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LASER MATTER INTERACTION AT GAS-SOLID INTERFACE: ELECTROMAGNETIC FIELDS FROM ELECTRON DENSITY

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Abstract. We developed a model giving the classical vector potential $\vec{A}$ of the electromagnetic field obtained from the unperturbed electron density at the gas-solid interface. The model uses a local dielectric function deduced from the experiment and varying with the laser energy. The derived potential was used to construct the laser-matter interaction Hamiltonian in the **Ap** formalism and was tested in the calculation of interaction terms for Al(001) surface, between 0.1 and 30 eV, using an elementary wave function. The results show that for the ***p*** polarization the normal to the surface, linear and quadratic, interaction terms dominate below approximatively 2 eV whereas the parallel to the surface linear interaction term dominates above this energy. The present model reproduces the bulk plasma frequency at about 15.3 eV.

Key words: potential vector, dielectric permittivity, electron density, continuous, **Ap** formalism, Al.

1. INTRODUCTION

In this paper we discuss the laser-matter interaction at a gas-solid interface in the long wave length (LWL) approximation, *i.e.*, for wavelength greater than 100 A° or energies less than 124 eV. This means that the laser only "sees" the sharp rise of the electron density at the gas-solid interface not the particular atoms. The rise of the electron density takes place at sub-nanometric scale and therefore the electromagnetic field in the present model is explicitly dependent on $\vec{r}$.

We propose a model for the classical vector potential $\vec{A}$ using the local dielectric function of the bulk at various frequencies taken from tables of Palik [1] and a relation between the dielectric function and the electron density at the gas-solid interface similar to the one known from the Drude theory. Consequently the present approach is called: electromagnetic fields from electron density, **EMED**.

The continuity relations for the electric and magnetic fields at abrupt interfaces are used to calculate the electric field. Then, the potential vector $\vec{A}$ is derived and used to construct the Hamiltonian in the $\vec{A}\cdot\vec{p}$ formalism (called **Ap**) that we demonstrated [2, 3] to be more appropriated, when treating interface problems, than the usual $\vec{E}\cdot\vec{d}$ formalism.

2. LIGHT AT INTERFACES

2.1. CONTINUITY AT INTERFACE

Let us now consider the continuity of the electric $\vec{E}$ and magnetic $\vec{B}$ fields of light at an abrupt interface. From now on the laser is monochromatic and the light polarization is linear. The problem of the continuity of the light at abrupt interfaces is given in classical textbooks [4–6]. Here we briefly reproduce the presentation given by Jackson [6] where the fields and potentials are taken to be classical and corresponding to absorption.

To fulfill the continuity at the gas-solid interface one requires: (a) the continuity of the normal components of the electric displacement $\vec{D}$ and magnetic induction $\vec{B}$; (b) the continuity of the tangential components of electric $\vec{E}$ and $\vec{H}$ magnetic fields.

The plane of incidence (POI), or xOz, is defined by z, the normal to the surface oriented towards the solid, and the wavevector $\vec{k}$ of light. The equality of the tangential component of $\vec{k}$ for the incident (i), the reflected (r) and the transmitted (t) waves at the interface, $\vec{k}^i_{//}=\vec{k}^r_{//}=\vec{k}^t_{//}$ gives the Snell law: $\sin\theta_i=\tilde{n}\sin\theta_t$, where $\tilde{n}=n_s/n_v$ and n_v and n_s are the refraction indices in vacuum and solid. Because of the absorption of the light in the solid, $\tilde{n}=\dfrac{(n_s+ik_s)}{n_v}$ is a complex number. The wave vector $\vec{k}$ is related to the refraction indices by $k=\dfrac{\omega n}{c}$ where ω is the laser pulsation.

When the electric field belongs to the POI, the polarization is known as ***p*** whereas for the vector field perpendicular to POI, the polarization is called ***s***. Because it is more complicated, in the following we consider only the case of ***p*** polarization. Using the continuity relations mentioned above, one deduces for the **tangential** component of the electric field

$$\begin{cases} E^p_x(\vec{r},t)=\cos\theta_i\left(E^p_i(\vec{r},t)-E^p_r(\vec{r},t)\right) & \text{vacuum} \quad z<0 \\ E^p_x(r,t)=\cos\theta_t E^p_t(\vec{r},t) & \text{solid} \quad z\geq 0 \end{cases} \tag{1}$$

while the **normal** component is

$$\begin{cases} E_z^p(\vec{r},t) = \sin\theta_i \left(E_i^p(\vec{r},t) + E_r^p(\vec{r},t) \right) & \text{vacuum} \quad z < 0 \\ E_z^p(r,t) = \varepsilon(z) \sin\theta_t E_t^p(\vec{r},t) & \text{solid} \quad z \geq 0 \end{cases} \tag{2}$$

In eqs. (1)–(2) above, $E_i^p(\vec{r},t)$ is the incident field at the angle θ_i, $E_r^p(\vec{r},t)$ is the reflected field, $E_t^p(\vec{r},t)$ the transmitted field and $\varepsilon(z)$ is the z-dependent relative dielectric function that will be derived in the next section. Since the correspondent refraction index $\tilde{n}(z) = \sqrt{\varepsilon(z)}$ is complex, θ_t is also complex and has no longer the simple significance of an angle of refraction [4]. Using eqs. (1)–(2) one writes the following relations for the reflected and the transmitted fields that can be calculated from the known incident field once $\tilde{n}(z)$ is derived

$$\begin{cases} E_i^p(\vec{r},t) = E_0 \exp\left[i\vec{k}^i\vec{r} - i\omega t \right] \\ E_r^p(\vec{r},t) = E_0 \dfrac{\tilde{n}(z)\cos\theta_i - \cos\theta_t}{\tilde{n}(z)\cos\theta_i + \cos\theta_t} \exp\left[i\vec{k}^r\vec{r} - i\omega t \right] \\ E_t^p(\vec{r},t) = E_0 \dfrac{2\cos\theta_i}{\tilde{n}(z)\cos\theta_i + \cos\theta_t} \exp\left[i\vec{k}^t\vec{r} - i\omega t \right]. \end{cases} \tag{3}$$

Here E_0 is the amplitude of the incident electromagnetic field related to the laser fluence.

2.2. ELECTROMAGNETIC FIELDS FROM ELECTRON DENSITY (EMED) MODEL

At an abrupt interface the material constants of the gas phase change suddenly to fit the bulk constants and if the light is absorbed in the solid, these quantities become complex. Here we present a simple model, called electromagnetic fields from electron density (EMED), based on the unperturbed electron density of the material that gives rise to continuous behavior of the material constants and of the electric field or vector potential at the interface. Such a model is justified in the solid and at interfaces because the laser intensities or fluences are weak and nondestructive, unable to produce a nonlinear behavior of the light by standard mechanisms known from gas phase. Even at low fluence such a nonlinear behavior can appear because of the band structure of the solid and/or non-equilibrium behavior of the electrons of the solid but in this paper the nonlinear behavior of the system is withdrawn.

In the solid state theory of Drude [4], one writes the relation between the solid dielectric constant ε^s, the conductivity σ at a given pulsation ω and the unperturbed electron density ρ_e of the solid

$$\varepsilon^s = \varepsilon_r + i\varepsilon_i = 1 + i\frac{\sigma}{\omega\varepsilon_0} = \tilde{n}^2 = (n_s + ik_s)^2 = n_s^2 - k_s^2 + 2in_s k_s \tag{4}$$

and

$$\sigma = \frac{\rho_e(z)e^2}{m(\gamma - i\omega)}. \tag{5}$$

Here γ is the friction force for the motion of these electrons of charge e and mass m.

The functional dependence given by eqs. (4)–(5) suggest the following simple analytic form for the dielectric function $\varepsilon(z)$ having a linear dependence on ρ_e:

$$\varepsilon(z) = 1 + (\varepsilon^s - 1)\rho_e(z) \tag{6}$$

with ε^s calculated from n_s and κ_s, the real and imaginary refraction indexes function of energy, taken from the tables of Palik [1]. The dispersion of n_s and κ_s gives rise to the energy dependence of $\varepsilon(z)$. One can verify that this expression displays the right asymptotic behavior in gas phase and deep in the solid.

In order to describe the z-variation of ε one have to model the free electron density $\rho_e(z)$. The electron density at the interface was estimated by Makinson [7, 8] many years ago and in the 70-ties this density was calculated by Lang and Kohn [9] for a jellium, using the density functional method. Here we fit the electron density given by [9] to an analytic form reproducing the Friedel oscillations. This form is derived by analogy to the energy behavior of the transmission factor through a potential barrier, considering the valence electrons of the surface as free electrons moving in a constant potential and bounded by finite barriers [10]. We started from the expression giving the transmission through a barrier of length L [11]:

$$\begin{cases} T = \dfrac{4E(E-U_0)}{4E(E-U_0) + U_0^2 \sin^2(qL)} & E > U_0 \\ T = \dfrac{4E(E-U_0)}{4E(E-U_0) + U_0^2 \sinh^2(\chi L)} & E < U_0 \end{cases} \tag{7}$$

where $q = \sqrt{(E-U_0)}$ and $\chi = \sqrt{(U_0 - E)}$. In fact these two forms correspond to a single function where the argument is allowed to be complex for $E < U_0$.

Here we use a function similar to the one given above but more flexible where we replaced $4E(E - U_0)$ by $\arctan[\lambda(z - z_0)]$ and added a damped function to attenuate the sine function:

$$\rho_e(z) = \frac{NA\arctan\left[\lambda\left(z - z_0\right)\right]}{A\arctan\left[\lambda\left(z - z_0\right)\right] + A_s \exp(\beta z)\sin^2\left[Lq(z)\right]} \tag{8}$$

where for $z < z_0$, as in eqs. (6), $q(z) = \sqrt{(z - z_0)}$ and sin become respectively, pure imaginary and sinh. None of the fit parameters used here (N, A, λ, z_0, A, β) have physical meaning.

In Fig. 1, we display the analytic (full curve) and numeric (dashed curves) electron densities function of z coordinate across the interface for a Wigner-Seitz radius ($r_s = \frac{3}{4\pi\rho_e^{1/3}}$) between 2 and 3 a.u. (for aluminium $r_s = 2.07$). Using the parameters: $N = 1.04$, $A = 50$, $\lambda = 0.5$, $z_0 = 0.53$, $A_s = 2.5$, $\beta = -0.01$ and $L = 2.2$, the agreement is quite fair between the above analytic function and the Lang and Kohn [9] calculations for $r_s = 2$ et 3 a.u.

Using the expressions of this subsection we can now construct a continuous electric field $\vec{E}$ or vector potential $\vec{A}$ all over the space. From $\vec{E} = -\frac{\partial \vec{A}}{\partial t} - \vec{\nabla} U$ assuming zero scalar potential U, the vector potential can be simply calculated as

$$\vec{A} = \frac{\vec{E}}{i\omega} \tag{9}$$

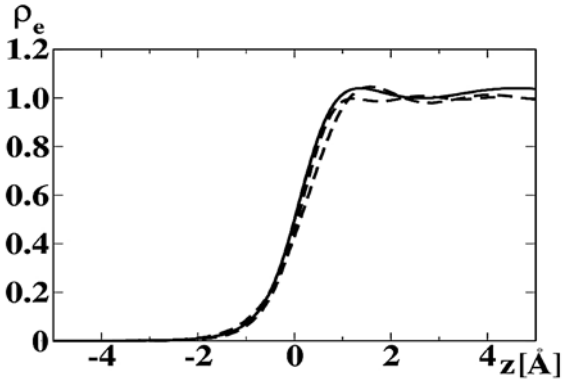

Fig. 1 – Electron density, at the gas-jellium interface: (full line) fitted electron density using the analytic form function eqs. (8); (dotted line) Lang and Kohn [9] calculations for $r_s = 2$ et 3 a.u.

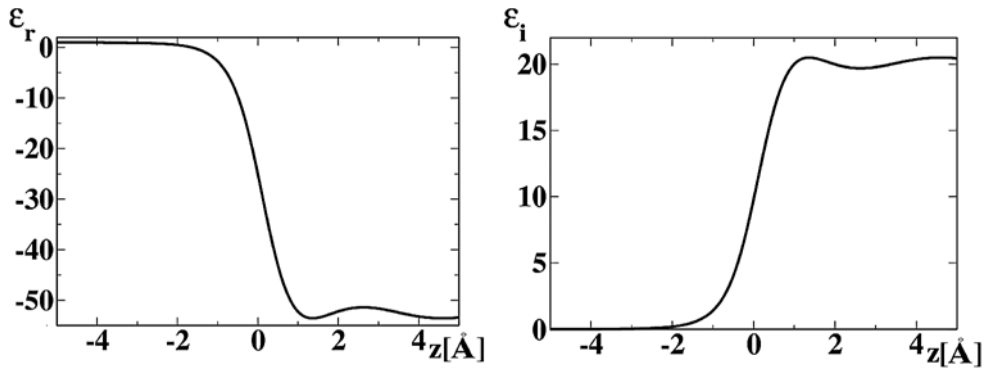

Fig. 2 – Behavior of the dielectric function with z calculated using eqs. (6).

The resulting variation in z of the vector potential for the $\boldsymbol{p}$ polarization is presented in Fig. 3. This variation is relatively sudden but continuous and, as the electron density, starts already in the gas phase near the surface. At the present laser energy of 2 eV the Friedel oscillations seems to not influence the spatial dependence of the vector potential but at greater energies (> 20 eV) the spatial dependence is clearly influenced as one would expected from chemisorption at metallic surfaces where the interaction potential clearly presents Friedel oscillations [12].

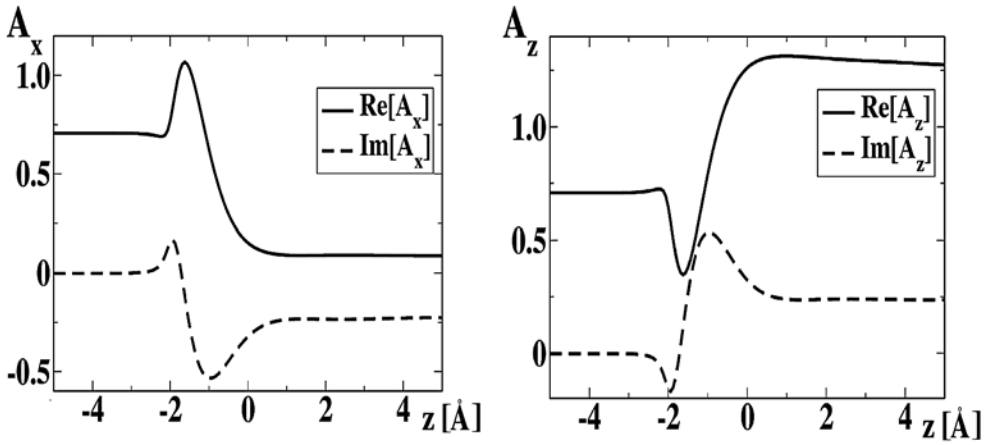

Fig. 3 – Variation with z of the spatial part of the vector potential near the interface for aluminium at oblique incidence of 15 degrees, for a laser of 2 eV, 30 J/m^2: left side represents the tangential (x) component and right side the normal (z) one.

3. LASER-MATTER INTERACTION HAMILTONIAN

The total Hamiltonian of a material system interacting with an external electromagnetic radiation reads

$$\begin{aligned} \hat{H} &= \frac{1}{2m}\left(\frac{\hbar}{i}\vec{\nabla} - e\vec{A}\right)^2 = \hat{H}_0^{cin} + \hat{H}_I \\ \hat{H}_I(\vec{r}) &= -\frac{\hbar e}{2im}\left(2\vec{A}(\vec{r})\vec{\nabla} + \vec{\nabla}\vec{A}(\vec{r})\right) + \frac{e^2}{2m}\vec{A}(\vec{r})\vec{A}(\vec{r}) = \hat{H}^L(\vec{r}) + \hat{H}^Q(\vec{r}) \end{aligned} \tag{10}$$

where e is the electron charge, $\hat{H}_0^{cin}$ is the zero kinetic operator and $\hat{H}_I$ is the interaction operator, $\hat{H}^L$ represents the linear term and $\hat{H}^Q$ represents the quadratic term. The time dependence of the potential vector is omitted here, being irrelevant for the performed calculations.

For $\boldsymbol{p}$ polarization, the detailed expressions for x and z terms of $\hat{H}^L$ and $\hat{H}^Q$ are:

$$\hat{H}^{L} = -\frac{\hbar q_e}{im}\left(\frac{\partial A_x(\vec{r})}{2\partial x} + A_x(\vec{r})\frac{\partial}{\partial x} + \frac{\partial A_z(\vec{r})}{2\partial z} + A_z(\vec{r})\frac{\partial}{\partial z}\right) = \hat{H}_x^L + \hat{H}_z^L \tag{11}$$

$$\hat{H}^{Q} = \frac{q_e^2}{2m}\left(A_x^2(\vec{r}) + A_z^2(\vec{r})\right) = \hat{H}_x^Q + \hat{H}_z^Q \tag{12}$$

We used for the initial and final states the same wave function, constructed as a product of two plane waves in the x and y directions and a sum of Gaussian functions in the z direction:

$$\psi(\vec{r}) = N\frac{1}{2\pi}\exp\left(-ik_x x\right)\exp\left(-ik_y y\right)\sum_{i=1}^{N_g}\exp\left(-\frac{\left(z-z_i\right)^2}{2\varsigma^2}\right) \tag{13}$$

where k_x and k_y are the momenta in the bulk solid varying in the first Brillouin zone from $-\pi/a_0$ to π/a_0 and the plane waves are normalized in the continuum to k (normalization factor $1/\sqrt{2\pi}$). The Gaussian functions have an identical exponent ($\varsigma = 1.5$), centered at the atomic positions z_i of the lattice, spaced by a_0. Such an arrangement corresponds to a (001) surface (for details see refs. [1, 2]).

4. LASER-MATTER INTERACTION TERMS WITH THE EMED METHOD

One of the best ways to test the laser-matter interaction model in the $\vec{A}\cdot\vec{p}$ formalism using the EMED expression for the vector potential $\vec{A}$ is to study the dependence of this interaction terms with the laser energy. In Fig. 4 we present the calculations for metallic aluminium, a nearly free electron metal. The calculations have been performed for a ***p*** polarized laser incident at 15 degrees having a moderate fluence of 30 J/m^2 and an energy variing between 0.1 and 30 eV. The energy of the material system, which should correspond to the used rudimentary wave function eqs. (13), is simply taken to be half of the laser energy. Then, by energy equipartition, this energy is split between the *x, y* and *z* degrees of freedom.

Analyzing the interaction terms presented in Fig. 4, one sees that the low energy edge domain (left graph) is dominated by the z (normal to the surface) components of the laser matter linear and quadratic interaction terms, $\left\langle \hat{H}_z^L \right\rangle$ and $\left\langle \hat{H}_z^Q \right\rangle$ (where $\left\langle \hat{H} \right\rangle = \left\langle \psi_f \middle| \hat{H} \middle| \psi_i \right\rangle$). The parallel to the surface, x, linear $\left\langle \hat{H}_x^L \right\rangle$ term dominates at other energies. The x quadratic term is small and negligible at all energies. At bulk plasma resonance, around 15 eV, some contribution from the linear term $\left\langle \hat{H}_z^L \right\rangle$ reappears. If one calculates this same interaction using spatially

independent vector potential, then the interaction would strongly diminish above 5 eV. From Fig. 5 one can see that if no attenuation of the laser field in the solid is

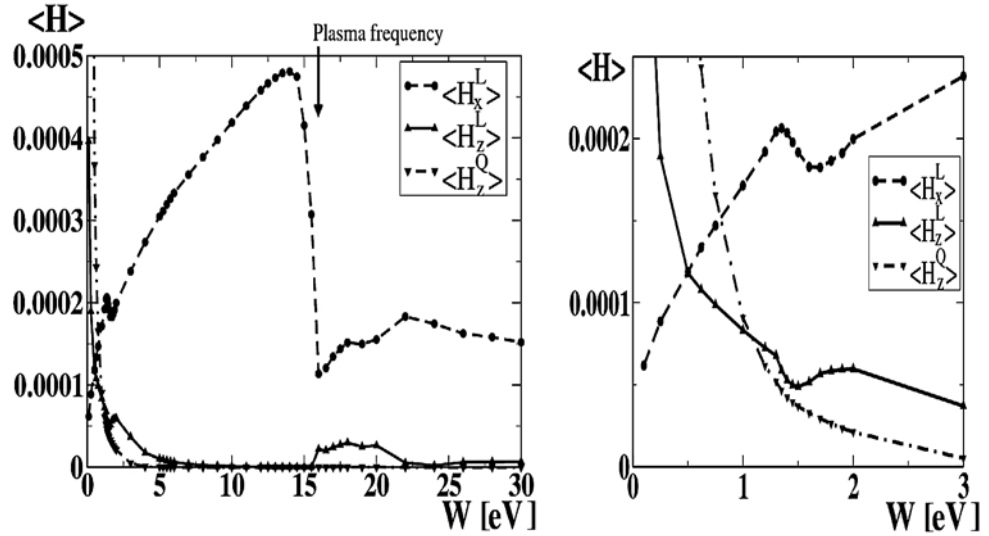

Fig. 4 – Expectation values for the linear terms $\left\langle \hat{H}_z^L \right\rangle$, $\left\langle \hat{H}_x^L \right\rangle$ and quadratic term $\left\langle \hat{H}_z^Q \right\rangle$ of metallic aluminium for a ***p*** polarized laser, as function of laser energy for: left panel, whole energy range and right panel, low energy range.

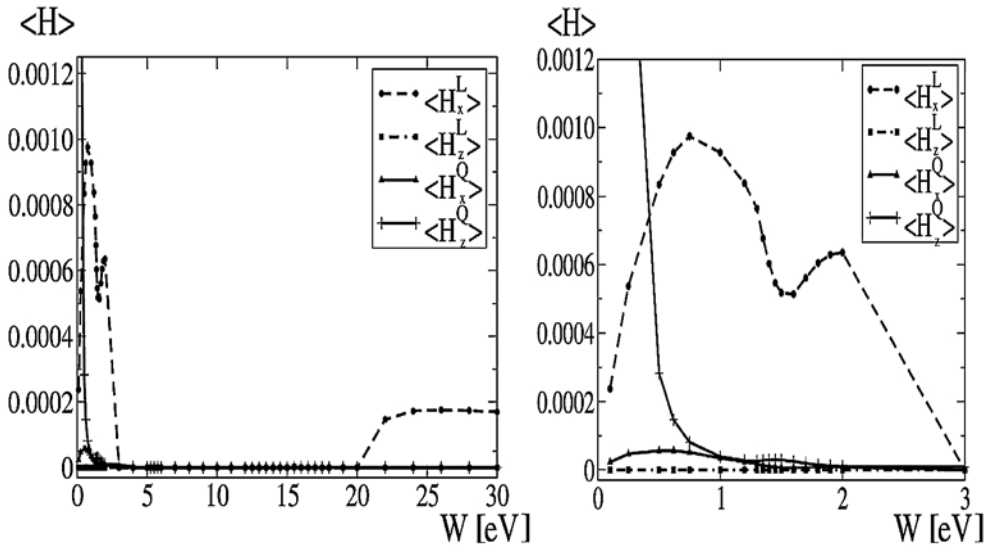

Fig. 5 – Expectation values of the linear terms $\left\langle \hat{H}_x^L \right\rangle$, $\left\langle \hat{H}_z^L \right\rangle$ and quadratic terms $\left\langle \hat{H}_x^Q \right\rangle$. $\left\langle \hat{H}_z^Q \right\rangle$ of metallic aluminium for a ***p*** polarized laser, disregarding the laser attenuation in the solid, as function of laser energy for: left panel, whole energy range and right panel, low energy range.

introduced, the interaction matrix elements are much larger and display very different energy dependence. Also the expectation value of the quadratic term $\left\langle \hat{H}_x^Q \right\rangle$ has some contribution contrasting with the preceding situation where it can be completely neglected.

5. CONCLUSIONS

The EMED model developed here is based on the local dielectric function taken from the experiment. This dielectric function depends, through the electron density, on the z coordinate normal to the surface. The connection between the dielectric function and electron density at the surface, together with the continuity conditions for the wave vectors of the electromagnetic fields allow us to construct a phenomenological vector potential of the electromagnetic field continuous through the surface.

Laser-matter interaction Hamiltonian is then obtained in the **Ap** formulation and the contributing terms are studied as function of laser energy for aluminium. The wave function used in the calculation of the matrix elements of the laser-matter interaction, product of plane waves parallel to the surface and sum of Gaussian functions perpendicular to it, is elementary but allow to estimate the interaction terms. We have calculated the corresponding expectation values of the interaction terms with the same function for the initial and final states. Therefore our results have to be seen as an upper limit to the actual interactions, but they conserve all their meaning when comparing the different contributions of the laser-matter interaction.

For the ***p*** linearly polarized electric field of 30 J/m^2, the z components of the laser-matter interaction dominate below 2 eV whereas the x linear term ρ dominates above this energy. For a structureless surface, or a surface having the same structure in x and y direction, one can infer the results for the ***s*** polarization from the present work because the operators $\hat{H}_y^a$ (with $a = L$ or Q) of ***s*** polarization are completely equivalent to the corresponding $\hat{H}_x^a$ for ***p*** polarization.

The present preliminary work with the EMED model will be extended in the near future to the time dependent calculation of the excitation cross section and electric current at the interface.

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