Interface modes of two-dimensional composite structures

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Abstract

The surface modes of a composite consisting of aligned metallic wires with square cross-sections are investigated, on the basis of photonic band structure calculations. The effective long-wavelength dielectric response function is computed, as a function of the filling fraction. The dependence of the optical absorption on the shape of the wires and the polarization of light is discussed, and the effect of sharp corners analyzed. The effect of the interaction between the wires on the localization of surface plasmons is also addressed. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Electron–solid interactions, scattering, diffraction; Interface states; Light scattering; Plasmons

1. Introduction

The interface electromagnetic modes of composite structures [1] are well-known to be of basic importance in the understanding of a number of physical problems such as optical absorption (see for example Ref. [2]), van der Waals attraction [3], surface-enhanced Raman scattering [4,5], and the response to incident charged particles [6–9]. Analytical descriptions of normal electric modes have been limited to slabs and isolated spheres [10–12] and spheroids [13], while numerical methods have been required for most shapes [14,15] including such simple cases as single cubes [16,17] and rectangles [17].

Dipolar modes in a system of identical spheres embedded in an otherwise homogeneous host have

been approximately described, for many years, from the knowledge of the effective transverse dielectric function first derived by Maxwell-Garnett [18,19] (MG) within a mean-field approximation valid for small values of the volume occupied by the spheres. Also, many attempts have been made to account, at large filling fractions, for higher multipole interactions, which are absent in the MG theory [20–24]. Recently, new techniques have been developed for solving Maxwell's equations in structured materials [25–28]. Based on these numerical solutions of Maxwell's equations, exact calculations of the long-wavelength limit of the effective transverse dielectric function of periodic dielectric [29] and metallic [30–32] structures have been performed. From these calculations, strengths and positions of surface modes of a composite made up of long metallic cylinders have been obtained [32], and the MG theory has been shown to offer a good approximation as long

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as the distance between the axis of neighboring cylinders is larger than twice the diameter of the cylinders.

2. Theory

In this paper we extend the work presented in Ref. [32] to the case of aligned metallic wires with square cross-sections. We consider a binary metal–dielectric mixture with a volume fraction $1-f$ of the insulator, with a real positive dielectric constant ϵ_0, and f of a periodic system of long metallic wires with square cross-sections arranged in a square array with lattice constant $a = xd$, as shown in Fig. 1. The cross-section of the wires is taken to be small in comparison to the wavelength of the electromagnetic excitation and large enough that a macroscopic dielectric function $\epsilon(\omega)$ is ascribable to the wires. Thus, the electromagnetic properties of the composite will be defined by a single effective dielectric function:

$$\epsilon_{\text{eff}}(\omega) = \frac{k^2 c^2}{\omega^2}, \tag{1}$$

the wave vector k being that corresponding to a Bloch wave propagating through the composite. For simplicity, the magnetic permeabilities are assumed to be equal to unity in both media.
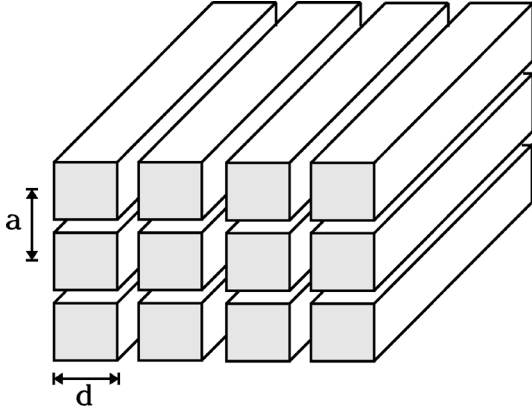

Fig. 1. Periodic system of metallic wires with square cross-sections of side d, arranged in a square array with lattice constant a. The wires are infinitely long in the y direction.

The dielectric function of metals is nearly real and negative below the plasma frequency down to frequencies comparable to the inverse of the conductivity mean free time. Hence, negative values of $\epsilon(\omega)$ give rise to a variety of surface resonances, which allow us to express the long-wavelength limit of the effective dielectric function of the composite in the following form [33,34]:

$$\epsilon_{\text{eff}}(\omega) = \epsilon_0 \left[1 - f \sum_v \frac{B_v}{u(\omega) - m_v} \right], \tag{2}$$

where

$$u(\omega) = [1 - \epsilon(\omega)/\epsilon_0]^{-1}, \tag{3}$$

with m_v ($0 \leq m_v < 1$) the depolarization factor associated with the vth normal mode, and B_v ($B_v \geq 0$) representing the corresponding strengths, which all add up to unity:

$$\sum_v B_v = 1. \tag{4}$$

Similarly:

$$\epsilon_{\text{eff}}^{-1}(\omega) = \epsilon_0^{-1} \left[1 + f \sum_v \frac{C_v}{u(\omega) - n_v} \right], \tag{5}$$

where $0 < n_v \leq 1$, $C_v \geq 0$, and

$$\sum_v C_v = 1. \tag{6}$$

{The minus sign in front of f in eq. (4) of Ref. [32] must be corrected and replaced by a plus sign, as in the present paper.} Both depolarization factors and strengths in Eqs. (2) and (5) depend only on the microgeometry of the composite material and not on the dielectric function of the components.

Optical absorption is directly given by the imaginary part of the effective dielectric function of Eq. (2), and coupling to charged particles is described by the imaginary part of the effective inverse dielectric function of Eq. (5). (If a broad beam of charged particles passes through the composite, the probability that they lose energy $\hbar\omega$ while undergoing a wave vector change $\boldsymbol{q}$ is proportional to the so-called energy-loss function, i.e. the imaginary part of the finite wavelength effective inverse longitudinal dielectric function. In typical electron energy-loss experiments the energy-loss function can sometimes be approximated by the $q \rightarrow 0$ limit,

and in this limit both longitudinal and transverse effective dielectric functions coincide.) In a homogeneous metal ($f=1$), there is only one mode strength different from zero, with depolarization factors $m_0=0$ and $n_0=1$, where Re $\epsilon \to \pm \infty$ and Re $\epsilon \approx 0$, respectively. Thus, for a metal described by the Drude dielectric function, $\epsilon(\omega)=1-\omega_p^2/\omega^2$, the bulk resonance at the plasma frequency, ω_p, can only be excited by penetrating charged particles.

In the case of two-dimensional composite structures (see Fig. 1), homogeneous along the axis of the inclusions, there are two different values of $\epsilon_{eff}(\omega)$ corresponding to different polarizations. If the electromagnetic (e.m.) wave incident on the structure is polarized along the axis of the inclusions (s-polarization), the presence of the interfaces does not modify the electric field and the response of the composite is easily found to be equivalent to that of a homogeneous medium having the effective dielectric function of Eqs. (2) and (5) with only one mode strength different from zero, $B_0=C_0=1$, and depolarization factors $m_0=0$ and $n_0=f$. {The same result is found in the case of plane parallel slabs aligned along the electric field (see e.g. Ref. [1]). If the electric field is perpendicular to the slabs, there is also only one mode strength different from zero, but now with depolarization factors $m_0=1-f$ and $n_0=1$. That the n_0 depolarization factor equals f and is, therefore, independent of the geometry of the two-dimensional structure is only valid, within the quasistatic approximation, for small values of the length d characterizing our two-dimensional structure. In the case of metallic wires of the order of 1 μm radius the self-inductance of the wire structure results in the n_0 depolarization factor decreasing, for a given filling fraction, as $\sim 1/d^2$ (see Ref. [35]).} Thus, for this polarization the energy-loss function, $Im[-\epsilon_{eff}^{-1}(\omega)]$, shows a single peak at a reduced plasma frequency. For a metal described by the Drude dielectric function, the reduced plasma frequency is $\omega = \sqrt{f}\omega_p$.

Now we focus on the response to e.m. waves polarized within the plane normal to the axis of the inclusions (p-polarization). For this polarization all depolarization factors m_v and n_v of Eqs.

(2) and (5) are found to satisfy the relation [32]:

$$n_v = 1 - (D-1)m_v, \qquad (7)$$

where D represents the dimensionality of the inclusions, i.e. $D=2$. As long as only dipole surface modes can be excited, $B_1=C_1=1$, a combination of Eqs. (2) and (5) with Eq. (7) results, for $D=2$, in the depolarization factors m_1 and n_1 to be given by

$$m_1 = \frac{1}{2}(1-f) \qquad (8)$$

and

$$n_1 = \frac{1}{2}(1+f). \qquad (9)$$

Eqs. (2) and (5) with $B_1=C_1=1$ and the depolarization factors of Eqs. (8) and (9) reduce to the two-dimensional MG effective dielectric function, as discussed in Ref. [32]. {For three-dimensional composite structures, $D=3$, a combination of Eqs. (2) and (5) with Eq. (7) results, if all multipolar modes are neglected, in the dielectric function originally derived by Maxwell-Garnett [18,19] for a system of spherical particles.} We note that multipolar modes can only be neglected in a few simple cases, in which the long-wavelength effective dielectric function is accurately given by the MG approximation. These are the cases when the two-dimensional structure is made of (a) a sparse ($f \to 0$) distribution of circular inclusions, and (b) a dense ($f \to 1$) distribution of square inclusions.

For a single inclusion embedded in a host material with dielectric constant ϵ_0, both $Im[\epsilon_{eff}(\omega)]$ and $Im[-\epsilon_{eff}^{-1}(\omega)]$ are proportional to the particle susceptibility, and satisfy the relation

$$Im[\epsilon_{eff}^{-1}(\omega)] = Im[\epsilon_{eff}(\omega)]/\epsilon_0^2, \qquad (10)$$

i.e. the spectral representations of Eqs. (2) and (5) coincide. Thus, one finds from Eq. (7) that, in the case of single inclusions, for any given mode with $m_v < 1/D$ there exists another mode with depolarization factor $m_v' > 1/D$ such that

$$m_v' = 1 - (D-1)m_v. \qquad (11)$$

In particular, for single circular inclusions only dipole surface modes can be excited, depolarization

factors being $m_1=n_1=1/2$, and both the optical absorption and the electron energy-loss exhibit a single strong maximum at the dipole resonance where $u=1/2$, i.e. $\epsilon(\omega)+\epsilon_0=0$. For single inclusions with shapes other than circular, this resonance splits into several peaks that occur at a set of frequencies according to Eq. (11).

In order to give a full quantitative description of all surface modes in our two-dimensional composite structure, we have followed the method developed in Refs. [27,28] for calculating dispersion relationships of Bloch waves in structured materials, and we have computed the effective dielectric function from Eq. (1). The insulating component of the composite has been chosen to have a real dielectric constant of $\epsilon_0=1$ and the dielectric properties of the metal inside the inclusions have been modeled by a Drude dielectric

function of the form

$$\epsilon(\omega)=1-\frac{\omega_p^2}{\omega(\omega+i\gamma)}, \qquad (12)$$

where ω_p represents the bulk plasma frequency of the metal and γ is an inverse electron relaxation time. The plasma frequency of the conduction electrons in aluminum has been used, $\hbar\omega_p=15.8$ eV, and the parameter γ has been chosen to be small enough so as to distinguish all multipolar excitations: $\gamma=0.1$ eV.

Figs. 2 and 3 show our numerical calculations of $\mathrm{Im}[\epsilon_{eff}(\omega)]/f$ and $\mathrm{Im}[-\epsilon_{eff}^{-1}(\omega)]/f$ as a function of the frequency ω, for various values of the ratio x between the lattice constant and the side of the wires. For $x\geq6$ our results are almost insensitive to the precise value of x, and they nearly coincide with
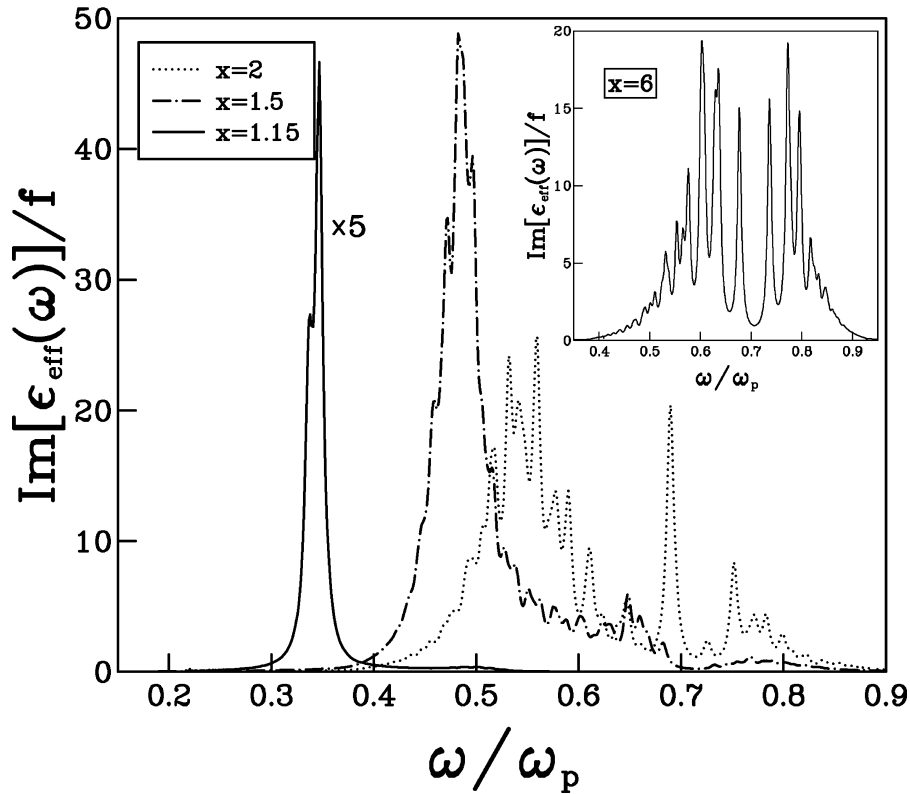

Fig. 2. Imaginary part of the effective long-wavelength dielectric function of the periodic system described in Fig. 1, for p-polarized electromagnetic excitations and various values of the volume filling fraction: 25.0% (dotted line), 44.4% (dashed line), and 75.6% (solid line). The results obtained for a volume filling fraction of 2.3% are represented in the inset.
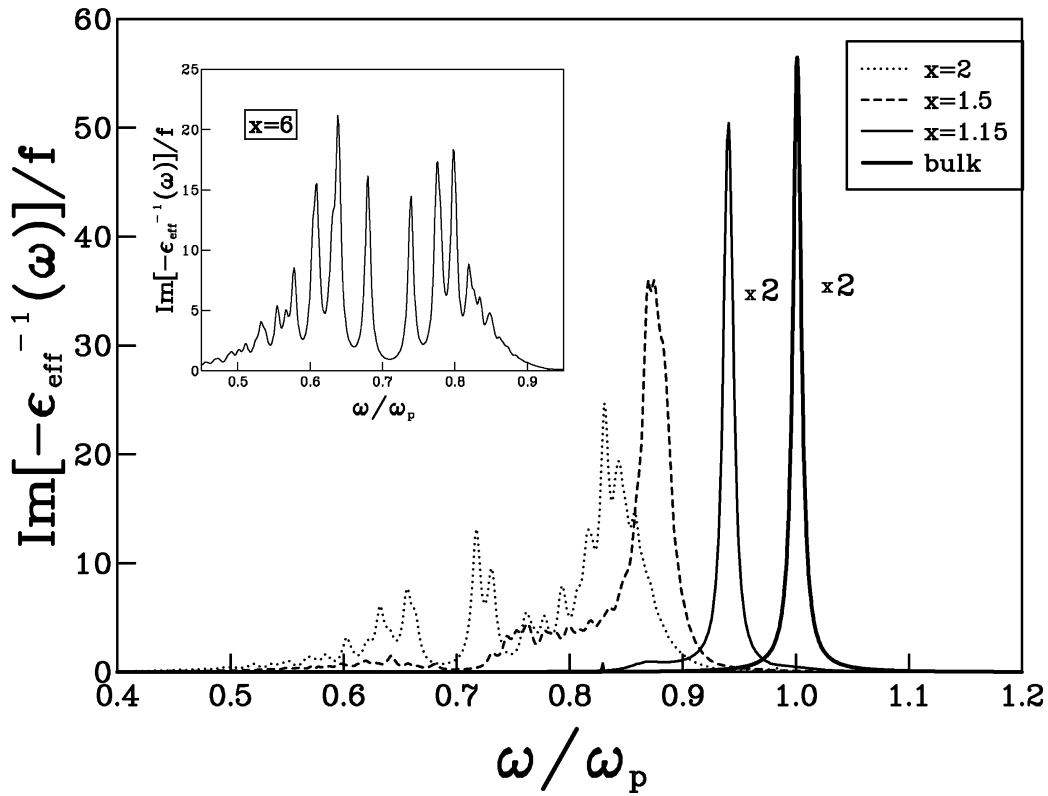
Fig. 3. Same as Fig. 2, for the effective energy-loss function. The bulk energy-loss function ($x=1$) is represented by a thick solid line.

the effective dielectric function of single wires. In this (dilute) limit, $\text{Im}[\epsilon_{\text{eff}}(\omega)]$ and $\text{Im}[-\epsilon_{\text{eff}}^{-1}(\omega)]$ coincide, and they both exhibit several resonances satisfying Eq. (11) (see the insets to Figs. 2 and 3). [Although the results presented in Figs. 2 and 3 are all well converged for $x \leq 2$, the results presented for $x=6$ (corresponding to a sampling mesh of $120 \times 1 \times 120$) might differ slightly at $\omega \sim \omega_{\text{p}}/\sqrt{2}$ from well-converged results.]

At higher concentrations of metal, both the optical absorption $\{\text{Im}[\epsilon_{\text{eff}}(\omega)]\}$ and the electron energy-loss $\{\text{Im}[-\epsilon_{\text{eff}}^{-1}(\omega)]\}$ show a dominant dipole-like mode and a band of multipole resonances. We note (see Figs. 2 and 3) that as x decreases the dipole-like resonance in $\text{Im}[\epsilon_{\text{eff}}(\omega)]/f$ and $\text{Im}[-\epsilon_{\text{eff}}^{-1}(\omega)]/f$ is shifted towards lower and higher frequencies, respectively, as a consequence of the interaction between one surface and another. This result, already predicted within the MG

approximation [see Eqs. (8) and (9)], is also found in the case of circular inclusions [32]. Also, it is interesting to notice that as long as x decreases multipole resonances tend to be negligible, thus dipolar depolarization factors approaching those predicted by Eqs. (8) and (9).

In the case of circular wires that are touching ($f=0.785$) the metal forms a connected medium, and the so-called energy-loss function $\text{Im}[-\epsilon_{\text{eff}}^{-1}(\omega)]$ shows, therefore, a bulk plasmon excitation at $\omega=\omega_{\text{p}}$ ($n_1=1$). Besides this peak, there is also a band in $\text{Im}[-\epsilon_{\text{eff}}^{-1}(\omega)]$ from multipole contributions to the effective response at $\omega_\nu = \sqrt{n_\nu(f)}\,\omega_{\text{p}}$ ($\nu=2, ...; n_\nu < 1$), which prevents our computed effective dielectric function from coinciding with the MG approximation (see Ref. [32]). However, the presence of flat surfaces with sharp corners, as in the case of square inclusions, results in all multipolar strengths being
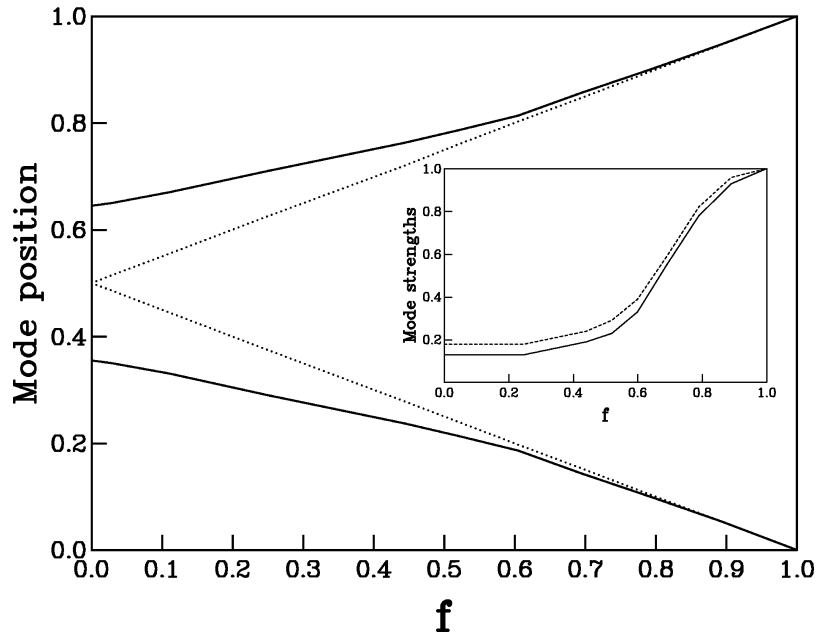
Fig. 4. Dipolar mode positions (depolarization factors), as a function of the volume filling fraction, for wires with square cross-sections. Solid lines: our numerical results for m_1 (the line below 0.5) and n_1 (the line above 0.5). Dotted lines represent the MG depolarization factors given by Eqs. (8) and (9). Dipolar mode strengths are represented in the inset: solid and dashed lines represent the coefficients B_1 and C_1 entering Eqs. (2) and (5), respectively. The MG dipolar mode strengths, B_1 and C_1, are both equal to unity.

negligible for $f > 0.75$ ($x < 1.15$); thus, our computed effective dielectric function is well described, in this case and for $x < 1.15$, by the MG approximation.

Finally, we have represented in Fig. 4 universal curves (solid lines) for the dipolar mode depolarization factors and strengths, versus the filling fraction, as determined with use of Eqs. (13) and (14) from our computed effective dielectric function of a system of infinitely long wires with square cross-sections. An inspection of this figure indicates that the trend with increasing filling fraction is for the dipolar peaks in $\mathrm{Im}[\epsilon_{\mathrm{eff}}(\omega)]/\mathrm{Im}[-\epsilon_{\mathrm{eff}}^{-1}(\omega)]$ to approach the MG results of Eqs. (8) and (9), also plotted in this figure (dotted lines); furthermore, the MG results nearly coincide with our numerical results for $f > 0.8$ ($x < 1.1$). Associated with the deviation, for smaller filling fractions, of the actual dipolar mode positions from the MG results is the reduction in the dipolar mode strengths B_1 and C_1, which are represented in the inset to Fig. 4. These mode strengths have

been determined by

$$B_v = \frac{\sqrt{m_v}}{fH} \, \mathrm{Im}[\epsilon_{\mathrm{eff}}(\omega_v)] \tag{13}$$

and

$$C_v = \frac{\sqrt{n_v}}{fH} \, \mathrm{Im}[-\epsilon_{\mathrm{eff}}^{-1}(\omega_v)], \tag{14}$$

where ω_v is the frequency associated with the vth normal mode, and H represents the peak height in the bulk energy-loss function [in the case of the Drude dielectric function of Eq. (12), m_v, $n_v = \omega_v^2/\omega_p^2$ and $H = \omega_p/\gamma$].

3. Conclusion

In summary, we have presented, on the basis of ab initio numerical solutions of Maxwell's equations in structured materials, exact calculations of the effective long-wavelength dielectric response

function of a periodic system of long metallic wires with square cross-sections. We have investigated the dependence of both the optical absorption and the electron energy-loss on the shape of the wires and the polarization of light. In the case of s-polarization the effective dielectric function does not depend on the shape of the wires and there is only one mode strength different from zero. In the case of p-polarization it had already been concluded [32] that MG results are good as long as the distance between the axis of neighboring circular cylinders is *larger* than twice the diameter of the cylinders. Now we have concluded that in the case of two-dimensional periodic structures consisting of square inclusions MG results are reliable as long as the ratio between the lattice constant and the side of the inclusions is *smaller* than $x \sim 1.1$. Both the presence of sharp corners in single wires and the interaction between circular wires result in absorption and energy-loss peaks to be broadened by the existence of multipolar surface resonances.

Acknowledgements

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