

Supplemental Material for

“Superradiance properties of a suspension of composite nanoparticles”

1. Local dipole fields in the spaser. Since the dye molecules does not penetrates deep into the shell, the distance from the molecules to the spherical gold core of the “spaser” is large enough to apply the dipole approximation. Let $\mathbf{P}_n = \mathbf{d}_n(R_n + R_n^*)$, $\mathbf{P}_0 = \mathbf{d}_0(B + B^*)$ are the polarization vector of dipole transitions of the dye molecules $n = 1, 2, \dots$ and metallic the kernel $n = 0$. R_n and B are the off-diagonal elements of the density matrix of a two-level media and plasmon transition, respectively.

After applying the approximation of a rotating wave and replacing $\rho_n = R_n e^{i\omega t}$, $b = B e^{i\omega t}$ we derive the Maxwell equations for the amplitudes of a quasi-monochromatic electric field

$$\nabla \times \nabla \times \mathbf{E} + k^2 \mathbf{E} = -\frac{\omega^2}{c^2} \sum_n \mathbf{P}_n \delta(\mathbf{r} - \mathbf{r}_n), \quad (1)$$

here $k^2 = \omega^2 \epsilon_h / c^2$, ϵ_h is the dielectric susceptibility of the environment.

The field at the point \mathbf{r} , created by the polarization of dipole transitions of dye molecules and gold nanoparticles (NP) at the points \mathbf{r}_j , $j \neq 0$ and \mathbf{r}_0 , respectively, has the form

$$\mathbf{E}(\mathbf{r}) = \frac{4\pi k^2}{\epsilon_h} \left[\sum_j \overset{\leftrightarrow}{\mathbf{D}}(\mathbf{r}, \mathbf{r}_j) \cdot \mathbf{d}_j \rho_j + \overset{\leftrightarrow}{\mathbf{D}}(\mathbf{r}, \mathbf{r}_0) \cdot \mathbf{M} b \right]. \quad (2)$$

Here $\overset{\leftrightarrow}{\mathbf{D}}(\mathbf{r}, \mathbf{r}')$ is the dyadic Green's function. The point denotes the scalar product.

For a small amplitude b of the plasmon transition excitation we have

$$\partial_t b + i(\omega_p - \omega - i\gamma_p) b = -i \frac{(\mathbf{M} \cdot \mathbf{E})}{\hbar}, \quad (3)$$

here \mathbf{M} is the effective dipole moment of the gold sphere. In the stationary limit, from Eqs. (1), (2) (see the main text) and Eq. (3) we find

$$b = \frac{- \left[\mathbf{M} \cdot \mathbf{E}_0(\mathbf{r}_0) + \sum_j \mathbf{M} \cdot \overset{\leftrightarrow}{\mathbf{D}}(\mathbf{r}, \mathbf{r}_j) \cdot \mathbf{d}_j \rho_j \right]}{\hbar (\omega_p - \omega - i\gamma_p)}. \quad (4)$$

The polarizability coefficient of the gold kernel $\alpha(\omega)$ is reduced to the form corresponding to spherical NP,

$$\alpha(\omega) = \frac{|\mathbf{M}|^2}{\hbar (\omega_p - \omega - i\gamma_p)} \rightarrow \frac{4\pi R_{\text{Au}}^3}{3} \frac{\epsilon_p - \epsilon_h}{\epsilon_p + 2\epsilon_h}. \quad (5)$$

2. Environment parameters. Analysis of the experimental data gave the following values of the physical parameters. The center of dye luminescence resonant to a plasmon mode dye posses the data: $\lambda_d(T_R) = 523$ nm, $\lambda_d(T_N) = 516$ nm, $\gamma_2(T_R) = 79.2$ meV, $\gamma_2(T_N) = 59.2$ meV. With cooling $T_R \rightarrow T_N$ the center of the line shifts to the left by $\delta\lambda_d = 7$ nm (32.1 meV). For spherical Au NP, the following data are obtained: $\lambda_p(T_R) = 520$ nm, $\lambda_p(T_N) = 519.6$ nm, $\gamma_p(T_R) = 305$ meV, $\gamma_p(T_N) = 255$ meV. Spectral characteristics of radiation of suspensions of the “spasers” are as follows: $\lambda_s(T_R) = 529.3$ nm, $\lambda_s(T_N) = 518.4$ nm, $\gamma_s(T_R) = 24.5$ meV, $\gamma_s(T_N) = 7.68$ meV. To fit the shape of absorption lines of Au nanospheres to experimental data the following expressions for the NP susceptibilities were used $\epsilon_p(\omega, T) = 11 - \omega_p^2(T)/(\omega^2 + i\gamma_p(T)\omega)$, $\alpha(\omega) = V_{NP}(\epsilon_p - \epsilon_h)/(\epsilon_p + 2\epsilon_h)$, $\epsilon_h = 1.75$, V_{NP} is the volume of the gold NP. Number of dye molecules in one spaser is $N_d = 3 \times 10^3$. It was believed that dye molecules are absorbed in the ~ 10 nm layer near the outer boundary of silica shell.

3. The effect of temperature on the parameters of the nanosystem. Change of the Au permittivity is described by the following formula

$$\epsilon_{\text{Au}}(T) = 11 - \frac{\omega_p(T)^2}{\omega^2 + i\Gamma_p(T)\omega} + LT, \quad (6)$$

where LT is a Lorentz term that does not depend on T and $\omega_p(T) = \omega(T_R)/\sqrt{1 - \kappa(T_R - T)}$, $\kappa = 4.3 \cdot 10^{-5} K^{-1}$. The dependence $\Gamma(T)$ has the form

$$\Gamma_p(T) = \Gamma(T_R) - \Gamma(T) - \Gamma_0, \quad \Gamma(T) = \Gamma_{e-e}(T) + \Gamma_{e-ph}(T). \quad (7)$$

For Au, the phenomenological value $\Gamma_0 \sim 0.3$ eV is found from experimental data.

Scattering of electrons on the NP surface for sizes $R > 10$ nm does not depend on temperature. Dependencies on temperature of relaxation constants, determined by the electron-electron Γ_{e-e} and electron-phonon scattering Γ_{e-ph} are the following (see, citations [19–21] in the main text)

$$\Gamma_{e-e}(T) = \frac{\pi^2}{24\hbar E_F} [(k_B T)^2 + (\hbar\omega)^2], \quad (8)$$

$$\Gamma_{e-ph}(T) = \Gamma_0 \left[0.4 + \frac{4T^5}{\theta_D^5} \int_0^{\theta_D/T} \frac{z^4 dz}{e^z - 1} \right], \quad (9)$$

where $\hbar E_F = 5.5$ eV is the Fermi energy, k_B is the Boltzmann constant, $\theta_D = 170$ K is the Debye temperature, $\Gamma_0 = 0.07$ eV.

The change in ω_p with temperature can be explained by a change of the volume of the NP and the corresponding change in the electron density n_e , included in the expression for $\omega_p \propto \sqrt{n_e}$, $\omega_p(T_N) = 1.00463\omega_p(T_R)$. From (8) and (9) we found $\Gamma_{e-ph}(T_N) = 0.03994$ eV, $\Gamma_{e-ph}(T_R) = 0.12295$ eV, $\Gamma_{e-e}(T_N) = 0.425173$ eV, $\Gamma_{e-e}(T_R) = 0.425217$ eV. Thus we have: $\Gamma_{e-e}(T_R) - \Gamma_{e-e}(T_N) = 0.00004$ eV, $\Gamma_{e-ph}(T_R) - \Gamma_{e-ph}(T_N) = 0.083$ eV.

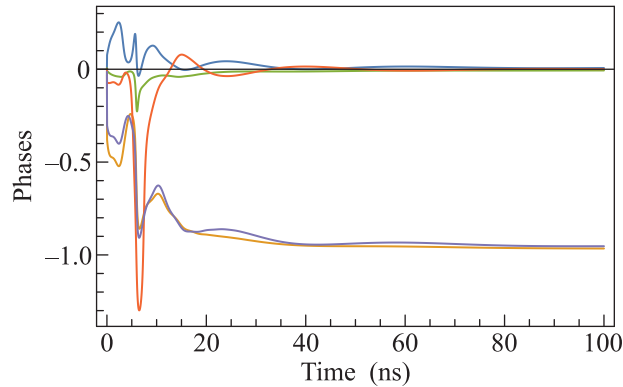


Figure 1: (Color online) Time dependences of the phase differences $(\phi_1(t) - \phi_n(t))/\pi$, nondiagonal elements of the density matrix ρ_n (ns). $\kappa = 0.2$, $F = 0.1$

The main contribution to the relaxation at the resonance frequency is made by electron-electron Coulomb interaction. Electron-phonon scattering makes relatively small contribution to the temperature dependence of relaxation. Change ω_p with temperature leads to a shift of the center of the luminescence line by less than 0.5 nm. The observed shift of the $> \sim 3$ nm (11 meV) line is not explained by temperature dependence of the medium parameters known from literature.

In the experiment, the dye molecules were pumped by pulses with an interval of 10 ns and duration ~ 7 ns. For study of synchronization of dye molecules polarizations we performed a numerical analysis of the system Bloch equations for $N_d = 10, 20$ molecules in the case of one pulse of outer field.

If $F = 0$ then during the time $< \sim 1$ ns the phases of all polarizations of dipole transitions become equal (“ferromagnetic state”) due to the action of the external field $E_0 \neq 0$ and (or) due to the nonlinear feedback. Fluctuations of $F \neq 0$ lead to desynchronization of polarizations. With increasing of the fluctuations amplitude F , a partial synchronization, in which, mixed states of the ferromagnetic and antiferromagnetic type arise, see Fig. 1. An increase in the amplitude of the fluctuations corresponding to temperature increasing leads to the destruction of states of the ferromagnetic type. This corresponds to decrease in the intensity of the output field $\propto |q|^2$ generated by dye molecules, see Fig. 2.

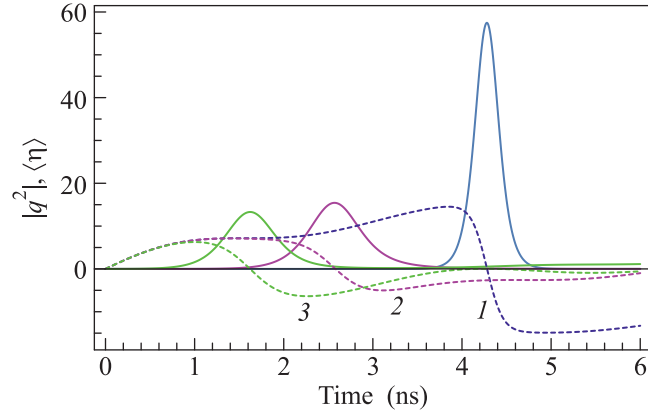


Figure 2: (Color online)(Color online) Dependence of $|q^2|$ (solid lines) and $\langle \eta \rangle$ (dashed lines) versus time (ns) for different fluctuation amplitudes F . The values $F = 0, 0.01, 0.1$ correspond to blue 1, pink 2 and green 3 lines, respectively. $\kappa = 0.125$

Numerical simulation of the effects of fluctuations of levels population, relaxation constants and input power density have shown that their contributions to the desynchronization of polarizations is much less than the contribution of the term $FG(t)$.