

Ultra-High Efficiency and Low Threshold in Random Lasers

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Abstract: Random lasers hold the potential for cheap and coherent light sources, however, improvements in terms of efficiency and laser threshold are required. In this paper, we show two new strategies to increase efficiency and decrease the laser threshold.

OCIS codes: (140.3380) Laser materials; (290.4210) Multiple scattering;

1. Introduction

In recent years, there has been dramatic progress in the photonics field in disordered media, due to their potential applications in random lasers [1–4], localization of light [5,6] and other novel photonic functions. Random lasers hold the potential for cheap, coherent light sources that can be miniaturized and molded into any shape with several other added benefits such as speckle-free imaging; however, they require improvements specifically in terms of efficiency and threshold. Here, we show, for the first time, two new kinds of random lasers, one with a threshold more than one order of magnitude lower than similar random lasers and one with efficiency that rivals diode pumped solid-state lasers.

The first random laser is composed by core-shell TiO₂@Silica suspended nanoparticles (NPs) of rhodamine 6G (R6G). The NP's concentration of [140x10¹⁰ NPs ml⁻¹] is equivalent to 10.6% filling fraction. R6G is diluted at [1x10⁻⁴ M]. The silica shell prevents the “optical” junction of the TiO₂ scattering surfaces (steric “optical” effect), preserving the scattering strength of the medium [1]. Additionally, the shell provides a light-coupling enhancement with TiO₂ scattering cores [2], which leads to an increase of the effective TiO₂ scattering cross section. Narrow peaks, all with similar intensity, were observed in a broad frequency range, showing a high degree of suppression of the interaction between these laser modes indicating operation at the phase transition to localization [3] and a very low random laser threshold. In the other random laser, diffusion and gain are separately controlled by using controlled, reaching a world record in optical efficiency of 50% [4]. This random laser (RL) consists in a powder pellet composed by a polydispersed particle size distribution (smaller particles between bigger ones) from a grinded and sieved 1.33 mol% yttrium vanadate doped with neodymium crystal with mean particle size of 54 μm. The smaller particles, trapped between large particles serves as absorption and gain centers whereas the large particles control mainly the light diffusion into the sample.

2. Results and discussion

Random lasing at localization transition. Figure 1a shows the RL emitted intensity, recorded from frontal and back collection, as a function of pumping energy fluence. The RL slope efficiency (RL_{eff}) for back collection (RL_{eff-back}) was constant without depletion (magenta line), whereas the RL_{eff} of frontal collection decreases from fluencies ≥ 12 up to 36 mJ cm⁻², where it keeps constant (blue line, fluencies ≥ 36 mJ cm⁻²). The saturated RL_{eff} of the frontal collection (blue line) corresponds to the RL_{eff} of the extended modes.

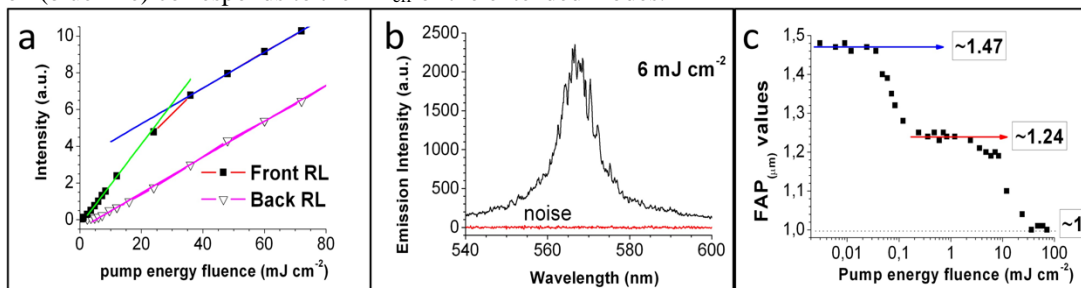


Fig. 1. a) Influence of the pump fluence on emitted intensity for frontal and back collection, b) emission spectra of RL collected from a small micrometric volume (<4 μm diameter and depth) for pumping fluencies of 6 mJ cm⁻² and c) FAP_(μm), as a function of the pumping energy fluence.

The RL emissions from a micrometric region (3.8 μm diameter) near the input-pumping surface (less than 4 μm depth) using a detection system that performs like a confocal microscope was also performed. Figure 1b shows the emission spectra for pumping fluencies of 6 mJ cm⁻². The emission spectra consisted of a flat-topped ASE band of 7.5 nm width with overlapping narrow peaks at frequencies close to the maximum of the spectrum. The minimum linewidth

observed for the narrowest peaks was ≤ 0.17 nm. The light of the pumping pulse reflected by the samples was measured with and without R6G. We designated the ratio between the pumping intensities reflected by the scattering medium with and without R6G as the fraction of absorbed pumping (FAP), which represents the absorbance of the sample, $\ln(\text{FAP})$ [1,3]. The FAP values from the small micrometric volume ($\text{FAP}_{(\mu\text{m})}$) were measured with the same confocal experimental setup. The $\text{FAP}_{(\mu\text{m})}$ values for fluencies (< 0.036 mJ cm^{-2}) well below the laser threshold ($\text{FAP}_{\text{bT}(\mu\text{m})}$) are constant (1.47), which represents a passive regime. For fluencies between 0.036 and 0.24 mJ cm^{-2} , the $\text{FAP}_{(\mu\text{m})}$ value decreases quickly down to 1.24, where it remains approximately constant up to a fluence of 2.4 mJ cm^{-2} . The latter implies that for this range of fluencies (0.24–2.4 mJ cm^{-2}), a stationary regime of the RL emission is reached in this micrometric volume near the input-pumping surface (< 4 μm depth), with RL threshold of 0.14 mJ cm^{-2} . This threshold value is more than 10x smaller than the threshold of any reported $\text{TiO}_2/\text{R6G}$ solution reported before.

Random lasing with polydispersed particle sizes. Ten groups of pressed pellets using monodispersed and polydispersed $\text{Nd}^{3+}:\text{YVO}_4$ powders have been prepared with different grain size distributions by using five sieves of different mesh size, having mean particle size ranging from 9.5 μm to 147 μm . The polydispersed groups had a strong participation of smaller particles whereas the monodispersed groups suffered an additional cleansing procedure that eliminated those smaller particles. Using a polydispersed powder mixture of 54 μm mean particle size, we achieved an increase of 3.1x in output power with respect to its respective monodispersed powder (96 μm mean particle size), as shown in figure 2a. Using absorption (FAP) and backscattering cone measurements, we calculated a similar mean photon path length in both groups (343 μm and 357 μm), which indicates that light dispersion within the pellets is mainly governed by the larger particles that are common to both groups. However, polydispersed groups have on average a 2% higher fill fraction due to additional small particles and therefore undergo 15% more scattering events than the corresponding monodispersed samples (because of the smaller transport mean free path). This means that the additional 15% of absorbed energy is located mainly within the pockets containing 2% of smaller particles which in turn corresponds to a five times higher amount of absorbed pump power within these pockets (for a fill fraction of 0.66). In addition to their optical properties (l , volume), these pockets are pumped by all sides and from any direction. As shown in figure 2a, the slope efficiency of this diode pumped laser is 54%, which is comparable to the best reported values for diode-pumped solid-state neodymium lasers.

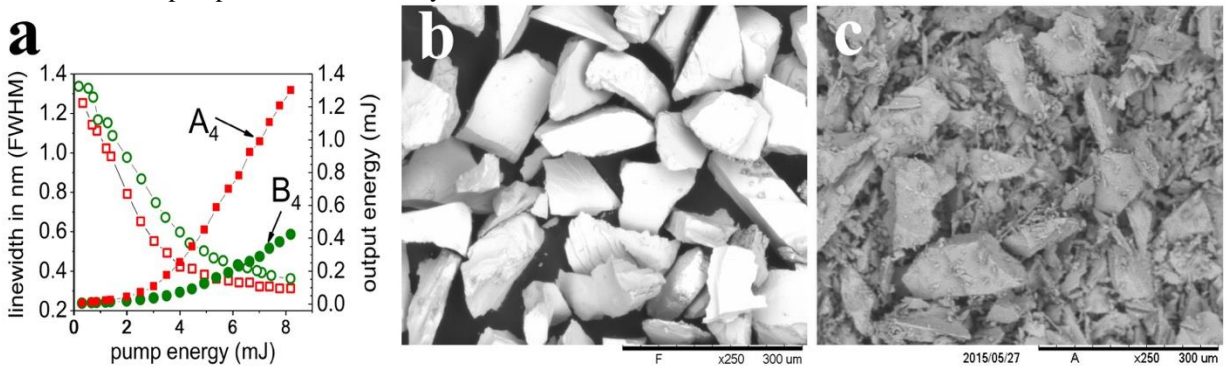


Fig. 2. a) comparison between the best sample from the polydispersed groups (A_4) and its monodispersed counterpart (B_4). Shown are linewidth narrowing (left axis) and laser output power (right axis). The slope efficiency of curve A_4 is 50%. b) and c) are SEM images of the monodispersed and polydispersed groups B_4 and A_4 , respectively.

3. References

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