

Modulation of Absorption & Gain in a Hybrid Semiconductor Quantum Dot – Metal Nano-Spheroid System: Impact of Structural Parameters

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INTRODUCTION & SCOPE OF THIS WORK: Modified nonlinear optical effects in exciton–plasmon hybrid systems have been extensively studied, demonstrating that their optical properties can be tuned by controlling key system parameters. Notably, the pump–probe response has already been investigated in a hybrid structure consisting of a two-level semiconductor quantum dot (SQD) and a spherical gold metal nanoparticle (MNP) under strong pump and weak probe fields [1]. The present study extends this work by examining absorption and gain while modifying the orientation and eccentricity of the MNP [2–4]. Using the Liouville equation under the rotating wave and dipole approximations, we perform an expansion of the density matrix elements with respect to the weak probe field and numerically solve the resulting equations in the steady state. We compute the optical susceptibilities of both the SQD and the MNP, focusing on how the absorption/gain spectral profile is influenced by the geometrical parameters of the nanostructure. A dressed-state framework supports our analysis of the pump-probe response.

METHOD

$$\text{Hamiltonian: } H = \sum_{i=0,1} E_i |i\rangle\langle i| - \hbar \left\{ \sum_{n=p,s} \left[\frac{\Omega_n}{2} e^{-i\omega_n t} + G_n \rho_{10}(t) \right] + c.c. \right\} (|0\rangle\langle 1| + |1\rangle\langle 0|)$$

$$\Omega_n = \frac{\mu E_n}{\hbar \epsilon_{effS}} \left(1 + \frac{s_{pol} \gamma a b^2}{R^3} \right) : \text{Rabi frequency}$$

$$G_n = \frac{\gamma_n}{4\pi \epsilon_0 \epsilon_{env}} \frac{s_{pol}^2 \mu^2 a b^2}{\hbar \epsilon_{effS}^2 R^6} : \text{self-interaction factor}$$

$$\gamma_n = [\epsilon_m(\omega_n) - \epsilon_{env}] / \{ 3\xi [\epsilon_m(\omega_n) - \epsilon_{env}] + 3\epsilon_{env} \}$$

$$\epsilon_{effS} = (2\epsilon_{env} + \epsilon_S) / (3\epsilon_{env})$$

$$\mu: \text{dipole moment}$$

$$\text{Liouville-Von Neumann equation: } i\hbar \dot{\rho}(t) = [H(t), \rho(t)]$$

$$\text{RWA: } \sigma = \rho_{10} e^{i\omega_p t}$$

Depolarization factor:

$$\xi = \frac{1}{1 - (a/b)^2} \left\{ 1 - (a/b) \frac{\arcsin[1 - (a/b)^2]}{\sqrt{1 - (a/b)^2}} \right\}$$

Non-linear density matrix equations

$$\dot{w}(t) = i(\Omega_p + \Omega_s e^{-i\delta t}) \sigma^*(t) - i(\Omega_p^* + \Omega_s^* e^{i\delta t}) \sigma(t) + 4i \text{Re}(G) \sigma(t) \sigma^*(t) - [w(t) + 1]/T_1 \quad (1)$$

$$\dot{\sigma}(t) = -\frac{i}{2} (\Omega_p + \Omega_s e^{-i\delta t}) w(t) - iGw(t) \sigma(t) - [i\Delta(t) + 1/T_2] \sigma(t), \quad G = G_p + G_s \quad (2)$$

$$\sigma(t) = \sigma^{(0)}(t) + \sigma^{(\delta)}(t) e^{-i\delta t} + \sigma^{(-\delta)}(t) e^{i\delta t}$$

$$w(t) = w^{(0)}(t) + w^{(\delta)}(t) e^{-i\delta t} + w^{(-\delta)}(t) e^{i\delta t}$$

First-order expansion of density matrix elements

Derivation of a set of 6 D.E.s solved in steady state

$$\text{Optical susceptibilities: } \chi_{SQD}^{(1)} = \frac{2\mu(\Gamma/V)}{\epsilon_0 E_s} \sigma^{(\delta)SS}, \quad \chi_{MNP}^{(1)} = 3\gamma_s \left[\frac{\epsilon_{env}}{\epsilon_0} + \frac{s_{pol}}{4\pi\epsilon_{effS}(\Gamma/V)R^3} \chi_{SQD}^{(1)} \right]$$

Γ/V : SQD optical confinement factor over volume

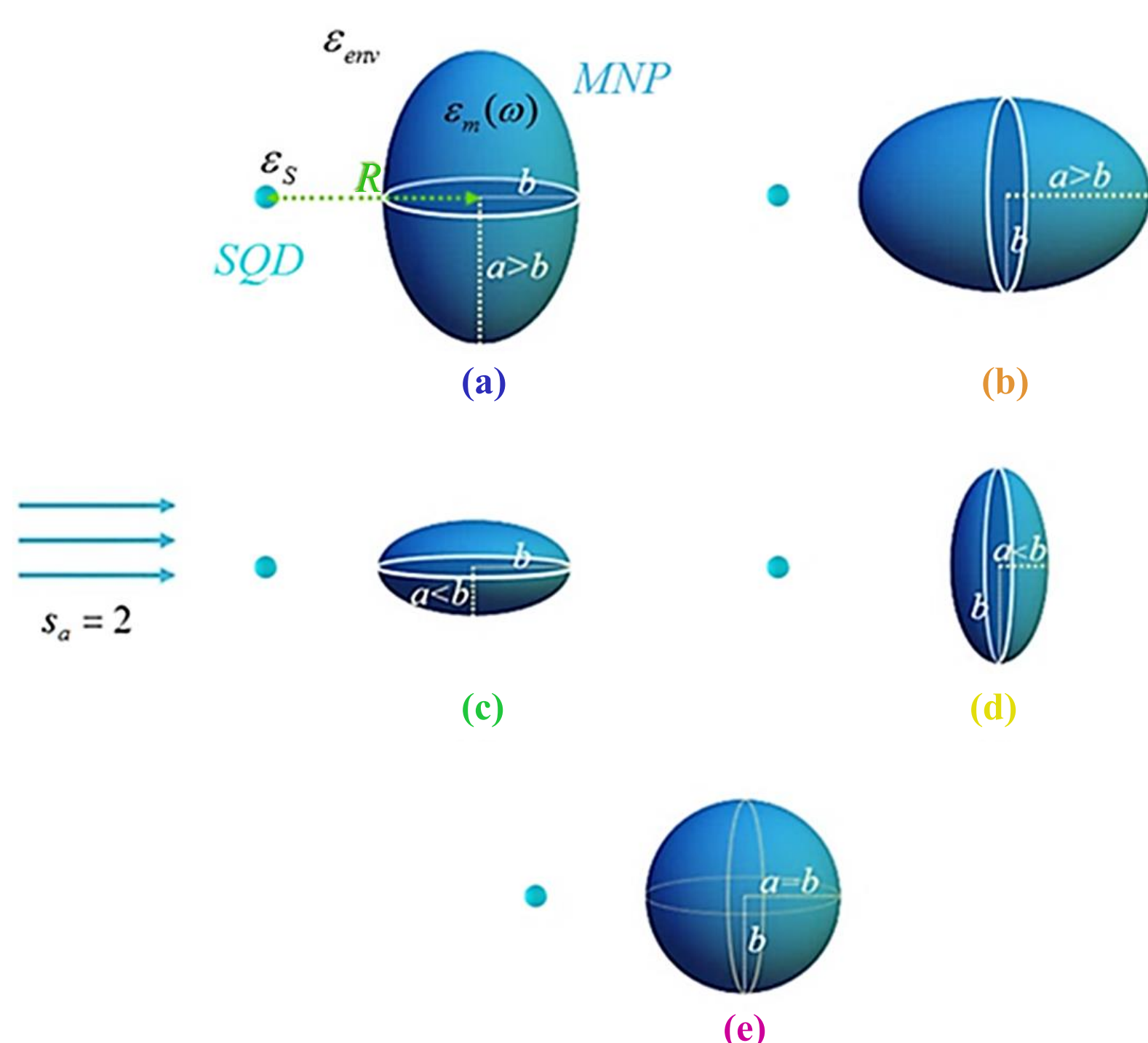


FIG. 1. Hybrid nanostructure composed of a SQD and a spheroidal MNP with fixed equatorial semiaxis length and variable polar semiaxis, while the polar semiaxis changes to form prolate (a), oblate (c, d), with $a/b=1.5$ & spherical (e) geometries. ϵ : relative dielectric constants.

RESULTS & DISCUSSION

- At $R=14 \text{ nm}$ (Fig. 3), confs. (c) and (d) lie in the Bright metastate; others fall in the Dark region.
- Fig. 4 shows a resonance triplet for (c) and (d), while other confs. exhibit single resonances.
- Gain is observed only in the Bright metastate, with conf. (c) showing the strongest amplification.
- The SQD exhibits a higher absorption peak in the Dark metastate than in the Bright.
- Conf. (d) shows reduced MNP absorption compared to the reference black curve (non-interacting components).
- In the Dark region, both components show exclusively absorption over a broader spectral range.
- Conf. (b) displays a pronounced leftward shift in its resonance peak.

CONCLUSION

- The optical absorption and gain of the SQD–MNP hybrid system are governed by precise tuning of the MNP’s eccentricity and orientation.
- These findings offer valuable guidance for the design of advanced nanophotonic and quantum devices, including nanosensors, nanoswitches, and energy transfer systems.

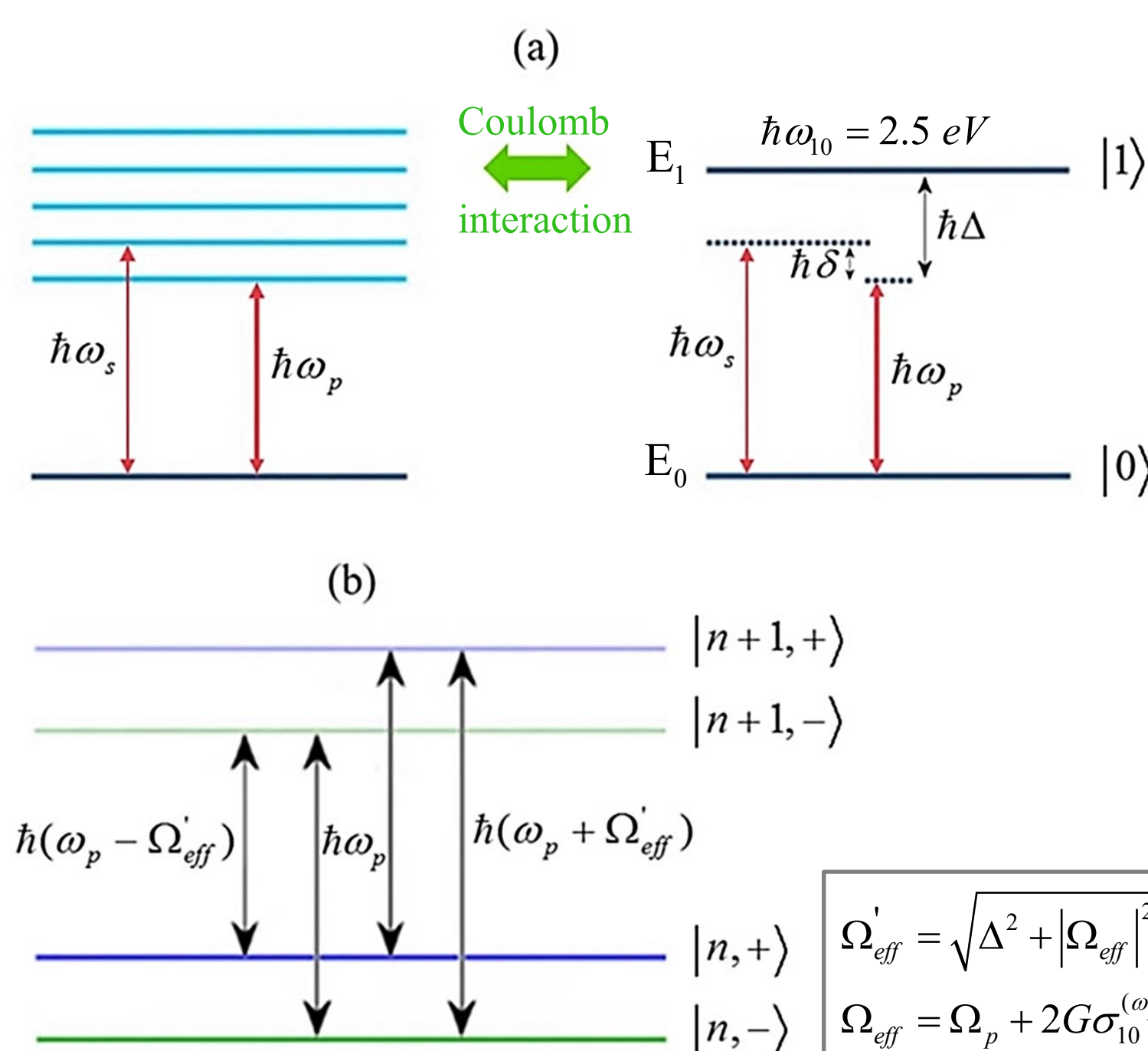


FIG. 2. (a) Energy-level diagram illustrating exciton and surface plasmon transitions in the hybrid nanosystem, under pump (ω_p) and probe (ω_s) fields. The MNP is modeled as a continuum of states. (b) Dressed state picture representation.

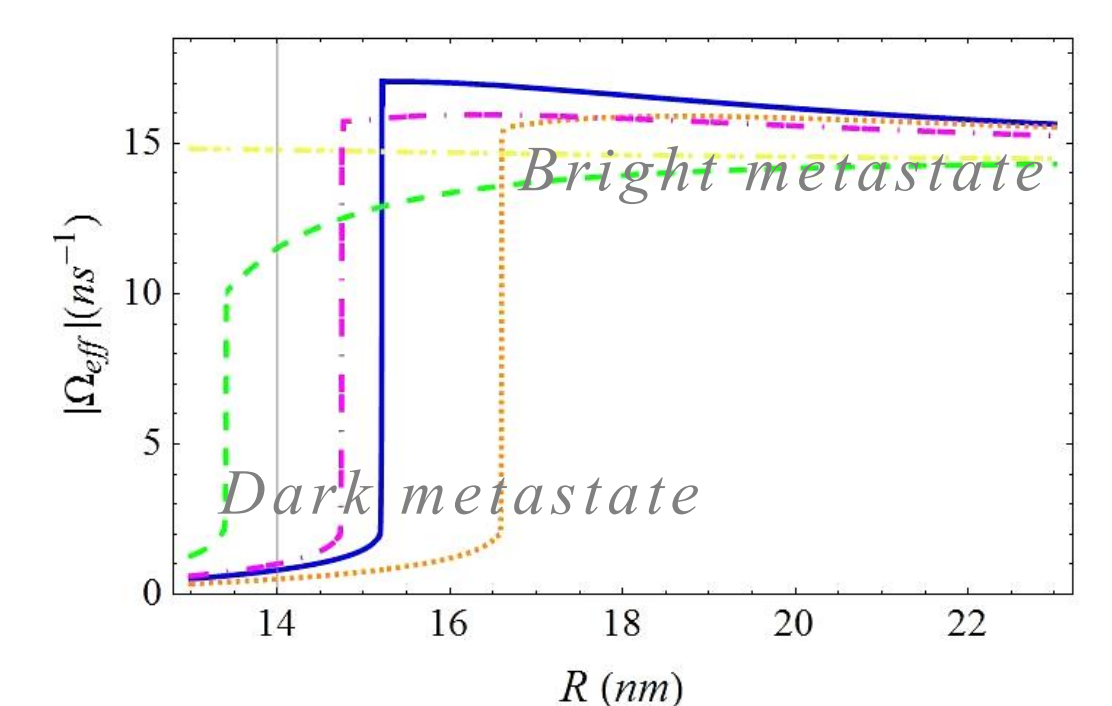


FIG. 3. Absolute value of the effective Rabi frequency, Ω_{eff} , as a function of the interparticle distance, R , for the configurations illustrated in FIG. 1. $b=7.5 \text{ nm}$, $\epsilon_s=6$, $\epsilon_{env}=1$, $\epsilon_m=-2.3+3.8i$, $T_1=0.8 \text{ ns}$, $T_2=0.3 \text{ ns}$, $\Delta=0$, $\mu=0.65 \cdot e \text{ nm}$.

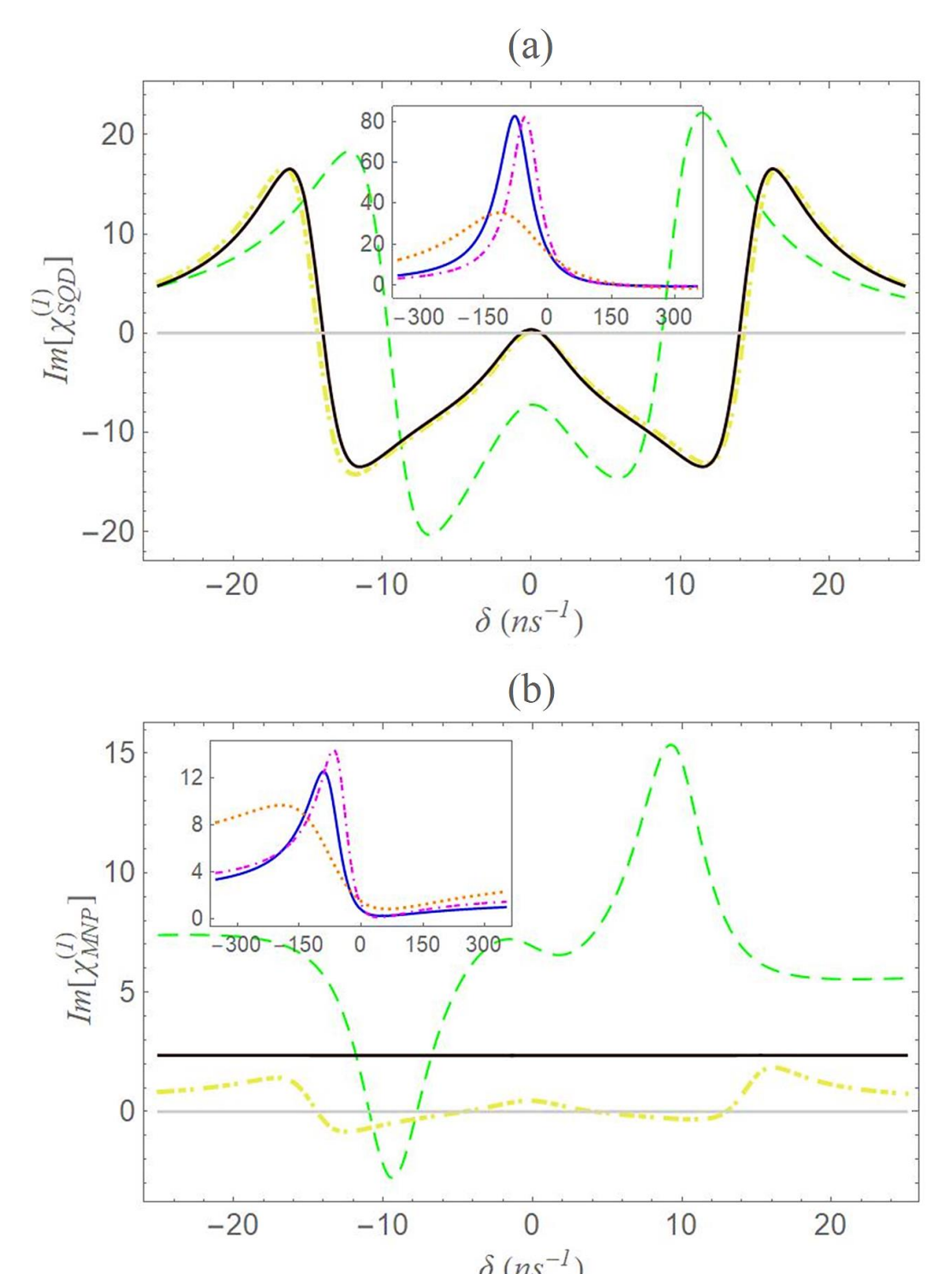


FIG. 4. Absorption spectra for the SQD & the MNP, at $R=14 \text{ nm}$, for the confs. illustrated in FIG. 1.

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