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Ion – Surface Scattering Processes in the Presence of Electromagnetic Field: The Concept of Temporal Effective Density of States

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Abstract

The interaction dynamics in the scattering process between surface and species were studied earlier using the one-electron Hamiltonian in the presence of a (monochromatic) electromagnetic field. This work defines a new concept, "the temporal effective density of states". This quantity is essential in obtaining the ion neutralization probability in the ion-surface scattering and is equivalent to the time average of the transient energy-dependent current through the system. Our applications ensure that this kind of the tunneling current can be controlled by the electromagnetic field parameters such as its frequency and its strength. Then, adiabatic and non-adiabatic electron tunneling regimes are investigated and discussed for the process. Also, in the limit of very low ion velocity one can find, an equivalent behavior for the temporal effective density of states.

Keywords: Electromagnetic field, Charge exchange process, and Scattering process.

عمليات الاستطارة لنظام ايون – سطح بوجود مجال كهرومغناطيسي : مفهوم كثافة الحالات المؤثرة زمنياً

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الخلاصة

تمت دراسة ديناميكيات التفاعل في عملية الاستطارة بين السطح والجسيمات في وقت سابق باستخدام هاملتونين – إلكترون واحد بوجود مجال كهرومغناطيسي (أحادي الطول الموجي). في هذا العمل نعرف مفهوم جديد هو " كثافة الحالات المؤثرة زمنيا ". هذه الكمية ضرورية في توضيح احتمالية تعادل الايون في استطارة ايون – سطح وتكافئ المعدل الزمني للتيار العابر المعتمد على الطاقة عبر النظام. اكدت تطبيقاتنا أن هذا النوع من تيار النفق يمكن التحكم به باستخدام معاملات المجال الكهرومغناطيسي مثل تردده وقوته. بعد ذلك تم التحقق ومناقشة مناطق نفق الإلكترون الاديباتيكية وغير الاديباتيكية لهذه العملية. أيضا في حدود سرعة الأيون الواطئة جدا يمكن أن نجد سلوكا مكافئا لكثافة الحالات المؤثرة زمنيا.

1. Introduction

Processes in the gas-phase metal surface that are assisted and supported by electromagnetic magnetic fields prove essential to understanding surface physics. In this field, Chemical reaction dynamics in gas-phase reactions and those that happen at the gas-solid interface have started to be dominated by chemists. The electromagnetic field had been used to study processes state - selected reactions, transition - state spectroscopy.

The electromagnetic field has many controllable parameters: propagation vector, polarization, intensity, and frequency. By controlling these parameters as well as the

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consistency of the radiation. Researchers use a supple stimulator to check the chemical dynamics [1, 2]. Much research has been done on how electromagnetic fields interact with matter in uniform systems [3-6]. However, The laser-induced surface phenomena, including dissociation, adsorption, reactions, and migration, for heterogeneous systems (such as atoms adsorbed on a solid surface or gaseous atoms near it), have been thoroughly researched and explored for various systems [7, 8]. (For more information, check references in [9-13].

The orthonormal basis set for electron states in atoms and metals was extended in terms of the total wave function, which satisfied the Schrodinger equation dependent on time. For the amplitudes of these processes, a system of interconnected ordinary equations was derived. Surface processes that are aided by an electromagnetic fields should occur at field intensities similar to those in related atomic collision reactions (~ 1016 W/m2).

There are two regimes of coupling between the external field and the electrons [14-17]. The first one is low frequency regime, in the adiabatic regime, where the tunneling rate $\Gamma/\hbar$ dominates the transport time and the external field frequency ω is smaller than the tunneling rate $\Gamma/\hbar$, electrons sense a static external field, i.e., the external field dependent on time only affected the electronic states adiabatically. The external field frequency is higher than the tunneling rate in the second, high frequency regime, i.e. $\omega >> \Gamma/\hbar$. Since energy quanta ($\hbar\omega$) can be absorbed or emitted by electrons during the non-adiabatic regime, which produces direct tunnel current, electrons can sense the oscillation of the external field during the transport time. The case that we have found in our previous works[9, 10].

Our aim is to use the earlier develop theoretical treatment [9, 10] related to the interaction of the monochromatic electromagnetic field with the atom-surface scattering process to define a new concept, which we call "the temporal effective density of states" and to discuss the adiabatic and non-adiabatic electron tunneling regimes for the process.

2. Formalism

One electron resonant process in collisions of slow atoms (ions) with a metal surface in the presence of electromagnetic field had been considered in Ref. [18]. The system's Hamiltonian (atom + surface + laser field) was expressed as follows:

$$\hat{H}(t) = \hat{H}_o + \hat{V}(t) \tag{1}$$

where, both the atom-metal interaction and the interaction with the electromagnetic field are included in operator $\hat{V}(t)$. The electromagnetic field effect on the ion-surface neutralization (SIN) mechanism during the ion-surface scattering process was studied [19-26] by extending the one-electron formalism developed by Easa and Modinos [22, 27] to enclose an additive term in the system Hamiltonian that required to describe the electromagnetic field- assisted charge transfer processes.

At time (t), the ion neutralization probability is given by [9, 10, 22, 28],

$$n_A(t) = \sum_{\mu'(occ.)} \left| C_{A\mu'}(t) \right|^2 \tag{2}$$

All occupied surface states are included in the summation., and the equation of motion of the amplitude is given by [9, 10],

$$\dot{C}_{A\mu'}(t) = -\frac{i}{\hbar}\varepsilon_{im}(t)C_{A\mu'}(t) - \frac{i}{\hbar}\left[\tilde{V}_{A\mu'}(t) + \tilde{W}_{A\mu'}(t)\right] - \frac{1}{\hbar^2}\sum_{\mu}\left[\tilde{V}_{A\mu'}(t) + \tilde{W}_{A\mu'}(t)\right]\int_{t_\circ}^{t}\left[\tilde{V}_{\mu A}(t') + \tilde{W}_{\mu A}(t')\right]C_{A\mu'}(t')dt' \quad (3)$$

Then, we define the following relations [9, 10, 20],

$$\begin{aligned} C_{A\mu'}(t) &= V_\mu C(\varepsilon, t) \\ \tilde{V}_{A\mu}(t) + \tilde{W}_{A\mu}(t) &= V\mu e^{-i(\varepsilon_\mu - \varepsilon_A^\infty)t} H(t) \\ H(t) &= V(t) + W(t) \\ \rho_s(\varepsilon) &= \sum_{\mu}\left|V_\mu\right|^2 \delta(\varepsilon_\mu - \varepsilon) \end{aligned} \quad (4)$$

where, $\rho_s(\varepsilon)$ represents the surface density of states. Then, Eqs. (2) and (3) become,

$$\dot{C}(\varepsilon', t) = -\frac{i}{\hbar}\varepsilon_{im}(t)C(\varepsilon', t) - \frac{i}{\hbar}e^{-i(\varepsilon' - \varepsilon_A^\infty)t}H(t) - \frac{1}{\hbar^2}H(t)\int_{t_\circ}^{t} dt' H(t')e^{i\varepsilon_A^\infty(t-t')}\int d\varepsilon\, \rho_s(\varepsilon)\, e^{-i\varepsilon(t-t')}\, C(\varepsilon', t) \quad (5)$$

and,

$$n_A(t) = \int \rho_s(\varepsilon')\left|C(\varepsilon', t)\right|^2 d\varepsilon' \quad (6)$$

If we put $\rho_s(\varepsilon) = \bar{\rho}$ (as constant, $\bar{\rho} = 1/4\beta$), then the definition of the delta function is only the remaining energy integral as $\int e^{-i\varepsilon(t-t')}\, d\varepsilon = 2\pi\delta(t - t')$ [22]. And finally we have,

$$\dot{C}(\varepsilon', t) = \left[-\frac{i}{\hbar}\varepsilon_{im}(t) - \frac{\bar{\rho}}{\hbar^2}\left|H(t)\right|^2\right]C(\varepsilon', t) - \frac{i}{\hbar}H(t)e^{-i(\varepsilon' - \varepsilon_A^\infty)t} \quad (7)$$

A wide-band approximation may be used to describe this approximation. Eq. (7) should be solved with the initial conditions, $C(\varepsilon', t_o) = 0$.

The matrix element of $H(t)$ in Eq. (4) can be read as,

$$H(t) = V(t) + e\varepsilon_0 W(t) \quad (8)$$

The atomic orbital, overlap ultimately only when the ion is close to the surface, and $W(t)$ has the same specific manner as that of $V(t)$. Thus the simplest model for $W(t)$ is analog to [26]:

$$V(t) = V_o\, e^{-\alpha_A u|t|}\ , W(t) = W_o\, e^{-\alpha_A u|t|}\left[e^{-i\omega t - i\delta} + e^{i\omega t + i\delta}\right],\ \alpha_A = \sqrt{2\left|\varepsilon_A^\infty\right|} \quad (9)$$

The laser coupling strengths and hopping are W_o and V_o, respectively. u is the species velocity's An ordinary component.

Our numerical analyses of the ion (survival) probability and investigation of the impact of the electromagnetic field on the electron transfer process in an ion-surface system use Eq. (7).

We offer the idea of the density of states at the effective level site, which is described by $\rho_{eff}(\varepsilon, t)$, to learn more and investigate the possible direction of the transfer of charge between the incident solid surface and the ion.

$$\rho_{eff}(\varepsilon,t) = \rho_s(\varepsilon)\left|C(\varepsilon,t)\right|^2 \tag{10}$$

Such that the ion level occupancy $\mathrm{n_A(t)}$ is given by,

$$P(t) = n_A(t) = \int_{u_o}^{0} \rho_{eff}(\varepsilon,t)d\varepsilon \tag{11}$$

In analogy to the static occupancy $\mathrm{n_A(z)}$ that defined by [22]:

$$n_A(Z) = \int_{u_o}^{0} \rho_A(\varepsilon,Z)d\varepsilon \tag{12}$$

Hence, the probability $\mathrm{P(t)}$ of charge exchange at any time t is calculated from the area under the $\rho_{eff}(\varepsilon,t)$ curve in the energy range from the bottom of the conduction band, $\mathrm{u_o}$, up to Fermi level position $\varepsilon_F = 0$. The amount of this area should depend on the shape of $\rho_{eff}(\varepsilon,t)$ at each time t (time-dependent).

However, the shape of this physical function depends on the interaction matrix elements, namely, the hopping one $\mathrm{V_o}$ and the one related to field strength $\mathrm{W_o}$. But, as it should be, at any time the following condition must be fulfilled,

$$\int_{-\infty}^{\infty} \rho_{eff}(\varepsilon,t)d\varepsilon = 1 \tag{13}$$

During the time of scattering, the non-stationary tunneling current will appear through the system which we define it as:

$$\mathrm{I(t)} = -\frac{\mathrm{dP(t)}}{\mathrm{dt}} \tag{14}$$

And one can find, for the period $\mathrm{t_f - t_o}$ (where $\mathrm{t_f}$ is final scattering time), the average tunneling current value which can be written as:

$$\left\langle \mathrm{I(t_f)} \right\rangle = \frac{1}{\mathrm{t_f - t_o}} \int_{\mathrm{t_o}}^{\mathrm{t_f}} \mathrm{I(t')dt'} = \frac{\mathrm{p(t_f)}}{\mathrm{t_f - t_o}} \tag{15}$$

Hence, one can define on the basis of Eq. (11) the average tunneling current as a function of the energy as,

$$\left\langle I(\varepsilon) \right\rangle = \frac{1}{t_f - t_o} \rho_{eff.}(\varepsilon) \tag{16}$$

then, this energy-dependent tunneling current is equivalent to the effective density of states Eq. (10) without the multiplication factor $\mathrm{1/(t_f - t_o)}$.

It is well-known that if the perturbation is harmonic time-dependent $\left(\propto \mathrm{e}^{\pm \mathrm{i}\omega \mathrm{t}}\right)$, then the system level $\varepsilon_o(t)$ will split to three levels, namely, $\varepsilon_o(t)$ and $\varepsilon_o(t) \pm \hbar\omega$. Here, we also expected that due to this kind of harmonic perturbation $\left(\propto \mathrm{e}^{\pm \mathrm{i}\omega \mathrm{t}}\right)$, which is applied to our system as it is given by the electromagnetic field strength formula Eq.(9), the system levels are generated from $\varepsilon_A(t)$ to obtaining $\varepsilon_A(t) \pm n\hbar\omega$, with n is an integer.

Some of these levels might be positioned above the Fermi level, while others might be positioned below the conduction band (bottom), where they might not actively participate in

the resonance charge exchange process. Therefore, we derive in static regime (velocity $\rightarrow 0$) the energy dependent static neutralization probability which is written as a summation of Lorantzian shape functions as [10],

$$p_{st..}(\varepsilon, Z) = \sum_{n=-2}^{2} g_n \frac{\Delta_c(Z) + 2\Delta_f(Z)}{(\varepsilon - \varepsilon_A(Z) + n\hbar\omega)^2 + (\Delta_c(Z) + 2\Delta_f(Z))^2} \tag{17}$$

with,

$$g_o = \frac{\Delta_C}{\Delta_C + 2\Delta_f}, \qquad g_{n\neq 0} = \frac{\Delta_f}{\Delta_C + 2\Delta_f} \tag{18}$$

The atomic level half width due to tunnel coupling with the metal surface in the presence of electromagnetic field, is $\Delta = \Delta_c + 2\Delta_f = \pi\bar{\rho}[|V_o|^2 + 2|W_o|^2]$ with the tunneling rate $\Gamma = 2\Delta$.

Theoretically, one also can use a appropriate incorporation of two parameters, electromagnetic power that related with W_o and the electromagnetic frequency ω, which could be selected to choose the adiabatic or the non-adiabatic regimes.

In the following, We thoroughly examine the effects of various system variables, such as coupling and hopping strengths, the electromagnetic field frequency and the normal component of the species velocity on the effective level density of states $\rho_{eff}(\varepsilon, t)$ in order to elaborate the way by which they enhance the charge exchange process.

3. The model parameterization

To calculate the ion neutralization probability, Eq. (7) is solved numerically with the initial conditions $C(\varepsilon', t_o) = 0$. Where, $\bar{\rho} = 1/4\beta$ and β is related to the band width $(= 4\beta)$. The atomic unit system is employed throughout the work, and all energies have been determined in relation to the Fermi level. The parameters used are $\beta = 0.2205$ a.u., the surface work function $\phi = 0.1837$ a.u. and $\varepsilon_A^{\infty} = -0.2756\ a.u.$ which is measured with respect to vacuum level.

The image effect is taken into account throughout the following relation [29],

$$\varepsilon_{im} = 1/[4(Z_o + u|t|] \tag{19}$$

Where $Z_o = 4.5353$ a.u. is the screening length.

Generally, the velocity of the projectile ion is taken to be in the range of $0.001\,\text{a.u.} \le u \le 0.01\,\text{a.u.}$ and, ω is fixed on the values 0.112, 0.056 and 0.0028 a.u.

The value of $n_A(\infty) = p(\infty)$ is the main quantity of interest due its experimental importance, so the values of $n_A(\infty)$ are calculated from Eq. (7) to investigate the adiabatic and non-adiabatic features of the charge transfer process in the presence of electromagnetic radiation as in the following.

Initially, the atomic energy level position ε_A^{∞} is one of the most important parameter in determining the direction of the charge transfer process in the absence of electromagnetic field. By varying ε_A^{∞} we have sweep the energy values for met stable state which are separated by $\hbar\omega$, see Figure 1. There is two plateau, the left corresponds to higher occupation $n_A(\infty) \approx 1$ and the right corresponds to lower occupation $n_A(\infty) \approx 0$, to the left and to the right of the

position of the atomic energy level $\varepsilon_A(Z_o)$. With the action of electromagnetic field, the left plateau is lying below nearly by the same amount that the right one is raised, with the same crossing points, indicating the atomic level sense to the electromagnetic field.

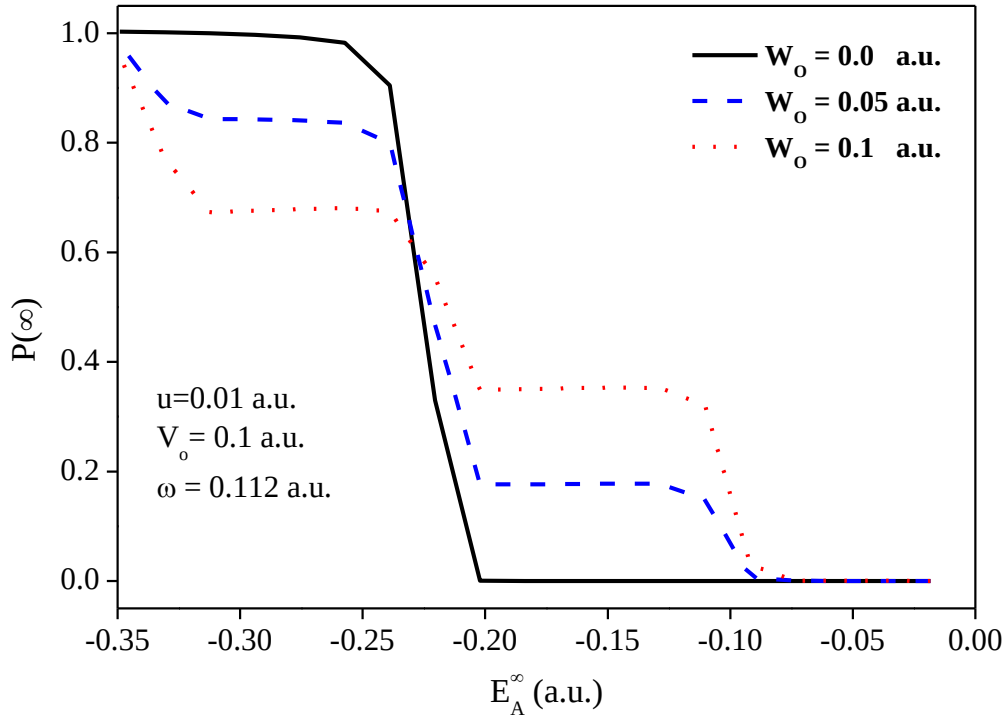

Figure 1. Shows the values of P(∞) as a function of ε_A^{∞} for three different values of $W_o = 0.0, 0.05, 0.1$ a.u. with u = 0.01 a. u. and $V_o = 0.1$a.u..

In the following, We thoroughly investigate the impacts of various system parameters, such as V_o, W_o (or consequently $\Gamma = 2\Delta$), and ω on the $\rho_{eff}(\varepsilon,\infty)$ to investigate the adiabatic and non-adiabatic regimes by which the tunneling process occurs.

Firstly, $\rho_{eff}(\varepsilon,\infty)$ is shown in Figure 2 for the case of very low velocity (i.e. $u = 0.001\,a.u.$). The figure shows two clear extra peaks (P1,P2 and P1',P2') positioned at lower and higher parts of the main peak (M) having relatively small height, showing the photon side peaks located at energies $\varepsilon = \varepsilon_A(Z_o) + n\hbar\omega$; $n = -2,-1,1,2$. Since, the position of the peak (P2) is above Fermi level, it will be ignored since it does not contribute to $P(t)$. While the position of the lower one (P2') affects the calculated of the area under the curve, but it may be of order of 10^{-1} of the area under the central one which may also be ignored in comparison. Consequently, these peaks (P2 and P2') may be ignored for such application that needs an analytical formalism to avoid mathematical complexes.

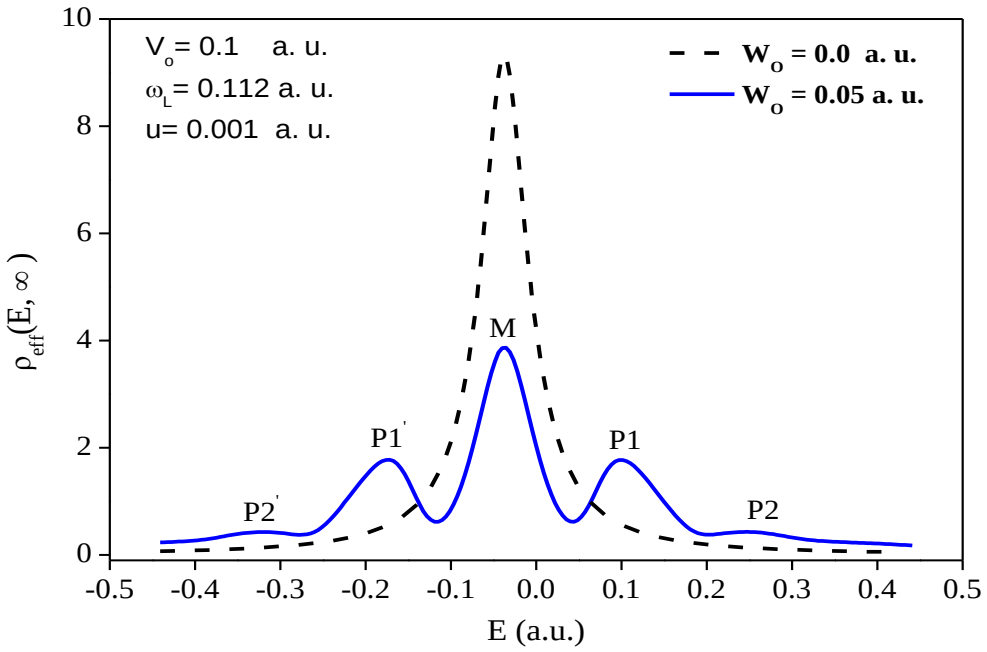

Figure 2. Shows the effective level density of states as a function of energy at low energy $u = 0.001\,\text{a.u.}$ with $W_o = 0.05\,\text{a.u.}$ and $V_o = 0.1\text{a.u.}$.

A simulation of $I(\varepsilon)(t_f - t_o) = \rho_{eff}(\varepsilon,\infty)$ based on Eq. (16) gives the similar curve. However, the actual $I(\varepsilon)$ characteristics will be more complicated than that shown in Figure 2. The symmetrical side peaks are a result of using the wide band approximation i.e. $\rho_S(\varepsilon) \to \bar{\rho}$.

However, the final ion level occupancy $n_A(\infty)$ depends on the shape of $\rho_{eff}(\varepsilon, t_f)$ which is needed to perform the integral in Eq. (15).

The effects of different parameters such as V_o, W_o and u on the peaks height as well as widths of the shapes as the ion is scattered off the surface $(t = \infty)$ will be discussed. The relative variation of the peaks height of the three peaks (M , P1 and P1') are as follow. On varying the values of V_o (keeping $u = 0.01\,\text{a.u.}$ and $W_o = 0.05\,\text{a.u.}$), the height of the central peak largely increases due to the effect of the hopping process, while the height of the other peaks decrease (Figure 3a). While one finds opposite features when W_o is increased.

On keeping $V_o = 0.1\text{a.u.}$ and $u = 0.01\text{a.u.}$ with W_o varying, the electromagnetic strength effects appear in increasing the heights of the left and right peaks (Figure 3b).

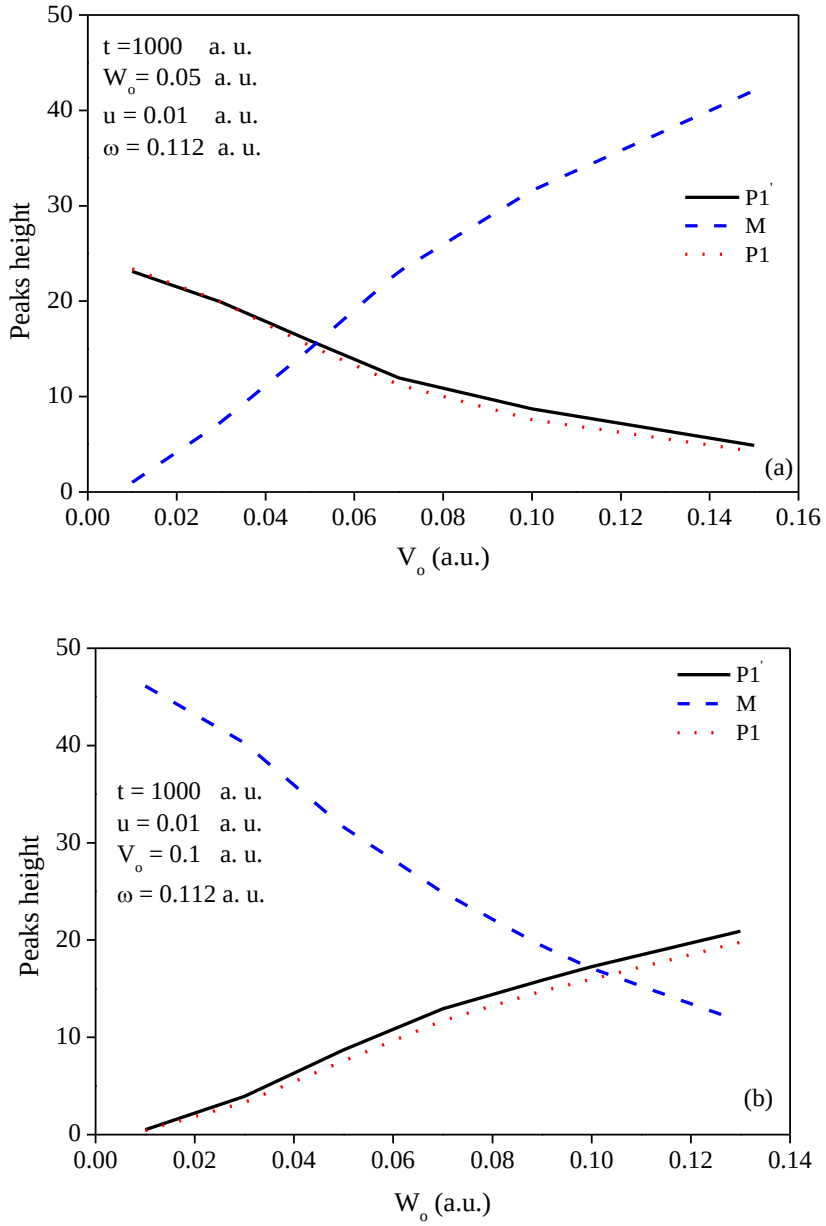

Figure 3. Shows the variation of peaks height as a function of V_o in (a); W_o in (b).

In the following, the adiabatic and non-adiabatic regimes will be discussed. The behavior of $\rho_{st.}(\varepsilon,0.0)$ is shown in Figure 4 such that by increasing Δ (by increasing the strength V_o) at fixed $W_o = 0.05 a.u.$ and $\omega = 0.112\,a.u.$, one notes that decreasing V_o makes the peaks height ratio changes. When $V_o = W_o$, the peaks height become equal (Figure 4c) .

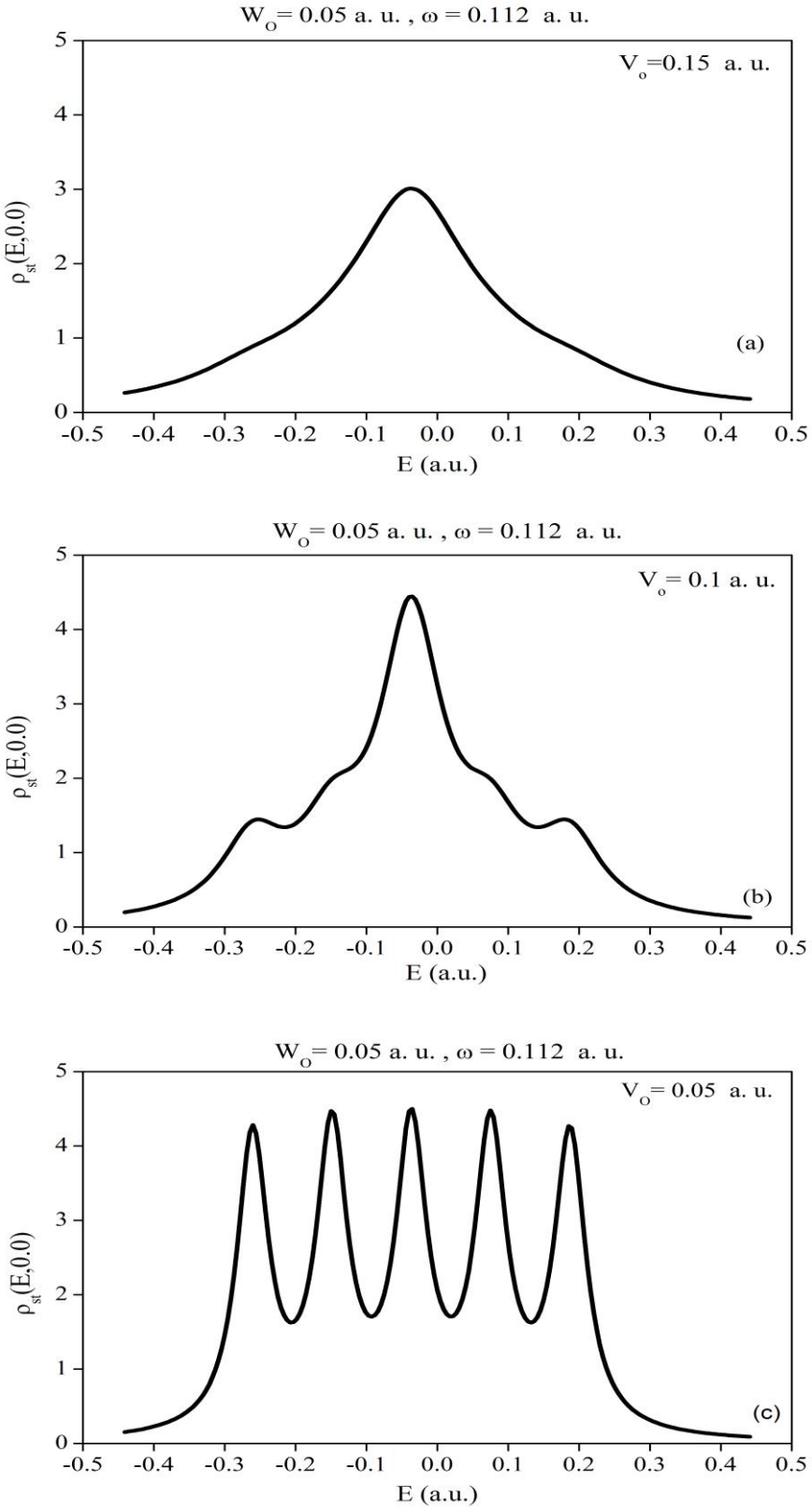

Figure 4. Shows the behavior of $\rho_{st.}(\varepsilon,0.0)$ in non-adiabatic regime when decreasing Δ by decreasing the strength V_o, where (a) $V_o = 0.15$ a. u. (b) $V_o = 0.1$ a. u and (c) $V_o = 0.05$ a. u at fixed $W_o = 0.05$a.u. and $\omega = 0.112$ a.u..

On transition from non-adiabatic regime to adiabatic regime by decreasing the frequency ω , we change the inequality between Δ and ω,this is obvious in the $\rho_{eff}(\varepsilon,\infty)$ behavior as seen in Figures 5a, 5b and 5c .

Figures 5a, 5d and 5e show also that by increasing the field strength W_o, one can change the relative height and width of the peaks of $\rho_{eff}(\varepsilon,\infty)$ .In this case, the dynamic one, we note that when $V_o = W_o$, the side peaks become higher than the central one in contrary to the steady state behavior in Figure 4. Figure 5f ensures that as W_o increases larger than 0.1 a.u. the distribution becomes lower and wider. Which explains how the field controls the ion – surface processes.

Finally, the transition from non-adiabatic regime to adiabatic regime but now by increasing the strength V_o and fixed $W_o = 0.1\text{a.u.}$ and frequency $\omega = 0.112\,\text{a.u.}$ as seen in Figures 5d and 5f. Figures 5d and 5f show physically logic feature, since as $V_o > W_o$, the electromagnetic field effect on the structure of $\rho_{eff}(\varepsilon,\infty)$ is reduced as compared with case $V_o < W_o$ (Figures 5a, 5d and 5e).

4. Conclusions

The one-electron Hamiltonian has to have two terms added in order to examine the electromagnetically assisted surface-ion charge transfer system. (see refs.[1,8]) It takes into account how the species and solid surface orbitals are coupled as a result of how they interact with the electromagnetic field. Thus, we use an appropriate arrangement of the two controllable parameters associated with electromagnetic field, these are electromagnetic frequency ω and its strength of coupling due to electromagnetic field W_o, which can be selected (with the parameter V_o) to discuss the adiabatic and non-adiabatic tunneling regimes in ion-surface scattering process. The process general features are checked throughout extensive model parameterization. At high values of W_o, a startling decline in the probability of neutralization is observed; this could be the result of the development of a quasi-molecular state. If we believe that the increase in W_o indicates an improvement in the total atomic level-broadening $\Gamma = 2\Delta$ caused by the electromagnetic field, then this drop might be regarded as an expected one. i.e. decreasing the "atomic level life-time". This may affect the tunneling of electrons from the surface to the atom (which determines the neutralization probability). This may also guide the tunneling to the adiabatic or the non-adiabatic regimes. The temporal effective density of states is an important and essential quantity, through energy integration, to obtain the ion neutralization probability after scattering from the surface. It simultaneously describes the time averaged of the transient energy-dependent current through the system. Finally, one should note that our results reflect the use of two approximations. The first one is the dipole approximation that used in obtaining $W(t)$ in Eqs. (8) and (9) which restricts the frequency ω values. And the second one is the wide band approximation $\rho_S(\varepsilon) = \overline{\rho}$ (Eq. (10)) which leads to the symmetrical shape of $\rho_{eff}(\varepsilon,t)$ and then to equal side peaks height.

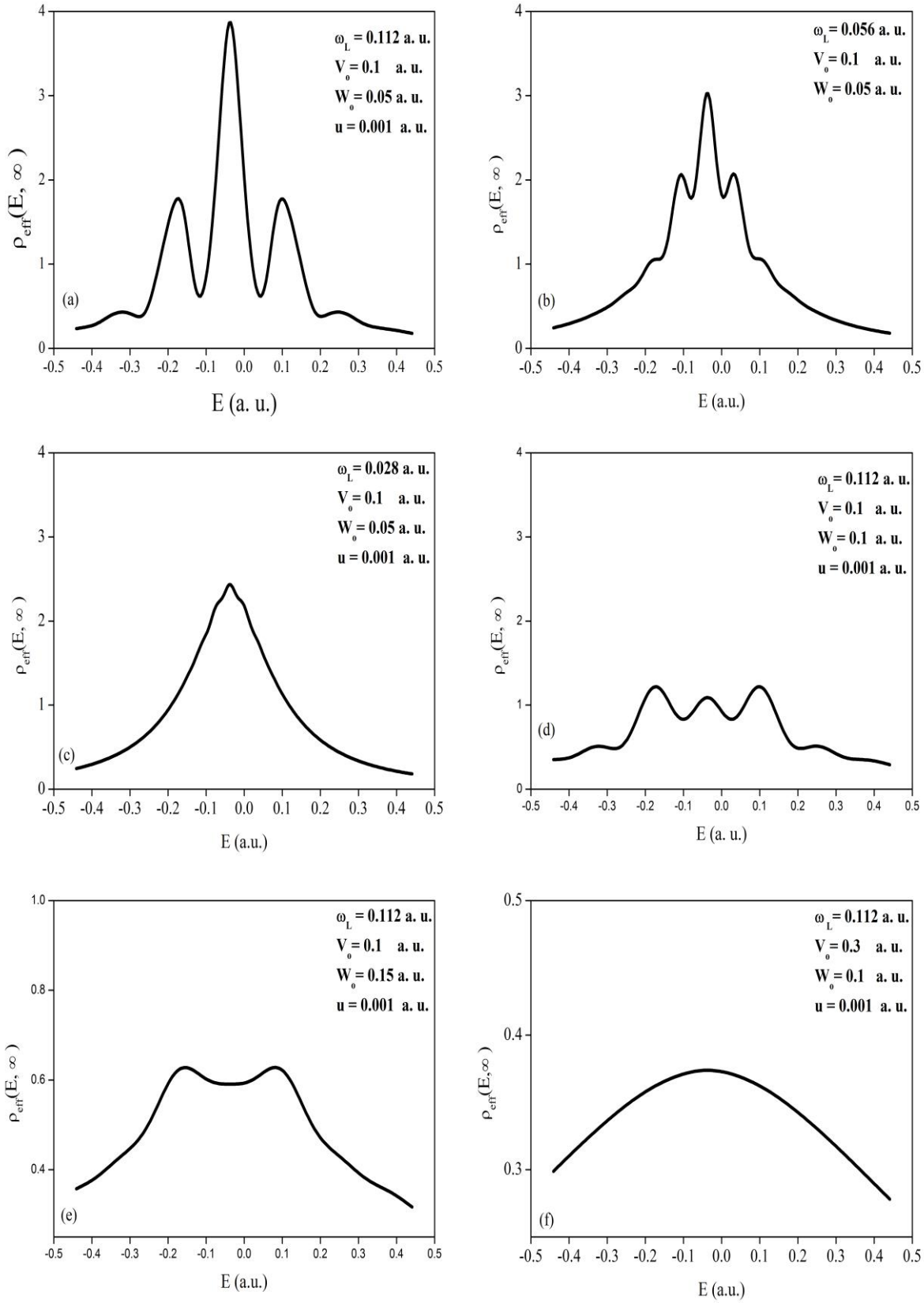

Figure 5. Shows the temporal effective density of states for different system parameters and electromagnetic fields characteristics.

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