Random laser action in aqueous solutions

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Abstract: We show experimental results on random laser action of rhodamine 6G in aqueous solutions containing alumina particles and SDS with high efficient energy conversion. The threshold is inversely proportional to the SDS concentration.

OCIS codes: 140.2050, 290.4210, 290.5850, 240.6670.

1. Introduction

Laser emission from random media occurs due to propagation of light in an amplifying medium containing scattering particles [1] or in systems consisted only of particles that work simultaneously as amplifiers and scatterers [2]. In the first case the system is usually made by dissolving laser dyes in non-polar or weak-polar solvents, such as methanol or ethanol, and by adding dielectric nanoparticle scatterers, such as TiO_2.

Many experimental works show that laser dye solutions using polar solvents like water do not show fluorescence due to the fact that this kind of solvent favors the formation of aggregates, mainly the H-type dimer, which drastically quenches the quantum efficiency of the solution. These aggregates are present even at very low laser dye concentrations. The rhodamine 6G (R6G), a xanthene dye, aggregates at concentrations around 10^{-5} M in water, forming stable ground state dimers via $\pi-\pi$ mixing of the xanthene ring orbitals. At 10^{-4} M R6G in water no longer lases because of the dimerization [3]. In ethyl alcohol solutions aggregation occurs at much higher dye concentrations and the dimers, of the J-type, are fluorescent. The fluorescence spectrum of the J-type dimers is red shifted in relation to the fluorescence spectrum of the monomers. As a consequence random laser bichromactic emission occurs at concentrations around 10^{-3} M [4].

Water has a lot of advantages over other solvents like ethanol or methanol [5, 6]. In this work we show that adding sodium dodecyl sulfate (SDS) to the water solves the problem of aggregation of R6G molecules and rises the random laser efficiency to the same level of ethanolic solutions.

2. The experiments

In the experiments we have used alumina, with an average diameter of 300 nm, as the scattering particles, and R6G as the amplifying medium. The samples were pumped by a pulsed Nd:YAG laser, operating at 532 nm, with pulse durations of 5 ns. In the first experiment we prepared a solution of R6G dissolved in ethanol and another one in distilled water plus SDS, both with the same dye concentration and density of particles, in order to make a comparison between them. All the measurements presented in this Letter were taken with a dye concentration of 1×10^{-4} M and the density of alumina of 4×10^{12} particles/cm^3. The spectra, shown in Fig. 1, taken at a pumping energy of 10 mJ, are practically identical except by a red shift presented by the aqueous solution sample.

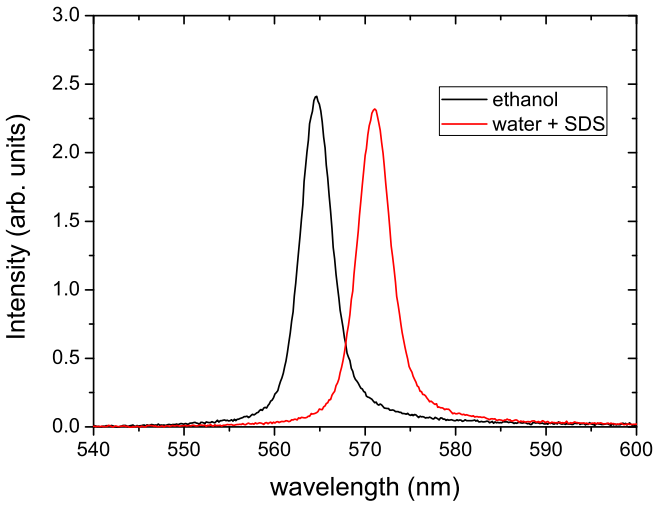

Fig. 1. Random laser emission from R6G dissolved in ethanol and water.

The similarity between these spectra extends to a large range of pumping energies, showing that similar random laser efficiencies may be achieved in ethanolic and aqueous solutions, at the same dye concentration and particle density conditions.

A detailed investigation was done, by measuring the linewidth as a function of the pumping energy for various SDS concentrations. The result from this analysis is shown in Fig. 2, where the threshold energy E_{th} as a function of the SDS concentration C is represented by the empirical fitting function

$$E_{th} = \frac{A}{C - C_0} + E_0 \tag{1}$$

where E_0 is the limit value of the threshold energy obtained at high SDS concentrations, and A and C_0 are fitting parameters. The fitted values are A = 0.0022 mJ.M, E_0 = 0.16 mJ, and $C_0 = 9.8\times10^{-3}$ M. Since the value of C_0 is close to the literature value of the critical micelle concentration (CMC) of SDS in pure water (8.1×10^{-3} M) [7], we may conclude that the effect of the SDS on the disaggregation of the laser dye molecules, and a consequent enhancement of the laser dye quantum yield, happens close to the region where the SDS micelle phase transition takes place. The inverse proportionality behavior presented by the random laser threshold as a function of SDS concentration is be closely related to the results of Peterson *et al.* [8] and Weber [9] in their investigations on dye laser threshold as a function of dye concentration. An investigation on the amount of R6G monomers as a function of SDS concentration is under way, and will help us to relate the random laser threshold and the R6G monomers concentration.

3. Conclusions

In conclusion, we demonstrate efficient laser action of R6G in aqueous solutions containing alumina scattering particles and the surfactant sodium dodecyl sulfate (SDS). We show that random laser efficiencies from aqueous solutions may be as high as that presented by ethanolic solutions when the SDS concentration is above the critical micelle concentration (CMC). We also show that the random laser threshold is inversely proportional to the SDS concentration above CMC, a behavior similar to that observed in experiments of the dye laser threshold as a function of dye concentration.

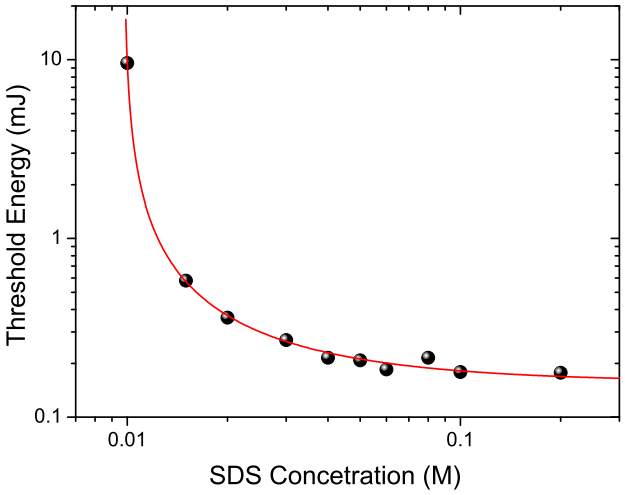

Fig. 2. Lasing threshold energy as a function of the SDS concentration.

Acknowledgments

This work was supported by CNPq, CAPES and CTPetro-FINEP, Brazilian agencies.

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