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Scattering of light by a periodic array of metallic nanoparticles on a waveguide

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Abstract. The optical response of a two-dimensional periodic array of metallic nanoparticles on a dielectric waveguide is investigated by means of numerical calculations using the on-shell layer-multiple-scattering method. We find that the strong interaction between particle-plasmon and waveguide modes influences drastically the extinction spectrum of the system. Our results explain successfully available experimental data and provide a transparent physical picture of the underlying processes.

The optical properties of materials can be efficiently manipulated by periodic structuring on a length scale comparable to the wavelength of light. Recent advances in electron-beam-lithography and self-assembly nanofabrication techniques allow one to prepare well-defined photonic structures of nanoparticles with a tailored shape, size and arrangement, and observe new, interesting and potentially useful physical phenomena. In the last years, considerable effort is devoted to the investigation of photonic nanostructures made of ionic, semiconducting or metallic materials which have a strongly dispersive, resonant optical response in the frequency region of interest, thus opening up further impressive possibilities for tailoring the light-matter interaction. In the present work we investigate the optical response of metallo-dielectric photonic crystal slabs consisting of a periodic array of gold nanoparticles on top of an indium tin oxide (ITO) film on a quartz substrate, by means of numerical calculations using the layer-multiple-scattering method [1, 2]. This method is ideally suited for the calculation of the transmission, reflection and absorption coefficients of an electromagnetic (EM) wave incident on a composite slab consisting of a number of layers which can be either planes of non-overlapping particles with the same two-dimensional (2D) periodicity or homogeneous plates. For each plane of particles, the method calculates the full multipole expansion of the total multiply scattered wave field and deduces the corresponding transmission and reflection matrices in the plane-wave basis. For homogeneous plates, these matrices are readily obtained in the plane-wave basis. The transmission and reflection matrices of the composite slab are evaluated from those of the constituent layers. The method applies equally well to non-absorbing systems and to absorbing ones; it can also deal with systems containing strongly dispersive materials such as real metals.

An ITO film, of refractive index $n_{\text{ITO}} = 1.90$, on a quartz substrate, of refractive index $n_{\text{sub}} = 1.46$, operates as a waveguide for transverse electric (TE) and transverse magnetic (TM) EM waves [3]. The dispersion curves of the waveguide modes for an ITO film 140 nm thick are
Figure 1. Left panel: Dispersion curves of the TE and TM waveguide modes for an ITO film, of thickness 140 nm, sandwiched between air and quartz. The straight lines $\omega = c q_{\parallel}$, $\omega = c q_{\parallel}/n_{\text{sub}}$, and $\omega = c q_{\parallel}/n_{\text{ITO}}$, which define the light cone in air, quartz, and ITO, respectively, are shown by dashed lines. Right panel: Dispersion curves of the virtual bound states associated with $k_{\parallel} = (k_x, 0)$, with electric field odd and even upon reflection with respect to the $xz$ plane, for a rectangular array ($a_x = 375$ nm, $a_y = 300$ nm) of non-absorbing Drude spheres ($\hbar\omega_p = 3.58$ eV, $\tau^{-1} = 0$), of radius 50 nm, on top of an ITO film, of thickness 140 nm, on a quartz substrate. The dashed lines show, separately, the corresponding dispersion curves of the waveguide modes in the reduced-zone scheme and of the particle-plasmon modes (schematically drawn as horizontal lines), in the absence of interaction between them.

shown in the left panel of figure 1. It can be seen that these modes lie outside the light cone in air and quartz and, therefore, they cannot be excited by an externally incident wave. These modes have the form of plane waves propagating with a wave vector $q_{\parallel}$ parallel to the surfaces of the waveguide (the $xy$ plane) while they are bound and decrease exponentially along the $z$ direction outside the waveguide, on either side of it. However excitation of these modes can be achieved if the waveguide is coated with a periodic monolayer of particles. In this case, because of the 2D periodicity of the structure, an incident plane wave of wave number $q$ generates a series of diffracted plane waves with wave vectors $K_g = k_{\parallel} + g \pm \sqrt{q^2 - (k_{\parallel} + g)^2} z$, where $g$ are the 2D reciprocal-lattice vectors and $k_{\parallel}$ the $xy$ component of the wave vector of the incident wave, reduced within the first surface Brillouin zone (SBZ). If $q < |k_{\parallel} + g|$ we obtain evanescent diffracted beams which can match continuously the corresponding guided waves of the same frequency, polarization, and of the same $q_{\parallel} = k_{\parallel} + g$, provided they have the proper symmetry. In this way, the waveguide modes are no longer bound within the ITO film, but leak in the outer region (virtual bound states) acquiring a finite lifetime (their eigenfrequency becomes complex). Such quasi-guided modes can be excited by an externally incident wave and manifest themselves
as resonance structures in the reflection spectrum. From another point of view, because of the 2D periodicity of the coating layer, the frequency bands associated with the waveguide modes are folded within the SBZ of the given lattice and acquire a small imaginary part because of their mixing with the extended (scattering) states.

\[ \varepsilon_s(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+i\tau)} \]

Let us consider as coating layer a rectangular array, of lattice constants \( a_x, a_y \), of metallic nanospheres described by a Drude dielectric function \( \varepsilon_s(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+i\tau)} \) \[4\]. The optical response of such a particle is characterized by a pronounced resonance due to the excitation of dipole particle-plasmon modes. These are virtual bound states, which correspond to collective electron oscillations at the surface of the particle that cause large enhancement of the local field and strong light absorption, effects which are interesting for a variety of applications in the surface-enhanced Raman scattering, nonlinear optics, optical tweezers, biosensors, solar energy absorbers, etc. The weak interaction between the dipole particle-plasmon modes of the periodically arranged spheres removes the threefold degeneracy of this mode of the single sphere and three relatively flat bands of particle-plasmon virtual bound states are formed.

**Figure 2.** Theoretical (solid lines, right axis) and experimental (dashed lines, left axis) extinction spectra of rectangular arrays of gold nanoparticles on top of an ITO film, 140 nm thick, on a quartz substrate, at normal incidence and polarization along the \( y \) direction. The sequential diagrams correspond to \( a_x \) from 350 nm up to 475 nm with a step of 25 nm, and \( a_y = 300 \text{ nm} \).

**Figure 3.** Theoretical (solid lines) and experimental (dashed lines) extinction spectra of a rectangular array (\( a_x = 425 \text{ nm}, a_y = 300 \text{ nm} \)) of gold nanoparticles on top of an ITO film, 140 nm thick, on a quartz substrate, for incidence in the \( xz \) plane at an angle from 0° to 30° and polarization along the \( y \) direction. The theoretical extinction is scaled to the experimental one. The different spectra are shifted upwards for clarity.
Superimposing these bands to those of the waveguide we obtain a band structure diagram for the virtual bound states of the composite system (see right panel of figure 1). These states, which have a relatively long lifetime (their eigenfrequency has a small imaginary part), can be excited, in general, by an externally incident wave and manifest themselves as pronounced peaks in the reflection and absorption spectra. Bands of the same symmetry interact and repel each other. This mechanism removes, for instance, the degeneracy of the waveguide states at the edges and the center of the SBZ (Bragg gaps). The same mechanism leads also to a hybridization between particle-plasmon and waveguide bands.

We shall now compare our results with available experimental data for rectangular arrays of gold nanodisks, of diameter 100 nm and thickness 20 nm, on top of an ITO film on a quartz substrate [5]. Figure 2 shows theoretical and experimental results for the extinction (negative logarithm of the transmittance) at normal incidence and polarization along the $y$ direction. It can be seen that there is very good agreement as far as the shape and position of the peaks are concerned. However, the magnitude of the calculated extinction is larger than the experimental one (note that theoretical and experimental curves refer to different vertical axes). This discrepancy is ascribed to the fact that, instead of disks, we have considered Drude spheres ($\hbar\omega_p = 3.58$ eV, $\hbar\tau^{-1} = 0.179$ eV) of the same diameter and the polarizability of the sphere is significantly larger than that of the disk [6, 7]. It is worth noting that at normal incidence and polarization along the $y$ direction, while one would expect excitation of the two odd waveguide modes at the center of the SBZ (they are separated by a Bragg gap as shown in the right panel of figure 1), the lower-frequency mode cannot be excited for symmetry reasons. In addition, in this case, the corresponding particle-plasmon mode is also excited and, therefore, two resonance peaks appear in the diagrams of figure 2. By increasing the lattice constant the SBZ shrinks and the waveguide states at the center of the SBZ are shifted to lower frequencies. In this way one can change the position of the waveguide states relatively to that of the particle-plasmon states (and therefore their interaction) in a controllable manner, as shown in figure 2.

Finally, in figure 3, we compare theoretical and experimental results for incidence at an angle in the $xz$ plane and polarization along the $y$ direction. In this case, two odd waveguide modes and one odd particle-plasmon mode (see right panel of figure 1) are excited. As can be seen, very good agreement between theory and experiment, concerning the shape and position of the particle-plasmon and the lower-frequency waveguide resonances, is obtained. However, the agreement is not so good for the higher-frequency waveguide resonance, especially at large angles of incidence. This is due to the fact that the refractive index of ITO decreases with frequency and its value at higher frequencies is considerably smaller than the average constant value $n_{ITO} = 1.90$ that we have assumed.

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