EVOLUTION OF LUMINESCENT EMISSION IN MEDIA WITH AMPLIFICATION AND MULTIPLE LIGHT SCATTERING

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We have carried out the experimental research and analysis of the new phenomenon of a stimulated radiation in media with amplification and multiple light scattering. It is shown that the strong multiple light scattering, at which the partial localization of photons occurs within the scope of a pumping area and the necessary amplification is realized, promotes the development of a stimulated radiation with all signs of superluminescence like that in ballistic modes for transparent laser media. The phenomenon runs without the catastrophes typical of resonator lasers. The localization light paradigms at multiple scattering and the light localization in resonator modes are discussed.

Introduction

The laser oscillation is customarily produced in optically transparent amplifying media placed inside a cavity ensuring a feedback on the highest-Q modes. The latter approximately correspond to the transmission/reflection resonances of an empty Fabry—Perot interferometer which arise owing to a multibeam interference of partial waves with a regular phase delay. In the same media at a large starting amplification, it is possible to excite a laser radiation named superluminescence [1, 2]. The last occurs when a feedback arises due to the spatially random intensity retro-reflection (respectively, there are no conditions for an interference) or the amplification of an initial noise (or a signal in optical amplifiers) takes place without feedback at all in the traveling wave mode [2]. Thus, in all the cases at a linear stage of oscillation (or amplification) evolution, the initial luminescence spectrum of a laser medium experiences the primary amplification in the envelope maximum and, respectively, is deformed or, exactly speaking, is spectrally narrowed. The spectral narrowing of a contour is lasted up to the gain saturation and the formation of a contour with hardly overwhelmed wings of a spectral distribution. Upon the achievement of a gain saturation, the envelope of a spectral contour of the conforming radiation begins to broaden. It is clear that, in the case of the system with a stable cavity, the transformation of a continuum luminescence spectrum is accompanied also by the origination of a discrete oscillation spectrum, a comb of modes with definite intermode spacings.

Thus, in laser amplifiers (without feedback) and in laser oscillators (a feedback exists), the original luminescence radiation is underwent to a sharp transformation in connection with the spectral characteristics and statistics.

The subject of investigation and discussion of our paper is the evolution of the luminescence characteristics of the known laser dyes in a polymer matrix (polyvinylacetate (PVA)) with high concentration (up to 30 wt. %) of scattering centers (fumed silica and other) under nanosecond laser pumping. The studied subject lies in a borderline field of problems of laser physics and optics of multiple light scattering systems. Because both the scientific fields are advanced within the framework of their own paradigms, the first observations of a lasing analog in the media with multiple light scattering were accompanied by a legal surprise of the advocates of conventional laser schools and have spawned hopes of the specialists in multiple light scattering for a new lasing appearance [3, 4]. The reason for different explanations and estimations of this phenomenon is the multiply checked rule that scattering losses in optically transparent laser media increase a lasing threshold, while, on the conclusions of other advocates, the Anderson localization of photons in a multiple scattering medium can become a new lasing appearance [5—8].

However, the localization of photons in a cavity laser mode is a prosaic fact. Moreover, in the case of lasers with a distributed feedback functioning due to the spatially periodic modulation of the complex-valued index of refraction, the laser oscillation is actually the implementation of a $1D$ photon crystal where the phase oscillation condition $\Lambda = l_{\mathrm{free}} = \lambda/2n$ [9]

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practically coincides with the condition for the Anderson localization existence (here, Λ, l_{free}, λ, and n are the spatial period, photon mean free path, wavelength, and refractive index, respectively).

However, in spite of the high level of comprehension and practical implementation of a lasing in the ensembles of microcavities and photon crystals [10—12], the nature of a stimulated light emission in gained multiple light scattering media [16—17] requires, in our opinion, a further investigation.

1. Experimental Observations and Analysis

We have observed and analyzed a number of emission patterns arisen at the pumping by the second harmonic of a laser Nd^{3+}:YAG (λ=532 nm, $\tau_p \approx 20$ ns, and the size of an excitation spot $s \approx 0.5 \div 2$ mm) from samples of PVA (a PVA film with the thickness $t \approx 1 \div 2$ mm) with scattering centers in the range of concentrations 6—30 wt. % (a powder of fumed silica, sapphire, diamond, etc.) activated by laser dyes (R6G, astrophloxyne at a concentration of $\approx 10^{-4} \div 10^{-3}$ mol/l). For the measurement of the true sizes of scattering powder particles used in the sample preparation, a particle-size analyzer LMS-30 (from the Sheishin corporation) was used. The measured widths of the distributions (over the base) were in limits from 8 up to 20 mkm (fumed silica) and 20—100 mkm (sapphire). The measured difference of the mean diameters of particles alongside with the difference of their indices of refraction did not lead to marked changes of the observed emission characteristics.

The light emission measurement from opaque samples activated by dyes was executed in the forward half-plane at different angles of the orientation of a photodiode detector (having the angular aperture of about 10 sq. grades) about the pumping beam. The spectral characteristics of the radiation were measured with a diffraction spectrometer SPM-2 with a resolution of 0.7 nm, a silicon photodiode FD-24 having linear response (3 orders) and a digital voltmeter. Thinner samples ($s < 1$ mm) on the transparent substrate allowed us to excite and to study the light emission in the front and back half-planes simultaneously. The radiation pattern in the front half-plane had approximately a spherical form with a point of birth at the pumping spot on the sample. Owing to large transverse dimensions of samples, no emission was observed in the plane of a layer outside a pumping spot. In Fig. 1, we present the main results of similar measurements reflected the changes of characteristics of the given radiation with increase in the pumping.

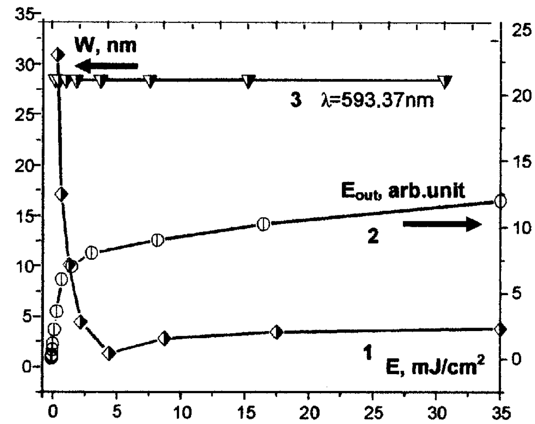

Fig. 1. Evolution of the R6G emission in PVA for 30 wt. % of fumed silica with pumping power: *1* — line-width of the emission band; *2* — emission power in the forward hemisphere under 45° in a solid angle ≈10 grade^2; *3* — spectral position of the emission peak

The radiation has no polarization, despite a linear polarization of the source of pumping, and its spatial coherence is as low as that of a fluorescence source under equivalent conditions of observation (the van Cittert—Zernike theorem). In particular, the radiation did not demonstrate "speckles" clearly observed for spatially coherent beams at the transverse size of a pumping beam ≈1 mm on the surface.

At a sample temperature decrease up to 77 K, the light emission parameter was conserved, but with a somewhat larger emission band narrowing. At the helium temperature $T = 4.2$ K, the radiation was excited on two adjacent (~ 18 nm split) simultaneously narrowing bands.

In Fig. 2, the spectrum widths of a studied emission (normalized on the width of fluorescence spectrum R6G) vs the pumping power density for different concentrations of fumed silica are presented at a constant pumping spot size on samples. This representative behavior of the width with pumping manifests a decrease of the second threshold of excitation of the studied light emission with increase in the scatterer concentration.

Thus, the most typical sign of the evolution of dye emission bands under study was a change of the width and contour of the emission band (curve *1*) with a growth of pumping since its representative value for fluorescence spectra R6G (40—45 nm) decreased up to minimum values (2.5—3.0 nm). The transition zone spreading to almost constant values of the width (by a level of pumping) coincides with a transition zone to a new dependence of the output power of the given

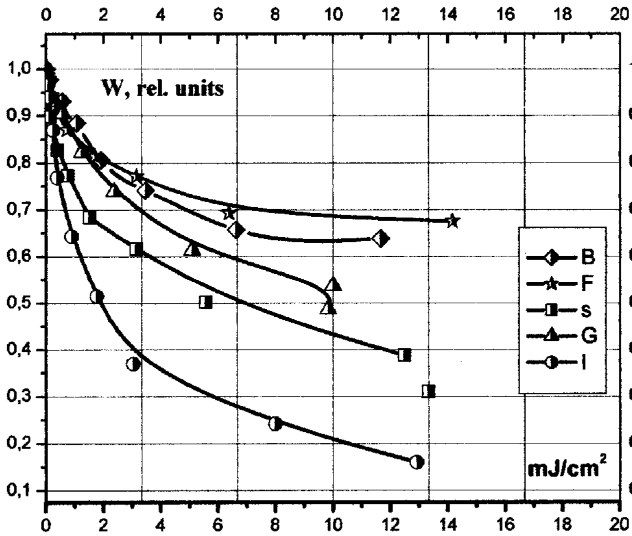

Fig. 2. R6G bandwidth emission evolution with pumping power for 6 different fumed silica concentrations in wt. %: I — 30, s — 24, G — 18, B — 12 , F — 6

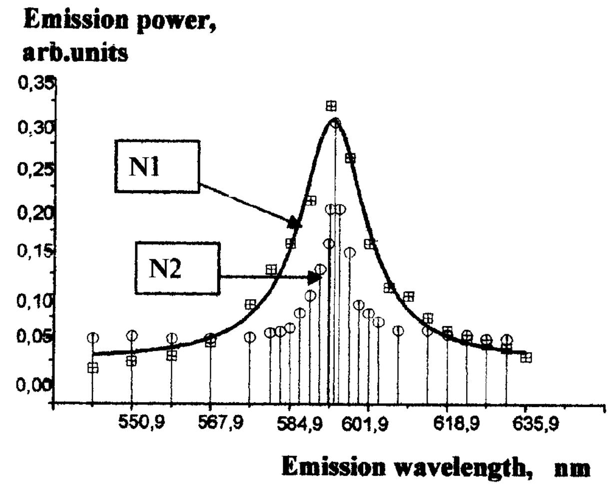

Fig. 3. Form-factor of the emission band in the case of multiple scattering media for 2 different amplification values: N1 corresponds to the middle of narrowing and N2 corresponds to the end of narrowing on line 1 of Fig. 1

radiation on pumping power (curve *2*, Fig. 1). The position of the emission band maximum remained practically invariable in the whole range of available pumping (relation *3*, Fig. 1).

In Fig. 3, the emission bands formed in the transient and second gain saturating areas are shown. For a small pumping, the radiation spectra met the well-known fluorescence spectra of our dyes. The approximation of the experimental contours of emission bands is managed in the best way with the application of the Lorentz function (we applied the nonlinear method of least squares from the package "Origin 6.1").

At last, we present the data on a radiation pattern (Fig. 4) in the medium with multiple scattering on the opaque substrate. It has a similar kind for the horizontal and vertical planes in the laboratory coordinate system. On the transparent substrate at a small absorbance of the medium, the similar hemisphere appeared also in the back half-plane.

Comparing the measured emission characteristics with those for a dye lasing in the optically transparent medium with a regular distributed feedback (a strictly given spatial frequency) or a laser with external stable cavity suggests following resemblances and distinctions:

— The observed emission spectral narrowing is representative for a linear stage of lasing development, and the measured bandwidth in our experiments is comparable to those for conventional pulse dye lasers with nondispersive cavities; the spectral distribution of intensity inside bands has the diffuse character, instead of the discrete one (as in lasers with a cavity), which makes it similar to that inside superfluorescence emission bands excited in transparent laser media without feedback, or in the traveling wave mode [2].

— Consider the onset of narrowing the luminescence spectrum as the first threshold of a stimulated radiation in media with multiple scattering, above which one observes an increase in the contribution of stimulated transitions comparing to that of spontaneous transitions. We note that its value is approximately equal to the dye lasing threshold in optically transparent media. However, all transient area prior to the onset of the second bend in the dependence of the bandwidth on pumping, when there comes a gain saturation, is rather broad on the scale of the pumping power. In typical laser oscillators, the linear stage of oscillation, on which the formation of a laser spectrum from a fluorescence noise takes place, is superposed with the "true" second oscillation threshold characterized by the jump-like transition to a nonlinear gain dependence on the oscillated power.

— The absence of a primary directivity and a spatial coherence of emission in the near-field of diffraction (Fig. 4) and the nonpolarized nature of a stimulated emission at a linearly polarized pumping can be a consequence of the diffusive photon movement in a multiply scattering gain medium, rather than a result of the wave character of the propagation of photons.

The considered resemblances and distinctions in the behavior of conventional lasers and the studied source are in the scope of the paradigm of a laser oscillation and a laser amplification of own luminescence noises [11].

The phenomenon of laser amplification of noises is well known for laser amplifiers as amplified spontaneous emission (ASE) or superluminescence (SL), often arising in optical amplifiers as an undesirable parasitic phenomenon. The transversal mode structure in the case of SL, determining the degree of a spatial coherence of radiation, depends on the Fresnel numbers of an amplification geometry; the spectral content of emission without the quantization of longitudinal modes is characterized by a continuous filling with the half-width dependent on the power of narrowing at a linear stage of amplification.

The amplification in a system with resonant feedback occurring at the implementation of a multibeam interference in Fabry—Perot interferometers and their analogs is given by the expression [11]

$$G(\lambda) = t_1 t_2 g(\lambda)/(1 - r_1 r_2 g^2(\lambda)), \quad (1)$$

where $t_{1,2}$, $r_{1,2}$ are the amplitude transmittance/reflection coefficients of cavity mirrors, $g = d_{m,n} \exp[\alpha(\lambda)L]$ is the amplification for a single passage of a signal through the amplifier without reflection, $\alpha(\lambda)$ is the spectrum-defined gain, $d_{m,n}$ is the real coefficient which is related to diffraction and equal to at most 1, L is the length of the active area equal to a spacing interval between mirrors. It is obvious that the second threshold of a lasing depends on $r_1 r_2 g(\lambda)$ and looks like a catastrophe starting when the denominator vanishes, whereas the beginning of a linear amplification and the narrowing of a noise spectrum have no sharp threshold and can be observed earlier, than the true oscillation begins. Changing the ratio of values $r_1 r_2/t_1 t_2$, it is possible to change the time interval between realizations of these thresholds.

It is clear that, in a high-Q cavity, the basic oscillation threshold is reached first of all. In a resonator without mirrors, the first threshold of laser amplification will be realized without a catastrophe. For a multiple scattering laser medium, it is necessary to answer the following questions:

1. Is the observed emission referred to the case of laser amplification or oscillating?
2. Which is the origin of the quantities L, r_i, and $\alpha(\lambda)$ in the case of a multiple scattering laser medium, and what defines them?

The lack of directivity in a definite solid angle for the observed emission (Fig. 4), being the customary and reference property of a laser oscillator, is not a sufficient condition for denying the oscillatory lasing. For example,

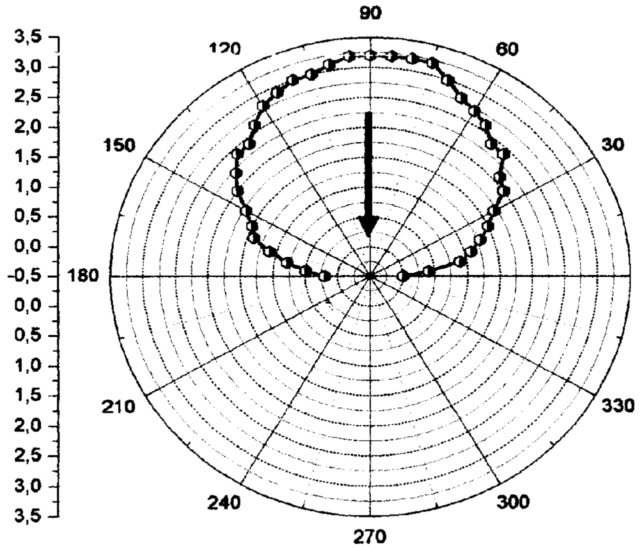

Fig.4. Set diagram for lasing in the multiple scattering media with amplification. (The arrow shows the pumping beam set)

the oscillation in laser material powders which form microcavities with a random space orientation also did not discovered a sharp threshold and directivity [11]. However, for an oscillatory lasing, the spectral narrowing under the excess of a pumping power above the threshold is atypical, because, in this case, the system passes in a nonlinear amplification mode considered as the main condition for steady-state oscillation. Respectively, for laser oscillators with a growth of the pumping power above the threshold, the spectral line-width broadening is observed. At the same time, a noise laser amplifier shows a linear amplification mode and a slow reduction of the spectral width of an amplified noise with a growth in the input power after the first threshold (if the spectral distribution of noise has no rectangular character).

Let's consider the narrowing of the own luminescence band in an optical amplifier, taking $r_i = 0$, $L = \text{const}$, $\alpha(\lambda) = A(\lambda) = \Lambda\ (\lambda)$ in (1). That is, the contours of a gain and a luminescence band are described by equivalent Lorentz distributions. The iterative procedure for the half-width of an emission line on the output of an optical amplifier as a function of the maximum gain was executed with application of the software package "Maple 7". In Fig. 5, the result of computations is given: at the zero amplification, the injection bandwidth is taken to be 500 cm^{-1}. The band gravity center corresponds to 500 nm, and the half-width of a reshaped emission band is equal to the difference of

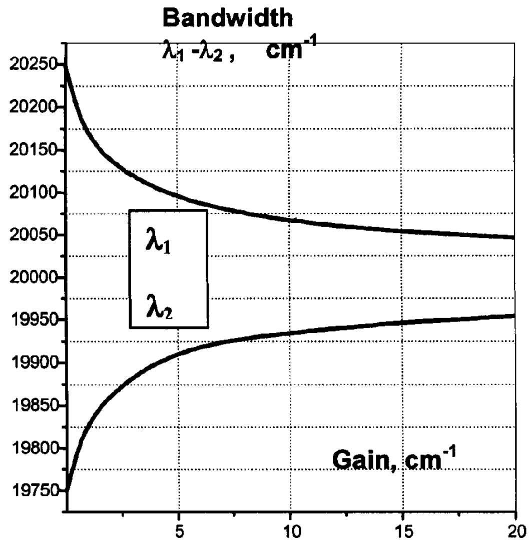

Fig. 5. Calculated dependence of the Lorentz bandwidth emission as a function of the Lorentz gain profile $\alpha(\lambda)L$

wavelengths on the wings of a band (the top and bottom curves in Fig. 5).

It is known that, for linear amplifiers, the product of the maximum amplification and the half-width of a pass-band is conserved [11]. This yields the observed spectral narrowing of SL.

Thus, the light emission by a multi-scattering gain medium in the region of spectral narrowing and prior to the onset of the second threshold can be described as the process of linear amplification in the relevant medium. The area with a stop of spectral narrowing can be considered as the region with gain saturation.

However, the gain saturation is a typical process not only for a oscillatory lasing. Therefore, the actually registered smoothly varying bandwidth vs pumping power (Figs. 1,2) indicates the state of a gain saturation without the powerful contribution of a feedback usually inducing the sharp threshold like a catastrophe. It is possible that the transition to a oscillatory lasing for a similar system can be accompanied by a deviation from the spherically symmetric radiation pattern presented in Fig. 4.

The subsequent analysis of the phenomena of both the laser amplification of noise and a possible laser oscillation should be based on the multiple light scattering mechanism in similar media allowing one to evaluate the efficient length of amplification L and the back-scattering [14—17]. For the logarithmic mean distribution of the particles of fumed silica over sizes, their most probable size $a = 10$ mkm gives $2ka \gg 1$, the condition at which the anisotropy parameter $\delta = \langle\cos\vartheta\rangle \approx 1$ and the mean length of the diffusive transport of a photon $l = l_{sc}/1-\delta$ can become notably more than the mean free path connected with scattering, $l_{sc} = 1/\alpha_{sc}$ [15]. The evaluated values of the light scattering coefficient for fumed silica in PVA in the used samples were in the scope of $(30\ldots150)$ cm^{-1}, which corresponds to l_{sc} from 66.7 to 13 mkm. The value l has sense of a spatial scale for the full chaotization of a wave traveling direction. The mean free path l_{sc} in a material with the mean index of refraction n is connected to the diffusion coefficient of light D in the following way:

$$D = lc/3n = c/3n(1-\delta)\alpha_{sc}. \qquad (2)$$

Here, c/n is the light phase velocity in a medium with a mean index of refraction n. The required effective length of amplification L is the sum of the large number of statistically independent steps N with length l_{sc}. Therefore, the computation of the number N will allow us to estimate of a non-saturated gain in multiple light scattering medium $g(\lambda)$ and to compare it to a value achievable traditionally in the optically transparent media on the base of a dye solution. To provide the laser amplification on each step of scattering, the mean free path l_{sc} should not exceed the reference size $\varnothing$ of a pumped area of the laser medium. Otherwise, such photons will leave from a "diffusive" resonator and will be lost. This positive role of strong multiple scattering is confirmed by a decrease of the observed threshold of spectral narrowing of the luminescence band for a row of fumed silica concentrations of 6—12—18—24—30 wt. % in dyed PVA (Fig. 2). From the condition

$$l = \sqrt{D\tau_{lum}} = l_{sc}\sqrt{N} \leq \varnothing_{pump}, \qquad (3)$$

we get the expression for the effective path of amplification

$$L_{eff} = l^2/l_{sc} = l_{sc}/(1-\delta). \qquad (4)$$

Depending on the anisotropy parameter δ, the value of L_{eff} can exceed the mean free path l_{sc} by $10 \div 100$ times and, respectively, bring up the amplification to values comparable with those for optically transparent media. If the pathway L_{eff} is closed in a loop, then one of the possible conditions for the localization of photons and the appearance of a feedback for the laser oscillation will be satisfied. In [16], upon the use of 2D samples, similar self-contained loops $\varnothing \sim$35 mkm were observed with a microscope under the optical excitation of microcrystals ZnO. However, the probability of the formation of closed circuits seems to be notably lower than that for open chains in the random realizations.

Respectively, the probability of a stimulated radiation such as superluminescence or ASE seems to be higher than that of a non-resonance lasing on the basis of systems with closed paths.

There is one more possibility for a lasing in the multiple light scattering medium at the expense of the mentioned distributed feedback [9] of a random type. Actually, the existence of a small peak of coherent back-scattering with a weak localization serves as a definite indication of the possibility of a similar DFB lasing. In [15], a coherent scattering peak was registered in a gain light scattering medium with contrast above the background much exceeding analogs in media without gain. However, since the efficiency of back-scattering hardly depends in this case on resonance conditions (the conservation of the spatial distance $\Lambda = m\lambda/2$ between scattering centers) and sharply drops for $m > 1$, the probability of such events is not high. It seems reasonable to search for a similar implementation in systems with Rayleigh multiple scattering ($ka < 1$), for the monodispersed type of scatterers and with a possible higher difference of refractive indices for the matrix and scattering centers. Both requirements are necessary for the increase of the backward wave power in a circuit with a random realization of equidistant scatterers.

Conclusion

We have conducted the experimental research and analysis of the new phenomenon of a stimulated radiation in media with amplification and multiple light scattering. It is shown that the strong multiple light scattering, at which the partial localization of photons occurs within the limits of a pumping area and the necessary amplification is realized, promotes the development of a stimulated radiation with all signs of a superluminescence like that in ballistic modes for transparent laser media. The phenomenon runs without the reference catastrophe signs for resonator lasers. However, it contains reference regions of the first and second thresholds of the line-width evolution, which are rather different from those of resonator lasers by the pumping power level. The localization light paradigms at multiple scattering and the light localization in resonator modes are discussed. Of a certain interest is the search for analogies with space lasers.

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ЕВОЛЮЦIЯ ЛЮМIНЕСЦЕНТНОГО ВИПРОМIНЮВАННЯ У СЕРЕДОВИЩАХ З ПIДСИЛЕННЯМ ТА БАГАТОРАЗОВИМ РОЗСIЯННЯМ СВIТЛА

Є.О. Тихонов, В.П. Ящук, В.I. Безродний

Р е з ю м е

Проведено експериментальне дослiдження та аналiз нового явища — вимушеного випромiнювання у середовищах з пiдсиленням та багаторазовим розсiянням. Показано, що сильне багаторазове розсiяння, за якого у межах областi накачки вiдбувається часткова локалiзацiя фотонiв i створюється необхiдне пiдсилення, сприяє розвитку вимушеного випромiнювання з усiма ознаками суперлюмiнесценцiї у балiстичних режимах для прозорих лазерних середовищ. Явище вiдбувається без катастроф, характерних для лазерiв з резонаторами, але мiстить характернi областi першого та другого порогiв еволюцiї ширини смуги, якi сильно рознесенi за рiвнем пiдсилення (накачки). Обговорено парадигми локалiзацiї свiтла в умовах багаторазового розсiяння i локалiзацiї у резонаторних модах.