





Conference Paper

The Features of Surface Plasmon-Polariton Pulses Generation Via Cooperative Effects in Waveguide Spaser

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Abstract

The problem of sub-picosecond plasmon-polariton pulse formation in metal/dielectric interface due to collective decay of excited quantum dots, placed in the dielectric layer near the metal surface, is considered. Theoretical approach to selection of semiconductor quantum dots and dielectric host medium to increase the energy transmission of quantum dot collective excitations into surface plasmon-polariton modes of waveguide spaser is developed.

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1. Introduction

Collective processes in a system of quantum emitters for a long time remain a subject of intensive studying [1] with both theoretical and experimental points of view. New opportunities of the well-known cooperative effects in optics can be associated with the collective behavior of the plasmonic oscillators pumped by a near-field of excited chromophores (semiconductor quantum dots, dye molecules, etc. [2]). The kinematics of individual localized systems "quantum dot+metal nanoparticle" [3], the core-shell nanocrystals [4] is well described in the framework of spaser theory [2]. However, generated plasmons in such systems are strongly localized and their collective dynamic is restricted to the near-field area of the plasmonic nanoparticles [5]. Suitable interfaces for observing collective processes with surface plasmon-polariton (SPP) can be planar metal/dielectric waveguides [6] which were already implemented in practice [7]. One of approaches to solving the problem of fast damping of plasmons in such systems is connected with use of photonic crystals as a dielectric layer [8]. In this case, the long-range SPPs with a maximum energy of the field in the dielectric region are formed. On the other hand, compensation of damping of plasmons in metal can be

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realized in the model of the dissipative waveguide spaser with a near-field pumping from the chromophores placed in the dielectric layer near a metal surface.

In this work the approach to choosing specific chromophores and the dielectric host medium to increase the energy transmission efficiency of collective excitations of chromophores to SPPs modes in metal/dielectric waveguide is proposed. Considering that the refractive index of the dielectric host medium is a complex value, we have defined such conditions when the spontaneous emission rate of the chromophores near the metal-dielectric interface [9], as well as the collective optical processes with quantum emitters [10, 11] are almost completely suppressed by the influence of the dielectric environment. Using model of the waveguide spaser the selfconsistent system of the equations describing dynamics of excitons and propagated SPP pulses was obtained. It is shown that in the mean-field approximation the self-consistent problem can be reduced to a modified pendulum equation with an additional term of nonlinear losses. A separatrix solution of the nonlinear equation, which corresponds to the formation of the single SPP pulse in wavequide spaser model, was obtained. A model of the waveguide spaser with an ensemble of CdS quantum dots placed in the dielectric layer near the metal surface for the realization of the predicted effects was proposed.

2. Master equation for collective process of SPP generation in wavegide spaser

Consider the model of an interface in Fig. 1a in the form of a metal/dielectric waveguide [12] with two-level chromophores located inside a thin dielectric layer, the transition frequency between the two levels $\omega_a = 2\pi c/\lambda_a$ being resonant with the SPP frequency $\omega_{SPP} = 2\pi c/\lambda_{SPP}$ (1-ground and 2-upper levels, respectively). By selecting a dielectric medium with appropriate dispersion characteristics and providing the initial excitation (inversion) of a dense ensemble of chromophores in this model, it is possible to produce the collective decay of excitons.

We assume that the characteristic size of the interaction region of the effective field of SPP and chromophores $h = L_x = L_y = L_z$ satisfies the inequality $h \ll \lambda_p$ and the inequality $L_z \ll l_d$ is also valid, where l_d is the SPP decay length along the *z* axis. The corresponding Rabi frequency can be written in the form $\Omega = -(A\nabla\phi\mu_{12}\epsilon)/\hbar$, where $A = \sqrt{\hbar S / \left(\epsilon_0 \epsilon_d \frac{\partial S}{\partial \omega}\right)}$, $\epsilon = \sqrt{N_p}$ is the SPP amplitude, N_p is the number of SPP modes in the interaction region, μ_{12} is the transition dipole moment in a chromophore, and ϕ is the scalar potential of the plasmon field linearly decreasing with distance from the





Figure 1: (a) Formation scheme of SPP pulses in a layered (planar) metal/dielectric waveguide pumped by CdS QDs; (b) dependence of the transition energy on the CdS QD size ($E_g = 2.42 \ eV$ at 0 K for a bulk); (c) parametric plane of the complex refractive index $n = n_R + in_I$ of a dielectric medium with separatrices $\Gamma_e = 0$ for the effective rate of radiative losses of QDs in this medium.

surface, \hbar is the Planck's constant. In the case of excitation of a mode of the plasmon field at frequency ω , using the normalization $\int |\nabla \phi|^2 dV = 1$ [13], the expression for the Rabi frequency can be approximated by the function

$$\Omega = \mu_{12} \sqrt{\frac{S_n}{\hbar \epsilon_d \epsilon_0 V \frac{\partial S_n}{\partial \omega}}} \epsilon = g \epsilon.$$

For a metal-dielectric boundary, the relation

$$\lambda_{SPP} = \sqrt{\frac{Re(\epsilon_m) + Re(\epsilon_d)}{Re(\epsilon_m)Re(\epsilon_d)}} \cdot \lambda_0$$

is valid, where the parameters ϵ_d and $\epsilon_m(\bar{\omega}) = 1 - \omega_p^2 / (\bar{\omega}^2 + i\gamma_s \bar{\omega})$ are the dielectric permittivities of dielectric (with QD) and metal, respectively. Here, $\omega_p = \sqrt{4\pi n_m e^2/m_0}$ is the plasma frequency in a metal, m_0 and n_m are the electron mass and concentration, respectively, γ_s is the electron collision frequency in the metal, $\bar{\omega} = 2\pi c/\lambda_0$. We chose the wavelength $\lambda_0 = 387 \ nm$ for our simulation. The spectral properties of the metal-dielectric interface can be described by use of the Bergman's parameter $S(\omega) = Re(\epsilon_d/(\epsilon_d - \epsilon_m(\omega)))$ [2].

We assume that the pumping volume V' is a dielectric containing QDs with the characteristic radius *a* and concentration $N >> 10^{21} m^{-3}$. Assuming that the refractive index $n = n_R + in_I$ of the dielectric environment of QDs is a complex quantity, where



 $n = \sqrt{\epsilon_d}$ and ϵ_d is the complex permittivity, expressions for the radiative relaxation rate Γ_a , the Rabi frequency Ω , and the effective frequency detuning Δ_a can be written in the form [14]

$$\Gamma_{\epsilon} = \Gamma_{a} \left(n_{R} l_{R} - n_{I} l_{I} + 2 \frac{\delta_{a}}{\Gamma_{a}} l_{I} \right), \tag{1a}$$

$$\Omega_0 = \Omega \cdot \sqrt{l_R^2 + l_I^2},\tag{1b}$$

$$\Delta_{\epsilon} = \delta_a \left(l_R - \frac{\Gamma_a}{2\delta_a} \left(n_I l_R + n_R l_I \right) \right) + \Delta_a, \tag{1C}$$

where $l(n) = l_R + i l_I$ is a complex function for which $l_R = (n_R^2 - n_I^2)/3$, $l_I = 2n_R n_I/3$; and δ_a is a small correction caused by the Lamb shift. It is assumed here that the function $l(n) = E_l/E_M$ coupling the Lorentz local E_l and Maxwell E_M fields will retain its structure in the case of the near field through which SPP are excited in the scheme in Fig. 1.

The parameter $\Gamma_a^* = 1/\tau_R + 1/\tau_F$ is the total rate of radiative (with the time $\tau_R = 1/\Gamma_a$) and nonradiative (with the time τ_F) losses for QDs in vacuum. In the semiclassical approximation, the system can be described similarly to the "metal nanoparticle in a dielectric with chromophores" spaser model [2] with the help of equations for elements of the density matrix ρ of a two-level chromophore:

$$\dot{\rho}_{12} = -\left(i\Delta_{\epsilon} + \frac{\Gamma_{\epsilon}}{2}\right)\rho_{12} + \left(i\Omega_{0}^{*} + i\xi_{0}u_{R}\rho_{12} + \xi_{0}u_{I}\rho_{12}\right)n_{21},$$
(2a)

$$\dot{n}_{21} = 2i \left(\Omega_0 \rho_{12} - \Omega_0^* \rho_{21} \right) - 4\xi_0 u_I \left| \rho_{12} \right|^2 - \Gamma_\epsilon \left(1 + n_{21} \right),$$
(2b)

where $\Delta_a = 2\pi c \left(1/\lambda_a - 1/\lambda_{SPP} \right)$, $n_{21} = \rho_{22} - \rho_{11}$. The Rabi frequency can be written as $\Omega_0 = g\epsilon \cdot \sqrt{l_R^2 + l_I^2}$, where $g = \mu_{12} \sqrt{S_n / \left(\hbar \epsilon_d \epsilon_0 V \frac{\partial S_n}{\partial \omega}\right)}$ is the coupling constant and $\epsilon = A_p \sqrt{\epsilon_d \epsilon_0 V \frac{\partial S_n}{\partial \omega} / (\hbar S_n)}$ is the normalized field with the amplitude A_p of the total field produced by the perturbed electron density in a metal and the electromagnetic field component in a dielectric.

The parameter $\xi_0 = N \mu_{12}^2 / (3\hbar\epsilon_0)$ in (2) determines the addition to the Rabi frequency appearing due to transition from the Maxwell E_M to the local field E_l [14] acting on a chromophore.

The dispersive and dissipative corrections

$$u_{R} = \left(l_{R}\epsilon_{R} + l_{I}\epsilon_{I}\right) / \left(\epsilon_{R}^{2} + \epsilon_{I}^{2}\right),$$



$$u_{I} = \left(l_{I}\epsilon_{R} - l_{R}\epsilon_{I}\right) / \left(\epsilon_{R}^{2} + \epsilon_{I}^{2}\right),$$

respectively, are expressed in terms of the real and imaginary parts of the permittivity of the host-medium [14] in which QDs are placed and have the physical meaning of the additional frequency modulation and the effects of absorption ($u_I < 0$) or amplification $(u_I > 0)$ due to the local field (Fig. 2).



Figure 2: (a) Profiles of SPP pulse amplitude squared obtained by the numerical simulation of system (2)-(3) in the following regimes: (a) neglecting the dissipation parameters $\Gamma_{e} = \Gamma_{a} = \gamma_{p} = 0 \ s^{-1}$; (b) the dissipative regime with $\Gamma_{e} = \Gamma_{a} = 6.3 \times 10^{11} \ s^{-1}$, $\gamma_{p} = 4.1 \times 10^{13} \ s^{-1}$; (c) the regime of QD radiative decay suppression $\Gamma_{e} = 0$ in the host medium with $n_{R} = 1.6$ and $n_{I} = 1.23$; (d) dynamics of the angle θ (solid curve) and coefficients $\cos(\theta)$ (dotted curve) and $-\cos(\theta)$ (dashed curve) for regime (a). The interaction parameters are $g = 8.1 \times 10^{11} \ s^{-1}$, $\xi_{0} = 6.3 \times 10^{9} \ s^{-1}$. The initial polarization of the medium is $\rho_{12}(0) = i\theta_0 = i/\sqrt{N_a} = 0.14i$, the normalization parameter is $\Lambda = 5.7 \times 10^{12} \text{ s}^{-1}$ for $N_a = 50$.

The equation of motion for the Rabi frequency of SPP, which in the case of the exact plasmon resonance has the form

$$\dot{\Omega}_0 = -\frac{i}{t_R^2}\rho_{12} - \gamma_p\Omega_0, \tag{3}$$

where

$$t_{R} = \frac{1}{g\sqrt{N_{a}}} = \sqrt{\frac{\hbar\epsilon_{d}\epsilon_{0}\frac{\partial S_{n}}{\partial\omega}}{S_{n}\mu_{12}^{2}N}}$$



determines the characteristic formation time for quantum correlations in Fig. 1a (compare with the optical problem [15] when emitters are located in the field formation region).

Note that the plasmon mode decay rate $\gamma_p = 1/\tau_J + 1/\tau_R$ is high and determined by the characteristic times τ_R and τ_J of radiative and "joule" losses, respectively. Under conditions $1/\tau_J \approx 30/\tau_R$ [16], radiative losses can be neglected, while "joule" losses are determined by the collision frequency in a metal, i.e., $\gamma_p \approx \gamma_s$, and in problem (3) in the absence of pump, the short-range SPP appear. The self-consistent problem (2)– (3) will be valid only under conditions when the characteristic establishment time t_R for correlations between QDs proves to be considerably shorter than τ_J . Because t_R is inversely proportional to the dipole moment of a chromophore, the relation $t_R < \tau_J$ can be valid for pumping a distributed waveguide spaser by QDs with their giant dipole transition moments.

We use the known dependence [17] of the $1S(e) \rightarrow 1S(h)$ transition energy on the QD diameter $D_{QD} = 2a$ (Fig. 1b) for regime of strong confinement

$$E_{1S(e)-1S(h)} = E_g + 2\frac{\hbar^2 \pi^2}{D_{QD}^2} \left(\frac{1}{m_e} + \frac{1}{m_h}\right),$$
(4)

where *e* is the electron charge, m_e and m_h are the effective electron and hole masses, respectively, in the volume of the QD material with the permittivity ϵ_{qd} and band gap energy E_g [18, 19]. The corresponding parameters for CdS are $m_e = 0.19m_0$, $m_h = 0.8m_0$ and $\epsilon_{qd} = 9$ [20], which gives $D_{QD} = 1.56 \ nm$. Bohr radius of exciton R_{ex} for CdS is 2.5 nm [21] therefore strong confinement regime [22] will be observed for the considered QDs, and energy sublevels of conductivity zone will be essentially separated. The dipole moment of the corresponding interband transition in QDs is assumed equal to $\mu = \mu_{12} = 5 \times 10^{-29} \ C \cdot m$ [2].

For chosen model parameters and the QD concentration $N = 7 \times 10^{21} m^{-3}$, the characteristic correlation time is $t_R = 124 fs$ and the delay time $t_D = 414 fs$ for the number of chromophores in the interaction region $N_a = 50$ (here we assume that $N_a = N_p$). The duration of a formed SPP monopulse is only about 200 fs for ideal conditions $\Gamma_e = \Gamma_a = \gamma_p = 0 s^{-1}$ in Fig. 2a. For dissipative regime in Fig. 2b we take into account the rate of decay $\Gamma_e = 6.3 \times 10^{11} s^{-1}$ for QD near the metal surface [23] and the rate of decay $\gamma_p = 4.1 \times 10^{13} s^{-1}$ for plasmon. In this case we also find the possibility for the formation of SPP pulses, since the characteristic time t_R is shorter than the characteristic decay times in the system.

However, taking (1a) into account, the choice of the appropriate dielectric hostmedium can partially or completely compensate the increase of Γ_a (Fig. 1c), but it



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is also obvious that the properties of natural media are strongly restricted. Thus, for silica at the wavelength under study $\lambda_{SPP} = 192 \ nm$, we have $n_R = 1.6$, $n_I = 5 \times 10^{-7}$ [24] and $\Gamma_e = 2.43\Gamma_a$. To completely compensate relaxation processes in (2a) ($\Gamma_e \equiv 0$), the required combination of dispersion-dissipative parameters should satisfy the condition $n_R l_R - n_I l_I = 0$ (be neglecting a small Lamb shift), which is satisfied, for example, for the choice $n_R = 1.6$ and $n_I = 1.23$. Such conditions can be fulfilled for an artificial microstructured dielectric material with specified dispersion- dissipative characteristics (the Cole-Cole diagram). They lead to the significant increase in the SPP pulse intensity, while energy transfer from chromophores to radiation proves to be suppressed (see Fig. 2c). In this case, the influence of the local field increases, the absolute values of its parameters increase (corrections $u_R = 0.37$ and $u_I = -0.158$ in (2)) and the formation dynamics of SPP pulses changes.

3. Collective dynamics of a waveguide spaser in the mean field approximation

To analyze the contribution of dissipative effects related to the imaginary part u_I of the local field correction, we can neglect the corresponding phase effects with u_R in (2) and decay in (2)–(3) and to pass in the mean field approximation to a simplified system of self-consistent equations for a medium

$$\dot{\rho}_{12} = \left(i\Omega_0^* + \xi_0 u_I \rho_{12}\right) n_{21},\tag{5a}$$

$$\dot{n}_{21} = 2i \left(\Omega_0 \rho_{12} - \Omega_0^* \rho_{21}\right) - 4\xi_0 u_I \left|\rho_{12}\right|^2$$
(5b)

and the effective field

$$\dot{\Omega}_0 = -ig^2 N_a \rho_{12} \tag{6}$$

formed in it.

By passing to the representation for the Rabi frequency and polarization in the form

$$\Omega_0 = \frac{1}{2} \left(U e^{-iK_0 t} + U^* e^{iK_0 t} \right), \ \rho_{12} = \frac{1}{2} R \cdot e^{-iK_0}$$

where $K_0 = \omega_{SPP}t - k_{SPP}z$, and assuming that $Z = n_{21}$, we can obtain the system of Maxwell-Bloch equations for a spaser taking into account the (dissipative) local response of the QD environment

$$\dot{Z} = -\frac{i}{2} \left(UR^* - U^*R \right) - \xi_0 u_I |R|^2,$$
(7a)



$$\dot{R} = i \left(U - i\xi_0 u_I R \right) Z, \tag{7b}$$

$$\dot{U} = -ig^2 N_a R. \tag{7C}$$

By passing to new dimensionless variables $\delta_0 = -i\frac{U}{\Lambda}$ and $\tau = t \cdot \Lambda$, where $\Lambda = g\sqrt{N_a}$ and setting $R^* = R$ and $\delta^* = \delta$, we represent system (7) in the form

$$\frac{\partial Z}{\partial \tau} = \delta_0 R - \frac{\xi_0 u_I}{\Lambda} \left| R \right|^2, \tag{8a}$$

$$\frac{\partial R}{\partial \tau} = -\delta_0 Z + \frac{\xi_0 u_I}{\Lambda} R Z, \tag{8b}$$

$$\frac{\partial \delta_0}{\partial \tau} = -R. \tag{8c}$$

The solution of system (8) can be written in the form $Z = \cos(\theta)$ and $R = \sin(\theta)$, where θ determines the angle of the so-called Bloch vector with coordinates Z and R and their substitution to (8) gives the equation for the angle

$$\dot{\theta} = -\delta_0 + \frac{\xi_0 u_I}{\Lambda} \sin\left(\theta\right).$$
(9)

By substituting the expression for δ_0 from (9) into (8c), we obtain a new variant of the pendulum equation with the nonlinear harmonic losses/decay term

$$\ddot{\theta} - K\cos(\theta) \cdot \dot{\theta} = \sin(\theta),$$
 (10)

where the amplitude of the decay coefficient is defined as $K = \xi_0 u_I / \Lambda$. In the absence of the loss modulation, when $K \cos(\theta) \cdot \dot{\theta} = K \cdot \dot{\theta}$, Eq. (10) is reduced to the usual nonlinear pendulum equation with losses [25]. Taking the modulation into account under the same conditions K < 0 ($\epsilon_I > 0$ and $u_I < 0$), the pendulum experiences the additional decay in intervals

$$\theta \in \left[0 + 2\pi m; \frac{\pi}{2} + 2\pi m\right], \ \theta \in \left[\frac{3\pi}{2} + 2\pi m; 2\pi + 2\pi m\right]$$

responsible for the formation of the leading and trailing edges of SPP pulse (see Fig. 2c), whereas in the interval

$$\theta \in \left[\frac{\pi}{2} + 2\pi m; \frac{3\pi}{2} + 2\pi m\right]$$

when the central part of SPP pulse is formed, the enhancement of pendulum oscillations is observed; m = 0, 1, 2...

In other words, the absorbing dielectric host-medium coherently preserves a part of the QD energy during the formation of the leading edge of the pulse and then



coherently returns this energy to SPP pulse during formation of the pulse peak. As a result, taking into account the compensation of the spontaneous relaxation rate of QDs ($\Gamma_{\epsilon} = 0$) and nonlinear terms with u_I in (5), the increase in the peak pulse intensity is observed with respect to the case when the response of the host-medium is neglected (see Figs. 2a,b).

The initial conditions in simulation of (10) are chosen equal to $\theta_0 = 1/\sqrt{N_a}$ for the initial oscillation angle and

$$v_{\theta} = \left. \frac{\partial \theta}{\partial t} \right|_{t=0} = \frac{2}{\cosh\left(\ln \frac{\theta_0}{4}\right)}$$

for the initial velocity of the pendulum.

Equation (10) is a particular case of the Lienard equation and its approximate analytic solution can be expressed in terms of elliptic integrals of the first kind. The numerical solution for the Rabi frequency of SPP pulse field obtained from (10) completely coincides with the results of the direct numerical simulation of system (5)–(6) under conditions of the suppression of spontaneous relaxation in QDs for the chosen values $n_R = 1.6$ and $n_I = 1.23$ (K = -0.0147) (see Fig. 2a).

4. Conclusions

We have proposed efficient method for the formation of short SPP pulses at the dielectric-metal interface containing QDs. The general conditions for selecting parameters of QDs and a dielectric host-medium are determined which provide the maximal collective energy transfer from a QD ensemble to SPP modes dominating over the radiative relaxation of individual chromophores. To tune the system parameters to the plasmon resonance more accurately, it is useful to employ experimental absorption and fluorescence spectra of giant ensembles of emitters [26, 27] in different host medium. The models presented in the paper can be useful for practical implementation of multiqubits entanglements [28] and quantum computations in macroscopic and mesoscopic [29] systems. However, for the realization of the external control in such systems one additionally requires the use of multiwave schemes [30, 31] of nonlinear coherent interaction by analogy with optics [32, 33]. Further development of our research is related to the investigation of collective spin effects based on the photon echo [34] in plasmonic structures, as well as the possibilities of control such effects [35].



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