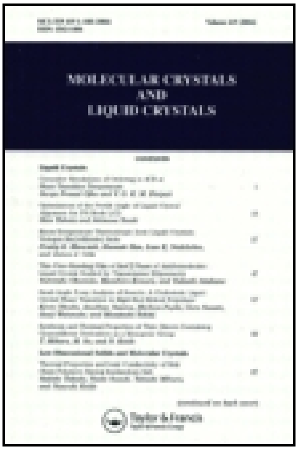

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LASING AND INTERMODE CORRELATION OF WHISPERING GALLERY MODE IN DYE-DOPED POLYSTYRENE MICROSPHERE

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Abstract We study the lasing behavior and photo induced quenching of dye doped polystyrene microspheres with diameter ranging from 10 μm to 92μm. Lasing of the spherical cavity modes is clearly observed. Emission from individual sphere placed on a glass plate is examined under the pumping of nanosecond pulses which are tuned to the absorption band of the dye. Intensity of each lasing line is strictly controlled by additional irradiation of monochromatic laser which can couple to resonance modes of the cavity. The line intensity plummets down when the additional beam frequency matches to the resonances of the modes which have the same radial profile of relevant mode.

Introduction

Ultra fast optical switching and data processing are one of the most important application of molecular system. In past decades, much work has been devoted to find efficient nonlinear optical materials which are suitable for such application among both inorganic semiconductors and organic materials. Although numerous kinds of materials have been studied, only limited numbers of devise structures, such as nonlinear Fabri-Perot type etalon and wave guide directional coupler have been studied. The advantage of the organic materials is their plasticity so that they can be made into any shapes. Thus it is possible to realize an ideal nonlinear optical device in the combination of appropriate material and device structure.

Recently, much attention has been paid on electromagnetic whispering gallery mode (WGM)[2] or morphology dependent resonance (MDR) mode in micro dielectric sphere.[1] The advantage of spherical resonator is its tremendously high quality factor (Q value). Such value provides us a very low threshold laser[3] and optical bistable device.[4] Dye Solution droplet is a very good candidate where high Q cavity modes are realized and it has been studied extensively.[1] For practical application, however, solid materials are necessary. Laser oscillation in solid spheres has been observed only in rather large spheres with diameters in several millimeters[5,6], except in one recent experiment by Wang in Nd doped glass spheres.[7] Polystyrene has rather large refractive index of 1.58 and can be made easily into spheres of required size. Benner et al. observed WG resonances in the fluorescence spectrum of dye doped polystyrene spheres.[8] Thus polystyrene sphere is suitable for the study of optical WGM's. In this paper, we report the evidence of lasing and nonlinear optical interaction between WGM's in dye doped polystyrene spheres with diameters ranging from 10 μm to 92 μm.

Experiment

Polystyrene spheres doped with organic dye molecule, nile red, having the concentration of 10^{-6} mol/g are placed on glass plate under microscope. One sphere is excited individually by nanosecond pulsed dye laser with 520 nm wave length excited by an excimer laser. The pulse duration is about 5 nanosecond and the pulse energy is about 100 nJ at maximum. We position optical fiber with core diameter of 200 μm directly below the sphere to collect the emission which is sent into 1 m monochromator (as shown in Fig.1).

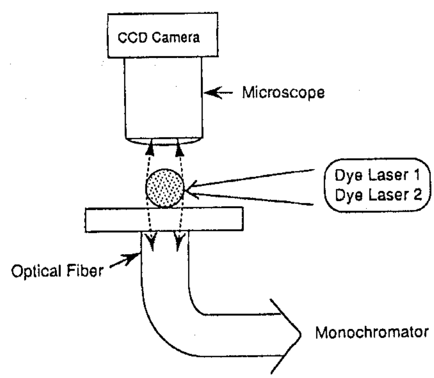

FIGURE 1 Schematic diagram of the experimental method.

Temporal responses of the emission are analyzed using fast response micro channel plate photomultiplier (MCP-PM) and a Boxcar integrator with high speed sampling head. Temporal resolution of our system is 0.5 nsec. Spatial profile of the emission pattern of the sphere is recorded with a CCD camera attached to the microscope. Another tunable nanosecond dye laser with spectral width of 0.05nm is also prepared for the photo induced quenching experiments.

Results and Discussions

Figure 2 shows the emission spectra from a 41μm sphere under various pumping intensity up to 150W of peak power. At low power excitation, emission spectrum is broad (a) and no pronounced structures are found. As pump power increases, sharp lines appear (b-d).

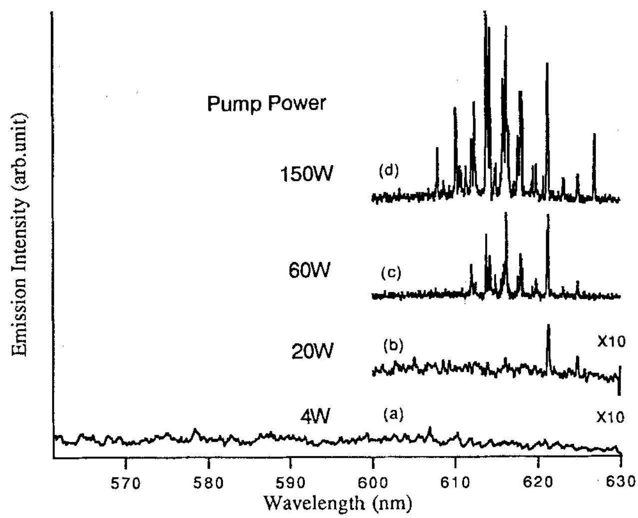

FIGURE 2 Pumping power dependence of emission spectra from a 41 μm dye-doped polystyrene sphere.

The lasing spectra look complicated because many lines have different mode number, *n*, and different order number, *l* , for both transverse electric (TE) mode and transverse magnetic (TM) mode. The indices *n* and *l* denote the *l*-th resonance of the *n*-th mode. *n* indicates the angular dependence of the mode and *l* does the radial distribution. The mode separation -- frequency deference between the neighboring modes of the same order number modes -- is proportional to the inverse of the diameter. When the excitation level is below the threshold, the image of the emission pattern is uniform. Above the threshold, the rim becomes brighter than the center. Temporal responses of the emission also show the lasing characteristics. The lasing line follows the pump laser pulse while the shorter wave length emission in broad spectrum region shows the slower decay of several nanosecond which corresponds to the spontaneous decay time of the dye molecule.

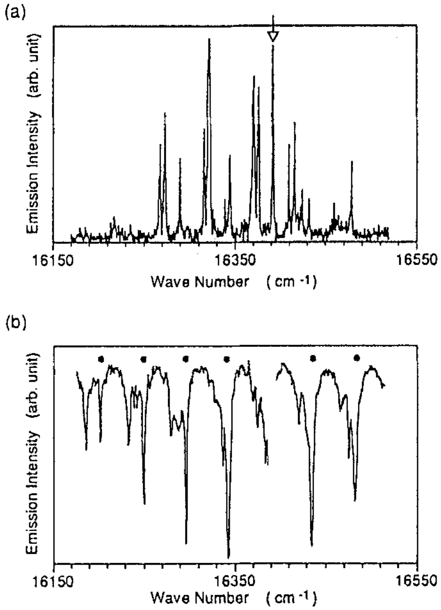

FIGURE 3 (a) Lasing spectrum from a 41μm sphere. (b) Intensity of a lasing line indicated with a downwards arrow in (a) as a function of the frequency of the additional beam.

To investigate the optical controllability of the laser, we examine the emission under the irradiation of the sample with additional monochromatic laser which is tunable in the frequency region of emission from spheres. The pulse energy of the additional beam is kept at as low as 5% of the pump beam. Figure 3 shows the result. Intensity of one lasing line, indicated by the downwards arrow in Fig.3 (a), is recorded as a function of the frequency of the additional beam. The emission is greatly reduced when the frequency of the additional beam reaches to certain frequencies as shown in Fig.3 (b). Those frequencies, marked with closed circles in the figure, are the WGM's resonances which have the same order number, *1*, as the mode of recorded line. This implies that the competition between modes with same order number is very strong but not between modes with different order numbers. This is because the modes of same order number have almost the same radial function and their interaction volume is large. Hence the strength of cross-mode interaction strongly depends on the combination of their mode indices.

Conclusion

We observed the lasing of whispering gallery modes in dye doped polystyrene spheres. The emission is strongly quenched by the irradiation of an additional beam resonant to the spherical cavity modes with same radial profile. Under the lasing condition, there is a salient cross-mode correlation between modes which have the same mode order. This feature enables us a new field of application such as highly efficient optical data processing using molecular systems.

Acknowledgement

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