Theoretical Calculation of Rate Constant of Charge Transport Processes in Au Metal/Semiconductor Interface System

الحسابات النظرية لمعدل انتقال الشحنة في سطح نظام معدن الذهب / شبه الموصل

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Abstract

The rates constant of the charge transfer in the interface between gold(Au) metal and ZnO, ZnS, and ZnSe semiconductor systems have been calculated with the reorientation free energy a according to a theoretical model which has been derived according quantum theory in terms of physical parameters. We present a quantum model and a Hush theory for reorientation energy to study of electron transfer across gold. The rate of charge transfer increase with increase the reorientation energy. The rate constant of electron transfer K_{et} depend on the barrier height of Au/ZnO, Au/ZnS, and Au/ZnSe system, this barrier formed during contact and depends on the work function and affinity of semiconductors.

Results of the rate constant of electron transfer K_{et} depends on reorientation energy λ, work function of metal Φ_m, affinity of semiconductors χ , crystal structure of semiconductors properties of metal and the coupling matrix element between metal and semiconductors.

Key Words: Charge transport processes, Au/ZnO, Au/ZnS, and Au/ZnSe Metal/semiconductor interface.

الخلاصة

ثابت معدل ألانتقال للشحنة عند السطح مابين نظام معدن الذهب وشبه الموصل(ZnO, ZnS, ZnSe) حسب مع الطاقة الحرة لإعادة الترتيب وفقا لأنموذج نظري اشتق وفقا للنظرية الكمية وبحدود البارامترات الفيزيائية. وحسب الأنموذج الكمي ونظرية هاش لطاقة إعادة الترتيب لدراسة الانتقال الالكتروني عبر سطح معدن الواهب / شبه الموصل وجد ان الانتقال الالكتروني يتزايد بتزايد طاقة إعادة الترتيب، حيث أن معدل الانتقال الالكتروني يعتمد ارتفاع حاجز الجهد لنظام Au/ZnO, Au/ZnS, Au/ ZnSe .

يتشكل هذا الجهد عند الاتصال ويعتمد على دالة الشغل للمعدن والألفة الالكترونية لشنه الموصل. نتائج معدل الانتقال الالكتروني تعتمد على طاقة إعادة الترتيب λ ودال الشغل للمعدن Φ_m والألفة الالكترونية لشبه الموصل χ والتركيب البلوري لشبه الموصل وصفات المعدن ومصفوفة العناصر المترابطة بين المعدن وشبه الموصل.

Introduction

Electron transfer (ET) reactions represent an elementary process which occurs in a large variety of molecules, ranging from small ions pairs up to large biological systems, Such charge transfer reaction are of vital importance to a variety processes in physical chemistry and biology [1]. This reaction occurs where an electron is transferred from an electron donor |D> to an electron acceptor |A>. The field of electron transfer has a rich history and continues to pose interesting problems for today's researchers. Electron transfer theory predicts reorientation energy to be one factor that determines the electrochemical potential of a metal[2].

Metal/ semiconductor contact from interfaces that give basic features of many metal –semiconductor devices. To construct the diagram of metal/ semiconductor contact, we consider the energy band diagram of metal /semiconductor, and align them using the same level [3] where a metal is placed in intimate contact with a semiconductor, the electrons from the conduction band in one material, which have higher energy, flow into the other material until the Fermi level on the two sides are brought into coincidence [4-5]. The energy level in the two materials are rearranged relative to the new common Fermi level [6]. However, the Fermi energy of the metal and semiconductor do not change right away . This yields the flat diagram [7]. The case of behavior of the contact between metal and semiconductor is illustrated in Figure(1) [8].

In this paper we have been calculated the rate constant and the reorientation energy of electron transfer in Au metal/semiconductor interface using a theoretical model derived for this system according on quantum model and a continuum Marcus-Hush theory. This model of electron transfer applied theoretically on Au/Zno, Au/ZnS and Au/ZnSe.

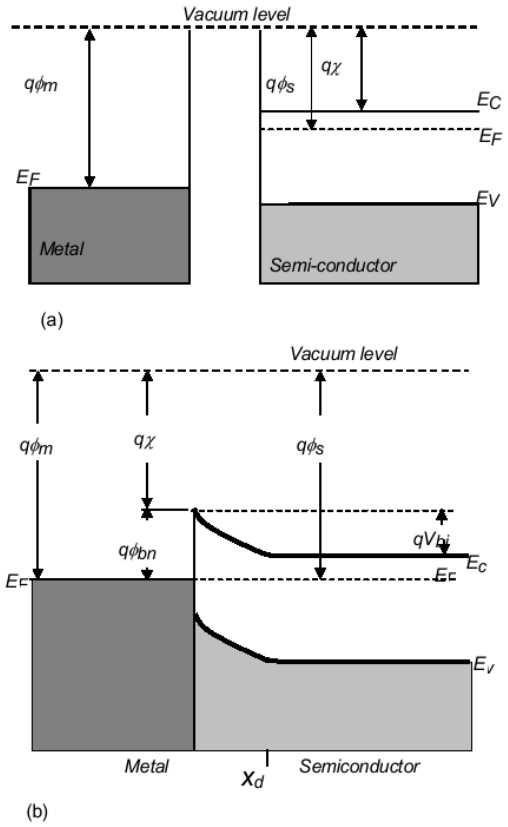

Figure (1):Metal/ semiconductor/interfaces (a) without contact, (b) with contact[8].

The nature of the contact depends on the relative height of the Fermi levels φ_m and φ_s [8].

Theory

A quantum mechanical model with first order perturbation theory for a transition from a donor state to accepter state used to derived expression for the rate electron transfer. A quantum system is described by the wave function [9].

$$\Psi(r,t) = \sum_n A_n \, e^{-\frac{iE_n t}{\hbar}} \Psi\Psi_n(r) = \sum_n A_n(t) \, |\Phi\Phi(t) > \quad \ldots\ldots\ldots\ldots \quad (1)$$

Where ħ is the Plank constant, model Hamiltonians operator used to describe the electron transfer in donor –acceptor system can be written as [10].

$$H = H_{op} + \mathrm{V} \qquad \ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

The rate of the transition probability per unit time is the rate constant of electron transfer i.e. [11]

$$K_{et} = \frac{d}{dt}P_n(t) = \frac{2\pi}{\hbar} \mid V_{DA}|^2 \rho(E) \qquad \ldots\ldots\ldots\ldots\ldots\ldots (3)$$

Where V_{DA} is the coupling coefficient and $\rho(E)$ is the density of states Such that [12]

$$\rho(E) = (4\pi\lambda k_B \mathrm{T})^{\frac{-1}{2}} \exp\frac{-(\lambda + \Delta \mathrm{G}_\circ)^2}{4\lambda \mathrm{k_B T}} \qquad \ldots\ldots\ldots\ldots\ldots\ldots (4)$$

Where λ is the reorientation energy for system and $\Delta G_\circ$ is effect free energy by substitute Eq.(4) in Eq.(3), then

$$K_{et} = \frac{2\pi}{\hbar} \mid V_{DA}|^2 (4\pi\lambda KT)^{\frac{-1}{2}} exp\frac{-(\lambda + \Delta G_\circ)^2}{4\lambda KT} \qquad \ldots\ldots\ldots\ldots\ldots\ldots (5)$$

The total electron transfer rate from semiconductor |D> state to the metal |A> state can be expressed as the sum of electron transfer rates to all possible accepting state in the metal which is given by [12]

$$K_{\mathrm{ET}} = \frac{2\pi}{\hbar}\int_{-\infty}^{\infty} \mathrm{f(E, E_F)}|\mathrm{V_{(k,r)}}|^2 (4\pi\lambda \mathrm{K_B T})^{\frac{-1}{2}} \exp\frac{-(\lambda + \Delta \mathrm{G_o})^2}{4\lambda \mathrm{k_B T}}\, \mathrm{dE} \qquad \ldots\ldots\ldots\ldots (6)$$

Where $\mathrm{V_{(k,r)}}$ is the electronic coupling coefficient between the donor state and accepter state and $f(E, E_F)$ is the Fermi- Dirac function related to electron by [13]

$$f(\mathrm{E, E_F}) = \frac{1}{1 + \exp\frac{(\mathrm{E_{CB} - E_F})}{\mathrm{K_B T}}} \qquad \ldots\ldots\ldots\ldots\ldots\ldots (7)$$

where E_{CB} is the conduction band energy and E_F is the Fermi level energy. Then Eq.(3) become

$$K_{\mathrm{ET}} = \frac{2\pi}{\hbar}\int_{-\infty}^{\infty} \frac{1}{1+\exp\frac{(\mathrm{E_{CB}-E_F})}{\mathrm{K_B T}}} \mid \mathrm{V_{(k,r)}}|^2 (4\pi\lambda \mathrm{KT})^{\frac{-1}{2}} \exp\frac{-(\lambda+\Delta \mathrm{G})^2}{4\pi \mathrm{K_B T}}\, \mathrm{dE} \ldots . (8)$$

The free energy ΔG related to occupied energy E by [14]

$$\Delta G = \Delta G_\circ - E \qquad \ldots\ldots\ldots\ldots\ldots\ldots (9)$$

Where $\Delta G_\circ$ is the standard free energy of reaction.

Substituting Eq.(9) in Eq.(8) then

$$K_{ET} = \frac{2\pi}{\hbar}\int_{-\infty}^{\infty}\frac{1}{1+\exp\frac{(E_{CB}-E_F)}{K_BT}}|V_{(k,r)}|^2(4\pi\lambda KT)^{\frac{-1}{2}}\exp\frac{-(\lambda+\Delta G_\circ - E)^2}{4\pi K_BT}dE \quad \ldots.(10)$$

The activation energy $\Delta G^{++}$is the height of the barrier of ideal potential contact between metal and semiconductor in the absence of surface state is equal the difference between the metal work function Φ_m and the electron affinity χ of the semiconductor and formulated by[15]

$$\Delta G^{++} = \frac{\left(e\Phi_m - e\chi\right)^2}{4\lambda K_B T} \quad \ldots\ldots\ldots\ldots\ldots\ldots(11)$$

The driving force free energy is given by[16]

$$\Delta G_0 = E_{CB} - E_{F,m} \quad \ldots\ldots\ldots\ldots\ldots\ldots(12)$$

Inserting Eq.(11) and Eq.(12) in Eq.(10) we gate

$$K_{et} = \frac{2\pi}{\hbar}(4\pi\lambda K_BT)^{\frac{-1}{2}}e\frac{-\left(e\Phi_m - e\chi\right)^2}{4\lambda K_BT}\int_0^{\infty}[1 + \exp\frac{\left(E_{CB} - eV_{app} - E_F^\circ\right)}{K_BT}]^{-1}|V(k,r)|^2 e\frac{-E^2}{4\lambda K_BT}\,dE \quad \ldots(13)$$

The Fermi-Dirac function in Eq.(13) reduced to the Boltzmann equation when $E_{CB} - E_F >> K_BT$ [17]

$$f(E,E_F)\,(E_{CB} - E_F \gg K_BT) \approx \exp\frac{-(E_{CB}-E_F)}{K_BT} \quad \ldots.\ldots.\ldots\ldots(14)$$

The square of the electronic coupling matrix element integrated over the distribution of the electronic state at the given E is [18]

$$H_{DA}(k,r)^2 = \frac{|V(k,r)|^2}{2\pi\delta(E_K - E)} \quad \ldots\ldots\ldots\ldots(15)$$

The metal density of state $\rho(E)$ can be defined as [18].

$$\rho(E) = \sum 2\pi\delta(E_K - E) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots(16)$$

After mathematical simplified the rate constant of electron transfer K_{et} depend exponentially on distance with a decay constant β,the K_{et} becomes [19].

$$K_{et} = \frac{1}{\beta}K_{et}\,(contact) \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots(17)$$

$$K_{et} = \frac{2\pi}{\hbar}(4\pi\lambda k_b T)^{\frac{-1}{2}} \frac{n_{eq}V}{\beta} \exp\frac{-(e\Phi_m - e\chi)^2}{4\lambda K_B T} \frac{\int_o^\infty \rho(E) f_0(E)|H_{DA}(k,r)|^2 e\frac{-E^2}{K_B T}\, dE}{\int_o^\infty \rho(E) f_0(E) dE} \quad \ldots.(18)$$

Where the volume of unit cell given by V=a.b×c**.**

The average coupling electronic coefficient of matrix element square is given by [20]

$$|\ H(E,r)|^2 = \frac{\int_0^\infty \rho(E) f_0(E)|H_{DA}(k,r)|^2 dE}{\int \rho(E) f_0(E) dE} \quad \ldots\ldots\ldots\ldots\ldots\ldots(19)$$

Substituted Eq.(19) in Eq.(18), we get

$$K_{et} = \frac{2\pi}{\hbar}(4\pi\lambda k_B T)^{\frac{-1}{2}} \exp\frac{-(e\Phi_m - e\chi)^2}{4\lambda K_B T} \frac{V}{\beta} |H(E,r)|^2 \quad \ldots\ldots\ldots(20)$$

Where $\hbar$ is the planck constant, Φ_m is the work function, χ is the affinity of semiconductor, K_B Boltzman constant, V the volume unit cell of semiconductor, T absolute temperature, $H(E,r)$ is the coupling matrix element and λ reorientation energy that given by

$$\boldsymbol{\lambda_\circ} = \frac{(\Delta e)^2}{2a_s}\left(\frac{1}{n_s^2} - \frac{1}{\varepsilon_s}\right) + \frac{(\Delta e)^2}{2a_m}\left(\frac{1}{n_m^2} - \frac{1}{\varepsilon_m}\right) - \frac{(\Delta e)^2}{4d_s}\left(\frac{n_m^2 - n_s^3}{n_m^2 + n_s^2}\ \frac{1}{n_s^2} - \frac{\varepsilon_m - \varepsilon_s}{\varepsilon_m + \varepsilon_s}\ \frac{1}{\varepsilon_s}\right) - \frac{(\Delta e)^2}{4d_m}\left(\frac{n_{s-n_m}^2}{n_s^2 + n_m^2}\ \frac{1}{n_m^2} - \frac{\varepsilon_s - \varepsilon_m}{\varepsilon_s + \varepsilon_m}\ \frac{1}{\varepsilon_m}\right) - \frac{2(\Delta e)^2}{R_{m_s}}\left(\frac{1}{n_s^2 + n_m^2} - \frac{1}{\varepsilon_s + \varepsilon_m}\right)$$

.....…….(21)

Where ε_s, ε_m, n_s^2, n_m^2, a_s, a_m, d_s,and, d_m are the dielectric constant for semiconductor and metal, refractive index for semiconductor and metal ,distance between metal semiconductor, radii of semiconductor and metal, and the distance for semiconductor and metal to electrode respectively.

Results

The electron transfer in metal/semiconductor have been investigated using a model derived according to a quantum theory and a Hush-Marcus theory of reorientation energy the rate constant of Au/ZnO, Au/ZnS ,and Au/ZnSe. One can find the reorientation free energy that the key factors in charge transfer . We have been estimated the reorientation free energy for Au/ZnO, Au/ZnS, and Au/ZnSe system by used equation (21) with value of refractive index n_{se} and ε_{se}dielectric static constant for semiconductor from table(2) and n_m , $\varepsilon_m$for metals from table(1), with R_m=1.6600A for Gold metal [21] ,R_{se}=3.8025A,5.4100A,and 5.6600A[23]for ZnO,ZnS,and ZnSe semiconductors, d_{se}= R_{se}+1,d_m= R_m+1 ,and R_{se+m}= R_m+R_{se},results are listed in table(3). Another important parameters is the coupling coefficient of

matrix element H(k,ϵ) for metal /semiconductor contact, that's overlap of the wave function of gold metal with k-state of semiconductors are calculated of the bridge and sequential formula of Hush and Marcus[20]. The value of $|H(k,\epsilon)|^2$are calculated for gold Au with typical semiconductors depending on result of typical schachi calculation[20], results are listed in table(4), a parameterization of volume of semiconductor are calculated according V=a.b×c.with lattice parameters of semiconductors from table(2),result the volume is V_{se}=5.49809 x10^{-29} ,1.583402 x10^{-28} m^3 for ZnO,ZnS,and ZnSe semiconductors. The driving force $\Delta G_{\circ}(eV)$ that is provided by the difference between the metal work function and affinity of semiconductor are calculated for three ZnO, ZnS, and ZnSe semiconductors with gold Au metal.Also the activation free energy for system are calculated according the values of ΔG and λ.

Inserting of all data of these parameters into designed a matlap program to calculate the rate constant of electron transfer in metal (Au) contact with ZnO, ZnS, ZnSe semiconductor. The rate constant have been calculated theoretically using eq(20) with values of work function of gold Φ_m , affinity of semiconductor χ ,reorientation free energy λ, volume of semiconductors and the coupling attenuation factors β≅ $0.8, 0.9, 1, 1.1, 1.2\ A^{-1}$. Inserting all these parameters in a matlab program that building to calculate these parameter and rate constant of electron transfer in gold (Au) metal and ZnO, ZnS, ZnSe semiconductors. Results of rate electron transfer are listed in tables(5),(6) and (7).

Table (1): Properties of Gold(Au) metals

Properties	Gold(Au) metal
Atomic weigth	196.97[21]
Atomic volume(cm^3/mol)	10.2[21]
Atomic radius(pm)	144[21]
Refractive index	1.658[22]
Crystal structure	Cubic face centered
Lattice constant(A^0)	4.080 [21]
Electron work function(ev)	5.1[21]
Density(g/cm^3)	19.32[21]
Fermi energy(eV)	7.32[21]

Table (2): Properties of semiconductors

semiconductor / Properties	ZnO	ZnS	ZnSe
Crystal structure	Wutzite[23]	Zinc blend[23]	Zinc blend[23]
Lattice constant(nm)	a=0.32495,c=o.5206[23]	0.541 [23]	o.566[23]
Density(g/cm^3)	5.66 [23]	4.08 [23]	5.42[23]
Dielectric constant	8.5 [23]	8.3 [23]	9.2[23]
Refractive index	2.00337[22]	2.52226[22]	2.62408[22]
Energy gab(eV)	3.4[23]	3.6 [23]	2.6[23]
Electron affinity(ev)	4.5[24]	3.9[24]	4.09[25]

Table (3): The reorientation energy for gold metal/semiconductor interface system

System	Reorganization energy λ(ev)			
	d=a+1.0	d=a+1.2	d=a+1.4	d=a+1.6
Au-ZnO	o.71502218	o.71900496	0.722411668	o.725354107
Au-ZnS	o.670556312	o.682074705	o.69202476	0.700700169
Au-ZnSe	o.673663776	O.685728201	o.696178713	o.705299904

Table (4): The Coupling matrix element $|H(k,\epsilon)|^2$ for gold(Au)

$\|V(\epsilon)\|^2$x10^{-11}	$\|H(k,\epsilon)\|^2$x10^{16} $\frac{(ev)^2}{m^3}$	E(ev)
0.4	5.889514591	-1.35
0.45	6.625703913	-0.90
0.5	7.361893239	-0.25
0.55	8.098082563	0
0.6	8.834271886	0.15
0.65	9.57046121	0.3
0.7	10.30665053	0.5
0.75	11.04283986	0.75
0.8	11.77902918	1

Discussion

Results of the reorientation free energy λ (eV) in table(3) indicate that when the semiconductor is not contact with metal leading large value of λ (eV) for all system. On the other hand this indicate are illatively week coupling when the distance d_m and

d_{se} increasing with R_m and R_{se} , this because of the barrier height is large in system with increasing of d. The results of λ is large for Au/ZnO system compare with Au/ZnS and Au/ZnSe , this leading to suggest that Au/ZnO possess a good matching system, also the ZnO have optical index smaller than ZnS and ZnSe. The structure is wutalazite is difference than ZnS and ZnSe have the Zinc blend structure [23]. One way to increase the rate constant of electron transfer , λ could be increase this showing when comparing between table (3) and (4) for λ and K_{et} for three system next, as for the metal / semiconductor interface a rearrangement of the conduction electron occurs at the interface .Here ,however some electrons flow from the semiconductor to the metal because $\Phi_m \cong 5.1$ (eV) for Au is large than $\chi \cong 4.5, 3.9, 4.09\ (eV)$ for ZnO,ZnS and ZnSe respectively ,this indicate the average energy of electrons in the semiconductor is greater than the average of those in metal. This means that when brought the metal to contact with semiconductor the difference in the average electron energy will transfer from semiconductor to the metal until the Fermi level coincide. Also when calculation free energy of driving force $\Delta G^{\circ} = \Phi_m - \chi$ is equal ΔG° (ZnO/Au)$\cong 0.6\ (eV)$, ΔG° (ZnSe/Au)$\cong 1.01\ (eV)$ and ΔG°(ZnS/Au)$\cong 1.2\ (eV)$, refers to large K_{et} for ZnO/Au system because the free energy charge associated with moving the electron from donating species to the acceptor species.

The barrier height is an important parameter that depending on the electrical characteristic of Au/ZnO,Au/ZnS,and Au/ZnSe contact system. Near metal / semiconductor due to energy level difference , the barrier is formed with main characteristic of two metal semiconductor material work function , affinity give the effect of electric field near surface, space charge region, velocity of charge, and reorientation free energy .It was found in our calculation that K_{et} is proportional to the barrier height value ΔG^{++}(ZnO/Au($\cong 0.1258\ (eV)$, ΔG^{++}(ZnSe/Au)$0.3785\ (eV)$, and ΔG^{++}(ZnS/Au)$\cong 0.5368\ (eV)$. However when energy of the tunneling electrons exceeds the barrier height which small for ZnO/Au$\cong 0.125$, the electrons are injected from semiconductor to metal is large than Au/ZnS, and Au/ZnSe system. This lead to large constant of electron transfer for Au/ZnO compared with other system. Finally we can find from results when raising the values of β leading to decreasing the rate of electron attenuation parameters β, for all system. The rate of electron transfer in tables (5),(6)and(7) for Au/ZnO, Au/ZnS,and Au/ZnSe show the rate constant of electron transfer increasing with decreasing barrier height and increasing the reorientation energy. The rate constant of electron transfer increasing with increase the driving free energy.

Conclusion

Al though the electron transfer model is heigh approximation it does provide a reasonable description for the varying in rate constant for metal / semiconductor system.

The present model show that the system have large rate constant when the reorientation free energy is large and vice versa. We concluded the rate constant for electron transfer K_{et} for metal / semiconductor system strongly depends on reorientation energy λ and type of semiconductor. Rate of electron transfer K_{et} is large for Au/ZnO have small value of driving force compare with Au/ZnS and Au/ZnSe. The rate constant K_{et} is inversely proportional to the barrier height. For more large barrier (0.53 eV) for Au/ZnS, the rate K_{et} are small and value for height barrier (0.125 eV) for Au/ZnO system lead to small electron transfer reaction. It can be concluded the reaction of electron transfer strongly depends on value of reorientation energy, driving free energy, barrier height. In summary we concluded the Au/ZnO system is move active media for electron transfer and possess a good matching compare with Au/ZnS and Au/ZnSe system.

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