

LIGHT SCATTERING AND PHOTON STATISTICS OF QUANTUM EMITTERS COUPLED TO METALLIC NANOPARTICLES

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ABSTRACT. We study theoretically the quantum optical properties of hybrid artificial molecules composed of an individual quantum emitter and a metallic nanoparticle. The coupling between the two systems can give rise to a Fano interference effect which strongly influences the quantum statistical properties of the scattered photons: a small frequency shift of the incident light field may cause changes in the intensity correlation function of the scattered field of orders of magnitude. The system opens a good perspective for applications in active metamaterials and ultracompact single-photon devices. We also demonstrate with accurate scattering calculations that a system constituted by a single quantum emitter (a semiconductor quantum dot) placed in the gap between two metallic nanoparticles can display the vacuum Rabi splitting.

1. Introduction

Control over the interaction between single photons and individual optical emitters deserves great importance in quantum science and quantum engineering. Recently, substantial advances towards the realization of solid state quantum optical devices have been made coupling single quantum dots (QDs) to high-finesse optical cavities [1]. An inherent limitation of cavity quantum electrodynamics (QED) is that the size of the cavity is at least half wavelength and practically much more than that owing to the presence of mirrors or of a surrounding photonic crystal. Unlike optical microcavities, metallic nanoparticles and metallic nanostructures are able to focus electromagnetic waves to spots much smaller than a wavelength [2]. In this way it is possible to increase the local density of the electromagnetic modes as in microcavities but with ultra-compact structures. The ability of metallic nanoparticles and nanostructures to control the radiative decay rate of emitters placed in their near field has been widely demonstrated. This ability stems from the existence of collective, wave-like motions of free electrons on a metal surface termed surface plasmons (SP). An outstanding demonstration of the cavity-like behavior of metallic nanoparticles is the recent realization of a nanolaser based on surface plasmon amplification by stimulated emission of radiation (spaser) [3]. Here we present very recent results on the nonperturbative interaction between quantum emitters (QEs) (e.g. semiconductor quantum dots (QDs)) and metallic nanoparticles (MNPs). In particular we present two cases: i) we describe the quantum optical properties of a single QE-MNP hybrid artificial molecule [4] (see Fig. 1a);

ii) we study the scattering properties of a single QE placed in the gap between two metallic nanospheres [5].

2. QE-MNP hybrid artificial molecule

We consider a spherical QD interacting with a spherical MNP of radius r_m , separated by a distance R (see Fig. 1). There is no direct tunneling between the MNP and the SQD ($R - r_m - r_{QD} > 2$ nm, being r_{QD} the QD radius). The coupling mechanism is due to dipole-dipole interaction. The QE is modeled as a two level system with a dipole moment μ , that is a good approximation for high-quality QDs when studying optical processes at frequencies resonant with the lowest energy excitonic transition. The coupling mechanism between the two units is the dipole-dipole interaction. Further details can be found in [4]. Figure 1b displays the dependence of the coupling rate g on the QD-MNP distance R . Throughout this section we use a dipole moment $\mu = er_0$ with $r_0 = 0.7$ nm (corresponding to 33.62 Debye), being e the electron charge and $\epsilon_b = 3$. In the following we use for the QD transition a linewidth $\gamma_x = 50\mu\text{eV}$. Figure 2a displays scattering spectra as

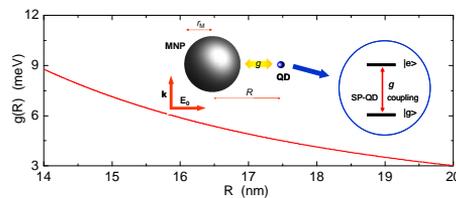


Figure 1. Interaction between a quantum dot and a silver nanoparticle: the applied electromagnetic field induces a polarization that causes dipole-dipole coupling. States $|g\rangle$ and $|e\rangle$ are coupled via the localized surface plasmon dipole mode with a strength g . Dependence of the coupling g on the metallic nanoparticle-quantum dot distance R

function of the frequency of the incidence light obtained for different QD-MNP distances R as indicated in the panel. The spectra in Fig. 2a have been calculated in the limit of very low excitation intensity, where the excitonic populations $\langle\sigma^\dagger\sigma\rangle \ll 1$. At $R = 14$ nm a Fano-like lineshape around the QD transition energy ω_x is evident. For a particular input frequency the scattered light is highly suppressed, while at slightly lower energy an enhancement of scattering due to constructive interference can be observed. For comparison the plot at $R = 14$ nm shows the scattering spectrum in the absence of the QD (dash-dotted line). Increasing the distance R , the Fano resonance narrows, due to the reduction of the MNP induced broadening of the QD linewidth. While at $R = 18$ nm the destructive interference remains almost complete, at larger distances ($R = 25$ nm), the Fano interference effect lowers and the suppression as well as the increase of the scattered light are reduced. Figure 2b displays scattering spectra obtained for QDs with different excitonic energy levels. In particular, each panel corresponds to different exciton-SP detunings $\Delta = \omega_x - \omega_{SP}$. Interestingly, the interference effect determining a strong suppression of scattering at specific wavelengths of the input field requires no special tuning unlike analogous effects in cavity QED. While SPs supported by the MNP can be described as harmonic oscillators, the single QD displays nonlinearities at single photon level. Panel 2b puts forward the

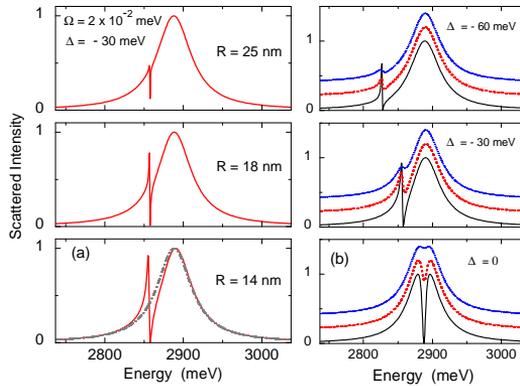


Figure 2. (a) Scattered light intensity spectra (red continuous line) calculated for different QD-MNP distances R at low density excitation power. For $R = 14$ nm, the scattered light without the presence of the QD (dot-dashed line) is also plotted. (b) Spectra calculated at $R = 14$ nm. Each panel shows calculations for a specific exciton-SP energy detuning (Δ) indicated in the figure. The black continuous line describes plots obtained for an input intensity field $\Omega = 0.02$ meV (corresponding to a photon flux $\Phi = 1.75 \mu\text{m}^{-2} \text{ps}^{-1}$), the red short-dashed line plots obtained at $\Omega = 0.4$ meV ($\Phi = 700 \mu\text{m}^{-2} \text{ps}^{-1}$), and the blue short-dotted line plots at $\Omega = 1$ meV. Plots are peak-normalized. The plot at $\Omega = 0.4$ meV was vertically shifted by 0.2; the one at $\Omega = 1$ meV by 0.4

dependence of light scattering on the intensity of the input field. The continuous lines describe low-field spectra obtained for a Rabi energy $\Omega = 2\mu E_0 = 2 \times 10^{-2}$ meV. Increasing the input field to $\Omega = 0.4$ meV, saturation effects appear (dashed line). At $\Omega = 1$ meV saturation is almost complete. The hybrid artificial molecule thus behaves as a frequency dependent saturable scatterer. This behavior has a profound impact on the statistics of the scattered photons [4].

3. Nanopolaritons

It is well known that the electromagnetic field in the gap region of a pair of strongly coupled nanoparticles can be drastically amplified, resulting in an extraordinary enhancement factor large enough for single-molecule detection by surface enhanced Raman scattering (SERS). We exploit this so-called hot spot phenomenon in order to demonstrate that the vacuum Rabi splitting with a single quantum emitter within a subwavelength nanosystem can be achieved. We employ a pair of silver spheres of radius $r_{Ag} = 7$ nm separated by a gap $l = 8$ nm embedded in a dielectric medium with permittivity $\epsilon_r = 3$. We consider a small spherical quantum dot with radius of the active region $r_{rmQD} = 2$ nm, whose lowest energy exciton is resonant with the dimer bonding mode. Fig. 3a display a sketch of the system and of the input field polarized along the trimer axis in order to provide the largest field enhancement at the dot position. Fig. 3b shows the extinction cross section spectra calculated for different dipole moments $\mu = er_0$, being e the electron charge. For $r_0 = 0.1$ nm a narrow hole in the spectrum occurs which could be confused with the appearance of a small vacuum Rabi splitting. Only for higher dipole moments $r_0 = 0.3$ a

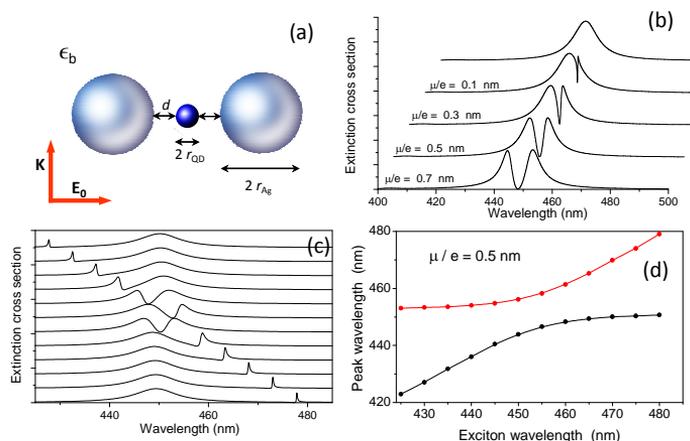


Figure 3. Vacuum Rabi splitting with a single quantum dot in the center of a dimer nanoantenna. (a) Sketch of the system and of the excitation. (b) Calculated extinction cross sections as function of the wavelength of the input field obtained for different dipole moments of the quantum dot. (c) Extinction cross sections spectra obtained for different resonant energies E_0 of the quantum dot exciton ($\mu/e = 0.5$ nm). A clear anti-crossing is observed owing to strong coupling between the dot and the localized bonding surface plasmon dimer-mode. (d) Dependence of the two Rabi-peaks (extinction cross sections) wavelengths on the exciton transition wavelength $\lambda_0 = hc/E_0$ ($\mu/e = 0.5$ nm).

significant splitting can be observed. Fig. 3c displays the extinction spectra for $r_0 = 0.5$ nm obtained changing the energy of the quantum dot exciton. At large detuning the peak arising from the quantum dot is significantly narrower from that originating from the SP bounding mode. Lowering the detuning increases the linewidth of the exciton-like peak while at the same time lowers that of the SP-like peak as a consequence of the strong coupling between the modes. Fig. 3d show the Dependence of the two Rabi-peaks (extinction cross sections) wavelengths on the exciton transition wavelength $\lambda_0 = hc/E_0$ ($\mu/e = 0.5$ nm). The anticrossing behaviour certifying true strong coupling is evident.

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