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Dynamics of fast electron beams and bounded targets

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ABSTRACT

We analyze the full relativistic force experienced by swift electrons moving close to planar films for the experimental conditions commonly used in electron energy loss spectroscopy in STEM. In metals the main effects derive from the dispersion of the surface plasmons, which are clearly observed in the EEL spectra. In insulators we explore the role played by the Cherenkov radiation (CR) emitted in the energy gap window. The focus is placed on the transverse force and different factors which may turn this force into repulsive, as reported in recent experimental and theoretical works.

1. Introduction

The retarding force experienced by electron probes in inhomogeneous media has attracted great attention in the last decades due to its interest in electron energy loss spectroscopy (EELS) in scanning transmission electron microscopy (STEM). Nowadays this technique has become a very accurate tool to characterize nanostructures and map plasmons with high resolution. The transverse component of the force has received less attention, but in general it is assumed to be attractive and small. Nevertheless, several experiments and theoretical predictions have questioned the expected behavior.

Early experiments [1,2] detected strong deflections when a 100 keV electron beam passed close to targets of MgO and Au of 100 nm side at a distance of around 1 nm, which stimulated different calculations of the image force within classical dielectric theory [3,4], considering infinite planar surfaces or spherical targets [5]. More recently, advances in electron microscopy have allowed to monitor the movement of nanometric gold particles under the action of a well focused electron beam, finding that the force experienced by the particle becomes repulsive at small impact parameters. These experiments have stimulated new relativistic calculations of the momentum transferred to nanoparticles (NPs), accounting for a precise description of the target [6–8], which reproduce the observed repulsive interaction, but the physics behind this effect is still unclear. In fact this repulsive force near small metallic particles at small impact parameter was also reported by García de Abajo [9] in a previous work. In a very recent paper Rebernik [10] predicted that the interaction between a planar surface and a charge packet moving parallel to it becomes repulsive above a critical relativistic energy.

In the present work the retardation effects on the force between 300 keV electron beams and metallic (Al, Au) or insulator (MgO) films are explored, with the focus on the sign of the transverse component, extending a recent study for a semi-infinite medium [11]. First we analyze the effects of retardation induced plasmon dispersion clearly observable in the EEL spectra. In the case of insulator targets it has been proved that most of the relativistic effects derive from Cherenkov radiation (CR). The behavior of the transverse force with impact parameter is finally studied, finding that it is always attractive for the common STEM setups, but different factors providing repulsive forces are discussed.

2. Theory

The Lorentz force experienced by a fast electron moving at velocity v and impact parameter b, parallel to a planar film of thickness a (see inset of Fig. 1(a)), characterized by a dielectric function $\epsilon(\omega)$, is calculated following the same method used for semi-infinite targets in a previous reference [11]. Both transverse and longitudinal components are written in terms of a single complex response function Σ, which depends on frequency ω and parallel component of the momentum k_y :

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$$F_x = \frac{-2}{\pi v}\int_0^\infty dk_y \int_0^\infty d\omega v_0 \mathrm{Re}[\Sigma(k_y,\omega)]e^{-2v_0 b} \quad (1)$$

$$F_z = \frac{-2}{\pi v}\int_0^\infty dk_y \int_0^\infty d\omega \frac{\omega}{v}\mathrm{Im}[\Sigma(k_y,\omega)]e^{-2v_0 b}, \quad (2)$$

where

$$\Sigma(k_y,\omega) = \frac{\epsilon-1}{\Delta_0}\left[\frac{2v_0}{\Delta}\Lambda - \frac{\omega^2}{c^2}\frac{1-\beta^2}{v_0}(1-e^{-2va})\right], \quad (3)$$

and

$$\Delta_0 = (v+v_0)^2 - (v-v_0)^2 e^{-2va}$$
$$\Delta = (v+v_0\epsilon)^2 - (v-v_0\epsilon)^2 e^{-2va}$$
$$\Lambda = (v+v_0)(v+v_0\epsilon) + (v-v_0)(v-v_0\epsilon)e^{-4va} - 2(v^2+v_0^2\epsilon)e^{-2va}, \quad (4)$$

with $v = \sqrt{k_y^2 + [1-\beta^2\epsilon(\omega)]\omega^2/v^2}, v_0 = \sqrt{k_y^2+\omega^2/\gamma^2 v^2}, \beta = v/c$ and $\gamma = (1-\beta^2)^{-1/2}$ the Lorentz factor. Notice that these general equations lead to the simpler expressions for the semi-infinite medium [11] as $a \to \infty$. From one side, for EELS applications in electron microscopy, it is helpful to consider the ω dependent integrand in the longitudinal force [12],

$$F_z = -\int_0^\infty \omega d\omega \frac{dP}{dzd\omega}, \quad (5)$$

which corresponds to the probability P (per unit length) of losing energy ω,

$$\frac{dP}{dzd\omega} = \frac{2}{\pi v^2}\int_0^\infty dk_y e^{-2v_0 b}\mathrm{Im}[\Sigma(k_y,\omega)]. \quad (6)$$

On the other side, in order to describe the nature (attractive or repulsive) of the interaction, it is illustrative to analyze the transverse momentum transfer p_x per unit length and energy,

$$\frac{dp_x}{dzd\omega} = \frac{F_x}{vd\omega} = -\frac{2}{\pi v^2}\int_0^\infty dk_y \mathrm{Im}[k_x\Sigma(k_y,\omega)]e^{-2v_0 b}. \quad (7)$$

Notice also that v_0 may be interpreted as the modulus of k_x, the transverse wave vector component associated to the evanescent fields, $k_x = iv_0 = \sqrt{\omega^2 c^{-2} - k_y^2 - \omega^2 v^{-2}}$.

The previous formalism provides a description of the scattering of the probe in terms of excitations of momentum $k = (k_x, k_y, \omega v^{-1})$ and energy ω, where $\Sigma(k_y,\omega)$ is a key function describing the probability (always per unit length) of exciting such a process. In [11] we described in detail the behavior of this function for metals and insulators in a semi-infinite medium. It is worth mentioning that its imaginary part is always positive in the whole $\omega - k_y$ space, while its real part changes its sign around the surface plasmon dispersion curve.

Finally, notice that the non-retarded limits are directly recovered from these expressions for $\beta \to 0$.

Following reference [11] we have also calculated the force experienced by the film, finding that, for any dielectric function, it is equal to the force on the probe. It is important to consider that the total momentum transfer to the film can contain some radiative contributions in the case of CR supporting media.

3. Results

First we analyze the energy loss probability, which is the magnitude commonly measured in EELS experiments, and the effects of retardation in Al slabs of different thicknesses, represented by a Drude dielectric function $\epsilon(\omega) = 1 - \omega_p^2/(\omega^2 + i\omega\gamma)$, with $\omega_p = 15.3$ eV and $\gamma = 1$ eV. The effects of retardation in metals derive mainly from the dispersion of the surface plasmon $\omega(k_y)$,

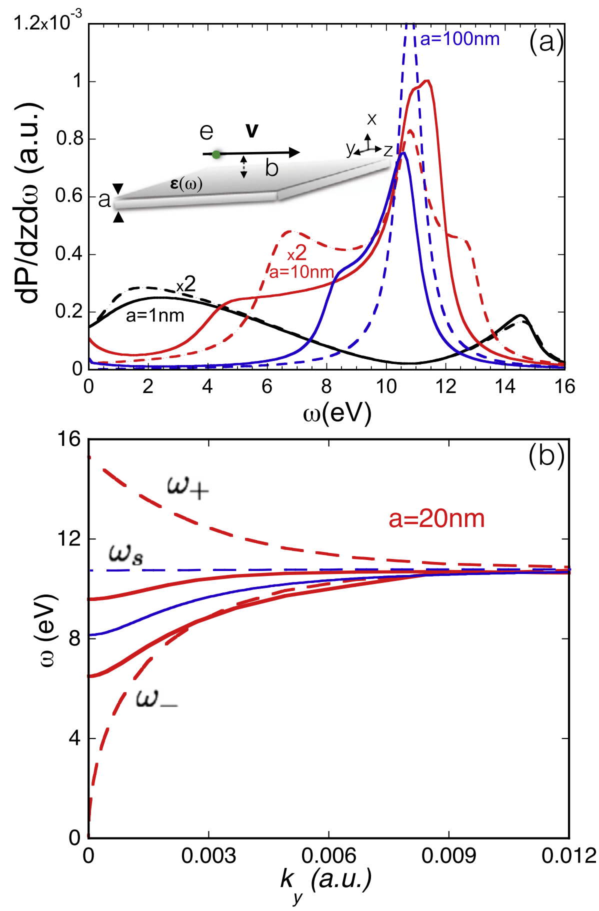

Fig. 1. (a) Energy loss probability per unit length for 300 keV electrons parallel to Al slabs of thicknesses a = 1, 10 and 100 nm at impact parameter b = 1 nm. The sketch in the inset represents the geometry of the problem. (b) Dispersion relation for 300 keV electron in front of an Al film of thickness a = 20 nm (red) and a semi-infinite medium (blue). Dashed lines correspond to non-retarded approximation in both (a) and (b). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

as described by Eqs. (1) and (2) and discussed in [11] for semi-infinite planar targets. In that case the surface plasmon is red-shifted as $k_y \to 0$. For films there are two surface plasmon modes [13], symmetric (ω_-) and antisymmetric (ω_+), which in the non-retarded limit are given by $\omega_\pm = \omega_p[(1 \pm e^{-qa})/2]^{1/2}$, where $q = (k_y^2 + \omega^2/v^2)^{1/2}$ is the parallel momentum. In the retarded case the modes (given by the zeros of the denominator of Σ function) exhibit a shift towards the surface plasmon frequency of the semi-infinite medium as $k_y \to 0$, as is clearly observed in Fig. 1(b).

This behavior is illustrated in the EEL spectra of Fig. 1(a) for three different values of the thickness a = 1, 10 and 100 nm, corresponding to 300 keV electrons at impact parameter b = 1 nm. In the last case of the thickest film the spectra are pretty similar to the ones obtained for a semi-infinite medium at the same impact parameter. Moreover, retardation produces a shift and broadening of the surface plasmon peaks, which is much stronger for thicker films. For a = 1 nm the non-retarded approximation works pretty well, as observed in the figure.

Furthermore, as the probe trajectory is more distant from the surface, the effects of retardation become stronger. This is because the small momenta have more weight in the loss spectrum, as inferred from the exponential factor in the k_y integral in Eq. (2), and it is just at small k_y where dispersion is more relevant, as shown by the dispersion curves described above.

In insulators, which are transparent in an energy range, there is an additional mechanism entering into play within the relativistic