Channeling light along a chain of near-field coupled gold nanoparticles near a metallic film

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Abstract: We study the propagation of light along a chain of 20-nm-spaced gold particles lying onto a silica substrate in which a metallic film can be incorporated. We first discuss, for a pure dielectric substrate, the specificities of the chain modes as compared to larger separation distances where far-field coupling dominates. We then show how the introduction of a buried metallic film allows a substantial increase in the propagation lengths. Finally, we discuss the crosstalk between two adjacent chains, with and without the buried metallic layer, for applications to ultra-compact interconnects.

OCIS codes: (130.3120) Integrated optics devices; (240.6680) Surface plasmons; (260.3910) Metal optics.

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1. Introduction

Guiding light through subwavelength sections still triggers huge efforts motivated by further miniaturization in integrated photonics. Among the potential configurations, periodic chains of metal nanoparticles have received lots of attention [1]. Such plasmonic chains allow strong lateral field confinement due to the electromagnetic coupling between individual localized surface plasmon (particle plasmons) supported by each of the particles. Their optical properties have been studied for several years, both theoretically and experimentally [2, 3, 4, 5, 6, 7, 8] showing abilities specially for focusing down to 3D (subwavelength) volumes [9, 10] and guiding within compact areas [11, 12, 13].

In this later context, a dipolar model is sufficient to show that propagation in chains can be supported both by transverse and longitudinal modes associated with different inter-particle coupling schemes [4]. However, for particles bigger than a few tens of nanometers, the shape and dimensions of the particles matter and more elaborated numerical methods are required to account for the multipolar character of each of the units [14]. Independently of the structural parameters, significant modification of the chain's transmittance could be achieved by changing the nature of the substrate on which the chain sits. In particular, the particle plasmon can be substantially affected by the presence of a nearby electromagnetic surface mode such as those introduced by a dielectric waveguide [15, 16] or a metal- dielectric interface [17]. The proximity of a nearby surface mode offers a new de-excitation channel for the particle plasmon [18] but can also be used to increase the interaction range between neighbor units [19]. One aspect of our work is to study how the introduction of a metallic layer allows to consequently increase the propagation lenth of the light along the chain.

In this article, full 3D calculations based on the Green dyadic method are used to investigate numerically the effect of different substrates (pure dielectric versus mixed metallic/dielectric) on the guiding efficiency of light along short and long chains of small gold nanoparticles. The chosen structure is closely related to the one studied in ref. [2], where it has already been shown experimentally and theoritically that the short distance between two particles induces a strong collective response of the chain. The full resolution of Maxwell equations first provides some new insights in the physics of light transmission through chains lying on pure dielectric substrates. In particular, we discuss the influence of the parity of the number of units forming the chain on the nature of the transmission modes. These results are discussed and corroborated

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by using a mechanical analogy with a spring-mass chain. We then show how the introduction of a noble metal layer at a determined distance below the chain enables a significant increase in the propagation length exploiting both surface plasmons and guided modes. In counter part, it is shown how this increase downgrades the lateral field confinement of the propagating fields.

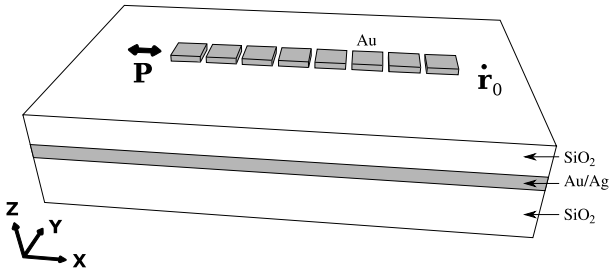

Fig. 1. A chain of gold particles deposited above a multilayered structure made of dielectric and gold. The system is illuminated by a dipole located 100 nm from the side of the first particle. The transmission of the chain is computed by evaluating the amplitude of the electric field at a point $\mathbf{r}_0$ located 100 nm from the side of the last particle. The particles are $100 \times 100 \times 20$ nm^3, and the period of the chain is 120 nm or 200 nm. The substrate is either a semi-infinite-silica space, or a 40-nm-thick layer of gold or silver separated from the chain by 100 nm of silica.

The system under study is formed by a chain of N gold nanoparticles, placed on a silica half-space in which a metallic film can be incorporated, few tens of nanometers beneath the particles level (Fig. 1). In all the following simulations we focus on parallelepiped particles with 100 nm $\times$100 nm basis and 20 nm high. The chain period is 120 nm or 200 nm, corresponding to a spacing between two adjacent particles of 20 nm and 100 nm, respectively.

The chain is optically addressed by a dipole at one of its ends and the transmitted light is detected at the other end as sketched in Fig. 1. This dipole couples to the chain mode arising from the electromagnetic coupling of the individual localized surface plasmons supported by each particle. The exact interaction of the dipole field with the chain was modeled using the Green's function method [20, 21]. The index of gold and silver are taken from Johnson and Christy [22].

2. Chain on pure silica substrate

In this section, we first look at a reference configuration where the substrate is a pure, semi-infinite silica space of refraction index $n = 1.5$. For the dimensions considered here, an isolated particle exhibits a localized surface plasmon resonance at $\lambda = 760$ nm.

Figure 2 plots the evolution with the number N of particles of the near field transmission (amplitude of the electric field at the observation point divided by the amplitude of the incident electric field 10 nm before the first particle) of the chain. Four different sets of curves associated to four different combinations of the interparticle distance and the polarization of the incident dipole are presented. In Figs. 2(a) and (b) the separation distance is 20 nm while in Figs. 2(c) and (d) it is 100 nm. The excitation dipole, parallel to the interface, is parallel to the chain axis (along the x axis) in Figs. 2(a) and (c) and perpendicular to it (along the y axis) in Figs. 2(b) and (d). These two illumination configurations are referred in the following as longitudinal and transverse illumination, respectively.

The four cases of Fig. 2 present a lot of noticeable differences. First, the number of resonances is smaller, for a same orientation of the excitation dipole when the distance between

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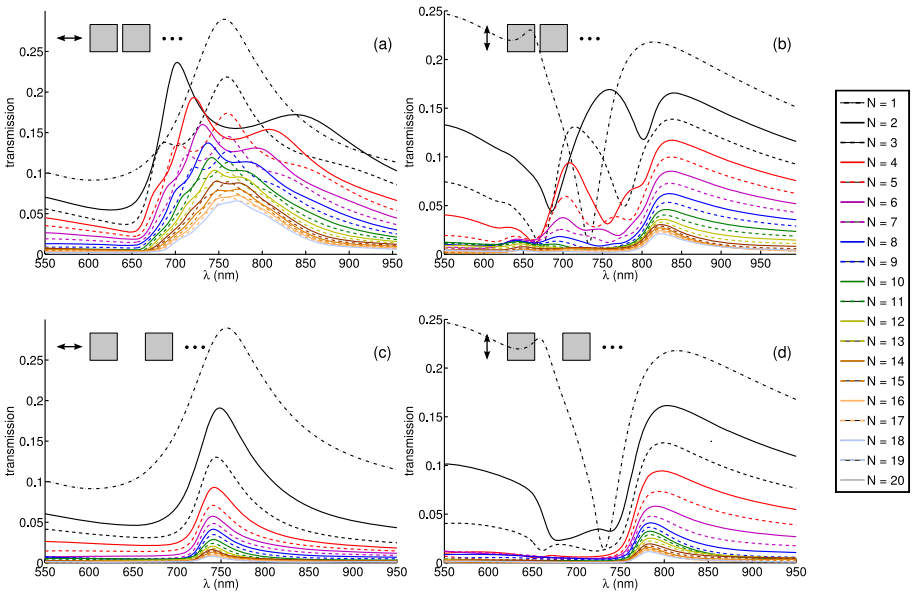

Fig. 2. Amplitude of the electric field at a point $\mathbf{r}_0$ located 10 nm above the surface and 100 nm away from the side of the last particle, as a function of the wavelength, and for an increasing number of particles ; (a) and (b), the spacing between two particles is 20 nanometers ; (c) and (d), the spacing is 100 nm ; (a) and (c) the excitation dipole is parallel to the direction of the chain (x axis) ; (b) and (d) the excitation dipole is perpendicular to the chain, and parallel to the interface (along the y axis).

two particles is 100 nm. The spectra feature essentially one main resonance, both for the longitudinal and the transverse modes (Figs. 2(c) and (d)). Conversely, the spectra are strongly multimodes when the distance is 20 nm (Figs. 2(a) and (b)), particularly for the longitudinal modes.

In this last particular case of Fig. 2(a), it appears clearly that the spectra are qualitatively different for chains with an even or odd number of particles. For example we can notice, for an odd number of particles, the presence of a central peak around 760 nm, whose position does not change with the length of the chain. A simple interpretation can be given considering a mechanical analogy with a system made of N masses relied by springs (see Fig. 3). A computation of the spectrum (for example the amplitude of the last mass under the excitation of the first) shows the same evolution with the mass number. The central-frequency resonance corresponds to the mode in which one mass out of two does not move, the movement of the two neighboring masses being symmetric. Hence, the moving masses are isolated from all the others and the mode frequency is the same as for the single mass. This situation can only occur for an odd number of masses, because the modes must be either symmetric or antisymmetric with respect to the middle of the chain. By analogy, in the optical case the light intensity is weaker inside one particle out of two, hence the neighboring particles are isolated from all the others as the coupling is the largest between the nearest neighbors: the chain behaves as $\approx N/2$ independent gold particles.

The amplitude of the electric field decays more slowly with the length of the chain in the longitudinal mode for a 20 nm interparticle distance. Indeed the coupling mechanism is not the same as with 100 nm gaps. When the distance is of a few tens nanometers, the particles are

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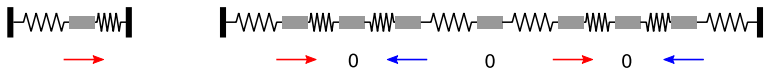

Fig. 3. Mechanical analogy of the longitudinal mode of the gold particles chain with a chain of small masses relied by springs. Left: single mass ; Right: displacement of the masses in the mode analog to the central peak in the transmission spectra of Fig. 2(a): one mass out of two is fixed, the two adjacent masses move in opposite direction.

coupled through the evanescent components of the electric field (near field coupling). It is a well-known fact that the electric field intensity is enhanced mainly outside the particle sides which are perpendicular to the direction of the dipole induced inside the particle: the electric field amplitude is enhanced by a factor equal to the ratio of the particle susceptibility to the background susceptibility. For this reason, the coupling coefficient is expected to be larger for an excitation dipole parallel to the chain than perpendicular to the chain, in the near field coupling regime.

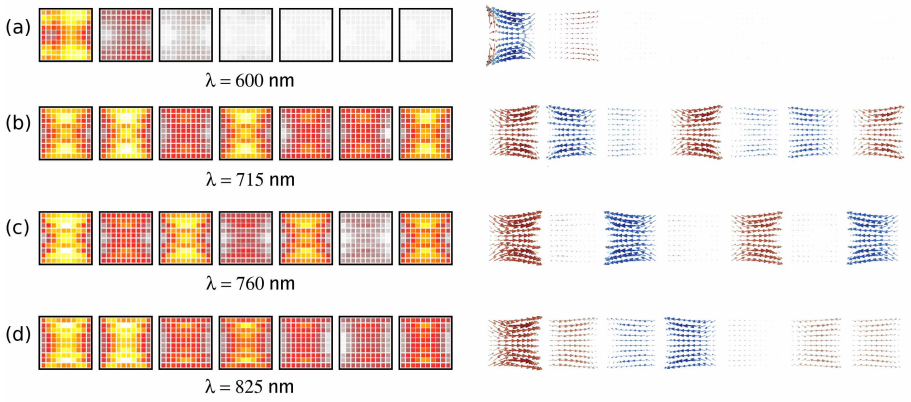

Fig. 4. Left: amplitude of the electric field inside a chain of 7 particles on a silica substrate, out of resonance (a), and for the three main resonances (b,c,d). Right: polarization of the electric field for the three same resonances. Blue arrows point to the left, whereas red arrows point to the right.

When the spacing increases to 100 nm, the particles are coupled mainly through the propagative components of the light, as the range of the near-field components is of few tens nanometers at these wavelengths. Since a dipole emits energy essentially perpendicular to its orientation, the coupling coefficient should be larger when the excitation dipole is perpendicular to the chain, making the propagation length shorter in the longitudinal mode compared to the transverse mode.

In Ref. [13], the transverse mode was found to guide the light clearly better than the longitudinal mode, whereas in the present case, they seem to have comparable propagation length, at least for chains up to 20 particles. Calculations carried out for larger N shows that the transmission decays slightly faster for the longitudinal mode than for the transverse mode. This difference of behavior with Ref. [13] could be attributed to the fact that each particle of the chain is 20−nm thick in this article, whereas it was 40−nm thick in Ref. [13]: a thicker particle increases the radiative coupling in the transverse mode and then increase the propagation length.

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It is interesting to plot the electric field amplitude and polarization inside the chain at the different resonance wavelengths of Fig. 2(a). In Fig. 4(a), $\lambda = 600$ nm, the excitation dipole is out of resonance, and the light does not propagate along the chain. In the mode plotted in Fig. 4(c), $\lambda = 760$ nm, one particle out of two is off, as explained previously. The polarization vector plot shows clearly that the electric field is opposite inside the two neighboring particles, similarly to what can be expected from the velocity direction in the mechanical analogy. This is a consequence of the symmetry of the associated mode with respect to the center of the chain which is a necessary condition to cancel the field inside one particle out of two. The structure of the two remaining modes (Figs. 4(b), $\lambda = 715$ nm, and (d), $\lambda = 825$ nm) are very similar concerning the amplitude of the electric field, but it appears clearly on the polarization plots that the sign of the electric field changes a larger number of times at $\lambda = 715$ nm (4 times) than at $\lambda = 825$ nm (2 times), which makes the first one a higher frequency than the second one.

3. Influence of a metallic film

With the aim of evaluating the possibility to increase the ligth propagation length along the chain, we study in this section the influence of a 40−nm-thick metallic film placed 100 nm under the chain. The main idea is to exploit the associated surface plasmon mode as an additional channel to assist the energy propagating along the chain. In addition to the surface plasmon at the gold-silica interface, the silica slab between the chain and the gold film is thick enough to support a TM guided mode, which can further contribute. Figure 5 shows plots similar to Fig. 2 for the new systems including the metal layer. Figures 5(a) and (b) correspond to a gold film, whereas Figs. 5(c) and (d) is for a silver film. Compared to the case of the silica semi-infinite substrate, the surface plasmon of the single particle is red shifted by 45 nm (single particle) because of its interaction with the metallic layer, both for silver and gold film. In general, the resonances of the chain are slightly more blue-shifted for the silver film compared to the gold film.

Let us at first consider the case of the longitudinal illumination. The spectra are very similar to what has been computed without the metallic film, at least for the short chains (up to 10 particles). The main noticeable difference occurs for chains longer than 10 particles for the gold film and 14 particles in the case of silver film. In both cases, a dip appears around $\lambda = 800 - 850$ nm. The position of this dip shifts slowly to the red while increasing the length of the chain. We can notice a second dip which appears for $N = 8 - 9$ particles around $\lambda = 660$ nm. When the gold film is replaced by a silver film, the dip is still present around the same position, but is not so important. In fact, calculations realized for longer chains show that in the case of a silver film the dip reaches a minimum value for 40 particles, against 25 particles for the gold film. The inset of Fig. 5(a) shows the distribution of the electric field amplitude inside the 18-particles chain (gold film) at 820 nm. It clearly appears that the amplitude is modulated by a cavity-type mode : the amplitude is minimum at the 4th or 5th particle and the 17th particle, which corresponds to a distance of 1440 nm between the two nodes. An effective wavelength of about $2500 - 3000$ nm can then be inferred, which does not correspond to the wavelength of any mode supported by the background (guided or surface plasmon). The only length scale which matches this value is the beating between these two modes. Indeed, the surface wavelength of the guided mode is 660 nm, whereas the surface wavelength of the surface plasmon mode is 522 nm. The corresponding beating (distance between two nodes) is about 2500 nm. Moreover, the dip and the cavity structure do not occur if the chain is coupled to a single silica slab, without metallic film, when is removed the contribution of the surface plasmon mode (not shown here). Hence, it appears that the intensity at the output of the chain is modulated by its interaction with the two modes supported by the substrate: when the node of the superposition of the two modes coincides with the end of the chain, the transmission reaches a minimum. Note that the

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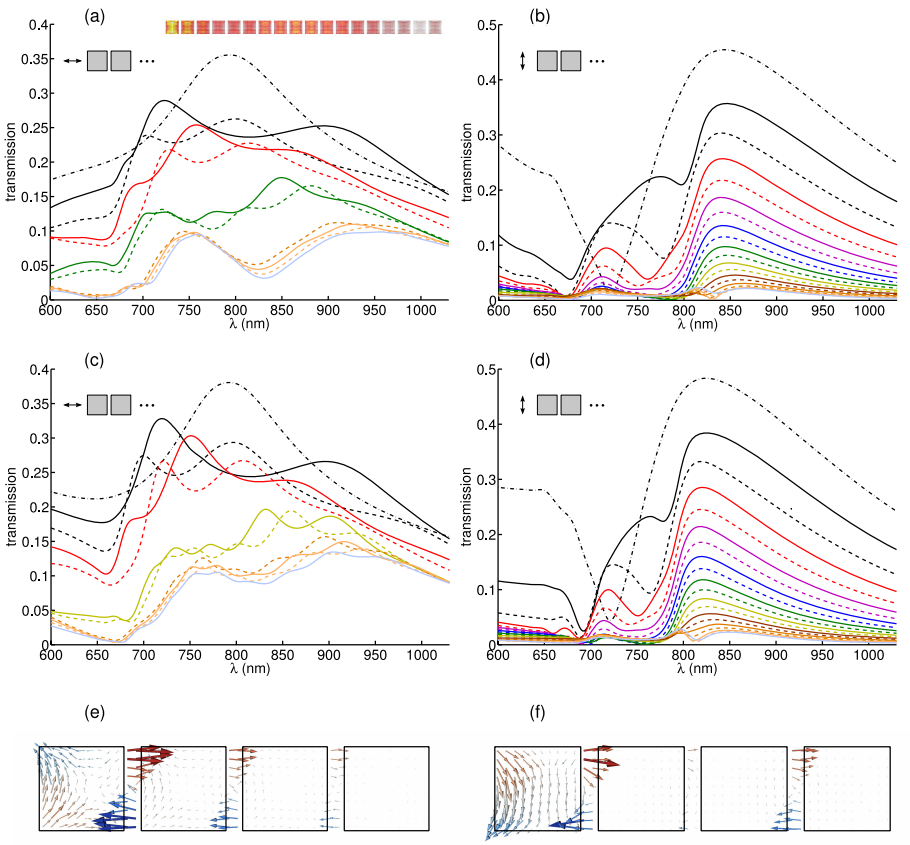

Fig. 5. Same as Fig. 2, but the chain is separated from a 40-nm-thick metallic film by a 100-nm-thick layer of silica and the distance between two particles is 20 nm ; (a) and (b): gold film ; (c) and (d): silver film ; (a) and (c) the excitation dipole is parallel to the direction of the chain (x axis, only few characteristic curves have been displayed, see Fig. 2 for legend) ; (b) and (d) the excitation dipole is perpendicular to the chain, and parallel to the interface (along the y axis) ; (e) and (f): amplitude and polarization map of the electric field for the two main transverses modes of a chain of 4 particles (gold film).

exact location of the minimum depends on the phase of the two guided modes supported by the background, which depends itself on a complex manner on the chain and source parameters.

In the case of transverse illumination (Figs. 5(b) and (d)), the spectra show much less differences with the half-infinite silica substrate curves. The positions of the resonances are almost not modified by the presence of the metallic film. However, a small lateral peak appears in the blue of the main resonance at 860 nm for chains longer than 17 particles. Calculations performed for longer systems show that this secondary maximum appears and disappears regularly when N increases, and induces a small increase of the transmission when the chain gets longer. However, the transmission stays much lower than in the longitudinal mode. In Figs. 5(e) and (f) are plotted the electric field distributions inside a chain of 4 particles, for the two main resonances of the spectra plotted in Fig. 5(b). The largest arrows correspond to the electric field inside the gap space in between two adjacent particles, which is enhanced compared to the value inside the particles due to the contrast of susceptibility between the air and the metal. This

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two figures show clearly that the two main resonances differ essentially by the phase relation between two successive cavities. In the smaller resonance (shorter wavelength), the cavities are in phase, whereas in the main resonance, the cavities are shifted by $\pi/2$.

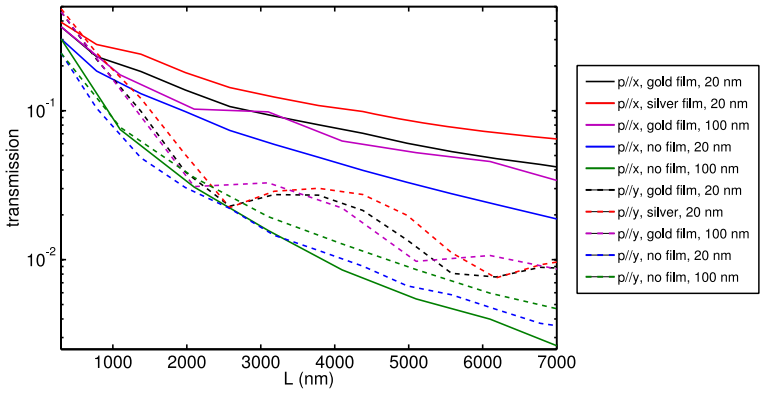

Fig. 6. Maximum of transmission as a function of the distance from the dipole source to the excitation dipole, for the different substrate types computed in the article.

Figure 6 synthetizes the evolution with the distance between emitting dipole and the observation point of the maximum of the transmission spectra for all the cases previously studied. The distance evolves from 300 nm (one particle) to 7000 nm (60 particles with a spacing of 20 nm, or 35 particles with a spacing of 100 nm). To perform this calculations, the discretization cell has been increased up to 20 nm, compared to 10 nm for the short chains calculations. This remains valid because the particles are very thin, the electromagnetic field is almost constant in the thickness of the particles. The increase of the mesh size results essentially in a small shift of the resonances to the red (by about 10 nm) and an overestimation of 5% to 10% of the electric field amplitude, but the overall structures of the transmission curves are in very good agreement. The curves of Fig. 6 have been computed for one particle and 5 to 60 particles by step of 5 for a spacing of 20 nm, and 5 to 35 particles for a spacing of 100 nm. The propagation of light is clearly longer in the longitudinal mode (solid curves) than in the transverse mode (dashed curves). The most efficient guiding is obtained when the chain is coupled to a metallic film, the best being a silver film because the losses are lower than for gold. In this last case, the propagation length was found to be 8.0 micrometers (amplitude) by fitting the end of the transmission curve by an exponential. Without the metallic film, the transmission drops significantly faster and is more sensitive to the distance between the particles. This is due to the shorter coupling range between two particles in absence of any guided mode and surface plasmon mode. The propagation length of the longitudinal mode is 3.5 micrometers when the spacing is 20 nm. The transverse mode allows a much lower transmission, although the coupling to a metallic film succeed in improving it. The oscillations of the transmission in presence of a metallic film are due to the periodic appearance of the secondary peak described previously. The amplitude of light at the observation point depends weakly on the distance between two particles in the transverse mode when there is no metallic film. This is a consequence of a coupling essentially based on the radiative components of the dipolar field, which decay slower than the near field.

Finally, an important parameter for the integration of photonic structures at the nanoscale is the minimum distance between two components under which they can operate independently without significant crosstalk. In the present case, this corresponds to the distance under which the light, injected inside one chain, starts to couple and propagate along an adjacent chain. Fig-

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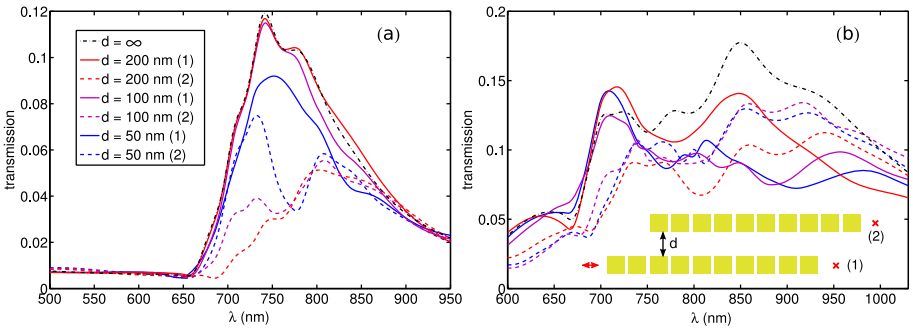

Fig. 7. Transmission spectra of the chain at the exit of the first chain (solid lines) or the second chain (dashed lines), for a space between the chains equal to 50 nm, 100 nm, and 200 nm.

ure 7 presents the transmission at the exit of two adjacent chains separated by a varying distance *d*, without (Fig. 7(a)) and with (Fig. 7(b)) the underlying gold film. When the chain lies on the silica half space, the distance has to be larger than 100 nm in order to minimize the perturbation due to the neighboring chain. However, the amplitude at the exit of the second chain is still not negligeable, particularly around 800 nm. For the resonance at 740 nm, the ratio of the electric field amplitude in (1) to the electric field amplitude in (2) is 4.3. When the distance is 50 nm, the two chains are clearly not isolated anymore, and the electric field amplitude in (2) equals at least 50% of the electric field amplitude in (1) on the whole spectrum. The coupling between the two chains occurs essentially through the radiative components of the light emitted from the first chain, which decreases as a power law of the distance *d*. When the gold film is added under the chain, a stronger crosstalk is observed, and the chains are still strongly coupled at a distance of 200 nm. This a consequence of the coupling occurs now essentially through both the guided and the surface plasmon mode supported by the background, whose ranges decay now at least as an exponential law, much slower than a power law at short distances from the source.

4. Summary

In this article we have studied the transmission properties of closely-packed-gold nanoparticles chains. Two cases were considered: in the first one, the chain lies on a half-infinite-silica substrate whereas in the second case, the chain is separated by 100 nm silica from a metallic film. For a short spacing (20 nm) between two particles, the spectra show complex longitudinal transmission spectra. Unlike what is known for a interparticle distance of 100 nm, here the longitudinal mode channels light further away than the transverse mode, because the coupling is obtained through the near field scattered by each particle. Comparison with [13] showed that the long-range dominance of either the transverse or the longitudinal mode depends noticeably on the distance and the volume of each particle. In particular, when the spacing is 100 nm, a thicker particle ([13]) increases the radiative coupling and then the range of the transverse mode, which can then propagate on a larger distance than the longitudinal mode. Also, for a small number of particles (less than 20), a clear distinction between chains with an odd or an even number of particles can be done, which originates in the symmetry of the excited modes. Beyond, we show that the addition of a metallic film under the chain allows to increase the propagation length of light. A silver layer allows the best transmission, because of its smaller propagation losses compared to gold. The propagation length is increased by a factor of two

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when the silver layer is added. Finally, we observed that the crosstalk between two adjacent chains is high, particularly when the chains are placed above a metallic film, because the two chains are then coupled through the surface modes supported by the substrate. This effect could be decreased by increasing the localization of light either using a higher index dielectric between the chain and the metallic film, or removing the material between the chains in order to prevent the perpendicular light propagation.

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