Enhanced spontaneous down-conversion in plasmonic and quantum emitter hybrid structures

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Metallic nanoparticles provide extreme localization and enhancement of optical fields. The existence of quantum emitter in the vicinity (hot spots) of these plasmonic particles introduces the weak hybridization in the excited state of plasmon and leads to create Fano resonances. Here we demonstrate that using such hybrid structures as the interaction centers in a nonlinear crystal, one can enhance generated down-converted fields. The results indicate that the intensity of down-converted field can be strengthened 6-orders of magnitude with reliable interaction strengths and by choosing the appropriate level spacing for the quantum emitter.
INTRODUCTION

Spontaneous down conversion (SDC) is a second-order non-linear process in which a single photon splits into pair photon of smaller frequencies commonly labelled as signal and idler [1]. Nonlinear materials like Barium Borate [2], KTP [3] are used to generate such kinds of photon pairs due to their large nonlinearities. The one of the key features of SDC is that it can exhibit entanglement in various degrees of freedom such as polarization, space, time, and orbital angular momentum [4]. Entangled photon pairs are essential for many fundamental quantum optics experiments [5] as well as a key resource in quantum communication including cryptography [6], quantum computation [7] and quantum information [8]. SDC can also be used in solar cell applications by means of reducing the energy losses with converting high energy photons into two or more lower energy photons [9, 10].

SDC efficiency (conversion rate) is one of the main disadvantage of generation photon pair [5]. Some works were done to increase efficiency including changing the geometry/structure of the crystals [11, 12] and using quantum dots to enhance down conversion rate [13]. The use of plasmonic structures to enhance SDC processes was theoretically studied in Refs [14, 15] and 5-orders of magnitude enhancement was predicted. It was also shown that integration of plasmonic nanostructures with nonlinear devices such as fiber glasses [16, 17] and photonic crystals [18–20] can be used to enhance nonlinear processes.

The property of metal nanoparticles (MNPs) to localize incident light into the small volumes (hot spots) provide strong light-matter interaction. Quantum emitters (QEs) placed at these hot spots couple to the polarization field of the plasmon excitation, which is several orders of magnitude larger compared to the coupling of the QE to the incident light. This strong coupling can be used to test many quantum optical applications such as quantum entanglement [21, 22], single-mode nonclassicality [23] and squeezing [24, 25]. A recent study [26] shows that it is possible to entangle many QEs which can also be utilized as entanglement source in quantum networks [27–29].

The existence of QEs in the vicinity of hot spots of MNPs introduces the weak hybridization in the excited state of plasmon and leads to create Fano resonances, a dip in the absorption spectrum. The phenomenon of Fano resonance is not only the appearance of a dip in the absorption spectrum. They can also increase the lifetime of plasmon oscillations [30, 31] leading the further accumulation of the field strength at the hot spots. This gives rise to much stronger field enhancements in the hot spots, dark-hot resonances [32].

In this work, we demonstrate that such scheme can be used to enhance down-converted field. We find that down-converted field intensity can be enhanced 6-orders of magnitude depending on the interaction strength between MNP and QE and also level spacing of QE ($\omega_{eg}$). The existence of MNP-QE hybrid structure introduces an extra term, which can be tuned properly. The new term can cancel the non-resonant term in the conversion and create enhancement in the response of nonlinear field. In other words, controllable constructive or destructive interface can be obtained by using appropriate QE and/or by tuning interaction strength between MNP and QE. In Figure 2 and Figure 3, we show enhancement of down-converted field intensity (around 6-orders of magnitude) depending on interaction strengths $f_c$ and $g$ respectively for a various QE level spacing.

STRUCTURE

We consider the structure as shown in Figure 1. The incident light of frequency $\omega$ interacts with nonlinear crystal and excites $a_2$-mode. Generated photons, $a_1$, interact with embedded MNPs and excites surface plasmons $a_p$ with interaction strength $g$. QEs with level spacing $\omega_{eg}$ are placed in the vicinity of MNPs. The level spacing of QE is chosen close to plasmon field oscillations ($\omega_{eg} \sim \Omega_p$). Here, we consider the strong interaction between MNP and QE ($f_c$) that we neglect the interactions between crystal fields and QE. We also assume the response of the MNP to the pumped field is weak.
and generates two photons oscillating with $\omega$ and generates two photons oscillating with $\omega/2$. Generated photons interact with MNP-QE hybrid structures.

**HAMILTONIAN AND EQUATIONS OF MOTION**

In this section, we first derive the effective Hamiltonian for embedded MNP-QE hybrid structure into the material having SDC property. We then obtain the equations of motion of this system, which is solved by numerically. Finally, we will discuss the results.

The interaction Hamiltonian for SDC process can be written as

$$\hat{H}_{dc} = \left\{ \int d^3r \chi_2(r) E_2^*(r) E_1^2(r) \right\} \hat{a}_2^\dagger \hat{a}_1 + H.c.$$  \hspace{1cm} (1)

where $E_2^*(r)$ ($E_1^2(r)$) is the positive (negative) frequency part of the electric field. $\hat{a}_j^\dagger$ ($\hat{a}_j$) is the creation (annihilation) operator of the j-th mode and $H.c.$ stand for Hermitian conjugation. The integral in the parenthesis, $\chi^{(2)} = \int d^3r \chi_2(r) E_2^*(r) E_1^2(r)$, is the overlap integral, which gives the strength of the SDC process. Here, one can consider $\chi_2(r)$ as a 3D step function which is zero outside the crystal.

The total Hamiltonian of the system can be written as the sum of the energy of the crystal oscillations of the pumped ($\Omega_2$) and down converted ($\Omega_1$) fields as well as plasmon oscillations ($\Omega_p$), QE ($\omega_{eg}$) and and the energy transferred by the laser source ($\varepsilon e^{-i\omega t}$).

$$\hat{H}_0 = h\Omega_2 \hat{a}_2^\dagger \hat{a}_2 + h\Omega_p \hat{a}_p^\dagger \hat{a}_p + h\omega_{eg} |e\rangle \langle e|$$  \hspace{1cm} (2)

$$\hat{H}_L = i\hbar (\varepsilon \hat{a}_2^\dagger e^{-i\omega t} - h.c.)$$  \hspace{1cm} (3)

Here $|g\rangle$, $(|e\rangle)$ is the ground (excited) state of the QE. The interaction part, $\hat{H}_{int}$, contains the interactions between down converted fields and plasmon mode of strength $g$, the interaction between QE and MNP ($f_c$) and the SDC process $\hat{H}_{dc}$ as defined in Eq.(1).

$$\hat{H}_{dc} = h\chi^{(2)}(\hat{a}_2^\dagger \hat{a}_1 + \hat{a}_1^\dagger \hat{a}_2)$$  \hspace{1cm} (4)

$$\hat{H}_{int} = h f_c (\hat{a}_p^\dagger |g\rangle \langle e| + |e\rangle \langle g| \hat{a}_p) + h g (\hat{a}_p^\dagger \hat{a}_1 + \hat{a}_1^\dagger \hat{a}_p).$$  \hspace{1cm} (5)

The dynamics of the system can be derived by using Heisenberg equation of motion (e.g. $i\hbar \dot{\hat{a}}_i = [\hat{a}_i, \hat{H}]$). Since we are interested in intensities but not in correlations, we replace the operators $\hat{a}_i$ and $\hat{\rho}_{ij} = |i\rangle \langle j|$ with complex numbers $\alpha_i$ and $\rho_{ij}$ respectively and desired equations of motion can be obtained as

$$\dot{\alpha}_2 = -(i\Omega_2 + \gamma_2) \alpha_2 - i\chi^{(2)} \alpha_1^2 + \varepsilon e^{-i\omega t}$$  \hspace{1cm} (6a)

$$\dot{\alpha}_1 = -(i\Omega_1 + \gamma_1) \alpha_1 - 2\chi^{(2)} \alpha_1^* \alpha_2 - i\omega_{eg} \rho_{ge}$$  \hspace{1cm} (6b)

$$\dot{\alpha}_p = -(i\Omega_p + \gamma_p) \alpha_p - i\omega_{eg} \rho_{ge} + i f_p \alpha_p (\rho_{ge} - \rho_{gg})$$  \hspace{1cm} (6c)

$$\dot{\rho}_{ge} = -(i\omega_{eg} + \gamma_{eg}) \rho_{ge} + i f_p \alpha_p (\rho_{ge} - \rho_{gg})$$  \hspace{1cm} (6d)

$$\dot{\rho}_{ee} = -\gamma_{ee} \rho_{ee} + i (f_p \rho_{ge} \alpha_p^* - c.c.)$$  \hspace{1cm} (6e)
where $\gamma_2$, $\gamma_1$, $\gamma_p$ and $\gamma_{ee}$ are the damping rates of pumped and down-converted fields, plasmon field and quantum emitter respectively. We have also additionally conservation of probability $\rho_{ee} + \rho_{gg} = 1$ and the off-diagonal decay rate of the QE $\gamma_{eg} = \gamma_{ee}/2$ as the constraints.

### Enhancement

The enhancement factor for down-converted field intensity can be defined

$$\text{Enhancement Factor} = \left| \frac{\alpha_1(f_c \neq 0, g \neq 0)}{\alpha_1(f_c = 0, g = 0)} \right|^2$$

in the presence ($f_c \neq 0, g \neq 0$) and absence ($f_c = 0, g = 0$) of the MNP-QE hybrid structure. By time evolving Eqs. (6a-6e) and using Eq. (7), one can examine the effect of MNP-QE hybrid structure to the output signal of the system.

In a realistic picture, playing with the oscillation parameter $\Omega_1$ is not practical for a given material, but it is possible to tune interaction strengths $g$ and $f_c$ by adjusting positioning of nanoparticles or by deforming geometry of the material. Furthermore, one can also use different type of QEs with various level spacing ($\omega_{eg}$). In Figure 3, we demonstrate how the interaction strength ($f_c$) between MNP and QE plays role in the enhancement of down-converted field intensity for different QE level spacing ($\omega_{eg}$). It can be seen that 6-orders of magnitude enhancement of the down-converted field intensity can be obtained. Frequencies are scaled with $\Omega_2$ and we use $\omega = 1.02 \Omega_2$, $\Omega_1 = \Omega_2/2$, $\Omega_p = 0.51 \Omega_2$, $\gamma_1 = \gamma_2 = 10^{-5} \Omega_2$, $\gamma_p = 0.1 \Omega_2$ and $\gamma_{eg} = 10^{-4} \Omega_2$. We take $\chi^{(2)} = 10^{-9} \Omega_2$. The choice of $\chi^{(2)}$ does not affect the relative enhancement ratios as long as it is small.

The interaction strength between MNP and down-converted field also plays significant role in the enhancement of the generated photon intensity. In Figure 3 we demonstrate this dependence for a various QE level spacing ($\omega_{eg}$). It can be seen from figure, when QE level spacing ($\omega_{eg}$) gets closer to resonance frequencies (i.e. $\omega_{eg} \sim \omega/2$), there is not significant change in the generated photon intensity. However, there emerges enhancement between 0.35 - 0.45 $\Omega_2$ values. The origin of such enhancement comes from the path interference effect in the nonlinear response. Since, presecence of MNP-QE defines extra terms which may lead constructive or destructive interferences. When $\omega_{eg}$ of the QE or the interaction strengths are tuned properly, the new term can cancel the non-resonant term in the conversion, which creates the enhancement. Depending on these parameters better cancellation can be made and larger enhancement at the output signal can be obtained.
FIG. 3. Enhancement factor for down-converted field intensity, $|\alpha_1|^2$, with respect to interaction strength $g$ and QE level spacing $\omega_{eg}$. Here, we take coupling strength between MNP and QE as $f_c = 0.001 \Omega_2$ and obtained 6-orders of enhancement. The rest of the parameters are the same as used in the previous figure. The results are obtained by numerical time evolution of Eqs. (6a–6e).

DISCUSSION AND CONCLUSIONS

This work is devoted to a theoretical study of an enhancement of down-converted field intensity using MNP-QE hybrid structures as the interaction centers. We find that it is possible to strength nonlinear responses of materials having inherently weak output. Plasmonic nanostructures embedded in nonlinear crystal can be used for increasing the collection of light. The existence of QE in the vicinity of MNP provide to control the field at the hot spots and output signal from these interaction centers. We demonstrate that with sufficient interaction strengths one can enhance down-converted field intensity 6-orders of magnitude. However, the enhancement does not always increase with a stronger interactions between MNP-QE, see Figure 2. The results obtained here may help to increase the efficiency of SDC process and generation of entangled photons, which is important for many applications, such as quantum computing and quantum optical experiments.

MET and MG acknowledges support from TÜBİTAK Grant No. 117F118. MET acknowledges support from TUBA-GEBİP award.

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