Steady state superradiance of a 2D-spaser array

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Abstract: We show that due to near-field interaction of plasmonic particles via gain particles, a two-dimensional array of incoherently pumped spasers can be self-synchronized so that the dipole moments of all the plasmonic particles oscillate in phase and in parallel to the array plane. The synchronized state is established as a result of competition with the other possible modes having different wavenumbers and it is not destroyed by radiation of leaking waves, retardation effects, and small disorder. Such an array produces a narrow beam of coherent light due to continuous-wave superradiance. Thus, spasers, which mainly generate near-fields, become an efficient source of far-field radiation when the interaction between them is sufficiently strong.

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1. Introduction

Recent developments of nanotechnologies incorporating plasmonic structures have led to the design of a new generation of components for optoelectronics operating in the deep subwavelength regime. Such devices include plasmonic waveguides, spectrometers and microscopes, in which local fields are enhanced by plasmonic resonance [1–3]. In 2003 Bergman and Stockman [4] proposed a quantum-plasmonic device, the spaser, that was demonstrated experimentally [5, 6]. Schematically, the spaser is an inversely populated two-level system (TLS), e.g. an atom, a molecule, or a quantum dot, placed near a plasmonic nanoparticle (NP) [4, 7]. Due to small length scales (d < 20 nm), energy transfer from the TLS to the NP is provided by near-fields [8–10]. Spasers have never been considered as efficient sources of light radiation but rather as systems that create high local intensity of the electric field and enhance nonlinear effects [11].

Boosting energy extraction from spasers is of special interest [12]. It is mainly concerned with excitation and support of eigenmodes of plasmonic transmission lines [2, 13–21]. In particular, a chain of plasmonic NPs with dipole-dipole interaction supports travelling waves of NP dipole moments [22, 23]. The electromagnetic field of such eigenmodes is strongly localized on the chain. The synchronized modes with zero and near-zero wave numbers radiate leaky waves, which cause the far-field interaction of NPs. However, this interaction changes the dispersion curve and destroys the synchronization [17]. When the wavenumber decreases and tends to that of free space, the eigenfrequency tends to zero and does not reach the plasmonic resonance.

Chains of spasers also support travelling waves [24]. Due to the nonlinear nature of such systems, mode competition leads to survival of only one wave with fixed frequency and wavenumber. For weak TLS-NP interactions, this is a solution with a high wavenumber q ($q \gg \omega / c = k_0$). In this case, only near-field interactions need to be taken into account [17]. For high values of the TLS-NP coupling constant, the theory predicts in-phase oscillations accompanied by radiation of leaking waves. A consistent description of such synchronized states requires taking into account radiation and retardation effects which is done in this paper. We show that in a large 2D array, spasers can be mutually synchronized despite radiation and the far-field interaction. This synchronization arises due to the interaction of the spaser's TLS with plasmonic particles of neighboring spasers. Synchronization results in superradiance. For arrays smaller than the free space wavelength, the interference of radiated fields is constructive in all directions, and the radiation intensity power depends on the number of spasers, N, as N^2. For larger systems, the interference becomes destructive for almost all directions and the total radiation power is linear in N. Only in the direction perpendicular to the array plane the interference is still constructive. In this direction, the power of radiation per solid angle is proportional to N^2.

2. Synchronization of the spaser array

As the 2D array of spasers, we consider inversely populated TLS's positioned near nano-holes in a metallic film, as shown in Fig. 1. A nano-hole with a dipole mode plays role of the NP. The spacing between holes Δ is much smaller than the wavelength. We consider a square array with sides L so that the number of spasers in the array is $N=(L/\Delta)^2$. The TLS can be electrically pumped via either a p-n junction or a quantum well parallel to the film. However, discrete TLS's are more convenient for modeling.

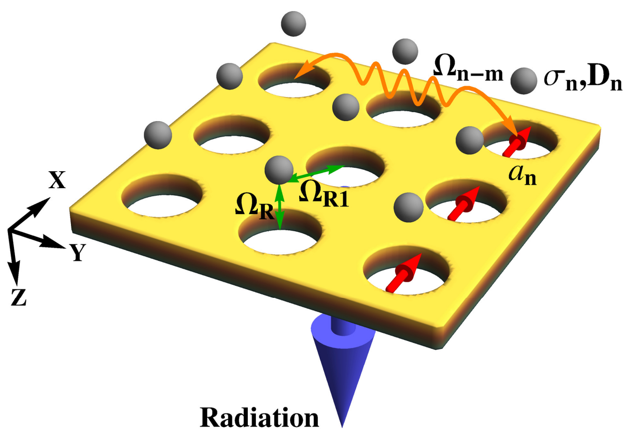

Fig. 1. Phase distribution of the plasmon oscillations in spaser arrays of (a) 5 × 5 and (b) 100 × 100 spasers. In all calculations, we use $\Delta = \lambda/20$, where λ is the free space wavelength.

The dynamics of a single spaser is described by the system of three equations for operators of the plasmon amplitude $a_{\mathbf{n}}$, the TLS polarization $\sigma_{\mathbf{n}}$ and the population inversion $D_{\mathbf{n}}$ [25, 26]. To properly describe radiation of the array, we have to take into account retardation while considering the interaction between the spasers. A spaser located at the point $\mathbf{n}=\{n_x,n_y\}$ "feels" the local field $\Omega_{\mathbf{n-m}}a_{\mathbf{m}}=\Omega\left((\mathbf{n}-\mathbf{m})\Delta\right)a_{\mathbf{m}}$ of a spaser, located at the point $\mathbf{m}=\{m_x,m_y\}$. This field includes both far and near-fields:

$$\Omega\left(\mathbf{e}R\right)=\tau_R^{-1}\frac{3}{2}\left(\frac{3(\mathbf{e}\cdot\mathbf{e}_x)^2-1}{\left(k_0R\right)^3}-i\frac{3(\mathbf{e}\cdot\mathbf{e}_x)^2-1}{\left(k_0R\right)^2}-\frac{(\mathbf{e}\cdot\mathbf{e}_x)^2-1}{k_0R}\right)\exp(ikR), \qquad (1)$$

where $k_0=\omega/c$ is the wave number of radiation in vacuum, $\mathbf{e}_x$ is the unit vector parallel to the dipole moments, $\mathbf{e}R$ is the vector connecting the dipoles. The local field given by Eq. (1) is the x-projection of the electric field created by a unitary dipole $\mathbf{e}_x$ pointed in x direction,

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expressed in units of the inverse radiation relaxation rate, τ_R^{-1}. It can be shown that at the dipole resonance frequency ω_r the value of τ_R^{-1} for a NP of any shape is related to polarizability $\alpha(\omega)$ as $\tau_R^{-1} = -2k_0^3/3\left(\partial\alpha^{-1}/\partial\omega\right)\Big|_{\omega=\omega_r}$. The expression for the polarizability of a hole in metal films was obtained in [27].

In the rotating wave approximation [28–31], the system of interacting spasers is described by the system of equations

$$\dot{a}_{\mathbf{n}} + \tau_a^{-1} a_{\mathbf{n}} = -i\Omega_R \sigma_{\mathbf{n}} - i\Omega_{R1} \sum_{|\mathbf{m}-\mathbf{n}|=1} \sigma_{\mathbf{m}} + i\sum_{\mathbf{m}\neq\mathbf{n}} \Omega_{\mathbf{n}-\mathbf{m}} a_{\mathbf{m}}, \tag{2}$$

$$\dot{\sigma}_{\mathbf{n}} + \tau_\sigma^{-1}\sigma_{\mathbf{n}} = i\Omega_R a_{\mathbf{n}} D_{\mathbf{n}} + i\Omega_{R1} \sum_{|\mathbf{m}-\mathbf{n}|=1} a_{\mathbf{m}} D_{\mathbf{m}}, \tag{3}$$

$$\dot{D}_{\mathbf{n}} + \left(D_{\mathbf{n}} - D_0\right)\tau_D^{-1} = 2i\Omega_R (a_{\mathbf{n}}^* \sigma_{\mathbf{n}} - \sigma_{\mathbf{n}}^* a_{\mathbf{n}}) + 2i\Omega_{R1} \sum_{|\mathbf{m}-\mathbf{n}|=1} \left(a_{\mathbf{m}}^* \sigma_{\mathbf{n}} - a_{\mathbf{m}} \sigma_{\mathbf{n}}^*\right), \tag{4}$$

where τ_σ and τ_D denote relaxation times of the polarization and the population inversion, respectively, D_0 describes pumping of a TLS and corresponds to the population inversion in absence of NPs. In Eqs. (2)-(4), Ω_R and Ω_{R1} (we consider $\Omega_R = 110\,\tau_R^{-1}$ and $\Omega_{R1} = 0.3\,\Omega_R$) characterizes the interaction of a TLS with the adjacent and neighboring NPs, respectively. Ω_R and Ω_{R1} are real-valued quantities which correspond to neglecting retardation and taking into account only near-fields in the TLS-NP interaction. The latter is justified by the small distance between the TLS and the NP. This interaction synchronizes the spaser array [24]. To proceed, we neglect quantum correlations and fluctuations and substitute operators by *c*-numbers. The decay rate of plasmonic modes is determined by the Joule loss in the NP and radiation, $\tau_a^{-1} = \tau_J^{-1} + \tau_R^{-1}$ (in our calculations $\tau_J^{-1} = 27\tau_R^{-1}$).

The local field given by Eq. (1) is characteristic for free space. The only effect of the metal film that we take into account by having an imaginary part in the wavevector, $k = k' + ik''$, is attenuation of the wave in space (for calculations we assume that $k' = k_0$ and $k''/k' = 0.2$).

Figure 2 shows phase distributions for small and large arrays, in which TLS's interact with NPs of neighboring spasers ($\Omega_{R1} \neq 0$). These distributions are obtained by solving dynamic Eqs. (2)–(4) numerically until the system reaches a stationary state. For both small and large arrays, the phase distributions are almost uniform. In the large system, a considerable deviation from the uniform distribution only occurs near boundaries of the array in the *x*-direction, which is parallel to the direction of oscillations of the dipole moments [Fig. 2(b)]. This is a boundary effect for which the length scale is of the order of the free space wavelength. In addition, the phase exhibits weak spatial oscillations along the direction perpendicular to the direction of dipoles. These oscillations increase in lossless systems. Both of these effects are due to a non-uniform change of the effective relaxation times of spasers and they do not lead to significant changes in the radiation pattern of large lossy spaser arrays.

The effect of synchronization is a result of competition of collective modes in which the oscillations of spasers in the array are modulated by the wavenumber $\mathbf{q} = \{q_x, q_y\}$. The solution of the dynamic equations found by computer simulation shows that the mode with $q = 0$ corresponding to the synchronized state wins the competition with other modes.

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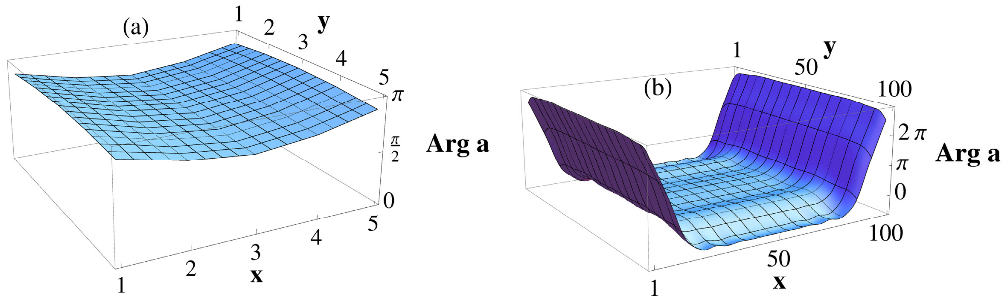

Fig. 2. Phase distribution of the plasmon oscillations in spaser arrays of (a) 5 × 5 and (b) 100 × 100 spasers. In all calculations, we use $\Delta = \lambda/20$, where λ is the free space wavelength.

The winning mode is determined by the minimum value of pumping threshold, $D_{th}(\mathbf{q})$ [24, 32]. For the spaser array the threshold pumping may be written in the form $D_{th}(\mathbf{q}) = \Omega_{R\_eff}^{-2}\tau_{a\_eff}^{-1}\tau_{\sigma}^{-1}\left(1+\delta_{eff}^{2}\tau_{a\_eff}^{2}\right)$, which is identical to that for a single spaser [25], with the only difference that Ω_R, τ_a, and frequency detuning, δ, are replaced by the values modified by interactions of a NP with TLS's and NPs of neighboring spasers:

$$\begin{aligned}
\Omega_{R_eff} &= \Omega_R + 2\Omega_{R1}\left(\cos q_x\Delta + \cos q_y\Delta\right),\\
\tau_{a_eff}^{-1} &= \tau_J^{-1} + \Delta^{-2}\int \Omega(\mathbf{e}R)\, d^2\mathbf{R},\\
\delta_{eff} &\approx 3\tau_R^{-1}\left(k_0\Delta\right)^{-3}\left(2\cos q_x\Delta - \cos q_y\Delta\right).
\end{aligned} \tag{5}$$

The integral in the second equation can be calculated in the case $\mathbf{q}=0$. In addition, if $k'' \to 0$, one obtains $\tau_{a\_eff}^{-1} \approx \tau_J^{-1} + 3\pi\tau_R^{-1}k_0^{-2}\Delta^{-2}$.

The value of the $D_{th}(\mathbf{q})$ is determined by three factors, $\Omega_{R\_eff}^{-2}$, $\tau_{a\_eff}^{-1}$, and $1+\delta_{eff}^{2}\tau_{a\_eff}^{2}$, given in Eqs. (5). The factor $\Omega_{R\_eff}^{-2}$ has a minimum at $\mathbf{q}=0$. Indeed, in the synchronized mode there is a constructive interference of the fields induced on a TLS by the neighboring NPs. Due to this interference, the corresponding coupling and the energy transfer are the most efficient. This mechanism of synchronization was considered in [24]. On the other hand, the second factor, $\tau_{a\_eff}^{-1}$, increases with vanishing k due to radiation losses, which causes increase of D_{th}. Due to this factor a desynchronized mode with minimal radiation should win in the mode competition. At the same time, increase of $\tau_{a\_eff}^{-1}$ makes the third factor, $1+\delta_{eff}^{2}\tau_{a\_eff}^{2}$, insignificant, because it becomes of the order of unity for any $\mathbf{q}$ for large arrays.

Thus, we have two competing mechanisms: (1) the synchronized mode extracts energy from TLS's more efficiently than the modes with $\mathbf{q} \neq 0$. This synchronizes the system and maximizes the radiation. (2) The synchronized mode has larger radiative losses and larger $\tau_{a\_eff}^{-1}$, which desynchronizes the system and minimizes radiation.

The first mechanism is dominant for our, quite realistic, parameters. Hence, the $\mathbf{q}=0$ mode provides minimum for $D_{th}(\mathbf{q})$ and wins the competition. This makes the spaser array self-synchronize.

For a smaller value of the interaction constant Ω_{R1}, the synchronization state is destroyed. The surviving mode becomes the one with $\mathbf{q} \neq 0$. In this case, the second mechanism determines the winning mode as the one with lower radiation losses [13].

The key role of the interaction of a TLS with neighboring NPs in the effect of synchronization is confirmed by our computer simulation of Eqs. (2)–(4). If $\Omega_{R1}=0$, almost ideally synchronized state turns into a state with the phase distribution that corresponds to absence of synchronization shown in Fig. 3.

The considered mechanism of synchronization is principally different from the one studied by Dicke [33]. Indeed, in our case, the synchronization occurs due to the near-field dipole-dipole interaction of the spasers, which destroys Dicke's synchronization.

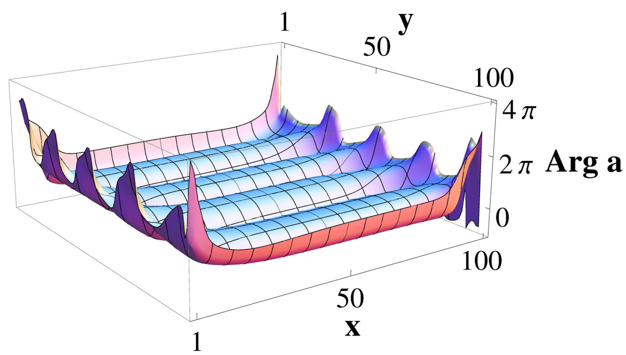

Fig. 3. Phase distribution of plasmon oscillations in the 100 × 100 spaser array with $\Omega_{R1}=0$.

3. Superradiance of the spaser array

The ability of the spaser array to self-synchronize solves the problem of the radiation extraction from spasers. In a synchronized array, interaction given by Eq. (1) forces spasers to emit radiation. This phenomenon is more evident for a system of a small size, $k_0R \ll 1$, for which Eq. (1) becomes

$$\Omega(\mathbf{e}R) \approx \tau_R^{-1}\left(\frac{3}{2}\frac{3(\mathbf{e}\cdot\mathbf{e}_x)^2-1}{(k_0R)^3}+i\right) \tag{6}$$

with $\operatorname{Im}\Omega(\mathbf{e}R)\approx\tau_R^{-1}$. When all the dipoles oscillate with the same phase and amplitude, the last term in Eq. (2) can be split into two parts:

$$i\sum_{\mathbf{m}\neq\mathbf{n}}\Omega_{\mathbf{n}-\mathbf{m}}a_{\mathbf{m}} \approx ia_{\mathbf{n}}\sum_{\mathbf{m}\neq\mathbf{n}}\operatorname{Re}\Omega_{\mathbf{n}-\mathbf{m}} - a_{\mathbf{n}}(N-1)\tau_R^{-1} \tag{7}$$

The second term at the right hand side of Eq. (7) leads to an increase in the relaxation rate of the **n**-th plasmon by the factor of $(N-1)\tau_R^{-1}$. As a result, the effective relaxation rate of a plasmon becomes $\tau_J^{-1}+N\tau_R^{-1}$. Equation (2) can now be rewritten as

$$\dot{a}_{\mathbf{n}}+\left(\tau_J^{-1}+N\tau_R^{-1}\right)a_{\mathbf{n}} = -i\Omega_R\sigma_{\mathbf{n}} - i\Omega_{R1}\sum_{|\mathbf{m}-\mathbf{n}|=1}\sigma_{\mathbf{m}} + i\operatorname{Re}\sum_{\mathbf{m}\neq\mathbf{n}}\Omega_{\mathbf{n}-\mathbf{m}}a_{\mathbf{m}} \tag{8}$$

Thus, all plasmonic NPs, in the area, which size is much smaller than the wavelength, contribute equally to the radiation rate giving an effective radiation rate of $\tau_{R\_eff}^{-1}=N\tau_R^{-1}$. The total intensity of radiation of N NPs is then proportional to N^2, which is characteristic for superradiance [34–36]. Let us note that to obtain this dependence we use the full dipole field, Eq. (1), including the retardation effects. Hence, all terms in Eq. (1) are required to correctly account for radiative damping in the dipole (spaser) array.

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The radiation power of the array can be evaluated by using the energy balance equation, which follows from Eq. (2) for the stationary regime:

$$\sum_{\mathbf{n}}\left[\Omega_R \operatorname{Im}\left(a_{\mathbf{n}}^{*}\sigma_{\mathbf{n}}\right)+\Omega_{R1}\sum_{|\mathbf{m}-\mathbf{n}|=1}\operatorname{Im}\left(a_{\mathbf{n}}^{*}\sigma_{\mathbf{m}}\right)\right]=\tau_J^{-1}\sum_{\mathbf{n}}\left|a_{\mathbf{n}}\right|^2+\sum_{\mathbf{n},\mathbf{m}}\operatorname{Im}\left(\Omega_{\mathbf{n}-\mathbf{m}}\right)\operatorname{Re}\left(a_{\mathbf{n}}^{*}a_{\mathbf{m}}\right). \quad (9)$$

According to Eq. (6), $\operatorname{Im}\Omega(\mathbf{e}R)\to\tau_R^{-1}$ for $R\to 0$. Therefore, the term with $\mathbf{n}=\mathbf{m}$ in the sum $\sum_{\mathbf{n},\mathbf{m}}$, can be evaluated with the account of $\operatorname{Im}\Omega_{\mathbf{n}-\mathbf{m}}=\tau_R^{-1}$. The left-hand side of Eq. (9) is proportional to the power supplied into the system by TLS's. The first term in the right hand side corresponds to Joule losses. It can be shown that the second term, up to a factor independent of the dipole amplitudes, equals the radiation power I,

$$I=\frac{m\omega^2}{e^2}\sum_{\mathbf{n},\mathbf{m}}\operatorname{Im}\left(\Omega_{\mathbf{n}-\mathbf{m}}\right)\operatorname{Re}\left(a_{\mathbf{n}}^{*}a_{\mathbf{m}}\right). \quad (10)$$

This term normalized by the radiation power of a single spaser is shown in Fig. 4.

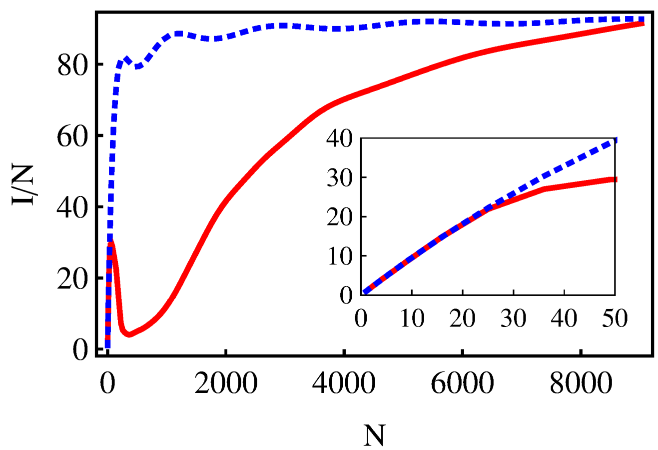

Fig. 4. The integral intensity of radiation per spaser for the array with calculated amplitude distribution (solid line) and the array of ideally synchronized dipoles (dashed line) as a function of the array size. The dependence for a small number of spasers is magnified in the inset.

As long as the array size is smaller than the half-wavelength ($N < 100$ for our parameters), all spasers are synchronized and oscillate in phase [Fig. 2(a)]. In this case, as one can see in Fig. 4, intensity growth with the array size is characteristic for superradiance, $I/N \propto N$. Also, for a small array, radiation from the array nearly coincides with the radiation of a system of synchronized classical dipoles with uniform amplitudes over the array equal to the average amplitude of a spaser $\langle a_{\mathbf{n}}\rangle$ (see inset in Fig. 4). The radiated intensity drops as the array size increases. The reason for this is a decrease of an effective system size due to boundary effects [see Fig. 2(b)]. An increase of the array size to $L \gg \lambda$ leads to synchronization of spasers for the most of the array, as shown in Fig. 2(b). Now, radiation per spaser of the array tends to catch up with the radiation of the ideally synchronized system ($N > 5000$ in Fig. 4). Due to destructive interference of radiation emitted by different parts of a large array, radiations of both spaser and ideally synchronized dipole arrays saturate, so that total radiation becomes proportional to the array size, $I \propto N$.

The important quantity for applications is the intensity per solid angle radiated in the direction normal to the array plane. This is the quantity measured by a detector of a small size.

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We calculate this intensity considering the spaser array as an antenna. The distribution of power radiated by the antenna into a solid angle $d\Omega$ in the direction of a unit vector $\mathbf{e}$, $I_\Omega(\mathbf{e})$, is referred to as the radiation pattern and can be determined by the Fourier transform of the field distribution in the antenna opening [37]. In our case, this is the dipole moment distribution in the array. Thus, the radiation pattern is equal to

$$I_\Omega(\mathbf{e}) = I_\Omega^{(0)}(\mathbf{e})\left|\sum_{\mathbf{n}} a_{\mathbf{n}} \exp\left(-ik_0 \mathbf{e}\cdot\mathbf{r}_{\mathbf{n}}\right)\right|^2, \tag{11}$$

where $a_{\mathbf{n}}$ is the stationary solution of Eqs. (2)–(4), $\mathbf{r}_{\mathbf{n}}$ is the position vector of the $\mathbf{n}$-th dipole in the array and $I_\Omega^{(0)}(\mathbf{e}) = (8\pi)^{-1} ck_0^4 \left|\mathbf{e}\times\mathbf{e}_x\right|^2$ is the radiation pattern for a unitary dipole. The integration of Eq. (11) over directions of $\mathbf{e}$ gives Eq. (10). For the normal direction, $I_\Omega^{(0)}(\mathbf{e})$ is just the sum $I_\Omega = I_\Omega^{(0)}(\mathbf{e}_z)\left|\sum_{\mathbf{n}} a_{\mathbf{n}}\right|^2$, which gives $I_\Omega / N \sim N$ for a synchronized array of any size.

Interestingly, the linear increase of I_Ω / N is determined by two effects. For small arrays, $L < \lambda$, this happens due to the growth of the integral intensity, I / N, resulting from superradiance. When $L > \lambda$, I / N saturates and the growth of I_Ω is caused by the narrowing of the radiation pattern. The latter is due to the growth of the aperture of the radiative system.

The considered mechanism of superradiance differs from ones that we are aware of because of its continuous-wave character. Dicke's superradiance of atoms [33] is a spontaneous radiation of synchronized molecules. It is of pulse nature because after photon radiation, all the atoms return to the ground state. Further pumping brings atoms back to the excited state but the next spontaneous radiation is uncorrelated in phase. The continuous-wave regime for Dicke's superradiance can be achieved only if pumping is coherent [35]. In a spaser, surface plasmons arise due to multi-quantum excitations. Therefore, after radiating a single photon, the spaser oscillations continue without phase change. Due to stimulated emission, the surface plasmon wasted for photon radiation is regenerated with the same phase. This phenomenon is closer to radiation of an array of semiconductor lasers. However, it differs from the latter because synchronization of lasers is performed by far-fields and requires additional optical equipment (lens, mirrors, etc.). In the array of spasers, the synchronization is performed by near-fields automatically. This kind of synchronization turns out to be stable; it is not destroyed by interference in the far-field and by effects of retardation. As a consequence, the synchronization of 2D array of spasers is not constrained by the array size and superradiance occurs even for large apertures.

The considered effects are rather robust. The presence of disorder affects both superradiance and synchronization of the spaser arrays. A small disorder only slightly suppress superradiance leading to an incoherent radiation component, which is proportional to the square of the spaser density fluctuation, $\left\langle \Delta n^2 \right\rangle$. This creates effective loss in the coherent component. Our computer simulations show that the synchronization is stable against disorder. Because variation of the spaser positions shift the NP-QD interaction frequencies, the presence of aperiodicity is to some extent equivalent to variations of Ω_{R1} in a periodic array. Modeling of the array with randomly variable Ω_{R1} shows that the effects do not disappear up to value of mean square deviation of Ω_{R1} up to 30%.

4. Conclusions

To summarize, we propose a highly directional continuous-wave coherent light source. It is based on a 2D array of incoherently pumped spasers, which are shown to mutually

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synchronize each other. This leads to superradiance, which can increase the radiation intensity by two orders of magnitude compared to a single spaser. We hope that the intrinsic property of spasers to self-synchronize will find the applications in optical communications. In this field the use of nanolasers, because of their fast response, may be a good alternative to the conventional laser arrays.

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