Electronic Supplementary Information

Continuous wave random lasing in naturally occurring biocompatible pigments and reduction of lasing threshold by using triangular silver nanostructures as scattering media

*Subrata Biswas and Prof. Pathik Kumbhakar **

Nanoscience Laboratory, Dept. of Physics, National Institute of Technology Durgapur, 713209 West Bengal, India
**Corresponding author, E-mail:nitdgpkumbhakar@yahoo.com & pathik.kumbhakar@phy.nitdgp.ac.in*

Contents

Table-S1: Chlorophyll content of different plant species (mg/l)*

Sl.No	Plants Name	Young leaves		Total Chl. Content young leaves
		Chla	Chlb	
1.	Mango (Magniferaindica)	3.24	3.09	6.48
2.	**Hibiscus (Hibiscus rosa- sinensis)**	**19.6**	**12.87**	**39.2**
3.	Gavua (Psidiumguajava)	6.27	5.40	12.54
4.	Almond (Prunusdulcis)	5.25	3.04	10.5
5.	Sapodilla (Manikarazapota)	6.04	7.33	12.08
6.	Bryophyllum(Bryophyllumpinnatum)	4.62	8.87	9.42
7.	Neem (Azadiractaindica)	18.09	9.76	36.18
8.	Ashoka (Polyalthialongifolia)	1.30	1.65	2.6
9.	Ficus (Ficusbenjamina)	6.66	4.7	13.32
10.	Datura (Datura metal)	13.96	8.84	27.92

***Ref:** P. N. Kamble, S. P. Giri, R. S. Mane and A.Tiwana.*Univers. J. Environ. Res. Technol*. 2015, **5**, 306-310.

Photo-physical properties of TNS under laser irradiation: To determine the effect, if any, of the CW pump laser radiation on properties of the TNS we have performed a control experiment in which we have measured the static PL spectrum of TNS after irradiating the TNS for different duration of same CW laser light which is later used as pump for RL. As PL emission spectrum is very much sensitive to the photo-physical changes in the sample, if any, we have used this to investigate any change in sample characteristics upon irradiation of the laser light. Figure S1 below shows the PL emission spectra of the TNS collected at different times and from the figure it is clearly seen that there is almost no such changes in PL emission spectra after laser irradiation for the duration upto 2 min. Therefore, we can discard the possibilities of any photo-physical changes in TNS on laser irradiation within the time duration of laser generation experiment.

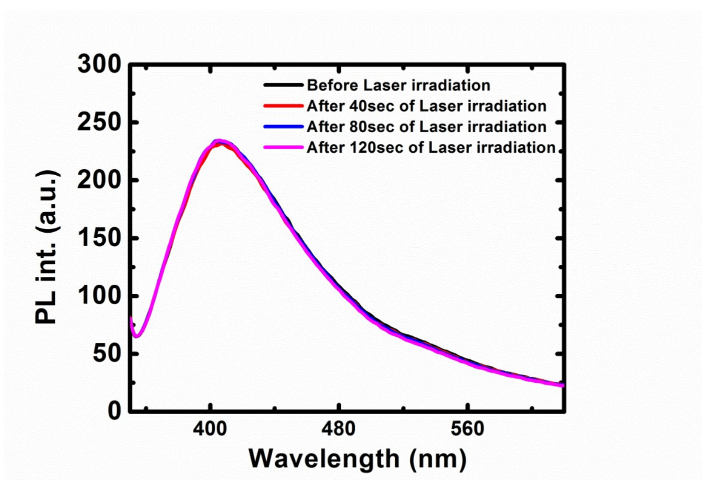

Fig. S1.The static PL emission behaviour of the TNS at different laser irradiation time.

Scattering length (l_{sc}):

Scattering lengths for different concentrations of scatterer have been calculated theoretically as well as experimentally, as elaborated in the main text and results are summarized below in Table-S2.

Table -S2. Experimentally and theoretically obtained values of scattering length for different concentrations of TNS.

TNS concentration (nos/mL)	l_{sc} from theory (cm)	l_{sc} from experiment (cm)
5×10^{14}	23.5	16.7
1×10^{15}	11.7	9.4
2×10^{15}	7.8	5.4
4×10^{15}	5.8	4.6
8×10^{15}	1.6	1.2

Quenching of PL of MB dye in the presence of TNS

PL emission spectra of HRLe dissolved in hexane have been measured under the excitation of ordinary light of 632nm wavelength and in the presence of TNS with concentrations varying between 0.5×10^{15} nos scatterer/mL to 2.0×10^{15} nos scatterer/mL. The results are shown in Fig. S2, which shows clearly that quenching of PL of MB dye is taken place but there is no effect on the position of PL emission peak.

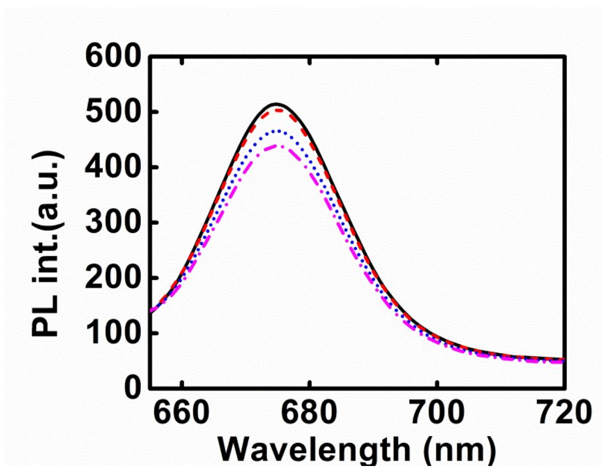

Fig. S2 Quenching of PL emission intensity of HRLe due to the presence of the TNS scatterer of different concentrations. Here black, red short dashed, blue short dotted line and pink dashed dot line corresponds to the PL spectrum of HRLe obtained without TSN scatterer, with 0.5×10^{15} nos /mL and 1×10^{15} nos /mL and 2×10^{15} nos /mL of TNS scatterer, respectively.

Random Laser Emission Spectra of HRLe with Different Concentrations of TNS

As elaborated in the main text RL generation experiments have been carried out in HRLe in the presence of different concentrations of TNS and with varying pump laser intensity. The emission spectra as obtained with four different concentrations of TNS (a) 5×10^{14} (b) 1×10^{15} (c) 2×10^{15} (d) 4×10^{15} nos./mL are shown in Fig. S3.

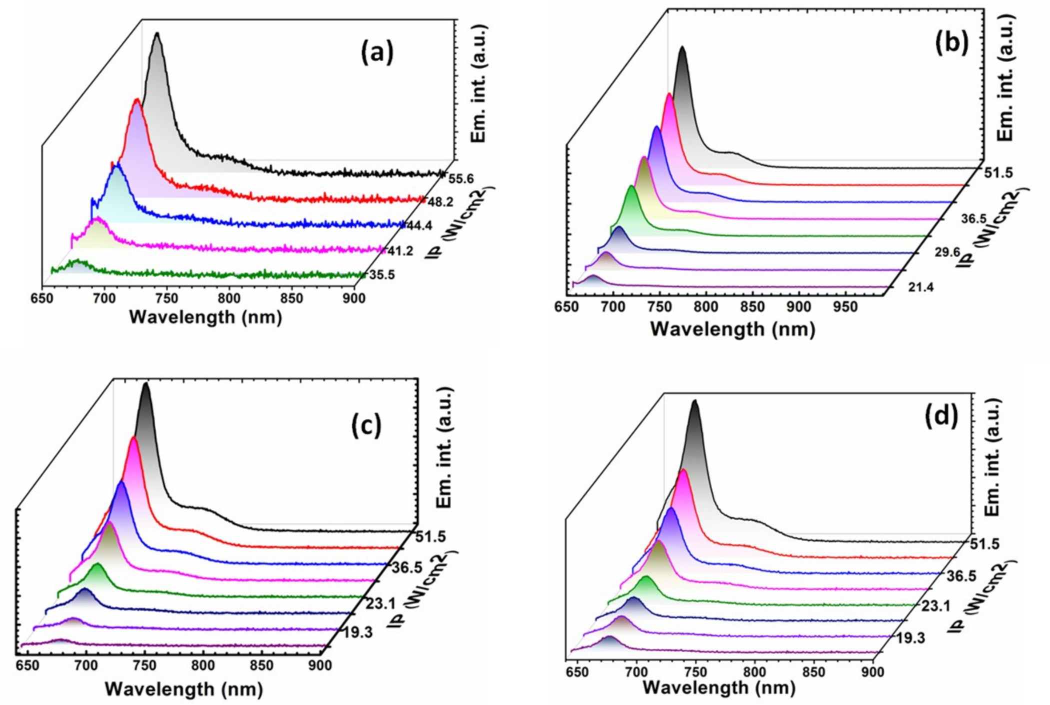

Fig. S3 Emission spectra of HRLe for four different values of N of (a) 5×10^{14}, (b) 1×10^{15}, (c) 2×10^{15} and (d) 4×10^{15} nos./mL, when it is pumped by a He-He laser of 632.8 nm wavelength with various pump intensities.

The line-widths (FWHM) of the PL emission peaks have been determined by Gaussian fitting of the emission spectra measured with various pump laser intensities but with constant scatterer concentration and those are plotted in Fig. S4. In Fig. S4(a) shows the variation of line-width of the emission spectrum of only HRLe with the pump intensity. The variations of line-width with the with the pump intensity corresponding to the RL emission spectrum measured in HRLe in presence of scatterer with five different concentrations of 5×10^{14}, 1×10^{15}, 2×10^{15}, 4×10^{15}, and 8×10^{15} nos./mL are shown in Figs. S4(b)-(f), respectively. The reductions of line-width after the lasing threshold as well as reductions of lasing threshold with the increasing concentrations of TNS are clearly observed.

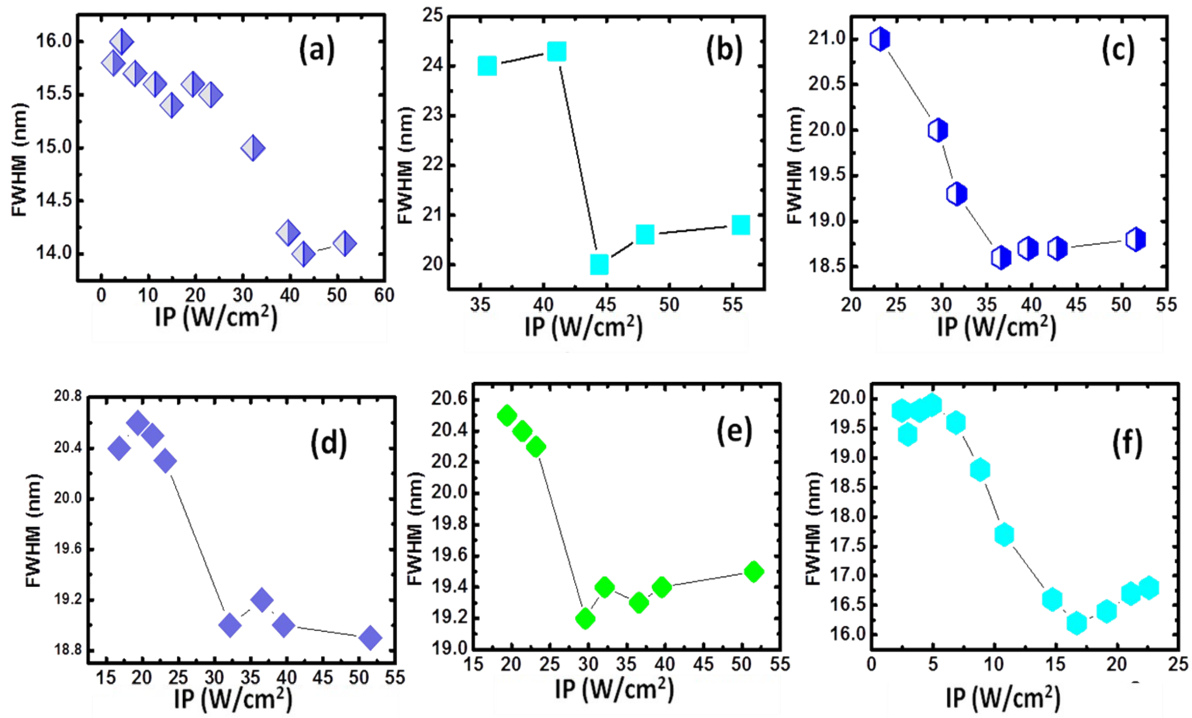

Fig. S4 Variation of line-width (FWHM) with the pump intensity. (a) Corresponds to the experiment with only HRLe. Figures marked as (b), (c), (d), (e) and (f) corresponds to the experiments in HRLe with N of 5×10^{14}, 1×10^{15}, 2×10^{15}, 4×10^{15}, and 8×10^{15} nos./mL, respectively.

Determination of the lasing threshold of RL generation in HRLe

The lasing threshold for RL generation in HRLe in the presence of the TNS scatterer of a given concentration have been determined by plotting the output power emitted at ca. 674 nm with the variation of the input pump laser intensity and the results are shown in Figs. S5(a)-(d).

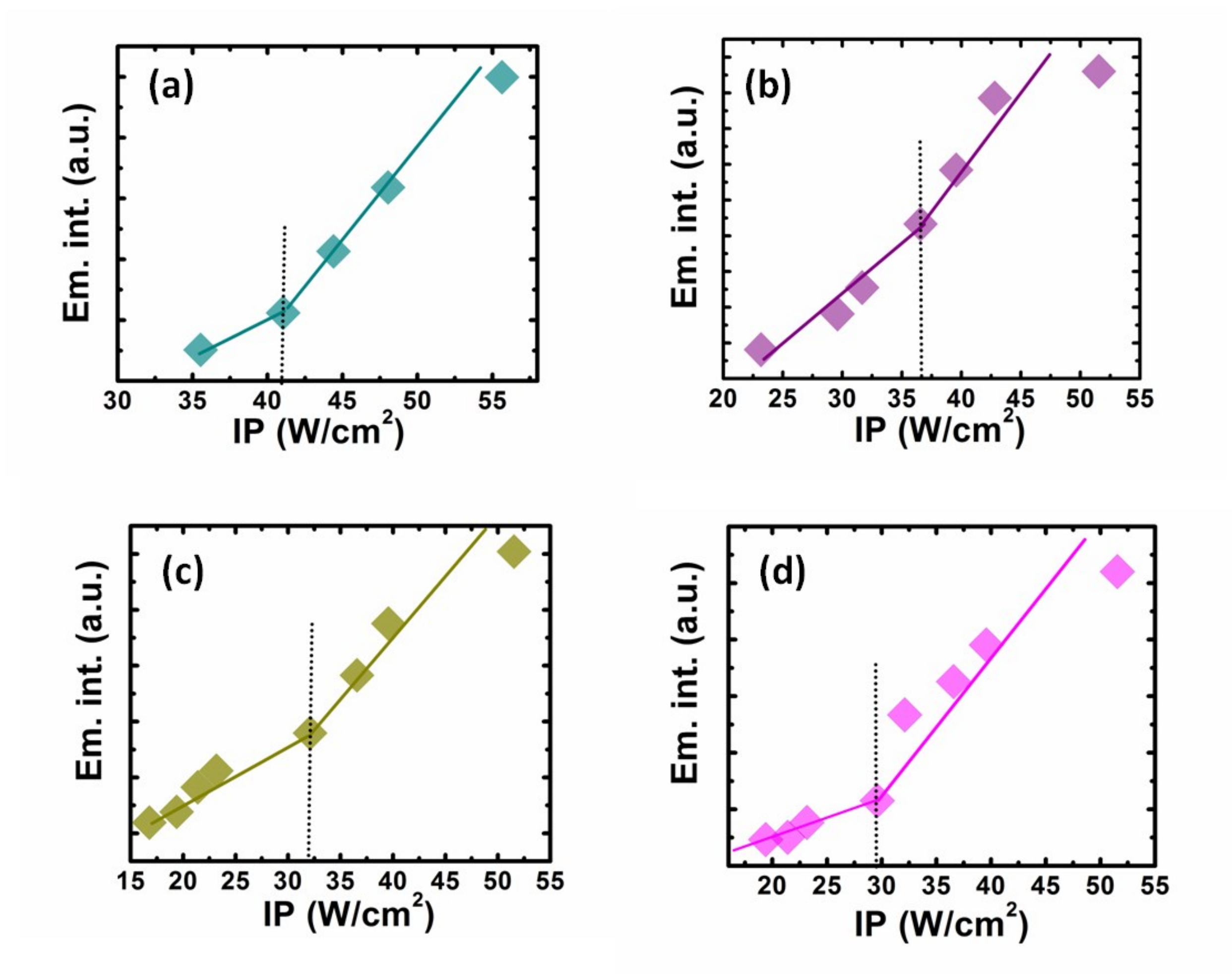

Fig. S5 Exhibits the variations of emission intensity with pump intensity for RL generation in HRLe for four different concentration (a) 5×10^{14} (b) 1×10^{15} (c) 2×10^{15} (d) 4×10^{15} nos/mL of TNS.

Determination of the polar intensity profile of emitted RL laser mode:

In order to determine the preferred direction of emission of RL radiation from HRLe at a scatterer concentration of 7.5×10^{15} nos./mL, the emission spectra, as shown in Fig. 6 (a), have been collected at different angles of 0° to 180° with respect to the incident laser direction. It is clearly seen from the collected spectra that mode pattern is appearing at different positions as we have changed the direction of collection of RL generation. The corresponding polar curve has been plotted over whole range of collection i.e. 0^o to 180^o and is shown in Fig. 6 (b). The enlarged section of the emission pattern of the RL at an angle 60^o is shown in the upper panel of the Fig. 6(a) where we can easily observe the generation of the RL mode of FWHM of 1.5 nm at ~ 674nm wavelength. Thus it can be concluded that the obtained RL generation is not unidirectional which can also considered as a signature of RL [6]. It is also evident that the emission intensity of RL generation at angle 60° is the highest so later we have collected RL spectra only at an angle of 60° with the incident laser direction.

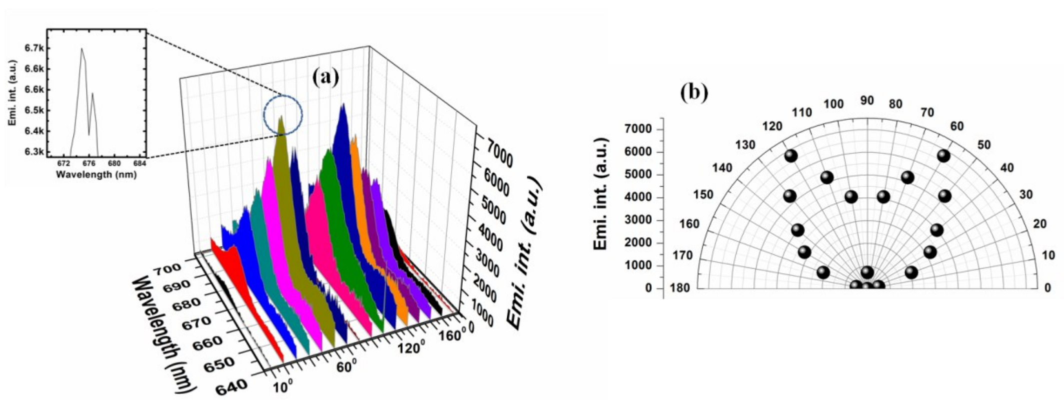

Fig. S6(a) Emission spectra of the HRLeat a scatterer concentration 7.5×10^{15}nos of scatterer/mL at different angles with the incident laser direction starting from 0^o to 180^o when the pumping energy is 24.2 W/cm^2. (b) The polar patterns shows the variation of the peak intensity of RL spectra at each given collection angle (θ) with the incident beam.

RL generation in MB dye and TNS doped polyvinyl alcohol (PMT) film

PMT films with various concentrations of TNS have been prepared for carrying out RL generation experiments under pumping of CW He-Ne laser light of 632.8 nm wavelength. Here, in Figs S7(a)-(d) the emission spectra obtained at various pump intensities has been shown. The lasing threshold for RL generation in PMT films in the presence of the TNS scatterer of a given concentration have been determined by plotting the output power emitted at ca. 691.7 nm with the variation of the input pump laser intensity and the results are shown in Figs. S9(a)-(d).

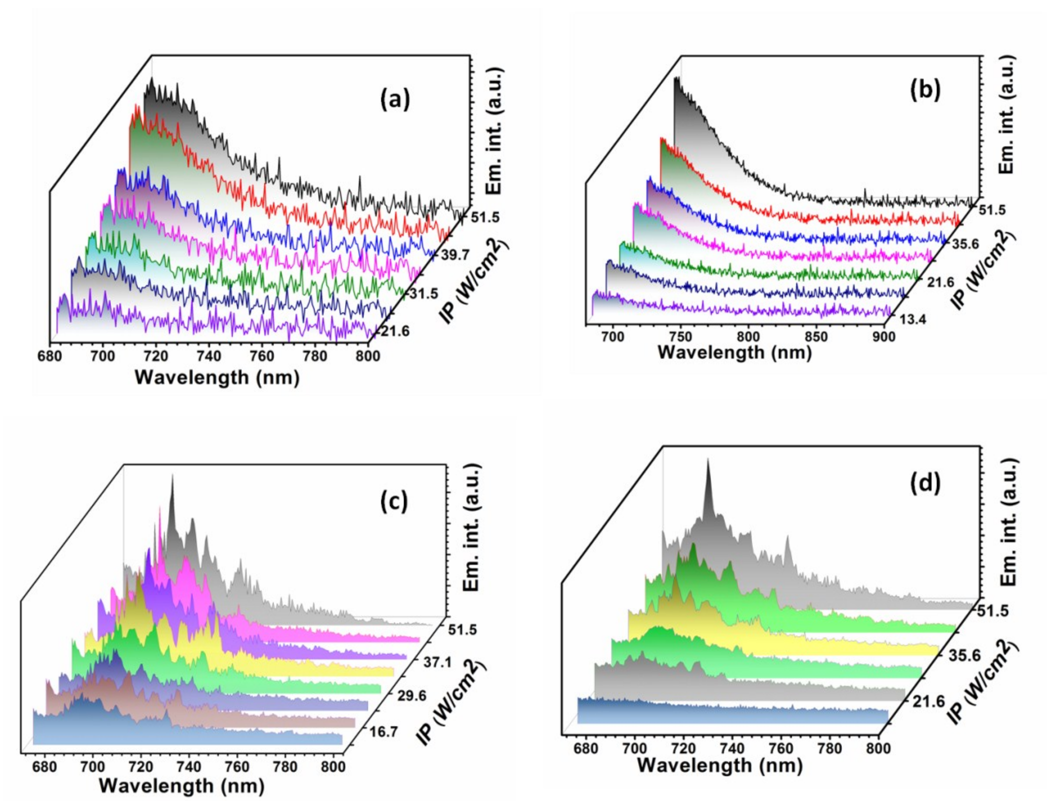

Fig. S7 Exhibits the RL generation in PMT films. Figures marked as (a), (b), (c), and (d) corresponds to the experiments in PMT with TNS as scatterer having number concentration of scatterer 5×10^{14}, 1×10^{15}, 2×10^{15}, and 4×10^{15} nos./mL.

SEM image of the PMT film

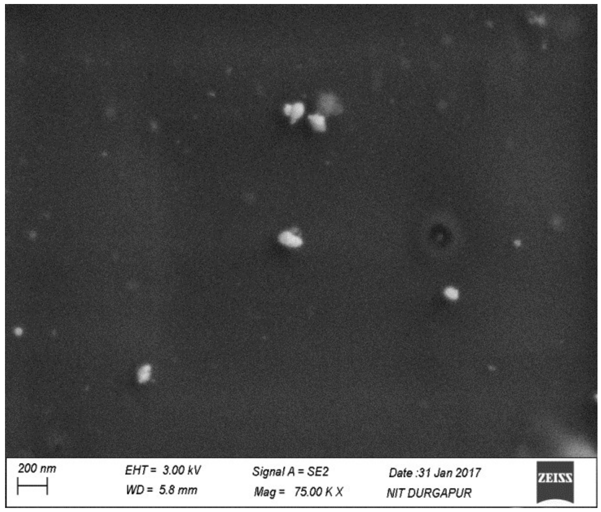

Fig. S8 SEM image of the PMT film which shows the TNS are evenly distributed in the film.

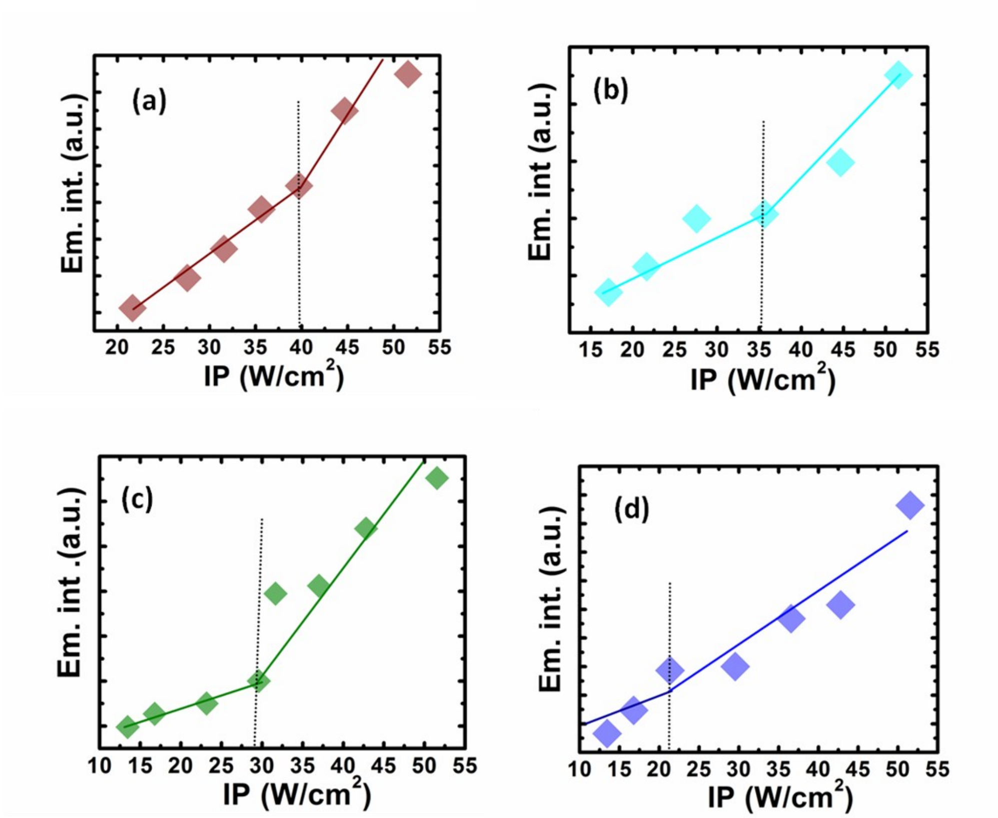

Fig. S9 Exhibits the variation of emission intensity with pump intensity for RL generation in PMT film for four different concentrations of TNS (a) 5×10^{14} (b) 1×10^{15} (c) 2×10^{15} (d) 4×10^{15} nos./mL

Just for comparison, in Fig. S10 we have plotted emission spectra obtained in an aqueous solution of MB dye in the presence of TNS of a typical number concentration of $2x10^{15}$ nos/mL and that of PMT film prepared with the same concentrations of TNS. Figure S10 shows that MB dye does not exhibit lasing except some broad amplified spontaneous emission (ASE).

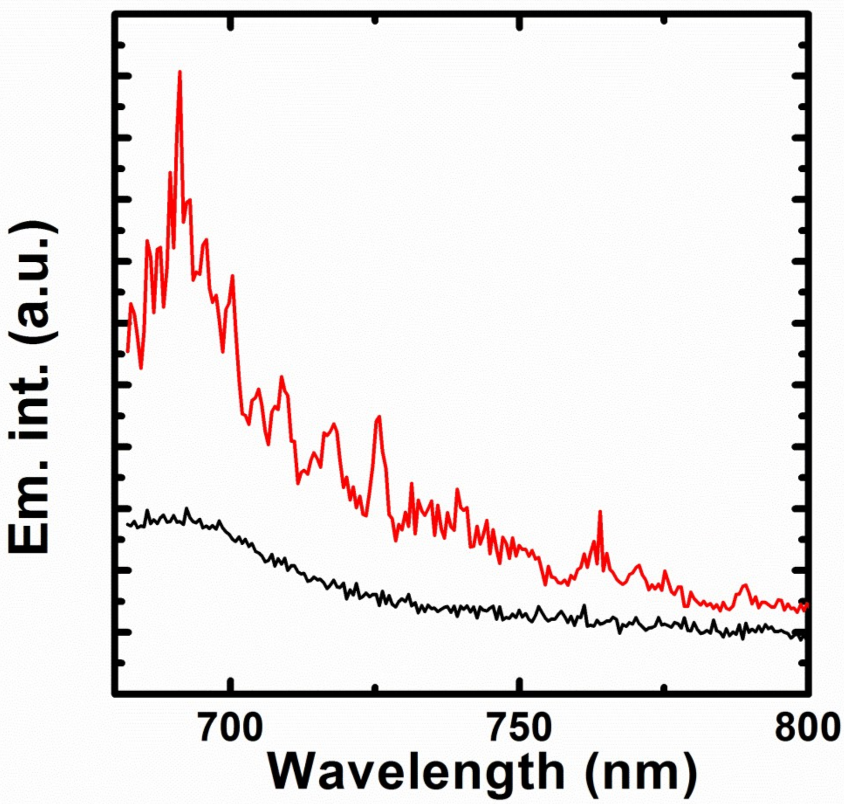

Fig. S10 A comparison of the emission spectra of MB dye solution with $2x10^{15}$ nos/mL of TSN (black line) and that of PMT film with same concentration of TSN (red line), the used pump intensity is $55W/cm^2$.