

RANDOM DYE LASING SUPPORTED BY ELASTIC MULTIPLE LIGHT SCATTERING²

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The elastic multiple light scattering, at which a partial confinement of photons within the area where amplification exists, promotes the development of a stimulated radiation with all tags of amplified stimulated emission (ASE). Because the spectral narrowing of ASE with increase in the pumping runs fluently without any catastrophe tags which are possible if arbitrary resonances (cavity modes, distributed feedback modes, or Anderson localization modes) are present, ASE conserves the initial smooth spectral distribution in spite of the strong spectral narrowing. The simple theoretical analysis of ASE under elastic multiple light scattering in connection with a band width evolution and scaled threshold conditions that is based upon the approximate determination of the mean free path in EMLS media as $l \approx (N\sigma_{\text{sct}} - \alpha_{\text{gain}})^{-1}$ and the Kubelka–Munk approximation is completed.

forbidden, and one says by analogy with the electron zone of a crystal about the forbidden and allowed energy zones in the handmade photonic crystals [1, 2].

Coherent Bragg scattering and non-coherent elastic multiple light scattering (EMLS) are both observable in media with periodic or chaotic spatial refraction index modulation in 1D, 2D, or 3D cases. Because oscillating modes in lasers are connected with energy transfer, they are spatially confined but never localized, and that is why the typical laser modes are dislocated around the energy band gap. If some defects in a photonic crystal structure appear, the localized mode states becomes allowed in the forbidden zone (by analogy with the electron zone theory and practice of crystals). In opposite to the usual cavity modes, the last are considered to be localized. That is why it was proposed the similar localized modes can appear in the case of EMLS in the amplifying media. The idea of the localized light mode appearance was born by analogy with the experimentally proved phenomenon of the Anderson localization of electrons in imperfect conductors [3–5].

1. Introduction

The laser oscillation is commonly produced with optically transparent amplifying media inside a cavity ensuring a positive feedback on the oscillating modes. The modes correspond approximately to the transmission/reflection resonances of a Fabry-Perot interferometer that is conceivable in 1D, 2D, and 3D performances. Resonances arise owing to a multibeam interference of coherent partial waves and support the coherent feedback.

The other type of cavities that support the coherent feedback is conceivable for the spatially periodic structures: the regular phase delay for partial adding waves arises due to the coherent multiple Bragg scattering of light. Because 1D, 2D, and 3D Bragg scatterings exist, one can build up similar 1D, 2D, and 3D laser cavities as well. They say these cavities allow one to establish the coherent spatially distributed feedback. When the coupling for forward-backward waves in the Bragg scattering gets enough strong, the light propagation for some direction becomes strongly

in the disordered case, the light waves will perform a random walk and their energy transports in the diffusion regime. The occurrence of interference effects is the case is less obvious to understand; however, the interference effects also turn out to revive in random systems. The interference of light in random dielectric systems influences the transport of light in a way that is similar to that where the interference occurs for electrons when they propagate in disordered conducting materials. The following phenomena that hang upon the coherence under conditions of EMLS have also been found to exist: the weak Anderson localization [6], the photonic Hall effect [7], optical magnetoresistance [8], and, last time, the above-mentioned strong Anderson localization under linear 1D [9] and nonlinear 1D–2D lasing conditions [10].

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V. Letokhov was first who predicted that one can excite a laser-like emission from amplifying media using a non-resonant feedback via the multiple scattering of light from a diffusive reflector [11]. This phenomenon was first observed in experiments on dye solutions without any cavity mirrors and was recognized afterwards as amplified spontaneous emission (ASE) [12]. The new splash of a research interest to the phenomenon began after publication [13], where the authors “spoiled” a transparent dye laser solution by advisedly adding the non-soluble powder TiO_2 and actualized the lasing on the opaque scattering medium without external cavity. Though the measured parameters of emission were the same as for the clear dye solution ASE, the results has excited the imagination to perform the laser optical analog of the Anderson localization and to observe the “frozen light”.

The following estimation was deposited in the expectation to oscillate the “frozen light” in a multiple scattering laser medium. The scattering strength of a disordered optical material is determined by its transport mean free path l defined as the scale length over which a propagating wave loses its memory of the propagation direction. A material is in the multiple scattering state, when l is considerably smaller than the system size L . In that case, the transport of light can be described by a random walk with step length l . If the scattering strength of a material is increased (and hence l decreased), eventually a phase transition into an Anderson localized state is expected to occur at $kl \rightarrow \pi$, where k is the wavevector of the light [3,4]. For $kl > \pi$, the transport is predominately diffusive, which is the case in most of the available disordered dielectric materials, whereas a fascinating scaling regime exists around $kl \sim \pi$, in which the transport become slow [14].

The subject of the investigation is the spectrum evolution of the known laser dyes in a polymer matrix (polyvinyl acetate) with high concentration (up to 30 weight %) of inaccurate (for size) scattering centers under nanosecond laser pumping. The studied subject lies in a borderline region of laser physics and optics of EMLS. Because both named scientific fields are advanced within the framework of their own paradigms, the first observations of a lasing clone in media with EMLS were accompanied by legal surprise of the advocates of conventional laser schools and have spawned expectations for a new lasing appearance of the EMLS specialists. Today it is possible to say both sides are right: ASE and the lasing on localized Anderson modes may exist independently and can be coexistent.

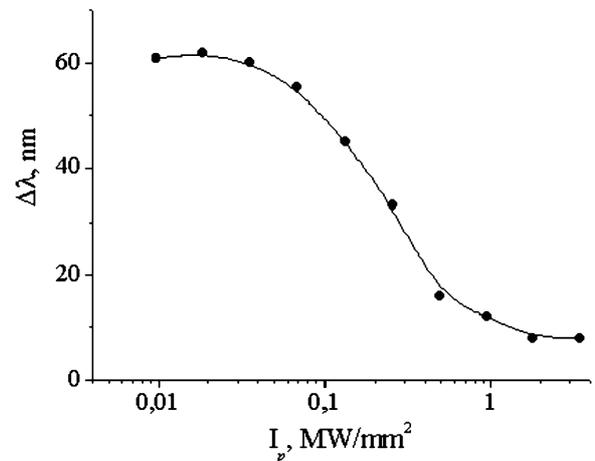


Fig. 1. Half-width emission dependence of R6G under the EMLS condition (PVA with fumed silica, concentration $3 \times 10^{11} \text{ cm}^{-3}$) on the pumping power

In spite of the high level of comprehension and practical implementation of a lasing in the ensembles of microcavities and a photon crystal [15], the nature of a stimulated light emission in amplifying multiple light scattering media [16] requires, according to our reckoning, the further investigation.

2. Experimental Observations and Analysis

We studied the emission bandwidth evolution of dyed polyvinyl acetate (a PVA film of $t \approx 1 \div 2$ mm in thickness) with elastic scattering particles in a range of concentrations 6–30 weight % (powder of fumed silica, sapphire, diamond, etc.) and laser dyes (R6G, astrophloxine at a concentration of $\approx 10^{-4} \div 10^{-3}$ mol/l) at the excitation by a second harmonic of a laser $\text{Nd}^{3+}:\text{YAG}$ ($\lambda = 532$ nm, $\tau_p \approx 20$ ns, the size of a excitation spot $s \approx 0.5 \div 2$ mm). The measurements of the sizes of powder particles were done with a size analyzer LMS-30 (Sheishin corporation). The size distributions lay in limits from 8 up to 20 μm (fumed silica) and 20–100 μm (sapphire). The difference of the mean diameters of particles and their index refraction were not developed in the evolution of the general pattern of emission.

The most typical behavior of the evolution of emission spectra as a function of pumping power (gain) is presented in Figs. 1 and 2. Let us pay attention that the spectrum narrowing begins from the bandwidth of R6G fluorescence and closes at the some definite pumping fluently without the catastrophe usual for a standard cavity laser. The envelope of the emission spectrum stays smooth without any sharp splash at all pumping

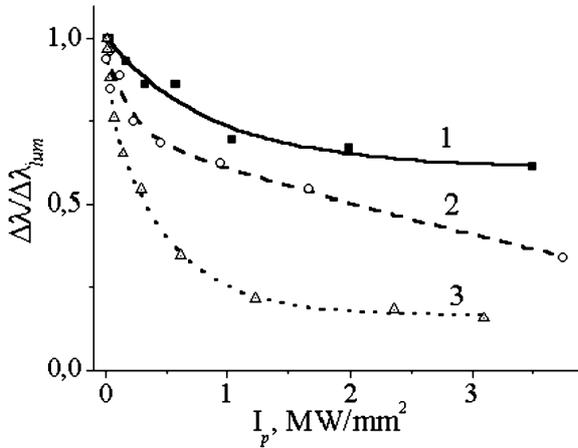


Fig. 2. Half-width emission dependence of R6G under the EMLS condition for 3 different fumed silica concentrations 6×10^{10} (1), 1.8×10^{11} (2), and $3 \times 10^{11} \text{ cm}^{-3}$ (3) on the pump intensity. The pump beam spot diameter $d = 0.32 \text{ mm}$ is invariable

magnitudes (2 orders). This characteristic behavior of the spectrum emission with pumping is similar to that of the known ASE emission in transparent media barring its radial beam-like character due to ballistic wave propagation [12].

It is known that the light scattering can be taken into account by combining the wave and diffusion equations. In this case, a propagation constant is complex-valued $\gamma^2 = k^2 - j\omega/D$, where $D = lc/3n$ – light diffusion coefficient, l – above-mentioned mean free path /17/. If it is so, we can consider the results in Figs. 1 and 2 as ASE in the diffusion approximation. The active medium in the absence of a back wave owing to the scattering and a feedback is characterized by the spectrally dependent single-pass amplification $\Gamma_1(\omega) \approx \exp[-(\omega - \omega_s + \alpha_0(\omega))jLn/c]$, where $\alpha_0(\omega)$ – gain coefficient for the given inverse population, whose formfactor is determined by the nature of a broadening, ω_s – frequency of a possible mode by definition $\exp(j\omega_s Ln/c) = \pm 1$, n – refractive coefficient of the laser medium, and Ln – the optical length of an amplifier. At the placing of such a medium in the cavity with 2 mirrors, described by the complex reflection/transmission factors r and t , the fluorescence emission will stand amplification on modes of the cavity. In the approximation of the most strong amplification of the modes which are near a maximum of $\alpha_0(\omega)$, the summation of partial waves excited owing to the multiple passes of radiation through the cavity gives [18]

$$\Gamma(\omega) \approx T^2 \Gamma_1(\omega) / (1 - R^2 \Gamma_1^2(\omega) [1 - 2jLn(\omega - \omega_s)/c]). \quad (1)$$

Here, $T = tt^*$ and $R = rr^*$ – energy transmittance and reflection of cavity mirrors, respectively. It is the Lorentz dependence with maxima of amplification on the proper modes of a given cavity. The catastrophic growth of amplification for the eliminated frequencies ω_s meets the excitation of oscillations on the proper modes of the system. The initial spectrum of an amplified luminescence in these cases is exchanged by the spectrum of oscillating modes with their natural formfactors. If the excitation of oscillations on the proper modes of the cavity due to the treatment of a denominator at zero does not occur, there is a rather smooth deformation of the initial contour in conformity to the frequency dependence of $\Gamma_1(\omega)$ in the numerator of (1).

It is seen from (1) that, while $\Gamma_1(\omega) < 1$, the power of a fluorescence in the cavity is attenuated. With the achievement of amplification $\Gamma_1(\omega) > 1$, there comes a stage of amplification of the spontaneous emission with the increasing contribution of the stimulated transitions. At the performance of resonant conditions in (1), there is a sharp magnification of the power at these resonances (modes) and a gain saturation will come:

$$\alpha_{\text{sat}}(\omega) = \alpha_0(\omega) / (1 + I_{\text{osc}}/I_{\text{cr}}) = \alpha_{\text{loss}}(\omega). \quad (2)$$

Here, $I_{\text{osc,cr}}$ – laser emission intensity and the critical intensity (dependent on spectroscopic parameters of the laser transition). Taking these considerations into account, it is possible to explain the characteristic behaviour of the ASE half-width under EMLS conditions as follows. In the lack of resonances of any nature, the occurrence of amplification is accompanied by an power increase of the initial fluorescence and the smooth diminution of its spectral width before the gradual approach to the gain saturation. For the quantitative description of such a dependence, we take advantage of regulations about a constancy of the product of the peak amplification by its half-width for homogeneously broadened transitions [18]:

$$\Gamma_1(\omega_{\text{max}}) \Delta_{05}(\omega) = \text{const}. \quad (3)$$

If a constant on the right-hand side is defined from the requirement that $\Gamma_1(\omega_{\text{max}}) = 1$ on the threshold of excitation and the spectrum bandwidth of the amplifier be equal to the fluorescence width Δ_{lum} , the width of ASE with growing amplification from 1 up to some value will change under the hyperbolic law:

$$\Delta_{05}(\lambda) \approx \Delta_{\text{lum}} / \Gamma_1(\lambda_{\text{max}}). \quad (4)$$

In the field of amplification outlying from any resonances, $\Gamma_1(\lambda) \approx \exp(\alpha_0(\lambda)aL)$, where aL – the

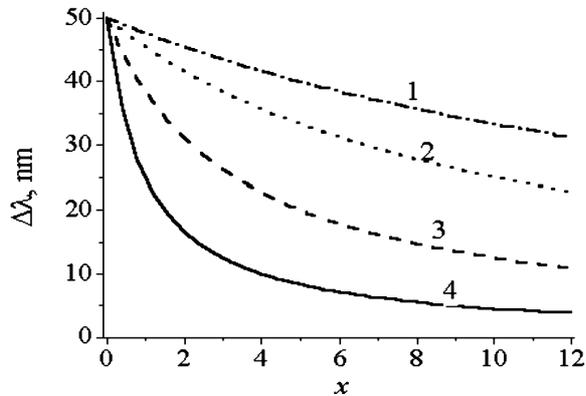


Fig. 3. ASE bandwidth dependence for a typical dye under EMLS conditions as a function of the virtual amplification $x = \exp(\alpha_{ge}(\lambda)aL$ at $a = 0.05$ (1); 0.1 (2); 0.3 (3); 1 (4)

effective length of amplification dependent on the multiplicity of scattering in the field of amplification; a – the parameter considering the efficiency of multiple scattering. The calculated dependence of the ASE bandwidth for typical dye with the initial luminescence at 50 nm is presented in Fig. 3.

The comparison of the calculated and experimental dependences (Figs. 1 and 3) specifies their qualitative consent. For convenience of a statement, the beginning of the luminescence spectrum narrowing is named below as the first oscillation threshold. The area of the second bend in the dependence of the ASE spectrum width on pumping is named as the second oscillation threshold. There are some reasons for the transition to the second bend with the trend to a constant value – gain saturation and ASE increment of a spectral edge steepness. The contribution of each of them should be analyzed carefully in experiments.

The second characteristic feature of the stimulated emission under EMLS conditions is connected with the scaling effect. As was noted above, the implementation of a multiple scattering depends on whether the inequality $L > l$ holds. In our case, L is the diameter of a pumping spot on the active medium surface: if the inequality is not fulfilled, the luminescence flows leave the activated volume at a minimal amplification; if this inequality holds, the flows remain partially inside the activated area due to the multiple scattering and repeatedly gain in power. For this reason, the scaling effect is possible: the dependence of the ASE first oscillation thresholds – beginning of the ASE spectral narrowing at a given scattering strength – on the pumping spot size. For a volumetric specimen R6G in the PVA matrix with fumed silica as a scatterer, the

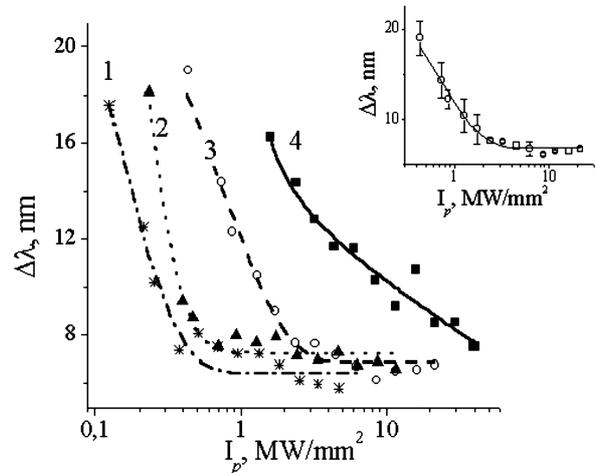


Fig. 4. Variation of the ASE spectral bandwidth for R6G in PVA doped by fumed silica (a concentration of $3 \times 10^{11} \text{ cm}^{-3}$) versus the pumping intensity at various beam spot sizes: $d = 0.32$ mm (1); 0.24 (2); 0.18 (3); 0.13 (4). The insert shows the errors of measurements

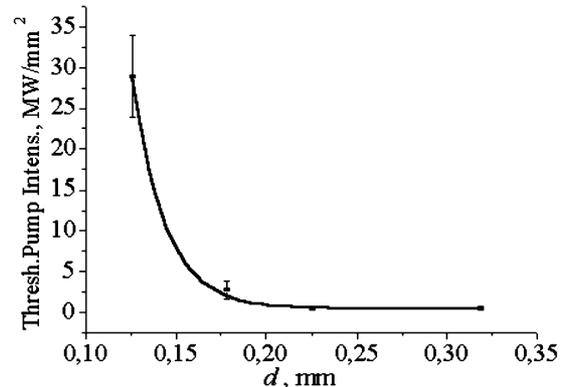


Fig. 5. Dependence of the second oscillation threshold on the pumping beam spot size

results are presented in Fig. 4. The experiment was carried out by the longitudinal shift of a specimen along the axis of a focusing lens with change of the power entry and the measurement of the spectrum width evolution with the pumping power. Having set, for example, a pumping level of 1 MW/cm², it is possible to see that the conditional second oscillation threshold is attained only with a pumping beam spot size of more than 0.2 mm. From pumping we refer to the area of constant bandwidth ASE in the second threshold of generation. In Fig. 5, the dependence of the second oscillation threshold on the pumping beam spot size gained from data of Fig. 4 is presented.

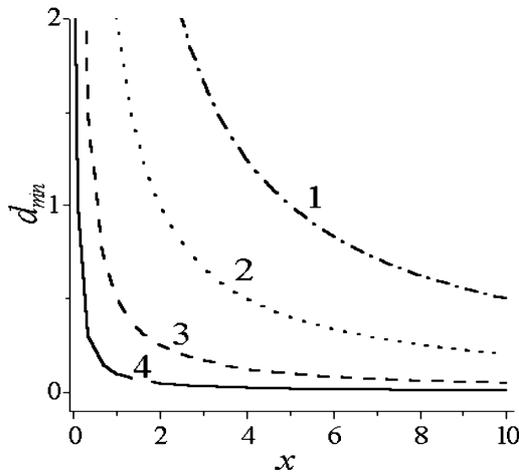


Fig. 6. Scaling effect: the minimum diameter of a pumping spot that is necessary for the first threshold achievement as a function of $x = N\sigma_s/\alpha_g - 1$ and $\alpha_l/\alpha_{\text{gain}}^2 = 5$ (1), 2 (2), 0.2 (3), 0.1 cm^{-1} (4)

The estimation of the critical size of the activation area (pumping beam spot size) about the first oscillation threshold can be obtained from the following reasons. The mean free path of a photon in the dissipative medium with total scattering and absorption losses is defined as:

$$\langle l \rangle = \int l \alpha_{\text{tot}} \exp(-\alpha_{\text{tot}} l) dl. \quad (5)$$

In that case, the photon mean free paths l in the presence of scattering, absorption, or amplification are

$$l_{\text{abs}} = (N\sigma_{\text{sct}} + \alpha_{\text{abs}})^{-1}, \quad (6)$$

$$l_{\text{gain}} = (N\sigma_{\text{sct}} - \alpha_{\text{gain}})^{-1}. \quad (7)$$

Therefore, the condition for the first threshold to be achieved can be written down as the inequality

$$|-\alpha_{\text{gain}} d_{\text{min}}| \geq |\alpha_{\text{loss}} l_{\text{gain}}| \quad (8)$$

or

$$d_{\text{min}} \geq \alpha_{\text{loss}}/\alpha_{\text{gain}}^2 (N\sigma_{\text{sct}}/\alpha_{\text{gain}} - 1). \quad (9)$$

Here, d_{min} , α_{loss} – minimally admissible diameter of a pumping spot size and the averaged losses in the active area of the wandering of photons. The graphic presentation of (9) is presented in Fig. 6.

For the considered system, we provide a little more strict analysis of ASE with the use of the mathematical

approach offered by Kubelka and Munk for the albedo analysis of strongly scattering surfaces [19]. It is the system of two nonpolarized counter-propagated light fluxes connected through the back scattering without taking into account the phase ratio owing to its averaging under multiple elastic scattering. The main media parameters – the amplification and scattering factors – determine the intensity ratio of these fluxes on the output. Amplification brings, indeed, a novelty aspect in the old model. The stationary one-dimensional problem of the flux power distribution along the z -direction can be posed as

$$dI/dz = (\alpha_{\text{gain}} - N\sigma_{\text{sct}})I + N\sigma_{\text{sct}}J, \quad (10a)$$

$$dJ/dz = -(\alpha_{\text{gain}} - N\sigma_{\text{sct}})J - N\sigma_{\text{sct}}I, \quad (10b)$$

where I, J – intensities of the counter-propagated fluxes in the active medium of thickness L . We introduce the designations $I/J = H$, $J/I = h$, $\alpha_{\text{gain}} - N\sigma_{\text{sct}} = G$, $N\sigma_{\text{sct}} = S$. Then we have

$$dI/I = Gdz + SH^{-1}dz, \quad (11a)$$

$$dJ/J = -Gdz - SHdz. \quad (11b)$$

Let's subtract (11b) from (11a). We dispart variables and make integration of the left and right parts in the limits from H_0 to H_L and from $L = 0$ to $L = L$. We obtain

$$\int dH / \left(H^2 + 2 \left(\frac{\alpha_{\text{gain}}}{S} - 1 \right) + 1 \right) = \int S dz.$$

For the case under consideration where $((\alpha/S - 1)^2 > 1)$, the above integral is equal to

$$\frac{1}{2(p-q)} \ln \left(\frac{H-p}{H-q} \right) \Bigg|_{H_0}^{H_L} = SL,$$

where p, q – roots of the equation in the integrand denominator which are $p = 0, q = -2G/S$. In view of the symmetry of light fluxes inside of a layer, the boundary conditions are defined as follows: $H_0 = I_{\text{lum}}/J_{\text{osc}}$ at $L = 0$ and $H_L = I_{\text{osc}}/J_{\text{lum}}$ at $L = L$. Then, after simple transformations, we get

$$\begin{aligned} & [1 + 2H_L(1 + \alpha_{\text{gain}}/S)]/[1 + 2H_0(1 + \alpha_{\text{gain}}/S)] = \\ & = \exp[4(\alpha_{\text{gain}} - S)L]. \end{aligned} \quad (12)$$

From (12) under the condition of $I_{\text{lum}} < I_{\text{osc}}$, it is possible to find the ASE intensity in the obvious form:

$$I_{\text{osc}} \approx \frac{SI_{\text{lum}}}{2(\alpha_{\text{gain}} + S)} \exp(4(\alpha_{\text{gain}} - S)L). \quad (13)$$

Formula (13) offers a number of simple criteria for the analysis of ASE in the elastic scattering medium in the presence of amplification:

1. ASE does not arise at all in the lack of scattering $S = 0$,
2. ASE fades at the excess of scattering over amplification $S > \alpha_{\text{gain}}$,
3. At the preset values of S and α_{gain} , there is some minimal critical length of the active layer L_{crit} , below which ASE also does not arise up to a level above the luminescent background. For this reason, there is the scaling effect considered above. In Fig. 7, we present the 3D-graphical dependence (13) at $L = \text{const}$ and the variables $X = \alpha_{\text{gain}}$ and $Y = S$.

3. Concluding Remarks

Taking the presented results into consideration, it is helpful to repeat the arguments in connection with the main question in the problem: Does the observed emission under EMLS conditions fall into the case of ASE or laser oscillator?

The lack of the directivity in a definite solid angle for the observed emission [20], as customaries and reference property of the laser oscillator, is not a sufficient badge for the denying of an oscillatory lasing. For example, the oscillation in a laser crystall powder, which formed nonreciprocal microcavities with a random space orientation, did not discovered the sharp threshold and the directivity as well [15]. However, the oscillatory lasing with spectral narrowing at above the threshold is not possible because, when the system passes into the gain saturated state, the oscillation spectral width should turn out to broaden as the pumping power grows. At the same time, the laser amplifier can only pinch the spectral width of an amplified emission up to the gain saturation and steepens the edges of the spectral band up to the rectangular formfactor. The last behavior has been shown in [20]. Thus, ASE under EMLS conditions in the spectral narrowing region and prior to the onset of the second threshold can be considered as the process of unsaturated amplification in a relevant medium. The area with spectral narrowing stop can be considered as a region with gain saturation or (and) rectangularization of the emission band formfactor. Because the gain

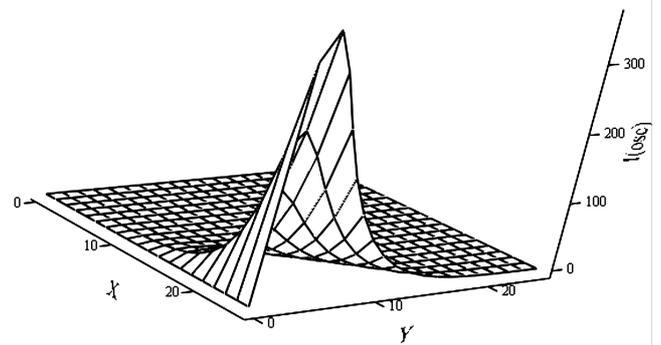


Fig. 7. Dependence of normalized ASE (on I_{lum}) in the EMLS amplifying medium on the gain ($X = \alpha_{\text{gain}}$) and scattering ($Y = S$) coefficients and the given interaction length L

saturation is typical of the amplification process also, the experimentally registered fluently running narrowing vs pumping power (Figs. 1, 2, and 4) indicates, indeed, the gain saturation (and rectangularization) without catastrophes due to the lack of any resonances. It is evident that the transition to the oscillatory lasing for a similar system can be accompanied by a deviation from the smooth spectral distribution in the radiation spectrum [20].

So, the main conclusions of the present work can be summed as follows:

- The strong elastic multiple light scattering, at which a partial confinement of photons within the limits of the area where the amplification exists promotes the development of a stimulated radiation with all tags of ASE or superluminescence like to the known one at the ballistic wave propagation in the transparent laser medium.
- Because the spectral narrowing with increase in the pumping runs without catastrophe tags possible in the case of arbitrary resonances (cavity modes, distributed feedback, or Anderson localization modes), ASE conserves the initial smooth spectral distribution in spite of the strong narrowing.
- The simple theoretical analysis of ASE in EMLS concerning the band width evolution and scaled threshold conditions, that is based upon the approximate determination of a mean free path in EMLS media as $l \approx (N\sigma_{\text{sct}} - \alpha_{\text{gain}})^{-1}$, is completed.
- The ASE power evolution in the Kubelka–Munk approximation for two coupled light fluxes propagating in the amplifying scattering medium is described. The analytic solutions forecast the reasonable behavior of ASE depending on the main medium parameters: gain and scattering and interaction lengths.

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ХАОТИЧНЕ ЛАЗЕРНЕ ВИПРОМІНЮВАННЯ
 БАРВНИКІВ В СЕРЕДОВИЩІ З ПРУЖНИМ
 БАГАТОРАЗОВИМ РОЗСІЯННЯМ
 ТА ПІДСИЛЕННЯМ СВІТЛА

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Резюме

Пружне багаторазове розсіяння світла в підсилюючому середовищі, коли внаслідок розсіяння відбувається часткова локалізація збуджуваної флуоресценції, створює умови для розвитку вимушеного випромінювання суперлюмінесценції (СЛ). Оскільки спектральне звуження полоси СЛ із зростанням підсилення протікає монотонно, без катастроф, які можливі за наявності резонансів довільного походження (моди звичайного або брегівського розподіленого резонатора або моди локалізації Андерсона), СЛ зберігає початковий гладкий розподіл у спектрі, незважаючи на значне звуження. Проведено теоретичний опис спектрального звуження та скейлінгового характеру порога збудження СЛ, що ґрунтується на визначенні середнього шляху вільного пробігу фотонів як оберненої різниці між коефіцієнтами розсіяння та підсилення середовища, $l = (N_{\text{sct}} - \alpha_{\text{gain}})^{-1}$, у наближенні Кубелки-Мунка.