Diffractively induced transparency and extraordinary light emission at the plasmon band edge

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Abstract

We demonstrate Electromagnetically Induced Transparency-like spectral line shapes for periodic arrays of optical antennas covered by a thin layer of quantum dots. The coupling of localized surface plasmons to diffraction orders radiating into the quantum dot layer leads to the opening of a plasmonic stop gap in the dispersion relation of the array. Standing waves with an enhanced density of optical states are formed at the band edge, leading to an extraordinary light emission enhancement across subwavelength apertures.

1. Introduction

The arrangement of materials with shape-dependent resonances in an orderly fashion has been the leading workhorse in the field of optical metamaterials [1]. Complex arrangements of metallic nanostructures often lead to non-trivial spectral line shapes arising from the interaction of Localized Surface Plasmon Resonances (LSPRs). Atomic physics phenomena leading to similar line shapes have been a major inspiration for understanding and developing these metamaterials. Seminal examples are Fano resonances [2] and Electromagnetically Induced Transparency (EIT) [3], which have re-emerged in the context of plasmonic metamaterials [4, 5]. In EIT, destructive quantum interference between different excitation pathways induces a narrow transparency window within an absorption band. Light propagating in such an atomic medium acquires a reduced or even vanishing group velocity, and enhanced nonlinearities, at frequencies of reduced absorption. Plasmonic metamaterials offer similar effects in a more simple, robust, and classical platform. Herein, we demonstrate that EIT-like behavior emerges from the interplay between surface plasmon resonances and diffraction orders radiating in the plane of an array, i.e., Rayleigh Anomalies (RAs). This form of Diffractively Induced Transparency (DIT) arises in the context of 3 coupled plasmonic resonances: one localized, and two lattice-induced. The mutual coupling of lattice resonances leads to the opening of a plasmonic stop gap [6]. By having resonant interaction between light-emitting Quantum Dots (QDs) in the vicinity of the array and the DIT/band-edge plasmons, we observe an extraordinary emission enhancement across subwavelength apertures.

2. Results and Discussion

We investigate a periodic array of silver nanorods fabricated by substrate conformal imprint lithography. A SEM image of the array is shown in Fig. 1(a). The rods have dimensions $290 \times 110 \times 20$ nm^3, and the lattice constants are $a_x = 350$ nm and $a_x = 200$ nm. A 20 nm layer of Si_3N_4 was deposited on top of the array to passivate the silver and prevent emission quenching. We spin-coated a 60 nm layer of colloidal CdSe/CdS core/shell QDs on top of the Si_3N_4 layer. Figure 1(b) depicts a side-view of the structure. Figure 1(c) shows the variable angle s-polarized extinction of the array, defined as $1 - T_0$, with T_0 the zeroth-order transmittance. The measurements are shown as a function of the incident photon

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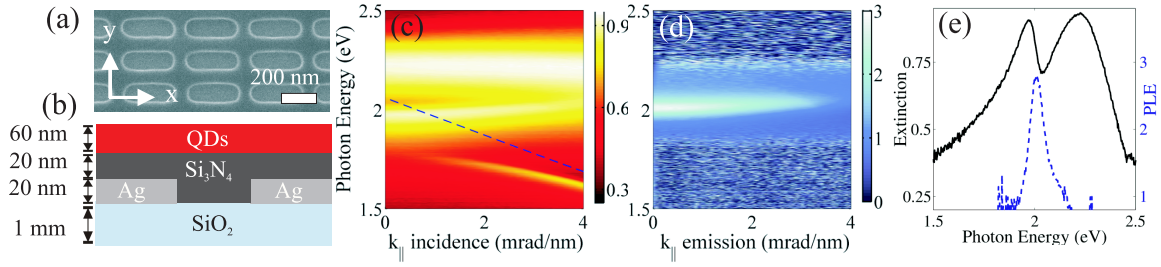

Fig. 1: (a) SEM image of Ag nanorod array, (b) side-view of the investigated structure, and dispersion relation in (c) extinction , and (d) PhotoLuminescence Enhancement (PLE) of s-polarized light, (e) cuts at $k_{||} = 0$ of (c) and (d) as solid black and dashed blue lines, respectively.

energy and wave vector component parallel to long axis of the antennas, i.e., $k_{||} = k_0 \sin(\theta_{in})\hat{x}$, with k_0 the magnitude of the free space wave vector and θ_{in} the angle of incidence. The extinction peak at 2.2 eV corresponds to the dipolar LSPR along the short axis of the nanorods. Its constant frequency and linewidth throughout the angular spectrum are the signature of localized resonances. The dispersion of the RAs follows from the conservation of the parallel component of the wave vector, i.e., $k_{out}^2 = (k_x \pm m_1 G_x)^2 + (k_y \pm m_2 G_y)^2$, with k_{out} the magnitude of the scattered wave vector, $k_{||} = (k_x, k_y)$ the wave vector components parallel to the surface, the integers (m_1, m_2) defining the order of diffraction, and $\vec{G} = (G_x = \frac{2\pi}{a_x}, G_y = \frac{2\pi}{a_y})$ the reciprocal lattice vector. The blue dashed line in Fig. 1(c) indicates the (-1,0) RA taking into account the frequency-dependent real part of the effective refractive index of the QD layer as determined from ellipsometry measurements. The coupling of LSPRs to RAs leads to plasmonic resonances on the low energy side of the RAs; these are known as Surface Lattice Resonances (SLRs) [7, 8]. SLRs associated with different diffraction orders may also couple to each other, opening a frequency gap in the dispersion relation of the array and forming standing waves at the band edge [6].

Figure 1(d) shows the variable angle s-polarized PhotoLuminescence Enhancement (PLE) of the array, defined as I_{in}/I_{out}, with I_{in} and I_{out} the emission of the QDs inside and outside the array, respectively. A 2.8 eV pump laser with a power far below saturation (confirmed by measurements not shown here) impinges from the top (see Fig. 1(b)), at an angle of 5 degrees. The variable angle emission is collected from below. This emitted light that leaks through the subwavelength apertures between the nanorods can be regarded as an emission analog to Ebbesen's extraordinary transmission in the complementary structure of subwavelength hole arrays [9]. Including the aperture/metal ratio in the plane of the array in the PLE measurements, as done in Ref. [9], would make the peak enhancement ~ 7 rather than ~ 3.

We shed light into the coupled nature of SLRs through an analogy with 3 mutually coupled harmonic oscillators [6]. The first oscillator represents the charge oscillation in the nanorods, driven by a harmonic force $F_0 e^{-i\omega t}$ representing the electromagnetic field. The second and third oscillators represent the RAs, i.e., the diffracted waves to which the nanorods couple. The equations of motion of the coupled system are,

$$\begin{aligned} \ddot{x_1} + \gamma_1 \dot{x_1} + \omega_1^2 x_1 - \Omega_{12}^2 x_2 - \Omega_{13}^2 x_3 &= F_0 e^{-i\omega t}, \\ \ddot{x_2} + \gamma_2 \dot{x_2} + \omega_2^2 x_2 - \Omega_{12}^2 x_1 - \Omega_{23}^2 x_3 &= 0, \\ \ddot{x_3} + \gamma_3 \dot{x_3} + \omega_3^2 x_3 - \Omega_{13}^2 x_1 - \Omega_{23}^2 x_2 &= 0, \end{aligned} \tag{1}$$

with x_j, γ_j, and ω_j $(j = 1, 2, 3)$ the displacement from equilibrium position, damping, and eigenfrequency associated with the j^{th} oscillator, and Ω_{jk} $(k = 1, 2, 3$ and $j \neq k)$ the coupling frequency between the j^{th} and k^{th} oscillator. We calculate the dissipated power by oscillator 1, $P(t) = F\dot{x_1}$, in analogy to the dissipated optical power by the electrons in the nanorod. Integrating $P(t)$ over one period of oscillation and scanning the driving frequency ω yields an absorbed power spectrum P_{abs}, which

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we compare in Fig. 2 to the extinction at $k_{||} = 0$ and $k_{||} = 2.5$ mrad/nm (displaced by 0.5). We set $\omega_2 = 2.04$ rad/s at $k_{||} = 0$, $\omega_2 = 2.05$ rad/s and $\omega_3 = 1.73$ rad/s at $k_{||} = 2.5$ mrad/nm. The dip in P_{abs} at ω_2 and ω_3 corresponds to the DIT effect. Figure 2(b) shows the phase of the oscillator, ϕ_1; a kink appears near ω_2 and ω_3. For frequencies far from these kinks, ϕ_1 increases from 0 to π, passing by $\pi/2$ near ω_1 - the expected resonance behavior for an uncoupled oscillator.

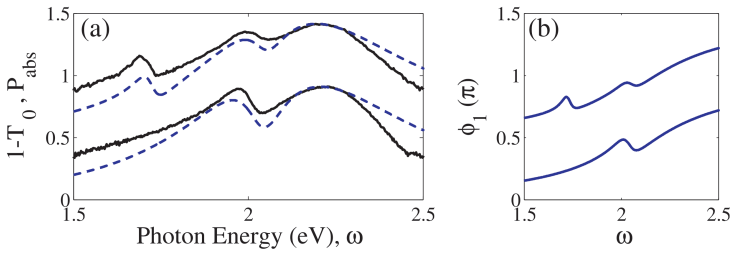

Fig. 2: (a) Extinction at $k_{||} = 0$, and $k_{||} = 2.5$ mrad/nm (displaced by 0.5) as solid black lines, and absorbed power calculated with the model described in the text as dashed blue lines. (b) phase of the first oscillator in the model yielding the spectra in (a).

3. Conclusion

In conclusion, we have demonstrated that EIT-like behavior emerges from the coupling of Rayleigh anomalies to surface plasmons in metallic nanorod arrays. We named this phenomenon Diffractively Induced Transparency, and showed its potential to achieve an extraordinary light emission enhancement across subwavelength apertures in nanorod arrays.

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