Nonlinear Optical Response of a Coupled Quantum Dot - Metal Nanoparticle System

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European Union
European Social Fund

OPERATIONAL PROGRAMME
EDUCATION AND LIFELONG LEARNING
investing in knowledge society
MINISTRY OF EDUCATION & RELIGIOUS AFFAIRS
MANAGING AUTHORITY

NSRF 2007-2013
programme for development
EUROPEAN SOCIAL FUND

Co-financed by Greece and the European Union

Plasmonics

- It has been known for several years that collective charge oscillations on the surface of metallic structures (usually termed surface plasmons) localizes electromagnetic radiation in very small volumes [1].
- The manipulation of electromagnetic radiation by coherent coupling of photons to surface plasmons is the main subject of the area of science and technology that is now called *'plasmonics'* [1,2].
- Several applications of plasmonic structures have been proposed, such as for example, in telecommunications, energy conversion and storage, high-density data storage, biological microscopy, medicine, photodetectors and sensors [1,2].

1. J.A. Schuller et al., Nature Materials **9**, 193 (2010).
2. See special issue in Plasmonics, Chem. Rev. **111**, Issue 6, p.p. 3667-3994 (2011).

Coupled Quantum and Plasmonic Structures

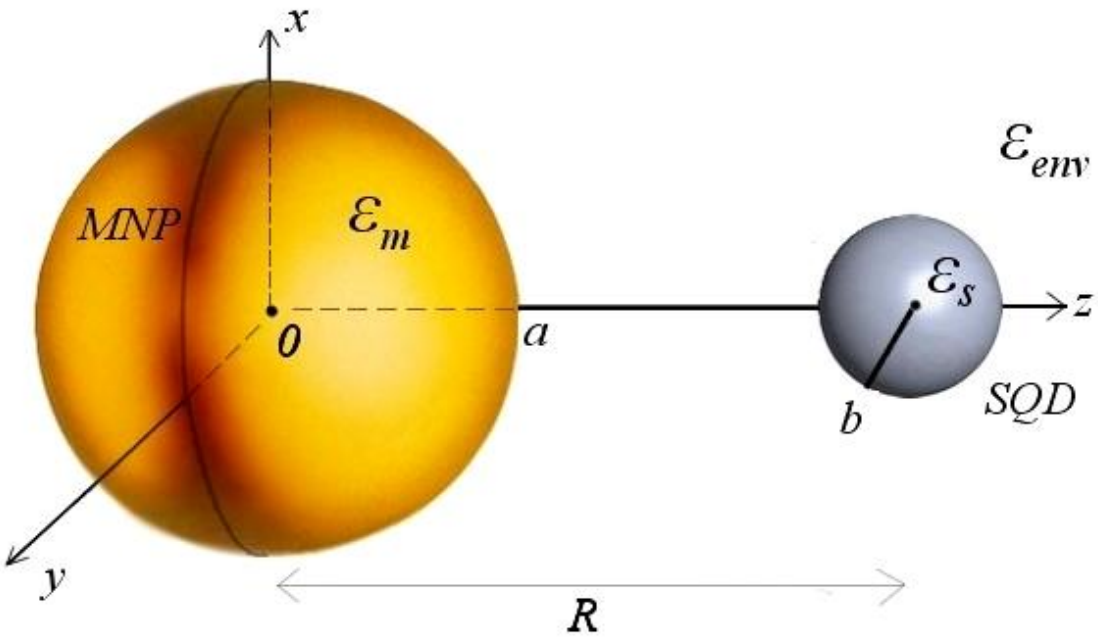
- A relatively new area of active research in photonics, termed quantum plasmonics [1], involves the study of the optical properties of complex photonic structures that involve both plasmonic nanostructures and quantum systems, such as atoms, molecules and semiconductor quantum dots.
- These complex (hybrid) structures are active photonic structures that are expected to have significantly enhanced optical response, in comparison to their constituents, if the properties of the two parts (plasmonic and quantum) are properly designed.
- The large fields and the strong light confinement associated with the plasmonic resonances enable strong interaction between the electromagnetic field and quantum systems near plasmonic nanostructures.
- In addition, using the quantum system one may achieve external control of the optical properties of the hybrid photonic structure.

1. M.S. Tame et al., Nature Physics **9**, 329 (2013).

Coupled Quantum Dot – Metal Nanoparticle System

- A system that has attracted particular attention is a hybrid nanocrystal complex composed of a semiconductor quantum dot coupled to a spherical metal nanoparticle via Coulomb interaction.
- Here, we study the nonlinear optical response arising from the interaction of this structure with a ***weak*** probe electromagnetic field of varying frequency and a ***strong*** pump electromagnetic field of fixed frequency.
- We calculate the effective third-order (Kerr-type) susceptibility and find that the spectrum of the nonlinear susceptibility depends strongly on the interparticle distance.
- We also present an explanation of the observed behavior in terms of an effective Rabi frequency.

Coupled Semiconductor Quantum Dot – Metal Nanoparticle Structure



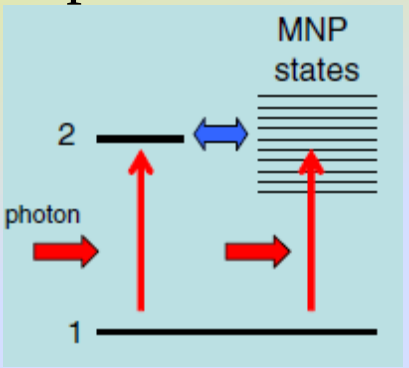
- b is the radius of the quantum dot.
- a is the radius of the metal nanoparticle.
- R $(>a)$ is the interparticle distance.

- ε_{env}, ε_s are the dielectric constants of the environment and the semiconductor quantum dot.
- $\varepsilon_m(\omega)$ is the dielectric function of the metal nanoparticle. Experimental values are used for the case of gold.

Four-Wave Mixing Effect – Basic Equations

• The system interacts with a linearly polarized strong pump field with fixed angular frequency ω_1 and a linearly polarized weak probe field with angular frequency ω_2.

• μ is the electric dipole matrix element.



$$E(t) = E_1 \cos(\omega_1 t) + E_2 \cos(\omega_2 t).$$

The electric field inside the quantum dot and is written as

$$E_{SQD} = \frac{\hbar}{\mu} \sum_{n=1,2} \left[\left(\frac{\Omega_n}{2} + G_n \sigma(t) \right) e^{-i\omega_n t} + \left(\frac{\Omega_n^*}{2} + G_n^* \sigma^*(t) \right) e^{i\omega_n t} \right]$$

$\sigma(t)$ is the slowly varying density matrix element.

$$G_n = \frac{1}{\pi \varepsilon_{env}} \frac{\gamma_n \mu^2 a^3}{\hbar \varepsilon_{effS}^2 R^6}, \quad \gamma_n(\omega_n) = \frac{\varepsilon_m(\omega_n) - \varepsilon_{env}}{\varepsilon_m(\omega_n) + 2\varepsilon_{env}},$$

$$\Omega_n = \frac{E_n \mu}{\hbar \varepsilon_{effS}} \left(1 + \frac{2\gamma_1 a^3}{R^3} \right)$$

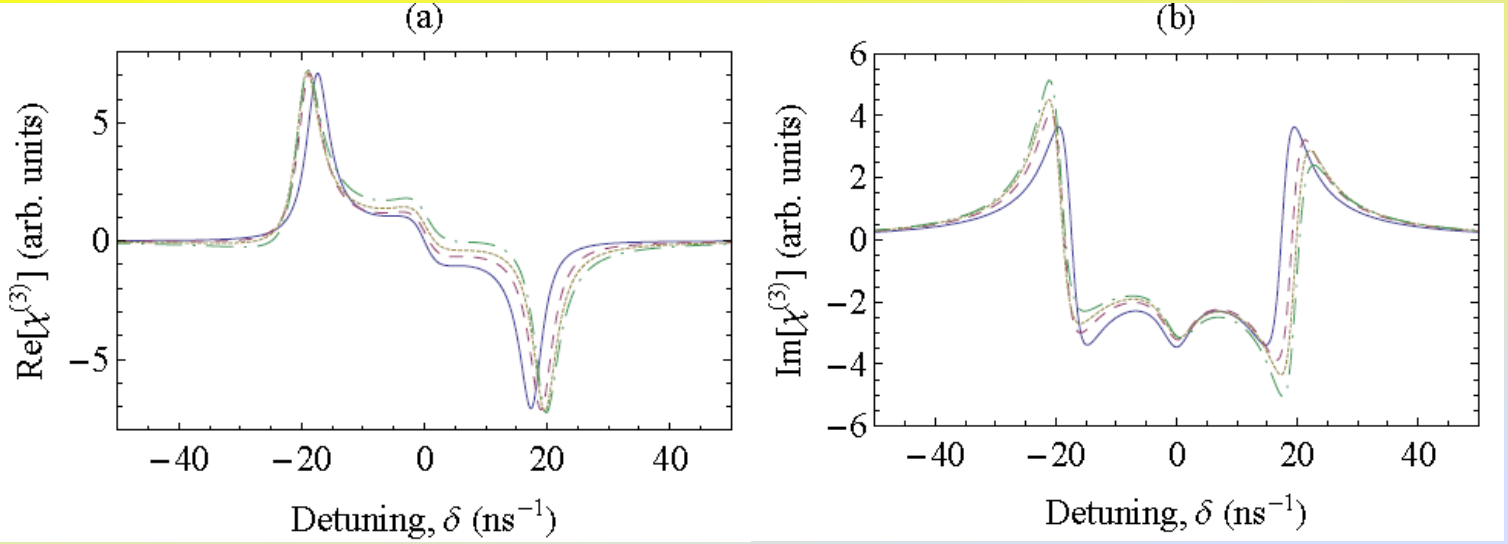
$$\varepsilon_{effS} = \frac{2\varepsilon_{env} + \varepsilon_S}{3\varepsilon_{env}}$$

• The parameter G_n describes the dipole-dipole interaction between excitons and plasmons.

Calculation Methodology for the Effective Third-order (Kerr-type) Susceptibility Spectrum

- We use density matrix equations for the description of the dynamics of the system.
- These equations, in addition to terms that describe the interaction of the system with the applied electromagnetic fields, contain also terms that describe the interaction between excitons and surface plasmons.
- We then write proper differential equations of the density matrix elements for the several nonlinear optical processes.
- In these equations the probe field is treated to ***first order*** and the pump field is treated to ***all orders***.
- The derived density matrix equations are nonlinear differential equations, so we adopt a numerical integration technique (4^{th} order Runge-Kutta method) for their solution and calculate the effective third-order (Kerr-type) susceptibility spectrum under steady state.

Results for Longer Distances: Exact Resonance



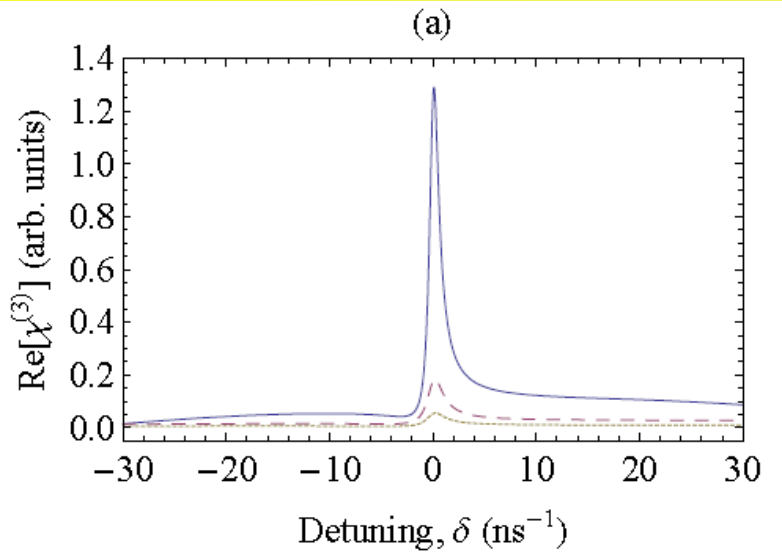
The parameters are a = 7.5 nm, $\varepsilon_{env} = \varepsilon_0$, $\varepsilon_S = 6\varepsilon_0$, $\hbar\omega_0$ = 2.5 eV, μ = 0.65 e nm, T_1 = 0.8 ns, T_2 = 0.3 ns.

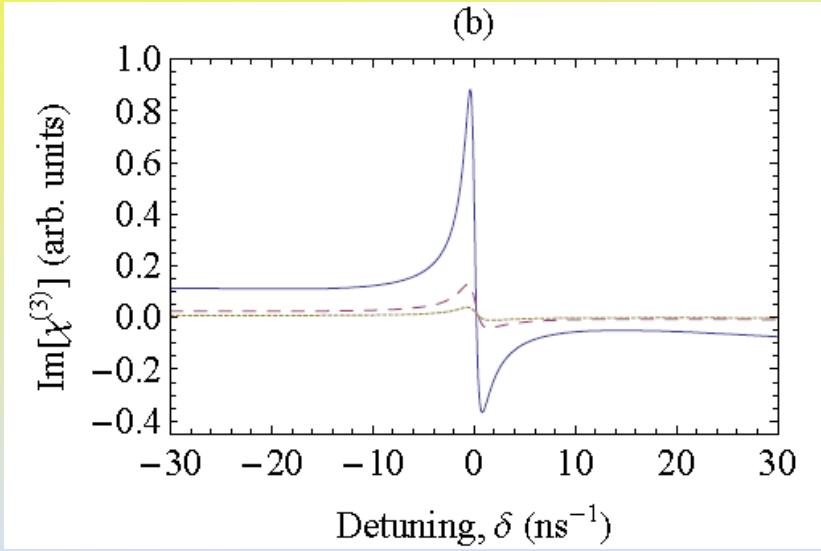
• The real part of $\chi^{(3)}$ in (a) and the imaginary part of $\chi^{(3)}$ in (b) in the coupled system as a function of the detuning $\delta = \omega_2 - \omega_1$ between the two fields. The intensity of the pump field is *3 10^2 W/cm^2*. Solid curve *R = 100 nm*, dashed curve *R = 20 nm*, dotted curve *R = 17 nm* and dot-dashed curve *R = 15 nm*. Typical values for CdSe-based quantum dot and gold metal nanoparticle (with radius 7.5 nm) are used. The pump field excitation is at exact resonance $\omega_1 = \omega_0$ with ω_0 being the single-exciton energy.

• The results are in agreement with that of previous studies [1,2].

1. J.-B. Li et al., Opt. Expr. **20**, 193 (2010).
2. J.-J. Li and K.-D. Zhu, Cr. Rev. Sol. State Mater. Sci. **39**, 25 (2014).

Results for Shorter Distances: Exact Resonance

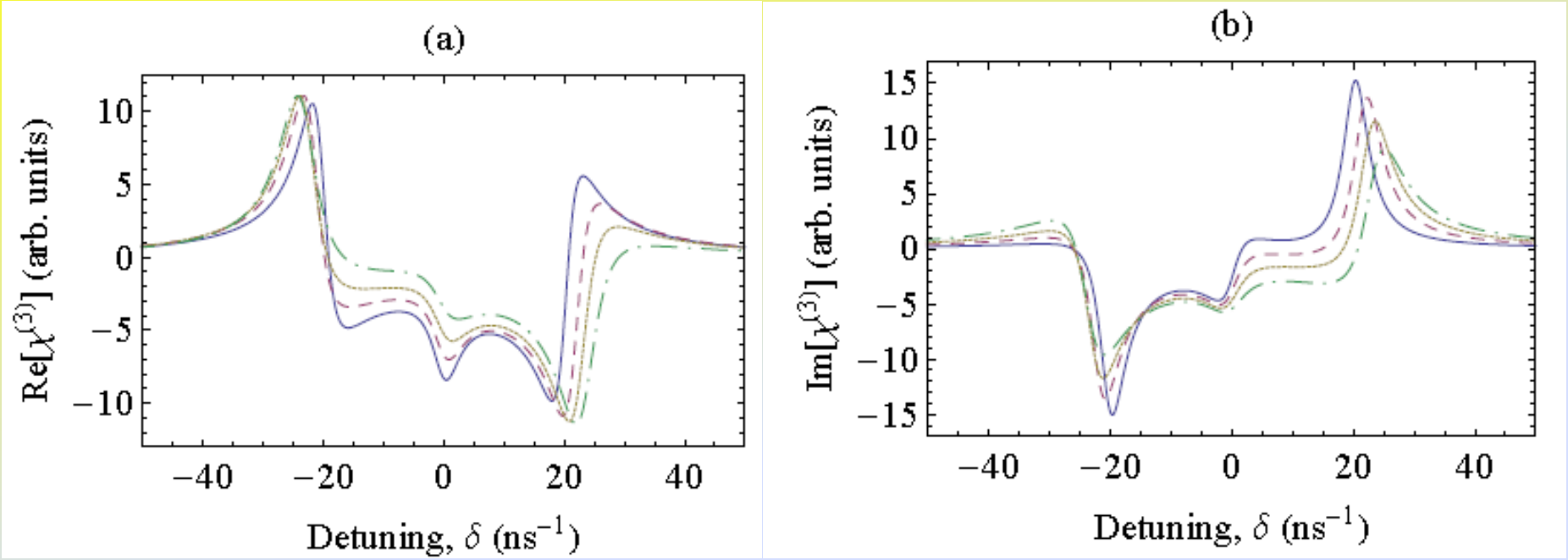




• The real part of $\chi^{(3)}$ in (a) and the imaginary part of $\chi^{(3)}$ in (b) in the coupled system as a function of the detuning $\delta = \omega_2 - \omega_1$ between the two fields. The intensity of the pump field is $3\ 10^2\ W/cm^2$. Solid curve $R = 14\ nm$, dashed curve $R = 13.5\ nm$ and dotted curve $R = 13\ nm$. The pump field excitation is at exact resonance $\omega_1 = \omega_0$ with ω_0 being the single-exciton energy.

• The effective $\chi^{(3)}$ nonlinearity spectrum changes form and is strongly suppressed. Also, as the interparticle distance decreases further the spectrum is further suppressed.

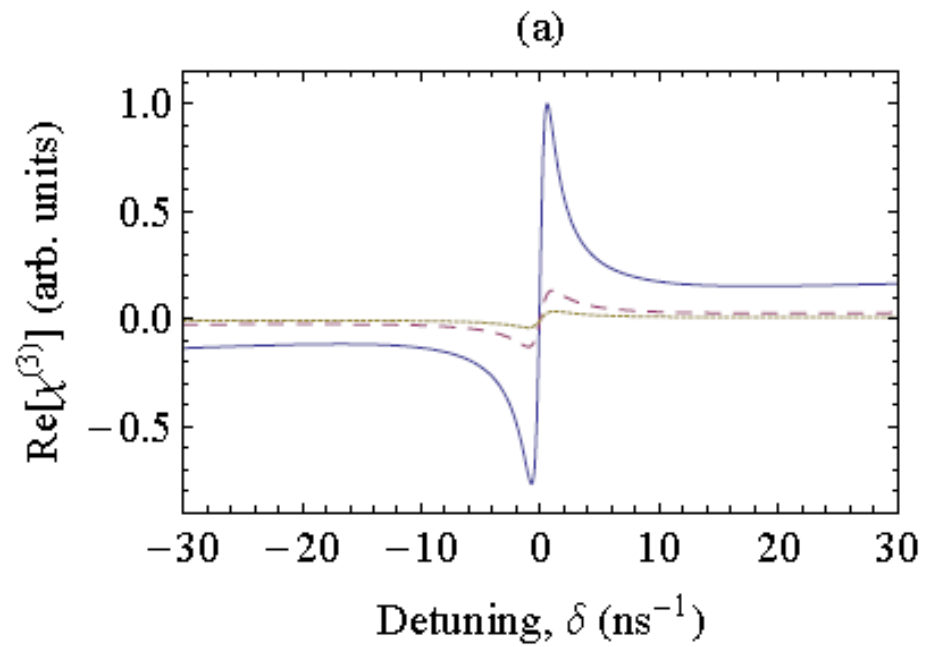
Results for Longer Distances: Detuned Excitation

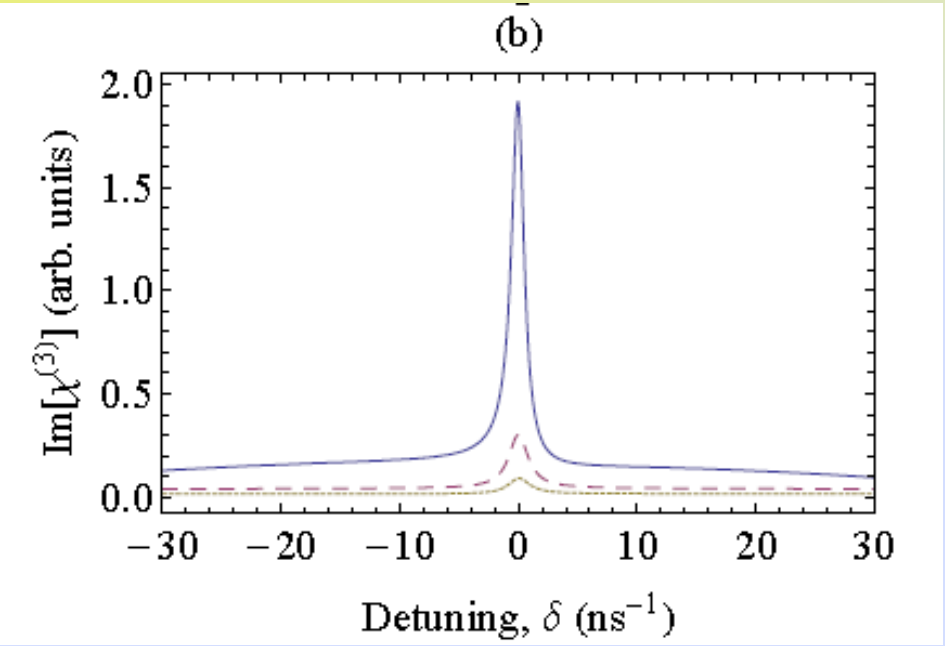


- The same as before but with the pump field excitation is detuned by $\omega_0 - \omega_1 =$ 10 ns^{-1}. Solid curve *R = 100 nm*, dashed curve *R = 20 nm*, dotted curve *R = 17 nm* and dot-dashed curve *R = 15 nm*.
- These results are also in agreement with that of previous studies [1,2].

1. J.-B. Li et al., Opt. Expr. **20**, 193 (2010).
2. J.-J. Li and K.-D. Zhu, Cr. Rev. Sol. State Mater. Sci. **39**, 25 (2014).

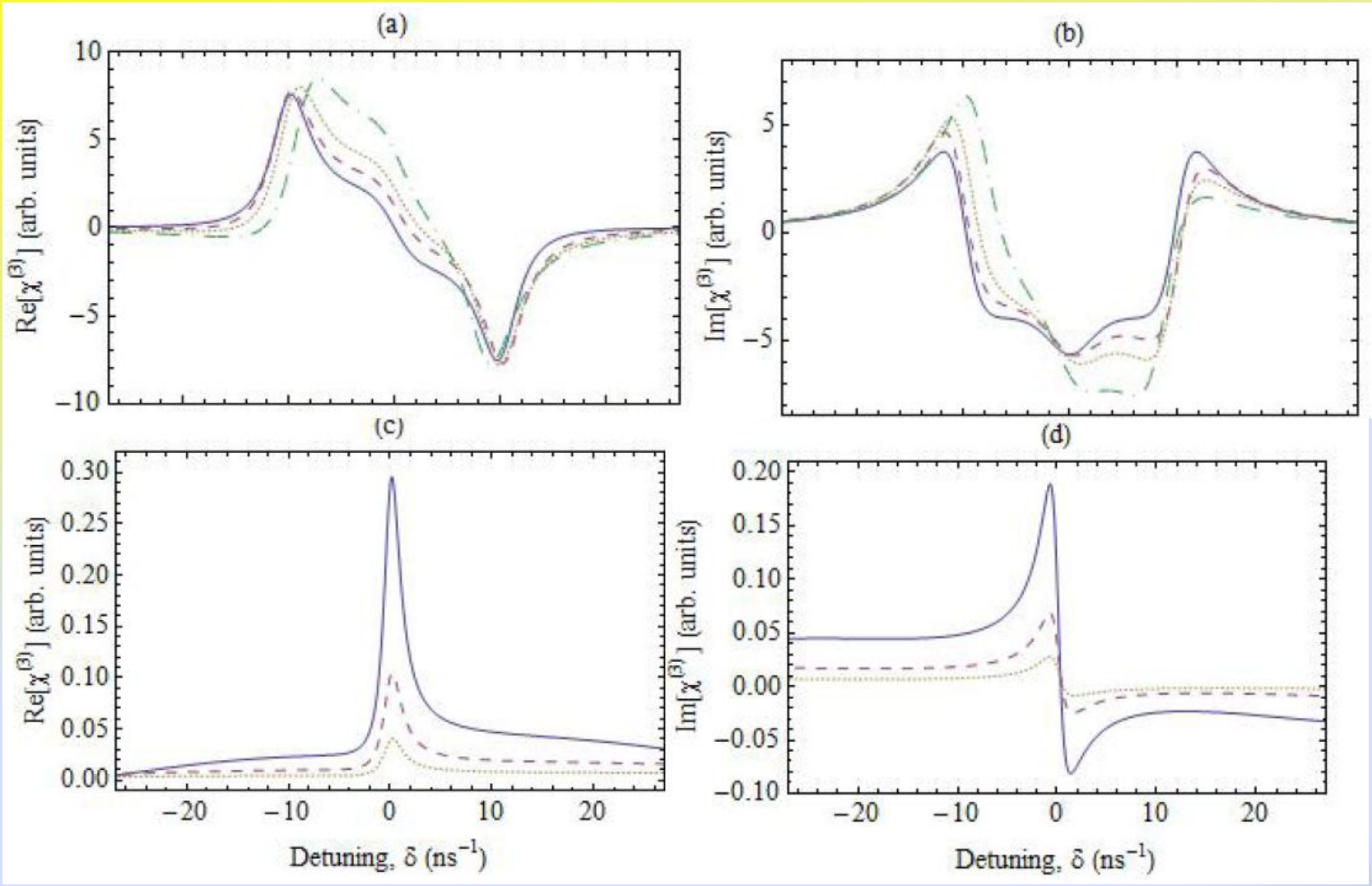
Results for Shorter Distances: Detuned Excitation





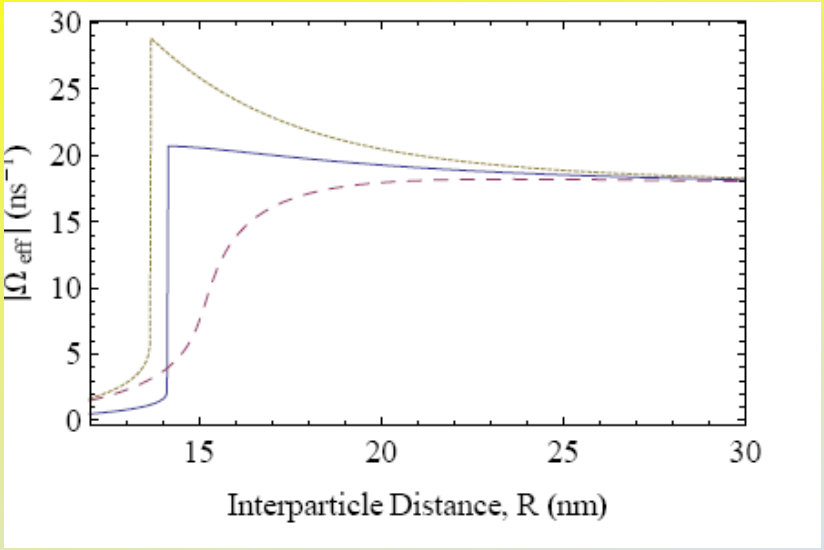
• The same as before but with the pump field excitation is detuned by $\omega_0 - \omega_1 = 10$ ns^{-1}. Solid curve $R = 13.5$ *nm*, dashed curve $R = 13$ *nm*, dotted curve $R = 12.5$ *nm*.

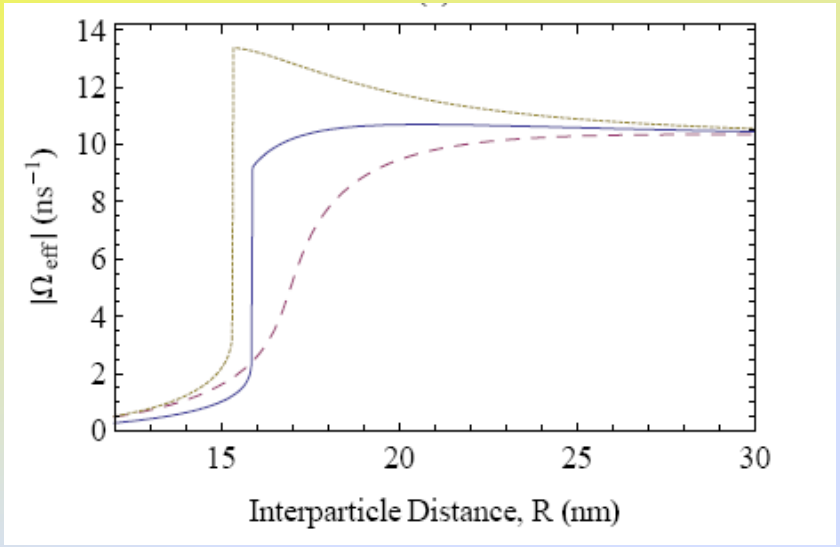
Results for Different Pump Field Intensity



• The real part of $\chi^{(3)}$ in (a), (c) and the imaginary part of $\chi^{(3)}$ in (b), (d). The intensity of the pump field is 10^2 W/cm^2. The pump field excitation is at exact resonance. (a) and (b) are for longer distances and (c) and (d) are for shorter distances.

Explanation via an Effective Rabi Frequency





$$\Omega_{eff} = \Omega_1 + 2G\sigma_{21}^{(\omega_1)^{ss}} \qquad \Omega_1 = \frac{\mu E_1}{\hbar \varepsilon_{effS}}\left(1+\frac{2\gamma_1 a^3}{R^3}\right)$$

• The absolute value of the effective Rabi frequency Ω_{eff} as a function of the interparticle distance R. Left figure: The intensity of the pump field is $I_1 = 3\ 10^2\ W/cm^2$ and the pump field detuning is $\Delta_1 = 0$ (solid curve), $\Delta_1 = -10\ ns^{-1}$ (dashed curve) and $\Delta_1 = 10\ ns^{-1}$ (dotted curve). Right figure: $I_1 = 10^2\ W/cm^2$ and $\Delta_1 = 0$ (solid curve), $\Delta_1 = -5\ ns^{-1}$ (dashed curve) and $\Delta_1 = 5\ ns^{-1}$ (dotted curve).

• $|\Omega_{eff}|$ is determined via a complex analytical formula that is determined by the analytical solution of the density matrix equations.

• The position of the Rabi sidebands is given approximately by $\delta = \pm\sqrt{\Delta_1^2 + |\Omega_{eff}|^2}$.

Summary

- We have studied the effective $\chi^{(3)}$ susceptibility in a coupled semiconductor quantum dot - spherical metal nanoparticle structure giving emphasis to the influence of the interparticle distance.
- We find that there is a critical distance that changes the form of the effective $\chi^{(3)}$ nonlinearity spectrum.
- Above this distance the $\chi^{(3)}$ spectrum has a regular form and relatively small changes of the magnitude and shape of the spectrum by changing the interparticle distance can be obtained.
- Below this critical distance the effective $\chi^{(3)}$ nonlinearity spectrum is strongly suppressed.
- The behavior of the system is explained by the effective Rabi frequency and the behavior of the effective Rabi frequency can be explained via an analytical solution of the density matrix equations.

Acknowledgements

- This research has been co-financed by the European Union (European Social Fund - ESF) and Greek national funds through the Operational Program "Education and Lifelong Learning" of the National Strategic Reference Framework (NSRF) - Research Funding Program: Archimedes III.

- We thank Dr. Sofia Evangelou for useful discussions and help.