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Gain-modulated plasmonic Rabi oscillations of coupled nanocomplex

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ABSTRACT

Strong coupling in nanostructures can bring intriguing optical phenomena such as ultrafast Rabi oscillation—periodical energy exchange phenomenon between two modes. Rabi splitting appears in the frequency-domain spectra for strong coupling system. However, in metallic nanosystems the time-domain Rabi oscillations are hard to be observed because the plasmon lifetime is limited by the heavy ohmic losses. Here we report a theoretical investigation of surface plasmon coupling behaviour of two gold nanorods with one being a core-shell rod filled with a gain material and find the periodic energy exchange phenomenon which recalls the concept of Rabi oscillation. The gain material-cored gold-shell structure dipolar mode hybridizes with the solid gold rod quadrupolar mode to form the Fano resonances. Energy exchange between the two rods happens through the near field coupling. Two approaches, to prolong plasmon lifetime by increasing the gain efficiency and to increase Rabi oscillation frequency by increasing the coupling strength, are suggested to increase the Rabi oscillation cycles. Our results offer a way to achieve unique control of light at the nanoscale and further to explore plasmonic Rabi oscillation phenomena in plasmonic nanosystems.

1. Introduction

Metallic nanostructures supporting local surface plasmon resonances have attracted large interest in recent years due to their unique properties of selected optical absorption and near field enhancement [1,2] and based on the advanced synthesis and characterization techniques [3–5]. The optical property of metallic monomer is determined by the shape and material [6,7]. The plasmon coupling between metallic nanoparticles is also of great importance to influence their optical properties [8–11]. Furthermore, the coupled plasmon modes will provide more favourable optical properties for technological applications, such as enhancing optical nonlinearity [12,13], photo catalysis [14], biological sensors [15], and solar energy harvesting [16].

Several phenomena, such as plasmon hybridization [8], Fano resonance [17,18], plasmonic splitting and energy transfer [19,20], have been found when the surface plasmon coupling happens in the spectra-overlapped nanostructures. The plasmonic Fano resonance that arises from the interference of a bright plasmon mode and a dark plasmon mode is of great interest. Various structures such as dolmen-type resonators [21,22], nano oligomers [23–27], ring-disk cavities [28–30], heterodimers [21–33], plasmon-exciton systems [34], Fanoshells [35], and split ring dimers [36,37] have been reported to support Fano resonance. Although the plasmonic nanostructures support excellent optical properties, they suffer the heavy ohmic losses at optical frequency range. From 2003, the creative idea of introducing optical gain material to the metallic nanostructures has enable us to overcome the challenges [38–45], and at the same time the surface plasmon amplification by stimulated emission of radiation (spaser) has been realized [39,40,43]. The concept of spaser was first put forward by Bergman and Stockman in 2003 via embedding either metal or semiconductor inclusions into an active medium to realize energy radiationless transitions from a two-level emitter to the plasmon quasi-static electric field [38]. In 2009, the nanoscale spaser was experimentally demonstrated [39,40]. Later works theoretically investigated the properties of metal nanoparticles with the gain inside and realized the extremely high local field enhancement and the sharp optical spectra [41–43]. This type of structure could be used for single-

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molecule detection [43].

Rabi oscillation could also be realized in plasmonic nanosystems. When two excited states are under the simultaneous excitation with a broad-band source, the periodical energy exchange happens between these two states, which is called Rabi oscillations [46–49]. The key element for the formation of Rabi oscillations is the coupling strength between the two energy dissipative resonators. The coupling strength between the oscillators should exceed their individual linewidths, so that the period of Rabi oscillations can be smaller than the oscillator lifetimes and the periodical energy exchange can be observed before the energy dissipation [46]. Plasmonic Rabi oscillations have been recently observed in the metal/J-aggregates nanostructures by monitoring the differential reflectivity spectrum as a function of time delay between the pump and probe pulses [50]. However, very few Rabi oscillation cycles have been observed because of the short plasmon lifetime which is caused by the strong ohmic losses in the metal at the optical frequencies.

In this paper, we investigate the coupling property of metallic nanostructures with an optical gain material inside. We find that the gain material-cored gold-shell structure shows the extreme local field enhancement and the extremely narrow extinction profile when the gain efficiency approaches a certain value as previously reported in Ref. [43]. By introducing a solid gold rod, whose quadrupole plasmon mode (dark mode) overlaps with the dipole one (bright mode) of the core-shell structure, the expected Fano resonance and the peak splitting is observed. By increasing the gain efficiency, the two extinction peaks become narrower while the splitting width is almost kept, which meets the requirements for Rabi oscillations. The Rabi oscillation is characterized by the near field intensity. The time-domain simulation results and the frequency-domain simulation results are consistent to show the Rabi oscillations in the nanodimer. With the large gain efficiency, the Rabi oscillation cycles are increased and the plasmon lifetime is extended. At different parts of the dimer, the two coupled modes show different strengths. Another approach to increase the Rabi oscillation cycles is to decrease the gap distance between the dimer. By decreasing the gap, the mode splitting width is increased while the linewidths of two hybridized modes are kept. Within the limited plasmon lifetime, the Rabi oscillation cycles are increased by the high energy exchange frequency. Furthermore, the anticrossing behaviour of the dispersion relation is displayed by changing the solid rod length.

2. Methods

The time-domain and frequency-domain simulations are performed with the software (FDTD Solutions and Comsol Multiphysics) based on the finite-difference time-domain (FDTD) method and finite element method (FEM) due to the advantages of their methods on time-domain simulations and on solving gain material problems, respectively. Perfectly matched layers are used to avoid spurious reflections at the outmost boundaries for both methods. For FDTD simulations, a total-field scattered-field (TFSF) source (a linearly polarized plane wave) is used to excite the plasmon. The absorption cross sections are obtained by monitoring the net power flowing inward and toward the source region, and the scattering cross sections are obtained by monitoring the power flowing outward the source region. For the FEM simulations, the absorption cross sections are obtained by integrating the resistive losses in the nanoparticles, and the scattering cross sections are obtained by integrate the Poynting vector at all the particles surface. The extinction cross section is the sum of the absorption and scattering cross sections.

The background medium dielectric constant ε_b is set to be 1.7689, and that of gold is taken from Ref. [51]. The real physical objects of the gain material with specified parameters can be molecules, semiconductor nanocrystals or rear-earth material, which are externally pumped at some proper frequency far away from the investigated frequencies. Without pumping, the material could not serve as gain. The coupling properties of exciton (without pumping) and plasmon were extensively studied and this subject is termed as plexcitonics, which are not the concerns of our work. The gain material is modelled with a single Lorentzian emission line shape [41]: $\varepsilon_h(\omega) = \varepsilon_b - \varepsilon_{gmax} \times \gamma_g/[2\ (\omega - \omega_g) + i\gamma_g]$, where ε_{gmax} denotes the extreme value of ε_h which is related to the gain efficiency. The negative ε_h value means it is not a gain material but a dissipative material. γ_g is related to the emission linewidth and set to be 4.25×10^{14} s^{-1}. ω_g is the emission center and set to be 2.31×10^{15} s^{-1}.

The schematic diagram of the dimer excited by external electromagnetic source and its geometry parameters are shown in Fig. 1a. For the core-shell structure, the outer diameter and the height (d and l_1) are both 40 nm, and the shell thickness t is 4 nm. For the solid gold rod, the high l_2 and the diameter d are 228 nm and 40 nm, respectively. The gap g is 20 nm. The incident light polarizes along the long axis of the nanorods. The simulation results when the gain efficiency is 0 for the two simulation methods are shown in Fig. S1 (Online Resource 1). The results from two methods are consistent, which confirms the validity of our calculations.

3. Results and discussion

We start our work by investigating the individual gain material-cored gold-shell structure in the frequency domain. The optical cross sections of the core-shell structure with different core gain efficiencies ε_{gmax} are shown in Fig. 1b. With zero gain, the optical cross sections show a broad peak at 745 nm. When ε_{gmax} reaches 0.6715, the scattering and absorption become extremely high and narrow (extreme plasmon amplification). The absorption becomes negative because the losses are compensated by the gain material. When ε_{gmax} is further increased to 0.9, the line widths of scattering and absorption cross section come back to a large value and the large plasmon amplification almost vanishes. The electric field distributions corresponding to the peaks in Fig. 1b are shown in Fig. 1c. The largest field enhancement happens at the point of 0.6715. Considering the cross sections and near field distributions, we can physically understand why with this value the large plasmon amplification happens. With low gain efficiency, the absorption is partially compensated by the gain material. The absorption gradually becomes weak and then negative when the gain efficiency is increased. The compensation of ohmic losses will result in the enhancement of near field, which is the signature of enhanced local surface plasmon resonance (LSPR). The enhanced LSPR will lead to the high scattering efficiency. With the gain efficiency being 0.6715, the high positive scattering and the high negative absorption lead to the zero extinction and the strong near field. With the gain efficiency exceeding this value, the extra energy emitted from the gain material may interfere with the LSPR, which hinders the local field enhancement as well as the light scattering and absorption efficiency. In short, with low gain efficiency, the loss hinders the plasmon amplification, and with high gain efficiency, the extra gain interrupts the amplification. We have also changed the emission profile of the gain material in Fig. S2 (Online Resource 1) and found that as long as the dielectric constant of the gain material at the angular frequency of 2.31×10^{15} s^{-1} is 1.33^2 - 0.6715i, the extreme plasmon amplification will happen. So the line shape of the emission spectra is not so important. The really important factor is the emission efficiency of the gain material around the