Out-of-plane plasmon-polariton modes in two-dimensional nanoparticle arrays

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Plasmon-polaritons in a square, two-dimensional lattice of noncontacting nanoparticles are studied using the coupled dipole model. The approach considers diffractive losses out of the plane and also for the vector nature of the electromagnetic field, and is thus a three-dimensional theory of the two-dimensional nanoparticle array. We consider the out-of-plane polarization, where long range coupling is present, and present the case where the plasmon energy crosses the light cone near the M-point in the Brillouin Zone. In this case, only a restricted set of nonradiative modes are present.

Keywords: Surface plasmon; nanofabrication; ultrasmall.

Se estudian los plasmares-polaritones en un arreglo cuadrado de partículas, sin tocarse unas con otras, usando el modelo del dipolo acoplado. La aproximación considera pérdidas difractivas fuera del plano y por la naturaleza del vector del campo electromagnético, siendo un modelo tridimensional de un arreglo tridimensional de nanopartículas. Consideramos la polarización fuera del plano, donde se presenta acoplamiento de largo alcance y presentamos el caso donde la energía del plasmon cruza el cono de luz cerca del punto-M de la zona de Brillouin. En este caso solo está presente un conjunto restringido de modos no radiactivos.

Descriptors: Plasmon superficial; nanofabricación; ultrasmall.

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1. Introduction

Arrays on noncontacting noble metal nanoparticles (NP) have garnered a great amount of recent interest. The Plasmon polariton (PP), a hybrid plasmonic optical mode, has the potential to allow for a number of applications, such as interconnects, detectors, and lenses. In this letter, we examine the square 2D nanoparticle array (NPA) in particular. These types of arrays are of interest due to the defined surface plasmon (SP) resonances that occur in nanoparticles, in comparison to the broad resonances of metal sheets.

Some studies of 2D arrays have appeared in the literature. Sakoda [3] simulated using the finite-difference time-domain method the dispersion curves of a square arrays of metal cylinders. The finite-element method was used in [4] to explore negative refraction in arrays. In Ref. 5, the multiple multipole method was used to analyze triangular lattices in addition to the square lattice. In all of these studies, the arrays were of composed infinite cylinders, and out-of-plane losses were not accounted for.

This paper examines the out-of-plane-Plasmon-polariton modes in square-lattice NPA’s using a coupled-dipole model. This model follows the work done with respect to 1D NP chains [13, 14]. We treat a NPA, with the NP diameter δ and the center-to-center inter-NP spacing d. The dipole approximation has been shown to be accurate except in the case of NP’s that are nearly in contact [17]. The NP’s are treated as oscillating point dipole and coupling within the array is accounted for by the the retarded dipole field. More specifically, we apply this technique to show that square arrays can be suitably designed such that all SPPs are radiative with the exception of a small region in the Brillouin Zone near the M-point.

2. Calculations

The dipolar Green’s function (GF) $G_{\alpha}(q, \varepsilon)$ for excitation wavevector $q$ and mode $\alpha$, can be used to describe the PP’s. This treatment captures both radiative PP’s [12], the fields

Figure 1. Diagram of the nanoparticle array.
scattered by the NPA, and nonradiative surface polaritons, the
nearfield standing waves. The PP’s are given by the singular-
ities of $G^{-1}_\alpha(q, \varepsilon)$,

$$G^{-1}_\alpha(q, \varepsilon)\delta_{qq'} = \left( \frac{(\varepsilon^2 - \varepsilon_p^2 - i\gamma_p)}{2\varepsilon_p} - \Sigma_0 - \Sigma_\alpha(q, \varepsilon) \right) \delta_{qq'}, \tag{1}$$

with $\varepsilon_p$ and $\gamma_p$ the the single-NP surface-plasmon reso-
nance energy and nonradiative damping, respectively. $\Sigma_0$ is
the radiative decay of a single NP SP, and the re-
tarded dipole-dipole coupling is contained within the radiative
self-energy (SE)

$$\Sigma_\alpha(q, \varepsilon) = \sum_{h,j=-\infty}^{\infty} \Sigma_{h,j,\alpha}(\varepsilon) e^{i(q_{hd}+q_{jd})},$$

which is simply the retarded dipole-dipole energy per NP (the
prime indicating that the $(h, j) = (0, 0)$ term is not included).
We divide the solution to $G^{-1}_\alpha(q, \varepsilon) = 0$ into real and imag-
inary components $E_\alpha(q)$ and $i\Gamma_\alpha(q)$ and use the pole ap-
proximations

$$E_\alpha(q) \approx \varepsilon_p + \text{Re}\Sigma_\alpha(q, \varepsilon_p)$$
and

$$\Gamma_\alpha(q) \approx -\text{Im}\Sigma_\alpha(q, \varepsilon_p)/\hbar.$$

The SE is the dynamic dipole-dipole energy associated
with NP’s $(h, j)$ and $(h', j')$.

$$\Sigma_{(h,j)-(h',j')}(\varepsilon) = \mathbf{E}_{(h,j),(h',j')} \cdot \mathbf{p}_{(h',j')}$$

with $\mathbf{E}_{(h,j),(h',j')}$ the oscillating [as $\exp(-i\omega t)$] electric field
associated with the SP of NP $(h, j)$ at the position of NP
$(h', j')$ and $\mathbf{p}_{(h',j')}$ is the dipole moment of the SP of NP
$(h', j')$. A schematic diagram of the NPA is shown in Fig.
1. Based on the approach of Refs. [13, 14, 18, 19], the nonvan-
ishing SE for the mode transverse to the plane of the array is

$$\Sigma(q, \varepsilon) = n\gamma_0 \sum_{h,j=-\infty}^{\infty} \left\{ \frac{1}{k^3(h^2 + j^2)^{3/2}} \right. \right.$$  
$$\left. \left. - \frac{i}{k^2(h^2 + j^2)} - \frac{1}{k(h^2 + j^2)^{1/2}} \right\} \right.$$  
$$\times e^{i\frac{k}{2}(h^2 + j^2)^{1/2}} e^{i(q_{hx}+q_{yx})}, \tag{2}$$

where $\gamma_0 = \omega_p^2 / (4\pi\sigma_0 c^3)$, $\hbar \omega = \varepsilon$, $\epsilon_0$ is the electric con-
stant, $k = d\omega e^{1/2}/c$ is the dimensionless optical wavevec-
tor in the background medium, $q = q d$ is the dimensionless
excitation wavevector, $\varepsilon$ is the dielectric constant, which in-
cludes the gain of the medium, and $n = \sqrt{\varepsilon}$ is the complex
index of refraction. It should be noted that the out-of-plane
mode SE is completely decoupled from modes that exist in
the plane of the NPA, and the PP’s are purely transverse. In
order to compute the SE in dimensionless form, we divide the
SE by $\gamma_0$ such that $\sigma(\bar{q}, \bar{k}) = \gamma_0^{-1} \Sigma(q, \varepsilon)$.

3. Results

We turn our focus on the complex SE. We assume reason-
able values for interparticle spacing $d = 75 \text{ nm}$ and nanopar-
icle size $\delta = 50 \text{ nm}$. We consider Au NP’s since the radia-
tive decay time is on the order of the nonradiative dephas-
ing time [22]. The embedding medium is chosen to have
Re $n = 3.5$, with complex dielectric constant $\epsilon = n^2$, and a
small amount of loss is added for numerical stability. We
approximate the shift of $\varepsilon_p$ due to the embedding medium:
The SP energy in air is taken to be \( (\varepsilon_p)_{air} \). The SP energy in air is taken to be \( (\varepsilon_p)_{air} = 2 \text{ eV} \) [20–22, 24].

Figure 2 shows the dispersion \( \text{Re}\sigma(\bar{q}, \bar{k}) \), and Fig. 3 shows the radiative loss \(-\text{Im}\sigma(\bar{q}, \bar{k})\) as functions of \( \bar{q} \) for the Z-mode, which is purely transverse. In this case, the photon dispersion has undergone zone folding almost everywhere in the BZ once it crosses the SP dispersion, except in a small region close to the M-point. We see that \( \text{Re}\sigma(\bar{q}, \bar{k}) \) exhibits overall negative phase velocity with an additional feature at the crossing with the light cone \( \bar{q} = \bar{k} \). This appears as a sharp decline beyond which the dispersion is mostly constant. The radiative width in a small region close to the M-point is such that the SP-like SPPs are nonradiative; elsewhere in the BZ, radiative decay is prevalent. At this particular interparticle spacing, beam forming can occur.

4. Conclusion

We have presented a theory for the out-of-plane polarized PP's in NPA's with square symmetry. We have examined a region where the light cone has undergone zone folding near the X-point, but not in a small region near the M-point. This \( \bar{q} \) dependence allows for the restriction of nonradiative dark modes that do not have radiative losses to a small region in the Brillouin Zone.

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