

## Theoretical Study in the Ion Grazing Scattering on Aluminum Surface: Neutral Fraction Calculation

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### Abstract

In this paper, we performed a theoretical study on the formation of alkali atom in its ground state formed in grazing scattering of  $Li^+$ ,  $Na^+$ ,  $K^+$  and  $Rb^+$  from (111)-Aluminum surface. Our treatment is based on the coupled angular- mode method which is applied to the treatment of the parallel velocity –assisted electron capture in grazing scattering. This can be explained from the "shifted Fermi sphere" model with respect to the Fermi energy.

The angular distributions and the natural fraction of the ground state as well as the excited states were calculated for static and with incorporating the velocity of incoming ion.

The calculating natural fraction are in reasonable agreement with the available experimental data.

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### Introduction

Charge exchange process from ion beams scattered from solid surfaces has long studied both experimentally and theoretically [1, 4]. One reason for this is that the charge exchange process offers spectroscopic techniques for probing electron state in the first layer of surfaces. In particular, a low energy ion with small velocity, less than Boher velocity, can capture a surface electron in the conduction band, this can be regarded as

elastic or inelastic quantum tunneling from the surface electron state to the atomic electron state. For an ion beam with few Kev/amu, the grazing angle at which the beam is incident on surface is chosen so that the interaction time is sufficiently long. Here the ion beam experience many small angle elastic scattering with surface atom, and can move nearly parallel to the surface.

This kind of scattering (grazing scattering) has essential difference from

other scattering with its fast motion parallel to the surface with a velocity component ( $V_{//}$ ) of a fraction of a.u. or higher, and is important in many fields of technology applications, such that in atomic spectroscopy<sup>[5]</sup>, nuclear physics<sup>[6]</sup>, electron emission<sup>[7]</sup>, epitaxial growth and structure of ultra thin films deposited solid surfaces<sup>[8]</sup>.

Since many established surface analytical tools are affected by electron loss and capture in a decisive manner, so, the calculation of neutral probability gives a shortest method to comparing the theoretical and experimental work for charge transfer interaction. Attempts were made to utilize the neutral fraction experimental results to form the transition rate function. Most of theoretical works<sup>[9]</sup> assumed this rate to have a general form defined as a simple exponential function, however different works<sup>[10]</sup> show that this is not completely give a good consequences especially at small distance ( $z$ ) from the surface. Then for grazing ion-surface collision, the rate equation approach can be used to describe the time evolution of the charge states of the scattered particle.

Electron states of the atomic particle and the surface are defined in different reference frames moving with respect to each other. Thus one expects that transition

factors affect electronic transitions between atomic particle and the surface.

### Model

The coupled angular mode method (CAM) is a scattering method allowing for calculating of atomic or molecular systems in front of a metal surface and it is already successfully applied for atomic or molecular levels that can be described as one electron levels, as well as for certain multi electron problems<sup>[11]</sup>, although, it has been used for negative ion system<sup>[12]</sup>, and it can also applied to neutral and positive ion systems<sup>[13]</sup>. So it is a well suited for our problem, where the CAM method was applied here to the case of neutral atom formation in positive ions-Al(111) surface collisions.

In CAM method one studies how an electron is scattered by a complex potential of an electron – atom and electron – surface interactions, where the electron – surface interaction is represented by a potential only function of  $z$  (the distance between ion and the surface), and it is taken from the work of Jennings<sup>[14]</sup>,

$$V_{es}(z) = -\frac{1}{4z} (1 - e^{-\lambda z}) \text{ if } z > 0 \quad \dots (1)$$

$$V(z) = -u_0 / (Ae^{Bz} + 1) \text{ if } z < 0 \quad \dots (1)$$

and to get a good linking at  $z = 0$ , A and B will be,

$$A = -1 + 4u_0 / \lambda \dots\dots\dots( 2 )$$

We choose  $AU_0$  and  $\lambda$  parameters to represents Al (111) surface ( $\lambda = a_0^{-1}$  and  $U_0 = \text{eV}$  ). While the electron – atom interaction is described

by using the effective density method, in which the space of the electron is divided into tow regions, the outer  $r > r_c$  ( $r$  is the e- atom distance) the e-atom interaction is described by a long range potential  $V_{ext}^{lm}(r)$  which is depends on the spherical wave function depends on  $l$  and  $m$  in the spherical harmonics  $Y_l^m(\theta, \phi)$  in such away,

$$V_{ext}^{lm}(r) = -\frac{\alpha_i}{2r^4} + \frac{\ell(\ell+1)}{2r^2} \dots\dots( 3 )$$

$\alpha_i$  is the polarizability of the state  $i$  of the atom,  $\ell$  is the electron angular momentum. The inner region dose not treated explicitly, so it will be replaced by energy independent boundary condition on the electron radial wave function at

$$r = r_c \text{ (critical radius).}$$

In grazing scattering, and due to the large parallel velocity component  $V_{||}$ , the e-atom and the e- surface potentials are supposed to be unperturbed to each other. In this limits one can assume that the ion is not modified by the metal surface, but at small distance from the surface the atomic orbitals are

modified by the surface potential and an overlapping between the atomic orbitals (e.g.  $n_s$  and  $n_p$ ) will occur.

The effective electron wave function is expanded over spherical harmonics  $Y_{lm}(\theta, \phi)$  ( $m$  is the projection of the angular momentum of the electron on z-axis which is perpendicular to the surface and going through the atom).

$$\psi = \sum_{\ell} \frac{1}{r} F_{\ell}(r) Y_{\ell}^m(\theta, \phi) \dots\dots\dots( 4 )$$

The angular distribution  $|\sigma_i(\theta, \phi)|^2$  is obtained from the eigenvector  $Q_{\ell m}$  of the time-delay matrix [15].

$$|\sigma_i(\theta, \phi)|^2 = \left| \sum_i Q_{\ell|m|}^i(z) Y_{\ell|m|}(\theta, \phi) \right| \dots\dots( 5 )$$

$i$  referred to the state  $n_s, n_p$  respectively.

The  $Q_{\ell m}$  transfer to a diagonal matrix equal to the exponential resonance occupation for the system, by assuming the state resonance is centered at  $\epsilon_a$  then

$$Q(\epsilon) = \hbar\Gamma \left[ (\epsilon_a - \epsilon)^2 + \Gamma^2 \right]^{-1} \dots\dots( 6 )$$

Where  $\Gamma$  is the level half width of the incoming atom, one of the eigen values of  $Q_{\ell m}$  matrix is large and have a positive value and exhibits a bell shaped resonance variation with energy. Then the level width connected with maximum  $Q(q_{max})$  is.

$$\Gamma = \frac{2}{q_{\max}} \dots\dots\dots( 7 )$$

The eigen functions associated with this maximum eigen value will contains all the information relevant to the parallel velocity effect.

The quantity  $|\sigma_i(\theta, \phi)|^2$  in equation (5) gives the angular distribution of the transition probability, It reflects the existence of the most transparent direction for electron tunneling and this function is peaked in a rather narrow  $\theta$  interval around the surface normal.

In calculating the angular distribution (eq. 5), we use for the eigen vector  $Q_{lm}$  an approximate formula  $Q_{lm} = q_o \exp(-\alpha z/4)$

, where  $q_o$  is an adjustable parameter and  $\alpha$  is a parameter related to the ionization level of the incoming ion and the surface parameters  $\epsilon_1$  and  $W$  and the image shifted dew to the polarization of the surface, fig.(1), give an example of the angular distribution calculated from equ. (5), normalized to one at maximum of (ns, np(m=0), np (m= ± 1)) states , where n=1,2,3,4 for Li, Na,K,Rb-ions respectively scattered from Al(111) surface. As it is seen from these figs., the np(m=± 1) states have an angular distribution equal to zero at  $\theta = 0$  direction ,this behaved reflected the structure of these states.

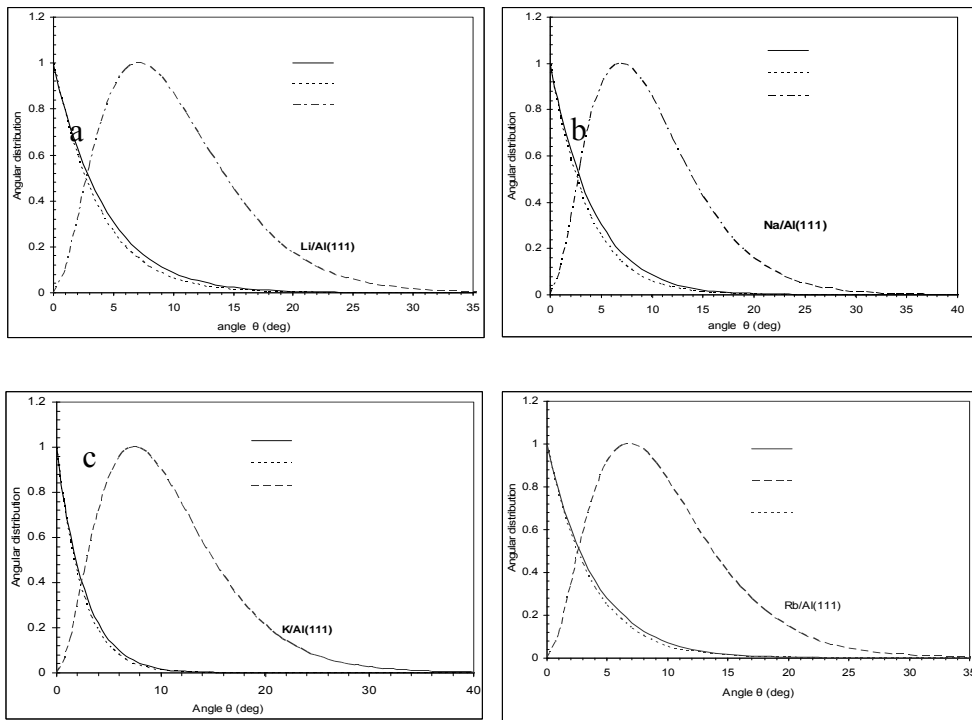


Fig.(1):-Angular distributions (Normalized to one at maximum)

for ,(a) Li-states, (b): Na-states, (c): K-states, (d): Rb-states ,in front of Al(111) surface. S-states, solid line, p ( $m=0$ ), dotted line, p ( $m=\pm 1$ ), dashed line.

In fig. (2), the calculated angular distribution to the nd-states, scattered from Al (111) surface, (  $2d(m=0), 2d(m=\pm 1), 2d(m=\pm 2)$  ) orbital for  $Na^+$  -ion,

$3d(m=0), 3d(m=\pm 1), 3d(m=\pm 2)$  orbital for  $K^+$  -ion, and (  $4d(m=0), 4d(m=\pm 1), 4d(m=\pm 2)$  ) orbital for  $Rb^+$  -ion respectively . All these results are taken at an ion – surface distance of  $18 a.u.$

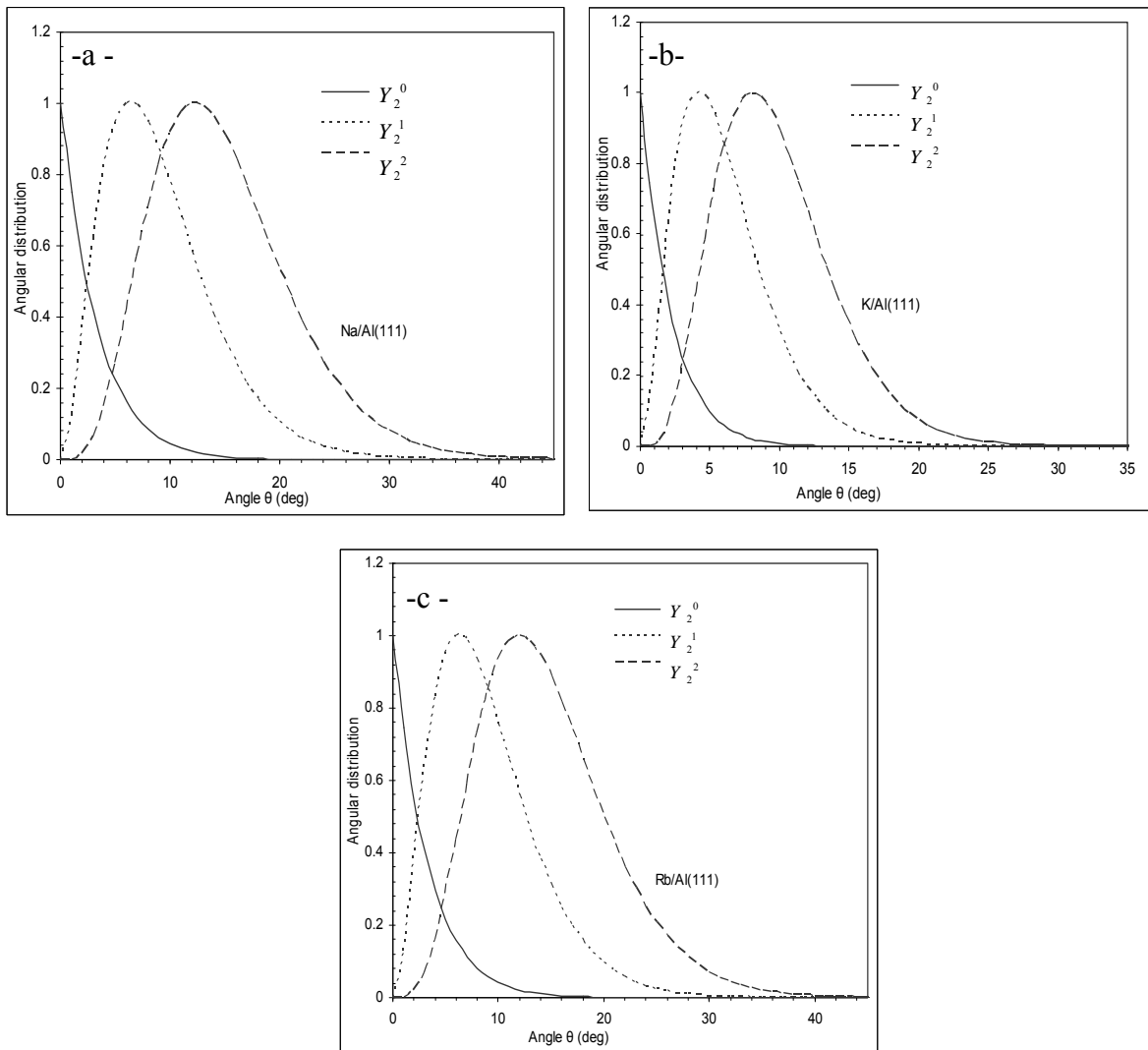


Fig.(2 ):-Angular distributions (Normalized to one at maximum) for ,(a) Na-

states, (b): Rb- States ,in front of Al(111) surface. S-states, solid line, d (m=0), dotted line, d (m=± 1), dashed line and d (m=±2).

The angular distributions for various ion- surface distance (  $z = 5 a.u., 8 a.u., 14 a.u$  measured from the image reference plane ) , have been normalized at  $\theta = 0$  , are shown in fig. (3)

for  $Rb^+$  on Al(111) surface. The three angular distributions are close to each other, and the distributions are strongly peaked around  $\theta = 0$  .

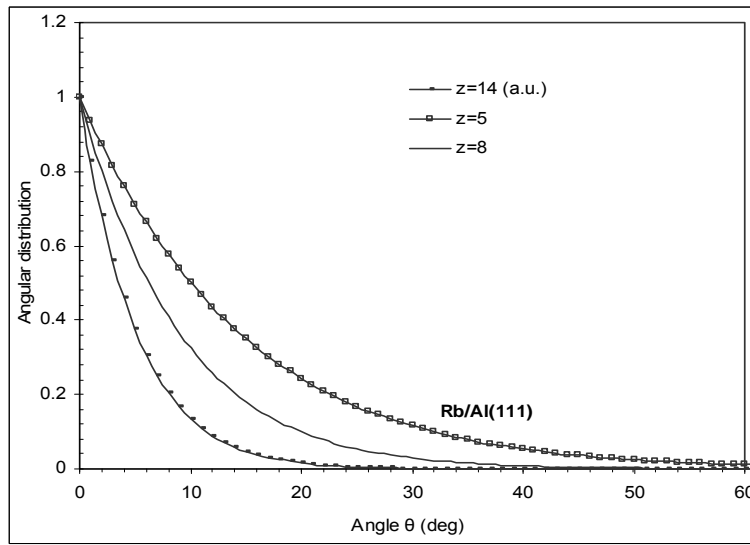


Fig.(3):- Angular distributions (Normalized to one at maximum) for  $Rb^+$  scattered from Al(111), for various distances  $z = 5 a.u., 8 a.u., 14 a.u$  ( measured from the image reference plane) .

The simple picture to described the resonant electron transfer between an atom and a continuum of metal states tunneling is based on the general concept of quantum mechanics density outlined by

Bardeen<sup>[16]</sup> ], where the density of tunneling electron current electrons is given by ,

$$j = \frac{dp_a}{dt} = 2\pi \sum_{k_a} |M_{ka}(z)|^2 [f_k(1 - p_a) - p_a(1 - f_k)] \delta(\epsilon_k - \epsilon_a) \dots\dots\dots(8)$$

Where  $p_a$  is the probability for the occupation of the atomic state, then  $j = \frac{dp_a}{dt}$  denoted the current through the potential barrier established between the ion and the surface,  $|M_{ka}(z)|^2$  is the transition matrix elements,  $k$  described the free electron metal with energies  $\epsilon_k$  and wave function  $\psi_k$ , and  $a$  described the atom with energies  $\epsilon_a$  and wave function  $\psi_a$ ,  $f_k$  is the Fermi-Dirac distributions.

The first term in the square brackets of eq. (8) represents tunneling of electrons from surface to the atom i.e., electron capture, so the second term represents the electron loss, so, the electron capture (loss) rate can be defined as,

$$\Gamma_c(z) = 2\pi \sum_k f_k |M_{ka}(z)|^2 \delta(\epsilon_k - \epsilon_a) \dots\dots\dots(9)$$

$$\Gamma_\ell(z) = 2\pi \sum_k (1 - f_k) |M_{ka}(z)|^2 \delta(\epsilon_k - \epsilon_a) \dots\dots\dots(10)$$

And the composition of  $\Gamma_l(z)$  and  $\Gamma_c(z)$  yields

$$\Gamma_T(z) = \Gamma_l(z) + \Gamma_c(z) \dots\dots\dots(11)$$

Where  $\Gamma_T(z)$  defined the total transition rate.

Equation (8) is called the transition rate equation, and if we added the statistical factors for capture and loss  $g^c$  and  $g^l$  that take into account the electronic spin and participation of more than a single electron in the tunneling

process, for example  $g^c = 2$  and  $g^l = 1$  for alkali atoms, then one obtained for eq. (8):-

$$\frac{dp_a}{dt} = g^c \Gamma_c(z) (1 - p_a) - g^l \Gamma_\ell(z) p_a \dots\dots\dots(12)$$

**Effect of the Parallel velocity on electron transfer**

The effect of parallel velocity  $V_{||}$  in this work is introduced by taken into account the frame transformation between atom and metal [17, 18]. We have to transform the momenta of free conduction electrons into the rest frame of the moving ion, where this kind of Galilean transformations is well established in the field of fast ion (atom) collisions. In such transformation we focus on calculating the wave functions for the conduction band  $\psi_k(\vec{r}, t) = \exp(-i\epsilon_k t)\psi_k(\vec{r})$  in the rest frame of the ion moving parallel to the surface with a velocity component  $V_{||}$ , i.e.,  $\vec{r} = \vec{r} - \vec{v}_{||} t$  and  $\vec{p} = \vec{p} - v_{||}$

So the wave function using Galilean transformation will be

$$\Psi_k(\vec{r} - \vec{v}_{||} t, t) = \exp[-iv_{||}(\vec{r} - \vec{p}t)]\Psi_k(\vec{r}, t) \dots\dots\dots(13)$$

If we defined  $q$  as the momentum of metal electrons such as :-

$$\vec{q} = -m_e \vec{v}_{||} = -\vec{v}_{||} \dots\dots\dots(14)$$

Where we use here ( $m = \hbar = 1$ ) in atomic units.

$$\Psi_k(\vec{r} - v_{ll}t, t) = \exp(i\vec{q} \cdot \vec{r}) - \exp[-i(\vec{q} \cdot \vec{k} + \vec{q}^2 / 2 + \epsilon_k)t] - \Psi_k(\vec{r}) \dots\dots\dots(15)$$

Where  $\epsilon_k = \vec{k}^2 / 2$  is the energy of state k in the free electron approximation, by make a simple comparison between eqs. ( 13 ) and ( 15 )one holds that

$$\psi_k(\vec{r} - v_{ll}t, t) = \psi_{k+Q}(\vec{r}, t) \dots\dots\dots(16)$$

This relation means that the static wave function  $\psi_k(\vec{r})$  can be used in this treatment by shifting momenta of metal electrons ( $\vec{k}$ ) by a constant momenta  $\vec{q}$ , so that the origin of the atomic momentum space is obtained from translation by  $-\vec{q}$ , i.e.  $\vec{k}' = \vec{k} + \vec{q}$  ( $\vec{k}'$  is the effective momenta).

By looking at the description of conduction phenomena in free- electron metals [19], then the effect of the parallel velocity motion on the occupation of metal states can be visualized by a Fermi sphere shifted in the atomic frame by a momentum  $\vec{q}$ .

$$f_{k+\vec{q}} = \begin{cases} 0 & 0 \leq \epsilon \leq (1 - \vec{q})^2 \\ 0.5 + \frac{1 - \vec{q}^2}{4\vec{q}} \epsilon^{-1/2} - \frac{\epsilon^{1/2}}{4\vec{q}} & (1 - \vec{q})^2 \leq \epsilon \leq (1 + \vec{q})^2 \\ 0 & \epsilon \geq (1 + \vec{q})^2 \end{cases} \dots\dots\dots(18)$$

where  $\epsilon = \frac{E_a}{\hbar^2}$ ,  $E_a = \epsilon_o + \Delta\epsilon$   
 $q = \frac{v_{ll}}{v_F}$ ,  $v_{ll}$  is the Fermi velocity.  
 $v_F = (\hbar^2 \epsilon_F)^{1/2}$  is the Fermi sphere velocity.  
 $\vec{k}'$  is fixed by the resonance condition i.e.  $k = \sqrt{e\epsilon_a(z)}$

Then,

And the shift of the sphere is equivalent to a modification of the effective occupation of metal states in the atomic rest frame .

At temperature  $T = 0$  and  $\vec{v}_n = 0$  ( $\vec{q} = 0$ ), the feature of Fermi Dirac distribution are

$$f_k = \begin{cases} 1 & k \leq k_F \\ 0 & k > k_F \end{cases} \dots\dots(17)$$

If  $\vec{q}$  have a finite value, then the fraction of occupied states for a given  $\vec{k}'$  where results from the overlap of surface sphere (have radius  $\vec{k}'$ ) with the volume of the Fermi sphere. From geometrical arrangements one obtains the Doppler – Fermi- Dirac distributions:-



Fig. (4) illustrate the Doppler- Fermi- Dirac distributions with parallel velocity (in a.u.),

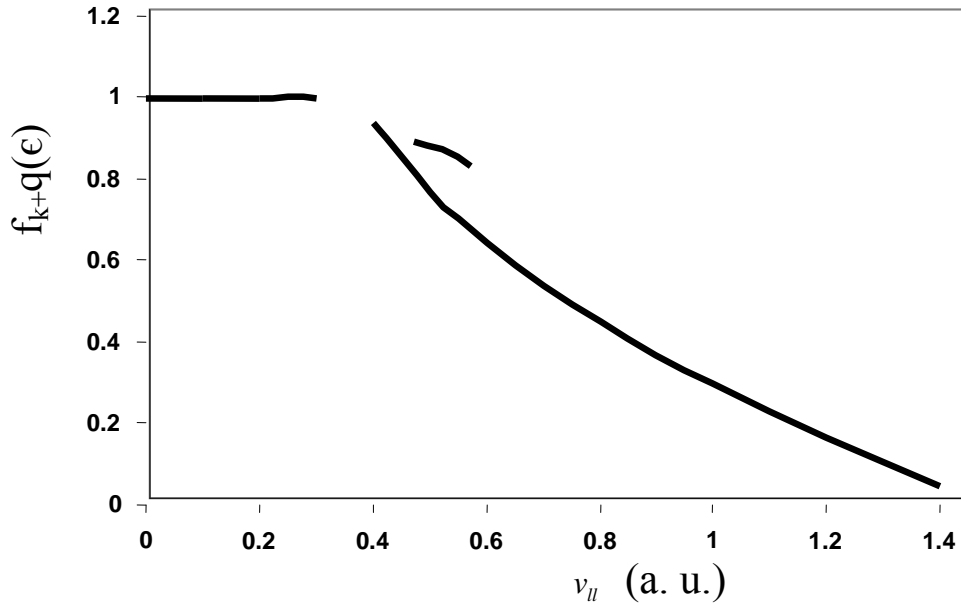


Fig. ( 4 ):- illustrate the Doppler- Fermi- Dirac distributions with parallel velocity.

As we see from this figure that for finite  $v_{||}$  we find occupied as well as empty metal states. This explained the important of grazing scattering collisions in charge exchange, since the atomic levels are brought into resonance with occupied as well as unoccupied metal states and the electron probabilities for capture or loss is

found, this leads to a new feature for charge transfer in atom (ion) –surface scattering.

By using the concept of shifted Fermi sphere , the definition in equation (18) , then the rate equations (9 , 10 ) for loss and capture can be deduced in the way,

$$\Gamma_c(z) = 2\pi \sum_k |M_{ka}(z)|^2 f_k \left( \epsilon_f - \frac{(\bar{k} + \bar{q})^2}{2} \right) \delta \left( \epsilon_f - \frac{(\bar{k} + \bar{q})^2}{2} - \epsilon_a(z) \right) \dots\dots\dots(19)$$

$$\Gamma_\ell(z) = 2\pi \sum_k |M_{ka}(z)|^2 \left(1 - f_k\left(\epsilon_f - \frac{(\bar{k} + \bar{q})^2}{2}\right)\right) \delta\left(\epsilon_f - \frac{(\bar{k} + \bar{q})^2}{2} - \epsilon_a(z)\right)$$

These relations will be introduced in equation (12) to get the occupation rate for the atomic level.

Eq. (19) contains a sum over all states  $\bar{k}$  degenerated with the ionic state and the weighting term  $|M_{ka}(z)|^2$  gives in fact the distribution over the metal state  $\bar{k}$  populated by the decay of the ionic state. This distribution is readily obtained in CAM method from the resonant eigen function  $\sigma$  of the  $Q$  matrix, indeed  $\sigma$  is a function of the electron scattering angle

$\theta$  and  $\phi$  and  $|\sigma_i(\theta, \phi, z)|^2$  gives the normalized angular distribution of the electrons ejected by the resonance i.e., exactly the same information as that contained in  $|M_{ka}(z)|^2$ .

One of the characteristics of the CAM in grazing scattering with the surface is enable us to calculate the angular distributions for transition probability as a rule to solve the rate equation where the loss and capture rates reads:-

$$\Gamma_c(z) = \Gamma(z) \int_0^{\pi/2} \sin \theta d\theta \int_0^{2\pi} d\phi |\sigma_i(\theta, \phi, z)|^2 f\left(\epsilon_f - \frac{(\bar{k}_i + \bar{v}_l)^2}{2}\right) \dots\dots\dots(20)$$

$$\Gamma_\ell(z) = \Gamma(z) \int_0^{\pi/2} \sin \theta d\theta \int_0^{2\pi} d\phi |\sigma_i(\theta, \phi, z)|^2 \left(1 - f\left(\epsilon_f - \frac{(\bar{k}_i + \bar{v}_l)^2}{2}\right)\right) \dots\dots\dots(21)$$

Where  $\sigma_i(\theta, \phi, z)$  gives the angular distribution for the transition probability between atom and metal states, and it is normalized using the condition,  $\int_0^{\pi/2} \sin \theta d\theta \int_0^{2\pi} d\phi |\sigma_i(\theta, \phi, z)|^2 = 1$ .

**Neutral Fraction calculation:**

The neutral fraction ( $P_o$ ) is the probability of the scattered projectile reaches the detector as an atom in its ground state, so the calculation of neutral

probability gives a shortest method to comparing the theoretical and experimental work for charge transfer interaction. Attempts were made to utilize the neutral fraction experimental results to form the transition rate function.

If  $p_a$  being the probability for the occupation of the atomic states, the current in equation (8) is directly related to  $\frac{dp_a}{dt}$  then

$$\frac{dp_a}{dt} = g^c \Gamma_c(z) - (g^c \Gamma_c(z) + g^l \Gamma_l(z)) p_a \dots\dots\dots(22)$$

Here we added the statistical factors for loss and capture that take into account the electron spin and the participation of more than one electron .

$$\frac{dp_a(t)}{dt} = -(\Gamma_l(t) + 2\Gamma_c(t)) p_a + 2\Gamma_c(t) \dots\dots\dots(23)$$

Then,

$$p_a(t) = p_a e^{\int_{t_0}^t [\Gamma_l(t') + 2\Gamma_c(t')] dt'} + \int_{t_0}^t 2\Gamma_c(t') \left[ e^{-\int_{t'}^t [\Gamma_l(t'') + 2\Gamma_c(t'')] dt''} \right] dt' \dots\dots\dots(24)$$

This integral is solved numerically to get the neutral fraction , and the related ionic fraction  $p^+(t)$  ( related to the probability of the scattered projectile reaches the detector as an ion) in the simple way  $p^+ = 1 - p_a(t)$

$$\dots\dots\dots(25)$$

To calculate the neutral fraction as a function of parallel velocity for alkali positive ions in front of Al(111) surface (it's work function  $W = 4.29eV$ . and  $\epsilon_F = 11.65eV$ .)  $\Gamma_T(z)$  is approximated to a simple exponential function equal to  $\Gamma = \Gamma_o e^{-\beta z}$ , where  $\Gamma_o$  defined as the level width at  $z = 0a.u.$  and  $\beta$  is an adjustable parameter .

If we assume a trajectory  $z(t) = v_z t$  ( $v_z$  is the normal component of the ion velocity) ,taking into account that  $\Gamma_l$  and  $\Gamma_c$  varied with time,

And to make a comparison with experimental data for grazing scattering ,the normal velocity component is taken to be equal to  $v_z = 2.65 * 10^{-3} a.u.$  for  $Na^+$  ions and  $v_z = 3.33 * 10^{-3} a.u.$  for  $Li^+$  ions. In fig. (5) , we presents the neutral fraction  $p_a(t)$  , normalized to one as a function of parallel velocity for  $Li^+(2s$  and  $2p)$  state ,  $Na^+(3s$  and  $3p)$  state  $K^+(4s$  and  $4p)$  state and  $Rb^+(5s$  and  $5p)$  state. At small values of  $v_{||}$  , the ions are neutralized completely into the  $s$  ground state. When the parallel velocity increased, the neutral fraction decreases flowing the ground state population evolution.

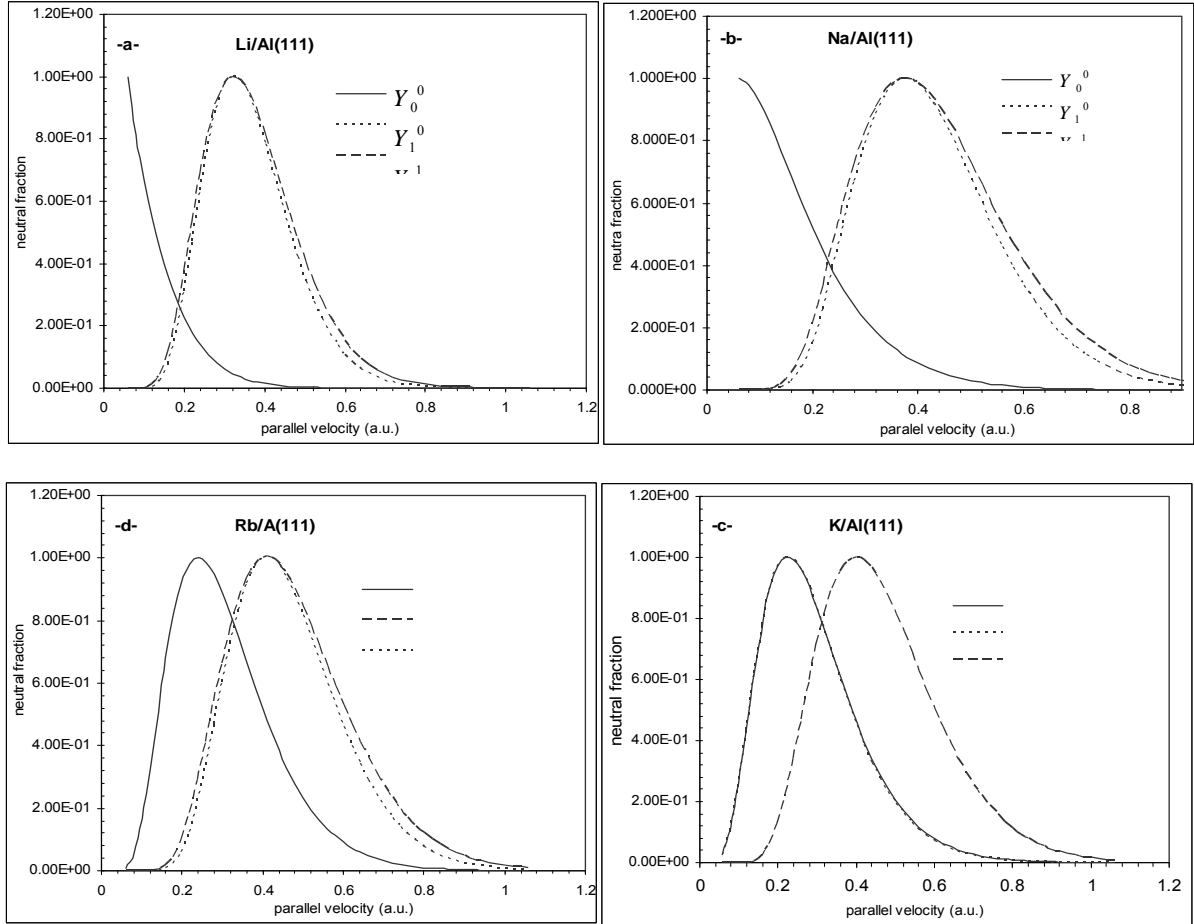


Fig.(5):- Neutral fraction as a function of parallel velocity . (a)  $Li^+ - Al(111)$   
 (b)  $Na^+ - Al(111)$ , (c)  $K^+ - Al(111)$ , (d)  $Rb^+ - Al(111)$  .

We see also in this figures, the ions in  $p$ -states population has a resonance shape with a maximum at different values for  $v_{||}$ , this behavior is different from the

dependence of the ground state population on  $v_{||}$  .

Fig. (6 ) shows a comparison with the experimental data taking from ref. (20 )

with that calculated here for  $Li^+(2s\text{-state})$  with  $v_z = 3.33 \cdot 10^{-3} a.u.$ .

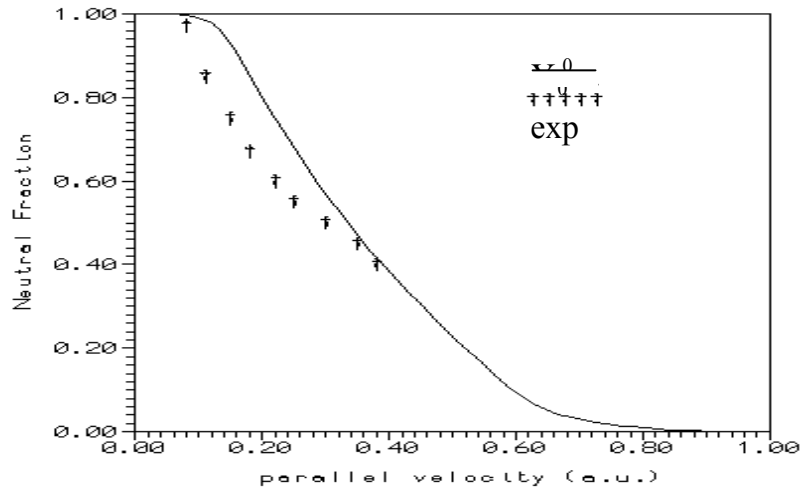


Fig (6 ):- Neutral fraction for  $Li^+ - Al(111)$  as a function of parallel velocity.our work , solid line and the data taken from ref. (20 ) solid dots with error bar.  $v_z = 3.33 \cdot 10^{-3} a.u$

The theoretical results and experimental data (ref. 21)for  $Na^+(3s)$  neutralization near  $Al(111)$  surface are compared in fig.(7) with

$v_z = 2.65 \cdot 10^{-3} a.u$  , a good agreement between our work and experimental data is obtained spatially at small parallel velocity.

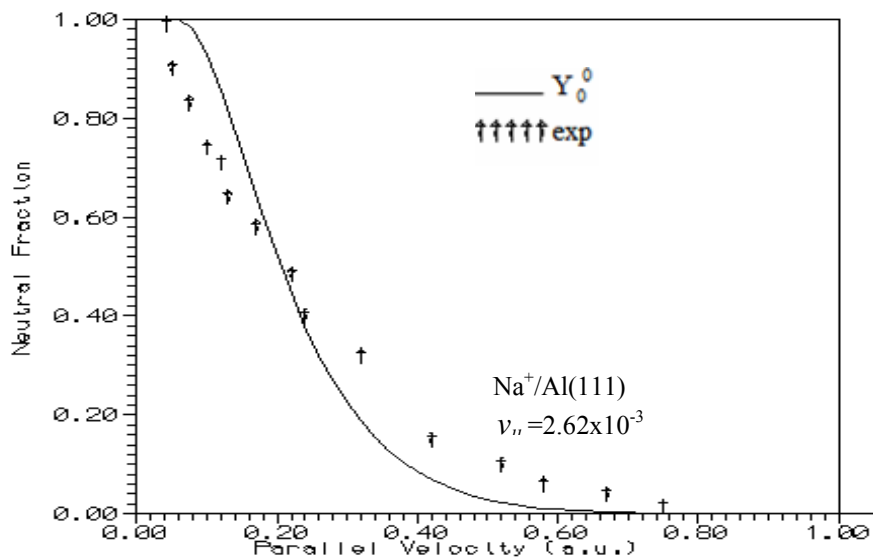


Fig (7):- Neutral fraction for  $\text{Na}^+/\text{Al}(111)$  as a function of parallel velocity. Our work ,solid line and the data taken from ref. (21) solid dots with error bar,  $v_z = 2.65 \cdot 10^{-3} \text{ a.u.}$  . While in fig.(8) we present the results for the population showing a good agreement between our work and that taken from ref. (22) for  $\text{K}^+ / \text{Al}(111)$  surface

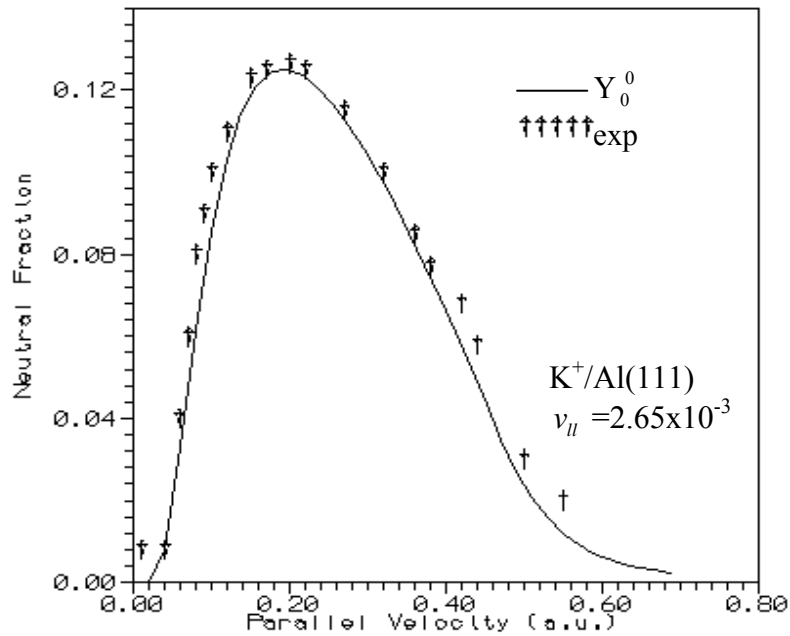


Fig.(8):- Neutral fraction for  $\text{K}^+/\text{Al}(111)$  as a function of parallel velocity. our work ,solid line and the data taken from ref. (22) solid dots with error bar  $v_z = 2.65 \cdot 10^{-3} \text{ a.u.}$

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