Two-Photon Pumping of a Random Laser

Alexander L. Burin, Hui Cao, and M. A. Ratner

***Abstract*—Random lasing originates from pumping disordered materials by high-intensity and high-frequency light. The performance of random lasers is restricted due to the absorption of pumping light within the interfacial layer. The lifetime of optical modes there is short due to the emission of photons to the outside, and it is hard to pump them sufficiently for lasing. The opportunity to excite the modes far from the material surface using two-photon absorption is investigated within the diffusion model. The authors show that lasing requires lower pump power for the two-photon pumping mechanism under conditions of negligible absorption of the emitted light. Experimental implications and restrictions are discussed.**

***Index Terms*—Inhomogeneous medium, random laser, two-photon absorption.**

RANDOM lasing has attracted much attention since its theoretical prediction by Letokhov [1], more than 30 years ago. It has a certain fundamental interest as the manifestation of coherent phenomena in random media with no electronic analog. Experimental demonstrations of amplified spontaneous emission from mirrorless random media [2] and recent discoveries of coherent random lasing from ZnO nano-powder [3] and disordered organic polymers [4] suggest that this effect can be used to construct a cheap and efficient microsize source of coherent light [5]. However, practically the pump intensity required for random lasing (lasing threshold) is so high ($I_{\rm thr} \geq 1$ MW/cm^2 [3]) that the system is not suitable for any real application.

A challenging problem is how to reduce the lasing threshold by orders of magnitude. In this paper, we examine the opportunity to do so using a two-photon pumping scheme. One can characterize light transport in an active random medium by its transport mean free path l_T (approximately the distance passed between two scattering events) and the gain length l_g (the average distance the photon travels before it stimulates the emission of one more photon). Lasing takes place when the spontaneously emitted photon stimulates the emission of one more photon before leaving the active domain. This happens when the random walk path length of a photon within the medium of size L reaches the gain length [1]

$$\frac{1}{l_g} = \frac{l_T}{L^2}. \tag{1}$$

Recent work on ZnO nano-powder [3], solid solutions of TiO_2 particles and rhodamine dye in PMMA [2], [3], [6], and some molecular materials [4] suggests experimental realizations of random lasing. The lasing instability emerges at sufficiently high pump intensity when the gain rate for some optical modes exceeds their decay rate, caused by light leakage or absorption [7], [8]. This is equivalent to the criterion (1), when the diffusion approximation is valid. Although it was demonstrated in [8] that the diffusion model fails to describe lasing in low-dimensional open systems, the situation is much better in case of closed geometry. The numerical analysis of the lasing modes behavior in a two-dimensional model of resonant dipoles formed by infinite rods, with polarization parallel to the plane (TE modes) [9] confirms the diffusion model validity to predict the lasing threshold (1). The analytical random matrix model [7] also supports the diffusion approximation.

The absorption of pumping light restricts the size L of the active part of the medium to the penetration length within the interface region. Light leakage is very strong there, so that the lasing threshold, i.e., the minimum pump intensity needed to reach lasing, is high. Pumping deep into the sample interior can reduce it remarkably because in that case the emitted light spends much more time in the active medium, as was demonstrated in several modeling considerations [8], [10]–[12]. In fact, even within the diffusion picture (1), simultaneous increase of the medium size $L \to aL$, $a > 1$ with the proportional increase of the gain length $l_g \to al_g$ (conserving the total absorbed pumping energy $\propto L/l_g$) reduces the lasing threshold (1).

Lowering of the random lasing threshold is very significant for fundamental and practical applications [5]. Therefore, any change of the pumping mechanism permitting the excitation of deep optical modes is of the great interest. One possible scenario, realized experimentally for TiO_2-dye solution by Zacharakis *et al.* [13], is based on using a two-photon pumping mechanism. Two-photon absorption is weaker than single photon absorption and the light goes much deeper into the material. The first experiment on a two-photon pumped random laser has been reported already [13]. The future development of this method for other systems is promising.

In this work, we investigate the lasing threshold in the regime of two-photon pumping within the diffusion model [1], [6] that is adequate to describe the regime of incoherent feedback lasing, similar to that observed in [13]. Our calculations permit direct comparison of the lasing thresholds for single and two-photon

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pumping. We demonstrate that two-photon pumping is generally more efficient than single photon pumping. The ratio of lasing thresholds for single-photon and two-photon pumping is defined by the ratio of corresponding scattering lengths for the pumping light of the initial frequency ω_p and the half-frequency light $\omega_p/2$ for two-photon pumping. This ratio is expected to be less than unity. Effects of deviations from the diffusion model due to mode fluctuations [8] and localization effects [10], [11] and practical and experimental restrictions and implications of multiphoton pumping are discussed.

To describe the occurrence of lasing in different pumping regimes we use the diffusion formalism suggested in [1]. The applicability of the diffusion model to both pumping and emitted light is assumed. This means that the scattering lengths of pumping and emitted light are smaller than their absorption or gain lengths. The gain length is determined by population inversion, which is proportional to the density of absorbed pump power for single photon and two-photon absorptions. The absorption of emitted light is neglected. This is reasonable, for instance, at the dye emitting frequency (rhodamine dye absorbing and emitting bands have almost zero overlap; see also arguments in [8], based on the experimental data). The calculations are done in parallel for both cases to enable direct comparison of results. They are performed in the steady state regime and for the semi-infinite medium $(x > 0)$ pumped by a laser pulse of the infinite diameter. This simple case can be resolved analytically, which is convenient for qualitative comparison.

The propagation of pumping light of intensity I_{01} (or I_{02}) for single (or two-photon) regimes obeys the diffusion equations with the absorption

$$0 = D_1 \frac{d^2 I_1}{dx^2} - \gamma I_1, \quad I_1(0) = I_{01} \tag{2}$$

for the single-photon pumping and

$$0 = D_2 \frac{d^2 I_2}{dx^2} - \Gamma I_2^2, \quad I_2(0) = I_{02} \tag{3}$$

for two-photon pumping, where D_1 and D_2 are diffusion coefficients for pumping light in the medium for single and two-photon pumping, respectively, γ and Γ characterize single- and two-photon absorption inside the medium. It is clear that the diffusion coefficient D_2 is greater than D_1 because the half-frequency light scattering is much weaker than that for the high frequency light that is normally close to the Mie resonance for scattering particles [8].

The solutions for the diffusion equations in the semi-infinite medium are straightforward and given by

$$I_1 = I_{01} \exp\left(-\sqrt{\frac{\gamma}{D_1}} x\right) \tag{4}$$

for the single-photon case and

$$I_2 = \frac{I_{20}}{\left(1 + \sqrt{\frac{\Gamma I_{20}}{6 D_2}} x\right)^2} \tag{5}$$

for the two-photon case. It is important that the pump intensity decreases with the depth x much more slowly for two-photon pumping.

The x-dependent gain coefficient can be expressed through the absorbed pump power as

$$g_1(x) = \eta\gamma I_1 = \eta\gamma I_{01} \exp\left(-\sqrt{\frac{\gamma}{D_1}} x\right) \tag{6}$$

and

$$g_2(x) = \eta\Gamma I_2^2 = \eta\Gamma \frac{I_{20}^2}{\left(1 + \sqrt{\frac{\Gamma I_{20}}{6 D_2}} x\right)^4} \tag{7}$$

for single- and two-photon pumping, respectively. Here η is the proportionality coefficient between the absorbed power and the gain rate. This is the simplest assumption for the relationship between the absorbed intensity and the gain strength. One should note that in the regime of saturated absorption the pump energy can go deeper and the consideration should be modified [9]. We do not think that this saturation effect is significant for weak two-photon absorption.

The propagation of the emitted light within the medium $x > 0$ can be described by the diffusion equation with gain

$$\frac{\partial I}{\partial t} = D \frac{\partial^2 I}{\partial x^2} + g(x) I, \quad I(0) = 0, \quad I(\infty) < \infty \tag{8}$$

where D stands for the diffusion coefficient of the emitted light. In both cases of single-photon and two-photon pumping the emission occurs at the same frequency defined by the resonant transition (e.g., exciton resonance for ZnO particles [3]). The zero boundary condition at the sample interface $x = 0$ corresponds to the regime of full absorption. For real materials it should be slightly modified (see e.g., [6]), but this modification is negligible in the case of the short scattering length compared to the characteristic penetration depth and the weak surface reflection. The gain rate g and the gain length l_g can be expressed through each other as $g = v/l_g$, where v is the speed of light within the medium. We consider only this simple case, although in the opposite regime of the strong surface reflection the results for the single and two-photon pumping change similarly.

Lasing instability emerges at sufficiently large gain, when (8) has a solution exponentially increasing with the time $I(t) \propto \exp(\lambda t)$, $\lambda > 0$ (this regime does not take place in reality because of the gain saturation, but gives an estimate for the lasing threshold). Formally, (8) is similar to the Schrodinger equation with imaginary time. The lasing instability is equivalent to the formation of the bounded state in the attractive potential $-g(x)$ having the zero amplitude at $x = 0$. At the lasing threshold the increment ("energy") is zero $(\lambda = 0)$, and (8) becomes time independent, while at larger gain (pump intensity) it grows exponentially with the time. The time-independent solution of (8) having the appropriate behavior at $x \to \infty$ can be found for both single- and two-photon pumping (6) and (7). In the case of single-photon pumping the solution can be expressed through the zeroth order Bessel function

$$I_{\mathrm{em1}}(x) \propto J_0\left(\frac{2\exp\left(-\frac{\lambda_1 x_1}{2}\right)}{\lambda_1}\right)$$
$$\lambda_1 = \sqrt{\frac{D}{\eta I_{10} D_1}}, \quad x_1 = x\sqrt{\frac{\eta\gamma I_{10}}{D}} \tag{9}$$

while in the two-photon case it takes the form

$$I_{\mathrm{em2}}(x) \propto (1+\lambda_2 x_2)\sin\left(\frac{1}{\lambda_2(1+\lambda_2 x_2)}\right)$$
$$\lambda_2 = \sqrt{\frac{D}{6\eta I_{20} D_2}}, \quad x_2 = x\sqrt{\frac{\eta\Gamma I_{20}^2}{D}}. \tag{10}$$

The lasing instability emerges for the minimum intensity satisfying the zero boundary condition at $x = 0$. This happens at pumping intensities

$$I_{10} \approx 1.44\frac{D}{\eta D_1} \tag{11}$$

for single-photon pumping and

$$I_{20} = \frac{\pi^2}{6}\frac{D}{\eta D_2} \approx 1.64\frac{D}{\eta D_2} \tag{12}$$

for two-photon pumping. Both thresholds are independent on the absorption strength for the pumping light. This is because the increase of absorption leads on the one hand to an increase of the absorbed intensity (increase of gain) and on the other hand to a decrease of the size of the gained domain. In this approach, these two effects exactly cancel each other.

For both situations the lasing threshold turns out to be inversely proportional to the diffusion coefficients for the pumping light. To compare the threshold intensities one can express the diffusion coefficient using the kinetics formula

$$D \approx \frac{v l_T}{3} \tag{13}$$

where v is the speed of light and l_t is the mean-free path in the random medium. The speed of light is the same for both pumping regimes, while the scattering length (defined by the inverse of scattering cross section) is different for the single-photon pumping (frequency ω_p) and the two-photon pumping (frequency $\omega_p/2$). For example, when the scattering takes place in the long wavelength limit for both pumping schemes the scattering cross section has the Rayleigh frequency dependence $\sigma \propto \omega^4$ so it is larger for the single photon pumping by a factor of 16. Accordingly, the scattering length (and the diffusion coefficient) is larger for the two-photon pumping by the same factor. Then the lasing threshold for the two-photon pumping should be one order of magnitude smaller than that for the single-photon pumping. A similar relationship is expected to hold even for different scattering mechanisms when the pumping frequency ω_p is close to the scattering particle Mie resonance while $\omega_p/2$ is much lower.

One can easily extend the above arguments to the general case of n-photon pumping. Then, the absorption term in the two-photon (3) has to be replaced with the general n-photon term $\Gamma_n I_n^n$. One can estimate the gain rate and the pumping domain depth as

$$g_n \sim \eta\Gamma_n I_{0n}^n, \quad 0 < x < L_n$$
$$L_n \sim \sqrt{\frac{D_n}{\Gamma_n I_{0n}^{n-1}}} \tag{14}$$

where I_{0n} is the pumping intensity and D_n is the diffusion coefficient for n-photon pumping. Then, using the approximate estimate (1) for the lasing instability we arrive at a criterion independent of the strength of the n-photon absorption, similar to (11) and (12)

$$I_{n0} \sim \frac{D}{\eta D_n}. \tag{15}$$

Since the diffusion coefficient increases with decreasing frequency $\omega_n \propto 1/n$ the increase of n should reduce the lasing threshold. If we assume that the diffusion is due to the Rayleigh scattering of light, then the diffusion coefficient D_n for the frequency ω_0/n scales as n^4 and we get $I_{n0} \propto n^{-4}$ for n-photon pumping lasing threshold (15).

Although the diffusion model criterion (15) predicts remarkable improvement of lasing performance for very low pumping frequencies, real systems are more complicated and the applicability of (8) is constrained by the transparency threshold for the medium and the finite thickness of the sample. It is convenient to relate the transparency threshold to the absorption of emitted light that is very small but finite. The pump power should be large enough to overcome this absorption and only after that it contributes to the population inversion. The absorption can be defined through the absorption coefficient γ_{abs} or the absorption length $l_{\mathrm{abs}} = v/\gamma_{\mathrm{abs}}$. The absorption can be ignored when the pumping strength g_n [in the domain $0 < x < L_n$; see (14)] exceeds the absorption rate γ_{abs} and the penetration length of pumping light is smaller than the sample thickness. The second condition can be easily satisfied by making a sufficiently thick sample, while the first constraint is more difficult. One can reformulate the transparency threshold requirement using the estimate (14) as

$$\eta\Gamma_n I_{0n}^{n-1} > \gamma_{\mathrm{abs}}. \tag{16}$$

To discuss real systems we consider the dye materials with the highest two-photon absorption cross section (e.g., trans-4-[p-(N-hydroxyethyl-N-methylamino)styryl]-N-methylpyridinium p-toluene sulfonate [18])

$$\sigma_{2\mathrm{ph}} \sim 10^{-46}\ \mathrm{cm^4 s\ photon^{-1}}. \tag{17}$$

It is more convenient to express the strength of the two-photon absorption in terms of the single photon absorption as

$$\Gamma_2 I^2 \sim \gamma_1 I \times \frac{I}{I_*} \tag{18}$$

where I_* is the formal (very high) intensity of pump light needed to have equivalent single- and two-photon absorption rates. We assumed the maximum single-photon absorption strength ($\sigma_1 \sim \lambda^2$). Making use of (17) one can estimate

$$I_* \sim 10^{11}\ \mathrm{J/(scm^2)}. \tag{19}$$

This intensity exceeds the threshold intensity found in two-photon pumping measurements by one to two orders of magnitude. Therefore, the gain length for two-photon absorption is by the factor of 10–100 larger than for the single-photon pumping. Since the typical value of the gain length for a TiO_2-based random laser is of the order of 10–100 μm the gain length for the two-photon pumping is about 1000 μm. It can be still less than the absorption length for the emitted light that is expected to exceed the size of the lasing mode estimated by as much as several hundred microns [14].

Real lasing materials show deviations from the predictions of the diffusion model [8]. These can be due to fluctuations of the optical mode lifetime, for instance, caused by the destructive interference of the emission from several resonant scattering centers [15] or random closed light paths holding the light for a long time inside the active medium. If these fluctuations are significant for the bulk modes they will show up in the two-photon pumping studies, leading to a lower lasing threshold than (15) (see [8]). The experimental study of these effects will be straightforward within the two-photon technique since the light goes much deeper and more modes are involved.

The limit of the very strong disordering leading to the Anderson localization of light (several experimental realizations of the light localization were reported in [16] and [17]) are of special interest for multiphoton pumping. The localized states can be attained very efficiently since their decay rate decreases exponentially with the distance x from the sample boundary [12], while the gain decreases with x by the power law (7). Therefore, the lasing instability can always be reached for localized states in the bulk, when the absorption of the emitted light is sufficiently small.

Summing up, we have demonstrated within the framework of the diffusion model that multiphoton pumping can be very efficient for attaining random lasing in random medium with small absorption. The TiO_2-based random lasers can be most appropriate to realize this mechanism. The great advantage of the multiphoton technique is that a high-frequency laser is not required to excite the lasing modes, while the disadvantage is the larger size of lasing material compared to that for the single photon pumping.

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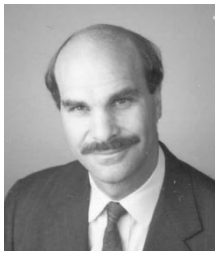

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