

Laserlike emission from rhodamine in a photonic crystal

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Abstract

Laserlike emission from optically pumped inverse opals photonic crystal infiltrated with a laser dye is reported for the first time. The data show strong modification of the dye fluorescence and a laser threshold of ~ 0.1 mJ/pulse. The nonlinear process studied is enhanced by local field effects due to the location of dye molecules inside the opal voids.

Introduction

Laserlike emission from strongly scattering media has been the subject of intense research since this phenomenon was observed in laser dye solutions containing TiO₂ nanoparticles [1]. The emission from such system, which was named as laser paint (LP), has characteristics of a multimode laser, although no resonant cavity is used. Of particular importance to obtain laser emission is the multiple scattering of light which occurs due to the difference between the refractive index of the liquid and the scatters. The multiple scattering events contribute to increase the effective pathways of the emitted photons favoring their interaction with very large of molecules.

An analogous process of laser generation is expected to be observed if a periodic arrangement of particles, such as a photonic crystal (PC) [2, 3], is infiltrated by a laser active medium and excited in a suitable manner. An appropriate PC structure to demonstrate such effect is the inverted opal which presents pores of well defined size and long-range spatial periodic ordering. Synthetic opals can be made of silica, polystyrene or other materials [4] and the radiative properties of dyes incorporated into such PC may change considerably as shown in ref. [5].

In this letter we report the observation of laser action in synthetic opals infiltrated with Rhodamine.

Experimental Setup

The experiments were carried out in opals fabricated using closed packed crystals of latex spheres as templates for silica polymerization [6]. Samples with f.c.c. crystalline structure of nanoholes as characterized by electron microscopy and Bragg diffraction of light were prepared. For the present experiments the opals were infiltrated with Rhodamine 6G by adding some drops of an ethanolic solution (concentration: 5.87×10^{-4} M.) over the samples and left to dry at room temperature for some days. We studied samples with different diameter's size of holes, 220 nm (sample A) separated by walls of 70 nm and 500 nm (sample B), with walls of 110 nm, respectively.

The second harmonic of a Nd:YAG laser (532 nm; 6ns; 5 Hz) was used to excite the samples. The linearly p-polarized laser beam is incident on the PC surface with an angle of incidence which could be adjusted with respect to its normal direction. The pump wavelength is off the pseudogap of the PC and thus the laser beam penetrates inside the crystal exciting molecules along its trajectory. Although silica presents no absorption in the visible range, the laser light is absorbed inside the opal structure due to the presence of dye molecules.

In the experiments performed the incident laser intensity was varied and the dye fluorescence was collected and sent throughout a monochromator to monitor its characteristics. The signals were recorded using a photomultiplier coupled to a storage oscilloscope connected to a computer.

Results and Discussions

Figure 1 shows the fluorescence spectra corresponding to sample A for two different pump intensities. The pseudogap of sample A (B) is located in the blue (red) region, as already shown in the literature [5] and the dye fluorescence band is outside of the gap. For the sample B (A), the fluorescence peak intensity for excitation at 0.92 (0.84) GW/cm^2 is 54 (7) times less intense than the peak intensity for excitation at 10.1 (10.5) GW/cm^2 , but the signals are normalized with respect to the maximum fluorescence intensity signal. Spectral narrowing was observed increasing the pump laser intensity as in the LP experiments. Note that at low-laser intensity the fluorescence linewidth is ~ 30 nm while above threshold it is reduced to ~ 8 nm.

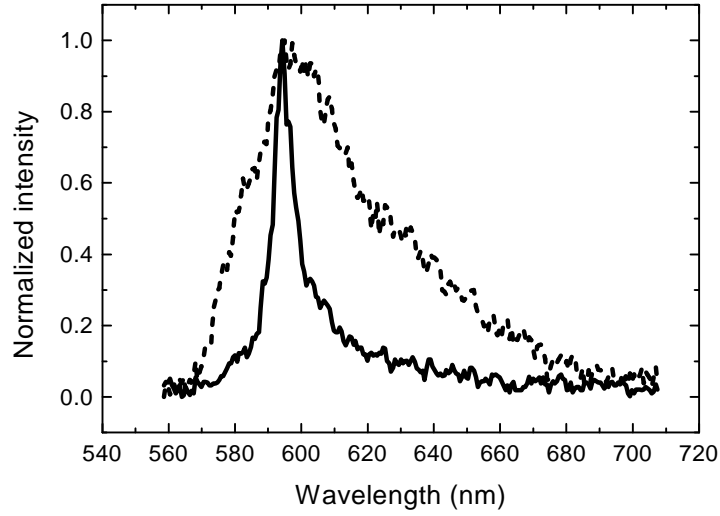


Figure 1: Normalized fluorescence spectra of the sample A. The dashed line corresponds to pump intensity of $0.9 \text{ GW}/\text{cm}^2$, while the solid line is for pump intensity of $10.1 \text{ GW}/\text{cm}^2$.

Figure 2 summarizes the intensity and linewidth behavior of the fluorescence as a function of the pump intensity for the sample B. The fluorescence peak intensity shows a weak dependence with the pump intensity up to $5 \text{ GW}/\text{cm}^2$ ($0.1 \text{ mJ}/\text{pulse}$) which represents the threshold for nonlinear growth of the fluorescence intensity. The peak intensity increases by one-order of magnitude while increasing the pump intensity from 4 to $10 \text{ GW}/\text{cm}^2$, for the sample B. Note in Fig. 2 that the linewidth starts at ~ 30 nm, which is the natural width of the dye, and narrows to ~ 7 nm when the laser intensity is increased to $\sim 12 \text{ GW}/\text{cm}^2$. We also note that the PC laser wavelength is off the pseudogap and thus the emitted light propagates inside the opal structure and amplified spontaneous emission occurs along the random pathway.

The emission characteristics from both samples (i.e. its wavelength, bandwidth and intensity) for different observation angles are similar and the features were reproduced well for different regions on the samples' surfaces.

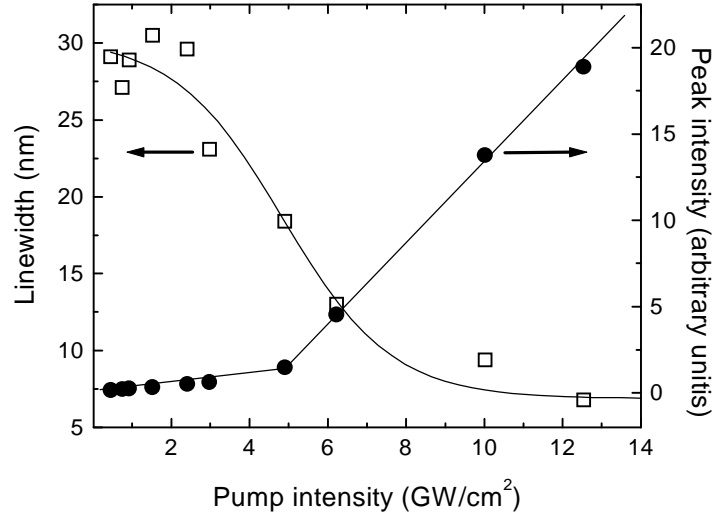


Figure 2: Linewidth (open squares) and peak intensity (solid circles) dependence with pump intensity for the sample A.

The present results can be compared with previous reports on LP and PC studies. Two kinds of systems were exploited in the LP experiments. Most of the works were performed with colloidal solutions containing a laser dye dissolved in alcohol and dielectric nanoparticles randomly distributed [1] but samples where the dye and the scatters are dispersed in a polymer sheet were also investigated [7, 8]. The laser threshold of ~ 0.1 mJ/pulse measured in the present experiment, is about the same observed in LP experiments [1, 7, 8] where two-orders of magnitude larger dye concentration was used. In LP experiments all feedback mechanisms that are due to morphological resonances are eliminated since the gain medium is outside the scatters. Local – field effects due to the scatters are not relevant in the LP experiments because most of the active molecules are located far from the particles surfaces.

In this work the dye molecules are inside nanoholes periodically arranged and thus the opal structure behaves as a PC with laser active lattice points. The walls of the nanoholes scatter light originating a random walk of the photons generated inside the structure before they reach the exit PC surface. Inside the holes the spatial distribution of dye molecules is expected to be non uniform and larger dye molecules concentrations occur nearby the walls due to adsorption. Although organic dyes adsorbed in semiconductors have their quantum efficiency lowered, in the present case this can be compensated by the effect of dielectric confinement of radiation. In fact, the difference between the refractive index of the hole wall and its inner space is such that local field effects may contribute to enhance the nonlinear process in contrast to the LP experiments. According to theoretical predictions [9] strong enhancement effects in the emission rate of atoms having transitions with optical frequencies outside the bandgap is expected even in structures with incomplete photonic bandgap. Therefore, local-field effects are attributed as the origin of the large laser-action enhancement observed in the present work in comparison with LP experiments [1, 7, 8]. On the other hand, the present results indicate that laser emission from dyes infiltrated PC made of inverse opals of titania could be observed in the previous experiments [5] if appropriate conditions have been exploited. Moreover, the results of ref. [5] which demonstrate large modification in the fluorescence spectrum of dyes molecules corroborate our assumption that local field effects play an important role in the present experiment.

A comparison with recently reported lasing in a liquid crystal based PC is also worthwhile. Ref. [10] presents an approach which relies on the properties of partially ordered liquid crystal phases which exists for certain temperatures. Although the laser threshold reported in ref. [10] is lower than in the present case, the approach of ref [10] is limited by the solubility of laser dyes in liquid crystals while there is no limitation to extend the operation of the present PC laser to other spectral regions were efficient dyes are already available.

Conclusions

In summary, we have observed for the first time laser emission from an optically pumped PC. The stimulated emission process is favored by the geometry of the inverted opal nanostructure and the location of dye molecules inside nanoholes.

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