

# Lasing of nanoemitters with nanorings having Drude dispersion

Nanoemisores láser con nanoanillos que tienen dispersión Drude

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## KEYWORDS:

Nanoemitters,  
nanorings, laser  
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polariton

## ABSTRACT

We systematically study the dynamics and laser emission of randomly distributed nanoemitters (NEs) integrated into two-dimensional (2D) array of metal nanorings (NR) enhanced by the plasmon-polariton (PP) excitations. It is shown that the transition time to the laser generation regime (instability) of NE optical fields significantly depends on the plasma frequency  $\omega_p$  of the NR. The PP fields is excited macroscopically in the whole system if  $\omega_p$  of NR exceeds a certain critical transition value  $\omega_p > \omega_c$ . The latter leads to a strong coupling of the NE emission with the PP fields and a critical increase of the total current in the NR array. We found the analytical dependence of the critical current amplitude  $I = I(\omega_p/\omega_c)$  as function of NR plasma frequency, which is in a good agreement with the numerical simulations.

## PALABRAS CLAVE: RESUMEN

Nanoemisores,  
nanoanillos,  
emisión láser,  
plasmon-polaritón

Estudiamos sistemáticamente la dinámica y la emisión láser de nanoemisores (NE) distribuidos aleatoriamente integrados en una matriz bidimensional (2D) de nanoanillos metálicos (NR) mejorados por las excitaciones de plasmón-polaritón (PP). Se muestra que el tiempo de transición al régimen de generación de láser (inestabilidad) de los campos ópticos NE depende significativamente de la frecuencia del plasma  $\omega_p$  del NR. Los campos PP se excitan macroscópicamente en todo el sistema si  $\omega_p$  de NR excede un cierto valor de transición crítico  $\omega_p > \omega_c$ . Esto último conduce a un fuerte acoplamiento de la emisión de NE con los campos de PP y a un aumento crítico de la corriente total en el conjunto de NR. Encontramos la dependencia analítica de la amplitud de la corriente crítica  $I = I(\omega_p/\omega_c)$  en función de la frecuencia del plasma NR, lo que concuerda bien con las simulaciones numéricas.

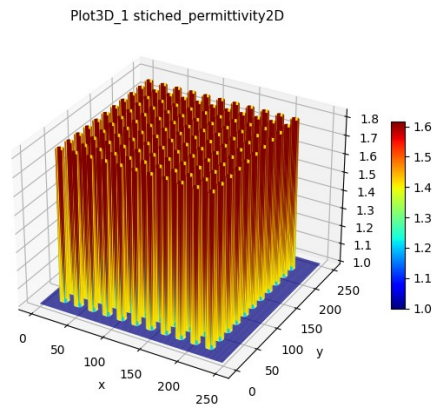
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## 1. INTRODUCTION

Nowadays, investigations of aligned carbon nanostructures have stimulated various spectroscopic studies on a spatial scale of several nanometers [1,2,3,4,5,6,7,8,9,10]

[11,12]. Considerable interest is devoted to study linear and nonlinear plasmonic properties of nanohybrids and nanocomposites made of metallic nanoparticles and quantum emitters (QEs) [13,14,15,16,17]. The inclusion of dispersed

single-walled carbon nanotubes (SWCNTs) in the working space of QEs or nanoemitters (NEs) considerably changes the properties of the electromagnetic field, whose structure depends significantly on the plasma frequency  $\omega_p$  of SWCNTs. In such a hybrid system, it becomes possible to control the properties of local optical fields that allows creating of miniature low threshold coherent sources with tunable polarization [18, 6]. For the low frequency spectrum, such a medium behaves like a system of dielectric cylinders with the well-known properties of Bloch waves [19]. But for high frequencies, in the system already dominate the dynamic properties of electromagnetic waves in plasma cylinders, where the surface plasmons can propagate. In the intermediate frequency region, the fields acquire a complex structure in the area of cut-off of plasmonic wave modes. The spectral properties of such modes in SWCNTs in the ideal case of lossless and infinite SWCNTs have been well-studied in quantum and quasiclassical approximations [20,21,22,23].



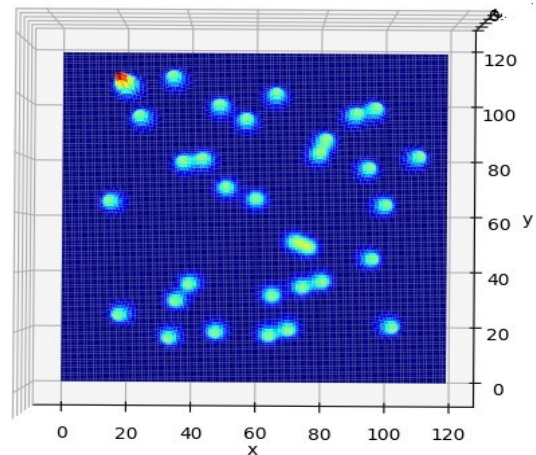
**Figure 1.** The array of nanorings (NR) with external radius  $R$  dielectric permittivity  $\epsilon^\infty = 1.8$  and width of the well  $w$ , periodically placed on a dielectric substrate.

However, in real case of finite and lossy SWCNTs, new factors become to play an important role. The plasmonic fields of dispersing nanotubes perturb the energy levels of the NE and, thus, the PPs in SWCNTs appear to be related to the inner degrees of freedom of the NEs. An important property of NEs is the nonlinearity of the field and the possibility of the laser generation [24]. In such a hybrid

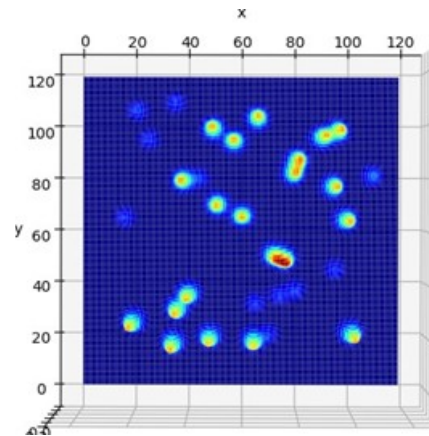
system, the lasing threshold in the NE depends on the spatial structure of randomly distributed NEs. It is found that all mentioned factors can establish of a resonant change in the field structure associated with the PP generation in SWCNTs.

## 2. GEOMETRY

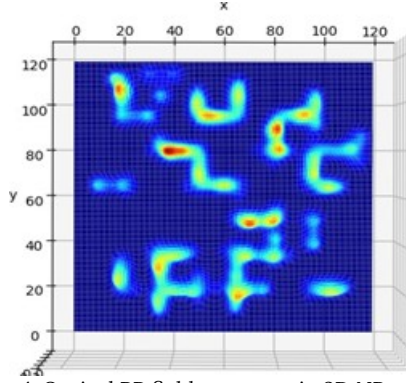
We consider the (metal) nanorings (NR) having external radius  $R$  and width of the well  $w$  periodically placed in 2D dielectric substrate, see Fig.1. (Here the colorbar indicates the static dielectric permittivity of ND).



**Figure 2.** Initial distribution of 33 NE in the 2D nanorings array (see Fig. 1).



**Figure 3.** Optical field PP structure in the 2D NR array (see Fig. 1) with NE at small time  $t = 19$  before the lasing of NE begins.



**Figure 4.** Optical PP field structure in 2D NR array (see Fig. 1) with lasing of NE ( $t = 39$ ) and the field interconnections between NR.

### 3. BASIC EQUATIONS

In considered hybrid system the Maxwell equations read:

$$\nabla \times \mathbf{E} = -\mu_0 \frac{\partial \mathbf{H}}{\partial t}, \quad \nabla \times \mathbf{H} = \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} + \mathbf{J}_e + \sigma_e \mathbf{E} + \sum_k I_k \mathbf{P}_k(\mathbf{R}_k, t) \delta_{r, \mathbf{R}_k}, \quad (1)$$

where  $\mathbf{J}_e$  is the electrical current in NR,  $t$  is the time,  $\mathbf{E}$  and  $\mathbf{H}$  are the electric and magnetic fields respectively, which obeys the material equation [25].

$$\mathbf{J}_e + \gamma_e \mathbf{J}_e = b_e \mathbf{E}, \quad (2)$$

where  $\gamma_e$  is the collision frequency in NR,  $b_e = \epsilon_0 \omega_p^2$ ,  $\omega_p$  is the plasma frequency,  $\sigma_e$  is conductivity, and,  $\epsilon_h$  is the dielectric constant of the host medium of NR. In Eq.(1)  $I_k = \frac{\partial \mathbf{P}_k}{\partial t}$  and  $\mathbf{P}_k(\mathbf{R}_k, t)$  ( $k=1, N$ ) is the polarization density produced by  $N$  NEs in positions  $\mathbf{R}_k$ . Following the single electron case, the equation for  $\mathbf{P}_k$  in the vicinity of the emitters is [24].

$$\frac{\partial^2 \mathbf{P}_k}{\partial t^2} + \Delta \omega_a \frac{\partial \mathbf{P}_k}{\partial t} + \omega_a^2 \mathbf{P}_k = \frac{6\pi\epsilon_0 c^3}{\tau_{21} \omega_a^2} (N_{1,k} - N_{2,k}) \mathbf{E}_k. \quad (3)$$

To complete the model, we add the rate equations [24] for the occupation levels of emitters  $N_{i,k} = N_i(\mathbf{R}_k, t)$  (following [17], we consider that the NEs are four-level quantum systems or quantum dots (QD)):

$$\frac{\partial N_{0,k}}{\partial t} = -A_r N_{0,k} + \frac{N_{1,k}}{\tau_{13}}, \quad \frac{\partial N_{3,k}}{\partial t} = A_r N_{0,k} - \frac{N_{3,k}}{\tau_{02}}, \quad (4)$$

$$\frac{\partial N_{1,k}}{\partial t} = \frac{N_{2,k}}{\tau_{32}} - M_k - \frac{N_{1,k}}{\tau_{13}}, \quad \frac{\partial N_{2,k}}{\partial t} = \frac{N_{1,k}}{\tau_{12}} + M_k - \frac{N_{2,k}}{\tau_{02}}, \quad (5)$$

$$M_k = \frac{(\mathbf{j} \cdot \mathbf{E})_k}{\hbar \omega_a}, \quad j_k = \frac{\partial \mathbf{P}_k}{\partial t}.$$

Here  $\Delta \omega_a = \tau_{21}^{-1} + 2T_2^{-1}$ , where  $T_2$  is the mean time between dephasing events,  $\tau_{21}$  is the decay time from the second atomic level to the first one,  $\omega_a$  is the frequency of radiation (see e.g. [24]), and  $A_r$  is a certain pumping rate, which is proportional to the pumping intensity in experiments. In our simulations we considered the gain medium with parameters close GaN powder, similar to Ref. [26]. The lasing frequency  $\omega_a$  is  $2\pi \times 3 \times 10^{13}$  Hz, the lifetimes are  $\tau_{32} = 0.3$  ps,  $\tau_{10} = 1.6$  ps,  $\tau_{21} = 16.6$  ps, and the dephasing time is  $T_2 = 0.0218$  ps. The nonlinear quasi-classical system (1)-(5) cannot be solved analytically in general; therefore, below we use the numerical Finite Difference Time Domain (FDTD) simulations (in the output we impose the absorbing boundary conditions (ABC)) [25]. The later allowing us to obtain the exact solutions to system (1)-(5). In what follows we use the dimensionless time  $t$  renormalized as  $t \rightarrow tc/l_0$ , where  $l_0 = 100 \mu m$  is the typical spatial scale and  $c$  is the light velocity in the vacuum.

### 4. PLASMONS IN NANORING

In this Section we start to discuss the properties of a simplest plasmonic modes in a single cylindrical nanoparticle [20,21,22] as a lossless, infinitesimally thin cylindrical shell with radius  $R$ , where the valence electrons are considered as a free-electron gas distributed uniformly over the cylindrical surface, with the density per unit area  $n_0$ . It is used the cylindrical coordinates  $r = (\rho, \varphi, z)$  and considered an electromagnetic wave with frequency  $\omega$ , propagating along the nanoring axis  $z$ . The homogeneous electron gas is perturbed by the electromagnetic wave and can be regarded as a charged fluid with the velocity field  $v(r_s, t)$  and the perturbed

density (per unit area)  $n_i(r_s, t)$ , where  $r_s=(\varphi, z)$  are the coordinates of a point at the cylindrical surface of the nanoring. The electronic excitations on the cylindrical surface surface can be described by the following (linearized) continuity equation and the momentum-balance equation respectively [22].

$$\frac{\partial}{\partial t} n_1 + n_0 + \nabla_{\parallel} v_1 = 0, \quad \frac{\partial}{\partial t} v_1 = \frac{-e}{m_e} E_{\parallel} - \frac{\alpha}{n_0} \nabla_{\parallel} n_1 + \frac{\beta}{n_0} \nabla_{\parallel} (\nabla_{\parallel}^2 n_1) \quad (6)$$

$$\frac{\partial}{\partial t} \rho_1 + n_0 + \nabla_{\parallel} j_1 = 0, \quad \frac{\partial}{\partial t} j_1 = \frac{-e^2 n_0}{m_e} E_{\parallel} - \frac{\alpha n_0}{n_0} \nabla_{\parallel} \rho_1 + \frac{\beta n_0}{n_0} \nabla_{\parallel} (\nabla_{\parallel}^2 \rho_1), \quad (7)$$

where  $E_t$  is the tangential component of the electromagnetic field,  $e$  is the element charge, and  $m_e$  is the electron mass,  $\rho_1 = en_1$ ,  $j_1 = en_0 v_1$ , and coefficients  $\alpha, \beta$  describe single electron excitations in the 2D electron gas. In particular,  $\alpha = v_F^2$  is the speed of propagation of the density disturbances in the electron gas with  $v_F = (2\pi n_0 a_B^2)^{1/2} v_B$  being the Fermi velocity of the 2D electron gas and  $\beta = (a_B v_B)^2 / 4$  describes single-electron excitations in the electron gas. Here  $a_B$  and  $v_B$  are the Bohr radius and the Bohr velocity, respectively.

In the follows we will use the simple Drude model, where the equation for electrical current  $j_k$  in  $k$ -th nanoring reads

$$\frac{\partial j_k}{\partial t} + \gamma j_k = \epsilon_0 \omega_p^2 E \quad (8)$$

where  $\omega_p^2$  is the plasma frequency,  $\gamma$  is the loss (phenomenological inverse relaxation time). The total electric current of the plasmons in all the nanorings in the system can be written as

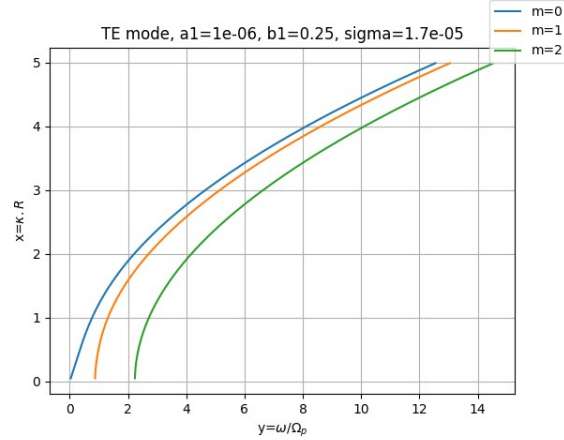
$$J = \sum_{k=1}^N j_{1k}$$

where  $N$  is the number of nanorings. Due to the cylindrical symmetry of a NP we replace  $\rho_1, j_1$  and the fields in the Maxwell's equations  $\nabla \times \mathbf{E} = -\partial \mathbf{B} / \partial t$ ,  $\nabla \times \mathbf{B} = c^{-2} \partial \mathbf{E} / \partial t$  by the Fourier forms

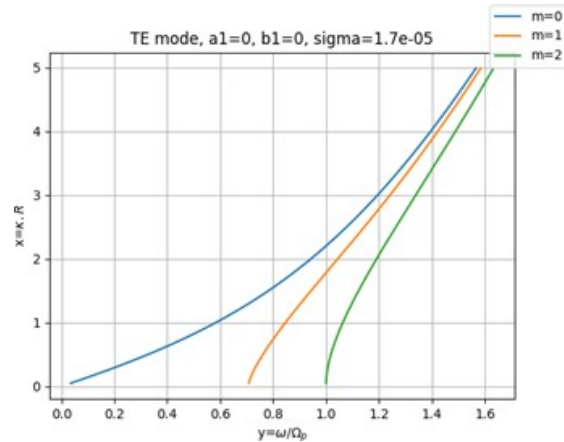
$$|E(r, \varphi, z, t), B(r, \varphi, z, t)| = \sum_{m=-\infty}^{+\infty} \int_{-\infty}^{\infty} dq [E(r, q), B(r, q)] \exp i(m\varphi + qz - \omega t),$$

where  $m$  is the integer azimuth quantum number, and  $q$  is the longitudinal wave vector. Substituting these expressions into boundary conditions, we obtain the dispersion relation between the frequency  $\omega$  and the wave number  $k$  (TE modes) in the following dimensionless form:

$$D_{TE}(x, y) \equiv y^2 - \alpha_1(x^2 + \sigma y^2 + m^2) - \beta_1(x^2 + \sigma y^2 + m^2)^2 - x I'_m(x) K'_m(x) = 0, \quad (9)$$



**Figure 5.** Dispersion dependence for TE waves for case of nonzero coefficients  $a_1$  and  $b_1$  in the dispersion Eq. (9).

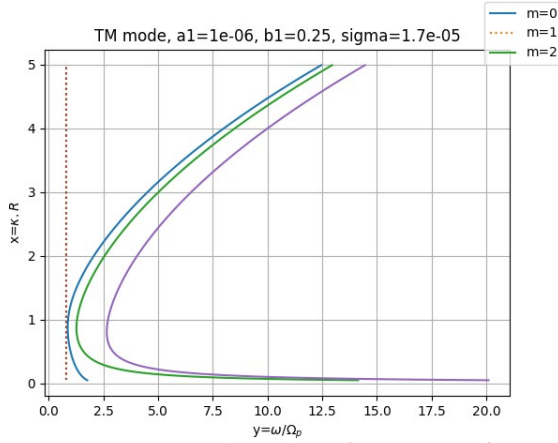


**Figure 6.** Dispersion dependence for TE waves for case of zero coefficients  $a_1$  and  $b_1$  in the dispersion Eq. (9).

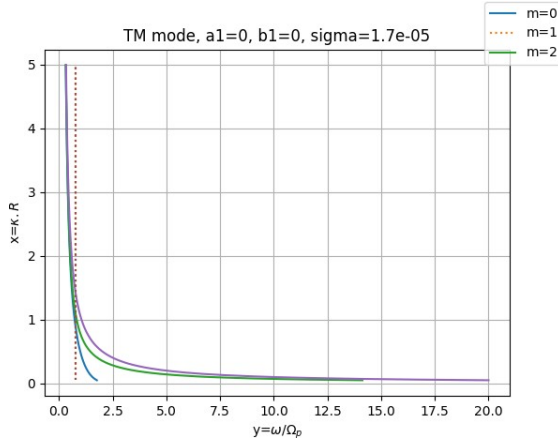
$$D_{TE}(x, y) \equiv y^2 - \alpha_1(x^2 + \sigma y^2 + m^2) - \beta_1(x^2 + \sigma y^2 + m^2)^2 - x^{-2}(x^2 + \sigma y^2 + m^2) I'_m(x) K'_m(x) = 0, \quad (10)$$

where  $y = \omega / \Omega_p$ ,  $x = \kappa R$ ,  $\kappa^2 = q^2 - k^2$ ,  $k = \omega / c$  is the wave number,  $c$  is the light speed,  $R$  is radius of nanoring. In Eq. (10)  $\Omega_p = (e^2 n_0 / \epsilon_0 m_e R)^{1/2}$ ,  $I'_m(x)$  and  $K'_m(x)$  denote the derivatives of the modified Bessel functions

with respect to argument [27], and  $\epsilon_0$  is the permittivity of free space,  $\alpha_1 = \alpha(\omega_p R)^2$ ,  $\beta_1 = (\Omega_p^2 R^4)$ , and  $\sigma = \Omega_p^2 R^2 / c^2$ .



**Figure 7.** Dispersion dependence for TM waves for case of non zero coefficients  $a_1$  and  $b_1$  in the dispersion Eq. (10).

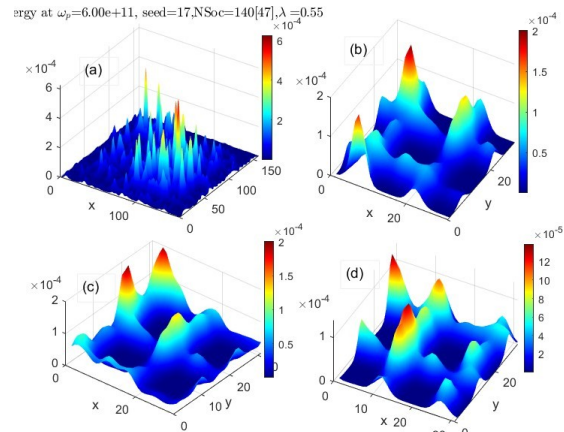


**Figure 8.** Dispersion dependence for TM waves for case of zero coefficients  $a_1$  and  $b_1$  in the dispersion Eq. (10).

The eigenvalues  $\omega^2 = [\omega_m(q)]^2$  obtained by solving (10) represent the plasmon dynamics in the infinitely long and lossless nanoring. Fig. 7 shows the solution of Eq. (10) for  $x = \kappa R$  as function of  $y = \omega / \Omega_p$  at different  $m$ . One can see that the dispersion curves  $\kappa R$  for the nanoring increase with increasing the value  $\omega$  of for all  $m \geq 0$ , and approach the plasmon frequency of the 2D electron gas  $y^2 = x/2$  [19]. Since the azimuth symmetric  $m=0$  plasmon mode normally does not contribute to the high-frequency plasmons [28], we will consider the frequency range  $\omega > \omega_c$ , where  $\omega_c$  is

the cut-off frequency for the first azimuthal asymmetric  $m=1$  mode of plasmon (at  $y \approx 0.7$ , see Fig 8) We evaluate  $\omega_c$  for the charge density  $n_0 = 2.15 \times 10^{13} m^{-2}$  and  $R = 2.4 nm$  when  $\Omega_p$  is about  $2\pi \times 0.34$  THz that, as Fig. 8 shows, for  $m=1$  corresponds to  $\omega_c = 2\pi \times 0.24$  THz.

In what follows, such a frequency  $\omega_c$  can be mentioned as a critical value at that the asymmetric plasmons can be generated and coupled to the field of embedded NEs. However the study of plasmon dynamics in a simple model of lossless, infinite nanorings is too simplified. Numerical studies are needed to investigate the details of PPs in a periodic array of finite and dispersive NR, where the field interconnections and coupling the plasmon field to embedded NEs are possible.

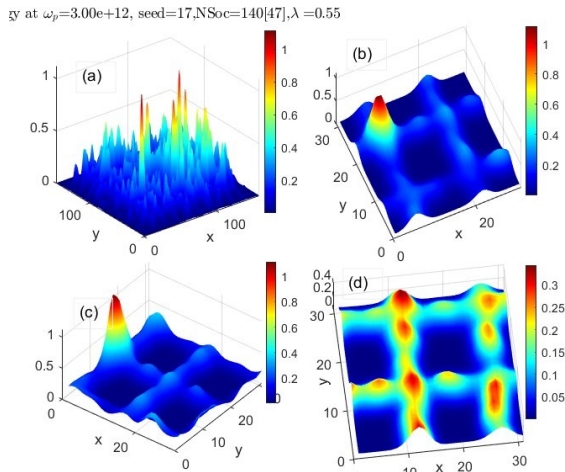


**Figure 9.** Spatial distribution of the average PP field energy (arbitrary units) at  $\omega_p = 0.6$  THz and  $N_e = 47$  radiated NE: (a) the energy distribution in the complete NR array; (b) views the energy in the local rectangular [30, 30; 60, 60]; (c) [35, 35; 65, 65]; (d) [40, 40; 70, 70] respectively. It is seen that the energy is concentrated mainly in the gaps and the field practically does not penetrate inside NRs. For such value  $\omega_p = 0.6$  THz the typical scale of the amplitude of PP field energy  $\approx 10^{-4}$  is negligible small with respect of cases with larger  $\omega_p > 1.0$  THz, see figures below.

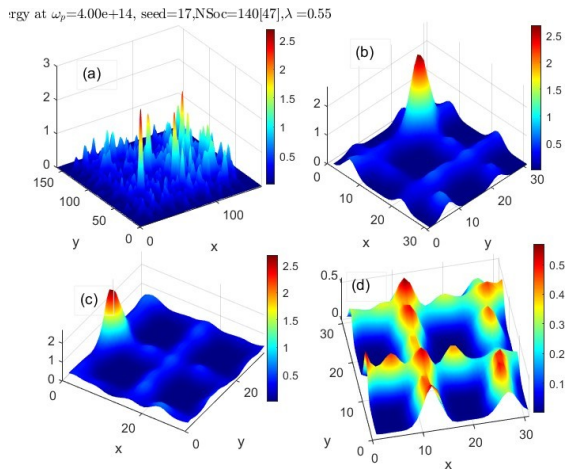
## CONCLUSION

We have studied the dynamics and laser emission of randomly distributed nanoemitters (NEs) integrated into two-dimensional (2D) array of metal nanorings

(NR) enhanced by the plasmon-polariton (PP) excitations. It is shown that the transition time to the laser generation regime (instability) of NE optical fields significantly depends on the plasma frequency  $\omega_p$  of the NR. The PP fields is excited macroscopically in the whole system if  $\omega_p$  of NR exceeds a certain critical transition value  $\omega_p > \omega_c$ . The latter leads to a strong coupling of the NE emission with the PP fields and a critical increase of the total current in the NR array.



**Figure 10.** The same as in Fig. 9 but for  $\omega_p=6.0\text{THz}$ . Besides, we observe that for this  $\omega_p=6\text{THz}$  the typical scale of the field energy is  $\sim 1$  that is at 4 orders larger with respect of the case  $\omega_p=0.6\text{THz}$  shown on Fig. 9.



**Figure 11.** The same as in Fig. 9 but for  $\omega_p=4.0 \times 10^2 \text{ THz}$ .

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