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Random lasing action in magnetic nanoparticles doped dye solutions

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ABSTRACT

Fe_3O_4@SiO_2 nanoparticles were used as scatters in Rhodamine B solutions, and coherent random lasing was achieved. It was revealed that Fe_3O_4@SiO_2 doped dye solution has a magnetically controllable feature. When external magnetic field is applied, the laser peaks would disappear if the diameter of Fe_3O_4 is relatively large (~100 nm), while the laser peaks would exist if the diameter of Fe_3O_4 is relatively small (~12 nm). This kind of random laser may has potential applications in fabricating magnetic sensors and integrated optical device.

1. Introduction

The prediction made by Letokhov [1] on laser emission from a disordered gain medium was clearly verified experimentally by Lawandy et al. [2], using colloidal solutions containing rhodamine 640 perchlorate dye and TiO_2 nanoparticles in methanol. Subsequently, several researchers have focused their attention on such new laser systems without cavities, called random laser (RL) which is characterized by multiple scattering of light and amplification by stimulated emission. A variety of RL systems have been reported so far, such as liquid laser dyes doped with nanoparticles (NPs) [3–5], dye-polymers embedded with scatters [6], dye-doped liquid crystals [7,8], ZnO powder [9], and porous media infiltrated with dyes [10].

The tunable laser is an important component in optical communications and other applications. Among all the reported RLs, the liquid crystal (LC)-based RLs show temperature [7] or voltage [8] tunable properties and have attracted much attention in recent years. However, LC-based RLs work unstably because they are sensitive to the environmental temperature.

NPs consisted of iron oxide NPs show promising prospects in the fields of magnetic resonance imaging and drug deliver [11]. Because of the superparamagnetism of embedded iron oxide nanocrystals, microbeads doped with iron oxide nanocrystals exhibit a high level of magnetization which allows the control of RL with external magnetic fields.

In the present work, we report the characterization of random lasing in dye solution doped with magnetic nanoparticles (MNPs), and the observation result that the RL spikes vanish when external magnetic field is applied. To our knowledge, this is the first investigation of magnetically-controlled nanoparticle doped random lasers. This kind of magnetic controlled random laser opens a new way for practical application of random lasers.

2. Experiment

The experimental setup for the random laser(RL) is shown in Fig. 1. The pump source is the second harmonic of a Nd:YAG laser (532nm, 10 ns, 10 Hz). The wavelengths of output light are 355 nm, 532 nm, and 1064 nm. The direction of the mixing light is changed by a right-angled prism and the mixing light is separated by a triple beams prism. An aperture is used to obtain a uniform part of the green light spot (532 nm) and the light beams with other wave lengths (355 nm and 1064 nm) are absorbed by an absorbing plate. Then the green light passes through a reflection mirror, green filter and Glan prisms in sequence. It is finally focused by a lens on a 1×1 cm^2 quartz cuvette which contains MNPs doped RhB solution, and the incident beam is normal to the cuvette. The external magnetic field is applied by putting a ferromagnet cling to the cuvette. A high resolution optical multi-channel analyzer (OMA) with a resolution of about 0.1 nm is used to measure the random lasing signal. The Glan prisms are placed in front of the lens to modulate the incident pulse energy. When the pump energy is above the threshold energy, the forward light spot of random

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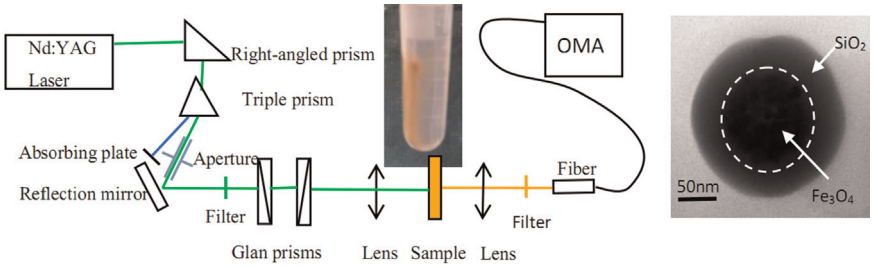

Fig. 1. experimental setup. Inset on the right shows the TEM image of a typical prepared Fe_3O_4@SiO_2 nanoparticle. Inset in the middle shows the particles separated by magnetic field.

lasing will be obtained immediately and detected by the OMA, as shown in Fig. 1.

The system is composed of rhodamine B(RhB) dye solution embedded with Fe_3O_4@SiO_2 MNPs forming one kind of random lasing media. The Fe_3O_4@SiO_2 core–shell MNPs are silica NPs enriched with iron oxide nanocrystal. The MNPs average 200 nm in diameter while the thickness of SiO_2 shell is 50 nm, as illustrated in Fig. 1. The dye solution (2.5×10^{-4} M in deionized water) doped with MNPs is put in an ultrasonic bath for 30 min before measurement, and the concentration of RhB is chosen in such a way that there is only a weak absorption of emitted light outside the pumped region.

3. Results and discussion

We first investigated the lasing resonance property of RhB dye solution embedded with Fe_3O_4@SiO_2 MNPs. Fig. 2(a) depicts the evolution of emission spectra as a function of the pump energy. The spectra shows a broad spontaneous emission of RhB molecules under a low pump energy (60 μJ/pulse). As the pump energy exceeds 100 μJ/pulse, well-distinguished sharp spikes with linewidth less than 0.2 nm appear around 590 nm, indicating that the coherent random lasing occurs. The appearance of sharp spikes implies the interference of waves which is usually resulted from intensified scattering in random lasing media. Fig. 2(b) shows the comparison between the emission spectra of pure dye solution and MNP doped dye solution under a pump energy of 120 μJ/pulse and a significant difference is observed. This result is due to the multiple scattering of MNPs, which provides the optical feedback of random lasing in the solution.

When an external magnetic field is applied, the emission spectra varies with the time. Fig. 3 shows the dependence of emission spectra on the applied time of an external magnetic field at a pump energy of 120 μJ/pulse. After about 5 minutes, the sharp random lasing peaks vanished and the emission of the dye solution only shows amplified spontaneous emission (ASE) property with FWHM of about 5 nm. It is because that most of the MNPs in the solution are effectively separated from the pump region when the magnetic field is applied [11], and there is nearly pure dye solution in the excited region, as shown in Fig. 1. In this case, the random lasing would be quenched off since the feedback provided by MNP scatters in the excited volume becomes very weak.

The experimental results demonstrate that the RL emission can be controlled by magnetic field. However, it was found that the controllability depends on the diameter of Fe_3O_4. A kind of Fe_3O_4@SiO_2 MNPs with relatively small iron oxide nanocrystals ($\approx$ 12 nm) and with silica shell of about 100 nm were prepared, and the emission of dye solution doped with this kind of MNPs was investigated. The RL emission intensity was nearly constant after the external magnetic field was applied for about 20 s, and the random lasing peaks would not disappear, as shown in Fig. 4. The corresponding dependence of the emission peak intensity on

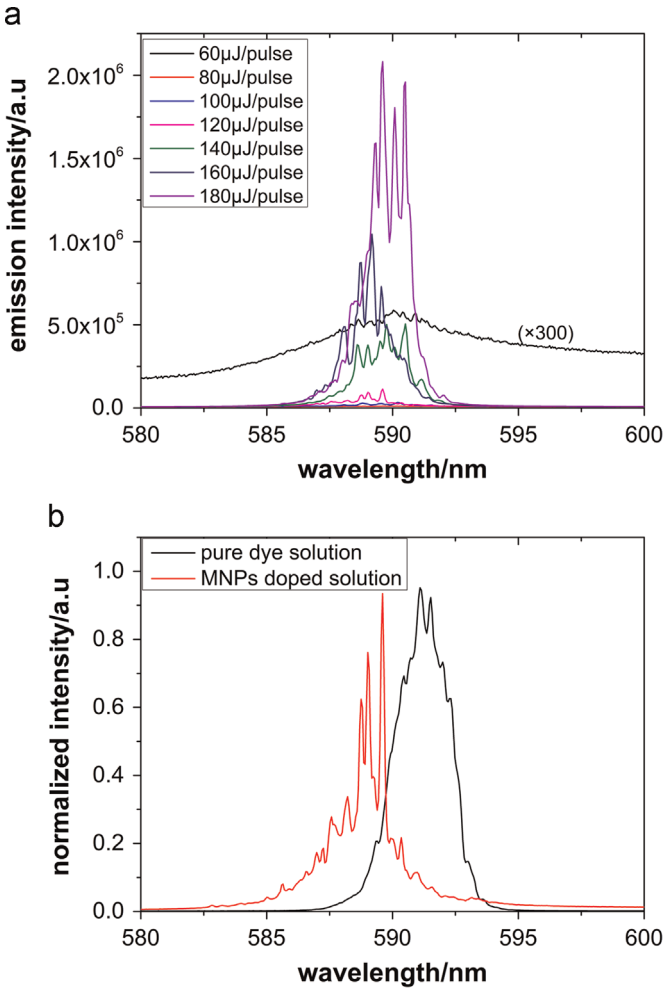

Fig. 2. (a) Evolution of emission spectra as a function of the pump energy. (b) Emission spectra of pure dye solution and MNPs doped dye solution under pump energy of 120 μJ/pulse.

the pump energy was measured, as shown in Fig. 5. Circles in Fig. 5 represent the input–output relationship of the sample when the magnetic field was applied for 30 min, which is distinct from that of the pure dye solution. This fact means that there are a part of MNPs that are not separated from the excited region due to their weak magnetism. These results indicate that a larger Fe_3O_4 core contribute to better magnetic controllability in this kind of MNP doped random laser.

It is known that in strong scattering regime, photons have long amplification paths that contribute to the stimulated emission, and the emission intensity would increase when the density of scatters becomes larger. However, the forward spot was found to increase while the scattering strength to decrease. In our experiments, the