   | 0.5088                              | 0.1127                                |
| Nitromethane       | 0.4959                   | 0.5060                              | 0.1119                                |
| Tetrahydrofuran(THF)| 0.3731                   | 0.3806                              | 0.0808                                |

The energetics coupling $\langle \mathfrak{M}_{mM} \rangle (eV)$ of Au metal-on-TFB contacts are estimated experimentally depending on literature [19] and approximately to $\langle \mathfrak{M}_{mM} \rangle^2 = (0.42 \ 0.4 \ 0.35 \ 0.5 \ 0.56 \ 0.6 \ 0.65 \ 0.7 \ 0.75 \ 0.8 \ 0.9) \times 10^{-11} eV^2$. Flow of electronic transfer rate has evaluated using Eq.(19) with MATLAB program and using results of transition energy in Table 2, Fermi energy of Au is 7.25 eV and ionization energy IE= 5.5 eV, results are showing in Table 4.
Table 4. Results of flow rate of electrons transfer calculation for Au/TFB at Ionization Energy IE= 5.5 eV

| Solvent type            | Coupling strength $|\langle \mathbf{m}_\text{mol} \rangle |^2 \times 10^{-11}$ eV/ state |
|-------------------------|---------------------|
|                         | 0.42 | 0.4  | 0.35 | 0.5  | 0.56 | 0.6  | 0.65 | 0.7  | 0.75 | 0.8  | 0.9  |
| 2,2,2-Trifluoroethanol  | 1.487 | 1.417 | 1.239 | 1.771 | 1.983 | 2.125 | 2.302 | 2.479 | 2.656 | 2.833 | 3.187 |
| Methanol                | 0.842 | 0.802 | 0.702 | 1.002 | 1.123 | 1.203 | 1.303 | 1.403 | 1.503 | 1.604 | 1.804 |
| Acetonitrile            | 0.910 | 0.866 | 0.758 | 1.083 | 1.213 | 1.300 | 1.408 | 1.516 | 1.625 | 1.733 | 1.949 |
| Diethyl ether           | 9.440 | 8.991 | 7.867 | 11.238 | 12.587 | 13.486 | 14.610 | 15.734 | 16.857 | 17.981 | 20.229 |
| Acetone                 | 1.314 | 1.252 | 1.095 | 1.565 | 1.753 | 1.878 | 2.034 | 2.191 | 2.347 | 2.504 | 2.817 |
| Ethanol                 | 1.246 | 1.187 | 1.038 | 1.484 | 1.662 | 1.780 | 1.929 | 2.077 | 2.225 | 2.374 | 2.670 |
| Nitromethane            | 1.281 | 1.220 | 1.067 | 1.525 | 1.708 | 1.830 | 1.982 | 2.135 | 2.287 | 2.440 | 2.745 |
| Tetrahydrofuran(THF)    | 4.959 | 4.722 | 4.132 | 5.903 | 6.611 | 7.084 | 7.674 | 8.264 | 8.854 | 9.445 | 10.625 |

4. Discussion

Quantum theory model used to examine the flow electron transfer rate at Au/TFB interfaces, and evaluate the flow rate, transition energy and potential barrier at interface. The energy levels alignment of Au metal contact with TFB are showing in Figure1, i.e. the interface of Au metal energy level alignment with HOMO-LUMO energy levels for TFB molecule strictly by the Fermi energy of Au and the HOMO-LUMO energy. The positions of the energy levels of TFB molecule relative to the Fermi level of Au are shown in Figure1. The transition energy controlled electronic transfer of Au metal contact on TFB molecule. In the majority of transition energy investigated here, the polarity estimation using the refractive index and dielectric constant in Table 3 effected forcedly on calculation the transition energy with different solvent. At the Au metal is aligned with TFB molecule, the transition energy increased with increased polarity of solvents with the fact that solvents generally form good media to level alignment with each other. Considering the fact that the transition energy are controlled to transfer of electrons from donor to acceptor we conclude that transition energy relative to the position of the energy level of highest orbitals molecule energy (HOMO), and the lowest orbitals molecule energy (LUMO) relative to Fermi energy are limited the flow electronic rate through the potential that created in interface of contact Au with TFB. The potential barrier, which simply defined as the energy difference between the ionization energy IE and Fermi level relative to transition energy. When the Fermi energy of the Au metal is relatively to ionization energy and transition energy low, making the interface potential barrier larger, it's shown that for Au/TFB with Methanol solvent has potential is equal to 0.1222 eV compare with Diethyl ether is equal to 0.0663 eV. However, when the transition energy is large lead to potential is high, the increased transition energy making the interfacial potential barrier higher because the increased transition energy indicate the driving energy decreased because the system has more energy to reorientation energy, or small flow electronic rate. The electronic transition are most occurs when potential is low this showing for Au/TFB with Diethyl ether solvent (9.440×10−9 to 20.229×10−9 cm3/sec) compare with less electron transfer with methanol solvent (0.842×10−9 to 1.804×10−9 cm3/sec). The transition energy of Au metal contact on TFB molecule interfaces is energy necessarily to transfer occurs with the energy level alignments at interfaces. Different solvent media are taken to examined the transfer of electrons at Au/TFB system. While methanol solvent media shows slow flow rate, Diethyl ether and Tetrahydrofuran (THF) solvents show strong electronic transfer from Auto TFB molecule, with low potential (0.0663 eV and 0.0808 eV) and low transition energy (0.3222 eV and 0.3806 eV) relatively to system with methanol has low flow electronic rate with large potential 0.1222 eV and large transition energy (0.5470 eV) for Au contact on TFB molecule.
5. Conclusion

We have determined the flow electronic transfer rate based quantum transition theory for interface of Au contact with TFB with different solvents have polarity ranging from 0.3158 to 0.5361. Most importantly effects on flow electronic transfer is the transition energy, that’s limited the produce electron density corresponding energies of electrons transfer. When increased the transition energy the flow rate increased electrons to transfer from donor to acceptor. In additional effect on increasing flow electronic transfer in the Au/TFB when the potential barrier is small (0.066 eV). The flow rate of electronic transfer decrease with increasing the potential energy and decreased the electronic rate when increased the potential energy at interface. The Diethyl ether is topic media with Au contact with TFB molecule found to yielding large flow electronic rate and low potential barrier than the bottom Methanol solvent founded to yielding small flow electronic rate and high potential barrier at interface.

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