Quantum Electronics **43** (1) 63–70 (2013)

NANOCOMPOSITES

PACS numbers: 42.55.Zz; 42.25.Dd; 78.67.Bf
DOI: 10.1070/QE2013v043n01ABEH014944

Random lasing in a nanocomposite medium

S.N. Smetanin, T.T. Basiev

Abstract. **The characteristics of a random laser based on a nanocomposite medium consisting of a transparent dielectric and scattering doped nanocrystals are calculated. It is proposed to use ytterbium laser media with a high concentration of active ions as nanocrystals and to use gases, liquids, or solid dielectrics with a refractive index lower than that of nanocrystals as dielectric matrices for nanocrystals. Based on the concept of nonresonant distributed feedback due to the Rayleigh scattering, an expression is obtained for the minimum length of a nanocomposite laser medium at which the random lasing threshold is overcome. Expressions are found for the critical (maximum) and the optimal size of nanocrystals, as well as for the optimal relative refractive index of nanocomposites that corresponds not only to the maximum gain but also to the minimum of the medium threshold length at the optimal size of nanocrystals. It is shown that the optimal relative refractive index of a nanocomposite increases with increasing pump level, but is independent of the other nanocomposite parameters.**

Keywords: *random lasing, nanocomposite medium, doped nanocrystals, Rayleigh scattering.*

1. Introduction

In recent years, increasing interest is attracted to random lasers [1–9], whose feedback is based on elastic scattering of laser radiation by scattering particles. The main advantage of these lasers is a simple design since they need no cavities. The idea of random (cavity-less) lasing in a laser medium with scattering particles was proposed by Letokhov as early as in 1966 [10]. Almost at the same time, the authors of [11, 12] reported on the operation of a ruby laser with nonresonant feedback achieved by using a diffuser instead of one of the cavity mirrors. The first experiments with a mirrorless laser were performed much later, in 1986, by Markushev et al. [13] using neodymium-doped dielectric powders. Later, lasing was obtained in various randomly inhomogeneous media. Lasing in these media is usually explained by the diffusion model proposed by Letokhov [14], which is valid when scattering makes the photon free path much shorter than the dimensions of the media, i.e., when $\mu^{-1} \ll d$, where μ is the distributed scattering loss coefficient and d is the minimal size of the medium. In this case, the motion of photons is diffusive due to multiple scattering in the amplifying medium, which leads to an increase in the photon lifetime in the random laser equivalent to the existence of noncoherent feedback in the absence of cavity.

Recently [15, 16], random lasing was experimentally achieved in a fibre Raman laser with feedback due to the Rayleigh scattering from random defects of the medium. The laser operation mechanism is not diffusion since the transverse size of the fibre (d) is very small compared to the photon free path upon scattering, i.e., $\mu^{-1} \gg d$. The feedback in this laser can be explained by light backscattering in a narrow oscillation angle, which is similar to the situation in distributed feedback lasers but without resonance effects since scattering centres are distributed chaotically.

Another interesting and rapidly developing scientific field is the creation of a new class of nanocomposite media with controlled optical and laser properties. Note that nanocomposites based on active laser nanoparticles incorporated into various dielectric media have noticeable advantages. In [17, 18], it was shown that the emission characteristics of nanocrystals are considerably different from the characteristics of bulk crystals. Choosing different nanocrystals, their morphology and sizes, dopant ions and their concentration, as well as varying the volume fraction of nanocrystals in a dielectric medium and its refractive index, one can control the properties of laser nanocomposite media and improve their characteristics.

In this work, we consider the possibility of random lasing in a nanocomposite medium consisting of a transparent dielectric and scattering doped nanocrystals such that the photon free paths is longer than the transverse size of the medium ($\mu^{-1} > d$).

2. Lasing threshold condition

Let us consider a nanocomposite laser medium with the length L containing doped nanoparticles with a size much smaller than the radiation wavelength. The nanoparticles are responsible for laser amplification and determine the useful and parasitic Rayleigh scattering losses. A portion of backscattered (in a narrow angle) light creates the positive feedback for random lasing. The lasing threshold condition corresponds to the gain and loss balance during a double pass of the medium. Figure 1 shows the scheme used to determine the random lasing threshold. The laser medium is divided into layers with a thickness Δz, whose interfaces, due to the Rayleigh scattering, have the backscattering coefficient $r = \varepsilon\Delta z$, where $\varepsilon = Q\mu$ is the Rayleigh backscattering coefficient responsible for the feedback necessary for lasing and Q is the

S.N. Smetanin, T.T. Basiev A.M. Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, 119991 Moscow, Russia; e-mail: ssmetanin@bk.ru

Received 19 July 2012; revision received 3 October 2012
Kvantovaya Elektronika **43** (1) 63–70 (2013)
Translated by M.N. Basieva

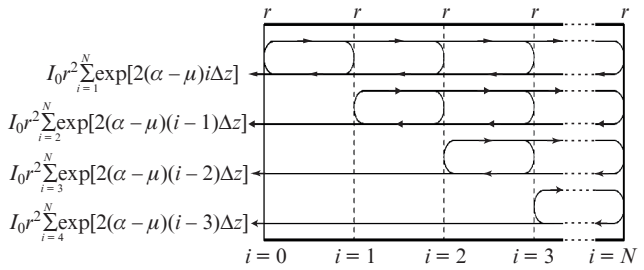

Figure 1. Scheme of determination of the random lasing threshold.

Rayleigh backscattering factor equal to the portion of laser radiation backscattered in the narrow lasing angle.

Let the seed radiation have the intensity I_0. In the case of balanced gain and loss, taking into account all reflections from the interfaces between layers, the radiation intensity after a double pass through the medium must restore its initial value I_0, i.e.,

$$I_0 r^2 \sum_{j=0}^{N-1} \sum_{i=j+1}^{N} \exp[2(\alpha-\mu)(i-j)\Delta z] = I_0, \tag{1}$$

where α is the laser gain, μ has the meaning of the Rayleigh scattering loss coefficient, and $N = L/\Delta z$ is the number of layers. Decreasing the layer thickness Δz to an infinitely small value, we obtain the integral threshold condition of random lasing in the form

$$\varepsilon^2 \int_0^L \int_z^L \exp[2(\alpha-\mu)(z'-z)]\mathrm{d}z'\mathrm{d}z = 1. \tag{2}$$

A similar equation was obtained in [16] for the gain and loss balance of random Raman fibre lasers with distributed feedback based on Rayleigh scattering from random defects of the medium.

At constant coefficients α and μ, condition (2) is integrated, and we obtain the lasing threshold condition in the form

$$\frac{\varepsilon^2}{4(\alpha-\mu)^2}\{\exp[2(\alpha-\mu)L] - 2(\alpha-\mu)L - 1\} = 1, \tag{3}$$

which is implicit with respect to the medium length L. However, in practically important cases, the medium gain is high, i.e.,

$$\exp[2(\alpha-\mu)L] \gg 2\,(\alpha-\mu)L + 1,$$

and condition (3) takes the form

$$\exp[(\alpha-\mu)L] \simeq \frac{2(\alpha-\mu)}{\varepsilon}, \tag{4}$$

which is close to the Letokhov formula $\exp(\alpha L) = \alpha/\varepsilon$ [10] at $\alpha \gg \mu, \varepsilon$. Then, we can obtain from (4) the medium length corresponding to the threshold lasing condition, which we denote as L_{th},

$$L_{\mathrm{th}} \simeq \frac{1}{\alpha-\mu}\ln\frac{2(\alpha-\mu)}{\varepsilon}. \tag{5}$$

3. Gain of nanocomposite laser media

The gain of a nanocomposite laser medium can be found by the formula [19]

$$\alpha = \sigma_{\mathrm{em}}^{\mathrm{nano}}(\lambda_{\mathrm{L}})N_2C - \sigma_{\mathrm{abs}}^{\mathrm{nano}}(\lambda_{\mathrm{L}})N_1C, \tag{6}$$

where $\sigma_{\mathrm{em,abs}}^{\mathrm{nano}}(\lambda_{\mathrm{L}})$ are the effective stimulated emission (em) and absorption (abs) cross sections of the nanocomposite medium at the laser wavelength λ_{L}, $N_{1,2}$ are the populations of the lower (1) and upper (2) laser multiplets in nanocrystals, and C is the volume fraction of nanocrystals in the nanocomposite medium. The expression found in [17, 18] for the stimulated emission cross section of a nanocomposite, in the case of a small volume fraction of nanocrystals in the nanocomposite ($C \leqslant 0.1$), is written in the form

$$\sigma_{\mathrm{em}}^{\mathrm{nano}} = \sigma_{\mathrm{em}}^{\mathrm{bulk}} n\left(\frac{3}{n^2+2}\right)^2, \tag{7}$$

where $\sigma_{\mathrm{em}}^{\mathrm{bulk}}$ is the stimulated emission cross section of the corresponding bulk crystal, $n = n_{\mathrm{crys}}/n_{\mathrm{med}}$ is the relative refractive index of the nanocomposite, n_{crys} is the refractive index of nanocrystals, and n_{med} is the refractive index of the dielectric host.

To obtain a high gain of a nanocomposite with a small volume fraction of nanocrystals, it is necessary to have a high concentration of dopant ions. Therefore, for lasing in the near IR region, it is preferable to use nanocrystals of ytterbium laser media, in which the effects of up-conversion and excited-state absorption are weak and cross-relaxation quenching is absent.

As an example of the initial material of nanocrystals, we consider ytterbium laser crystals Yb^{3+}:YAG (n_{crys} = 1.82) and Yb^{3+}:SrF_2 (n_{crys} = 1.44), while air ($n_{\mathrm{med}} \approx 1.0$), alcohol ($n_{\mathrm{med}} \approx 1.36$), and solid dielectrics ($n_{\mathrm{med}} > 1.4$) are used as dielectric hosts. It should be noted that the laser properties of the chosen ytterbium media have some specific features [20]. The Yb^{3+}:YAG crystal has a relatively large gain cross section $\sigma_{\mathrm{em}}^{\mathrm{bulk}}(\lambda_{\mathrm{L}}) = 2.03 \times 10^{-20}$ cm^2 (λ_{L} = 1.031 μm). Although the gain cross section of Yb^{3+}:SrF_2 laser crystals is an order of magnitude lower, $\sigma_{\mathrm{em}}^{\mathrm{bulk}}(\lambda_{\mathrm{L}}) = 0.16 \times 10^{-20}$ cm^2 (λ_{L} = 1.024 μm), the radiative lifetime of these crystals is 2.5-fold longer. Because of this, the minimal threshold cw pump intensity for the Yb^{3+}:SrF_2 crystal is close to that for the Yb^{3+}:YAG crystal. Note also that the properties of Yb^{3+}:SrF_2 only slightly differ from the properties of other ytterbium-doped difluorides, Yb^{3+}:BaF_2 and Yb^{3+}:CaF_2.

For example, according to expression (7), for suspensions of Yb^{3+}:YAG and Yb^{3+}:CaF_2 nanocrystals in alcohol, we have $\sigma_{\mathrm{em}}^{\mathrm{nano}} \approx 0.84\sigma_{\mathrm{em}}^{\mathrm{bulk}}$ and $\sim 0.98\sigma_{\mathrm{em}}^{\mathrm{bulk}}$ and respectively, while for suspensions of the same nanocrystals in air $\sigma_{\mathrm{em}}^{\mathrm{nano}} \approx 0.58\sigma_{\mathrm{em}}^{\mathrm{bulk}}$ and $\sim 0.78\sigma_{\mathrm{em}}^{\mathrm{bulk}}$, i.e., the stimulated emission cross section of nanocomposites insignificantly differs from that of bulk crystals.

The population N_2 of the upper laser multiplet in a doped nanocrystal can be determined by the rate equation [19]

$$\frac{\partial N_2}{\partial t} = [\sigma_{\mathrm{abs}}^{\mathrm{nano}}(\lambda_{\mathrm{p}})N_{\mathrm{l}} - \sigma_{\mathrm{em}}^{\mathrm{nano}}(\lambda_{\mathrm{p}})N_2]\frac{I_{\mathrm{p}}}{\hbar\omega_{\mathrm{p}}} - \frac{N_2}{\tau_{\mathrm{nano}}}, \tag{8}$$

where I_{p}, $\hbar\omega$, and λ_{p} are the intensity, photon energy, and wavelength of the pump radiation, respectively; τ_{nano} is the radiative lifetime of the excited state in the nanocomposite; and $\sigma_{\mathrm{em,abs}}^{\mathrm{nano}}(\lambda_{\mathrm{p}})$ are the effective stimulated emission (em) and absorption (abs) cross sections of the nanocomposite at the pump wavelength λ_{p}. Assume that the nanocomposite absorption cross section $\sigma_{\mathrm{abs}}^{\mathrm{nano}}$ is determined from the absorption cross section of the bulk laser crystal $\sigma_{\mathrm{abs}}^{\mathrm{bulk}}$ similar to expression (7). The population of the lower laser multiplet can be

determined as $N_1 = N_{ions} - N_2$, where N_{ions} is the concentration of active ions in nanocrystals.

In [17, 18], it was shown that the radiative lifetime of the excited state τ_{nano} in a nanocomposite medium strongly depends on the relative refractive index n. At a small volume fraction of nanocrystals in a nanocomposite ($C \leqslant 0.1$), we have

$$\tau_{nano} = \tau_{bulk} n \left(\frac{n^2+2}{3}\right)^2, \quad (9)$$

where τ_{bulk} is the radiative lifetime of the excited state in the corresponding bulk crystal. According to (9), the radiative lifetime in a nanocomposite is longer than that in a bulk crystal and increases with increasing n [17, 18]. For example, for suspensions of Yb^{3+}:YAG and Yb^{3+}:CaF_2 nanocrystals in alcohol, we have $\tau_{nano} \approx 2.14\tau_{bulk}$ and $\sim 1.15\tau_{bulk}$, respectively, while for suspensions of the same crystals in air we obtain $\tau_{nano} \approx 5.71\tau_{bulk}$ and $\sim 2.66\tau_{bulk}$, i.e., the radiative lifetime τ_{nano} in nanocomposites can considerably increase with increasing relative refractive index n.

For a rectangular pump pulse with the intensity I_p and the duration t_p, Eqn (8) has the analytical solution

$$N_2(t_p) = N_2^{\infty}\left\{1 - \exp\left[-\frac{t_p}{\tau_{nano}} \times \left(\frac{\sigma_{abs}^{nano}(\lambda_p) + \sigma_{em}^{nano}(\lambda_p)}{\hbar\omega_p} I_p \tau_{nano} + 1\right)\right]\right\}, \quad (10)$$

where

$$N_2^{\infty} = \frac{N_{ions}}{1 + \sigma_{em}^{nano}(\lambda_p)/\sigma_{abs}^{nano}(\lambda_p) + I_{sat}^{nano}/I_p} = \frac{N_{ions}}{1 + f_p + (i_p n^2)^{-1}} \quad (11)$$

is the maximum population of the upper laser multiplet at the given pump level $i_p = I_p/I_{sat}^{bulk}$;

$$f_p = \frac{\sigma_{em}^{nano}(\lambda_p)}{\sigma_{abs}^{nano}(\lambda_p)} \equiv \frac{\sigma_{em}^{bulk}(\lambda_p)}{\sigma_{abs}^{bulk}(\lambda_p)}$$

is the relative stimulated emission efficiency at the pump wavelength; and

$$I_{sat}^{nano} = \frac{\hbar\omega_p}{\sigma_{abs}^{nano}(\lambda_p)\tau_{nano}} = \frac{I_{sat}^{bulk}}{n^2}, \quad I_{sat}^{bulk} = \frac{\hbar\omega_p}{\sigma_{sat}^{bulk}(\lambda_p)\tau_{bulk}} \quad (12)$$

are the pump absorption saturation intensity for the nanocomposite (I_{sat}^{nano}) and the corresponding bulk crystal (I_{sat}^{bulk}). As follows from (12), the intensity I_{sat}^{nano} quadratically decreases with increasing relative refractive index n of nanocomposites. For example, for suspensions of Yb^{3+}:YAG and Yb^{3+}:CaF_2 nanocrystals in alcohol, $I_{sat}^{nano} \approx 0.56 I_{sat}^{bulk}$ and $\sim 0.89 I_{sat}^{bulk}$, respectively, while for the suspensions of the same crystals in air we have $I_{sat}^{nano} \approx 0.30 I_{sat}^{bulk}$ and $\sim 0.48 I_{sat}^{bulk}$.

With increasing the pump duration t_p, the factor in the braces in (10) tends to unity, and the upper laser multiplet population is $N_2(t_p) \approx N_2^{\infty}$. Note that an increase in the pump level, which is favourable for increasing the inverse population, occurs not only due to an increase in the pump pulse intensity, but also due to the much lower absorption saturation intensity of nanocomposites compared to bulk crystals ($I_{sat}^{nano} < I_{sat}^{bulk}$), which decreases the required pump pulse energy.

Substituting (7) and (11) into (6), we obtain the following expression for the small-signal gain in the case of $N_2(t_p) \approx N_2^{\infty}$:

$$\alpha = \sigma_{em}^{bulk}(\lambda_L) N_{ions} C n \left(\frac{3}{n^2+2}\right)^2 \left[\frac{1+f_L}{1+f_p+(i_p n^2)^{-1}} - f_L\right], \quad (13)$$

where $f_L = \sigma_{abs}^{bulk}(\lambda_L)/\sigma_{em}^{bulk}(\lambda_L)$ is the relative resonance absorption efficiency at the laser wavelength λ_L. The existence of the resonance absorption at this wavelength ($f_L > 0$) is a drawback of quasi-three-level laser media, for example, ytterbium-doped materials. In this case, the pump level is limited from below [19], i.e., there exists a minimal required pump level i_p^{min}, which can be found by equating the first and second terms in the square brackets in (13) and taking into account that $1 + f_p \ll (i_p^{min} n^2)^{-1}$,

$$i_p^{min} = \frac{f_L}{(1+f_L)n^2}. \quad (14)$$

From (14) it follows that the use of a nanocomposite with a relative refractive index n decreases the minimum required pump level i_p^{min} by a factor of n^2. In particular, Yb^{3+}:YAG and Yb^{3+}:CaF_2 laser crystals have $f_L \approx 0.06$ [19], and, for these bulk crystals ($n = 1$), we obtain $i_p^{min} \approx 0.057$. For suspensions of Yb^{3+}:YAG and Yb^{3+}:CaF_2 nanocrystals in alcohol ($n = 1.338$ and 1.059), $i_p^{min} \approx 0.032$ and ~ 0.050, respectively, while for the suspensions of the same nanocrystals in air ($n = 1.82$ and 1.44), we have $i_p^{min} \approx 0.017$ and ~ 0.027.

Figure 2 shows the dependences of the gain α on the relative refractive index n calculated by formula (13) for Yb^{3+}:YAG and Yb^{3+}:CaF_2 nanocrystals with the dopant concentration $N_{ions} = 1.3 \times 10^{21}$ cm^{-3}, the volume fraction of nanocrystals in the nanocomposites $C = 0.1$, and different pump levels.

Figure 2 demonstrates that dependences of the gain have maxima corresponding to the optimal relative refractive index of the nanocomposite n_{opt} (these maxima in Fig. 2b lie out of the region restricted by the maximum possible relative refractive index of nanocomposites $n = n_{crys} = 1.44$). The existence of the gain maxima is explained by the fact that, with increasing n, the population of the upper laser multiplet N_2 (10) increases and the gain cross section σ_{em}^{nano} (7) decreases. The value n_{opt} decreases with increasing pump level. Differentiating expression (13) and equating the derivative to zero, we obtain

$$n_{opt} \approx \sqrt{\frac{\sqrt{4i_p^2 + 68i_p + 1} + 2i_p - 1}{6i_p}}, \quad (15)$$

which is valid even in the case of high $f_{L,p}$ (no higher than 1.0), since the graph of function (13) noticeably shrinks along the ordinate axis with increasing $f_{L,p}$, but only slightly changes along the abscissa axis. In particular, we have $n_{opt1} \approx 1.47$ at $i_p = 0.4$ [see curves (*1*) and (*1'*); as curve (*1*) continuation of curve (*1'*) in the forbidden region $n > n_{crys}$ has a maximum at n_{opt1}] and $n_{opt2} \approx 1.64$ at $i_p = 0.2$ [see curves (*2*) and (*2'*); as curve (*2*) continuation of curve (*2'*) in the forbidden region $n > n_{crys}$ has a maximum at n_{opt2}]. According to (15), with decreasing pump level i_p, the optimal refractive index of nanocomposites n_{opt} increases to n_{crys} and higher, because of which under these conditions it is preferable to choose a dielectric medium with a lower refractive index $n_{med} = n_{crys}/n \to 1$. Hence, as a nanocomposite medium, one can use a suspension of nanocrystals in air or another gas, for example, in helium, which is also a good refrigerant. On the contrary, at a high pump level ($i_p > 5$), the refractive index n_{opt} decreases to a value smaller than unity, i.e., it is better to use a dielectric matrix with the refractive index $n_{med} > n_{crys}$.

Note that the optimal refractive index (15) of a nanocomposite does not depend on the volume fraction C on nanocrystals in the medium.

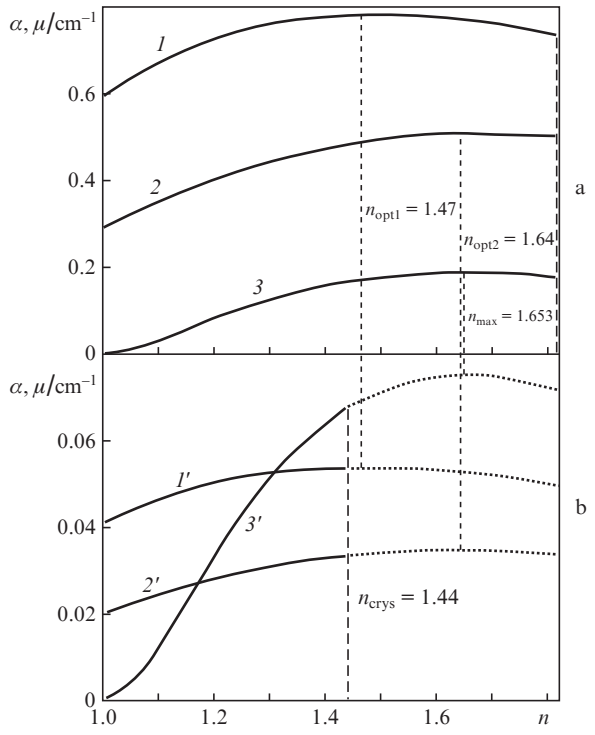

Figure 2. Calculated dependences of the gain α (*1*, *2*, *1′*, *2′*) and Rayleigh scattering loss coefficient μ (*3*, *3′*) on the relative refractive index n of nanocomposites with Yb^{3+}:YAG (a) and Yb^{3+}:SrF_2 (b) at the concentration of active ions in nanocrystals $N_{ions} = 1.3 \times 10^{21}$ cm^{-3}, the volume fraction of nanocrystals in nanocomposites $C = 0.1$, and the pump level $i_p = 0.4$ (*1*, *1′*) and 0.2 (*2*, *2′*).

4. Rayleigh scattering loss coefficient in nanocomposites

The Rayleigh scattering (elastic scattering from nanoparticles with a size smaller than radiation wavelength) cross section is determined as [21]

$$\sigma_{sc} = \frac{8}{3}\pi k^4 \alpha_p^2, \tag{16}$$

where α is the nanoparticle polarisability (the particle is assumed to be nonabsorbing, and, hence, the polarisability is real) and $k = 2\pi n_{med}/\lambda_L$ is the wave number. The validity of the approximation of a nonabsorbing (non-amplifying) particle for describing the Rayleigh scattering must be proved. As was shown in [21], when the polarisability is isotropic, the total cross section for an absorbing particle can be represented as the sum of scattering (16) and absorption cross sections, because of which we will consider the laser processes (amplification of laser radiation and pump radiation absorption) separately.

As was shown by Lorentz, the polarizability for a spherical particle with a radius a is found by the formula [19]

$$\alpha_p = a^3\left(\frac{n^2-1}{n^2+2}\right). \tag{17}$$

Substitution of (17) into (16) yields the following expression for the cross section of scattering by spherical nanocrystals in a medium with the refractive index n_{med}:

$$\sigma_{sc} = \frac{8}{3}\pi\left(\frac{2\pi n_{med}}{\lambda_L}\right)^4 a^6\left(\frac{n^2-1}{n^2+2}\right)^2. \tag{18}$$

The loss coefficient due to the Rayleigh scattering by nanocrystals is determined by the formula

$$\mu = \sigma_{sc} N_{crys}, \tag{19}$$

where N_{crys} is the concentration of scattering crystals in the nanocomposite medium.

The content of spherical nanocrystals in a nanocomposite is convenient to determine by their volume fraction

$$C = VN_{crys} = \frac{4}{3}\pi a^3 N_{crys}. \tag{20}$$

The scattering loss coefficient μ (19) can be expressed through the volume fraction of nanocrystals (20). Then, using the substitution $n_{med} = n_{crys}/n$ to consider suspensions of nanocrystals of a particular material with the refractive index n_{crys} in different dielectric media, we obtain

$$\mu = 2\left(\frac{2\pi}{\lambda_L}\frac{n_{crys}}{n}\right)^4\left(\frac{n^2-1}{n^2+2}\right)^2 a^3 C. \tag{21}$$

Note that the Rayleigh scattering loss coefficient (21), as well as the nanocomposite gain (13), is directly proportional to the volume fraction of nanocrystals C, because of which the loss and gain balance condition, which determines the lasing threshold conditions, does not depend on C.

Figure 2, in addition to the dependences of the gain, shows the dependences of the loss coefficient μ on the relative refractive index n of nanocomposites calculated by formula (21) for Yb^{3+}:YAG and Yb^{3+}:SrF_2 nanocrystals with the volume fraction $C = 0.1$ and the radius $a = 7$ nm. It is seen that the loss coefficient μ has a maximum at the relative refractive index $n_{max} = \sqrt{1+\sqrt{3}} \approx 1.653$ (this value is determined from the extremum of (21) and does not depend on λ_L, n_{crys}, a, and C).

The Yb^{3+}:SrF_2 laser crystal has a very low gain cross section, because of which the dependences of the gain α in Fig. 2b lie below the dependences of the loss coefficient μ in the region $n > 1.34$ at a high pump level ($i_p = 0.4$) and in the wider region with $n > 1.19$ at a low pump level ($i_p = 0.2$). In these regions, amplification and lasing under the given conditions are impossible. In any case, an increase in losses at n close to n_{max} requires an increase in the pump level i_p and/or in the concentration of active ions N_{ions}. In particular, a twofold increase in the concentration N_{ions} [the gain α (13) linearly depends on N_{ions}] in Yb^{3+}:SrF_2 crystals ensures an excess of gain over losses at any n even at a low pump level ($i_p = 0.2$). It should be noted that an increase in the volume fraction of nanocrystals, which also leads to a linear increase in the gain [see expression (13)], cannot solve this problem, because the scattering loss coefficient in this case also linearly increases with increasing C [see expression (21)].

5. Critical size of nanocrystals in a nanocomposite laser medium

Amplification of laser radiation in a nanocomposite laser medium under optical pumping will occur if the laser gain is higher than the loss coefficient due to scattering from nanoparticles. Equating the gain (13) and loss (21) coefficients, we obtain the critical (maximum) radius of nanocrystals

$$a_{cr} = \sqrt[3]{\frac{1}{2}\sigma_{em}^{bulk}(\lambda_L) N_{ions}\left(\frac{2\pi}{\lambda_L}\frac{n_{crys}}{n}\right)^{-4} n\left(\frac{3}{n^2-1}\right)^2} \times$$

$$\times \sqrt[3]{\frac{1+f_L}{1+f_p+(i_p n^2)^{-1}}-f_L}, \qquad (22)$$

which is independent of their volume fraction C.

Figure 3 presents the dependences of the critical radius of nanocrystals a_{cr} on the relative refractive index of nanocomposites n calculated by formula (22) for $Yb^{3+}:YAG$ and $Yb^{3+}:SrF_2$ nanocrystals at different pump levels and the concentration of active ions in the nanocrystals $N_{ions} = 1.3 \times 10^{21}$ cm^{-3}. It is seen that the critical radius of nanocrystals a_{cr} at first rapidly decreases with increasing n (at $1 < n < 1.2$), then decreases much more slowly, and, at $n > 1.3$ becomes almost constant and equal to 11.5 and 6.8 nm for $Yb^{3+}:YAG$ and $Yb^{3+}:SrF^2$ nanocrystals, respectively, at the pump level $i_p = 0.4$. The decrease in i_p to 0.2 leads to $a_{cr} = 10$ and 6 nm for the $Yb^{3+}:YAG$ and $Yb^{3+}:SrF_2$ nanocrystals, respectively. Note that the critical size of nanocrystals increases with increasing pump level. It is also seen that the a_{cr} for $Yb^{3+}:YAG$ nanocrystals is by a factor of 1.72 higher that that for $Yb^{3+}:SrF_2$ nanocrystals at any n. This is explained by the higher gain cross section and refractive index of $Yb^{3+}:YAG$.

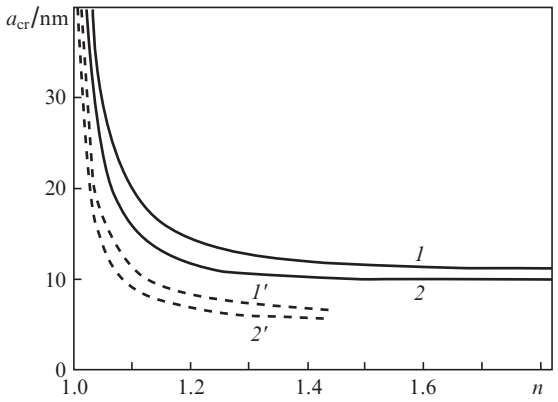

Figure 3. Calculated dependences of the critical radius of nanocrystals a_{cr} on the relative refractive index n of nanocomposites with $Yb^{3+}:YAG$ (*1*, *2*) and $Yb^{3+}:SrF_2$ (*1′*, *2′*) nanocrystals at the pump level $i_p = 0.4$ (*1*, *1′*) and (*2*, *2′*) and the concentration of active ions in nanocrystals $N_{ions} = 1.3 \times 10^{21}$ cm^{-3}.

Figure 4 presents the calculated dependences of the critical radius a_{cr} of nanocrystals on the pump level i_p for $Yb^{3+}:YAG$ and $Yb^{3+}:SrF_2$ nanocrystals at different relative refractive indices of nanocomposites. The index $n = 1.05$ in the case of $Yb^{3+}:YAG$ nanocrystals corresponds to $n_{med} \approx 1.73$, which is achieved when using oxide or oxysulfide crystals or glasses as a dielectric matrix, while for $Yb^{3+}:SrF_2$ nanocrystals at $n = 1.05$, we have $n_{med} \approx 1.7$, i.e., as a dielectric matrix, one can use alcohol or simple fluoride crystals. In the case of $n = 1.4$, we obtain $n_{med} \approx 1.3$ (alcohol or simple fluoride) for $Yb^{3+}:YAG$ nanocrystals and $n_{med} \approx 1.0$ (air, helium, or another gas) for $Yb^{3+}:SrF_2$ nanocrystals.

Figure 4 shows that an increase in the pump level i_p leads to an increase in the critical radius a_{cr} of nanocrystals, but this dependence levels off at $i_p > 3$. Therefore, to increase the critical size of nanocrystals, it is reasonable to increase the pump radiation intensity to three absorption saturation intensities to obtain the critical radius of nanocrystals larger than 20 nm for $Yb^{3+}:YAG$ and 14 nm for $Yb^{3+}:SrF_2$ even at a large relative refractive index of nanocomposites ($n = 1.4$).

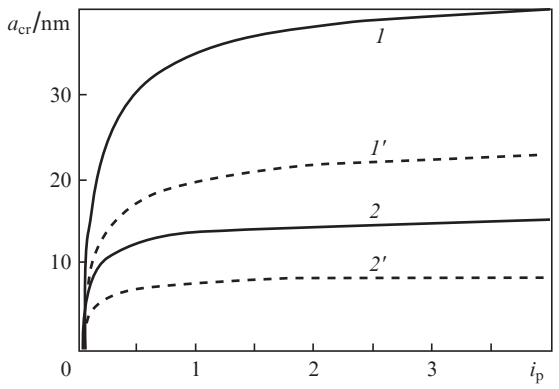

Figure 4. Calculated dependences of the critical radius of nanocrystals a_{cr} on the pump level i_p for $Yb^{3+}:YAG$ (*1*, *2*) and $Yb^{3+}:SrF_2$ (*1′*, *2′*) nanocrystals at the relative refractive index $n = 1.05$ (*1*, *1′*) and 1.4 (*2*, *2′*) and the concentration of active ions in nanocrystals $N_{ions} = 1.3 \times 10^{21}$ cm^{-3}.

6. Rayleigh backscattering coefficient

The Rayleigh backscattering coefficient ε providing the feedback needed for lasing can be determined as

$$\varepsilon = Q\mu, \qquad (23)$$

where Q is the backscattering factor equal to the portion of radiation Rayleigh-backscattered in a narrow oscillation angle θ. The backscattering factor Q can be determined as the ratio of the solid angle $\Omega = 2\pi(1-\cos\theta)$, corresponding to the oscillation angle θ, to 4π, i.e.,

$$Q = \frac{1-\cos\theta}{2}, \qquad (24)$$

and the oscillation angle can be expressed via the geometric parameters of the laser medium as

$$\theta = 2\arctan\frac{d}{2L} \qquad (25)$$

where d and L are the diameter and length of the laser medium.

It is of interest to obtain random lasing in a nanocomposite optical fibre. Although the ratio d/L for optical fibres is very small, the medium length L is almost unlimited. In this case, the total internal reflection returns into the lasing channel the radiation scattered in a larger angle than follows from formula (25).

The authors of [16] measured the Rayleigh backscattering factor Q responsible for random lasing for a single-mode optical fibre 2 km long and for a few-mode optical fibre 11 km long, which was turned out to be 0.002 in both cases. Thus, due to the total internal reflection, the backscattering factor Q in optical fibres does not depend on their length L.

In the subsequent analysis, we take the backscattering factor to be 0.002, which corresponds to an optical fibre or an active element with the ratio $d/L = 0.09$ according to formulas (24), (25).

It should also be noted that the approximation of the constant (along the length) gain α, which is used in formula (5), is adequate only for transverse pumping of a medium. In the case of longitudinal pumping, one must solve initial equation (2).

7. Parametric optimisation of nanocomposites

Now, we have all the parameters needed for estimating the threshold length L_{th} of an active medium by formula (5). For example, taking $Q = 0.002$, for Yb^{3+}:YAG nanocrystals with the concentration of active ions $N_{ions} = 18$ at % (2.6×10^{21} cm^{-3}), their volume fraction $C = 0.001$, and the radius a = 10 nm (which is smaller than the critical radius (22) equal in this case to 14.8 nm) in a dielectric medium with the refractive index $n_{med} = 1.3$ (for example, alcohol) at the pump level $i_p = 0.4$, we obtain the threshold length $L_{th} \approx 74$ cm. The active medium diameter d (according to (24) and (25), the factor $Q = 0.002$ corresponds to $d/L = 0.09$) must be larger than $0.09L_{th} \approx 6.7$ cm.

To decrease the size of a nanocomposite medium, it is necessary to increase the volume fraction C of nanocrystals, because, according to (5) and taking into account (13), (21), and (23), the threshold length L_{th} is inversely proportional to C. In particular, an increase in the volume fraction C from 0.01 to 0.1 leads to an increase in the medium size also by an order of magnitude. Then, in our example, we obtain $L \geqslant 7.4$ cm and $d \geqslant 6.7$ mm.

Figure 5 presents the dependences of the threshold length L_{th} on the radius a of doped scattering nanocrystals calculated by formula (5) using expressions (13), (21), and (23) for suspensions of Yb^{3+}:YAG and Yb^{3+}:SrF_2 nanocrystals in air and alcohol at the pump level $i_p = 0.2$, $Q = 0.002$, $C = 0.1$, and different concentrations N_{ions}. It is seen that the threshold lengths for Yb^{3+}:YAG are an order of magnitude smaller than for Yb^{3+}:SrF_2. This is caused by an order of magnitude higher gain cross section of Yb^{3+}:YAG than of Yb^{3+}:SrF_2 (2.03×10^{-20} cm^2 versus 0.16×10^{-20} cm^2), i.e., laser media with larger cross sections are preferable. The dependences of the threshold length have a minimum (L_{min}) corresponding to the optimal nanocrystal radius a_{opt}. At $a \to 0$, the threshold length L_{th} increases approximately twofold compared to L_{min}. At $a > a_{opt}$, the threshold length also increases and tends to a vertical asymptote corresponding to the critical radius a_{cr} (22) of nanocrystals.

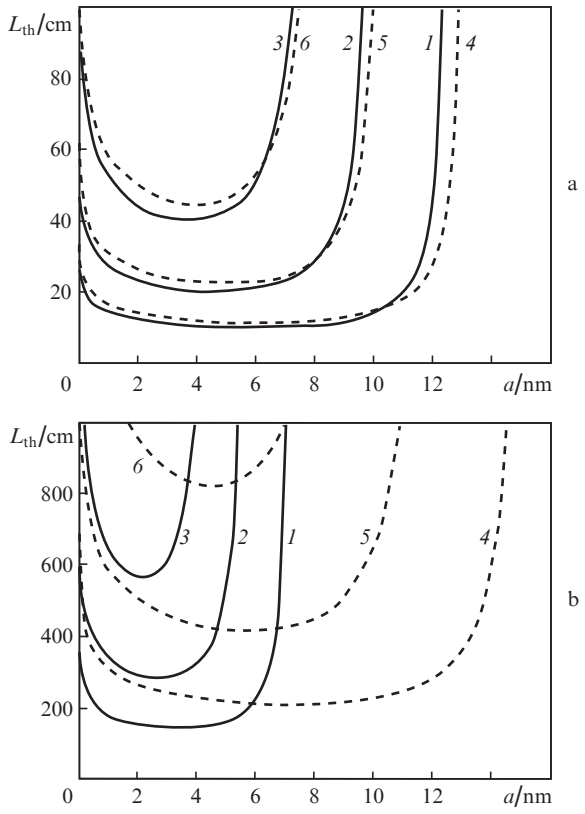

Figure 5. Calculated dependences of the threshold length of the medium L_{th} on the radius a of Yb^{3+}:YAG (a) and Yb^{3+}:SrF_2 (b) scattering nanocrystals suspended in air (*1–3*) and alcohol (*4–6*) at the pump level $i_p = 0.2$ and the medium parameters $Q = 0.002$, $C = 0.1$, and $N_{ions} = 2.6 \times 10^{21}$ (*1*, *4*), 1.3×10^{21} (*2*, *5*), and 0.65×10^{21} cm^{-3} (*3*, *6*).

Figure 5 also shows that the optimal radius a_{opt} corresponding to the minimum threshold length L_{min} is under any conditions two times smaller than the critical radius a_{cr} (22), i.e.,

$$a_{opt} \approx \frac{a_{cr}}{2} = \frac{1}{2}\sqrt[3]{\frac{1}{2}\sigma_{em}^{bulk}(\lambda_L) N_{ions}\left(\frac{2\pi}{\lambda_L}\frac{n_{crys}}{n}\right)^{-4} n\left(\frac{3}{n^2-1}\right)^2}$$

$$\times \sqrt[3]{\frac{1+f_L}{1+f_p+(i_p n^2)^{-1}} - f_L}\,. \qquad (26)$$

Note that, with increasing nanocrystal radius to $a \approx 0.89a_{cr} \approx 1.78a_{opt}$, the threshold length of the medium increases by two times with respect to the minimal length, i.e., $L_{th} = 2L_{min}$. Note also that the threshold length is comparable with μ^{-1} at $a \approx a_{opt}$, and becomes an order of magnitude larger than μ^{-1} as a increases to $1.78a_{opt}$.

As the concentration of active ions N_{ions} decreases (Fig. 5), the dependences go up (L_{min} increases) and become narrower (a_{cr} decreases). According to (22), this narrowing is proportional to $\sqrt[3]{N_{ions}}$. Note that the minimum threshold length L_{min} is inversely proportional to N_{ions}. Substituting the optimal radius (26) into relation (21), we find the optimal loss coefficient to be $\mu_{opt} \approx \alpha/8$. Taking this into account, we indeed find from (5) the inversely proportional dependence of L_{min} on N_{ions} in the form

$$L_{min} \approx \frac{8}{7}\alpha^{-1}\ln\frac{14}{Q} = \frac{8}{7}[\sigma_{em}^{bulk}(\lambda_L) N_{ions} Cn]^{-1}\left(\frac{n^2+2}{3}\right)^2$$

$$\times\left[\frac{1+f_L}{1+f_p+(i_p n^2)^{-1}} - f_L\right]^{-1}\ln\frac{14}{Q}. \qquad (27)$$

The length L_{min} decreases with increasing pump level. In addition, there exists only a weak dependence of L_{min} on n with a minimum corresponding to the optimal refractive index n_{opt} (15) determined from the gain maximum.

It should also be noted that the dependences for suspensions of Yb^{3+}:YAG nanocrystals in air and alcohol (Fig. 5a) are close to each other, while the corresponding dependences for Yb^{3+}:SrF_2 nanocrystals are strongly different (Fig. 5b). The dependences for Yb^{3+}:SrF_2 nanocrystals in air are considerably narrower, i.e., the critical radius a_{cr} is smaller. The decrease in the a_{cr} is caused by the smaller $\sigma_{em}^{bulk}(\lambda_L)$ for Yb^{3+}:SrF_2 than for Yb^{3+}:YAG [if the gain cross section decreases by an order of magnitude, then the a_{cr} decreases approximately by $\sqrt[3]{10} \approx 2$ times according to (25)]. However, the dependences for Yb^{3+}:SrF_2 nanocrystals suspended in alcohol are even wider than for Yb^{3+}:YAG. This occurs because the refractive index of SrF_2 ($n_{crys} = 1.44$) is close to the refractive index of alcohol ($n_{med} \approx 1.36$), i.e., the relative refractive index n for this nanocomposite is close to unity, which leads to an increase in the critical and optimal radii of

nanocrystals a_{cr} (22) and a_{opt} (26) due to the increasing term $(n^2 - 1)^{-2}$.

The choice of a dielectric medium with n_{med} close to n_{crys}, is an original method for increasing the optimal size of nanocrystals with a small gain cross section (both $Yb^{3+}:SrF_2$ and any other nanocrystals). However, to decrease the threshold length of the medium, according to (27), it is necessary to increase the concentration of active ions in nanocrystals [the minima of dependences (*1*) and (*4*) in Fig. 5b corresponding to a large $N_{ions}$are close to each other], their volume fraction, or the pump level.

According to (27), the minimum threshold length L_{min} (corresponding to the optimal radius of nanocrystals) is inversely proportional to the volume fraction C of nanocrystals in the medium. According to (5) and taking into account (13), (21), and (23), the threshold length L_{th} (at a nonoptimal radius of nanocrystals) also turns out to be inversely proportional to the volume fraction C of nanocrystals. Note that the results obtained for $C = 0.1$ can be easily extended to other values of C.

It should also be noted that, at a nonoptimal choice of the radius of nanocrystals, the threshold length L_{th} is higher than L_{min}. This is especially pronounced in the case of the weakly amplifying $Yb^{3+}:SrF_2$ laser medium. To explain this fact, we should look at Fig. 2b, which shows that the loss exceeds the gain at $n > 1.34$ for a high pump level ($i_p = 0.4$) and at $n > 1.19$ for a low pump level ($i_p = 0.2$), and, hence, amplification and lasing at these n cannot occur. Therefore, nanocrystals with a small gain cross section can be used if their size is close to optimal.

Let us discuss the dependence of the threshold length on the concentration of active ions N_{ions} in nanocrystals. According to (27), this dependence for the minimum threshold length L_{min} , which corresponds to the optimal size of nanocrystals a_{opt}, is inversely proportional, i.e., the reciprocal length $1/L_{min}$ linearly increases as the argument increases from zero.

The dependence of the reciprocal threshold length $1/L_{th}$ on N_{ions} at a nonoptimal size of nanoparticles ($a \neq a_{opt}$) plotted by formula (5) is also close to linear in the case of large N_{ions} but deviates from linear at small N_{ions} and is equal to zero ($L_{th} \to \infty$) at $N_{ions}^{min} > 0$. The limit $L_{th} \to \infty$ corresponds to $a = a_{cr}$, which allows one to find the concentration N_{ions}^{min} from expression (22).

8. Conclusions

Thus, we calculated the characteristics of a random laser based on nanocomposite media consisting of a transparent dielectric with scattering doped nanocrystals.

As nanocrystals, it is proposed to use ytterbium laser materials with a high concentration of active ions, and, as a dielectric matrix for these nanocrystals, it is possible to use gases (air, helium), liquids (alcohol), or solid dielectrics with refractive indices lower than that of nanocrystals.

It is shown that, at a fixed volume fraction of nanocrystals in a medium, the laser gain does not depend on the size of nanocrystals, while the dependence of the scattering loss coefficient on this size is cubical. This allows one to control the size of nanocrystals in order to provide the best conditions for amplification and lasing.

Based on the concept of nonresonant distributed feedback due to the Rayleigh scattering, an expression is obtained for the minimum (threshold) length of a nanocomposite laser medium at which the random lasing threshold is overcome. The threshold length of the medium is inversely proportional to the gain cross section of nanocrystals, concentration of active ions in these nanocrystals, and their volume fraction in the medium. For example, for suspensions of $Yb^{3+}:YAG$ nanocrystals (18% at %) with a radius of 20 nm in alcohol at the pump level $i_p = 0.4$ (ratio of the pump intensity to the absorption saturation intensity), the threshold length of the medium decreases from 74 to 7.4 cm as the volume fraction of nanocrystals increases from 0.01 to 0.1.

An expression is obtained for the optimal relative refraction index n of nanocomposites (ratio of the refractive index of nanocrystals to the refractive index of the dielectric medium) corresponding both to the maximum gain and the minimum threshold length of the medium at the optimal size of nanocrystals. It is shown that the optimal relative refractive index of nanocomposites decreases with increasing pump level, does not depend on the nanocomposite parameters, and is 1.64 and 1.47 at $i_p = 0.2$ and 0.4, respectively.

Expression are derived for the critical (maximum) and optimal sizes of nanocrystals, which are found to be independent of the volume fraction C of nanocrystals in the medium at $C < 0.1$. The critical size of nanocrystals corresponds to the balance of gain and scattering loss at which the threshold length of the medium tends to infinity. At the relative refractive index n tending to unity, the critical size of nanocrystals tends to infinity, but, at $n > 1.3$, it has an almost constant small value $a_{cr} = 10$ and 6 nm for $Yb^{3+}:YAG$ and $Yb^{3+}:SrF_2$ nanocrystals, respectively, at the pump level $i_p = 0.2$. An increase in i_p to 3 leads to an increase in the critical radius to 20 nm for $Yb^{3+}:YAG$ and 14 nm for $Yb^{3+}:SrF_2$ nanocrystals at $n > 1.3$. The optimal size of nanocrystals, which corresponds to the minimum threshold length of the medium, is two times smaller than the critical size at any nanocomposite parameters.

The obtained results can be used for purposive search and development of new nanocomposite laser media with improved characteristics.

Acknowledgements. The authors thank K.K. Pukhov for discussion of the results. This work was supported by the Program of the Presidium of the Russian Academy of Sciences No. 24P 'Fundamentals of nanostructures, nanotechnologies, and nanomaterials'.

References

1. Noginov M.A. *Solid State Random Lasers* (Berlin: Springer, 2005).
2. Cao H.J. *Physica A*, **38**, 10497 (2005).
3. Noginov M.A., Novak J., Grigsby D., Deych L. *J. Opt. A: Pure Appl. Opt*., **8**, S285 (2006).
4. Markushev V.M., Ryzhkov M.V., Briskina Ch.M. *Kvantovaya Elektron*., **37** (9), 837 (2007) [*Quantum Electron*., **37** (9), 837 (2007)].
5. Zhu G., Small C.E., Noginov M.A. *J. Opt. Soc. Am. B,* **24**, 2129 (2007).
6. Wiersma D.S. *Nat. Phys*., **4**, 359 (2008).
7. Garcia P.D., Ibisate M., Sapienza R., Wiersma D.S., Lopez C. *Phys. Rev. A*, **80**, 013833 (2009).
8. Guerin W., Mercadier N., Brivio D., Kaiser R. *Opt. Express*, **17**, 11236 (2009).
9. El-Dardiry R.G.S., Mosk A.P., Muskens O.L., Lagendijk A. *Phys. Rev. A*, **81**, 043830 (2010).
10. Letokhov V.S. *Pis'ma Zh. Exp. Teor. Fiz*., **4**, 477 (1966) [*JETP Lett.*, **4**, 321 (1966)].

11. Ambartsumyan R.V., Basov N.G., Kryukov P.G., Letokhov V.S. *Pis'ma Zh. Exp. Teor. Fiz*., **3**, 261 (1966) [*JETP Lett*., **3**, 167 (1966)].
12. Ambartsumyan R.V., Kryukov P.G., Letokhov V.S., Matveets Yu.A. *Zh. Exp. Teor. Fiz*., **53**, 1955 (1967) [*JETP*, **26**, 1109 (1968)].
13. Markushev V.M., Zolin V.M., Briskina Ch.M. *Kvantovaya Elektron*., **13** (2), 427, (1986) [*Sov. J. Quantum Electron*., **16** (2), 281 (1986)].
14. Letokhov V.S. *Pis'ma Zh. Exp. Teor. Fiz*., **5**, 262 (1967) [*JETP Lett*., **5**, 212 (1967)].
15. Turitsyn S.K., Babin S.A., El-Taher A.E., Harper P., Churkin D.V., Kablukov S.I., Ania-Castanon J.D., Karalekas V., Podivilov E.V. *Nat. Photonics*, **4**, 231 (2010).
16. Vatnik I.D., Churkin D.V., Babin S.A., Turitsyn S.K. *Opt. Express*, **19**, 18486 (2011).
17. Pukhov K.K., Basiev T.T., Orlovskii Yu.V. *Pis'ma Zh. Exp. Teor. Fiz.*, **88**, 14 (2008) [*JETP Lett*., **88**, 12 (2008)].
18. Basiev T.T., Orlovskii Yu.V., Pukhov K.K. *Ros. Nanotekhnol*., **3**, 66 (2008) [*Nanotechnol. Russ*., **3**, 551 (2008)].
19. Svelto O. *Principles of Lasers* (New York: Springer-Verlag, 2004).
20. DeLoach L.D., Payne S.A., Chase L.L., Smith L.K., Kway W.L., Krupke W.F. *IEEE J. Quantum Electron*., **29**, 1179 (1993).
21. Van de Hulst H. *Light Scattering by Small Particle*s (New York: Wiley, 1957).