Third Order Nonlinear Optical Effects in a Strongly Driven Semiconductor Quantum Dot Coupled to a Metallic Nanosphere

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SCOPE OF THIS WORK: A relatively new area of active research that combines quantum optics and nanophotonics involves the study of the quantum and nonlinear optical properties of complexes of metallic nanoparticles (MNPs) and semiconductor quantum dots (SQDs). One of the structures that have attracted particular attention is a hybrid nanocrystal complex composed of a SQD and a spherical MNP that interacts with a weak probe field of varying frequency and a strong pump field of fixed frequency [1-5]. In this system slow light [1], gain without inversion [2], third order nonlinearity [3,4], and four-wave mixing [5] have been theoretically studied. These studies have shown that the interparticle distance plays an important role in the nonlinear optical response of the nanocrystal complex. An important result was found by Sadeghi [2], who showed that when the interparticle distance reaches a critical value, plasmonic meta-resonances are created [6] and an abrupt formation of a significant amount of gain without inversion in the SQD occurs. In this work we also address theoretically the problem of the nonlinear optical response in a coupled SQD-spherical MNP structure that interacts with a weak probe field of varying frequency and a strong pump field of fixed frequency and give emphasis to the behaviour of the real and imaginary part of the third order nonlinearity of the SQD with the interparticle distance. We find that there is a critical interparticle distance that strongly modifies the third order nonlinearity. The derived spectra show strongly different behaviour above and below the critical distance. This critical distance is the one that creates plasmonic meta-resonances in the hybrid structure [6].

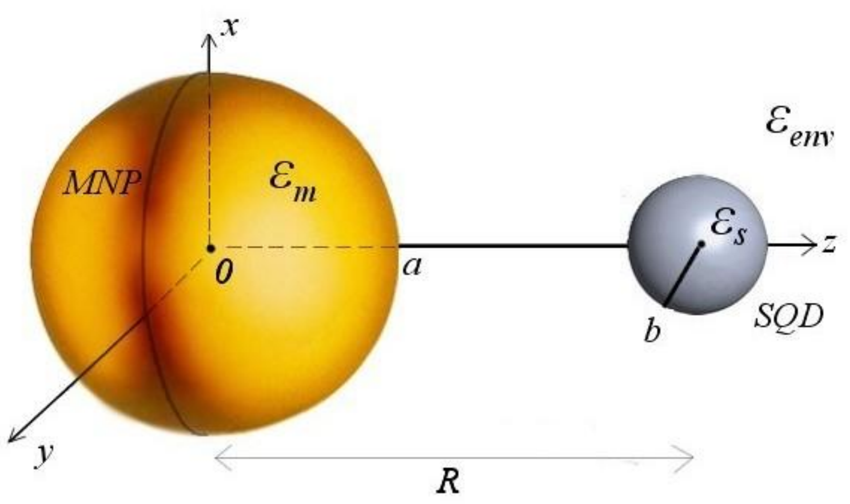

Figure 1: A coupled system consisting of a SQD and a spherical MNP. The system interacts with two electromagnetic fields, a linearly polarized strong pump field with angular frequency ω_1 and a linearly polarized weak probe field with angular frequency ω_2. Both fields are taken polarized parallel to the interparticle axis.

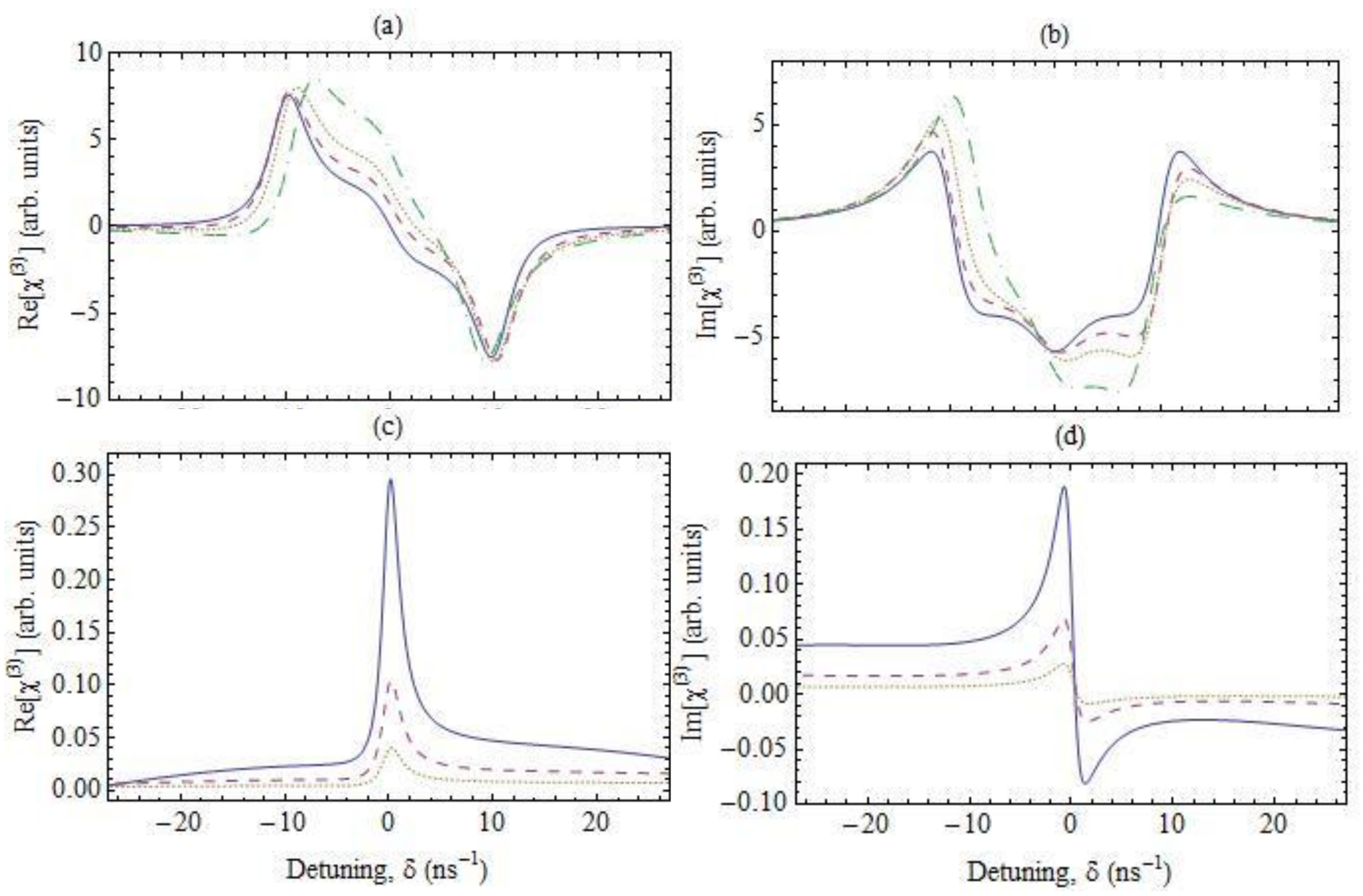

Figure 2: The real part of $\chi^{(3)}$ in (a) and (c) and the imaginary part of $\chi^{(3)}$ in (b) and (d) in the coupled SQD-MNP system as a function of the detuning $\delta = \omega_2 - \omega_1$. For the calculation we extend the density matrix methodology (see, e.g., refs. [4,7]) for the present system. The interaction between excitons and surface plasmons is taken into account in the calculations [1-6]. The pump field is strong and it is treated to all the orders, while the probe field is weak and it is treated to first order [4,7]. The intensity of the pump field is taken 10^2 W/cm^2. Typical values for CdSe-based SQD and gold MNP (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. In (a) and (b): solid curve R = 100 nm, dashed curve R = 20 nm, dotted curve R = 18 nm and dot-dashed curve R = 16.5 nm. In (c) and (d): solid curve R = 15 nm, dashed curve R = 14.5 nm and dotted curve R = 14 nm. The change (mainly enhancement) with distance in figures (a) and (b) and the strong suppression in figures (c) and (d) can be explained using the plasmonic meta-resonances concept [5,6].

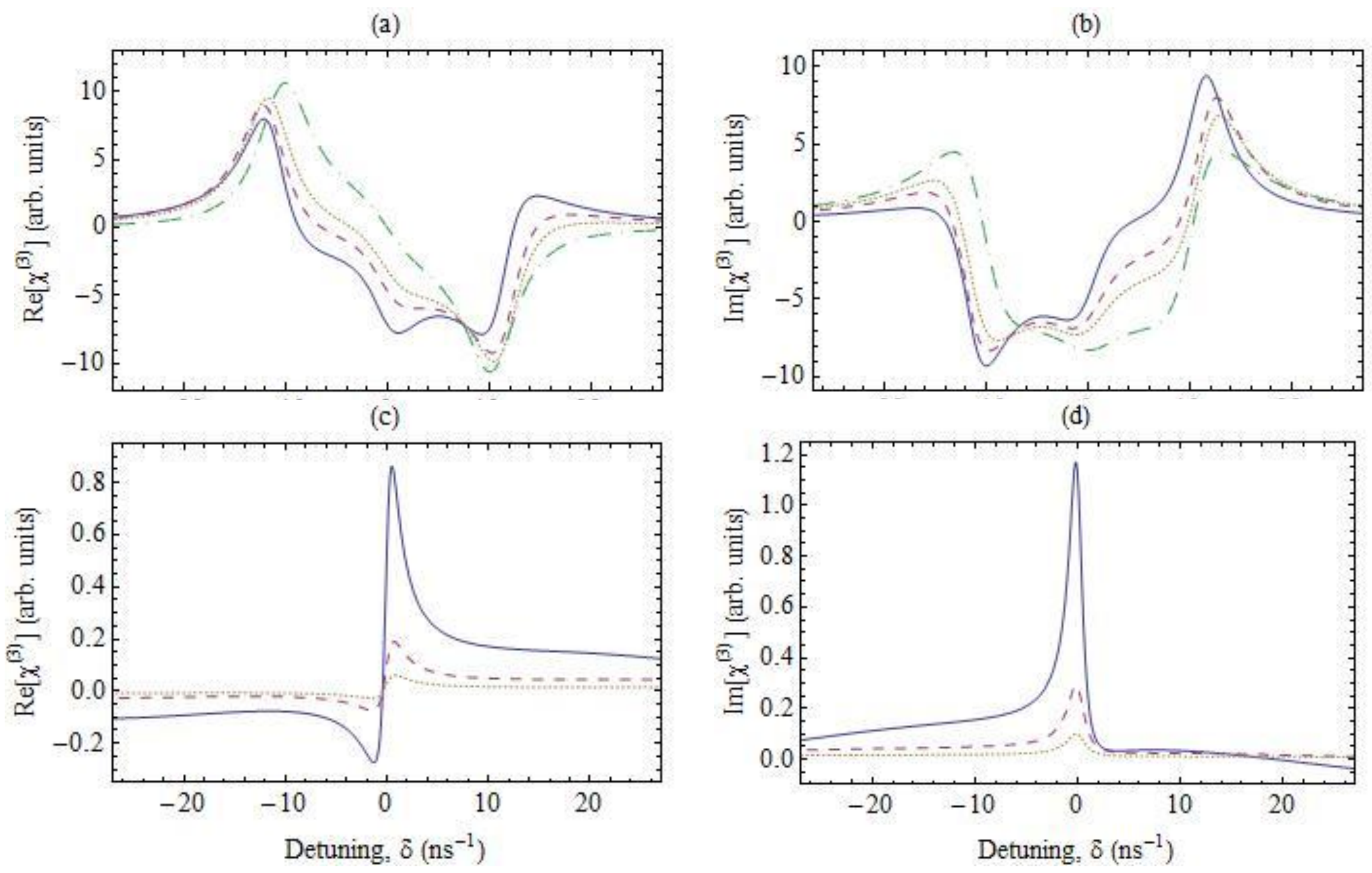

Figure 3: The same as in Fig. 2 but with the pump field detuned by $\omega_0 - \omega_1$ = 5 ns^{-1}. In (a) and (b): solid curve R = 100 nm, dashed curve R = 20 nm, dotted curve R = 18 nm and dot-dashed curve R = 16 nm. In (c) and (d): solid curve R = 15 nm, dashed curve R = 14.5 nm and dotted curve R = 14 nm.

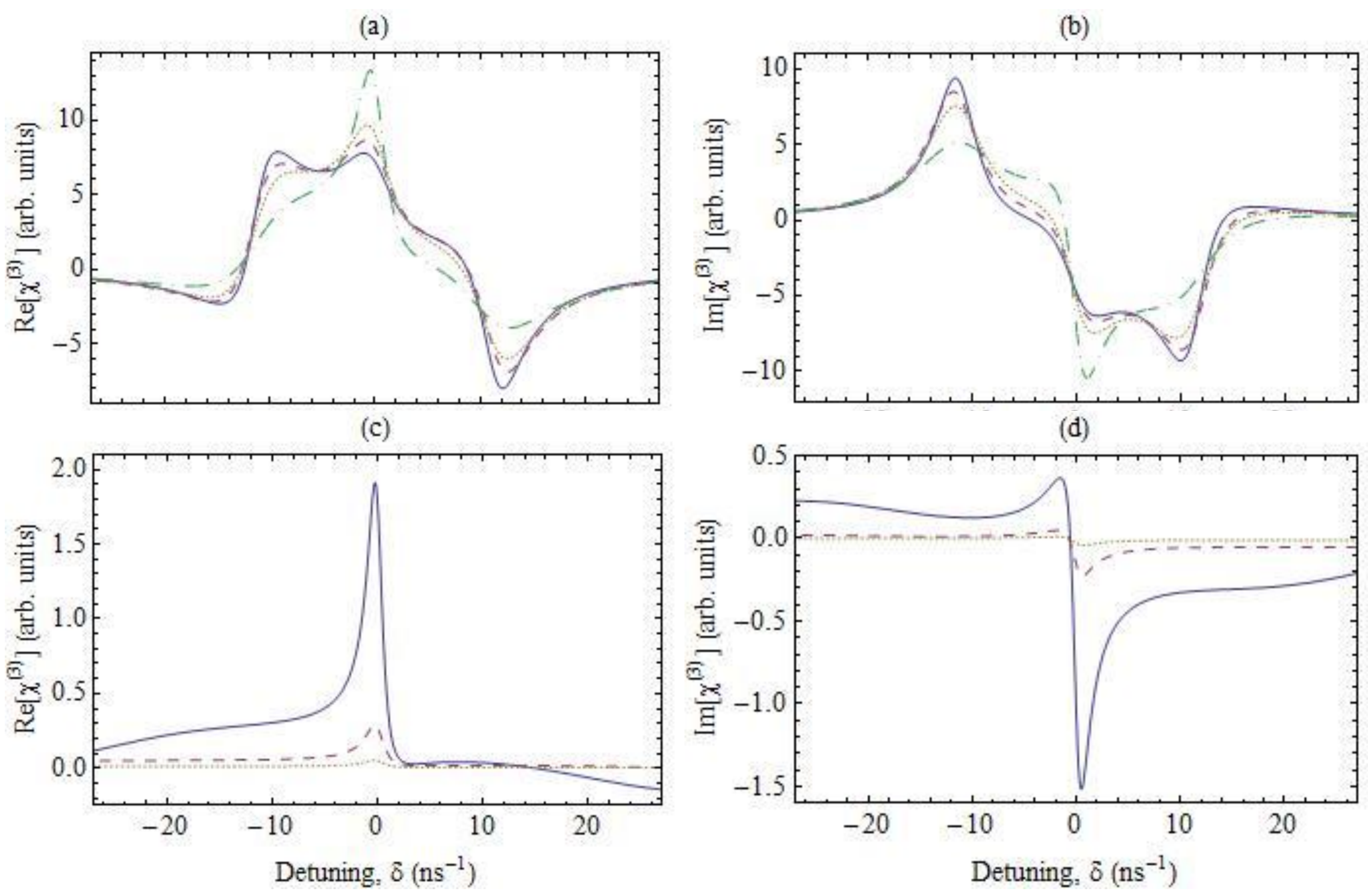

Figure 4: The same as in Fig. 2 but with the pump field detuned by $\omega_0 - \omega_1$ = -5 ns^{-1}. In (a) and (b): solid curve R = 100 nm, dashed curve R = 22 nm, dotted curve R = 20 nm and dot-dashed curve R = 18 nm. In (c) and (d): solid curve R = 16 nm, dashed curve R = 15 nm and dotted curve R = 14 nm.

REFERENCES

1. Z. Lu and K.-D. Zhu, J. Phys. B **41**, 185503 (2008).
2. S. M. Sadeghi, Nanotechnology **21**, 455401 (2010); Phys. Rev. A **88**, 013831 (2013).
3. Z. Lu and K.-D. Zhu, J. Phys. B **42**, 015502 (2009).
4. J.-B. Li, N.-C. Kim, M.-T. Cheng, L. Zhou, Z.-H. Hao, and Q.-Q. Wang, Opt. Express, **20**, 1856 (2012).
5. E. Paspalakis, S. Evangelou, S. G. Kosionis, and A. F. Terzis, submitted, (2013).
6. S. M. Sadeghi, Phys. Rev. B **79**, 233309 (2009).
7. S. G. Kosionis, A. F. Terzis, and E. Paspalakis, J. Lumin. **140**, 130 (2013).

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