Effect of Dispersion in Surface Plasmon-Mediated Ion Neutralization at Metal Surfaces

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Abstract

A mechanism for ion neutralization at metal surfaces was previously suggested. In this mechanism the ion is neutralized upon scattering from a metal surface via exciting a surface plasmon. It was shown that this mechanism is of comparable probability as the other two mechanisms, namely, Auger and resonance tunneling neutralizations. In the previous calculations, the dispersion of the surface plasmons that arise from the neutralization of an ion scattered from a metal surface was not taken into account. In the present work dispersion was considered by allowing the surface plasmon frequency to be dependent on the wave vector. This makes the calculation valid for large values of the wave vector q. The calculations show that the orthogonalization correction to the neutralization rate is found to be important at small distances from the surface when applied to the scattering of protons from aluminum surface.

Introduction

It was recently discovered that the neutralization of ions scattered from metal surfaces by surface plasmons is an important electron transfer process especially for ions carrying high potential energy [1-3]. This mechanism was originally suggested by the author as a possible mechanism of neutralization in addition to the fully studied resonance and Auger neutralization mechanisms [4]. The experimentally measured time for neutralization of a proton scattered from a metal surface is about 10^{-15} seconds. For aluminum, for example, the energy of the surface plasmon is about 10.6 eV. This implies a period of oscillation of about 10^{-16} sec. It can be concluded that a collective response from the metal by exciting a surface plasmon is quiet possible. Also plasmons can

be excited because the valence electrons cannot respond instantaneously to screen the moving charge. Recently a good number of theoretical papers have been published on potential excitations of surface plasmon during surface neutralization [5-9]. The transition rate for the process is competitive with that for the other two processes [6,11-12]. It was seen that surface plasmon mediated ion neutralization at metal surfaces is very important. Since surface plasmons detection in experiment is indirect, relying on the observation of the ejected electrons released from plasmon decay [13], most of the work in this field is done theoretically. However, some experimental work that measures the energy distribution of electrons ejected by ion impact confirmed the reality of the mechanism of ion neutralization by surface plasmon excitation. For He ions on Mg the electron structure due to plasmon decay is more important than that of Auger neutralization [1].

In an earlier work the author suggested the mechanism of ion neutralization at metal surfaces by surface plasmon excitation. In that calculation a unitary transformation was used in order to able to work with the Hamiltonian of the interaction between the charged particle and the potential of the surface plasmons. In that calculation the dispersion of the surface plasmons was neglected, that is, it is assumed that the surface plasmons frequency is not a function of the wave vector. Although the value of energy is in good agreement with experimental values, this result is only good for small wave vector q. In other words all interactions are assumed instantaneous and the oscillations are entirely longitudinal in character. In other words the speed of light is infinite. For large q the transverse character of the wave must be taken into account. This is done by solving the full Maxwell's equations and doing the matching on the surface. The result is

$$\omega_s^2 = \tfrac{1}{2}\omega_p^2 + q^2 - (\frac{\omega_p^4}{4} + q^4)^{\frac{1}{2}} \qquad (1)$$

This form of dispersion relation can also be derived using the model in which the hydrodynamic Bloch equation

$$n_0 m \frac{\partial \vec{v}}{\partial t} = n_0\, e\, \vec{E} - m\, \beta^2\, \vec{v}\, n \qquad (2)$$

is used with Maxwell's equations. This form of dispersion will be taken into account in the calculation of the transition rate and neutral fraction in this paper.

Model

The second quantized Hamiltonian of our model is

$$H = \int d\bar{r}\, \psi^{\dagger}(\bar{r})\, T(\bar{r})\, \psi(\bar{r}) + \frac{1}{2}\int d\bar{r}\, d\bar{r}'\, \psi^{\dagger}(\bar{r})\, \psi^{\dagger}(\bar{r}')\, V'_{el-el}(\bar{r},\bar{r}')\, \psi(\bar{r}')\, \psi(\bar{r})$$

$$+ \int d\bar{r}\, \psi^{\dagger}(\bar{r})[V_b(\bar{r}) - |\bar{r}-\bar{s}|^{-1}]\psi(\bar{r}) + \sum \omega_s\, \hat{c}_q^{\dagger}\, \hat{c}_q + \int d\bar{r}\, \psi^{\dagger}(\bar{r})\, \Phi(\bar{r})\, \psi(\bar{r})$$

(3)

where s is the position of the proton and $\Phi(\bar{r})$ is the second quantized potential of the surface plasmons give by

$$\Phi(\bar{r}) = \sum g\, e^{-q|z|}(e^{i\bar{q}\cdot\bar{R}}\, \hat{c}_q + e^{-i\bar{q}\cdot\bar{R}}\, \hat{c}_q^{\dagger})$$

(4)

The coupling constant g is given by

$$g = [\frac{\pi\, \omega_s}{q\, A}]^{1/2}$$

(5)

where ω_s is now taken as a function of q as in equation (1). The interaction term in the Hamiltonian that gave rise to the mechanism being studied is given by the last term in (3), namely,

$$\int d\bar{r}\, \psi^{\dagger}(\bar{r})\, \Phi(\bar{r})\, \psi(\bar{r})$$

(6)

In the equations above $\hat{c}_q$ and $\hat{c}_q^{\dagger}$ are the surface plasmon annihilation and creation operators. The prime on the summation implies $q < q_c$ where q_c is the plasmon cutoff wave vector (maximum plasmon q). No volume plasmons are assumed since at low ion energies considered herein, the ion does not penetrate the metal surface and

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hence couples mainly to surface modes. In the Hamiltonian (3), $T(\vec{r})$ is the electron kinetic energy, $V_b(\vec{r})$ is the potential of the positive background, $V'_{el-el}(\vec{r},\vec{r}')$ the electron-electron interaction and $\hat{\psi}(\vec{r})$ and $\hat{\psi}^\dagger(\vec{r})$ are the electron annihilation and creation operators. The z-axis is perpendicular to the metal surface, the half plane $z<0$ constitutes the jellium metal and $z>0$ constitutes the exterior region. The electron position vector $\vec{r}$ in (3) may be either inside or outside the metal. This allows for tunneling of the electron. Throughout this paper atomic units will be used.

The electron field operator will be expanded in terms of the complete orthonormal set of orbitals and the corresponding annihilation operators $\hat{\psi}_k$ take the form :

$$\hat{\psi} = \sum \phi_k(\vec{r})\,\hat{\psi}_k \tag{7}$$

The metal electrons wave functions would be taken as solutions of the Schrodinger equation with a potential $V(z)$, which is constant inside and outside the metal with a step of height V_0 at the surface ($z=0$)

$$V(z) = V_0\,\vartheta(z) \tag{8}$$

Here $V_0 = F + W$ with F the Fermi energy and W the work function, and energies are measured from the bottom of the conduction band. The corresponding orthonormal eigenfunctions $\phi_k(\vec{r})$ are

$$\phi_k(\vec{r}) = \frac{1}{k_v\,(V)^{-1/2}}[e^{i\vec{k}\Box\vec{r}}\{(k'_z + ik_z)e^{(ik'_z z)} + (k'_z - ik_z)e^{(-ik'_z z)}\}]\quad z<0$$

$$= \frac{1}{k_v\,(V)^{-1/2}}[2\,k_z\,e^{i\vec{k}\Box\vec{r}}\,e^{-k_z z}\qquad\qquad z>0 \tag{9}$$

where V is the volume of the metal and k'_z , k'_z and k_v are defined by

$$(k'_z)^2 = 2 E_{k'} \qquad ; \ k_z^2 = 2 (V_0 - E_{k'}) \qquad ; \ k_v^2 + (k'_z)^2 = 2 V_0$$

with

$$E'_k = E_k - (1/2)^2 \vec{K}$$

E_k is the eigenvalue of ϕ_k and $\vec{K}$ is the component of $\vec{k}$ parallel to the surface. Within the conduction band $E_k < F$ one has

$$0 < E'_k < E_k < F < V_0$$

so that the wave functions are oscillatory inside the metal and decay into z direction outside. The same is true for the unfilled levels with $F < E_k < V_0 + \frac{1}{2} \vec{K}^2$. The wave functions with $E_k > V_0 + \frac{1}{2} \vec{K}^2$ oscillate with z outside as well as inside the metal. They also require a different normalization, but this is irrelevant since the plasmonic neutralization matrix element will involve conduction band electrons. Since the ϕ_k constitute a complete set, the bound atomic wave function ϕ_{at} in the final states can be expended in terms of them.

In order to overcome the problem of lack of orthogonality between the final atomic wave function and the initial conduction band wave function, a unitary transformation to a new representation wherein the atom is described by a state orthogonal to all conduction band states was used. The appropriate unitary transformation is of the form

$$\overset{\square}{U} = e^{(\frac{\pi}{2}) \overset{\square}{F}} \tag{10}$$

where the appropriate unitary operator is

$$\overset{\square}{F} = \sum (A_v^\dagger \psi_v - \psi_v^\dagger A_v) \tag{11}$$

And

$$A_\nu^\dagger = \int d\vec{r}\, \phi_\nu(\vec{r}-\vec{s})\, \psi_\nu^\dagger(\vec{r}) \qquad (12)$$

This unitary transformation rotates the Fock space by $\pi/2$ into a new space called the ideal space. The transformation being unitary, preserves the matrix elements and the Hermiticity of the Hamiltonian. The problem of the final atomic state being not orthogonal to the initial band function is solved automatically in this formalism. The final atomic states will be taken as the unperturbed ground $1s$ state of hydrogen

$$\phi_{1s} = \frac{1}{\sqrt{2}}\, e^{-|\vec{r}-\vec{s}|} \qquad (13)$$

Acting on the Hamiltonian in (3) a transformed Hamiltonian in which the matrix elements of the possible reactive channels are orthogonalized will be produced. Each matrix element will contain two parts, the first being the usual matrix element of the process, and the second is the orthogonalization term. In (12) $A_\nu^\dagger$ is the creation operator for an electron in a bound hydrogen orbital $\phi_\nu(\vec{r}-\vec{s})$ centered on the proton (position s) and ν stands for the atomic quantum numbers $\nu=(nlm)$.

The physical states on which the transformed Hamiltonian acts are of the form

$$|...\rangle = U^{-1}\, \psi^\dagger(\vec{r}) \qquad (14)$$

is any standard Fock state represented in terms of electron $|...\rangle$ where $|o\rangle$ acting on the vacuum state $\psi^\dagger(\vec{r})$ creation operators

Using (10-12) and making use of the commutation rules of $\hat{\psi}_\nu$ and , $\hat{\psi}(\vec{r})$ the electron field operators transform as follows

$$\hat{U}^{-1}\hat{\psi}(\vec{r})\hat{U} = \hat{\psi}(\vec{r}) - \int d\vec{r}' \Delta(\vec{r}-\vec{s},\vec{r}'-\vec{s})\hat{\psi}(\vec{r}') + \sum \phi_\nu(\vec{r}-\vec{s})\hat{\psi}_\nu \qquad (15)$$

where $\Delta(\vec{r}-\vec{s},\vec{r}'-\vec{s})$ is the hydrogen bound state kernel

$$\Delta(\vec{r}-\vec{s},\vec{r}'-\vec{s}) = \sum \phi_\nu(\vec{r}-\vec{s})\phi_\nu^*(\vec{r}'-\vec{s}) \qquad (16)$$

Transforming the Hamiltonian in (3) using the unitary transformation in (10), it can be seen that all channels of scattering are represented; including those of reactive scattering that are sought. Transforming the last term in the Hamiltonian, the surface plasmon-mediated ion neutralization channel would arise.

The transformed term takes the form

$$\int d\vec{r}\, \hat{U}^{-1}\hat{\psi}^\dagger(\vec{r})\hat{\Phi}(\vec{r})\hat{\psi}(\vec{r})\hat{U} \qquad (17)$$

This can be manipulated by inserting an identity operator $\hat{U}^{-1}\hat{U}$ in between the operators in this way

$$\int d\vec{r}\, \hat{U}^{-1}\hat{\psi}^\dagger(\vec{r})\hat{U}\hat{U}^{-1}\hat{\Phi}(\vec{r})\hat{U}\hat{U}^{-1}\hat{\psi}^\dagger(\vec{r})\hat{U} \qquad (18)$$

The terms take the form

$$\hat{T}_{11} = \sum \int d\vec{r}\, \phi_\nu^*(\vec{r}-\vec{s})\hat{\psi}_\nu^\dagger\, \hat{\Phi}(\vec{r})\, \hat{\psi}(\vec{r}) \qquad (19)$$

$$\hat{T}_{12} = -\sum \iint d\vec{r}\,d\vec{r}' \, \phi_\nu^*(\vec{r}-\vec{s})\,\Delta(\vec{r}-\vec{s},\vec{r}'-\vec{s})\,\hat{\psi}_\nu^\dagger \, \hat{\Phi}(\vec{r})\,\hat{\psi}(\vec{r}) \qquad (20)$$

Inserting (7) in the two terms $\hat{T}_{11}$, $\hat{T}_{12}$ one finds the following expressions for the perturbation $\hat{H}_{in}$ leading to the channel of neutralization. For surface plasmon-mediated neutralization

$$\hat{H}_{in} = \sum (\nu,q|H_3|k)\,\hat{\psi}_\nu^\dagger \, \hat{c}_q^\dagger \, \hat{\psi}_k \qquad (21)$$

where the matrix elements are given by

$$(\nu,q|H_3|k) = g \int d\vec{r}\, \phi_\nu^*(\vec{r}-\vec{s})\,\phi_k(\vec{r})\,e^{-q|z|}\,e^{-i\vec{q}\Box\vec{r}}$$
$$\qquad\qquad - \iint d\vec{r}\,d\vec{r}'\, \phi_\nu^*(\vec{r}-\vec{s})\,\Delta^*(\vec{r}-\vec{s},\vec{r}'-\vec{s})\,\phi_k(\vec{r}')\,e^{-q|z|}\,e^{-i\vec{q}\Box\vec{r}} \qquad (22)$$

This form of the matrix elements shows clearly the advantage of using the unitary transformation. It includes (the second term) an orthogonalization term that comes out automatically with the theory. The orthogonalization term takes care of orthogonalizing the metal orbitals to all bound atomic orbitals. The matrix elements in (22) are corrected forms of the Born approximation to the exact T-matrix elements for the scattering process, and due to the inclusion of the orthogonalization term it is a better approximation. This term was found to be important in the ion neutralization at surfaces [4,11-12]. In this work it has the corrected form for the surface plasmon frequency, which is dependent on the wave vector q.

Calculations

The evaluation of the matrix elements will be carried out using (22). The transition rate of the scattering p is given by

$$P = 2\pi \sum \left|(1s|H_{in}|k)^2\right| \delta(\tfrac{1}{2}k^2 - E(1s)) \qquad (23)$$

where the final hydrogen state has been taken to be the $1s$ state. The prime on the summation sign indicates the restriction that the k sum is over the interior of the filled Fermi sea only. The matrix elements M for the neutralization channel are now taken from the actual evaluations, thus giving

$$P = 2\pi \sum |M|^2 \, \delta(\tfrac{1}{2}k^2 - E(1s)) \tag{24}$$

Changing the sum into an integral we get

$$P = \frac{4\pi V}{(2\pi)^3} \int d\vec{k} \, |M|^2 \, \delta(\varepsilon_i - \varepsilon_f) \tag{25}$$

Here $\varepsilon_i = \tfrac{1}{2}k^2$ is the metal electron energy and $\varepsilon_f = E(1s)$ is the atomic electron energy, V is the volume and 2 is for the double spin of the electron.

$$P(s) = \frac{V}{2\pi^2} \iint d\vec{K} dk_z \, \vec{K} |M|^2 \, \delta(\varepsilon_i - \varepsilon_f) \tag{26}$$

The delta function is used now to evaluate the integral over $\vec{K}$ where $\vec{K}$ is the component of $\vec{k}$ parallel to the surface.

$$P(s) = \frac{V}{2\pi^2} \int dk_z \, |M|^2 \tag{27}$$

with the matrix element evaluated at

$$\tfrac{1}{2}\vec{K}^2 = \varepsilon_f - \tfrac{1}{2}k_0^2 = E(1s) + \omega_s - \tfrac{1}{2}k_z^2 \tag{28}$$

It should be stated that the atomic energies are shifted upward by V since energies are measured from the bottom of the conduction band. Hence the value of $E(1s)$ should be given by

$$E(1s) = V_0 - 1/2 = F + W - 1/2 \qquad (29)$$

To calculate the neutral fraction we will assume that the proton follows a classical trajectory. This is an acceptable approximation, since protons of low energies for which the de Broglie wave length is small compared to the atomic dimensions are considered here [14]. As in all earlier calculations of the neutral fraction of low energy ions scattered from surfaces the following assumptions will be made. First, it is assumed that the particle follows a straight classical path such that the perpendicular distance from the surface s is simply given by $s = v_\perp t$. Second, specular reflection will be assumed and that the perpendicular velocity $v_\perp$ is constant up to the point of reflection where it changes its direction. The total neutralization probability can then be obtained by integrating over this orbit. Following previous work [4,11-12], the number $N(s)$ of un-neutralized incident protons at distance s from the surface was calculated using the rate equation

$$\frac{dN(s)}{ds} = -\frac{P(s)N(s)}{v_\perp(s)} \qquad (30)$$

where $v_\perp$ is the perpendicular component of the ion velocity; and $P(s)$ is the neutralizing rate . We will assume constant V and specular reflection to get

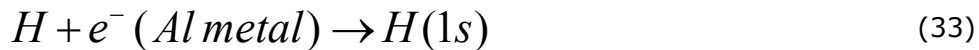$$N(s) = N_0 \exp[-\tfrac{1}{v_\perp} \int P(s)\,ds] \qquad (31)$$

The neutral fraction will be given by

$$f^0 = 1 - \frac{N(0)}{N_0} = 1 - \exp[-\tfrac{2}{v_\perp} \int P(s)\,ds] \qquad (32)$$

The 2 in the exponential comes from the assumption of specular reflection.

Results and Discussion

To apply the theory developed here we assume the scattering system

$$H + e^- \, (Al\,metal) \rightarrow H(1s) \tag{33}$$

The aluminum is chosen because it best satisfies the assumptions made in the theory. First it can be well approximated by a jellium model. Second its Fermi surface is very close to the free electron surface for a face centered cubic monatomic Bravais lattice with three conduction electrons per atom. Third surface plasmons are well defined for aluminum and their existence has been demonstrated experimentally. Fourth the existence of experimental work on this system [15] in addition to the theoretical work where orthogonalization is not taken into account [16-19].

The parameters used for aluminum are: 0.9261 for the Fermi wave vector k, 0.5862 for the surface potential V. The ground state in the hydrogen atom $H(1s)$ lies energetically within the conduction band of aluminum. All other states lie above the Fermi level. This makes Auger and resonant tunneling possible neutralization mechanisms for this system. Also, experimental and theoretical (not including orthogonalization term in the matrix elements) calculations on this system are available for comparison with the theory of this work [20-21].

Equation (25) is used to calculate the transition rate P as a function of the distance s of the proton from the surface. The integration over k is calculated numerically. All other calculations are done analytically using the inverse Fourier transform integrals and making use of the calculus of residues [4,11-12]. Figure (1) shows the neutralization rate P as a function of the distance s from the surface (fine curve for the case when dispersion is included while the thick curve represents the case with no dispersion). From the figure it is seen that the dispersion

correction is important, decreasing the transition rate by one order of magnitude. The decrease can be interpreted as a result of the neglect of the transverse oscillations of the surface plasmons. This is acceptable only for small wave vectors q. And
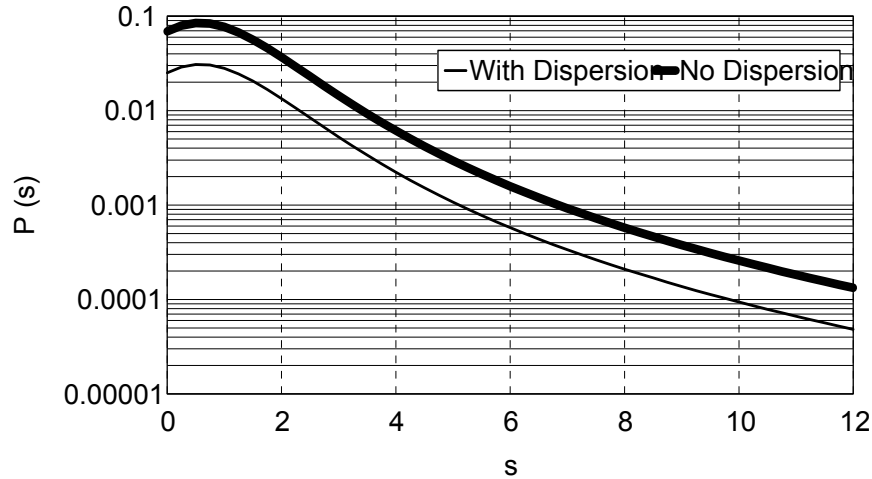

Figure (1): Transition rate P as a function of the distance s from the metal surface for a proton scattered from aluminum surface.
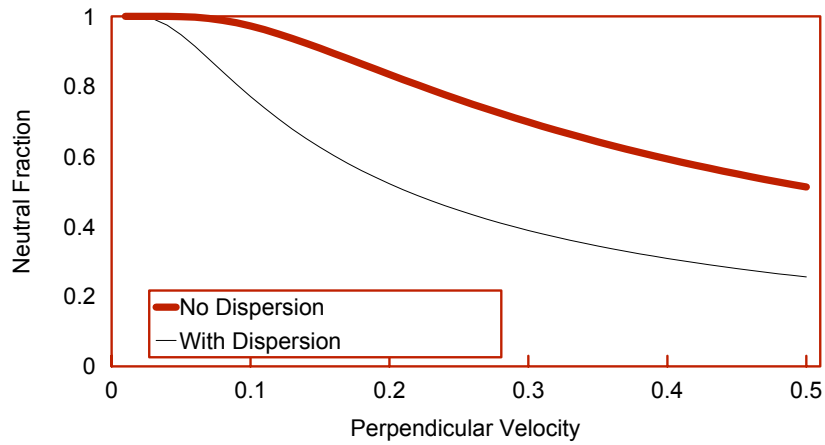

Figure (2): The neutral fraction f^0 for a proton scattered from aluminum surface as a function of the perpendicular velocity $v_\perp$ of the proton.

this process of ion neutralization -surface plasmon mediated- is to be compared with other competing ion neutralization processes. Full consideration of all cases should be taken into account. Figure (2) shows the results for the calculations for the neutral fractions f^0 defined in equation (32). This is also decreased due to the inclusion of the dispersion of the surface plasmonic frequency in the wave vector q .

Acknowledgement

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تم سابقا اقتراح إجراء جديد يتم فيه تعادل الأيونات عند سطوح المعادن. ﰲ هذا الأجراء يتم تعادل الأيون عند تشتته من سطح المعدن عن طريق تحفيز بلازمون سطحي. و قد تم بيان أن لهذا الأجراء احتمالية مقاربة للإجراءين الآخرين و هما تعادل أوجيه و التعادل بالرنين النفقي. ﰲ الحسابات السابقة لم يتم الأخذ ﰲ الاعتبار اعتماد التردد الزاوي ω للبلازمونات السطحية على المتجه الموجي k . ﰲ هذه الورقة البحثية يتم أخذ هذا الأمر ﰲ الاعتبار و ذلك بالسماح للتردد الزاوي ω للبلازمون السطحي بالاعتماد على المتجه الموجي k . و هذا الأمر يجعل الحسابات النظرية صحيحه للقيم الكبيرة من المتجه الموجي . أظهرت النتائج أن تصحيحات التعامد المأخوذ ﰲ الاعتبار فيها اعتماد التردد الزاوي على المتجه الموجي تكون مهمة للقيم الصغيرة لبعد الأيون عن سطح المعدن ﰲ حال تطبيق هذا الموضوع على تشتت أيون من سطح معدن الألمنيوم.