Cumulative plasmon field enhancement in finite metal particle chains

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The dramatic field enhancement at the extremity of finite chains of strongly coupled gold nanoparticles illuminated under total internal reflection is investigated numerically. We demonstrate that high enhancement factors can be achieved by exploiting the in-plane forward scattering of the particles, with geometries achievable by state-of-the-art lithographic techniques. © 2005 Optical Society of America

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Optical addressing of isolated nanometer-scale entities remains one of the main challenges of nano-optics. Matching the confinement of the illuminating fields with the dimensions of the specimen is required for maximizing matter–light interaction. For this purpose, it has been proposed to use the low dimensionality of plasmon fields bound to resonant metal nanostructures for focusing light from a propagating laser beam down to a subwavelength (sub-λ) volume.[1]

Essentially, two main configurations have been investigated to address this issue. The first approach relies on strong field gradients generated under appropriate illumination conditions at the extremity of a sharply elongated metallic tip.[2,3] The metal geometry gives rise to a strong concentration of surface polarization charges at the tip apex that is responsible for dramatic field enhancement with regard to the incidence. This effect, which was recently corroborated,[4] is used in apertureless scanning near-field optical microscopy for probing surface fields with a sub-λ resolution.[5]

The second configuration uses near-field coupling between closely spaced metallic nanoparticles.[6] The hot spot observed in the interparticle nanometer gap can be seen as a consequence of the hybridization of the single-particle localized plasmon modes.[7] Recent calculations suggest that a bowtie nanoantenna geometry significantly improves the field enhancement factor.[8] However, the latest experiments yielded lower values than expected from the theory mainly because current electron-beam lithography tools do not allow for accurate control of the spacing distance with nanometer precision.[9] Another elaboration of this concept based on decreasing particle radius ensembles has been proposed.[10] The results show a cascade enhancement effect where the last and smallest particle benefits from near-field scattering from its neighbors. Although the enhancement factors that were reported reached unprecedented values, here again the structural parameters may render experimental implementation of this second configuration difficult.

In this Letter, keeping in mind the limitations of nanofabrication, we propose a configuration based on finite chains of identical strongly coupled gold nanoparticles. Such a configuration has been investigated

previously for light-energy transfer from a local source.[11–13] It is shown that, under extended illumination, this configuration can provide an intense and non-diffraction-limited free-space hot spot of similar magnitude to that reported for nanometer gap systems (e.g., Refs. 6 and 8).

The configuration under study is sketched in Fig. 1. A regular chain of N gold nanoparticles lying on a glass substrate is illuminated by a p-polarized plane wave under an incidence angle Θ. The nanoparticles are defined by parallelepipeds with a 100 nm side square basis and a height of 20 nm. Despite its simplicity, this geometry has shown to be well suited to restoring the experimental near-field optical response of metal dots fabricated by electron-beam lithography.[14,15]

The interparticle distance is fixed at 20 nm. For this value, consecutive particles strongly interact through near-field coupling, so each of them experiences a field that is the sum of the plane-wave illumination and the scattering from its nearest neighbors. In the particular case of total internal reflection incidence, the resulting in-plane forward scattering of the particles is expected to create a cumulative
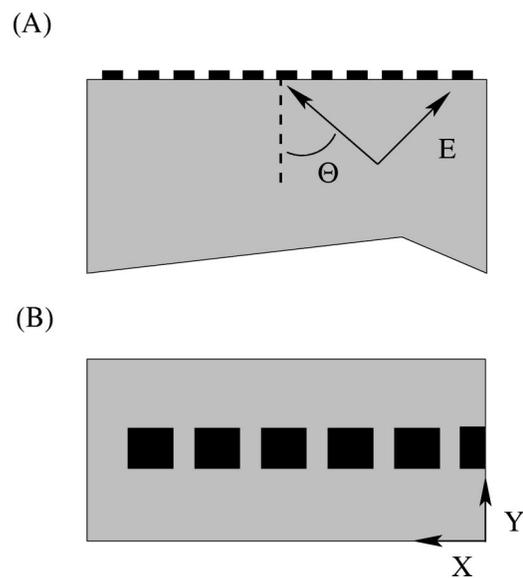
(A)

(B)

Fig. 1. Description of the optical configuration: (A) side view, (B) top view.

field increase along the chain and to give rise to an intense field at its extremity.

To study the optical properties of our system, the Green dyadic method is used to model both its far-field spectroscopy and the associated optical near-field map. This formalism, also known as the classical-field susceptibilities method, provides a self-consistent resolution of the Maxwell equations, accounting for the multipolar response of metallic nanoparticles.[16] We have recently shown that consideration of multipolar particle modes is required for a full description of near-field coupling in a chain of metallic nanostructures with the dimensions considered here.[13]

To start our numerical analysis, we calculate the evolution of the scattering power spectrum of a finite gold particle chain as a function of the number N of particles when $\Theta = 60°$.[17] The results, plotted in Fig. 2, indicate the presence of two main regimes. For short chains ($N \leq 5$), the spectrum is dominated by the successive order of multipolarlike modes resulting from the coupling of the localized surface plasmons of the single particles [Fig. 2(a)]. For $N=2$, the dipolar peak that dominates the spectrum of an isolated particle in the wavelength range that we consider is dramatically redshifted from 720 to 810 nm. This well-known effect can be explained by the neutralization of the polarization surface charges formed at the particle surface.[18] A further shift occurs when a third particle is added, together with the appearance of a quadrupolarlike resonance that becomes significant for $N=4$ and dominates for $N=5$. For longer chains ($N \geq 10$), a different regime that is less sensitive to N and is characterized by the presence of two peaks is built [Fig. 2(b)]. At lower wavelengths the main peak is slightly redshifted when the chain length is increased, whereas the secondary peak displays the opposite tendency.

To obtain a further understanding of the far-field properties of the longest chains, we calculated the influence of the incidence angle Θ (Fig. 3). For Θ values [Fig. 3(a)] smaller than the critical angle for total reflection $\Theta_c = 41.8°$, much smoother spectra are observed, confirming the implication of forward scattering of the particles in the chain response. The sharp resonant features observed in Fig. 2(b) appear for $\Theta > \Theta_c$ only when the forward scattering becomes parallel to the substrate interface [Fig. 3(b)] and consequently contributes to the coupling with the first
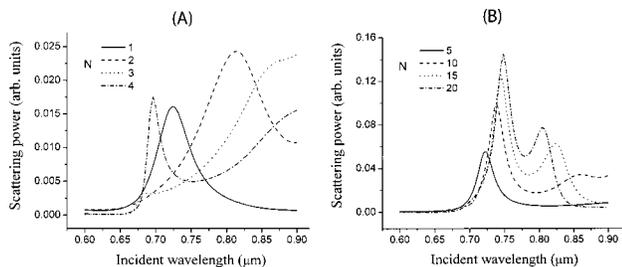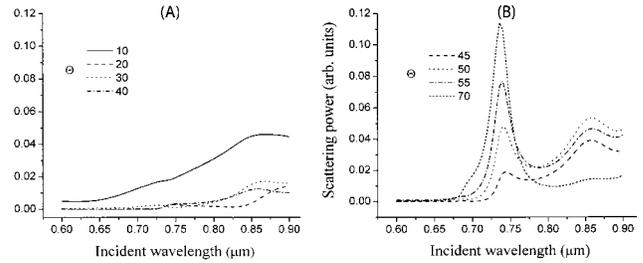

Fig. 3. Evolution of the scattering spectrum of a 10-particle chain as a function of the incidence angle Θ.
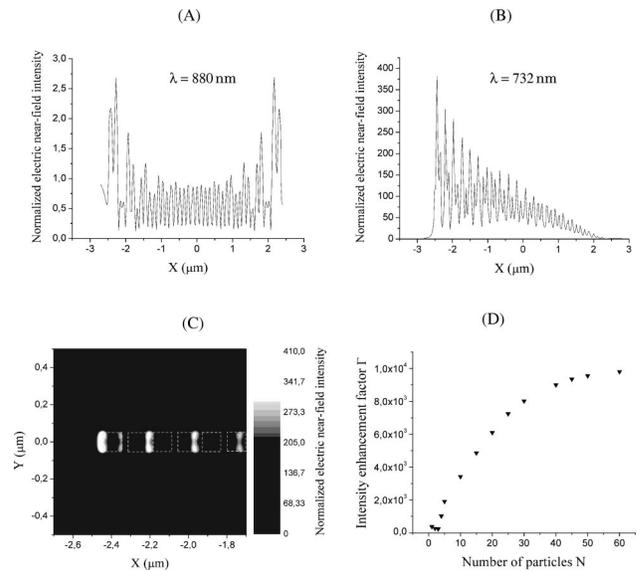

Fig. 4. Electric near-field intensity profiles ($Y=0$) calculated 20 nm above a 40-particle chain for (A) $\Theta=0$ ($\lambda = 880$ nm) and (B) $\Theta = 60$ ($\lambda = 732$ nm). (C) Near-field intensity map corresponding to (B). (D) Evolution of the enhancement factor Γ with the number N of particles.

neighbor. The strength of the resonances increases with the incident angle before reaching a threshold for $\Theta = 70°$. Despite the discrete character of the chain, the blueshift of both peaks when the incidence angle is increased, and thus the in-plane incident wavelength $\lambda_{\parallel}$ is decreased, suggests some kind of string mode. This hypothesis is corroborated by the appearance of this regime from a minimum chain length.

From the far-field analysis of the finite gold particle chains, we calculate the corresponding near-field optical distribution. Figures 4(a) and 4(b) show the electric field intensity profile, 20 nm above a 40-particle chain ($Y=0$), for $\Theta=0$ and $\Theta=60°$ at their respective main resonances. The normalization has been done with respect to the incident field intensity at the same position in the absence of any gold structure.

Under normal incidence, the electric near-field extends symmetrically over the whole chain with a maximum at both extremities. These maxima, independent of the incidence direction, result from the sudden jump in the dielectric function. An illumination under total reflection dramatically increases the

Fig. 2. Evolution of the scattering power spectrum of the chain as a function of the number N of gold particles for $\Theta = 60°$.

asymmetry of the field profile. The cumulative effect makes the field intensity increase from the first pad to the last. The maximum value localized at the left extremity corresponds to a dramatic field enhancement with regard to the incidence (i.e., a gain of ~ 150 times compared with $\Theta = 0$). The fact that the periodicity of the oscillation does not fully match the pitch of the chain, together with the overall modulation, seems to confirm the string-mode-type resonance. We have additionally observed that, for an incident wavelength corresponding to the secondary resonance, the field intensity is lower and displays a different distribution.

As expected, the highest field intensity enhancement is found at the half-height of the chain in front of the last pad. Its evolution as a function of the chain length has been computed for $\Theta = 60°$ (Fig. 4). The curve $\Gamma = f(N)$ follows a linear increase before reaching an asymptotic value $\Gamma = 10^4$ for $N \geqslant 40$. To rule out any effect from edges, the method of detuning in Ref. 19 has been applied. These values are much higher than for a single particle ($\Gamma = 300$) and are commensurable with what has been reported for dimers displaying a nanometer separation distance.[6,8,20]

To conclude, we have shown that the optical properties of finite chains of gold nanoparticles are strongly dependent on the illumination conditions. The in-plane forward scattering of the particles under total internal reflection contributes in strengthening their coupling, leading to sharp resonances in their scattered light. These resonances attributed to a stringlike mode correspond in the near-field zone to an intense and localized hot spot at the chain extremity that may be of interest for applications such as surface-enhanced Raman scattering or biosensing.

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