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Interference effects in the plasmon fields excited by a diatomic molecule

S. Segui [a,1], J.L. Gervasoni [a,b,*,1]

[a] Centro Atómico Bariloche (Comisión Nacional de Energía Atómica), Av. Bustillo 9500, 8400 San Carlos de Bariloche, Río Negro, Argentina
[b] Instituto Balseiro (Universidad Nacional de Cuyo and Comisión Nacional de Energía Atómica), Argentina

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ABSTRACT

We study the fluctuations of the plasmon field associated to the wake potential generated by a dimer formed by two identical charged particles (such as two nuclei of the ionized hydrogen molecule) traveling through a semiinfinite dielectric medium. We use coherent states to describe bulk and surface plasmons as wave packets that raise fluctuations in the electronic density of the material. We analyze different configurations of interest, taking into account various trajectories and orientations of the dimer.

1. Introduction

Plasmon excitation constitutes one of the most relevant processes in the interaction of swift charged particles with solid systems. These excitations have been extensively studied for ions and electrons interacting with surfaces, thin films and nanoparticles [1–5]. Less abundant is the literature devoted to study the excitation of plasmons with structured projectiles, such as molecules traveling inside or in the proximity of a polarizable medium [6–9]. On the other hand, these interactions have regained interest due to the development of surface plasmon resonance techniques, where laser-excited surface plasmons are used to detect and analyze the composition of molecules or structures adsorbed on a metallic surface [10].

The purpose of this work is to study the effects on the plasmon field when the incident particle has a simple structure, as a diatomic molecule, using coherent states [11,12]. This formalism provides a very simple tool to analyze different quantities of interest by means of a hamiltonian formulation of the interaction of the plasmon field with the incident particle. We calculate the wake potential and the fluctuations in the electronic density $\Delta\rho$, and compare them with the results obtained for a single particle. We take into account different configurations, regarding the trajectory as well as the orientation of the molecular axis with respect to the movement direction.

The structure of the paper is as follows: in Section 2 we write the basic formulas used to describe the excitation of a plasmon field due to the passage of a swift dimer formed by two identical charged particles. In Section 3 we show calculations of the wake potential and the charge density fluctuations, for bulk and surface excitations in aluminum. Finally, we present some concluding remarks in Section 4.

2. Theoretical model: coherent states

We consider the dimer or molecule as formed by two point particles with charge Ze, separated by a given distance d and traveling along a rectilinear trajectory with constant velocity v. We neglect the interaction between the charges, while each charge interacts with the electron gas of the material. In this work we consider a semiinfinite solid with planar interface, but the method can be easily extended to other geometries. We also assume that the valence electrons of the bulk and the electrons at the surface interact independently with the external charges, generating the *bulk* and *surface* plasmons, each with their own characteristic frequencies $\omega_{\mathbf{k}}^{b,s}$.

We make use of the coherent states formulation to describe the interaction of the incident particles with the plasmon field. Within this frame, plasmon modes form a set of harmonic oscillators with characteristic frequencies ω_ν determined by the composition and geometry of the studied system [2,3] and described by an unperturbed hamiltonian H_0 in terms of creation and annihilation operators $a_{\mathbf{k}}^\dagger$, $a_{\mathbf{k}}$ and with $|n_{\mathbf{k}}\rangle$ spanning the possible states of the plasmon

* Corresponding author at: Centro Atómico Bariloche (Comisión Nacional de Energía Atómica), Av. Bustillo 9500, 8400 San Carlos de Bariloche, Río Negro, Argentina.
[1] Also member of the Consejo Nacional de Investigaciones Científicas y Técnicas (CONICET), Argentina.

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field with n plasmons in the mode $\mathbf{k}$. The interaction of the incident particles with the electron gas is given by

$$H_{int}(t) = \int d^3r \phi(\mathbf{r},t)\rho_{ext}(\mathbf{r},t) \quad (1)$$

where $\rho_{ext}(\mathbf{r},t)$ is the external charge density,

$$\rho_{ext}(\mathbf{r},t) = Ze(\delta(\mathbf{r}-\mathbf{R}_1(t)+\delta(\mathbf{r}-\mathbf{R}_2(t))),$$

with $\mathbf{R}_i$ the position of the ith particle, and $\phi(\mathbf{r},t)$ is the potential associated to the plasmon field, in terms of $a^\dagger_{\mathbf{k}}$ and $a_{\mathbf{k}}$. The time evolution of the field interacting with the external particles can be obtained within the interaction representation using the Schrödinger equation

$$i\hbar\frac{\partial \mid \Psi(t)\rangle}{\partial t} = H_{int} \mid \Psi(t)\rangle \rightarrow \mid \Psi(t)\rangle = e^{-i\sum_{\mathbf{k}}(I_{\mathbf{k}}(t)a_{\mathbf{k}}+I^*_{\mathbf{k}}(t)a^\dagger_{\mathbf{k}})} \mid \Psi(-\infty)\rangle \quad (2)$$

The coefficients $I_{\mathbf{k}}(t)$ depend on the trajectory of the incident particle, and $|\Psi(-\infty)\rangle$ is the initial state of the field, in terms of the eigenstates $|n_{\mathbf{k}}\rangle$; the states $\mid \Psi(t)\rangle$ constitute a set of *coherent states*, describing the plasmon field as a wavepacket that propagates in a classical electromagnetic way. This formalism allows us to calculate the wake potential as:

$$\phi_W(\mathbf{r},t) = \langle\Psi(t) \mid \phi \mid \Psi(t)\rangle, \quad (3)$$

as well as the variation of electronic density $\Delta\rho$ as

$$\Delta\rho = -\frac{1}{4\pi}\langle\Psi(t) \mid \nabla^2\phi \mid \Psi(t)\rangle \quad (4)$$

and other quantities of interest such as the energy lost by the incident particles dW/dt.

3. Results

3.1. Bulk excitations

First, we consider bulk excitations generated by a dimer traveling along a rectilinear trajectory with constant velocity $\mathbf{v} = v\hat{z}$, with its axis forming an arbitrary angle with z-axis. We use the plasmon pole approximation to describe the dielectric properties of the material, with a hydrodynamic dispersion relation for $\omega_{\mathbf{k}} = \omega_p^2 + \beta k^2 + \hbar^2k^4/2m$. In this expression, ω_P is the plasma frequency of the electron gas, and $\beta = \sqrt{3/5}v_f$ is a constant related to the Fermi velocity v_f of electrons in this gas. The wake potential calculated from Eq. (3) yields

$$\phi^b_W(\mathbf{r},t) = \frac{Ze\omega_P^2}{2\pi^2}\Re\left[\int\frac{d^3k}{k^2\omega_k}\cos(\mathbf{k}\cdot\mathbf{r}_0)\frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{v}t)}e^{\eta t}}{\mathbf{k}\cdot\mathbf{v}-\omega_k t+i\eta}\right] = 2\cos(\mathbf{k}\cdot\mathbf{r}_0)\tilde{\phi}^b_w(\mathbf{r},t) \quad (5)$$

where η is a small damping constant and $\mathbf{r}_0$ is the position of one of the particles constituting the dimer with respect to the center of mass (CM). $\tilde{\phi}^b_w$ corresponds to the wake potential generated by a single charge Ze following the trajectory of the center of mass.

Fig. 1 shows the variation of the wake potential generated by the dimer along the z axis for various internuclear separations d, and two orientations of the dimer's axis (perpendicular and parallel to the velocity), in a homogeneous aluminum medium ($\omega_P = 0.55$ a.u.). For distances sufficiently small (say $d < 3$ a.u. for the parameters illustrated here), the differences with respect to the one-particle case are insignificant; for larger values of d, interference effects get important and modify the wake structure, specially in the adjacencies of the incident dimer.

Fig. 2 shows the wake potential on a plane containing the trajectory and the dimer axis, for the case of parallel and perpendicular orientations, and a interatomic distance of $d = 5$ a.u. These calculations are made at $t = 0$, and can be viewed as the potential accompanying the dimer in a suitable coordinates system.

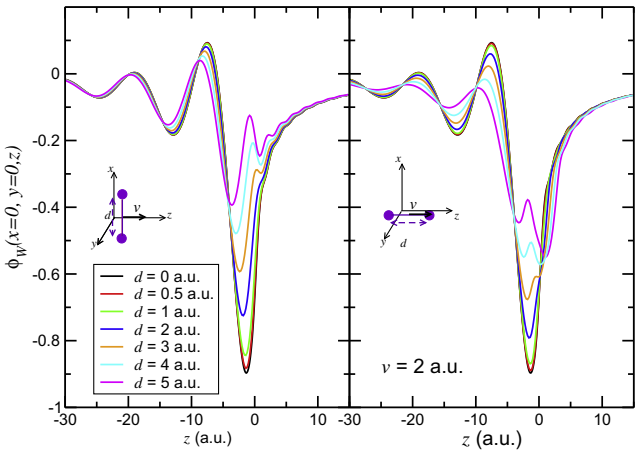

Fig. 1. Wake potential due to bulk excitations for various interatomic distances, for a dimer traveling with velocity $v = 2$ a.u. and its axis perpendicular (left) and parallel (right) to the trajectory.

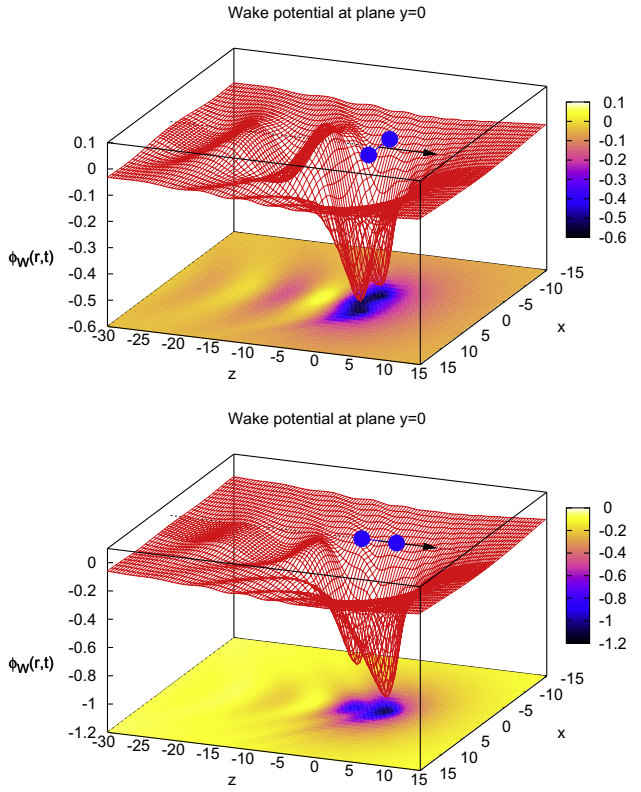

Fig. 2. Wake potential on the plane $y = 0$ for a dimer traveling with velocity $v = 2$ a.u. and different orientations of its axis with respect to the velocity.

3.2. Surface excitations

Now, we turn to the case of a semiinfinite medium ($z < 0$), with a planar surface at $z = 0$. In this work, we consider only the case of the dimer traveling parallel to the surface, with a fixed distance z_0 from the CM (now we take $\mathbf{v} = v\hat{x}$). In this configuration, different orientations can be of interest; as an illustrative example, we calculate the wake potential due to the passage of the dimer with its internuclear axis on a plane parallel to the surface too. We solve

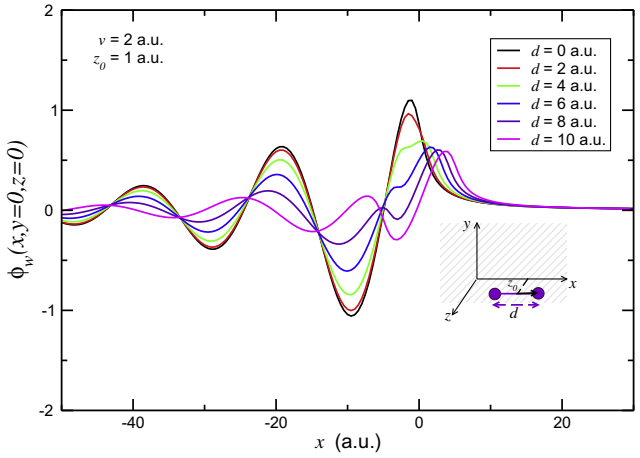

Fig. 3. Variation of the wake potential on the surface ($z = 0$) with the interatomic distance for a particle traveling parallel to the surface ($z_0 = 1$ a.u.), with its axis parallel to **v**.

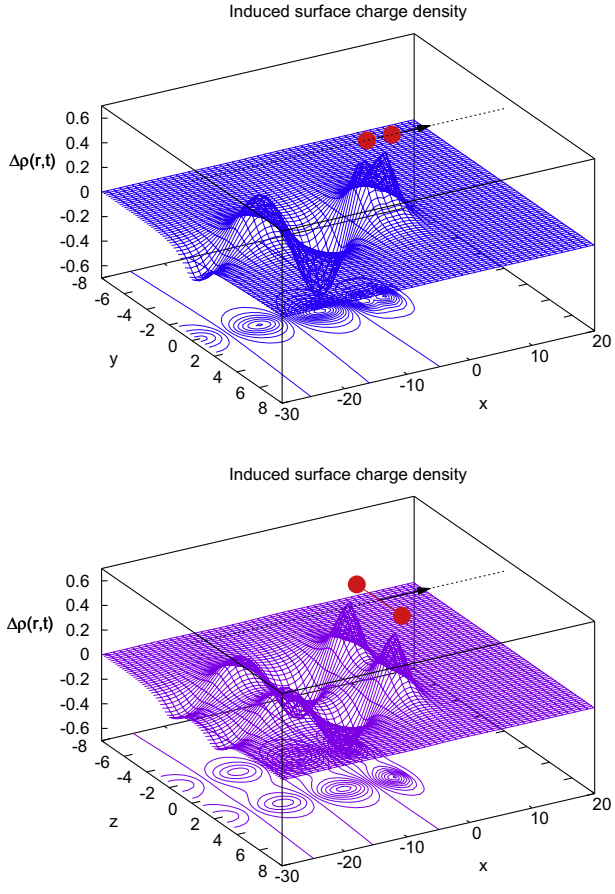

Fig. 4. Comparison of the induced charge density in the case of trajectories parallel to the surface, with the dimer axis parallel to the velocity (top) and perpendicular to it (bottom). The distance to the surface is $z_0 = 1$ a.u.; the velocity is $v = 2$ a.u.; the interatomic distance is $d = 4$ a.u.

the equations with a simpler dependence for the dispersion relation ($\omega_{\mathbf{k}} = \omega_S = \omega_P/\sqrt{2}$), and obtain the following expression for ϕ_W^s:

$$\phi_W^s(\mathbf{r},t) = \frac{Z}{4\pi} 2\omega_s e^{-\eta t} \times \Re\left\{ i \int \frac{d^2q}{q} e^{-qz_0} 2\cos(\mathbf{q}\cdot\mathbf{r}_0) \frac{e^{i\mathbf{q}\cdot(\mathbf{v}t-\mathbf{r})}}{\mathbf{q}\cdot\mathbf{v} - \omega_s + i\eta} \right\} \tag{6}$$

Fig. 3 shows the dependence of the wake potential due to surface excitations for different interatomic distances, along a line following the trajectory of the dimer on the interface. The distance from the trajectory to the surface is $z_0 = 1$ a.u., with its axis parallel to **v**. The interatomic distance for which differences from the case of a single charged particle start to be significant, varies with the distance between the trajectory and the surface.

The induced charge density is obtained from Eq. (4), which is easily evaluated when combined with the result of Eq. (6) for the wake potential. $\Delta\rho$ accompanies the wake potential on the surface and has approximately the same dependence on the distance to the surface and orientation of the incident molecule. Fig. 4 shows the induced charge density on the interface due to the passage of a dimer traveling on a rectilinear trajectory parallel to the surface, with its axis oriented parallel and perpendicular to the velocity. The distance to the surface is $z_0 = 1$ a.u. and the distance between the particles is $d = 4$ a.u.

4. Conclusions

We have applied the coherent states formalism to describe the plasmon excitation for a diatomic molecule with two identical charges interacting with a semiinfinite solid in different configurations. We have studied different quantities of interest, such as the wake potential and the charge density, and their dependence on various parameters of the system: interatomic distance, molecule's trajectory, etc. Within a first order of approximation, the calculations are useful to determine critical values of the parameters for which it is important to take the incident particle structure into account.

The coherent states formalism used to describe the interaction of the plasmon field with the incident charges has proved to be most adequate for a straightforward evaluation of different quantities of interest, providing an insight of the contribution of the different modes excited. The present results and their potential applications motivate us to go on with the study of new configurations and geometries of the surface.

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