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Mode distribution in coherently amplifying random media

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Abstract

We investigate the statistics of lasing modes in a disordered one-dimensional active system. Dielectric scatters are randomly embedded in an amplifying medium to increase the optical length of photon. The lasing mode of each individual sample is obtained numerically by carefully locating the frequency and the threshold gain at which the reflection and the transmission coefficient first become divergent, a characteristic of the oscillation pole. As the density of scatters increases, the characteristic threshold gain for lasing decreases while the distribution of mode frequency broadens around the spontaneous emission frequency of the gain medium. The averaged gain threshold is indeed found to be proportional to the localization length, signifying the enhancement of light amplification due to multiple scattering. Interestingly, we find that the actual value of the gain threshold larger than previous theoretical predictions by a factor of 3–5, in agreement with a recent experiment. © 2001 Published by Elsevier Science B.V.

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Recent observation of laser-like emission in solutions containing dye molecules and titania (TiO_2) particles [1–8] strongly indicate that random gain media can be a novel effective revenue for coherent light generation. The latest experiment [9] on semiconductor powder with both high gain and strong scattering provided the strongest evidence to date for the existence of coherent laser action in a disordered amplifying medium. In these experiments, an optically pumped active medium showed an onset of intensified output of light at certain pumping intensity accompanied by spectrum narrowing, a characteristic of a coherent laser system. These phenomena are of great interest for their implications in both fundamental physics and potential applications. The conventional coherent laser systems rely upon a well-defined resonant mode enforced by a feedback mechanism, provided either by optical mirrors placed at the two ends or through distributed feedbacks over the entire system. The random laser system, however, is expected to derive its feedback from random multiple scattering of photons by disorder within the microstructure [10]. The nature of the randomness makes the relevant physical quantity such as the mode frequency and the threshold for lasing action configuration dependent and highly statistical, analogous of the well-known sample-to-sample fluctuations in the conductance of disordered mesoscopic electron systems [11]. The possibility of multimode lasering in random laser was also suggested based on an analysis of the emission spectrum in

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the experiment [9]. In this work, we investigate numerically the lasing mode frequency and threshold distribution in a one-dimensional (1D) disordered dielectric micro-structure embedded in a gain medium.

Static disorder does not preclude coherence of waves in the sense that phase information, although highly altered by the interaction of wave with random media, is still kept intact, a lesson we learned in the context of electron localization in disordered systems. Multiple scattering of light from disorder, however, increases the optical path of photon and thus can enhance amplification [10] when the medium is active. This idea formed the basis of the earlier theoretical studies [10,12–16] on light amplification in random gain media. Based on the diffusive description of the photon propagation, the studies predicted a reduction of the gain threshold for laser action in random system due to the enhancement of the optical path. A recent study [16] also verified the narrowing of spectrum when the gain approaches the threshold. However, the neglect of important phase information and interference effects in the wave propagation means that the true lasing phenomenon and the statistical nature of the problem cannot be addressed by such approaches.

The coherent nature of the light propagation has only begun to be taken into account recently [17–21]. Starting from the time-independent wave equation accounting for both disorder and amplification (or absorption), these studies focused on the transmission properties and the interplay between localization of light from disorder and amplification (absorption) of light from gain (loss). Unfortunately these studies do not directly address the issue of the lasing mode and the gain threshold in the experiment [1–4,9]. The threshold was only inferred indirectly from the divergence of the averaged reflection or transmission coefficient [18,19].

In this study, we investigate directly the mode frequency and gain threshold of the amplified light and their distributions for a random gain medium. Note that a complete description of lasing actions requires full quantum mechanical descriptions of spontaneous emission and coupling of light with matter [22,23]. All lasing phenomena such as spectra narrowing and distribution changes are associated with the dynamic interaction between the amplified light and the gain media. The gain depends on the population inversion and thus couples to the lasing light intensity, making the problem nonlinear. However, below threshold, the gain can be taken approximately as a fixed value, independent of the light intensity, thus making classical electrodynamic theory a good approximation.

The system we investigate is essentially a one-dimensional simplification of the real experiment. It consists of fixed-thickness dielectric layers (titania with $\varepsilon = 12$) randomly placed in a gain medium (solutions of dye molecules). The whole system is placed inside a thin glass container. The overall length of the system is taken to be $L = 590\ \mu\text{m}$. The distance between the neighboring dielectric layers obeys the Poisson distribution, $P(d) = \mathrm{e}^{-d/d_0}/d_0$, with the average distance given by d_0. The thickness of the dielectric layers is $d_\mathrm{a} = 270\ \mu\text{m}$.

In semi-classical theory, the output of the amplified spontaneous emission is the sum of the contributions from emission at all segments of the active region. The laser oscillation condition of the amplified system is given by the pole of the Green's function of the linearized system. A distinct property of these poles is the divergence of the total reflection and transmission coefficient [23–25]. To locate these poles, we have calculated the total transmission and reflection coefficient for a random configuration of dielectric layers for different frequency and gain strength. The coherent wave propagation of light in the system is obtained by solving the transfer matrix equation that relates the wave amplitudes on the two sides the dielectric layers in the frequency domain.

The propagation of light at frequency ω from spontaneous emission is governed by the following wave equation for the electric field $E(z)$:

$$\frac{\mathrm{d}^2E(z)}{\mathrm{d}z^2} + \frac{\omega^2}{c^2}\varepsilon(z)E(z) = 0, \tag{1}$$

where the dielectric constant $\varepsilon(z) = \varepsilon'(z) + \mathrm{i}\varepsilon''(z)$ is complex with the imaginary part signifying amplification ($\varepsilon'' < 0$) or absorption ($\varepsilon'' > 0$). ε' takes either the value of nanoparticles or that of the solution. The active region, the solution, is described with a constant (uniform pumping) negative ε'' which leads to light amplification.

The pumping light density is assumed to be uniform over the whole sample, thus the gain in the solution varies only in frequency described by the spectrum function of the spontaneous emission with the standard Lorentzian distribution, $\varepsilon'' = \varepsilon''_0/(1 + ((\lambda - \lambda_0)/\Delta\lambda)^2)$, where ε''_0 is the maximum gain which is proportional to the pumping light intensity. $\lambda_0 = 620$ nm and $\Delta\lambda = 12$ nm are the wavelength and the line width of the spontaneous emission of the dye molecule, respectively. The container glass is assumed to be 50 nm thick with $\varepsilon = 3$.

The system consists of alternating segments of dielectric (nonactive) particles and solutions (active). The wave solution in each segment has the form $E(z) = A_n \exp[ik_n(z - z_n)] + B_n \exp[-ik_n(z - z_n)]$, describing plane waves propagating in both directions. Eq. (1) can be solved by the transfer matrix method, with the transfer matrix relating the wave amplitude $\{A_n, B_n\}$ between the two sides of the interface. The total transmission and reflection coefficient can then be obtained in a straightforward manner by multiplying the transfer matrix of each layer and solving for the amplitudes of the reflected and transmitted waves. We note that our recent study [20,21] indicates that time-independent wave equations may not be able to describe the transmission and reflection in disordered media at large gain, when the output from the linear wave equation actually diverges. However, we believe that the lasing threshold still correctly given by the poles of the transmission coefficient in the complex dielectric plane from the time-independent wave equation.

For each individual configuration, as the overall pumping intensity increases, the spontaneous emission from the dye molecules will increase with its spectrum distributed around the frequency ω_0. When the pumping reaches certain level, a singular mode, specific to that configuration, will achieve lasing threshold, characterized by the divergence of the transmission and reflection coefficient. Fig. 1a shows how the transmission coefficient changes as

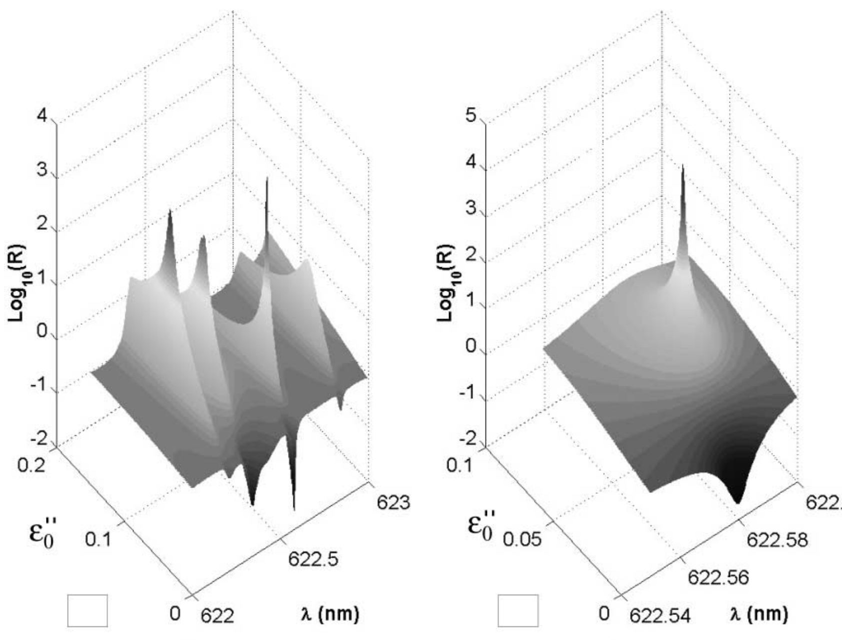

Fig. 1. The reflection coefficient at different frequencies and different gain for a random system with alternative dielectric and gain media. The possible lasing modes are indicated by the divergence of both the reflection and transmission (not shown) coefficient. The average distance between the randomly placed dielectric nanoparticles are 110 μm. (a) Several modes around the spontaneous emission wavelength, $\lambda = 620$ nm, and (b) the mode corresponding to the lowest gain threshold.

the gain increases for a particular random configuration generated with $d_0 = 110$ nm. The transmission coefficient and reflection coefficient are calculated with the transfer matrix method for different frequency and gain, assuming the output is finite. It is obvious from Fig. 1a that many modes exist around the spontaneous emission frequency ω_0. These modes are extremely narrow in spectrum when the gain approaches threshold values. Consequently spectrum narrowing will be observed in the output light.

It is interesting to compare our system with a distributed feedback laser when there is no disorder. We note that a distributed feedback laser system also exhibit similar behavior as shown in Fig. 1. The difference is that the modes are regular in frequency domain in a distributed feedback laser and their fields are extended over the whole system. For a random system, the field will become more and more localized at some spatial location as scatter density increases. The feedback mechanism is provided by local recurrent scattering events.

It is also important to note that the reflection coefficient shown in Fig. 1 is valid only before the gain reaches threshold. Beyond threshold, the time-independent field equation Eq. (1) is not adequate to describe the behavior of the field [20,21]. The fall of reflection and transmission coefficient for large gain is an artifact of the assumption of a stable finite output which is no longer true above threshold within the linear theory.

Among these different modes, the one with the lowest threshold (Fig. 1b with wavelength close to 622.58 nm) is a true lasing mode because it will lase first and is, therefore, the most relevant to the experiment. Once the system goes above that threshold, the output power is totally dominated by the coherence of the laser at that mode frequency. However, below the threshold, all frequencies within the Lorentzian window of the gain medium contribute. Note that because the distribution of the scatters is random, both the lasing frequency and its associated threshold will be configuration dependent.

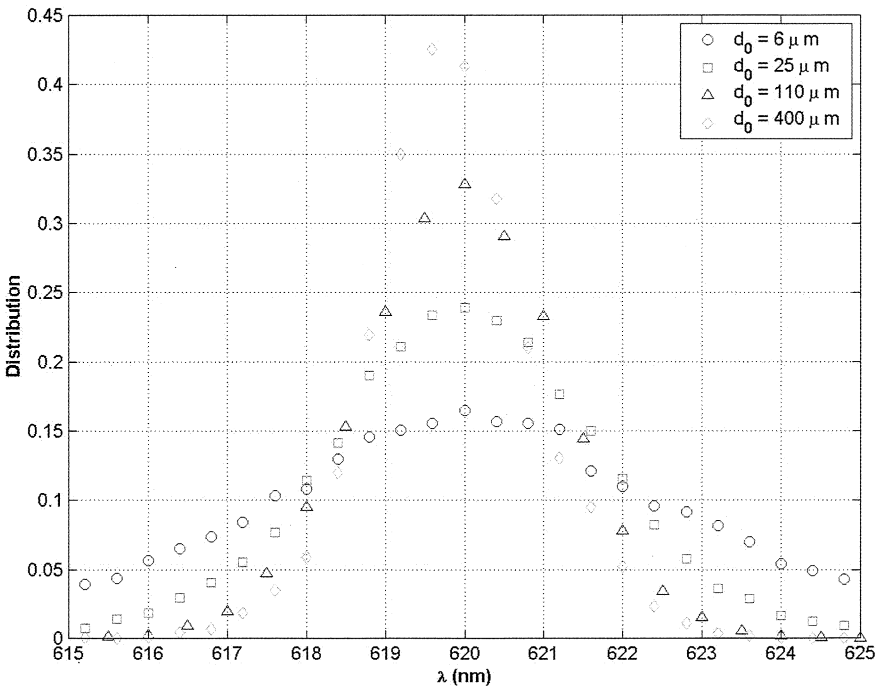

Fig. 2. The mode distribution in a disordered system with gain. The spontaneous emission wavelength is centered around $\lambda = 620$ nm. The average distance between the randomly placed dielectric nanoparticles (TiO_2) are 400, 110, 25 and 6.4 μm. The total length of the system is 550 μm. The data represents statistics from many thousands of independent random configurations.

To investigate the interplay between disorder and amplification, we investigate the distribution of the mode and the corresponding threshold over an ensemble of many configurations, for several density of dielectric layers, with average separation $d_0 = 400$, 110, 25, and 6 μm. Many independent configurations are examined by sweeping the frequency and gain with small increment to locate the mode with the lowest lasing threshold. The resultant distribution of mode frequency is shown in Fig. 2, for the four density of scatters. Clearly the distribution centered around 620 nm, reflecting basically the Lorentzian dependence of the gain with frequency. Interestingly, the distribution broadens as the particle density increases. Physically, with increased disorder, highly localized modes with frequency away from the center of the spontaneous emission began to become more and more competitive compared with those mode around the center frequency. Their enhanced optical paths can more than compensate the reduction in gain value from the Lorentzian distribution. This, we believe, is the underline reason for the broadening of the mode distribution in frequency.

More significantly, there is a reduction of threshold gain as scattering increases. This is supported by examining the corresponding distribution in threshold gain (Fig. 3). Clearly, the weight of the distribution shifts towards lower gain value as the density of the TiO_2 nanoparticle increases, signifying enhancement of the optical path for photon due to scattering and reduction of threshold gains. The shape of the distribution also changes from a well-defined distribution around some threshold at low density to a more Poisson-like distribution with a broad tail at higher scatter density. The spike at $\varepsilon_0 = 0.12$ for the distribution at $d_0 = 400$ μm is an artifact due to the dilute nature of the scatters.

An important question is how this reduction of threshold gain is related to the increases of localization as the scatter density increases. Theoretically, we expect that the gain threshold should be inversely proportional to the localization length. In Fig. 4, we show in a log–log plot how the mean, the

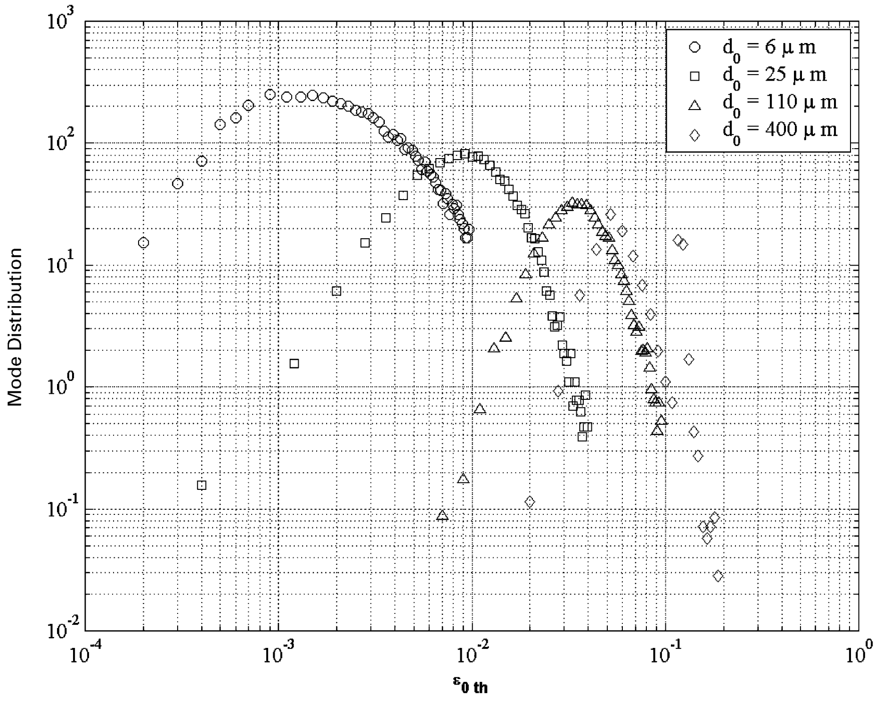

Fig. 3. The lasing threshold gain distribution in a disordered system with gain, corresponding to the system in Fig. 3.

median, and the most probable (peak) threshold gain of the distribution vary with the localization length. The localization length L_c is calculated with the same transfer matrix technique with the relation $T \sim e^{-2L/L_c}$ for long 1D systems.

Theoretically, one expects the lasing action to occur when the system size L is larger than the threshold length [18], $L_{th} = \sqrt{l_g L_c}$, where the gain length $l_g = (1 + d_0/d_a)\lambda\sqrt{\varepsilon_b}/(2\pi\varepsilon'')$. The additional factor is a correction from the geometry consideration since gain exist only in the solution. Equating L_{th} with the sample size L leads to a relationship between ε''_{th} and L_c. Our data are consistent with the theoretical expectation that the gain $\varepsilon''_{th} \sim L_c^{1/2}$. The magnitude of the most probable threshold gain, ε''_{0th} however, is about a factor of 3–5 larger than the theoretical prediction. It is worse if we use either the mean or the median value for the threshold. A recent experiment also found that the measured critical volume was about factor of 5 larger than the theoretical estimate [9]. Given the fact that the theory considers only gain without any frequency dependence while our gain in Fig. 4 is the maximum gain of the Lorentzian distribution, the agreement is remarkable.

In conclusion, the threshold and mode frequency distribution of the random gain system is investigated by taking into account the coherent nature of the light. The distribution of threshold shifts towards low value of gain while the distribution of the mode frequency broadens when the density of the scatter increases. It is interesting to investigate experimentally and theoretically whether true lasing action can be achieved in random gain systems. Whether it is possible to achieve a stable coherent light generation through such a mechanism is still open. However, given the fact that in strongly disordered systems different modes may correspond to different spatial locations, mode coupling may not be an important factor in random lasing systems.

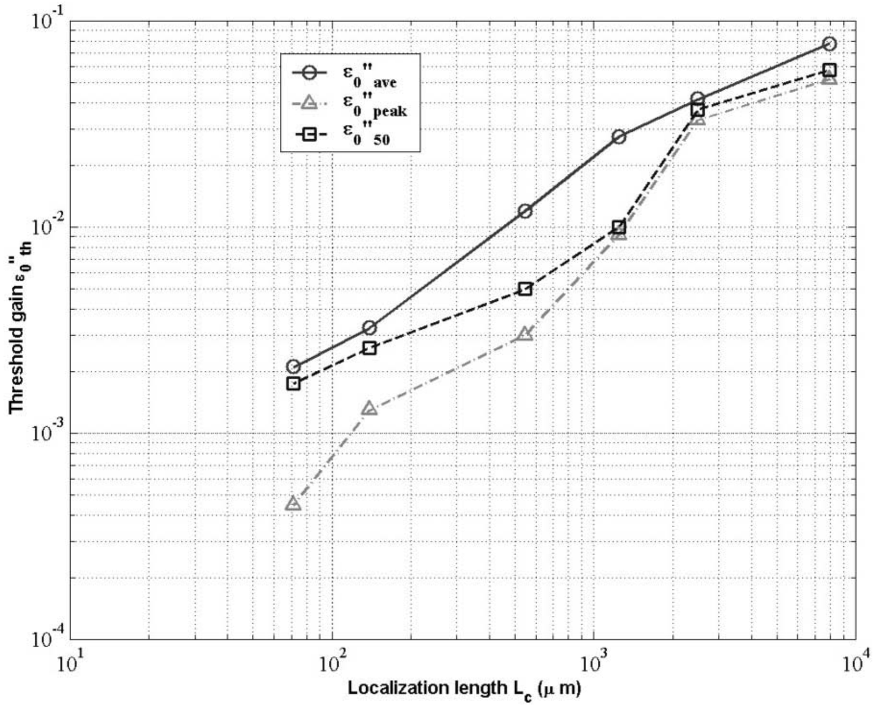

Fig. 4. The mean, the median, and the most probable gain threshold as a function of the localization length, for several systems with different particle density. The average distances used are $d_0 = 3.2$, 6.4, 25, 55, 110, and 400 μm. The data are consistent with the theoretical prediction $\varepsilon''_{th} \sim \sqrt{L_c}$ but the magnitude is at least 5 times larger than the theoretical estimate.

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References

[1] N.M. Lawandy, R.M. Balachandran, A.S.L. Gomers, E. Sauvaln, Nature 386 (1994) 436.
[2] N.M. Lawandy, R.M. Balachandran, Nature 373 (1995) 204.
[3] R.M. Balachandran, N.M. Lawandy, Opt. Lett. 20 (1995) 1271.
[4] W.L. Sha, C.-H. Liu, R.R. Alfano, Opt. Lett. 19 (1994) 1922.
[5] D.S. Wierma, M.P. van Albada, A. Lagendijk, Nature 373 (1995) 203.
[6] A.Z. Genack, J.M. Drake, Nature 368 (1994) 400.
[7] Daozhong Zhang et al., Opt. Commun. 118 (1995) 462.
[8] W. Zhang, N. Cue, K.M. Yoo, Opt. Lett. 20 (1995) 1023.
[9] H. Cao et al., Phys. Rev. Lett. 82 (1999) 2278.
[10] V.S. Letokhov, Zh. Eksp. Teor. Fiz. 53 (1967) 1442 [Sov. Phys. JETP 26 (1968) 835].
[11] Ping Sheng, Introduction of Wave Scattering, Localization and Mesoscopic Phenomena, Academic Press, New York, 1995.
[12] E. Akkermans, P.E. Wolf, R. Maynard, Phys. Rev. Lett. 56 (1986) 1471.
[13] A.Yu. Zyuzin, Europhys. Lett. 26 (1994) 517.
[14] A.Yu. Zyuzin, Phys. Rev. E 51 (1995) 5274.
[15] A.Yu. Zyuzin, JETP Lett. 61 (1995) 990.
[16] S. John, G. Pang, Phys. Rev. A 54 (1996) 3642.
[17] P. Pradhan, N. Kumar, Phys. Rev. B 50 (1994) 9644.
[18] Zhao-Qing Zhang, Phys. Rev. B 52 (1995) 7960.
[19] C. Beenakker et al., Phys. Rev. Lett. 76 (1996) 1368.
[20] X. Jiang, C.M. Soukoulis, Phys. Rev. B 59 (1999) 6166.
[21] X. Jiang, Qiming Li, C.M. Soukoulis, Phys. Rev. B 59 (1999) R9007.
[22] A.E. Siegman, Lasers, University Science Books, California, 1986.
[23] A. Yariv, Optical Electronics in Modern Communications, Oxford University Press, Oxford, 1997.
[24] C.H. Henry, J. Lightwave Technol. LT-4 (1986) 288.
[25] T. Makino, J. Glinski, IEEE J. Quantum Electron. 24 (1988) 1507.