Formation of Plasmon-Polariton Pulses in the Cooperative Decay of Excitons of Quantum Dots Near a Metal Surface

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The formation of pulses of surface electromagnetic waves in a metal/dielectric interface is considered in the process of cooperative decay of excitons of quantum dots distributed near a metal surface in a dielectric layer. It is shown that the efficiency of exciton energy transfer to excited plasmons can be increased by selecting the dielectric material with specified values of the complex permittivity. It is found that in the mean field approximation the semiclassical model of formation of plasmon pulses in the system under study is reduced to the pendulum equation with the additional term of nonlinear losses.

I. INTRODUCTION

Collective energy emission processes by a system of quantum emitters such as optical superradiation have long been studied, both theoretically and experimentally [1–4]. The new possibilities of known effects can be related to the collective behavior of plasmon oscillators pumped by the near field of excited chromophores such as semiconductor quantum dots (QDs), dye molecules, etc. [5]. In the case of localized quantum dot + metal nanoparticle systems [6] or individual combined core-shell nanocrystals [7], their kinematics is well described by the spaser theory [5]. However, plasmons formed in this case are strongly localized and their collective dynamics is restricted by the region of action of the near field of plasmon nanoparticles [8].

When a system is extended to the case of a 1-D array (chain) of localized spasers, the appearing collective nonlinear regimes can lead to the considerable narrowing down of emission lines and the simultaneous compensation of optical losses [9]. In the case of 2-D arrays of localized spasers such as an ensemble of QDs near a metal surface with defects, there is a region of the collective behavior of the system caused by the self-synchronization of individual chromophores due to the near-field interaction between them [10]. As a variant, chromophores can be synchronized by an external pump beam, which enhances the efficiency of induced processes in the system under study, resulting in the formation of a narrow coherent optical beam [11] perpendicular to the metal surface. As the external pump, the near field of the tip of a scanning tunneling microscope can be used [12].

Significant interest is the alternative possibility related to the coherent amplification of the near field of propagating surface plasmon-polaritons (SPP) due to collective effects with chromophores under conditions of the partial or complete suppression of processes of their radiative relaxation. The problem of the propagation of a plasmon field appears, in particular, in 1-D systems such as a metal groove [13] or a pyramid [14, 15] with nearby QDs and is solved by analyzing Maxwell Bloch equations. However, when the decay rate γp of plasmons in a metal is significant, the development of collective coherent processes involving SPP is much less efficient than emission processes in optical modes, in particular, in the superradiation mode.

At the same time, as convenient interfaces for observing coherent processes with SPP, planar metal/dielectric waveguides already realized in practice can be used, in which the transverse focusing of plasmon modes is performed by analogs of Bragg mirrors [16]. The solution of the problem of plasmon decay in such systems can be related to the use of photonic crystals as a dielectric layer [17] when long-range SPP are formed in the system with the field energy maximum considerably shifted to the dielectric region.

Another way for compensating plasmon decay in a metal can be the model of a waveguide spaser with near-field pumping from chromophores located near a metal surface [18]. The processes of interaction of chromophores with the effective field of a plasmon-polariton wave for such a scheme are described in detail in [19, 20] by the example of solving problems on the self-induced transparency and formation of dissipative solitons for plasmon-polariton pulses. The author of [21] proposed to realize such a scheme of a distributed spaser using a dielectric metamaterial film doped with QDs. However, it is necessary to take into account that the efficiency of the exciton energy transfer to a plasmon mode strongly depends on the ratio r/λp, where r is the distance from a chromophore to the metal surface and λp is the wavelength of a generated plasmon [22]. At the same time, for r/λp ≪ 1, the rate Γa of the spontaneous radiative decay of the chromophore tends to the limiting value Γa = (2/3) Γ0, where Γ0 is the rate of the radiative decay in vacuum. Then, under the condition Γa ≪ γp , the rate of the radiative decay in a QD could be neglected for this problem. However, in the presence of a dense (about 1015 cm−3) ensemble of adjacent excited chromophores, Γa can significantly increase [23, 24] in the initial stage of the system evolution due to dipole-dipole
interactions. This can lead to the undesirable transfer of a part of chromophore energy to optical modes and can initiate the development of cooperative optical effects, including superradiation \[8, 23\]. Thus, the partial or complete suppression of relaxation processes determined by the radiative decay rate \(a\) of excitons in QDs becomes the additional necessary condition for the observation of collective processes involving surface plasmons.

In this paper, we propose an approach for selecting particular chromophores and an appropriate dielectric host-medium to increase the efficiency of energy transfer from collective excitations of chromophores to SPP modes in a planar metal/dielectric waveguide. The condition for observing the process is a considerable decrease in the effective value of \(\Gamma_a\), which can be caused by local field effects \[26\] appearing upon the disposition of a dense ensemble of chromophores in a specially selected dielectric matrix. Our model assumes that the permittivity of the dielectric host-medium is complex, which allows us to completely compensate the spontaneous re-
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dielectric matrix. Our model assumes that the permit-

\[\begin{aligned}
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\end{aligned}\]

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electric (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, \(\gamma_s\) is the collision frequency in the metal, \(\tilde{\omega} = 2\pi c/\lambda_0\). The spectral properties of the metal-dielectric interface can be described by use of the Bergman’s parameter \(S(\omega) = \text{Re}(\varepsilon_d/\varepsilon_m(\omega))\) \[3\].

We assume that the pumping volume \(V'\) is a dielec-
tric containing QDs with the characteristic radius \(a\) and concentration \(N \gg 10^{21}\) \(\text{m}^{-3}\). The condition \(\lambda_0 > a\) allows us to remain within the dipole approximation, but the large value of dipole transition moments of QDs \[31\] requires the consideration of a local field acting on emitters \[27, 28\]. At the same time, the effects of exchange dipole-dipole interaction \[32\] between individual QDs, which are important in the superradiation problem of a localized spaser \[12\], are neglected in our problem.

Assuming that the refractive index \(n = n_R + in_I\) of the dielectric environment of QDs is a complex quantity, where \(n = \sqrt{\varepsilon_d}\) and \(\varepsilon_d\) is the complex permittivity, expressions for the radiative relaxation rate \(\Gamma_a\), the Rabi frequency \(\Omega\), and the effective frequency detuning \(\Delta_a\) can be written in the form \[33\]

\[\begin{aligned}
\Gamma_a &\approx \Gamma_a \left(n_Rl_R - n_Il_I + 2\frac{\delta a}{\Gamma a}\right), \\
\Omega &\approx \Omega \sqrt{l_R^2 + l_I^2}, \\
\Delta_a &\approx \delta a \left(l_R - \frac{\Gamma a}{2\delta a} (n_Il_I + n_Rl_R)\right) + \Delta_a,
\end{aligned}\]

in the region \(V'\) in the metal causes induced processes in chromophores located in the symmetric region \(V = h^3\) in the dielectric. Then, assuming that SPP modes are quasistatic within the volume under study \[30\], the corresponding Rabi frequencies can be written in the form

\[\begin{aligned}
\Omega = -\left(\frac{\partial \varphi}{\partial t}\right)_{\mu_2} \frac{\partial \varphi}{\partial n_0} / \hbar, \\
\text{where} A = \sqrt{hS/\varepsilon_0 d}, \quad a_0 \text{ is the plasmon amplitude,} \\
\mu_2 \text{ is the transition dipole moment in a chromophore,} \\
\phi \text{ is the scalar potential of the plasmon field linearly decreasing with distance from} \\
\text{the surface,} \quad \hbar \text{ is the Planck’s constant. In the case of} \\
\text{excitation of a mode of the plasmon field at frequency} \omega, \\
\text{using the normalization} \int |\nabla \varphi|^2 \, dV = 1 \[30\], \text{the expression} \\
\text{for the Rabi frequency can be approximated by the function}
\end{aligned}\]

\[\begin{aligned}
\Omega &\approx \mu_2 \sqrt{\frac{S_n}{\varepsilon_0 d^2}} \varepsilon = \gamma_s', \\
\varepsilon &\approx \sqrt{\varepsilon_0} \text{ and} \quad \varepsilon &\approx \sqrt{\varepsilon_0} \text{ and} \\
N_0 &\text{ is the number of plasmons in} \\
\text{the interaction region.}
\end{aligned}\]

For a metal-dielectric boundary, the relation

\[\begin{aligned}
\omega &\approx \omega_0 \left(\frac{\varepsilon_m}{\varepsilon_d}\right), \\
\text{is valid, where the parameters} \varepsilon_d \text{ and} \varepsilon_m(\tilde{\omega}) \text{ are the dielectric permittivities of} \\
\text{dielectric (with QD) and metal, respectively. Here,} \omega_0 = \sqrt{4\pi n_m e^2/m_0}\text{ is the plasma frequency in a metal,} \\
\text{and}\gamma_s \text{ is the collision frequency in the metal,} \\
\tilde{\omega} = 2\pi c/\lambda_0 \text{. The spectral properties of the metal-dielectric interface can be described by use of the Bergman’s parameter} \\
\text{S(}\omega\text{) = Re(}\varepsilon_d/(\varepsilon_m(\tilde{\omega}))) \text{[3].}
\end{aligned}\]

Consider the model of an interface in Fig. 1a in the form of a metal/dielectric waveguide \[29\] with two-level chromophores located inside a thin dielectric layer, the transition frequency between the two levels \(\omega_0 = 2\pi c/\lambda_a\) being resonant with the plasmon frequency \(\omega_{SPP} = 2\pi c/\lambda_{SPP}\) of the metal. 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. The difference of this situation from the model of an emitting spaser \[11\] is that the dipole moments of chromophores are oriented in the direction perpendicular to the waveguide plane, which leads to the coherent transfer of their energy predominantly to SPP modes propagating along the axis. In this case, the process can be localized in the \(y\) direction using the system of additional waveguides operating based on the Bragg reflection of SPP (antiresonant-reflecting optical waveguide, ARROW \[16\]).

By considering the problem in the 3-D approximation, we assume that the characteristic size of the interaction region of the effective field of plasmons 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 plasmon decay length along the \(z\) axis. In this case, the time-dependent perturbation of the electron density appearing

\[\begin{aligned}
\Gamma &\approx \Gamma_a \left(n_Rl_R - n_Il_I + 2\frac{\delta a}{\Gamma a}\right), \\
\Omega &\approx \Omega \sqrt{l_R^2 + l_I^2}, \\
\Delta &\approx \delta a \left(l_R - \frac{\Gamma a}{2\delta a} (n_Il_I + n_Rl_R)\right) + \Delta_a,
\end{aligned}\]
where \( l(n) = l_R + il_I \) is a complex function for which \( l_R = \left( n_R^2 - n_I^2 \right) / 3, l_I = 2 n_R n_I / 3 \); and \( \delta_a \) 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 plasmons 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 \( \tau_F \)) losses for QDs in vacuum. By using annealing technology [34], the time \( \tau_F \) can be increased to values comparable to the radiative time [35]. At the same time, when a dense (more than \( 10^{17} \text{ cm}^{-3} \)) ensemble of chromophores is located near the metal boundary, the spontaneous emission rate can considerably change and, in particular, increase [22, 24]. Note that the problem of temporal stability of single QDs during collective energy transfer to SPP in the configuration in Fig. 1 remains open, similarly to the blinking problem of emitting QDs [36].

In the semiclassical approximation, the system can be described similarly to the metal nanoparticle in a dielectric with chromophores spaser model [5] with the help of equations for elements of the density matrix \( \rho \) of a two-level chromophore:

\[
\dot{\rho}_{12} = - (i \Delta + \frac{\Gamma}{2} ) \rho_{12} + (i \Omega_{0} + i \varepsilon_0 u_R \rho_{12} + \varepsilon_0 u_I \rho_{12}) n_{21},
\]

\[
\dot{n}_{21} = 2i \left( \Omega_{01} \rho_{12} - \Omega_{02} \rho_{21} \right) - 4 \varepsilon_0 u_I |\rho_{12}|^2 - \Gamma_e (1 + n_{21}),
\]

where \( \Delta = 2 \pi c (1 / \lambda_a - 1 / \lambda_{SP}) \), \( n_{21} = \rho_{22} - \rho_{11} \). The Rabi frequency can be written as \( \Omega_0 = g \varepsilon \cdot \sqrt{I_R + I_l} \), where \( g = \mu_{12} \sqrt{S_n / (\hbar \varepsilon_0 V \partial S_{\varepsilon_0} / \partial \varepsilon_0)} \) is the coupling constant and \( \varepsilon = A_p \sqrt{\varepsilon_0 \partial S_{\varepsilon_0} / (\hbar S_n)} \) 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. In the general case, the relation between these components can be found only during the simultaneous solution of the evolution equation for the electron density in a conductor with the specified geometry and Maxwell’s equation [20].

The parameter \( \varepsilon_0 = N \mu_{12}^2 / (\hbar \varepsilon_0) \) in [2] determines the addition to the Rabi frequency appearing due to transition from the Maxwell \( E_M \) to the local field \( E_l \) acting on a chromophore.

The dispersion and dissipative corrections \( u_R = (l_R \varepsilon_R + l_I \varepsilon_I) / (\varepsilon_R^2 + \varepsilon_I^2) \) and \( u_I = (l_R \varepsilon_R - l_I \varepsilon_I) / (\varepsilon_R^2 + \varepsilon_I^2) \).
The characteristic formation time for quantum correlations in the chosen volume $V'$ in Fig. 1a (compare with the optical problem) 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 $\tau_R$ and $\tau_J$ of radiative and joule losses, respectively. Under conditions $1/\tau_J \approx 30/\tau_R$, radiative losses can be neglected, while joule losses are determined by the collision frequency in a metal, i.e., $\gamma_p = \gamma_S$, and in problem (3) in the absence of pump, the shortrange SPP appear.

In the presence of the maximum of a surface wave energy in the metal, the self-consistent problem (2)–(3) will be valid only under conditions when the characteristic establishment time $t_R$ for correlations between plasmons proves to be considerably shorter than $\tau_J$. 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.

In a simple case $\xi_0 = 0$ and in the absence of external excitation $\varepsilon_{ex}(t) = 0$, the collective behavior of the system, in particular, excitation of superradiation, as in an optical scheme, is stimulated by a small initial polarization $\rho_{12}(0) = \text{Im}(\rho_{12}(0)) \neq 0$ of the system initiating the growth of the real part of the Rabi frequency according to the relation $\rho_{12}(0) = \tau_R / \rho_{12}(0)$ (see 3). Thus, the front wing of a surface electromagnetic pulse (SPP pulse) will be formed. Then, according to $\rho_{12}(t) = \rho_{12}(0) n_{21}$ from (2) and under the condition $n_{21} > 0$, the growth of $\rho_{12}(t)$ is observed (Fig. 2a), which leads to the excited-level decay according to

$$n_{21} = -4 \left( \rho_{12}(0) \rho_{12}(t) + \rho_{12}(0) \rho_{12}(t) \right) \approx -4 \rho_{12}(0) \rho_{12}(t)$$

from (2b). The process will also continue with the beginning of saturation in the system. However, because of the change in the sign of the population difference $n_{21}$, the parameter $\rho_{12}(t)$ gradually decreases to zero. The condition $\rho_{12}(t_D) = 0$ is fulfilled at characteristic times $t_D = t_R \ln (4/\theta_0) \approx \tau_R (\text{where } \theta_0 = 1/\sqrt{N_a} \text{ and } N_a \text{ is the number of chromophores in the interaction region (3)})$ when a plasmon pulse is formed. Then this process is repeated but already in the region where the parameter $\rho_{12}(t)$ is negative, which leads to the formation of the rear edge of the pulse (Fig. 2). A similar picture is also observed in the case of a low initial stochastic coherence of the system $\rho_{21}(0) = \text{Re}(\rho_{21}(0))$ in (3).

The use of QDs for pumping with their giant dipole moments at the operating transition can result in a significant shortening of the establishment time for quantum correlations and in the proportional decrease in the

\[
\Omega_0 = -\frac{i}{t_R} \rho_{12} - \gamma_p \Omega_0, \tag{3}
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delay time $t_D$ and duration $t_W$ of SPP pulses generated in the system. As a model medium, we use CdS QDs located in a dielectric film near the gold surface. Taking into account the plasmon frequency of gold $\omega_p = 1.37 \times 10^{16}$ s$^{-1}$ and choosing the condition $\lambda_0 = 387$ nm, the wavelength of generated SPP is $\lambda_{SPP} = 192$ nm. To determine the QD size in the case of the exact resonance $\Delta_\varepsilon = 0$, we use the known dependence $E_{1S(e)} = E_{1S(h)} = E_0 + 2 \frac{\hbar k^2 \pi^2}{D_{QD}^2} \left( \frac{1}{m_e} + \frac{1}{m_h} \right) - \frac{3.56 \cdot e^2}{\varepsilon_{qd} \cdot D_{QD}}$, \hspace{1cm} (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 $\varepsilon_{qd}$ and band gap energy $E_g$. The corresponding parameters for CdS are $m_e = 0.19m_0$, $m_h = 0.8m_0$ and $\varepsilon_{qd} = 9$, which gives $D_{QD} = 1.56$ nm. Bohr radius of exciton $R_{ex}$ for CdS is 2.5 nm therefore strong confinement regime will be observed for the considered QDs, and energy sublevels of conductivity zone will be essentially separated. To tune the QD size to the plasmon resonance more accurately, it is useful to employ experimental curves $E_{1S(e)} - 1S(h)$ for particular synthesized QDs. The dipole moment of the corresponding interband transition in QDs is assumed equal to $\mu = 5 \times 10^{-20}$ C · m.

For chosen model parameters and the QD concentration $N = 10^{24}$ m$^{-3}$, the characteristic correlation time is $t_R = 10$ fs and the delay time $t_D = 60$ fs for the number of chromophores in the interaction region $N_a = 7 \times 10^3$. The duration of a formed SPP monopulse is only about 14 fs; for regime 1 in Fig. 2. taking into account the uncompensated rate of radiative losses, $\Gamma_e = \Gamma_a = 6.3 \times 10^{11}$ s$^{-1}$ for QDs near the metal surface. The additional consideration of the decay rate of plasmons in gold even under the condition $\gamma_p = 4.1 \times 10^{13}$ s$^{-1}$ does not strongly affect on the development of the formation dynamics of the plasmonic pulse (regime 3 in Fig. 2).

However, taking (2) into account, the choice of the appropriate dielectric host-medium can partially or completely compensate the increase of $\Gamma_a$ (Fig. 1b), but it 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 $n_R = 1.6$, $n_I = 5 \times 10^{-7}$ and $\Gamma_e = 2.43\Gamma_a$. To completely compensate relaxation processes in (2) (\Gamma_e \equiv 0), the required combination of dispersion-dissipative parameters should satisfy the condition $n_R m_R - n_I m_I = 0$ (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 regime 2 in Fig. 2). 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.

III. COLLECTIVE DYNAMICS OF A WAVEGUIDE SPASER IN THE MEAN FIELD APPROXIMATION

To analyze the contribution of dissipative effects related to the imaginary part $\mu_I$ 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

$$\dot{\rho}_{12} = (i\Omega_0 + \xi_0 u_I \rho_{12}) n_{21},$$ \hspace{1cm} (5a)

$$\dot{n}_{21} = 2i(\Omega_0 \rho_{12} - \Omega_0^* \rho_{21}) - 4\xi_0 u_I |\rho_{12}|^2,$$ \hspace{1cm} (5b)

and the effective field

$$\hat{\rho}_0 = -i\hat{g}^2 N_a \rho_{12}$$ \hspace{1cm} (6)

formed in it.

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

$$\Omega_0 = \frac{1}{2}(U e^{-i\phi} + U^* e^{i\phi}), \hspace{1cm} \rho_{12} = \frac{1}{2} R e^{-iK_0}$$

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

$$\dot{Z} = -\frac{i}{2} (UR^* - U^* R) Z - \xi_0 u_I |R|^2,$$ \hspace{1cm} (7a)

$$\dot{R} = i (U - \xi_0 u_I R) Z,$$ \hspace{1cm} (7b)

$$\dot{U} = -i\hat{g}^2 N_a R.$$ \hspace{1cm} (7c)

The system of equations (7) was derived using the rotating wave approximation by neglecting high-frequency terms with phase factors $e^{\pm 2i\omega_0 t}$. By passing to new dimensionless variables $\delta_0 = \frac{iK_0}{\omega_0}$ 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

$$\frac{\partial Z}{\partial \tau} = \delta_0 R + \frac{\xi_0 u_I}{\Lambda} |R|^2,$$ \hspace{1cm} (8a)

$$\frac{\partial R}{\partial \tau} = -\delta_0 Z + \frac{\xi_0 u_I}{\Lambda} R Z,$$ \hspace{1cm} (8b)

$$\frac{\partial \delta_0}{\partial \tau} = -R.$$ \hspace{1cm} (8c)

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

\[
\dot{\theta} = -\delta_0 + \frac{\xi_0 u_I}{\Lambda} B \sin(\theta).
\] (9)

By substituting the expression for \( \delta_0 \) from (9) into (8), we obtain a new variant of the pendulum equation with the nonlinear harmonic losses/decay term

\[
\ddot{\theta} - \frac{\xi_0 u_I}{\Lambda} B \cos(\theta) \cdot \dot{\theta} = B \sin(\theta).
\] (10)

The second term in the left-hand side of (10) is responsible for processes initiated by the local response of the medium and synchronized with the change in the angle \( \theta \). By using the separatrix condition \( B = 1 \) corresponding to the passage from the rotational motion of the pendulum to vibrational, Eq. (10) can be written in the form

\[
\ddot{\theta} - \frac{\xi_0 u_I}{\Lambda} \cos(\theta) \cdot \dot{\theta} = \sin(\theta).
\] (11)

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. (11) is reduced to the usual nonlinear pendulum equation with losses (8). Taking the modulation into account under the same conditions \( K < 0 \) \( (\xi_I > 0 \) and \( u_I < 0 \)), the pendulum experiences the additional decay in intervals

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

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

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

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

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 \((\Gamma_e = 0)\) 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 Fig. 2a).

It seems that in the presence of strong resonator effects in the dielectric host-medium near the QD resonant wavelength, the peak intensity of generated pulses can be additionally increased due to terms with \( u_R \) and \( u_I \) in system (2). But because these corrections are obtained assuming that the spacing wavelength \( \lambda_{SPP} \) lies at the wing of the absorption line of a dielectric film [33], this theory cannot be applied to the given case. Nevertheless, such amplification can be achieved, for example, using dielectric metamaterials [48] with a specially selected geometry doped with QDs [49, 51]. In this case, it is possible to excite longrange surface plasmon polaritons [17] at a metal/(QDdoped metamaterial) boundary with their simultaneous amplification due to QD pumping. However, problem (2)–(3) becomes considerably complicated in this case, because of the necessity of describing the field components in a dielectric, the consideration of the geometry of individual scattering centers [52] and the influence of the inhomogeneous structured microenvironment on the spontaneous decay rate in QDs [53].

For the case \( u_I > 0 \) \((K > 0)\) [33], when a host-medium with the background amplification exists, on the contrary, energy transfer from the medium to emitters doped into it should occur in the initial stage of the plasmon pulse formation. This causes the advance of the pulse generation in the medium and a partial loss in the pulse intensity compared to the case \( u_I = 0 \).

In the known case \( \xi_0 = 0 \), the separatrix solution of Eq. (11) has the form \( \theta = 4 \arctan (e^{-t_D}) \), where the dimensionless delay time is \( t_D = \ln (4/\theta_0) \) with the initial angle \( \theta_0 = 1/\sqrt{N_a} \). This corresponds to the formation of a monopulse with the Rabi frequency modulus squared that can be written in the form

\[
|\Omega_0|^2 = \frac{\Lambda^2}{|\cosh (\Lambda (t - t_D))|^2}
\] (12)

at the real time scale (regime 1 in Fig. 2b).

To simulate Eqs. (3)–(11), we considered a particular regime assuming that the plasmon phase is \( \varphi(t) = -\pi/2 \). In this case, as considered in Section I the initial polarization of the system is only imaginary \( \rho_1(0) = iR/2 \) and the Rabi frequency can be written as \( \Omega_0 = -U^{(j)} \), where the expansion \( U = U^{(R)} + iU^{(I)} \) is used. Under such conditions, the only real part of the Rabi frequency of the pulse is formed and system (3) is transformed to the system

\[
\frac{\partial Z}{\partial \tau} = 2i\delta_0 R - \frac{\xi_0 u_I}{\Lambda} |R|^2,
\] (13a)

\[
\frac{\partial R}{\partial \tau} = -2i\delta_0 Z + \frac{\xi_0 u_I}{\Lambda} RZ,
\] (13b)

\[
\frac{\partial \delta_0}{\partial \tau} = \frac{i}{2} R.
\] (13c)

However, the form of Eq. (11), to which (13) is reduced, does not change under the new normalization condition \( \delta_0 = -iU^{(I)}/\Lambda \). The initial conditions in simulation of (11) are chosen equal to \( \theta_0 = 1/\sqrt{N_a} \) for the initial oscillation angle and

\[
u_0 = \frac{\partial \theta}{\partial t} \bigg|_{t=0} = \frac{2}{\cosh \left( \ln \frac{4}{\theta_0} \right)}
\]

for the initial velocity of the pendulum.

Equation (11) 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 (11) 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 regime 2 in Fig. 2a).

IV. INFLUENCE OF CONCENTRATION AND DISSIPATIVE EFFECTS ON THE DEVELOPMENT OF COLLECTIVE REGIMES OF A WAVEGUIDE SPASER

The solutions of Eq. (11) are obtained under conditions of the suppression of spontaneous emission of excited QDs near a metal/dielectric boundary, whereas the violation of relations obtained for parameters \( n_R \) and \( n_I \) should lead to the increase in the rate of relaxation processes and the weakening of SPP pulses. In this case, the efficiency of the QD energy transfer to the superradiation mode nonlinearly depends on the parameter \( \delta_{\text{R}} = n_I/n_R \) characterizing the relative deviation of the loss coefficient of the host-medium from the specified level \( n_R \) for which the condition \( \Gamma_\varepsilon = 0 \) is exactly fulfilled for fixed \( n_R \) (see Fig. 3b). In particular, the decrease in \( n_I \) leads to the increase in the relaxation rate \( \Gamma_\varepsilon \) and the related decrease in the area

\[
S = \frac{\mu}{k} \int_0^{2t_D} A_\rho dt = \int_0^{2t_D} |\Omega_0| dt \quad \text{(rad)}
\]

of SPP pulses presented in Fig. 3, and calculated by direct numerical simulation of system (2)–(3). On the other hand, the change in the QD concentration under the condition \( \Gamma_\varepsilon = 0 \) in the system, allows one to control the delay time \( t_D \) and pulse duration \( t_{\text{sup}} \) at the medium output. Thus, the increase in the QD concentration for CdS leads to the rapid nonlinear shortening of the delay time \( t_D \) in the model under study (Fig. 3c) with the emission more and more intense pulses (Fig. 3b).

To analyze nonlinear phase effects during the generation of plasmon pulses, it is necessary to take into account the spatial dynamics of collective processes by introducing the longitudinal coordinate \( x \) into Eq. (3). The corresponding solution at the output of a medium of length \( L \) with the isotropic distributions of chromophores will have the form

\[
\varepsilon'(L,t) = \varepsilon_{\text{ex}}(t) \cdot \exp(ikL) + i \frac{1}{\tau_R g \varepsilon_0 L} \int_0^L \rho_{21} \cdot \exp(ik|L-x'|)dx', \quad (14)
\]

where \( \tau_R = t_R^2 \frac{g}{L} \) is determined by the new characteristic formation time of quantum correlations, \( \varepsilon_{\text{ex}}(t) \) is the amplitude of an additional trigger SPP pulse at the entrance of waveguide, and the factor \( \exp(ik|L-x'|) \) gives phase shifts which, unlike the case considered in Section III, depend on the coordinate \( x \).

It is convenient to study the influence of nonlinear dispersion effects on the spectral features of SPP pulses beginning from the numerical solution of the joint system of equations (2) and (14) under conditions \( \varepsilon_{\text{ex}}(t) = 0 \) when a trigger pulse at the medium input is absent and also by neglecting dissipative terms of the local field \( u_L = 0 \).

The real spectral shape of pulses generated in such approximation is determined by the inverse Fourier transform from the corresponding Rabi frequency at the medium output

\[
F(L,\omega) = |\Omega_0(L,\omega)|^2 = \left| \frac{g}{\sqrt{2\pi}} \frac{\sqrt{t_R^2 + L^2}}{2\pi} \int_{-\infty}^{\infty} \varepsilon'(L,t)e^{-i\omega t} dt \right|^2 \quad \text{(15)}
\]

In a simple case in the absence of the frequency modulation and neglecting delay effects, the full width at half maximum \( \Delta \nu_{1/2} \) of a spectrally limited pulse is determined only by its duration \( \tau_{1/2} \). For a pulse in the form of a hyperbolic secant \( \nu \), the relation \( \Delta \nu_{1/2} \tau_{1/2} = K_0 \) is valid, where \( K_0 = 0.315 \).

Figures 4a,b show that the spontaneous relaxation \( \Gamma_\varepsilon \equiv 0 \) is suppressed rate \( (n_R = 1.6, n_I = 1.23) \) but the effective frequency detuning \( \Delta_\varepsilon \) is simultaneously formed...
FIG. 4. Time dependences for the population difference $n_{21}$ (dashed curves) and the imaginary part of polarization $\rho_{12}^{(I)}$ (solid curves) in the collective formation regime of SPP on a gold surface initiated by the decay of excitons in a dense ensemble of CdS quantum dots in a dielectric matrix near the metal-dielectric interface under condition $\Delta_\epsilon = -1.48 \times 10^{10}$ s$^{-1}$ ($\Delta_a = 1.04 \times 10^{12}$ s$^{-1}$) by neglecting $(u_R = 0)$ (a) and taking into account (c) nonlinear local-field dispersion effects for $u_R = 0.375$, and also the corresponding frequency spectra (b, d) calculated by (15). The insets in Figs. 4b, d show the profiles of SPP pulses and Fig. 4c — the frequency modulation shape $\Omega_{\nu u}$ of the pulse in the regime with $\Delta_\epsilon = 0$ and $u_R = 0.02$. The interaction parameters are as in Fig. 2.

in the system (see (12)) corresponding to the appearance of linear dispersion. It leads, according to (14), to the formation of dispersion delays and the appearance of characteristic oscillations of the parameter $\rho_{21}^{(I)}$ (and also of $\text{Im}(\omega_0)$) at frequency $\Delta_\epsilon$. The relation between the spectral width $\Delta\nu_{1/2}$ and duration $\tau_{1/2}$ of such a modulated pulse changes to $\Delta\nu_{1/2} = K_{SF}$, where $K_{SF} = 0.8$ (see Figs. 4c, 4d). However, nonlinear phase effects corresponding to the contribute of terms with $u_R$ into (2a) were neglected in Figs. 4a, 4b.

On the other hand, the consideration of only nonlinear terms $\xi_0 u_R \rho_{21} (\rho_{22} - \rho_{11})$ in (4b) in the absence of linear dispersion ($\Delta_\epsilon = 0$) leads to a strong nonlinear frequency modulation of the parameter $\rho_{21}^{(I)}$, which, according to (12) is superimposed on the profile of a generated plasmon pulse. The specific feature of such a modulation is manifested in the change of its sign with displacement from the wing of SPP pulse, where $n_{21} = 1$, to its peak, where $n_{21} = -1$, and in the formation of the characteristic profile of the imaginary component of the Rabi frequency (see the inset in Fig. 4b for arbitrarily chosen $u_R = 0.02$).

Under model conditions, for the chosen values $n_R = 1.6$ and $n_I = 1.23$, the calculated value of $u_R$ will be 0.375 and the effects of linear and nonlinear dispersion will act simultaneously. As a result, a mixed regime with the phase modulation rate nonlinearly increasing from the pulse front to its tail appears in the system (see Fig. 4b). The relation between the duration $\tau_{1/2}$ of such a modulated pulse and its spectral width $\Delta\nu_{1/2}$ takes the form $\Delta\nu_{1/2} = K_{SF}$, where $K_{SF} = 440$, and its spectrum significantly broadens, becoming in fact rectangular (Fig. 4d). This result obtained for a distributed waveguide spaser in a pulsed regime considerably differs from the case of localized spaser with the characteristic spectral narrowing effect [55]. The spectral broadening regime for SPP pulse for the interface presented in Fig. 1 can find new applications in the problems of the development of broadband electromagnetic sources [54], similarly to the generation of laser combs in optics [57].

V. FEATURES OF THE TRIGGERED REGIME OF A WAVEGUIDE SPASER

The feature of the triggered regime in the scheme in Fig. 1, similarly to triggered optical superradiation (TSR), is related to the possibility of controlling the development of cooperative process in a system when the establishment of quantum correlations between individual chromophores is initiated by the external pump pulse. In this case, the development dynamics, the radiation pattern and the shape of a superradiation pulse are completely determined by the parameters of this trigger pulse. In optics, such a regime was first observed in gas medium in [58]. However, only the realization of this effect in solids [59] provided the basis for using TSR for the development of optical memory and optical computing devices [60]. The translation of this problem to plasmonics offers a number of advantages, retaining, on the one hand, optical data processing rates and, on the other hand, considerably simplifying the integration of individual plasmonic devices in circuits and providing their coupling with electronic computing devices.

In the problem (2)–(4), the triggered regime of generation of plasmon pulses can be achieved in the presence of a trigger SPP pulse

$$\varepsilon_{ex}(t) = \varepsilon_0 e^{-(t-nT_0)^2/(2\tau_0^2)}$$

with duration $T_0$ and time delay $nT_0$ ($n \in R^+$) with respect to the beginning of the free evolution of the system due to a relaxation process (see Section III). The trigger pulse can be obtained by transforming an external optical pulse on a metal grating, as in [61] (Fig. 4a). As in the optical case, the specific feature of the regime is the possibility of controlling the delay time of the main SPP pulse [62], which in the classical formulation of the problem by neglecting local field effects is determined by
and its delay duration pulse duration (16) becomes close to the characteristic virtually or completely overlaps the envelope of the generated pulse. Under such conditions, the field intensity

$$\tau_D = \tau_R \ln \left( \frac{1 + \cos \theta}{1 - \cos \theta} \right)$$

(17)

and depends on the trigger pulse area

$$\theta = \frac{2\mu_I}{h} \int A_p e^x dt.$$ 

In this case, the amplitude $A_p e^x$ of the optical trigger pulse can be recalculated to parameters \(16\) according to the relation

$$e_{ex} = A_p e^x \sqrt{\varepsilon_n \frac{\varepsilon_0 V \bar{S}_n}{\hbar S_n}}$$

for the ideal case when 100% of the optical pulse energy transfer to a surface wave.

Figure 5 presents the results of simulating system (2) + (14) in the form of a set of the time dependences of the Rabi frequencies of main pulses produced under the action of input trigger pulses with different areas with increasing their peak intensity $I_0 e^x = (A_p e^x)^2 C_0$, where $C_0 = c\varepsilon_0 / (2nR)$. The corresponding dependences for the delay times of SPP pulse formation are also approximated by expression (17) taking into account that recalculation expressions between the normalized time $\tau$ and the real time $t$ are analogous to the passage from system (7) to (8). For the chosen combination of the QD concentration, the duration and power of trigger pulses, the profile of the main formed pulse in Fig. 5 remains virtually invariable.

The regime will qualitatively change when the trigger pulse duration (16) becomes close to the characteristic duration $t_W$ of the main SPP pulse (transition regime) and its delay $nT_0$ is selected so that its envelope partially or completely overlaps the envelope of the generated pulse. Under such conditions, the field intensity at the medium output can exhibit a multipeak structure (see Figs. 5, 6).

A similar superradiation regime is well known in optics and is caused by the nonmonotonic decay of the excited state of continuous media (63). However, in the case under study for $\Gamma_e = 0$ and $\Delta_e = 0$, the effect is caused by modulation instabilities in the process of QDs excitons decay resulting in the appearance of Rabi oscillations (see the upper inset in Fig. 6) neglecting dispersion effects with $u_R$.

When nonzero frequency detunings $\Delta_e$ are taken into account in system (2), the time synchronization of oscillations of the polarization component $\rho_{21}$ and the population difference $\rho_{21}$ is violated. As a result, the amplitude of oscillations appearing in the system is modulated by a decreasing function of time proportional to the inverse detuning frequency $\Delta_e$ (see the lower inset in Fig. 6) in the approximation $u_R = 0$.

The consideration of the influence of a dielectric host-medium with the dispersion coefficient $u_R$ again leads to a strong nonlinear frequency modulation of the produced pulse (similarly to Figs. 4, 5). However, in the case of its interference with trigger pulse (16) with appropriate duration (as in Fig. 5), the envelope of the resulting pulse acquires a strong high-frequency amplitude modulation, which is absent for $u_R = 0$ in Fig. 6.
recorded by performing the inverse transformation of surface waves to an optical signal on a metal grating [29]. Note that a noticeable change in the permittivity of a metal due to optical excitation of electrons is observed at the energy density on the order of 0.5 mJ/cm² [61]. This allows one to realize pure plasmon nonlinearities and perform direct signal-pump experiments with surface plasmon-polaritons. However, after conversion to dimensional parameters, the energy density of emitted pulses in Fig. 5 does not exceed 0.2 × 10⁻⁹ mJ/cm² and therefore conditions for these nonlinear regimes are not achieved in this work.

It is necessary to note, that the contribution of dissipative effects of the local field to problem (24) can be estimated as

\[
\delta_{\text{loc}}^{(I)} = \frac{\xi_0 u_I \rho_{21}^{(R)}}{\Omega_0^I} = \frac{2}{3} \frac{c u_I}{\omega_a I^{(R)}},
\]

where

\[
I^{(R)} = \int_0^L \exp(ik_a |L - x'|)dx'
\]

and for \( L \approx \lambda_a \), we have \( \delta_{\text{loc}}^{(I)} \approx \frac{2}{3} u_I \). Similarly, we can obtain the estimate for the dispersion coefficient

\[
\delta_{\text{loc}}^{(R)} = \frac{\xi_0 u_R \rho_{21}^{(R)}}{\Omega_0^R} \approx \frac{2}{3} u_R,
\]

determining the relative contribution of local field effects to the frequency modulation of the produced signal. Thus, the contribution of local field effects to the kinematics of the system under study depends only on the introduced coefficients \( u_I \) and \( u_R \) determined only by the material parameters of the host-medium, but not by its geometry.

VI. CONCLUSIONS

We have proposed new efficient methods for the formation and external control of short SPP pulses at the interface of a metal and a QD-doped dielectric medium. The 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. By the example of a model medium with CdS nanocrystals, the dimensional and concentration dependences of the effect are studied and the amplitude and spectral features of SPP pulses generated in the system are determined. The presented model and studied regimes can be used, in particular, for solving a practical problem of increasing the characteristic coherent lengths of the SPP field.

Our approach can be realized in experiments by using dielectric films doped with semiconductor QDs with diameters selected to provide the equality of energies of interband transition and plasmons excited at the metal-dielectric interface. However, it is necessary to take into account that the efficiency of energy transfer from excitons to plasmon modes can be affected by blinking, as in the case of luminescent QDs [38]. In addition, the physical characteristics of QDs significantly differ from perfect and strongly depend on the method of their synthesis and characteristics of the host-medium [65]. In this case, the use of organic molecules can serve as an alternative for interface pumping (Fig. 1, 66).

The models presented in the paper can be useful for practical applications such as the development of plasmonic integrated circuits for quantum computations. In particular, considered collective effects can be used as a basis for multiqubits register initialization in the process of formation the quantum correlations between QDs. The advantage of the realization of such a register in the plasmon-exciton systems to atomic-optical systems is the ability to implement an effective addressing schemes by coupling of each quantum dot with localized plasmon modes on the nanoscale. However, this requires complication of the circuit shown in Fig. 1. Besides, important problems of the direct connection of such systems with all-optical data communication systems remain open. In particular, one of the problems is increasing the efficiency of mutually reversible conversion of the light wave field and plasmon polaritons formed in layered structures [67]. Final answers to these problems can be obtained in relevant experiments, in particular, using epiluminescence spectromicroscopy of single quantum emitters [68–70].

Another important technical problem is achieving very high QD concentrations in a matrix which for the maximum concentration \( N = 4 \times 10^{24} \) m⁻³ used in this paper (Fig. 8) will amount to 1.5% of the concentration \( N_{QD}^D = D_{QD}^{-3} \) of the closest packing of QDs with diameter \( D_{QD} = 1.56 \) nm. One of the solutions can be using the self-organization of QDs with different sizes during their evaporation from colloid solutions [71] on a substrate. However, the prospects for using such structures under conditions of the problem under study require special studies due to a considerable dispersion of QDs in size.

Note in conclusion that it is also important to obtain a more general nonlinear equation describing the propagation of ultrashort SPP pulses in experiments taking into account nonstationary terms of the nonlinear dispersion type, etc. [20, 72]. Such terms can appear due to modification of the metal permittivity by high-power pump femtosecond pulses of an external optical pump [29] and due to nonlinear effects in semiconductor QDs [73] and in a dielectric host-medium containing them [74]. Such an equation can serve as a starting point for searching and determining the stability conditions [75] for its soliton solutions and the development of new schemes of active nanoplasmonics [76] with dissipative SPP solitons.
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