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Radiation emission from an asymmetric quantum dot coupled to a plasmonic nanostructure

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Abstract

We propose a scheme for controlling the absorption and RFS of a quantum dot (QD) with broken inversion symmetry interacting with a plasmonic nanostructure. The QD is described as a two-level atom-like system with a permanent dipole moment in the excited state. A linearly polarized laser field drives the optical transition of the QD and produces localized surface plasmons in the MNP. The influence of plasmonic effects between the nanoparticle and the dot is analyzed using the Green tensor method. We found terahertz (THz) emission in the resonance fluorescence spectrum when the laser frequency lies above the transition frequency. The position and strength of the THz peaks is controlled by means of the dot-nanoparticle separation. The quantum nature of the emitted THz photons is analyzed by evaluating the second-order fluorescence intensity correlation function. We found a periodic alternation of photon bunching and anti-bunching with a period that depends on the polarization of the driving field.

Keywords: quantum dot, plasmonic, terahertz radiation, permanent dipole moment

On the other hand, in QD–MNP hybrid systems, when the exciton energy of the QD is on resonance with the plasmon peak of the MNP, strong exciton–plasmon coupling can be observed on the power absorption [11, 12]. It is well-known that nanometer-scale metallic structures can dramatically modify the optical properties of various optically active objects of similar dimensions such as atoms, molecules, or QD’s. The underlying physical mechanism relies on the fact that a MNP, whose size is smaller than the wavelength of light in vacuum, exhibits strong dipolar excitations in the form of localized surface plasmon resonances. These are non-propagating excitations of the conduction electrons coupled to the light field [11, 13]. These localized surface plasmons are tightly spatially confined, leading to a resonant modification of the local field felt by the QD [14–17]. In addition, the local density of states are dramatically altered by the MNP, which results in a modification of the spontaneous emission rates of the QD’s optical transitions [17–21]. Strong local fields are particularly important for nonlinear optical processes, such as...
surface-enhanced Raman scattering and second harmonic generation [22, 23]. The possibility of reaching the quantum regime using plasmonic systems has also been addressed [18–20]. The effect of a plasmonic nanoparticle on the resonance fluorescence spectrum (RFS) of a two-level atom was first analyzed in [24, 25]. There, the modification of the atom radiation field caused by the presence of the nanoparticle and the change in the local field in the vicinity of the nanoparticle caused by the particle’s remission (i.e., the re-normalization of the Rabi frequency) were taken into account.

All these experiments have been analyzed in the framework of symmetric QD’s with vanishing mutual dipole moments (PDM’s). In any case, the pump field should be considered to be aligned both with the transition and PDM of the QD. The laser field can transfer an electron in the QD from the valence band into the conduction band, thus the ground state |1⟩ corresponds to the absence of free carriers while the excited state |2⟩ is associated to the creation of a direct exciton. The QD is placed in close proximity to a spherical MNP. We consider a single QD in close proximity to a MNP. For instance, PDM’s have been experimentally observed in exciton states of several kinds of QD’s [26–29], in excited quantum rings [30], in the exciton and bi-exciton states [31], and in InGaN/GaN QD’s excitonic complexes [32].

It has been shown that the response of a quantum system (atom/molecule) to an external electromagnetic field can be significantly modified if the system possesses PDM’s [33–37]. When the inversion symmetry of these artificial atoms is broken, the selection rules do not apply [38], and microwave-induced transitions between any two energy levels in multi-level superconducting quantum circuits are possible. Thus, multi-photon and single-photon processes can coexist in such artificial multilevel systems [38, 39]. PDM’s considerably influence the saturation of absorption on intersubband transitions. More interestingly, PDM’s enable two-level second-harmonic generation [40] and allow correlated photon pairs generation at variable frequencies [41]. PDM’s also enhance multi-photon resonant excitation of a quantum system subjected to a strong laser field [42–44].

In the current paper, we consider a single QD in close proximity to a MNP. For symmetric QD’s such system can be described by the well-known Jaynes–Cummings Hamiltonian [45]. This model predicts the transformation of the emission spectrum from the Rabi doublet to the Mollow triplet as pump intensity increases [46–49]. The incorporation of the asymmetry into the quantum system changes radically its emission pattern and leads to new optical transitions which were forbidden in the symmetric case. In particular, optical transitions at the Rabi frequency in QD’s placed in a strong external laser field have been found [50]. A similar effect occurs for asymmetric quantum wells placed inside a planar microcavity [51, 52]. In view of this, the aim of this paper is to develop a general theoretical approach to study the interaction of an hybrid system formed by an asymmetric QD and a MNP. We emphasize on the presence of large PDM’s of the electronic states of the QD. The influence of plasmonic effects between the MNP and the QD is analyzed using the Green tensor method. We study the terahertz (THz) emission from the QD–MNP hybrid system driven by a monochromatic classical field. We present numerical simulations for a representative case of QD, taking values of the oscillator strengths, decay rates and the characteristic energy levels from experimental studies. Both the strength and the frequency of the THz radiation may be conveniently modified and controlled by changing the separation distance between the MNP and the QD.

The paper is organized as follows: section 2 establishes the model, i.e., the Hamiltonian of the system and the time-evolution equations of the atomic operators. We incorporate the effects of the field enhancement and the modification of the decay rates by making use of the Green tensor method. We also derive a second-order atom-plasmon Hamiltonian taking into account the counter-rotating terms introduced by the PDM’s of the QD. Section 3 deals with the numerical simulations of the spectral properties of the fluorescence photons. We analyze the RFS, the temporal properties of the second-order correlation functions of such fluorescence photons and the absorption spectrum of a weak probe field. Finally, section 4 summarizes the main conclusions.

2. Basic equations

We consider an asymmetric QD which can be modeled as a two-level quantum system with ground (excited) state |1⟩ (|2⟩) and energy ω1 (ω2). The transition frequency is ωT = ω2 − ω1 and the transition electric dipole moment is μ12. Due to the breaking of inversion symmetry in asymmetric QDs, the levels |1⟩ and |2⟩ exhibit unequal PDMs (μ11 ≠ μ22). This leads to new physical effects discussed hereafter. The optical transition |1⟩ → |2⟩ is driven by a polarized laser field of the form

$$\vec{E} = \frac{1}{2} (E_0 e^{-i\omega_1 t} + E_0^* e^{i\omega_1 t}) \hat{u},$$

where $\omega_1$ and $\omega_2$ being a unit vector which allows for the specification of the polarization of the laser field and the corresponding angular frequency. We will consider the laser field parallel (perpendicular) to the QD–MNP separation vector, i.e., $\hat{u} = \hat{z}$ ($\hat{u} = \hat{\chi}$). In any case, the pump field should be considered to be aligned both with the transition and PDM of the QD. The laser field can transfer an electron in the QD from the valence band into the conduction band, thus the ground state |1⟩ corresponds to the absence of free carriers while the excited state |2⟩ is associated to the creation of a direct exciton.

The QD is placed in close proximity to a spherical MNP with radius $a$ as depicted in figure 1(a). The MNP is characterized by the (dispersive and lossy) dielectric function $\varepsilon_m(\omega)$.

The Hamiltonian that governs the dynamics of the coupled QD–MNP hybrid system can be written as

$$H = H_A + H_F + H_{int} + H_{Ext}. \quad \text{(2)}$$

The free Hamiltonian $H_A$ of the two-level QD system reads as

$$H_A = \hbar \omega_1 \sigma_z. \quad \text{(3)}$$
Figure 1. (a) A QD with transition frequency $\omega_0$ is placed at a distance $R$ of a spherical MNP of radius $a$. (b) Energy level diagrams for the QD and the MNP. The laser field excites surface plasmons in the MNP and excitions in the QD. The two-level scheme illustrates the QD and the MNP. The laser field excites surface plasmons in the MNP and the wavy lines indicate radiative decay rates.

$H_F$ is the Hamiltonian of the medium-assisted electromagnetic field

$$H_F = \hbar \int d\vec{r} \int_0^{\infty} d\omega \omega \mathcal{J}^\dagger_{\lambda}(\vec{r}, \omega) \mathcal{J}^\dagger_{\lambda}(\vec{r}, \omega),$$

which is expressed in terms of a set of bosonic fields $\mathcal{J}^\dagger_{\lambda}(\vec{r}, \omega)$ and $\mathcal{J}^\dagger_{\lambda}(\vec{r}, \omega)$ which play the role of variable of the electromagnetic field and the medium, including a reservoir associated with the losses in the medium [53]. The field operators obey the usual commutation rules

$$[\mathcal{J}^\dagger_{J\lambda}(\vec{r}, \omega), \mathcal{J}^\dagger_{K\lambda'}(\vec{r}', \omega')] = \delta_{JK} \delta_{\lambda\lambda'} \delta(\omega - \omega') \delta(\vec{r} - \vec{r}'), \quad j, k = x, y, z,$$

$\lambda$ being an index for polarization.

The interaction Hamiltonian between the QD and the medium-assisted electromagnetic field reads as

$$H_{\text{int}} = -\hbar (\sigma^+ + \sigma^-) \int_0^{\infty} d\omega \left[ \frac{\mu_{12}}{\hbar} \cdot \vec{E} (\vec{r}_A, \omega) + \text{h.c.} \right],$$

where $\vec{r}_A$ is the molecular center-of-mass position. Here we have introduced the usual atomic operators $\sigma^+ = |2\rangle \langle 1|$, $\sigma^- = |1\rangle \langle 2|$ and $\sigma_z = |2\rangle \langle 2| - |1\rangle \langle 1|$. The electric field $\vec{E} (\vec{r}, \omega)$, excluding the external driving field, can be expressed in terms of the bosonic variables through [53]

$$\vec{E} (\vec{r}, \omega) = i \frac{\hbar}{\pi c^2} \int d\vec{r}' \sqrt{\epsilon(r', \omega)} \vec{G} (\vec{r}, \vec{r}', \omega),$$

In equation (7), $\vec{G} (\vec{r}, \vec{r}', \omega)$ is the dyadic Green’s tensor which is a solution of the Helmholtz equation

$$\nabla \times \nabla \times \frac{\omega^2}{c^2} \epsilon (\vec{r}, \omega) \vec{G} (\vec{r}, \vec{r}', \omega) = i \delta (\vec{r} - \vec{r}'),$$

together with the boundary condition $\vec{G} (\vec{r}, \vec{r}', \omega) \rightarrow 0$ for $|\vec{r} - \vec{r}'| \rightarrow \infty$, and $I$ being the unit dyadic. In addition, the Green’s tensor obeys the useful integral relation

$$\int d\vec{r} \frac{\omega^2}{c^2} \epsilon (\vec{r}, \omega) \vec{G} (\vec{r}, \vec{s}, \omega) \vec{G}^\ast (\vec{s}', \vec{s}, \omega) = \text{Im} \{ \vec{G} (\vec{r}, \vec{r}', \omega) \},$$

which follows directly from the Helmholtz equation [53]. Here, $\epsilon_m (\vec{r}, \omega) = \epsilon_r (\vec{r}, \omega) + i \epsilon_i (\vec{r}, \omega)$ stands for the complex permittivity of the MNP at frequency $\omega$.

Finally, $H_{\text{Ext}}$ is the part of the Hamiltonian which accounts for the external coherent interaction of the laser field with the QD–MNP system, namely

$$H_{\text{Ext}} = \hbar (\Omega_x \sigma^+_x e^{-i \omega t} + P_{\text{dust}} \sigma^+_x e^{-i \omega t} + \text{h.c.}),$$

with the effective Rabi fields $\Omega_x = \mu_{12} \cdot \vec{E}_{\text{pump,ext}}(\vec{r}_A)/2 \hbar$, and $P_{\text{dust}} = (\mu_{22} - \mu_{11}) \cdot \vec{E}_{\text{pump,ext}}(\vec{r}_A)/2 \hbar$. The pump field contains the direct pumping field term plus the scattered field from the MNP

$$\vec{E}_{\text{pump,ext}}(\vec{r}_A, \omega_L) = \vec{E}_0 (\vec{r}_A, \omega_L) + \int_{V_{\text{MNP}}} d\vec{r} \left( \epsilon_m (\omega_L) - 1 \right) \vec{G} (\vec{r}, \vec{r'}, \omega_L) \vec{E}_0 (\vec{r'}, \omega_L),$$

where $\vec{E}_0 (\vec{r}_A, \omega_L) = \frac{1}{2} \tilde{\theta} \vec{E}_0 e^{-i \omega t} + \text{c.c.}$ is the incident field operator, and $V_{\text{MNP}}$ stands for the volume of the MNP. Note that for an intense incident driving field ($\vec{E}_0 (\vec{r}_A, \omega_L)$) it can be treated as a $c$-number, so that the effective Rabi field can be expressed as

$$\Omega_x = \Omega_x^0 \left[ 1 + \int_{V_{\text{MNP}}} d\vec{r'} \left( \epsilon_m (\omega_L) - 1 \right) \tilde{\theta} \vec{G} (\vec{r}, \vec{r'}, \omega_L) \tilde{\theta} \right] = \Omega_x^0 F_{\alpha\alpha},$$

where $\tilde{\theta} = \frac{1}{2} \tilde{\theta} \vec{E}_0 e^{-i \omega t} + \text{c.c.}$ is the incident field operator, and $\Omega_x^0$ is the renormalized Rabi frequency associated with the driving field and the field produced by the MNP which acts back upon the QD. $\Omega_x^0 \equiv \mu_{12} E_0/(2 \hbar)$ stands for the free space Rabi frequency, i.e., the Rabi frequency which would drive the QD transitions in the absence of the MNP and $P_{\text{dust}}^0 \equiv (\mu_{22} - \mu_{11}) E_0/(2 \hbar)$.

In this work we are interested in the dynamical evolution of the QD, so we only need the reduced density matrix of the
system. For this purpose we shall derive an effective Hamiltonian for the atom–plasmon coupling. Let us consider a unitary transformation \( U(t) \). If we define \( \rho^{(T)} = U^{\dagger} \rho U \), then

\[
\frac{\partial \rho^{(T)}(t)}{\partial t} = -i \frac{\hbar}{\hbar} [H_{\text{T}}, \rho^{(T)}(t)],
\]

where \( H_{\text{T}} = U^{\dagger} H U - i \hbar U^{\dagger} \frac{\partial U}{\partial t} \). First, we perform the unitary rotating transformation given by

\[
U_{1} = \exp \left[ -i \int_{0}^{t} \text{d} \tau S_{1}(\tau) \right],
\]

where

\[
S_{1} = \frac{\hbar \omega}{2} \sigma_{z} + \hbar \frac{P_{d,u}}{\omega_{L}} \sigma_{z} \sin(\omega_{L} t) + \hbar \int_{0}^{\infty} \omega \text{e}^{+}(\bar{r}_{a}, \omega) \tilde{E}(\bar{r}_{a}, \omega) \text{d} \omega
\]

and we arrive at the Hamiltonian:

\[
H_{1} = \hbar \Omega_{p}^{2} e^{-i(\omega_{0} - \omega_{L}) t} \text{e}^{-i \varphi_{0}^{(T)}(t)} + \hbar \int_{0}^{\infty} \omega \text{d} \omega \left( \frac{\hat{m}_{12}}{\hbar} \cdot \sigma_{+} \tilde{E}(\bar{r}_{a}, \omega) e^{-i(\omega_{0} - \omega_{L}) t} \right) + \hbar \int_{0}^{\infty} \omega \text{d} \omega \left( \frac{\hat{m}_{12}}{\hbar} \cdot \sigma_{-} \tilde{E}(\bar{r}_{a}, \omega) e^{-i(\omega_{0} + \omega_{L}) t} \right) + \text{h.c.}
\]

Note that in deriving equation (17) the rotating-wave approximation has not been considered. Equation (17) can be further simplified when using the well-known expansion of the exponential functions in terms of Bessel functions

\[
e^{i\beta \sin \gamma} = \sum_{m} J_{m}(\beta) e^{i m \gamma},
\]

\( J_{m}(\beta) \) being the Bessel function of the first kind of order \( m \). This results in

\[
H_{1} = \hbar \sum_{m} J_{m}(\beta) \Omega_{p}^{2} e^{-i(\omega_{0} - \omega_{L} + m \omega_{L}) t} + \hbar \int_{0}^{\infty} \omega \text{d} \omega \left( \frac{\hat{m}_{12}}{\hbar} \cdot \sigma_{+} \tilde{E}(\bar{r}_{a}, \omega) e^{-i(\omega_{0} - \omega_{L}) t} \right) + \hbar \int_{0}^{\infty} \omega \text{d} \omega \left( \frac{\hat{m}_{12}}{\hbar} \cdot \sigma_{-} \tilde{E}(\bar{r}_{a}, \omega) e^{-i(\omega_{0} + \omega_{L}) t} \right) + \text{h.c.},
\]

where \( \beta \equiv 2 \Re[P_{d,u}] / \omega_{L} \). \( \Re[\cdot] \) denotes the real part of the magnitude enclosed in square brackets.

It follows from equation (19) that the effects of PDM’s are included through the parameter \( \beta \) in the argument of the Bessel functions. We stress that the above Hamiltonian is applicable for arbitrary multi-photon transitions and is very difficult to obtain a general solution. In what follows, we will obtain an effective Hamiltonian for the cases of one-photon process. Therefore we assume the following resonance condition

\[
|\omega_{0} - \omega_{L}| \ll \omega_{L}.
\]

The presence of rapidly oscillating terms in equation (19) requires some kind of averaging to eliminate the fast oscillating terms. The so-called Krylov–Bogolyubov–Mitropolsky technique of averaging (54, 55) allows to derive a slowly-varying in time effective Hamiltonian. The resulting Hamiltonian describing one-photon transitions is given by (details are provided in appendix A)

\[
H^{e} = H_{0}^{e} + H_{B}^{e} + H_{\text{ext}}^{e} + H_{\text{BS}}^{e},
\]

where \( H_{0}^{e} \) is the dressed QD-driving Hamiltonian, \( H_{B}^{e} \) is the environment term, \( H_{\text{ext}}^{e} \) is the interaction of the QD with the external field, and \( H_{\text{BS}}^{e} \) represents the QD–environment coupling. They are explicitly given by

\[
H_{0}^{e} = \frac{\hbar}{2} \left( \omega_{0} - \omega_{L} + \frac{\Omega_{p}^{2}}{2 \omega_{L}} \right) \sigma_{z},
\]

\[
H_{B}^{e} = \hbar \int_{0}^{\infty} d \omega (\omega - \omega_{L}) f_{a}^{\dagger}(\bar{r}, \omega) f_{a}(\bar{r}, \omega),
\]

\[
H_{\text{ext}}^{e} = \hbar \left[ J_{0}(\beta) + J_{2}(\beta) \right] (\Omega_{a} \sigma_{+} + \Omega_{p}^{e} \sigma_{-}),
\]

\[
H_{\text{BS}}^{e} = \hbar \left[ J_{0}(\beta) \left( \frac{\hat{m}_{12}}{\hbar} \right) \cdot \sigma_{+} \tilde{E}(\bar{r}_{a}) \right] + J_{2}(\beta) \left( \frac{\hat{m}_{12}}{\hbar} \right) \cdot \tilde{E}(\bar{r}_{a}) \sigma_{z}
\]

\[
+ J_{4}(\beta) \left( \frac{\hat{m}_{12}}{\hbar} \right) \cdot \sigma_{-} \tilde{E}(\bar{r}_{a}) e^{-i \omega_{L} t} + \text{h.c.} \right],
\]

where we have defined \( \tilde{E}(\bar{r}_{a}) = \int_{0}^{\infty} d \omega \tilde{E}(\bar{r}_{a}, \omega) \).

Now we can derive from the effective one-photon Hamiltonian (21) the equations of motion of both matter and radiation. Since the quantized electric field \( \tilde{E}(\bar{r}_{a}, \omega) \) is proportional to \( f_{a}^{\dagger}(\bar{r}, \omega) \) (see equation (7)), the equation of the field operator \( f_{a}^{\dagger}(\bar{r}, \omega) \) can be obtained from the corresponding Heisenberg’s equation of motion:

\[
\frac{\partial f_{a}^{\dagger}(\bar{r}, \omega)}{\partial t} = -i \frac{\hbar}{\hbar} \left[ f_{a}^{\dagger}(\bar{r}, \omega), H^{e} \right].
\]

A formal integration results in the following expression (see appendix B):

\[
f_{a}^{\dagger}(\bar{r}, \omega) = f_{a0}^{\dagger}(\bar{r}, \omega) e^{-i(\omega_{0} - \omega_{L}) t} - \pi J_{0}(\beta) \mu_{12} \left( \frac{\Omega_{p}^{e}}{2 \omega_{L}} \right)
\]

\[
\delta G_{E}(\bar{r}, \bar{r}, \omega, \sigma_{z}(t)) \delta(\omega - \omega_{L})
\]

\[
- \pi J_{0}(\beta) \mu_{12} \delta G_{E}(\bar{r}, \bar{r}, \omega, \sigma_{+}(t)) \delta(\omega - \omega_{L} - \frac{\Omega_{p}^{e}}{2 \omega_{L}})
\]

\[
- J_{2}(\beta) \mu_{12} \delta G_{E}(\bar{r}, \bar{r}, \omega, \sigma_{-}(t)) \delta(\omega - \omega_{L} + \frac{\Omega_{p}^{e}}{2 \omega_{L}}).
\]
where $G_E(\vec{r}, \vec{r}', \omega)$ is related to the classical Green’s tensor,
$G(\vec{r}, \vec{r}', \omega)$ by

$$G_E(\vec{r}, \vec{r}', \omega) = \frac{i\omega}{c^2} V_{\epsilon(\vec{r}', \omega)} \cdot G(\vec{r}, \vec{r}', \omega).$$

(25)

The radiation field (see equation (24)) consists of four different terms. The first one corresponds to the free evolution of the field; the second one represents an emission of a photon at the laser frequency $\omega_L$; the third term represents an emission at the frequency $\omega_L + [\Omega_m]^2/(2\omega_L)$, taking into account the Bloch–Siegert shift. Finally, the last term corresponds to an emitted photon at the frequency $\omega_L - \omega_0 - [\Omega_m]^2/(2\omega_L)$. The last term is proportional to $J_1(\beta)$ and the polarization operator $\sigma^+$. This term disappears in symmetric QDs since $\mu_{22} - \mu_{11} = 0$ ($\beta = 0$) and then $J_1(\beta) = 0$. However, as we mentioned above, PDM’s are non-null in asymmetric QDs. By using realistic QDs the frequency $\omega_L - \omega_0 - (\Omega_m)^2/(2\omega_L)$ lies in the THz range, therefore the emission spectra will contain THz radiation. It should be noted that if the argument $\beta$ of the Bessel functions in equation (24) is weak ($\beta \ll 1$), the Bessel functions approximate to $J_0(\beta) \approx 1$ and $J_1(\beta) \approx \beta/2$, and the equation of motion for the field operator (equation (24)) reduces to the one obtained by Oster et al. [41].

A master equation for the reduced density matrix of the QD $\rho_S(t)$ can be obtained by tracing the density operator of the total system over the radiation reservoir variables, i.e., $\rho_S(t) = Tr_{R}[\hat{\rho}_{SR}(t)]$. Using the Born–Markovian approximation, it can be shown that the reduced density matrix obeys the following equation of motion

$$\frac{\partial \rho_S(t)}{\partial t} = \frac{i}{\hbar} [H_{0\text{eff}} + H_{\text{ext}}, \hat{\rho}_S(t)] - \frac{1}{\hbar^2} Tr_{R} \int_0^\infty dt' [H_{0\text{eff}}(t'), [H_{0\text{eff}}(t'), \hat{\rho}_S(t)\hat{\rho}_B(t')]],$$

$$= \frac{i}{\hbar} [H_A + H_{\text{ext}}, \hat{\rho}_S(t)]$$

$$- \frac{2}{\hbar^2} \sum_{n=1}^4 \frac{\gamma_{R, n} I_n^2(\beta)(\sigma^+ \sigma^- \hat{r}_n - 2\sigma^- \hat{r}_n \sigma^+ + \hat{r}_n \sigma^+ \sigma^-)}{2\omega_L}$$

$$- \frac{2}{\hbar^2} \sum_{n=1}^4 \frac{\gamma_{T, n} I_n^2(\beta)(\sigma^- \hat{r}_n \sigma^+ - 2\sigma^- \hat{r}_n \sigma^- + \hat{r}_n \sigma^- \sigma^+)}{2\omega_L}$$

$$- \frac{1}{\hbar^2} \sum_{n=1}^4 \frac{\gamma_{L, n} I_n^2(\beta)\Omega_m}{2\omega_L}(\sigma_n \gamma_n \sigma_n \gamma_n + \Omega_m^2 \pi)[\gamma_n \sigma_n \gamma_n]$$

$$- \frac{1}{\hbar^2} \sum_{n=1}^4 \frac{\gamma_{L, n} I_n^2(\beta)\Omega_m}{2\omega_L}(\sigma_n \gamma_n \sigma_n \gamma_n + \Omega_m^2 \pi)[\gamma_n \sigma_n \gamma_n].$$

(26)

Here, $\gamma_{R, n}$ ($R, L, T$) stands for the spontaneous emission of the QD modified by the presence of the MNP [53, 56–59], which is explicitly given by

$$\gamma_{R, n} = \frac{\hat{r}_n \omega_m^2}{c^2 \hbar \epsilon_0} \cdot \hat{I} \cdot \text{Im}$$

$$\times [G(\hat{r}_n, \hat{r}_n, \omega_m)] \cdot \hat{I}, \quad (j = R, T, L), \quad (u = x, z),$$

(27)

where $\omega_{R,u} = \omega_0 + [\Omega_m]^2/(2\omega_L)$, $\omega_{T,u} = \omega_L - \omega_0 - [\Omega_m]^2/(2\omega_L)$, and $\omega_{L,u} = \omega_L$. In view of equation (26) it is easy to derive the Bloch equations, which read as

$$\partial \rho_{21} = - \left[ \frac{\gamma_{R, u}}{2\omega_L} I_1^2(\beta) + \frac{\gamma_{T, u}}{2\omega_L} I_1^2(\beta) + i(\omega_0 - \omega_L) \right] \rho_{21}$$

$$+ i\gamma_{L, u} J_0(\beta) \rho_D$$

$$+ \Omega_m \frac{\gamma_{R, u}}{4\omega_L} I_1^2(\beta) \rho_D + \frac{\gamma_{T, u} + \gamma_{L, u} \Omega_m}{4\omega_L} \rho_{21},$$

$$\partial \rho_{20} = - \left[ \frac{\gamma_{R, u}}{2\omega_L} I_1^2(\beta) + \gamma_{T, u} I_2^2(\beta) \right] \rho_{20}$$

$$+ \Omega_m \left[ \frac{\gamma_{L, u}}{2\omega_L} I_1^2(\beta) - 2i \right] \rho_{12}$$

$$+ \Omega_m \frac{\gamma_{L, u}}{2\omega_L} I_2^2(\beta) + 2i \rho_{21} + \gamma_{T, u} I_1^2(\beta) - \gamma_{R, u} \rho_{21}.$$

(28)

A close inspection of equation (28) reveals that the plasmonic interaction induces a modification of the spontaneous decay rates and an enhancement of the Rabi frequency which drives the QD according to the expression given in equation (12). Furthermore, the presence of the PDM’s allows for the emission of terahertz radiation for proper values of the detuning $\omega_L - \omega_0$. As we will show later, these features allow us the possibility of controlling the THz emission by varying the QD–MNP distance $R$ and the driving field polarization. The resulting THz radiation can be characterized by means of the so-called RFS of the QD–MNP hybrid system. This spectrum is proportional to the Fourier transformation of the steady-state correlation function $\lim_{t \to \infty} \langle E^+(r, t' + t) \cdot E^-(r, t) \rangle$, where $E^-(r, t)/E^+(r, t)$ is the negative/positive frequency part of the radiation field in the far zone. In the current system it has been shown (see equation (24)) that the radiation field consists of an optical field at the frequency $\omega_0 + [\Omega_m]^2/(2\omega_L)$, which is proportional to the atomic polarization operator $\sigma^+$, and a THz field at the frequency $\omega_L - \omega_0 - [\Omega_m]^2/(2\omega_L)$, which is proportional to the atomic $\sigma^-$ term. In the case of the fluorescence photons in the optical range, the RFS is nothing but the Mollow-triplet modified by the presence of the MNP. However, in the current system we obtain photons in the THz range as a result of the polar character of the QD. In this work we focus our analysis on the RFS in such range, which can be expressed in terms of the following atomic correlation function

$$S_{\text{THz}}(\omega) \propto \hat{p}_{12}^2 \mathfrak{R} \left[ \int_0^\infty t \lim_{t \to \infty} \langle \sigma^-(t + \tau) \cdot \sigma^+(t) \rangle e^{-i(\omega - \omega_0) t} d\tau \right].$$

(29)

where $\mathfrak{R} \{ \}$ denotes the real part of the magnitude enclosed in square brackets. It is worth noting that equation (29) is different to the normal definition of resonance fluorescence spectra encountered in general textbooks that have the atomic operators $\sigma^-(t)$ and $\sigma^+(t)$ in the reverse order [60]. In our case, the atom has to be excited from the ground state into the excited state to emit a THz-photon, contradicting the normal
process, in which a photon is emitted just when the two-level system is already excited [41].

The quantum nature of the THz generated photons can be revealed by analyzing their statistical properties. This can be accomplished by computing the second-order correlation function [60] given by

\[ g_2^{(2)}(\vec{r}, t, \vec{r}, t + \tau) = \frac{\langle \hat{E}(\vec{r}, t)\hat{E}(\vec{r}, t + \tau) \rangle^2}{\langle \hat{E}(\vec{r}, t)\hat{E}(\vec{r}, t) \rangle \langle \hat{E}(\vec{r}, t + \tau)\hat{E}(\vec{r}, t + \tau) \rangle}. \]  

(30)

Since \( \hat{E}(\vec{r}, t) \) is proportional to \( \hat{f}^+ \), and in view of equation (24), for the THz emission we have that \( \hat{E}(\vec{r}, t) \) is proportional to the atomic operator \( \sigma^- \). Therefore, the probability to detect an optical photon at time \( t + \tau \) when at time \( t \) a THz photon was produced reduces to

\[ g_{01}^{(2)}(\vec{r}, t, \vec{r}, t + \tau) = \frac{\langle \sigma^-(t)\sigma^-(t + \tau)\rangle^2}{\langle \sigma^-(t)\sigma^-(t) \rangle \langle \sigma^-(t + \tau)\sigma^-(t) \rangle}. \]  

(31)

and the probability to detect an optical photon followed by a THz photon reads

\[ g_{21}^{(2)}(\vec{r}, t, \vec{r}, t + \tau) = \frac{\langle \sigma^-(t)\sigma^-(t + \tau)\rangle^2}{\langle \sigma^-(t)\sigma^- \rangle \langle \sigma^-(t + \tau)\sigma^- \rangle}. \]  

(32)

Therefore, the different magnitudes in equations (31) to (32) can be evaluated under stationary conditions as a function of the atomic operators, and for \( \tau = 0 \) result in

\[ g_{01}^{(2)}(0) = \frac{2}{1 - \langle \sigma^-(\infty) \rangle^2} \]  

and

\[ g_{21}^{(2)}(0) = \frac{1}{1 - \langle \sigma^-(\infty) \rangle^2}. \]

Oster et al [41] have shown that for the case of an isolated molecule with PMD’s strongly driven out of resonance, the Cauchy–Schwarz inequality is violated. This demonstrates the non-classical character of the pair of photons (the optical photon and the THz photon), i.e., the following results holds:

\[ g_{01}^{(2)}(0) \neq g_{21}^{(2)}(0) \neq (g_{12}^{(2)}(0))^2. \]

Here we will analyze the time dynamics of \( g_{12}^{(2)}(\tau) \) and how the presence of the MNP influences the static of the emitted photons.

We end our analysis of the properties of the hybrid system by computing the steady-state absorption spectrum of a very weak probe field in the THz range. Such spectrum is given by

\[ A_{\text{THz}}(\omega) \propto \mathcal{R} \left[ \int_{-\infty}^{\infty} \lim_{t \to \infty} \langle [\sigma^- (t + \tau), \sigma^-(t)] \rangle \right. \]

\[ \times e^{-i\omega(\omega_{\text{opt}} + \omega_{\text{MNP}})} d\tau, \]

(33)

\( \omega \) being the angular frequency of the probe field. The calculation of \( S_{\text{THz}}(\omega), g_{01}^{(2)}(\vec{r}, t, \vec{r}, t + \tau), \) and \( A_{\text{THz}}(\omega) \) can be carried out from the equation of motion of the density matrix of the system and by invoking the quantum-regression theorem [60].

3. Numerical results

We consider a dipole emitter located at a distance \( R \) of a gold spherical MNP with radius \( a = 40 \) nm. As a realistic example, we choose a QD molecule which consists of two layers with identical GaAs QD’s separated by a AlGaAs spacer. This structure can be experimentally obtained via multi-step self-assembly as described in [61–63]. The energy gap between the ground state and excited state at a temperature of 10 K is \( \omega_0 = 1.517 \) eV (2.3 \times 10^{15} \text{ rad s}^{-1}). To simplify the problem, we assume that the transition dipole moment \( \hat{\mu}_{12} \) and PDM \( \hat{\mu}_{11} \) and \( \hat{\mu}_{22} \) are aligned with the direction of the laser polarization \( \hat{u} \) (\( \hat{u} = \hat{x} \) or \( \hat{u} = \hat{z} \)). The transition dipole moment is \( |\hat{\mu}_{12}| = 1.7 \) e nm and the PDM are \( |\hat{\mu}_{11}| = 0 \) and \( |\hat{\mu}_{22}| = 0.4 \) e nm [61, 64, 65]. The QD was chosen so that the resonance frequencies of the MNP and the QD satisfy the condition \( \omega_{\text{MNP}} \equiv \omega_0 \). The lifetime of the exciton is taken to be 1 ns.

As a first step, we focus our attention on the modifications of the spontaneous emission rate at frequency \( \omega_L \) due to the presence of the MNP. We also address how the presence of the QD modifies the effective Rabi frequency felt by the TD through the parameter \( [\hat{F}_{\text{QD}}] \). The numerical results have been obtained by using the procedure described in [66]: in this computational approach, the surface of the MNP is discretized by a set of triangles and the electromagnetic potentials are matched at the triangle centers. By fulfilling the boundary conditions imposed by Maxwell’s equations through auxiliary surface charges and currents, we can estimate the optical properties of the MNP with the selected shape embedded in a dielectric environment as depicted in figure 1. We achieved a spatial resolution of approximately 1 nm. The dielectric function of the gold MNP was extracted from optical data [67]. The QD–MNP is embedded in a surrounding medium with permittivity \( \varepsilon_B = 2.25 \). The results obtained for \( \hat{u} = \hat{z} \) and \( \hat{u} = \hat{x} \) are displayed in figures 2(a) and (b), respectively.

We find a very different behavior of the decay rate \( \gamma_{\text{L,au}} \) depending on the relative orientation of the transition dipole moment \( \hat{\mu}_{12} \) with regard to the line joining the centers of the MNP and the QD. In the case that \( \hat{\mu}_{12} \) is parallel to the Z-axis there is a larger variation of the decay rate versus distance than in the case that it is parallel X-axis. This asymmetric behavior has its origin in the difference between the surface charge oscillations produced in the two situations considered: since the MNP acts like a nanoscale cavity, the vacuum fluctuations strongly depend on the relative orientation of the dipole emitter to the MNP. The plasmon modification to the effective Rabi frequency felt by the TD is also strongly influenced by the orientation of the laser field polarization. For the case with \( \hat{z} \) polarization the field correction remains below unity for all the distances considered, whereas it is enhanced for the case with \( \hat{x} \) polarization. In the limit of large distances we obtain that \( \lim_{r \to \infty} [\hat{F}_{\text{QD}}] \to 1 \) as expected.

Numerical computations indicate us that the values of \( \gamma_{\text{R,au}} \) are very close to the ones of \( \gamma_{\text{L,au}} \). The discrepancies are in the second decimal place. Taking into account that the radiative decay rates of a transition scales to the third power
were determined by $\Delta = 5 \times 10^{12}$ rad s$^{-1}$. Solid line obtained for the isolated QD (without MNP), and dashed/dashed-dotted line in the presence of the gold MNP located at $R = 60$ nm from the QD for the case with polarization $\hat{z}$.

of the frequency, the values of $\gamma_{L,u}$ were determined by properly scaling the ones for $\gamma_{L,u}$.

Let us analyze the steady state population inversion as a function of the free space Rabi frequency $\Omega^0_c$, while keeping in mind that, in order to produce THz radiation, the magnitude of the Bloch–Siegert shift $|\Omega^0_c|^2/(2\omega_L)$ should not exceed that of the difference between the laser frequency and the two-level system frequency, i.e., $|\Omega^0_c|^2/(2\omega_L) < (\omega_2 - \omega_0) \equiv \Delta$. We choose a detuning of $\Delta = 5 \times 10^{12}$ rad s$^{-1}$. With these values the above condition is satisfied for Rabi frequencies below $1.5 \times 10^{13}$ rad s$^{-1}$ which corresponds to an intensity of 1.8 GW cm$^{-2}$. In addition, the non-perturbative resonant approach used here establishes the restriction $\Omega^0_c/\omega_L \ll 1$, which is also satisfied.

Figure 3 displays the results obtained both in the absence and in the presence of the MNP. Solid line corresponds to the isolated QD and reveals that in the regime of low Rabi frequencies the QD remains in the ground state. At values greater than $2 \times 10^{13}$ rad s$^{-1}$ the upper level has a non-vanishing probability of being in the excited state. When the Rabi frequency exceeds $2 \times 10^{13}$ rad s$^{-1}$ saturation is achieved. The presence of the gold MNP modifies this situation: in the case that the polarization of the incident field is $\hat{x}$ (dashed line) the curve shifts to the right, which indicates that saturation is obtained for a free space Rabi frequency larger than in the case of the isolated QD. This result is expected since the field enhancement factor $|F_{u,x}|$ is lesser than unity (see right axis in figure 2(b)). Then, the effective Rabi frequency felt by the QD $\Omega_x$ (see equation (12)) is lower than $\Omega^0_c$. In the case that the polarization of the incident field is $\hat{z}$ (dashed–dotted line), the resulting curve shifts to the left and saturation is obtained for a free space Rabi frequency lower than in the case of the isolated QD. This behavior is easily explained by taking into account the field enhancement factor $|F_{u,z}|$ (see right axis in figure 2(a)) which shows that the effective Rabi frequency $\Omega_{u,z}$ is larger than $\Omega^0_c$.

Now we turn our attention to analyze how the plasmonic interaction modifies the steady-state RFS. To this end, we have selected different MNP–QD separations, which results in different values of either $\gamma_{L,u}$ and the field enhancement factor $F_{u,z}$ as depicted in figure 2. We used a free space Rabi frequency $\Omega^0_c = 20 \times 10^{12}$ rad s$^{-1}$, which corresponds to an intensity of 47.8 MW cm$^{-2}$.

Figure 4 displays the RFS of the low-frequency spectrum via $\log_{10} S_{THz}(\omega)$ for four different MNP–QD separations when the polarization of the incident electric field is set to $\hat{u} = \hat{x}$ (figure 4(a)) and $\hat{u} = \hat{z}$ (figure 4(b)). Solid line shows the RFS when the MNP is not present or very far from the QD. The spectrum shows the well-known behavior consisting in a Lorentzian central peak at $\omega_f = 4.9 \times 10^{12}$ rad s$^{-1}$, with a natural width $\gamma_{p,z}$ and a blue outer sideband at $\omega_b = 45 \times 10^{12}$ rad s$^{-1}$.

The outer sideband is located at $\omega_b = \omega_c + \sqrt{(2\Omega^0_c)^2 + \Delta^2}$, as predicted by evaluating it in the dressed state picture. The red sideband is not produced since for the current values of the parameters used in the simulation, the
corresponding frequency $\omega - \sqrt{(2\Omega)^2 + \Delta^2}$ reaches a negative value. Thus, such a sideband has no meaning from a physical point of view. The other curves are obtained for three different MNP–QD separations. In the presence of the MNP the sideband is located at $\omega_{s,x} = \omega_L - \omega_0 - |\Omega_x|^2/(2\omega_L)$ and it is dependent on the polarization of the incident field. In the case with polarization of the incident field $\hat{u} = \hat{x}$ (see figure 4(a)), the plasmonic effects manifest as a negative shift for the blue sideband as the MNP–QD distance decreases. The tunability of the frequency of the sideband is estimated to be around 4 THz. This behavior is expected since $|F_{ex}|$ remains below unity and decreases as the distance $R$ decreases (see figure 2(b)). In addition, the central line frequency moves slightly to positive frequencies reaching a frequency of $\omega_{c,x} = 5 \times 10^{12}$ rad s$^{-1}$ for the shortest distance. The central line of the RFS is located at $\omega_{c,x} = \omega_L - \omega_0 - |\Omega_x|^2/(2\omega_L)$ which is dependent on the distance $R$ as predicted by the theoretical model (see equation (12) and figure 2(b)). In the case with polarization of the incident field $\hat{u} = \hat{z}$ (see figure 4(b)), the central line is located at $\omega_{c,z} = \omega_L - \omega_0 - |\Omega_z|^2/(2\omega_L)$ which is red-shifted in comparison with the absence of MNP. This is due to the high enhancement of the field felt by the QD. The frequency of the sideband is blue-shifted with regard to the case without MNP and the tunability of the frequency of the sideband is estimated to be around 12 THz. To better analyze the behavior of the sideband of the THz photons we compute its spectral position as a function of both the MNP–QD separation distance $R$ and the free space Rabi frequency $\Omega_0^2$. The result is depicted in figures 4(c) and (d) in the case that the polarization of the incident field is set to $\hat{x}$ and $\hat{z}$, respectively. A large degree of tunability for the frequency of the THz photons is achieved. This can be controlled by means of $\Omega_0^2$. Note that the detuning of the laser

Figure 4. (a), (b) Steady-state RFS ($S_{THz}(\omega)$) versus $\omega$ for the hybrid QD–MNP for different distances: $R = 50$ nm (dashed line), $R = 60$ nm (dashed–dotted line), $R = 80$ nm (dotted line), and $R = \infty$ nm (solid line). The incident field is polarized along the $X[Z]$ axis. (c), (d) Spectral position of the sidebands of the terahertz photons $\omega_{s,x} / 2\pi$ as a function of the distance $R$ between the QD and the MNP and the free space Rabi frequency $\Omega_0^2$, when the incident field is polarized along the $X[Z]$ axis, and $\Delta = 5 \times 10^{12}$ rad s$^{-1}$. Numbers indicate the value of $\omega_{s,x} / 2\pi$ in THz.
field $\Delta$ can be also externally changed, thus providing a full set of knobs to adjust the production of THz radiation. In summary, the plasmonic interaction results in a huge shift of the frequency of the sideband which can be adjusted by proper selection of the distance $R$, the free-space Rabi frequency ($\Omega_{0}^R$), and the detuning $\Delta$ of the driving laser. Such interaction is accompanied by a weak modification of the frequency of the central line of the spectrum of the THz photons (the Bloch–Siegert shift depends indirectly on $R$ through the effective Rabi field $\Omega_{0}$).

It is worth mentioning that besides the production of THz photons, the strongly detuned driven QD also produces optical photons. The steady-state RFS of such photons can be computed using the corresponding two-time correlation function: $S_{opt}(\omega) \propto \langle \hat{c}_{1} \hat{c}_{2} \rangle \sum_{ \omega_{R} \omega_{\ell} } \langle \sigma^{\dagger} (t + \tau) \sigma (t) \rangle e^{-i\omega_{R} - i\omega_{\ell} \tau} d\tau$. In this case the order of the operators appearing in the two-time correlation is the usual (see for example [60]). The results obtained for the optical photons can be summarized as follows: in the absence of the MNP we recover the usual Mollow-triplet with a central line and two additional sidebands. The gold MNP produces a shifting of the sidebands of the optical photons (not shown). The sign of the shift depends on the polarization of the incident field.

In order to bring to light the quantum nature of the THz photons we evaluate the second-order intensity correlation $g_{2}(\tau)$. We used the free space Rabi frequency value used above, i.e., $\Omega_{0}^R = 20 \times 10^{12}$ rad s$^{-1}$. Figure 5(a) presents the results obtained in the absence of MNP (solid line) and in the presence of the gold MNP when the polarization of the driving field is $\hat{x}$ (dashed/dashed–dotted line). The horizontal axis corresponds to the time elapsed after the emission of the THz photon normalized to the period of the incident field $T$, where $T = 2\pi/\omega_{L}$. We can see that the intensity–intensity correlation starts at $\tau = 0$ with a positive value and alternates periods of emission and darkness (bunching/anti-bunching) which is a clear signature of the quantum character of the process. The period of such oscillations is enlarged (shortened) for $\hat{\alpha} = \hat{x}$ ($\hat{\alpha} = \hat{z}$). This is expected since the period depends on the effective Rabi field $\Omega_{0}$ which varies with the polarization of the incident field. Note that almost half of the time in a period, the correlation flips from bunching to anti-bunching. If we select a free space Rabi frequency an order of magnitude lower than the previous one, we move close to the region where the system is weakly excited (see figure 3). The results for $\Omega_{0}^R = 2.5 \times 10^{12}$ rad s$^{-1}$ are displayed in figure 5(b). We observe that in the absence of MNP (solid line) the two-time correlation oscillates but keeps values in the bunching regime. This behavior is also reproduced in the presence of the gold MNP when the polarization of the incident field is $\hat{x}$ (dashed line). However, when the polarization of the incident field is $\hat{z}$ (dashed–dotted line) we recover a periodic transition from bunching to anti-bunching, although the interval of time in the latter regime is lesser than $T/2$.

The dark periods appearing in figure 5(a) correspond to the instants of time when the system has a non-vanishing probability to be in the upper state. Thus, it is unlikely to obtain an optical photon preceded by a THz photon since those photons are produced mainly when the system is in the lower state. To see this effect we plot in figure 5(c) the time evolution of inversion. There is a one-to-one correspondence between the dark periods of the second-order correlation and the periods where population inversion is obtained. Figure 5(d) shows the population inversion when the system is weakly excited. In this case, the absence of dark periods in the second-order correlation (see solid and dashed lines in figure 5(b)) are correlated with the time evolution of $\sigma_{z}(\tau)$. There is a nearly vanishing probability to find the system in the upper state, thus the signals remain in the bunching regime. The situation changes dramatically for the polarization $\hat{\alpha} = \hat{z}$ (see dashed–dotted line in figures 5(b) and (d)), where a brief period of darkness is obtained which corresponds to those instants of time where $\sigma_{z}(\tau)$ reaches positive values.

Finally we estimate the refractive properties of a weak probe field in the THz range interacting with the hybrid system. The susceptibility for such field can be computed by evaluating equation (33). The results obtained when the incident field is polarized along the $X$ axis are depicted in figure 6(a). To better appreciate the narrowness of those spectral features we shifted the horizontal axis for each curve by the corresponding frequency of the sideband $\omega_{s,x}$, which are coincidental with those appearing in the RFS of the THz spectra, and the results are displayed in figure 6(b). Here we can see that besides the red-shifting of the sideband due to the presence of the MNP, the peak of gain is strongly enhanced as a result of the plasmonic interaction. However, for the shortest distance ($R = 50$ nm, dashed line) the peak is reduced. This effect has its origin in the fact that for such short distance the plasmonic interaction results in the enhancement of the decay rate $\gamma_{j,x}$ ($j = R, L$) as shown in the left axis of figure 2(b), whereas for the other considered distances the plasmonic interaction results in a lowering of $\gamma_{j,x}$ ($j = R, L$) compared to free space values of the $\gamma$’s. The larger the time expended in the excited state, the greater the amplification obtained. The FWHH of the gain curves follows the dependence of $\gamma_{j,x}$ ($j = R, L$) on the distance $R$ between the MNP and the QD. The amplification of the weak probe is dramatically modified when the incident field is polarized along the $Z$ axis as shown in figure 6(c) where the presence of the MNP results in the decrease of gain. We know from left axis in figure 2(a) that the plasmonic interaction results in a huge enhancement of the $\gamma$’s and, consequently, the reduction of the time the system remains excited, thus the obtention of wider curves for the probe gain should not come as a surprise. The behavior found indicate that the use of an incident field along the $X$ axis is most favorable for the obtention of large amplification of the probe signal.
4. Conclusions

We propose a scheme for controlling the absorption spectrum and RFS of a QD with broken inversion symmetry interacting with a plasmonic nanostructure. The QD is described as a two-level atom-like system with a permanent dipole moment in the excited state. A linearly polarized laser field drives the optical transition of the QD and produces localized surface plasmons in the MNP. The presence of the MNP leads to local field corrections and modifies the spontaneous emission rate of the allowed optical transition. The influence of plasmonic effects between the MNP and the QD is analyzed using the Green tensor method. It is shown that these effects result into dramatic modifications of the optical properties of the fluorescence photons. If the laser frequency lies above the transition frequency of the QD, a new set of peaks at THz frequencies appear in the RFS. The position and strength of these peaks can be controlled by the separation between the QD and the MNP. The physical origin of this behavior can be understood in terms of the induced Bloch–Siegert shift and its modification by the plasmonic interaction. The quantum nature of the emitted THz photons is analyzed by means of the second-order intensity correlation. It is shown that the intensity–intensity correlation starts at $\tau = 0$ with a positive value and alternates periods of emission and darkness (bunching/anti-bunching) which is a clear signature of the quantum character of the process. The period of such oscillations can be controlled by the polarization of the driving field. Finally we estimate the refractive properties of a weak probe field in the THz range interacting with the hybrid system. It is shown that a red-shifting of the absorption sideband accompanied by an enhancement or decreasing of the peak of gain take place depending on the QD–MNP separation.

Figure 5. Intensity–intensity correlation function $g_{12}(\tau)$, describing the probability to detect an optical photon after a THz photon as a function of time elapsed after the emission of the terahertz photon $\tau = t/T$. Solid line corresponds to the intensity–intensity correlation in the absence of MNP. Dashed/dashed–dotted line corresponds to the intensity–intensity correlation for a gold MNP located at $R = 60$ nm from the QD when the incident field is polarized along the $X/Z$ axis ($\hat{a} = \hat{x} \hat{z}$). The detuning of the driving field is $\Delta = 5 \times 10^{12}$ rad s$^{-1}$, and the free space Rabi frequency is set to $\Omega_0^E = 20 \times 10^{12}$ rad s$^{-1}$ in (a), and to $\Omega_0^E = 2.5 \times 10^{12}$ rad s$^{-1}$ in (b). (c)/(d) Time evolution of inversion $\langle \sigma_z(t) \rangle$ for the curves depicted in (a)/(b).
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Appendix A. Effective one-photon Hamiltonian

In order to eliminate the fast oscillating terms appearing in the Hamiltonian in equation (18) and calculate the effective Hamiltonian, we apply a perturbation theory using the Krylov–Bogolyubov–Mitropolsky procedure of averaging over fast oscillations [54]. In particular, given the Hamiltonian $H(t)$ and retaining the second-order terms in the interaction, we can obtain the effective Hamiltonian as follows [54, 55]

$$H^e = H^{(1)}_e + H^{(2)}_e,$$  \hspace{1cm} (A.1)

where

$$H^{(1)}_e = \langle H_l(t) \rangle$$

$$H^{(2)}_e = \frac{1}{2} \left\{ \int_0^t d\tau [H_l(\tau) - \langle H_l(t) \rangle, H_l(t)] \right\}. \hspace{1cm} (A.2)$$

For the case of a Hamiltonian made up of a sum of harmonic terms of the form

$$H_l(t) = \sum_m \hbar \omega_m e^{-i\omega_m t} + \text{h.c.},$$  \hspace{1cm} (A.3)
it is easy to see that the $H_c$ is given by [55]

$$
H^c = \langle H(t) \rangle + \sum_{n,m} \frac{1}{\Delta \omega_n} \mathcal{A}^n \beta^m \left[ h_{nm}^T \right] + \frac{1}{\Delta \omega_n} \mathcal{A}^n \beta^m \left[ h_{nm}^T \right] + h.c.
$$

(A.4)

In order to apply the averaging method to the multi-photon-resonance Hamiltonian given in equation (19), we assume the one photon-resonance condition

$$
|\omega_0 - \omega_L| \ll \Delta \omega_n.
$$

(A.5)

Thus, all the terms in the sum in equation (19) oscillate very fast, with the exception of terms that are identified as the resonant or quasi-resonant terms. The effect of the non-resonant terms therefore can be neglected when studying the long-time system dynamics. With this condition in mind, we arrive to the following effective Hamiltonian:

$$
H^e = \Omega_0 \sigma^+ e^{i (\omega_0 - \omega_L)t} + \Omega_0 \sigma^- e^{i (\omega_0 - \omega_L)t}
$$

$$
+ \frac{\hbar \Omega_0^2}{2 \omega_L} \sigma_z
$$

$$
+ \hbar \left[ J_0(\beta) \int_{-\infty}^{\infty} d\omega \left( \frac{\mu_{12}}{\hbar} \right) \sigma^+ \tilde{E}(r_0, \omega) e^{-i(\omega_0 - \omega_L)t} \right]
$$

$$
+ \left( J_0(\beta) \Omega_0^2 \int_{-\infty}^{\infty} d\omega \left( \frac{\mu_{12}}{\hbar} \right) \sigma^- \tilde{E}(r_0, \omega) \right]
$$

$$
+ \left( J_0(\beta) \int_{-\infty}^{\infty} d\omega \left( \frac{\mu_{12}}{\hbar} \right) \sigma^+ \tilde{E}(r_0, \omega) \right]
$$

$$
\times \left[ e^{-i(\omega_0 - \omega_L)t} + h.c. \right].
$$

(A.6)

We make use of the following unitary transformation

$$
U_2 = \exp \left[ -\frac{i}{\hbar} \int_{0}^{\infty} d\tau S_1(\tau) \right].
$$

(A.7)

where

$$
S_1(t) = \frac{\hbar}{2} \left[ \frac{\omega_0 - \omega_L}{\Delta \omega_n} + \int_{-\infty}^{\infty} d\omega (\omega - \omega_L) \right]
$$

$$
\times \tilde{E}^+(r_0, \omega) \tilde{E}(r_0, \omega)
$$

(A.8)

and the Hamiltonian given in equations (21) to (22) is obtained.

### Appendix B. Equation of motion of the operator of the photon subsystem

The equation of the motion for the field operator $\mathcal{F}_A(t)$ (r, \omega) can be obtained from the corresponding Heisenberg’s equation of motion:

$$
\frac{\partial \mathcal{F}_A(t, r, \omega)}{\partial t} = \frac{i}{\hbar} [\mathcal{H}_A(t, r, \omega), \mathcal{F}_A(t, r, \omega)].
$$

(B.1)

Taking into account the expression for the effective Hamiltonian (equation (22)) and recalling the commutation rules for the field operator $\mathcal{F}_A(t, r, \omega)$:

$$
\mathcal{F}_A(t, r, \omega) \mathcal{F}_A(t', r', \omega) \mathcal{F}_A(t, r', \omega) \mathcal{F}_A(t', r, \omega)
$$

$$
= \delta(t - t') \delta(r - r') \delta(\omega - \omega') \delta(r - r')
$$

(j, k = x, y, z), it is easy to derive the Heisenberg equation of motion:

$$
\frac{\partial \mathcal{F}_A(t, r, \omega)}{\partial t} = i(\omega - \omega_L) \mathcal{F}_A(t, r, \omega) - J_0(\beta) \mu_{12} \left( \frac{\Omega_0^2}{2 \omega_L} \right)
$$

$$
\times \sigma_+(t) \sigma^- \mathcal{F}_E(t, r, \omega) - J_0(\beta) \mu_{12} \sigma^+(t) \sigma^- \mathcal{F}_E(t, r, \omega)
$$

$$
- J_0(\beta) \mu_{12} \sigma^+(t) \sigma^- \mathcal{F}_E(t, r, \omega).
$$

(B.2)

where $\mathcal{F}_E(r, r', \omega)$ is related to the classical Green’s tensor, $\mathcal{G}(r, r', \omega)$ by

$$
\mathcal{G}(r, r', \omega) = \frac{\omega^2}{e^2 \pi \epsilon_0} \mathcal{F}_A(t, r, \omega).
$$

(B.3)

Formal integration of equation (B.2) results in

$$
\mathcal{F}_A(t, r, \omega) = \mathcal{F}_A(0, r, \omega) e^{-i(\omega_0 - \omega_L)t}
$$

$$
- J_0(\beta) \mu_{12} \left( \frac{\Omega_0^2}{2 \omega_L} \right) \int_{0}^{\infty} dt' \sigma_+(t') e^{-i(\omega - \omega_L)(t - t')}
$$

$$
\times \sigma^- \mathcal{F}_E(t, r, \omega)
$$

$$
- J_0(\beta) \mu_{12} \sigma^+(t) \sigma^- \mathcal{F}_E(t, r, \omega)
$$

$$
- J_0(\beta) \mu_{12} \sigma^+(t) \sigma^- \mathcal{F}_E(t, r, \omega).
$$

(B.4)

In order to pick out the resonant interactions from the equation of motion, we introduce slowly varying amplitude operators $\sigma^Z(t) = \sigma^Z(t) e^{i(\omega_0 - \omega_L)t}$ and apply the Markov approximation. This involves taking the slowly varying amplitude out of the integral at the upper time $t$. Thus equation (B.4) reads

$$
\mathcal{F}_A(t, r, \omega) = \mathcal{F}_A(0, r, \omega) e^{-i(\omega_0 - \omega_L)t}
$$

$$
- J_0(\beta) \mu_{12} \left( \frac{\Omega_0^2}{2 \omega_L} \right) \sigma^Z \mathcal{F}_E(t, r, \omega)
$$

$$
\sigma^- \mathcal{F}_E(t, r, \omega)
$$

$$
- J_0(\beta) \mu_{12} \sigma^+(t) \sigma^- \mathcal{F}_E(t, r, \omega)
$$

$$
- J_0(\beta) \mu_{12} \sigma^+(t) \sigma^- \mathcal{F}_E(t, r, \omega).
$$

(B.5)

We further make the replacement

$$
\int_{0}^{\infty} dt e^{-i(\omega_0 - \omega_L)(t - t')}
$$

$$
\approx \pi \delta(\omega - \omega_0)
$$

(B.6)
and neglect the Lamb shifts. In this way we obtain

\[
\begin{align*}
\tilde{r}_A^i (\vec{r}, \omega) &= \int_0^1 \int_0^1 \int_0^1 \int_0^1 \frac{\Omega_{ii}^2}{2\omega_l} \Delta G_E (\vec{r}, \omega) \delta (\omega - \omega_l) \\
&- \pi \delta (\omega - \omega_l) \left( \sum_{i} \Omega_{ii}^2 \right) \Delta G_E (\vec{r}, \omega) \sigma_i (t) \delta (\omega - \omega_l) \\
&- \pi \delta (\omega - \omega_l) \left( \sum_{i} \Omega_{ii}^2 \right) \Delta G_E (\vec{r}, \omega) \sigma_i (t) \delta (\omega - \omega_l) \\
&- J_i (\beta) \mu_{12} \Delta G_E (\vec{r}, \omega) \sigma_i (t) \delta (\omega - \omega_l) \\
&\times \left( \omega - \omega_l + \frac{\Omega_{ii}^2}{2\omega_l} \right).
\end{align*}
\]

Taking into account the resonance condition, we can approximate the exponentials in equation (B.7):

\[
\left( \omega_0 - \omega_l + \frac{\Omega_{ii}^2}{2\omega_l} \right) \approx 0,
\]

thus equation (24) in the main text is obtained.

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