



## Light Amplification and Scattering by Clusters Made of Small Active Particles: The Local Perturbation Approach

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### Author's contribution

The sole author designed, analyzed and interpreted and prepared the manuscript.

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## ABSTRACT

The light amplification by finite active media is used extensively in modern optics applications. In this paper, the light amplification and scattering by the cluster of small active particles is studied analytically and numerically with the help of the local perturbation method and phenomenological laser theory. It is shown that light amplification is possible even for one small particle, and that the amplification is more profound when the light frequency nears the frequency of the cluster's morphological resonance. Theoretical discussions are supplemented by numerical results for scattering by clusters which particles positioned at ordered and at slightly disordered positions.

Keywords: Light amplification; lasing; small particles; cluster.

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## 1 INTRODUCTION

Light amplification by stimulated emission of radiation (laser) is well known phenomenon routinely employed in solids-state, gas, and dye lasers [1]-[3]. With the development of micro and nano technology the new kind of lasers, so-called photonic crystal lasers, emerged [4]-[7]. The common peculiarity of the photonic crystal lasers is that they are made of finite number of particles or cells ordered at least in one dimension. These lasers are extremely versatile: they can be made from active host medium filled by passive scattering particles, or from active scatterers immersed into passive host medium, or both. Moreover, the particles in such lasers can move or be static. From practical point of view, the photonic crystal lasers and amplifiers can be used as light sources to compensate the optical losses in metamaterials [8]-[10].

To the author's knowledge, the light generation by a scattering medium with negative resonance absorption was initially studied in work [11] where it was shown that the lasing is possible in such medium. The scattering by individual active spheres and cylinders was studied analytically and numerically in a number of works (see, for example [12]-[16] and references therein). At the same time, the discussion of the scattering by a cluster of active particles is somewhat limited to very small clusters made of few particles [17] or to periodic structures [18]. Scattering by many active particles occurs in so-called random lasers ([19]- [25]), while analytical predictions are difficult to made for such systems due to extremely large number of particles.

Recently, the scalar wave scattering by dispersive particles was studied by using the local perturbation method (LPM) in the work [26]. In reality, however, the vector wave scattering occurs.

To the author's knowledge, the cluster amplifier made of small particles was not studied analytically and numerically in the literature before.

In this paper the light amplification and scattering by active particles with the size smaller than the incident wavelength is studied with the help of the

LPM [27]-[30] and the phenomenological laser theory [1]-[3]. As an example, two important cases are studied numerically: scattering by ordered cluster and by weakly disordered cluster. It was shown that the light amplification is possible in both cases, however it is severely affected by the size of the particles, by the concentration of the doped active atoms, by interaction between the particles, and by morphological resonances.

## 2 THE LPM FORMALISM

The formalism used in this section is described in a number of papers ([27]-[28]) and it will be briefly presented here for convenience and consistency.

Consider the cluster positioned at the origin of the coordinates and made of  $N$  identical active particles which characteristic size  $L$  is small compared to the incident wavelength  $\lambda$ . The frequency-domain fourier transform  $\tilde{\mathbf{E}}(\mathbf{r}, \omega)$  of the electric field  $\mathbf{E}(\mathbf{r}, t)$  propagating in the host medium filled with the particles is described by the following equation [29]

$$(\Delta - \nabla \otimes \nabla + k^2) \tilde{\mathbf{E}}(\mathbf{r}, \omega) + \frac{k^2}{\varepsilon_h} \sum_{n=1}^N f(\mathbf{r} - \mathbf{r}_n) (\varepsilon_{sc,n} - \varepsilon_h) \tilde{\mathbf{E}}(\mathbf{r}_n, \omega) = \mathbf{S}(\mathbf{r}), \quad (2.1)$$

where  $\mathbf{r}$  and  $\mathbf{r}_n$  are the radius vectors of the observer and the  $n$ -th particle respectively, and

$$k \equiv \frac{2\pi}{\lambda} = \frac{\omega}{c} \sqrt{\varepsilon_h}, \quad f(\mathbf{r} - \mathbf{r}_n) \equiv \begin{cases} 1, & \mathbf{r} \in V_n \\ 0, & \mathbf{r} \notin V_n \end{cases}. \quad (2.2)$$

Here  $\Delta$  and  $\nabla$  are the Laplacian and nabla operators,  $\otimes$  defines tensor product,  $k$  is a wave number in the host medium ( $\omega$  is the angular frequency and  $c$  is the speed of light in vacuum),  $\varepsilon_{sc,n}$  and  $\varepsilon_h$  are the relative (in respect to vacuum) permittivities of the  $n$ -th particle and the host medium respectively,  $f$  is the function describing the shape of the scatterers,  $V_n$  is the volume of the  $n$ -th particle, and  $\mathbf{S}$  is the source of the field. The permittivity  $\varepsilon_{sc,n}$  of the active particles is, in principle, a complex function depending on the electric field  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega)$  inside the particle, the frequency  $\omega$ , and other parameters. We will discuss this topic in greater detail in the next section.

It should be noted, that the equation (2.1) is an approximate one and it is correct only when the small scatterers ( $kL \ll 1$ ) are considered.

The solution of the equation (2.1) can be expressed in the form

$$\tilde{\mathbf{E}}(\mathbf{r}, \omega) \equiv \tilde{\mathbf{E}}_{in}(\mathbf{r}, \omega) + \tilde{\mathbf{E}}_{sc}(\mathbf{r}, \omega), \quad (2.3)$$

where the scattered field  $\tilde{\mathbf{E}}_{sc}$  is

$$\tilde{\mathbf{E}}_{sc}(\mathbf{r}, \omega) = \frac{k^2}{\varepsilon_h} \left( \hat{\mathbf{I}} + \frac{\nabla \otimes \nabla}{k^2} \right) \times \sum_{n=1}^N (\varepsilon_{sc,n} - \varepsilon_h) \tilde{\mathbf{E}}(\mathbf{r}_n, \omega) \Phi_n(\mathbf{r}), \quad (2.4)$$

and

$$\Phi_n(\mathbf{r}) \equiv \int_{-\infty}^{\infty} \frac{\tilde{f}(\mathbf{q}) e^{i\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}_n)}}{(q^2 - k^2)} d\mathbf{q}$$

$$\tilde{f}(\mathbf{q}) \equiv \frac{1}{8\pi^3} \int_{-\infty}^{\infty} f(\mathbf{r}) e^{-i\mathbf{q} \cdot \mathbf{r}} d\mathbf{r}.$$

Here  $\hat{\mathbf{I}}$  is the  $3 \times 3$  unitary tensor in polarization space and  $\mathbf{r}_n$  is the radius vector of the  $n$ -th particle. The function  $\tilde{f}$  is the Fourier transform of the function  $f$ . The incident field  $\tilde{\mathbf{E}}_{in}$  is created by the source  $\mathbf{S}$  in the host medium (more information can be found in [31]).

The formula (2.4) is rather general one and it describes the field scattered by the cluster made of small particles of arbitrary form. The resonance properties and interference between the scatterers are taken into account by the fields  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega)$  inside the particles. The fields  $\tilde{\mathbf{E}}(\mathbf{r}_n)$  should be found by solving the system of  $3N$  linear equations obtained by substituting  $\mathbf{r} = \mathbf{r}_n$  into (2.3).

The scattered field (2.4) can be simplified when the observer is outside of the cluster, such that  $\mathbf{r} \neq \mathbf{r}_n$ . In this case the integrals (2.5) can be calculated explicitly and the scattered field (2.4) can be presented in the following form

$$\tilde{\mathbf{E}}_{sc}(\mathbf{r}, \omega) = \frac{k^2 V}{4\pi\varepsilon_h} \left( \hat{\mathbf{I}} + \frac{\nabla \otimes \nabla}{k^2} \right) \times \sum_{n=1}^N (\varepsilon_{sc,n} - \varepsilon_h) \tilde{\mathbf{E}}(\mathbf{r}_n, \omega) \frac{e^{ikR_n}}{R_n}, \quad (2.5)$$

where

$$R_n \equiv |\mathbf{r} - \mathbf{r}_n|, \quad \mathbf{r} \neq \mathbf{r}_n. \quad (2.6)$$

Here  $R_n$  is the distance between the observation point  $\mathbf{r}$  and the radius vector  $\mathbf{r}_n$  of the  $n$ -th scatterer,  $V$  is the scatterer's volume.

In many practical cases the distance between the cluster and the observer is much larger than the size of the cluster, i.e.  $|\mathbf{r}| \gg \max(|\mathbf{r}_n|)$ , and in addition, the condition  $k|\mathbf{r}| \gg 1$  is satisfied. In this case the field (2.5) can be simplified and it can be rewritten in the following form

$$\tilde{\mathbf{E}}_{sc}(\mathbf{r}, \omega) = \frac{k^2 V}{4\pi\varepsilon_h} \frac{e^{ikr}}{r} \left( \hat{\mathbf{I}} - \mathbf{l} \otimes \mathbf{l} \right) \times \sum_{n=1}^N (\varepsilon_{sc,n} - \varepsilon_h) \tilde{\mathbf{E}}(\mathbf{r}_n, \omega) e^{-ik\mathbf{l} \cdot \mathbf{r}_n}, \quad (2.7)$$

where

$$\mathbf{l} \equiv \mathbf{r}/r, \quad r \equiv |\mathbf{r}| \gg \max(|\mathbf{r}_n|), \quad kr \gg 1. \quad (2.8)$$

We note that the formula (2.7) is the final expression for the field scattered by the cluster of small particles, and it will be used in the following discussion.

### 3 THE PERMITTIVITY OF THE ACTIVE PARTICLES: STEADY STATE SOLUTION

In this section we will study the permittivity  $\varepsilon_{sc,n}$  of the active particles which characteristic size is much smaller than the incident wavelength ( $kL \ll 1$ ). The particles are active due to homogeneously distributed doped active atoms. The density of the active atoms is  $M$ . We note that the permittivity  $\varepsilon_{sc,n}$  in formula (2.7) can be complex number with negative or positive imaginary part, and in this case one can study wave scattering with gain or loss in active media [12]-[14]. The problem with such approach is two-fold. First, the value of the imaginary part is not related to the properties of the actual medium, and second, the permittivity is the same for all particles, that is not true for real systems. As it was suggested in [15], such approach is valid for quantitative estimations of lasers before threshold. For more accurate investigation one should use rigorous methods taking into account atomic transitions and pump dissipation.

It is important to acknowledge that when the

particles are small the number of active atoms in upper state is constant within the particle, while this number can be different for other particles. We assume that all the scatterers have the same density  $M$  of the active atoms. It is not limiting assumption, and it is very convenient one. We also assume that the amplifier is at steady state, so the densities of the active atoms in the upper and low states are time independent. We approximate the active atoms as two-level systems excited by the optical pump with the frequency  $\omega_p$  and relaxing with the wide range of the frequencies  $\omega_j$  ( $j = 1, 2, \dots, p, \dots, N_\omega$ ). We can present the permittivity  $\varepsilon_{sc,n}$  of the small particles in the following form

$$\varepsilon_{sc,n} \equiv \varepsilon_{sc,n}^0 + \varepsilon'_{sc,n}(\mathbf{r}_n, \omega), \quad (3.1)$$

where  $\varepsilon_{sc,n}^0$  is the permittivity of the  $n$ -th scatterer without the active atoms and  $\varepsilon'_{sc,n}$  is the permittivity of the  $n$ -th particle due to the presence of the active atoms. The latter can be expressed in the following form [1]

$$\varepsilon'_{sc,n}(\mathbf{r}_n, \omega) = i \frac{c}{\omega} \sqrt{\varepsilon_{sc,n}^0} (M_L(\mathbf{r}_n) \sigma_a(\omega) - M_U(\mathbf{r}_n) \sigma_e(\omega)), \quad (3.2)$$

where the emission and the absorption cross sections of the active medium respectively are

$$\sigma_e(\omega) \equiv \frac{2\pi e^2 \gamma_e \alpha}{mc \sqrt{\varepsilon_{sc,n}^0} ((\omega - \Omega_e)^2 + \gamma_e^2)}, \quad (3.3)$$

$$\sigma_a(\omega) \equiv \frac{2\pi e^2 \gamma_a \alpha}{mc \sqrt{\varepsilon_{sc,n}^0} ((\omega - \Omega_a)^2 + \gamma_a^2)}, \quad (3.4)$$

and the density of the active atoms is

$$M = M_U(\mathbf{r}_n) + M_L(\mathbf{r}_n). \quad (3.5)$$

Here  $M_U$  and  $M_L$  are the densities of the active atoms in the upper and low states respectively, and  $M$  is the total density of the active atoms. The frequencies  $\Omega_a$  and  $\Omega_e$  are the resonance frequencies for absorption and emission respectively, and  $\alpha$  is the oscillator strength. The frequencies  $\gamma_a$  and  $\gamma_e$  are the dipole relaxation frequencies for the absorption and the emission respectively (typical values are about  $10^{13}$  Hz [32]), and  $e$  and  $m$  are the electron's charge and mass respectively.

The importance of the formula (3.2) is that it allows us to use experimentally measured emission and absorption cross sections ( $\sigma_e$  and  $\sigma_a$ ) when the density  $M_U$  is known. This approach will be used in the following section

where the results of the numerical calculations will be presented. It should be noted also that we neglected by the real part of the permittivity  $\varepsilon'_{sc,n}$  because it is small compared to the optical contrast  $\varepsilon_{sc,n}^0 - \varepsilon_h$ , especially near the resonance. We note that the formulae (3.3) were calculated assuming that  $\omega \sim \Omega_e, \Omega_a$ .

### 3.1 Rate Equation Approximation

The expressions (3.1) and (3.2) for the permittivity of the particles suggest that the densities  $M_U$  and  $M_L$  should be known. We can find them by using the rate equation approximation (see for example, [1]).

When the pulse duration exceeds the dipole relaxation time (typically  $10^{-13}$  s), the density  $M_U$  of the upper level atoms can be calculated by using the rate equation approximation in which the dopants respond so fast that the induced polarization follows the optical field adiabatically [32]. We use the following rate equation [33]

$$\frac{dM_U(\mathbf{r}_n)}{dt} = \sum_j^{N_\omega} I_j(\mathbf{r}_n, \omega_j) \times [M_L(\mathbf{r}_n) \sigma_{aj} - M_U(\mathbf{r}_n) \sigma_{ej}] - M_U(\mathbf{r}_n) / \tau, \quad (3.6)$$

where

$$\sigma_{aj} \equiv \sigma_a(\omega_j), \quad \sigma_{ej} \equiv \sigma_e(\omega_j), \quad (3.7)$$

$$I_j(\mathbf{r}_n, \omega_j) \equiv \frac{c \varepsilon_h}{4\pi \hbar \omega_j} |\tilde{\mathbf{E}}(\mathbf{r}_n, \omega_j)|^2 \Delta\omega^2. \quad (3.8)$$

Here  $\sigma_{aj}$  and  $\sigma_{ej}$  are the absorption and the emission cross sections at the frequency  $\omega_j$  respectively, and  $I_j$  is the flux of photons at the frequency  $\omega_j$ , and the  $|\dots|$  brackets denote the absolute value. The parameter  $\tau$  is the relaxation time of the excited atom (typically  $10^{-3} - 10^{-6}$  s [32]),  $\hbar$  is the reduced Planck constant,  $\Delta\omega = \omega_{j+1} - \omega_j$  is the frequency bin, and  $N_\omega$  is the total number of the frequency bins.

Since we consider only steady state solutions when

$$\frac{dM_U(\mathbf{r}_n)}{dt} = 0, \quad (3.9)$$

the solution of the rate equation (3.6) is

$$M_U(\mathbf{r}_n) = \frac{M \sum_j^{N_\omega} I_j(\mathbf{r}_n, \omega_j) \sigma_{aj}}{1/\tau + \sum_j^{N_\omega} I_j(\mathbf{r}_n, \omega_j) (\sigma_{aj} + \sigma_{ej})}, \quad (3.10)$$

and the formula (3.2) for the permittivity  $\varepsilon'_{sc,n}$  has the form

$$\varepsilon'_{sc,n}(\mathbf{r}_n, \omega) \equiv i \frac{cM}{\omega} \sqrt{\varepsilon_{sc,n}^0} \frac{\sigma_a(\omega)/\tau + \sum_j^{N\omega} I_j(\mathbf{r}_n, \omega_j) [\sigma_a(\omega)\sigma_{ej} - \sigma_e(\omega)\sigma_{aj}]}{1/\tau + \sum_j^{N\omega} I_j(\mathbf{r}_n, \omega_j)(\sigma_{aj} + \sigma_{ej})}, \quad (3.11)$$

The formula (3.11) is the main result of this section and it shows that the permittivity of the small active scatterer is a complex function of the intensities of the fields  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega)$  inside the particles, frequency  $\omega$ , position of the particles  $\mathbf{r}_n$ , and absorption and emission cross sections  $\sigma_{aj}$  and  $\sigma_{ej}$ . When the light intensity changes (due to increased reflection from the scatterer's boundaries or due to decreased amount of the scattered light from all other particles, for example), it will affect the permittivity  $\varepsilon'_{sc,n}$  (3.11). We note that the photon fluxes  $I_j(\mathbf{r}_n, \omega_j)$  should be found by solving the system of nonlinear equations with respect to the fields  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega_j)$  inside the particles.

### 3.2 Usage of Weak Scattering

As the expression (3.11) for the permittivity of the small active particles suggests, the field scattered by the cluster should be found by solving the system of nonlinear equations with respect to the fields  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega)$  inside the particles. However, this tedious task is essentially simplified in our case, because we consider the scattering by small particles. As the result, the scattered field in the sum (3.11) is small compared to the incident one, and it is natural to use perturbation theory where the intensities  $I_j$  are small compared to the pump intensity  $I_p$ . We distinct pump and signal (anything but a pump) frequencies  $\omega_p$ , and  $\omega_j$  respectively.

When the signal is so small that

$$1/\tau + I_p(\mathbf{r}_n, \omega_p)(\sigma_{ap} + \sigma_{ep}) \gg \sum_{j \neq p}^{N\omega} I_j(\mathbf{r}_n, \omega_j)(\sigma_{aj} + \sigma_{ej}), \quad (3.12)$$

$$\sigma_{ap} \equiv \sigma_a(\omega_p), \quad \sigma_{ep} \equiv \sigma_e(\omega_p), \quad (3.13)$$

the permittivity (3.11) can be expressed in the approximate form

$$\varepsilon'_{sc,n}(\mathbf{r}_n, \omega) = -i \frac{cM}{\omega} \sqrt{\varepsilon_{sc,n}^0} \times \frac{\sigma_a(\omega)/\tau + I_p(\mathbf{r}_n, \omega_p) [\sigma_a(\omega)\sigma_{ep} - \sigma_e(\omega)\sigma_{ap}]}{1/\tau + I_p(\mathbf{r}_n, \omega_p)(\sigma_{ap} + \sigma_{ep})}. \quad (3.14)$$

For some estimations it can be sufficient to use the permittivity (3.14), while for rigorous numerical calculations one can apply general formula (3.11) where fluxes  $I_j$  are found by using the method of successive approximations.

When emission and absorption spectra are very distinct and separated such that  $\sigma_{ep} = \sigma_{as} = 0$ , we can simplify the formula (3.14) for the pump and the signal respectively

$$\begin{aligned} \varepsilon'_{sc,n}(\mathbf{r}_n, \omega_p) &= i \frac{cM}{\omega_p} \sqrt{\varepsilon_{sc,n}^0} \times \\ &\frac{\sigma_{ap}/\tau + I_s(\mathbf{r}_n)\sigma_{ap}\sigma_{es}}{1/\tau + I_p(\mathbf{r}_n)\sigma_{ap}}, \\ \varepsilon'_{sc,n}(\mathbf{r}_n, \omega_s) &= -i \frac{cM}{\omega_s} \sqrt{\varepsilon_{sc,n}^0} \times \\ &\frac{I_p(\mathbf{r}_n)\sigma_{es}\sigma_{ap}}{1/\tau + I_p(\mathbf{r}_n)\sigma_{ap}}. \end{aligned} \quad (3.15)$$

The important feature of the formulae (3.15) is the sign flip: for the pump it is positive (the pump is absorbed) and for the signal it is negative (the signal is amplified).

## 4 INTENSITY OF THE SCATTERED FIELD AND THE LIGHT AMPLIFICATION

We define the intensity of the scattered field as  $I_{sc} \equiv |E_{sc}|^2$ , and by using the formula (2.7) we can present the intensity in the following form

$$I_{sc}(\mathbf{r}, \omega) = \frac{k^4 V^2}{16\pi^2 \varepsilon_h^2 r^2} \left| \left( \hat{I} - \mathbf{1} \otimes \mathbf{1} \right) \sum_{n=1}^N (\varepsilon_{sc,n} - \varepsilon_h) \tilde{\mathbf{E}}(\mathbf{r}_n, \omega) e^{-ik\mathbf{l} \cdot \mathbf{r}_n} \right|^2, \quad (4.1)$$

where

$$l \equiv \mathbf{r}/r, \quad r \equiv |\mathbf{r}| \gg \max(|\mathbf{r}_n|), \quad kr \gg 1, \quad (4.2)$$

and the permittivity  $\varepsilon_{sc,n}$  is described by the expression (3.11) or by (3.14). The expression (4.1) suggests that the light amplification (related to the imaginary part of the permittivity  $\varepsilon_{sc,n}$ ) is due to step wise amplification inside each active particle, and it is coded in the fields  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega)$ . Below we consider the fields  $\tilde{\mathbf{E}}(\mathbf{r}_n, \omega)$  in grater detail for one active particle.

#### 4.1 The Light Amplification by Small Active Sphere

Consider the light amplification by small active sphere. In this case the intensity of the scattered field is described by the expression (4.1) where the field  $\tilde{\mathbf{E}}(\mathbf{r}_1, \omega)$  has the following form [34]

$$\tilde{\mathbf{E}}(\mathbf{r}_1, \omega) = \frac{\tilde{\mathbf{E}}_{in}(\mathbf{r}_1, \omega)}{D(\omega)}, \quad (4.3)$$

and the denominator  $D$  is

$$D(\omega) = 1 + \frac{(\varepsilon_{sc,1} - \varepsilon_h)}{3\varepsilon_h} \left( 1 - L^2 k^2 - i \frac{2}{3} L^3 k^3 \right). \quad (4.4)$$

The resonance frequency  $\omega_r$  is found from the following equation

$$\text{Re } D(\omega_r) = 0, \quad (4.5)$$

and the resonance width  $\xi$  is defined as

$$\xi \equiv \left| \frac{\text{Im } D(\omega)}{\frac{\partial \text{Re } D(\omega)}{\partial \omega}} \right|_{\omega=\omega_r}. \quad (4.6)$$

In accordance with formulae (4.5) and (4.6) the resonance width  $\xi$  and the resonance frequency  $\omega_r$  respectively are

$$\xi = \frac{\sqrt{\varepsilon_h} L \omega_r^2}{3c} + \frac{3c^2 \text{Im}(\varepsilon_{sc,1} - \varepsilon_h)}{2\omega_r L^2 \text{Re}^2(\varepsilon_{sc,1} - \varepsilon_h)}, \quad (4.7)$$

and

$$\omega_r = \frac{c\sqrt{3}}{L\sqrt{\text{Re}(\varepsilon_{sc,1} - \varepsilon_h)}} \left( 1 + \frac{\text{Re}(\varepsilon_{sc,1} - \varepsilon_h)}{3\varepsilon_h} \right)^{1/2}. \quad (4.8)$$

When the permittivity  $\varepsilon_{sc,1}$  of the particle is real, the resonance width is defined by the first term in Eq. (4.7). When the imaginary part of the

permittivity  $\varepsilon_{sc,1}$  is taken into account and it is negative or positive, the resonance width can be slightly decreased or increased respectively. The formulae (4.7) and (3.2) suggest that when  $M_U \sigma_e < M_L \sigma_a$ , the resonance width increases (with respect to the one in the passive medium) and it decreases when  $M_U \sigma_e > M_L \sigma_a$ .

This decrease (or increase) corresponds to effective gain (or loss) of the field scattered by the particle at the resonance frequency.

We note that similar conclusions can be drawn for the clusters consisting of two and more particles, while the analytical investigation of such systems is much more complicated.

## 5 TWO NUMERICAL EXAMPLES: LIGHT AMPLIFICATION BY ORDERED AND BY WEAKLY DISORDERED SPHERICAL CLUSTER

In this section the light amplification and scattering by active clusters (clusters made of an active material) is studied numerically. The normalized intensity  $\mathfrak{R}$  of the scattered field is calculated. The normalized intensity is defined as

$$\mathfrak{R}(\mathbf{r}, \omega) \equiv I_{sc}(\mathbf{r}, \omega) / I_{inc}(0, \omega). \quad (5.1)$$

All the used clusters are 3D structures (as shown, for example, on the Fig. 1) made of cubes doped with  $Yb^{3+}$  (active material). The absorption and emission cross sections are taken from [35] and the other parameters are from [33]. The incident field is generated by the point source described by the following formula

$$\tilde{\mathbf{E}}_{in}(\mathbf{r}, \omega) = \mathbf{E}_0 \frac{e^{ik(\mathbf{r}-\mathbf{r}_s)}}{4\pi |\mathbf{r}-\mathbf{r}_s|}, \quad k |\mathbf{r}-\mathbf{r}_s| \gg 1, \quad (5.2)$$

where the field  $\mathbf{E}_0$  is polarized along  $z$  direction. The source is positioned at  $\mathbf{r}_s = \{1, 0, 0\}$  and the center of the cluster is positioned at the origin of coordinates  $\mathbf{r} = \{0, 0, 0\}$ . The pump wavelength is selected to be  $\lambda_p = 911$  nm, and at this specific wavelength the field  $\mathbf{E}_0$  is artificially increased

by several orders of magnitude to simulate the pump. Finally, the perturbation theory is used to calculate the density of the upper level atoms.

### 5.1 The amplification and scattering by ordered spherical cluster

Consider the scattering by the spherical cluster made of the small active cubes organized into simple cubic lattice. The size of the cubes in the cluster is  $L = 25$  nm, the period is  $d = 2.2L$ , the radius of the cluster is  $5d$ . The permittivity of the scatterers is  $\varepsilon_{sc,n} = 4.2466$  and the permittivity of the host medium is  $\varepsilon_h = 1$ . Such combination of the cluster's permittivity and dimensions creates optical resonance of the passive cluster (cluster without any active material) near  $kL = 0.16$  ( $\lambda = 982$  nm).

The Fig. 2 shows the normalized intensity  $\mathfrak{R}$  of the scattered field for the active clusters with  $Yb^{3+}$  density  $M = 5 * 10^{27} \text{ m}^{-3}$ ,  $M = 2.5 * 10^{27} \text{ m}^{-3}$ , and  $M = 10^{27} \text{ m}^{-3}$ . For comparison, the intensity of the scattered field from the passive cluster with  $Yb^{3+}$  density  $M = 0 \text{ m}^{-3}$  is also presented.

The Fig. 2 shows that light is significantly amplified at the selected frequency  $kL = 0.16$  for the doping exceeding  $M = 10^{27} \text{ m}^{-3}$ . For relatively low  $M$ , when the doping increases 2.5 times (from  $M = 10^{27} \text{ m}^{-3}$  to  $M = 2.5 * 10^{27} \text{ m}^{-3}$ ), the intensity grows only 1.6 times. However, for relatively higher  $M$ , when the doping increases only 2 times (from  $M = 2.5 * 10^{27} \text{ m}^{-3}$  to  $M = 5 * 10^{27} \text{ m}^{-3}$ ), the intensity grows 2.25 times. Additional simulations (not presented here) suggest that at even higher doping, the light amplification increases several orders of magnitude while the doping increases only few times. It is important to realize that by using correct design, the light amplification can be produced by cluster made of few hundreds of small active particles. This phenomenon opens a way for novel applications in nanomedicine, nanooptics, and security.

We realize that the size of the cluster used in our calculation is too small to made a lasing with the conventional values of the doping  $M$  ( $M \sim 10^{25} \text{ m}^{-3}$ ) and that is why we have presented only results with  $M$  smaller than physically realistic limit ( $\sim 10^{28} \text{ m}^{-3}$ ).

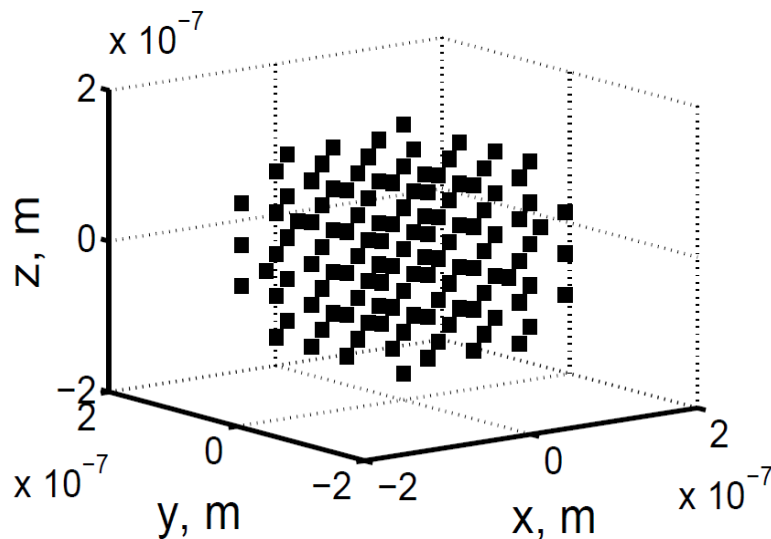


Fig. 1. The schematic representation of the spherical cluster made of 123 small cubes. The period of the cluster is  $d=2.2L$ , and the characteristic size of the cubes is  $L = 25$  nm.

## 5.2 The Amplification and Scattering by Spherical Cluster with Weak positional disorder

In this subsection we consider the light amplification and scattering by the active cluster which particles are randomly positioned near predefined positions. The predefined positions are the nodes of the cubic lattice with the period of  $d = 2.2L$ , and the particles are positioned not further than  $0.1L$  from the nodes to avoid a collision. We note that the distance  $0.1L$  is actually 2.5 nm, that is much less than the size of the particle  $L$ , and that is why we call this cluster weakly disordered one.

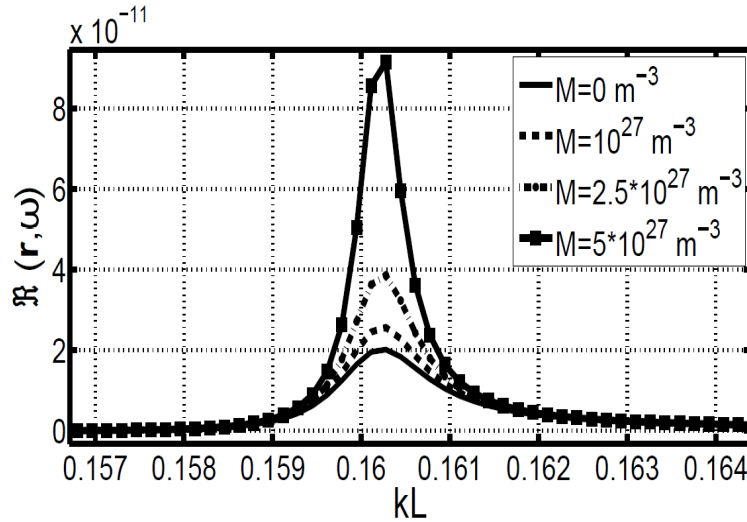
We note that the density of the active atoms in the cluster is  $M = 5 * 10^{27} \text{ m}^{-3}$ . The permittivity of the particles and the host medium is  $\varepsilon_{sc,n} = 4.2466$  and  $\varepsilon_h = 1$  respectively, the characteristic size of the cubes is  $L = 25 \text{ nm}$ , and the total number of the particles in the cluster is  $N = 515$ .

The results of the calculations are presented on the Fig. 3. The figure shows the results of two simulation runs for the disordered cluster and one

result for ordered cluster for comparison.

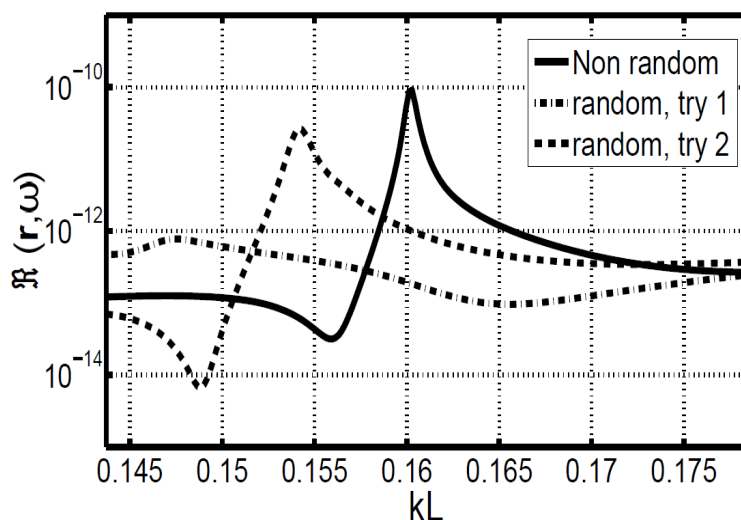
The figure suggests that despite weak positional disorder the random positioning significantly influences the scattering and amplification by the cluster. In our particular case, two scattering peaks compete with each other: one near  $kL = 0.154$  ( $\lambda = 1020 \text{ nm}$ ) and another is near  $kL = 0.147$  ( $\lambda = 1069 \text{ nm}$ ). This feature is probably related to the emission spectrum of active material ( $\text{Yb}^{3+}$ ) which has two crests: very narrow one near 980 nm and broad one near 1030 nm.

The random amplifier differs from the nonrandom one in a number of ways. The first difference is the absence of well defined boundaries, which in turn, govern the morphological resonances. Thus, the amplification (or lasing) can be at several frequencies simultaneously. The second difference is the random structure inside the cluster, affecting the interaction between the particles and the total gain as the result. It is important to realize that even small randomness will significantly affect the light amplification in the cluster made of small active particles, and that proposed theory can explain and predict the related effects.



**Fig. 2.** The normalized intensity of the scattered field  $\Re$  versus normalized frequency  $kL$  for the spherical cluster made of small cubes with different density of the active atoms  $M$ . The period of the cluster is  $d=2.2L$ , and the permittivity of the particles and the host medium is  $\varepsilon_{sc,n} = 4.2466$  and  $\varepsilon_h = 1$  respectively, the characteristic size of the cubes is  $L = 25 \text{ nm}$ , and the total number of the particles in the cluster is  $N = 515$ .





**Fig. 3. The normalized intensity of the scattered field  $\mathfrak{R}$  versus normalized frequency  $kL$  for slightly disordered spherical cluster made of small cubes. The results of two runs are presented, and the scattering by the ordered cluster is also shown for comparison. The period of the cluster is  $d=2.2L$ , and the density of the active atoms is  $M = 5 * 10^{27} \text{ m}^{-3}$ . The permittivity of the particles and the host medium is  $\varepsilon_{sc,n} = 4.2466$  and  $\varepsilon_h = 1$  respectively, the characteristic size of the cubes is  $L = 25 \text{ nm}$ , and the total number of the particles in the cluster is  $N = 515$ .**

When the allowed distance from the particles to the nodes increases (while the period is fixed or also increases), the effective size of the cluster grows. As the result, the interaction between the particles in the cluster will, in average, drop down. In addition, the effective permittivity of the cluster will decrease, reducing the reflection of the light inward from the surface of the cluster. Under these conditions, the lasing will become much more difficult.

## 6 CONCLUSIONS

The light amplification and scattering by the cluster made of the small active particles have been studied analytically and numerically.

The permittivity of the small active particles has been calculated in steady state by using the rate equation approximation.

The light amplification has been discussed for small active particle. It has been suggested that the amplification (or loss) effectively occurs in the

active particle due to narrowing (or broadening) of the resonance.

The light scattering by the ordered and slightly disordered clusters of small active particles has been calculated numerically. The numerical simulations have been shown that the light amplification occurs near the morphological resonances which are governed by the shape of the cluster and its optical contrast. It was shown that even small randomness can significantly affect the light amplification of the cluster made of small active particles.

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## COMPETING INTERESTS

The author declares that no competing interests exist.

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