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Negative ion formation during scattering of H-atom from-Al (111) Thin Films: Using Perturbative method

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Abstract

In this work, electron capture by H- projectiles in grazing scattering from Al(111) thin films [with difference thickness, which can be taken as a discrete set of values corresponding to different amount of atomic plane within Al(111)],is studied theoretically using Perturbative method. The resonance charge transfer(RCT) on thin film appears to be quite different from that on a semi - infinite free- electron approximation metal, this distinction is related to the difference of the electron structure of thin- metal films and the semi-infinite metal. The size quantization in this metallicfilms allows through the variation of the film thickness. Some of the feature of RCT on thin films have already been discussed based on non Perturbative methods (example CAM method) studied , so we treat the same systems using CAM method for comparison .

Key words: Thin films, resonance charge transfer, grazing scattering, negative ion fraction.

دراسة في تشكيل ايون الهيدروجين السالب عند استطارة ذرة الهيدروجين بالقرب من سطح *Al(111)* المصنع بشكل غشاء رقيق: بأستخدام الطريقة المضطربة

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المستخلص

في هذا العمل ،تمت دراسته اقتناص الاكترون من قبل ذرة الهيدروجين عند استطارتها من اغشيه رقيقه ذات سمك مختلف من سطح Al (111) باستخدام الطريقه المضطربه . وقد بدا واضحا ان انتقال الشحنه الرنين (RCT) بين ذره الهيدروجين والغشاء تكون مختلفه عند استطاره الرنين بين ذره الهيدروجين وسطح المعدن شبه اللا نهائي وذالك بسبب التكميم لحركه الاكترون باتجاه العمود عن الغشاء. ان الكثير من التطبيقات على الاغشيه الرقيقه قد تم دراستها مسبقا باستخدام نمط ازدواج الزخم الزاوي (CAM) لهذا قمنا باستخدام طريقه (CAM) النفس النظام في هذا البحث للمقارنه.

1- Introduction

Charge exchange phenomena between ions and solid surfaces have been studied Extensively[1 – 6] . Many experimental studies on charge exchange between atom (ion) and metal surfaces have been performed by scattering fast atoms or ions off clean surfaces under grazing angle of incident [7 – 11] In these studies, the electron exchange depends essentially on the quantity of the parallel velocity of the projectile ($v_{//}$). This dependence is explained by the fact that electron structure of solid change in system of coordinate relative to the projectile moving parallel to the surface and affects character and intensity of charge exchange process. Most of the experimental studies on the formation of negative ion of metal surfaces has been described by the small binding energies like H^- [12 , 13] and Li^- [14 , 15] , the affinity level of these projectiles are always lying over the Fermi- level of metals, so the RCT process will take place with the unoccupied states of the metal, then the electron will loss from atom to the surface results in a small negative ion yield.In order to increase these yields we must, either lowering the surface work function, or using grazing scattering with small indecent angle. In the 2nd case the kinematics affect of parallel velocity bridges the energy gap between the affinity level of the projectile and the occupied states of the metal surface "$v_{//}$ effect"[16 , 17] Perturbative method has been widely used for calculation of the transition rates and negative ion fraction of atomic levels at large distance in front of metal surface. Last investigation years, some nonperturbative methods was successfully employed for of charge exchange under grazing scattering, for example (CAM) Coupled Angular mode method (Teillet et.al., 1990) was used for treatment of experimental results for formation of negative – ion under grazing scattering from metal surface [19]

2- Quantum Size Effect (QSE)

The field of surface science has developed very quickly during past few years, along with advances in fabrication and measurement technologies at nanometer length scales. The dramatic reduction in size brought along the concepts of reduced dimensionality, reduced symmetric and change in structural geometry [20 , 21]. A good example to (QSE) is a thin metallic film grown by depositing a metal layer on a dielectric substrate with large band gap lying on the conduction band of the metal. When the thickness of a metal film reaches nanoscale dimensions, QSE can cause many material properties which differ greatly from those of the bulk [22 – 25] . The electron gas within a film is confined in the direction perpendicular to the plane of the film, these confined states called quantum well (QW), with energy levels, where $E(k_{//})$ is the energy $E = E_n + E(k_{//})$ measured from the bottom of the well associated with motion parallel to the film surface and,

$$E_n = (\frac{\hbar^2}{2m})(\frac{n\pi}{L})^2 \qquad \text{.....(1)}$$

The change of these energies as a function of film thickness (L) have been linked to many unique properties of ultra thin films and multilayer. charge exchange process between the incoming particle and thin metal films is differ from charge exchange with semi- infinite metal [23], this distinction is related to the difference of the electron structure of thin- metal films and the semi- infinite metal. The size quantization in this metallic films allows through the variation of the film thickness. In the present work, the influence of $v_{//}$ on electron exchange process is consider in case of grazing scattering at thin Al films using Perturbative method.The probability of H^- ion outer shell electron transfer from n- th level of the film is calculated during approach of H-atom to the film from infinity ,with different thickness of the metal film (L) as a function of atom- surface distance (d) and the atom velocity parallel component ($v_{//}$). These results show a pronounced dependence of the negative- ion fraction on the method we applied, on velocity component parallel to the surface and on the film thickness.

3- Electron Structure of Metal Film / Model Potential

Electron structure of Al thin films is described by free electron model (jelly model) [23], it depends only on distance between electron and surface (z) " where z correspond to vacuum". $z\rangle 0$ calculated from image surface

$$V_{e-film(z)} = V_j(|z| - L/2) \qquad \text{.....(2)}$$

Where $V_j(z)$ defined the potential of semi-infinite metal , L is film thickness , z is the distance between electron and middle of the thin film. For thin film, the electron is moving freely along parallel to the surface, so the interaction potential in equation 2 is electron's coordinate parallel to the surface independent. Within the potential in equation 2 electrons of the film have discrete set of energy state in the perpendicular to the surface direction, which correspond to proper value of energies of the potential well (equation 1).

4- Wave Functions Definition

The Proper function of the film (thickness of several monolayer) are given by

$$\phi_{n,k_{//}} = e^{i k_{//} \cdot r_{//}} \Psi_n(z)$$

$$\phi_{n,k_{//}} = e^{i k_x x} e^{i k_y y} \Psi_n(z) \qquad \ldots\ldots\ldots\ldots (3)$$

with kx and ky for x and y coordinates parallel to the surface, however , the wave function dependence on z is given by ψn(z), where ψn(z) are normalized wave function with discrete energies En . To described a thin metal film, it is useful to consider a deep potential well with a thickness L is small while the two other dimensions are very large, then ψ n(z) are calculated using elementary quantum mechanics [26]

$$\Psi_n(z) = A_n \cos\sqrt{2E_n(z)} \qquad \ldots\ldots\ldots\ldots (4)$$

A_n is the normalization constant which is function of the film thickness (L) and equal. $(2\pi/\sqrt{L})$ to The wave functions of metal electrons are:-

$$\langle r | k \rangle = e^{i k_{//} \cdot r_{//}} \psi_{k_z}(Z) \qquad \ldots\ldots\ldots\ldots (5)$$

$$\Psi_k(z) = \sqrt{2}\cos(k_z z) \qquad \ldots\ldots\ldots\ldots (6)$$

In the range distance z from the center of the film to $+\infty$, $V_{e\text{-film}}(z) \equiv V_{e\text{-metal}}(z)$ [27]. So by made a comparison between the wave function in equations 4 and 6 then the wave function of the film will take the form;

$$\Psi_n(z) = \frac{A_n}{\sqrt{2}} \Psi_{k_z^n}(z) \qquad \ldots\ldots\ldots\ldots (7)$$

$$k_z^n = \sqrt{2E_n} \qquad \ldots\ldots\ldots\ldots (8)$$

5- The Transition Matrix Elements for Thin Film Using Perturbative method

The transition matrix elements between metallic states $\langle \phi_{n,k_{//}} |$ of thin film and the atomic state $|a\rangle$ for H^-/metal system are defined by,

$$M^{e-film}(n, k_{//} \to a : d) = \langle \phi_{n,k_{//}} | V_{e-a} | a \rangle \qquad \ldots\ldots\ldots\ldots (9)$$

Where $k_{//} = \sqrt{2(E_n - E_a(d)}$ at the resonance condition, E_n are the discrete energies for thin film, $d = v_z t$. Following the argument of [30] and assuming the perturbation to be between a pure atomic and metallic state (e^- -atom interaction). V_{e-a} is unperturbed core potential of the neutral atom, and it can be replaced by some constant value, $V_{e-a} = -\lambda$ then equation 12 is replaced by

$$M^{e-film}(n, k_{//} \to a : d) = -\lambda \langle \phi_{n,k_{//}} | a \rangle \qquad \ldots\ldots\ldots\ldots (10)$$

This introduced an overlap integral multiple by a scaling constant. This reduction is particularly valid for atom – surface distance larger than about $3a_o$. The corresponding bound state wave function for bound state of the $H^- - ion$, is taken from [31]

$$\langle a | = \frac{1}{\sqrt{4\pi}} \frac{[2\alpha\beta(\alpha+\beta)]^{1/2}}{(\beta-\alpha)} \frac{e^{-\alpha r} - e^{-\beta r}}{r} \qquad \ldots\ldots\ldots\ldots (11)$$

α *and* β are the effective core charge and they are chosen such that the binding energy of H^- and the effective range of the $e^- - H$ interaction are reproduced. for H^-- ion , $\alpha = 0.2355$, $\beta = 0.5315$ and $\lambda = \frac{(\alpha+\beta)^2}{2} = 0.5315$. The wave functions in equations 3 and 14 are brought into equation 19 to compute the matrix elements of the perturbing potential,

$$M^{e-film}_{a,k_{//},n}(d) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \phi_a \cdot -\lambda \cdot \phi_{n,k_{//}} \, dx \, dy \, dz \qquad \ldots\ldots\ldots\ldots (12)$$

The calculation of $M^{e-film}_{a,k_{//},n}(d)$ can be partially performed analytically (the integral of x and y) such as;

$$M^{e-film}_{a,k_{//},n}(d) = -A_o \int_{-\infty}^{\infty} \psi_n(z) \frac{e^{-C_o[z-d]}}{C_o} \, dz \qquad \ldots\ldots\ldots\ldots (13)$$

And for a fixed distance from the film (z-d) is calculated from the image plan z_{img}

Where $A_o = \lambda\sqrt{2\pi}\sqrt{\frac{\alpha\beta}{\alpha+\beta}}$, $C_o = (k_{//}^2 + \beta^2)^{1/2}$

and the wave function associated to the energy E_n for the quantized states in the film $\psi_n(z) = A_n \cos\sqrt{2E_n(z)}$.

6- H^- Fraction Calculation

Let's now consider scattering under grazing angle of H- atom on Aluminum thin films with perpendicular to the surface velocity v_z and parallel velocity $v_{//}$ and with using the rates for electron loss and capture for thin film which is defined by equations similar to that for semi-infinite metal [29]

$$\begin{Bmatrix}\Gamma_c^{film}(d)\\ \Gamma_l^{film}(d)\end{Bmatrix} = \frac{1}{2\pi}\sum_n \left|M^{e\text{-film}}_{(n,k_{//}^{res},d)}\right|^2 \int_0^{2\pi} d\phi \begin{Bmatrix} f(E_F - E_n - \frac{(\vec{k}_{//}^{res} + \vec{V}_{//})^2}{2}) \\ 1 - f(E_F - E_n - \frac{(\vec{k}_{//}^{res} + \vec{V}_{//})^2}{2}) \end{Bmatrix} \quad \ldots\ldots (14)$$

While the master equation for the evaluation of the negative ion population P^- along the outgoing part of the trajectory can be computed by integrating the following rat equation:-

$$\frac{dP^-}{dt} = -\Gamma_L(Z(t))\ P^-(t) + \Gamma_C(Z(t))\ (1 - P^-(t)) \quad \ldots\ldots\ldots (15)$$

$$P^-(z) = P^- e^{-\int_{-\infty}^{\infty} \frac{[\Gamma_l(z') + \Gamma_c(z')]}{V_z} dz'} + \int_{-\infty}^{\infty} \frac{\Gamma_c(z')}{V_z} e^{-\int_{-\infty}^{\infty} \frac{[\Gamma_l(z') + \Gamma_c(z'')]}{V_z} dz''} dz \quad \ldots\ldots (16)$$

This equation is solved numerically and the integration is solved for $Z(t) = v_z.t$. V_z is the normal component of the atom velocity and we take the experimental value 0 .015a.u. for the hydrogen atom , this condition is equivalent to an energy for the normal motion of about 5eV. [28]

7- Results and Discussions

With theoretical concepts outlined in the previous section, the matrix elements $M^{e-film}_{n,k_{//}\to a}(d)$, the transition rates Γ_c , Γ_l , $\Gamma^{film}_{total}(d)$ and finally the negative hydrogen ion fractions at Al(111) thin films are calculated for different thickness (1ML, 2ML , 3ML, 4ML, 5ML, 6ML and 9ML). The Fermi energy and the metal work function differs from one of semi- infinite metal and depends upon thickness of the film, based on condition that in the volume $L\times L\times L$ of film and semi- infinite metal is located equal quantity of electrons (Shestakov et.al.,2009),

$$E_F = \frac{1}{n_{max}}\left\{\frac{L}{\pi}\cdot\frac{k_F^3}{3} + \sum_{n=1}^{n_{max}} E_n\right\} \quad \ldots\ldots\ldots (17)$$

$E_n \langle E_F$ and $n = 1,2,3,....n_{max}$, k_F is the Fermi vector in the case of semi- infinite metal. Table (1) contain the calculated Fermi energy E_F, Work function W and the film thickness L "which can be taken as discrete set of values corresponding to different amount of atomic plane within Al(111) " .

Table(1): The calculated Fermi energy , work function and the film thickness for Al(111) thin films

Al Film	Film Thickness a.u.	E_F(eV.)	W(eV.)
3ML	13.3	12.43	3.51
5ML	22.165	12.26	3.68
7ML	31.033	12.08	3.86
9ML	39.8	11.92	4.02
∞	——	11.65	4.2

Fig.(1) and fig.(2) we presents the square resonance matrix elements for H^- ion infront of Al (111) thin films with different thickness (1Ml- 9Ml), as a function of the distance between ion and film [measured from the image plane $z_{im} = \pm(\frac{L}{2} + 1a.u)$ [23]] .The figures also include comparison with the similar results obtained for semi- infinite metals " our calculation too".In the static situation (fixed ion- surface distance) it is important to find the ion. Level width (the total transition rates) $\Gamma^{film}(d)$ calculated from the summation of Γ_l with Γ_c or $\Gamma^{film}(d) = \frac{\Gamma_c}{g^-} + \frac{\Gamma_l}{g^o}$, and we use the statistical factors equal $g^o = 1$ and $g^- = \frac{1}{2}$ so fig. (3) and fig.(4) presented the results of $\Gamma^{film}(d)$ for $H^- - ion$ on Al (111) with different thickness (1ML-9ML) these results include also $\Gamma^{film}(d)$ for semi –infinite free electron metal surface. We notice here that these results differ from the case of film, this distinction is associated with the different of electron

structure of metal film from semi- infinite metal . With increasing the film thickness the width $\Gamma^{film}(d)$ converge towards the results for the semi- infinite metal, this means, by increasing the film thickness one should recover the limit of a semi- infinite free-electron surface, i.e. the density of quantized states in the 2D-film increases leading to 3D traveling states in the semi- infinite limit. The level width for all films and for the systems H^-/Al(111) displays an exponential behavior with the ion- film surface reaching very large values in eV. range at small distances. No step structures in the $\Gamma^{film}(d)$ dependence on the distance is observed, the same behavior exists in Na+-ion neutralization at Al films [30].

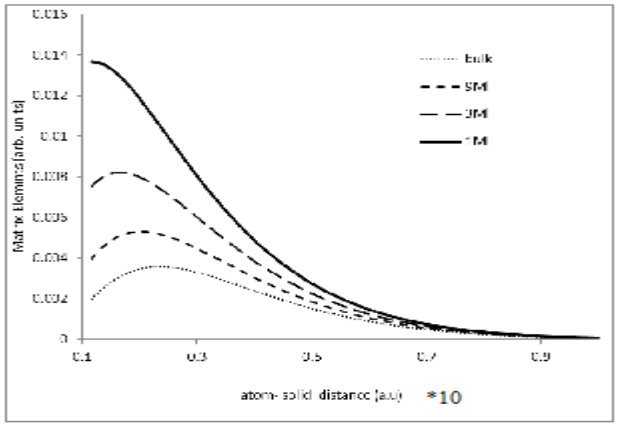

Fig.1: Matrix elements for H^- - ion in front of Al (111) thin films using the Perturbative method.

8-H--Fraction under Grazing Angle Scattering on Al(111)Thin Films/ Dynamical Calculations

The dependence of H^- ion formation probability upon value of parallel to the surface velocity ($v_{//}$) during scattering from thin metal film was calculated numerically using the Perturbative method (equation 16). This dependence is calculated for films of different thickness (1ML-9ML), and presented in fig.3 for Al(111) metal ,with atom normal velocity (v_z =0.015 a.u.) . The figure also represent our calculation for the bulk metal. It is interesting to remark that H^--fraction yield during scattering on the films (1ML- 6ML) thickness is more than ion yields in case of thick films ,this is due to the lower work function for thin film which is related to the quantization of the states in thin film. The H^- ion fractions shows the dependence of a kinematics resonance, where the small affinity level (-0.75eV.) for H^- -ion compared to the work function of the metal leads to small fractions (P^- =10^{-4}).During increase of film thickness the maximal value of negative ions fraction decreases to be approached the maximal value of bulk metal, where during increase the film thickness the density of it's two dimension levels increases,and reaches the case of three dimensional band of metal . Moreover, during increase of film's thickness the maximum of dependency for both systems shifted toward higher values of $v_{//}$, which is connected with the dependence of Fermi energy on the film thickness.

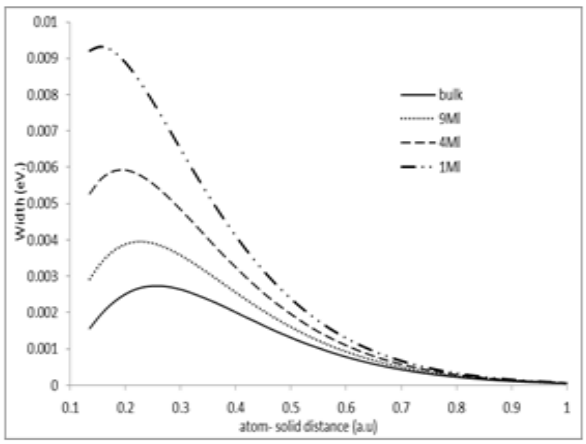

Fig.2: Total transition rates for H- / Al (111) thin films using the Perturbative method

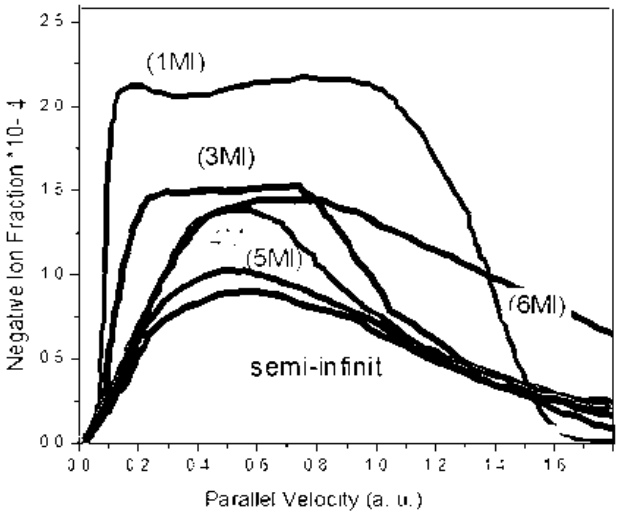

Fig.3: Negative ion fraction for H^- / Al (111) thin films using the Perturbative

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