

# Strong Coupling of Gold Dipolar Nanoantennas by Symmetry-Breaking in Evanescent Wave

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**Abstract:** Observing the resonance wavelengths of nanoantennas (NAs) with changing incident angles in TM and TE polarization. Extinction cross section shows the dark and bright coupling modes at resonance wavelength of NAs with symmetry breaking oblique incidence. The plasmonic enhancement is stronger under evanescent wave in total internal reflection.

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## 1. Introduction

Localized surface plasmon resonance (LSPR) for metallic nanoparticle dimers have catch a lot attention in recent years[1-6]. The plasmonic resonance between metal-dielectric interface has been investigated in many applications, such as bio-detection[7], fluorescence enhancement[8], active optoelectronic components[9], metamaterials[10], and surface enhanced Raman scattering (SERS)[11]. Nanoantennas (NAs), the metallic nanoparticle dimers, have shown the different resonance modes because of the dipolar bonding mode and antibonding mode[12, 13]. Numerous investigations attempt to analyze the plasmonic coupling mode with the particle-dimer-like-array structure because of its high-potential application[14, 15].

For identical nanoparticle dimers, the resonance wavelength of different coupling modes will be changed to the corresponding phase of electric displacement field by TM and TE polarization fields in different oblique incidence angles. In the case of metallic nanoparticle dimers, the in-phase (out-of-phase) response of two dipoles is called the bonding (antibonding) mode or bright (dark) mode, resemble to molecular orbital theory [16, 17]. The bonding mode could be coupled easily by normal incidence, but in symmetric structures of NAs, the antibonding mode could not be coupled by normal incidence. The antibonding mode resonance of NAs still gather a lot of attention owing to the slower radiative decay and narrower linewidths. The antibonding mode resonance of NAs can be predicted by the plasmonic Hamiltonian theory.

The antibonding mode resonance of NAs can be predicted by the plasmonic Hamiltonian theory. The electronic Hamiltonian of identical metallic particle dimers could be written as[18],

$$H = H_{pl} + H_{eh} + H_{pl-eh} \quad (1)$$

The plasmonic part in Hamiltonian equation is as,

$$H_{pl} = \sum_{n=1}^2 \hbar \tilde{\omega}_n b_n^\dagger b_n + \hbar \Omega f(\theta) (b_1^\dagger b_2 + b_1 b_2^\dagger + h.c.) \quad (2)$$

where the index n is used to identify each sphere, neutral nanoparticles of radius  $a_n$  (each contain  $N_n$  electrons). The localize surface plasmon (LSP) frequency is,

$$\omega_n = \sqrt{\frac{3N_n e^2}{m_\epsilon a_n^3 (\epsilon_d^{(n)} + 2\epsilon_m)}}, \quad \tilde{\omega}_n = \omega_n \sqrt{1 - \frac{N_{out,n}}{N_n}} \quad (3)$$

The two nanoparticles through their near fields, are giving rise to second term in the right-hand side of Equation 2, where

$$\Omega = \frac{1}{2} \Pi \left( \frac{\tilde{\omega}_n}{1 - N_{out,n} / N_n} \right)^{1/2} \left( \frac{a_n}{d} \right)^{3/2} \quad (4)$$

and

$$f(\theta) = 1 - 3 \cos^2 \theta \quad (5)$$

here,  $d$  is the center-to-center nanoparticle distance and  $\theta$  is the angle formed by the polarization of the nanoparticles and the dimers axis joining the two nanoparticles.

For the general case of unequal frequencies,

$$\omega_{\pm} = \sqrt{\frac{\tilde{\omega}_1^2 + \tilde{\omega}_2^2}{2} \pm \sqrt{4\Omega^2 \tilde{\omega}_1 \tilde{\omega}_2 f^2(\theta) + \left( \frac{\tilde{\omega}_1^2 - \tilde{\omega}_2^2}{2} \right)^2}} \quad (6)$$

The two plasmonic eigenmodes corresponding to the coherent oscillation of the two nanoparticles are shown in **Figure 1**. For  $\theta = 0$ , the low-energy (high-energy) mode with frequency  $\omega-$  ( $\omega+$ ) can be thought of as the in-phase (out-of-phase) motion of the two nanoparticles. Vice versa for  $\theta = \pi/2$  the  $-$  and  $+$  modes correspond to the out-of-phase and in-phase motions, respectively.

## 2. Result and discussion

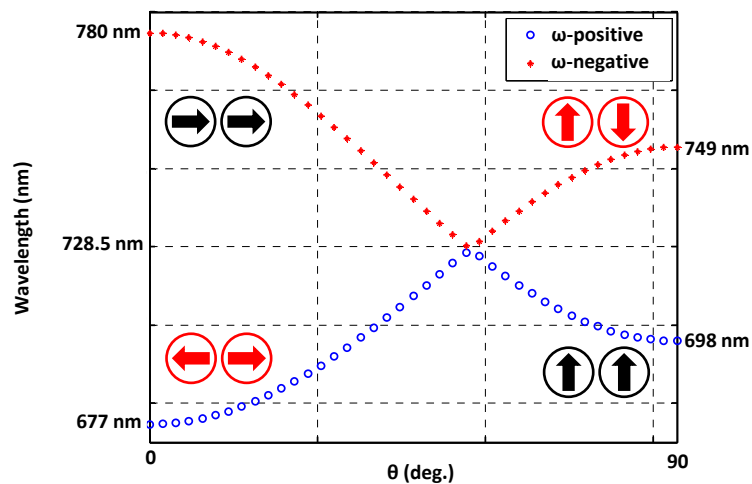


Fig. 1. The theoretical analysis results. Resonance wavelengths are calculated by the Eq. (6). By consider Eq. (6), two frequencies could be observed. The higher frequency is named  $\omega$ -positive (circle), and the other one is named  $\omega$ -negative (star).

The different direction of dipole resonance existing in each nanoparticle have been shown in Fig. 1 respectively. The four types of plasmonic coupling modes in Fig. 1 are longitudinal bonding (LB) mode, longitudinal antibonding (LA) mode, transverse bonding (TB) mode, transverse antibonding (TA) mode, and these modes corresponding to the wavelength 780 nm, 677 nm, 698 nm, and 749 nm, respectively.

The extinction spectra in both FEM simulation and experiment results are compared at wavelengths from 600 to 880 nm. The refractive index of the substrate is 1.52 (glass), the surrounding medium is water ( $n = 1.33$ ). Comparing Fig. 1 with Fig. 2(d), 2(e), and 2(f), the theoretical analysis is fitted well with the FEM modeling not only for the case of single particle, but also for the case of NAs. In Fig. 2(e), the red shift of the resonance wavelength could be observed when the incidence angle is larger than  $30^\circ$ . At the normal or small angle oblique incidence ( $< 30^\circ$ ), the resonance wavelength is at 780 nm which corresponds to the LB mode in Fig. 1. When the incidence angle is larger than  $30^\circ$ , the other resonance mode happens and is located at 680 nm, which corresponds to the LA mode in Fig. 1. According to the analysis, the great agreement has been shown not only for the resonance wavelength, but also for the plasmonic coupling mode, too.

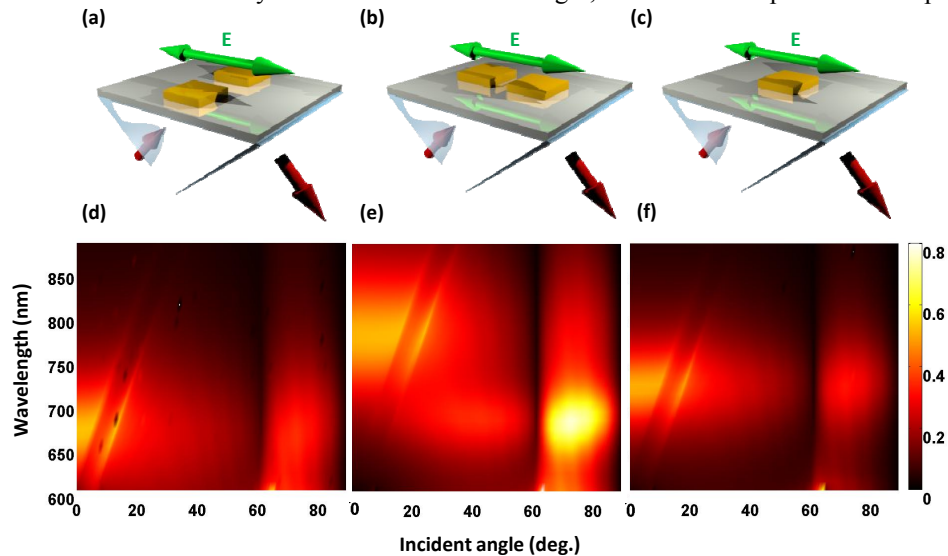


Fig. 2. Two-dimensional mapping extinction spectra with TM-polarize incidence light for incidence angles ranging from  $0^\circ$  to  $89^\circ$  and wavelengths ranging from 600 to 880 nm of (a), (d) TM transverse type antennas, (b), (e) TM longitudinal type antennas and (c), (f) TM type single particle. Red arrows indicate the direction of light.

Very interestingly, in Fig. 2(h), when incidence angle is larger than critical angle, the strong enhancement of coupling efficiency in antibonding mode has been shown.

### 3. Conclusion

Hybridization model of plasmonic NAs and the coupling resonance wavelengths could be predicted by solving the plasmonic Hamiltonian equation. Different types of the coupling modes could be found when changing incidence angles. In addition, by comparing with the reflectance spectra and near-field distribution, the enhancement of coupling efficiency to antibonding mode of NAs under the evanescent wave could be explained without the complex molecular orbital theory. Not only the simulation and theoretical analysis, the experiment has been applied to improve the credibility of this study. Bonding (bright) mode and antibonding (dark) mode could be coupled in both TM and TE polarization wave in NAs array. In future application, the antibonding mode could apply to improve the sensitivity for the sensor or quantum optics because of the high quality factor [3] and the slower radiative decay.

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