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Abstract. The article is focused on the investigation of features of quantum dynamics for localized plasmons in spaser systems consisting of metal nanoparticles and semiconductor quantum dots. The non-classical plasmon states generation in a three-particle spaser system with nonlinear plasmon-exciton interaction is predicted.

1. Introduction
Chains of near-field coupled nanoparticles (NPs) and quantum dots (QDs) are of significant interest as a platform for quantum computing [1, 2]. The bunching (antibunching) effects can be observed for emitted photons from self-assembled QD structures [3]. On the other hand, if QDs are coupled with shared NP [4, 5], direct transmission of quantum correlations from bound excitons to plasmons can be achieved. One of the main advantages of such correlated plasmons is the possibility of simpler external control and manipulation of their carriers – NPs. In particular, this addressing can be carried out using the epifluorescence microscopy technique for single quantum objects [6].

In this paper we propose a model of three nanoobjects (NP-QD-NP) coupled by nonlinear dipole-dipole interactions in the presence of an external magnetic field. The nonlinear regime of this ensemble corresponds to the two-quantum processes of the QD biexciton decay in the case $|\delta| > \Omega_{1,2}, |\Delta|$, where $\delta = \bar{\omega} - \omega$ and $\Delta = \bar{\omega} - \omega_p$, $\bar{\omega}$ is the spasing frequency, and $\Omega_{1,2}$ are the Rabi frequencies of dipole-dipole interactions between the QD and NPs. As a result of this nonlinear process, one can expect the appearance of strongly correlated plasmon pairs.

In the technical framework, the presented spaser systems can be used to generate nonclassical states of the electromagnetic field at the nanoscale. Such systems can be integrated in the individual plasmonic waveguides [7] and plasmonic circuits for quantum information processing [8, 9].

2. Entangled plasmon generation in the nonlinear regime of a three-particle spaser
Nonlinear regimes of interaction between NP and QD can be realized, firstly, in the presence of a two-photon pump in the system [10, 11] and secondly, under the condition that the coupling energy between two electron-hole pairs is of the same order of magnitude as the internal coupling energy of a single pair. In this situation the coupled states of two electron-hole pairs (biexcitons
of QDs) can appear [10]. The energy of biexciton state \(XX\) differs from the double energy of the exciton \(X\) by the biexciton binding energy \(\Delta_{XX}\) (see Fig. 1).

We choose such parameters of the spaser for which a cascade process is not realized in the system, but a pure two-quantum transition occurs [12]. In the conditions corresponding to scheme in Fig. 1 this regime is realized due to far-off-resonant interaction in the case \(|\delta| > \Omega_{1,2}\). Such a process can lead to the generation of nonclassical states of plasmons and it is described by the following Hamiltonian [13]:

\[
H = \hbar \omega_{p1} \hat{c}_1^+ \hat{c}_1 + \hbar \omega_{p2} \hat{c}_2^+ \hat{c}_2 + \frac{\hbar \omega_{XX}}{2} D + \hbar \Omega^{(2)} \left( \hat{c}_1 \hat{c}_2 \hat{S} + \hat{c}_1^+ \hat{c}_2^+ \hat{S} \right),
\]

where the last term in the brackets comprises the annihilation operator \(\hat{S}\) of the biexciton \(XX\) state and the creation operators \(\hat{c}_1^+\) and \(\hat{c}_2^+\) of a pair of plasmons, whose energies differ slightly for the different intermediate levels \(X_+\), \(X_-\) with frequencies \(\omega_{X+}\), \(\omega_{X-}\). The basis states \(|g\rangle_1 = |1S(h), m_s = -1/2\rangle\), \(|g\rangle_2 = |1S(h), m_s = +1/2\rangle\), \(|e\rangle_1 = |1S(e), m_s = +1/2\rangle\), \(|e\rangle_2 = |1S(e), m_s = -1/2\rangle\) for electrons and holes of QDs in Fig. 1 differ in the presence of (c) external magnetic field \(\Delta_{X,XX}\) and \(\Delta_{m}\) of (b) and in the absence of (a) the presence of (b) external magnetic field \(\Delta_{X,XX}\) and \(\Delta_{m}\) of (a) and in the absence of (c) external magnetic field \(\Delta_{X,XX}\) and \(\Delta_{m}\).

\[
\begin{align*}
\hat{c}_1 &= i \left( \frac{\Delta + i}{\tau_c} \right) \hat{c}_1 - i \Omega^{(2)} \hat{c}_2^+ \hat{S} + \hat{F}_{c1}, \\
\hat{c}_2 &= i \left( \frac{\Delta + i}{\tau_c} \right) \hat{c}_2 - i \Omega^{(2)} \hat{c}_1^+ \hat{S} + \hat{F}_{c2}, \\
\hat{S} &= i \left( \delta^{(2)} + i \frac{1}{\tau_S} \right) \hat{S} + \Omega^{(2)} \hat{c}_1 \hat{c}_2 D + \hat{F}_S, \\
\hat{D} &= 2 \Omega^{(2)} \left( \hat{c}_1 \hat{c}_2^+ \hat{S} - \hat{S}^+ \hat{c}_1 \hat{c}_2 \right) - \frac{D}{\tau_D} \hat{D}_0 + \hat{F}_D,
\end{align*}
\]

Figure 1. (a) A model of a three-particle spaser consisting of two NPs and single QD (NP-QD-NP), in which the generation of entangled plasmons is realized due to the QD’s biexciton states decay. The mapping of exciton and plasmon energy levels is in the presence of (c) external magnetic field \(\Delta_{X,XX} = 2kB^2_{m}\).
where the decay rates of different plasmon modes are assumed to be equal each other, i.e. \( \tau_{\text{e1}} = \tau_{\text{e2}} = \tau_{\text{c}} \). We also assume that the pump \( D_0 \) has a rate of \( 1/\tau_D \) for the exciton mode.

The amplitude of plasmons for a stationary solution (2) takes a form:

\[
\bar{c}_i = \frac{e^{i\phi_i}}{2} \sqrt{\frac{\tau_c}{\tau_D}} (D_0 - D).
\]  

These stationary solutions are defined up to the plasmon phase \( \phi_i \). The amplitude of exciton mode takes the form

\[
\bar{S} = -\frac{i\Omega^{(2)}\tau_c}{4B_1}\bar{D} (D_0 - D) e^{i(\phi_1 + \phi_2)}.
\]  

Finally, the corresponding stable solution for population imbalance takes the form:

\[
\bar{D} = \frac{D_0}{2} - \frac{1}{2} D_0^2 + \frac{16\tau_D}{\Omega^{(2)}^2} \frac{\tau_c}{\tau_S} \left( \Delta \delta^{(2)} - \frac{1}{\tau_c \tau_S} \right).
\]  

Initializing our system in the absence of a magnetic field, we assume \( \bar{\omega}_0 = \omega_{XX0}/2 \), where \( \omega_{XX0} = \omega_{XX}|_{D_0=0} \). Then we get \( \delta^{(2)}_0 = 0 \) and the required significant value of detuning is \( \delta = -\Delta_{2b}/2 \) and additional condition \( \Delta = 0 \). Then the frequency of the transition in the QD can be expressed as \( \omega_{XX} = \omega_p + \Delta_{2b}/2 \). We choose \( \lambda_p = 520 \text{ nm} \) for gold and \( \Delta_{2b} = 2.881 \times 10^{14} \text{ s}^{-1} (0.19 \text{ eV}) \) for the CdSe QD [15, 16]. We determine the size of the QD \( D_{QD} = 4.635 \text{ nm} \) to satisfy the conditions of two-quantum transition in the scheme. The dipole moment of corresponding transition in the QD will take new value \( \mu_{QD} = 0.303 \times 10^{-28} \text{ C} \cdot \text{m} \).

The simulation parameters correspond to \( \tau_{\text{e}} = 5 \times 10^{-12} \text{ s}, \tau_{\text{s}} = 4 \times 10^{-11} \text{ s} \) and frequency detuning to \( \delta_0 = -1.441 \times 10^{-14} \text{ s}^{-1} \). The distance between the NP and QD equals \( r = 5 \text{ nm} \). For a NP radius \( a_{NP} = D_{QD}/2 \), the single-plasmon Rabi frequency is equal to \( \Omega_1 = \Omega_2 = \Omega = 1.534 \times 10^{13} \text{ s}^{-1} \), while the two-plasmon Rabi frequency equals \( \Omega^{(2)} = 1.634 \times 10^{12} \text{ s}^{-1} \). These values of the Rabi frequencies approximately correspond to the study [17], where the NP-QD coupling factor is \( 1.516 \times 10^{12} \text{ s}^{-1} \).

The action of the magnetic field on the spaser system occurs at the spin moment of the electron and hole, which leads to a Zeeman splitting of the exciton energy. Then the frequencies of the excitons \( X_+ \) (\( X_- \)) will take the form \( \omega_{X_+} = \omega_{XX} - \alpha B_m (\omega_{X_+} = \omega_{XX0} + \alpha B_m) \), where \( \alpha = g^F \mu_B/\hbar \) [18] and \( g^F \) is the Lande g-Factor, \( \mu_B = 9.27 \times 10^{-24} \text{ J/T} \). The parameter \( g^F \) depends on the QD radius [19]. For very small QDs, this parameter almost coincides with \( g^F = 2 \) for the free electron and decreases to \( g^F_{\text{CdSe}} = 0.68 \) [20] for the CdSe bulk semiconductor.

However, the resulting biexciton frequency \( \omega_{XXX0} \) and the effective detuning \( \delta^{(2)}_0 \) do not change due to Zeeman splitting [18]. This is due to the fact that the Zeeman shifts for \( X_+ \) and \( X_- \) compensate each other. If, however, the diamagnetic shift in the QD is taken into account, then the exciton and biexciton energies take the forms \( \omega_{X_+} = \omega_{XX0} - \alpha B_m + kB_m^2 \) (\( \omega_{X_+} = \omega_{XX0} + \alpha B_m + kB_m^2 \)) and \( \omega_{XXX} = \omega_{XXX0} + 2kB_m^2 \), where \( k = \sqrt{\frac{\mu^*}{4\pi \varepsilon_0}} \). The parameters \( \mu^* = \left( \frac{1}{m_e} + \frac{1}{m_h} \right)^{-1} \) and \( \alpha_{ex} = \sqrt{\frac{1}{2} \left( r_e^2 + r_h^2 \right)} \) are determined by the mass and radius of the exciton [18], where \( r_e \) and \( r_h \) are the effective radiiues of the electron and hole respectively.

Then the corresponding detunings will take the forms \( \delta_{1,2} = \bar{\omega} - \omega_{X_-,X_+} \) and \( \delta^{(2)} = 2\bar{\omega} - \omega_{XXX0} - 2kB_m^2 \), where \( \bar{\omega} \) determines the new spaser frequency in the system, taking into account the magnetic field. Expression for \( \bar{\omega} \) can be obtained in the next form:

\[
\bar{\omega} = \frac{\tau_c \omega_p + \tau_S \omega_{XX}}{\tau_c + 2\tau_S}.
\]
Taking into account the solution (6) and in the presence of magnetic field with magnitude $B_m = 5 \text{ T}$, the spaser frequency becomes equal to the value $\tilde{\omega} = 3.625 \cdot 10^{15} \text{ s}^{-1}$ ($\delta_1 = -1.433 \cdot 10^{14} \text{ s}^{-1}$ and $\delta_2 = -1.448 \cdot 10^{14} \text{ s}^{-1}$, $\Delta = 8.678 \cdot 10^{10} \text{ s}^{-1}$, $\delta^{(2)} = -1.085 \cdot 10^{10} \text{ s}^{-1}$). The frequency detunings $\delta_1$ ($\delta_2$) satisfy to inequality $|\delta_{1,2}| > \Omega_1 \frac{1}{\tau_1}$, $\frac{1}{\tau_2}$ for the nonlinear regime in the spaser. Then the corresponding Rabi frequencies will take the values $\Omega_1^{(2)} = 1.642 \cdot 10^{12} \text{ s}^{-1}$, $\Omega_2^{(2)} = 1.625 \cdot 10^{12} \text{ s}^{-1}$.

In this paper we analyze the dynamics of the parameter

$$G_{12}^{(2)}(t, \tau) = \frac{\langle \hat{c}_1^+ (t) \hat{c}_1 (t) \hat{c}_2^+ (t + \tau) \hat{c}_2 (t + \tau) \rangle}{\langle \hat{c}_1^+ (t) \hat{c}_1 (t) \rangle \langle \hat{c}_2^+ (t + \tau) \hat{c}_2 (t + \tau) \rangle}, \quad (7)$$

which corresponds to the cross-correlation function and is a criterion for establishing correlations between plasmonic modes $\hat{c}_1$ and $\hat{c}_2$. In particular, the condition $G_{12}^{(2)} \equiv G_{12}^{(2)}(t, 0) > 1$ is associated with the intermode plasmon bunching. Moreover, the violation of the Cauchy-Schwarz inequality

$$C' \equiv \frac{G_{12}^{(2)}(t, 0)^2}{g_1^{(2)}(t, 0) g_2^{(2)}(t, 0)} \leq 1$$

indicates the nonclassical character of the correlations between plasmonic modes, where $g_i^{(2)}(t, 0) = G_i^{(2)}(t, 0) + 1$, $i = 1, 2$. This is the necessary condition for the generation of entangled plasmons in the spaser system. In order to study the dynamics of the correlation parameters, we have derived a self-consistent system of equations for bi-linear combinations on the basis of plasmonic and excitonic modes. The value of parameter $G_{12}^{(2)}$ in such stationary conditions reaches the level 3.15, which demonstrates strong intermode bunching between modes $\hat{c}_1$ and $\hat{c}_2$. Moreover, the maximum value of $C'$ parameter is $C_{\text{max}} = 15.15$ and the stationary regime value is $C_{\text{st}} = 1.2$, which demonstrates the nonclassical character of correlations between plasmon modes in spaser system, see inset in Fig. 2. This corresponds to the case when the initial entanglement between plasmons $\hat{c}_1$ and $\hat{c}_2$ is completely absent. Thus, the main result of our simulation is a demonstration of the development of quantum correlations between two localized plasmonic
modes and the possibility for formation of an entangled state during the process of biexciton state decay in a nonlinear NP-QD-NP spaser system.

3. Conclusion
We have proposed a novel mechanism for the control of plasmons’ quantum properties in the spaser, where a pair of NPs is coupled by nonlinear near-field interactions with a single QD (NP-QD-NP spaser).

We have optimized the system parameters providing the stationary regime of the entangled plasmons generation. Further development of this work may be aimed on a complex simulation of nonlinear collective processes [21] with non-classical states of surface plasmon-polaritons (SPP) [22] and SPP waves structures [23].

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