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In–out asymmetry and interference effects in plasmon excitation by swift charged particles traversing a surface

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We investigate the excitation of plasmons by a fast charged particle moving in the vicinity or traversing the surface of a solid along an arbitrary trajectory. We use both quantum-mechanical and semiclassical dielectric formulations to study how the particle couples with the bulk and surface excitations. We pay special attention to the differences and similarities between incoming and outgoing trajectories, finding some novel oscillatory structures that can be ascribed to an interference effect between direct and reflected plasmon excitations. Copyright © 2013 John Wiley & Sons, Ltd.

Keywords: in–out asymmetry; plasmon excitation; interference effects

Introduction

It is very well known that one of the most relevant processes in the interaction of swift charged particles with condensed matter is the phenomenon of plasmon excitations, both at the surface and in the bulk of the material [1,2]. These collective electronic excitations are of interest in studies of electron and ion interactions with surfaces, thin foils and small solid samples, even of nanoscopic dimensions, and in reflection electron energy loss spectroscopy, among others.

In previous studies, we have considered in detail the interrelations between external charged particles with materials. We deal with the semiclassical dielectric model for the external charged particle, using the plasmon pole approximation and the quantum description based on Hamiltonian model, for bulk and surface plasmon excitations in the material [3,4]. The continued development in high-resolution spectroscopy [5,6] maintains the interest in these processes; in particular, Ding [7] has made a complete analysis of the processes taking place for a charged particle leaving a solid ([7]). More recently, Salvat-Pujol and Werner [8] have analyzed different trajectories of a particle traversing a solid surface, focusing their work in the calculation of the inelastic mean free path.

The purpose of this work is to describe the energy loss due to the excitation of plasmons by external charged particles, analyzing in detail the asymmetries rising for incoming and outgoing trajectories. In Theoretical Description section, we give a brief account of the method used for the calculation of the plasmon excitations in macroscopic systems. In Bulk Contribution section, we analyze the results for incoming and outgoing trajectories at normal incidence in the case of a planar surface, and compare these results. In the Conclusions section, we discuss the main features of the results and comment on some perspectives for further research.

Theoretical description

The potential induced in a solid with the dielectric function $\varepsilon(q,\omega)$ located on the semi-space $z<0$, by a particle of charge Z moving along an arbitrary trajectory $\mathbf{R}=\mathbf{R}(t)$ is given by [4]

$$\phi^{ind}(\mathbf{r},\omega)=\frac{Z}{2\pi}\int\frac{d\mathbf{q}}{q}\int_{-\infty}^{\infty}dt' e^{i\omega t'}e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{R}')}\left[\frac{1-\varepsilon(q,\omega)}{1+\varepsilon(q,\omega)}e^{-q\left(|\mathbf{r}\cdot\hat{\mathbf{z}}|+|\mathbf{R}'\cdot\hat{\mathbf{z}}|\right)}+\frac{1-\varepsilon(q,\omega)}{\varepsilon(q,\omega)}\Theta(-\mathbf{r}\cdot\hat{\mathbf{z}})\Theta(-\mathbf{R}'\cdot\hat{\mathbf{z}})\times\left(e^{-q|(\mathbf{r}-\mathbf{R}')\cdot\hat{\mathbf{z}}|}-e^{-q|(\mathbf{r}+\mathbf{R}')\cdot\hat{\mathbf{z}}|}\right)\right] \tag{1}$$

where $\mathbf{R}'=\mathbf{R}(t')$, $\hat{\mathbf{z}}$ is the unit vector normal to the surface; ω and $\mathbf{q}$ are the usual time and space Fourier variables; $\Theta(x)$ is the Heaviside function. We recover the time dependency of the induced potential by means of the Fourier transform,

$$\phi^{ind}(\mathbf{r},t)=\frac{Z}{(2\pi)^2}\int\frac{d\mathbf{q}}{q}\int_{-\infty}^{\infty}dt' e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{R}')}\left[W_S(t'-t)e^{-q\left(|\mathbf{r}\cdot\hat{\mathbf{z}}|+|\mathbf{R}'\cdot\hat{\mathbf{z}}|\right)}+W_B(t'-t)\Theta(-\mathbf{r}\cdot\hat{\mathbf{z}})\Theta(-\mathbf{R}'\cdot\hat{\mathbf{z}})\left(e^{-q|(\mathbf{r}-\mathbf{R}')\cdot\hat{\mathbf{z}}|}-e^{-q|(\mathbf{r}+\mathbf{R}')\cdot\hat{\mathbf{z}}|}\right)\right] \tag{2}$$

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where we have defined

$$W_S(t) = \int_{-\infty}^{\infty} d\omega e^{i\omega t} \frac{1-\varepsilon(q,\omega)}{1+\varepsilon(q,\omega)} \quad \text{and}$$
$$W_B(t) = \int_{-\infty}^{\infty} d\omega e^{i\omega t} \frac{1-\varepsilon(q,\omega)}{\varepsilon(q,\omega)}$$

The energy lost by the particle per unit time is evaluated as

$$\frac{dW}{dt} = -Z \frac{\partial \phi^{ind}(\mathbf{r},t)}{\partial t}\bigg|_{\mathbf{r}=\mathbf{R}(t)} = iZ\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \omega e^{-i\omega t} \phi^{ind}(\mathbf{r},\omega) \tag{3}$$

which in conjunction with the particle velocity v determines the stopping power. Now, defining $\dot{W} = \partial W/\partial t$, namely

$$\dot{W}_S(t) = i\int_{-\infty}^{\infty} d\omega\, \omega e^{i\omega t} \frac{1-\varepsilon(q,\omega)}{1+\varepsilon(q,\omega)} \quad \text{and}$$
$$\dot{W}_B(t) = i\int_{-\infty}^{\infty} d\omega\, \omega e^{i\omega t} \frac{1-\varepsilon(q,\omega)}{\varepsilon(q,\omega)}$$

we obtain

$$\frac{dW}{dt} = \frac{Z^2}{(2\pi)^2}\int \frac{d\mathbf{q}}{q}\int_{-\infty}^{\infty} dt' e^{i\mathbf{q}\cdot(\mathbf{R}-\mathbf{R}')}\Big[\dot{W}_S(t'-t)e^{-q(|\mathbf{R}'\cdot\hat{\mathbf{z}}|+|\mathbf{R}\cdot\hat{\mathbf{z}}|)}$$
$$+\dot{W}_B(t'-t)\Theta(-\mathbf{R}\cdot\hat{\mathbf{z}})\Theta(-\mathbf{R}'\cdot\hat{\mathbf{z}})$$
$$\left(e^{-q|(\mathbf{R}-\mathbf{R}')\cdot\hat{\mathbf{z}}|} - e^{-q|(\mathbf{R}+\mathbf{R}')\cdot\hat{\mathbf{z}}|}\right)\Big] \tag{4}$$

These expressions can be analytically integrated only for some simple models of the dielectric function. Let us consider – for instance – the classical frequency-dependent dielectric function [9]

$$\varepsilon(q,\omega) = 1 - \frac{\omega_P^2}{\omega(\omega + i\gamma)} \tag{5}$$

as an instructive approximation. The parameter $\omega_P = \sqrt{3/r_S^3}$ is the plasma frequency of the medium in atomic units, with r_S the one-electron radius in the electron gas describing the solid; γ is an effective damping rate, which accounts for the finite lifetime of the plasmons. Inserting this model dielectric function into the previous equations, we obtain

$$W_B(t) = \int_{-\infty}^{\infty} d\omega e^{i\omega t}\frac{1-\varepsilon(q,\omega)}{\varepsilon(q,\omega)}$$
$$= \frac{\omega_P}{2}\int_{-\infty}^{\infty} d\omega \left(\frac{e^{i\omega t}}{\omega + i\gamma/2 - \omega_P} - \frac{e^{i\omega t}}{\omega + i\gamma/2 + \omega_P}\right)$$
$$= -\frac{\omega_P}{2} 2\pi i\left(-\Theta(-t)e^{i(\omega_P - i\gamma/2)t} + \Theta(-t)e^{i(-\omega_P - i\gamma/2)t}\right)$$
$$= -2\pi\omega_P\Theta(-t)e^{\gamma t/2}\sin(\omega_P t), \tag{6}$$

and

$$W_S(t) = -2\pi\omega_S\Theta(t-t')e^{\gamma t/2}sin(\omega_S t)$$

Thus, we obtain

$$\phi^{ind}(\mathbf{r},t) = \frac{Z}{2\pi}\int \frac{d\mathbf{q}}{q}\int_{-\infty}^{t} dt' e^{i\mathbf{q}\cdot(\mathbf{r}-\mathbf{R}')}e^{-\gamma(t-t')/2}$$
$$\left[\omega_S \sin[\omega_S(t-t')]e^{-q(|\mathbf{r}\cdot\hat{\mathbf{z}}|+|\mathbf{R}'\cdot\hat{\mathbf{z}}|)}\right.$$
$$+\omega_P \sin[\omega_P(t-t')]\Theta(-\mathbf{r}\cdot\hat{\mathbf{z}})\Theta(-\mathbf{R}'\cdot\hat{\mathbf{z}})$$
$$\left.\left(e^{-q|(\mathbf{r}-\mathbf{R}')\cdot\hat{\mathbf{z}}|} - e^{-q|(\mathbf{r}+\mathbf{R}')\cdot\hat{\mathbf{z}}|}\right)\right] \tag{7}$$

Now, we integrate over the orientation of the vector $\mathbf{q}$, with $\boldsymbol{\rho}$ as the projection of the position $\mathbf{r}$ onto the interface plane (i.e. in a direction perpendicular to $\hat{\mathbf{z}}$). Similarly, $\mathbf{R}_{||}$ and $R_\perp$ are the projections of the trajectory $\mathbf{R}$ on directions parallel and perpendicular to the surface. We have also defined the usual cutoff in the plasma response function,

$$q_c^{B,S} = \omega_{P,S}\sqrt{\frac{1}{v_F^2} - \frac{1}{v^2}}$$

where $v_F = (9\pi/4)^{1/3}/r_s = (3\sqrt{\pi}\omega_P/2)^{2/3}$ is the Fermi velocity, and v is the mean velocity of the projectile. The integral in the orientation of $\mathbf{q}$ can be analytically evaluated in terms of the Bessel function of order 0 ($J_o(x)$),

$$\phi^{ind}(\mathbf{r},t) = Z\int_0^{q_c} dq\int_{-\infty}^{t} dt' e^{-\gamma(t-t')/2}J_o(q|\boldsymbol{\rho} - \mathbf{R}'_{||}|)$$
$$\Big[\omega_S \sin[\omega_S(t-t')]e^{-q(|z|+|R'_\perp|)}$$
$$+\omega_P\Theta(-z)\Theta(-R'_\perp)\sin[\omega_P(t-t')]$$
$$\left(e^{-q|z-R'_\perp|} - e^{-q|z+R'_\perp|}\right)\Big] \tag{8}$$

A similar calculation leads to the following expression for the energy loss per unit time:

$$\frac{dW}{dt} = \left(\frac{dW}{dt}\right)_B + \left(\frac{dW}{dt}\right)_S \tag{9}$$

with

$$\left(\frac{dW}{dt}\right)_S = Z^2\omega_S^2\int_0^{q_c} dq\int_{-\infty}^{t} dt' e^{-\gamma(t-t')/2}$$
$$\cos[\omega_S(t-t')]J_o(q|\mathbf{R}_{||} - \mathbf{R}'_{||}|)e^{-q(|\boldsymbol{R}_\perp|+|R'_\perp|)} \tag{10}$$

and

$$\left(\frac{dW}{dt}\right)_B = Z^2\omega_P^2\Theta(-R_\perp)\int_0^{q_c} dq\int_{-\infty}^{t} dt' e^{-\gamma(t-t')/2}$$
$$\cos[\omega_P(t-t')]J_o(q|\mathbf{R}_{||} - \mathbf{R}'_{||}|) \tag{11}$$

As a first illustrative example, let us review the case of a particle of charge Z that crosses the surface in a normal trajectory, $\mathbf{R}(t') = v\hat{\mathbf{z}}t'$, which remains undisturbed by the plasmon excitation events. This approximation holds for sufficiently large kinetic energies, namely $mv^2/2 \gg \hbar\omega_p$. Both cases, of a particle entering ($v < 0$) and leaving ($v > 0$) the solid are considered, as illustrated in Fig. 1.

We obtain the following contributions to the induced potential

$$\phi^{ind}(\mathbf{r},t) = \phi_S^{ind}(\mathbf{r},t) + \phi_B^{ind}(\mathbf{r},t) \tag{12}$$

where

$$\phi_S^{ind}(\mathbf{r},t) = Z\omega_S\int_{-\infty}^{t} dt' e^{-\gamma(t-t')/2}$$
$$sin[\omega_S(t-t')]\int_0^{q_c} dq J_o(q\rho)e^{-q(|z|+|v||t'|)} \tag{13}$$

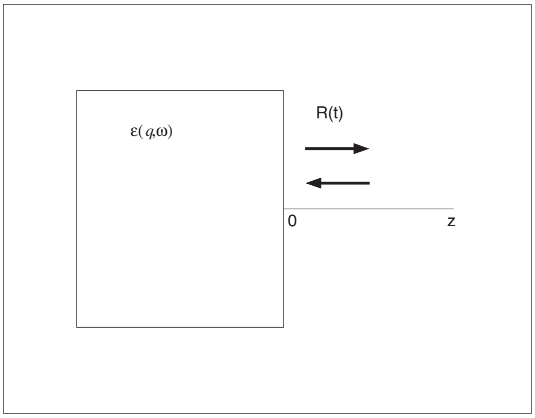

Figure 1. Illustration of the studied system, for two perpendicular asymmetric external trajectories, moving away and toward the solid–vacuum interface, respectively.

and

$$\phi_B^{ind}(\mathbf{r},t) = Z\omega_P\Theta(-z)\int_{-\infty}^{t} dt' e^{-\gamma(t-t')/2}\sin\left[\omega_P(t-t')\right]\Theta(-vt') \times \int_0^{q_c} dqJ_o(q\rho)\left(e^{-q|z-vt'|} - e^{-q|z+vt'|}\right) \quad (14)$$

ρ is the projection of the position $\mathbf{r}$ in a direction perpendicular to $\hat{\mathbf{z}}$. We clearly see that the surface contribution is independent of the sign of v. Thus, it is the same for incoming and outgoing trajectories.

The case of the bulk contribution is far from being so evident. Let us change the integration variable in order to write the induced potential in the following way:

$$\phi_B^{ind}(\mathbf{r},t) = Z\omega_P\Theta(-z)\int_0^{\infty} d\tau e^{-\gamma\tau/2}\sin(\omega_P\tau)\Theta(-v(t-\tau)) \times \int_0^{q_c} dqJ_o(q\rho)\left(e^{-q|z-v(t-\tau)|} - e^{-q|z+v(t-\tau)|}\right)$$

The terms of the interface in the brackets contain two components. We see that, except for the Heaviside function, one term can be changed into the other by replacing $z \leftrightarrow -z$, namely

$$\phi_B^{ind}(\mathbf{r},t) = \Theta(-z)\left[\tilde{\phi}_B^{ind}(\mathbf{r},t) - \tilde{\phi}_B^{ind}(\mathbf{r}-2z\hat{\mathbf{z}},t)\right]$$

where

$$\tilde{\phi}_B^{ind}(\mathbf{r},t) = Z\omega_P\int_0^{\infty} d\tau e^{-\gamma\tau/2}\sin(\omega_P\tau) \Theta[v(\tau-t)]\int_0^{q_c} dqJ_o(q\rho)e^{-q|(z-vt)+v\tau|}$$

is the potential induced by a charged particle of charge Z and velocity $\mathbf{v} = v\hat{\mathbf{z}}$ in an infinite medium. Now, because this potential is not symmetric (or even antisymmetric) in the velocity v, the potential is not the same for incoming and outgoing trajectories. In Fig. 2, we show these features for an electron crossing an aluminum–vacuum interface with velocity $v = 2$ a.u., at different positions along these trajectories.

Now we analyze the surface contribution,

$$\phi_S^{ind}(\mathbf{r},t) = Z\omega_S\int_0^{q_c} dqJ_o(q\rho)e^{-q|z|}\int_0^{\infty} d\tau e^{-\gamma\tau/2}\sin[\omega_S\tau]e^{-q|v||t-\tau|} \quad (15)$$

If the damping constant can be neglected ($\gamma = 0$), some simple algebra leads to

$$\phi_S^{ind}(\mathbf{r},t) = Z\int_0^{q_c} dqe^{-q|z|}J_o(q\rho)\frac{\omega_S}{(qv)^2+\omega_S^2} \left(\omega_S e^{-q|v||t|} + 2\Theta(t)q|v|\sin\omega_S t\right) \quad (16)$$

Because we are not particularly interested in the z-dependence of the induced surface potential, we shall usually particularize to the plane $z = 0$, namely

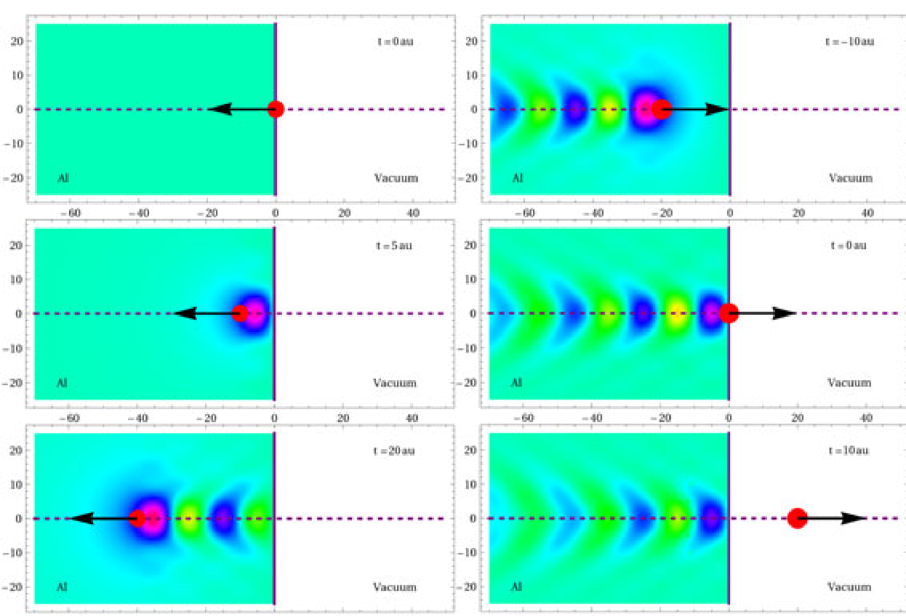

Figure 2. Induced potential for a charged particle traveling through an aluminum–vacuum interface with velocity $|v| = 2$ a.u., at different positions along incoming (left) and outgoing (right) trajectories. Lengths are given in atomic units.

$$\phi_S^{ind}(\rho,t) = Z\int_0^{q_c} \mathrm{d}qJ_o(q\rho)\frac{\omega_S}{(qv)^2+\omega_S^2}\left(\omega_S e^{-q|v||t|}+2\Theta(t)q|v|\sin\omega_S t\right) \quad (17)$$

that we can write as

$$\phi_S^{ind}(\rho,t) = \frac{Z\omega_S}{|v|}\left[G_o(\omega_S\rho/|v|,\omega_S t)+2\Theta(t)G_1(\omega_S\rho/|v|,0)\sin\omega_S t\right] \quad (18)$$

in terms of an auxiliary function,

$$G_n(u,x) = \int_0^{q_c|v|/\omega_S} J_o(uy)y^n\frac{e^{-xy}}{1+y^2}\mathrm{d}y \quad (19)$$

Starting from the previous induced potential, we can evaluate the surface contribution to the energy lost per unit time,

$$\begin{aligned}\left(\frac{\mathrm{d}W}{\mathrm{d}t}\right)_S &= -Z\frac{\partial\phi_S^{ind}}{\partial t}\bigg|_{\mathbf{r}=v\hat{z}t} \\ &= Z^2\omega_S\int_0^{q_c}\mathrm{d}qe^{-q|z|}J_o(q\rho)\frac{q|v|\omega_S}{(qv)^2+\omega_S^2}\left(\mathrm{Sgn}(t)e^{-q|v||t|}-2\Theta(t)\cos\omega_S t\right) \\ &= \frac{Z^2\omega_S^2}{|v|}\left[\mathrm{Sgn}(t)f_{11}(2\omega_S|t|)-2\Theta(t)f_{11}(\omega_S|t|)\cos(\omega_S t)\right]\end{aligned} \quad (20)$$

with

$$f_{nm}(x) = \int_0^{q_c|v|/\omega_S}\frac{y^n}{(1+y^2)^m}e^{-xy}\mathrm{d}y$$

Here, we have also defined the Sign function, Sgn (t) = 1 when $t \geq 0$ and Sgn (t) = − 1 when $t < 0$.

Bulk contribution

Let us first analyze the energy lost per unit time, given by Eqn (11) for a trajectory along the surface normal, $\mathbf{R}(t) = v\hat{\mathbf{z}}t$. We write

$$\left(\frac{\mathrm{d}W}{\mathrm{d}t}\right)_B = -Z^2\omega_P^2\Theta(-vt)\int_0^{q_c}\mathrm{d}q\int_{-\infty}^{t}\mathrm{d}t' e^{-\gamma(t-t')/2}\cos\left[\omega_P(t-t')\right] \times\Theta(-vt')\left(e^{-q|v|(t-t')}-e^{-q|v||t+t'|}\right) \quad (21)$$

Let us now consider the cases $v > 0$ and $v < 0$ separately. For $v < 0$ and $\gamma = 0$, we obtain

$$\left(\frac{\mathrm{d}W}{\mathrm{d}t}\right)_B = -Z^2\omega_P^2\Theta(t)\left(-2\int_0^{q_c}\mathrm{d}qe^{-q|v|t}\frac{q|v|\cos(\omega_P t)}{(qv)^2+\omega_P^2}+\int_0^{q_c}\mathrm{d}q\left(1+e^{-2q|v|t}\right)\frac{q|v|}{(qv)^2+\omega_P^2}\right) \quad (22)$$

We write this expression in terms of the auxiliary functions f_{11} as follows:

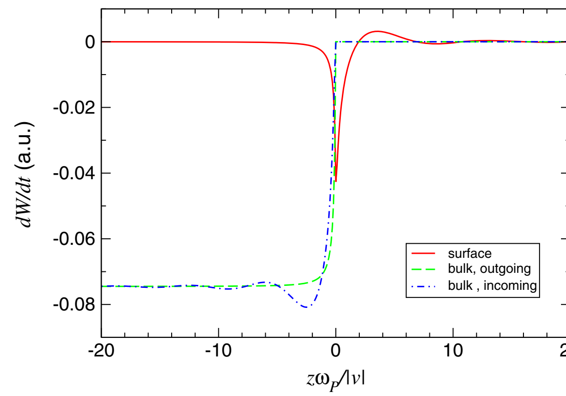

Figure 3. Energy lost per time unit by a charged particle traversing an interface with aluminum perpendicularly with velocity v = 10 au.

$$\left(\frac{\mathrm{d}W}{\mathrm{d}t}\right)_B = -\frac{Z^2\omega_P^2}{|v|}\Theta(t)\left[f_{11}(0)+f_{11}(2\omega_P t)-2f_{11}(\omega_P t)\cos(\omega_P t)\right] \quad (23)$$

Similarly, for the case $v > 0$ (and $\gamma = 0$), we obtain

$$\left(\frac{\mathrm{d}W}{\mathrm{d}t}\right)_B = -\frac{Z^2\omega_P^2}{|v|}\Theta(-t)\left[f_{11}(0)-f_{11}(2\omega_P|t|)\right] \quad (24)$$

We join both equations into a single one:

$$\left(\frac{\mathrm{d}W}{\mathrm{d}t}\right)_B = -\frac{Z^2\omega_P^2}{|v|}\Theta(-vt)\left[f_{11}(0)-\mathrm{Sgn}(v)f_{11}(2\omega_P|t|)-2\Theta(-v)f_{11}(\omega_P t)cos(\omega_P t)\right) \quad (25)$$

Figure 3 shows calculations of the surface and bulk contributions to the energy lost per unit time, both for incoming and outgoing trajectories of a particle traversing an aluminum–vacuum interface with velocity 10 a.u. We clearly see that, although the surface contribution is the same for both incoming and outgoing trajectories, the bulk contribution shows an oscillatory term that is only present for incoming trajectories.

Conclusions

In this work, detailed calculations show that the excitation of plasmons is influenced by changes in the trajectory of the electron beam, even in those that seem equivalent as incoming and outgoing normal trajectories. This result gives some novel oscillatory structures that can be ascribed to an interference effects between direct and reflected plasmon excitations.

This information about asymmetrical electron beam trajectories must be taken into account to calculate the average number of plasmon excitations and its phenomena of interference, an important aspect due to the increasing requirement of devices of reduced dimensions for their application in different areas. Comparison with experiments using reflection electron energy loss spectroscopy [10] is under way.

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