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MULTIPLE SPECTRAL HOLE BURNING AT ROOM TEMPERATURE WITH DYE-MOLECULES-DOPED SPHERICAL DIELECTRIC MICROPARTICLES

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Abstract Multiple spectral hole burning at room temperature was realized for Nile blue molecules-doped spherical polymeric microparticles based on morphology-dependent resonances. Hole parameters, sidehole generation, dynamical process of hole formation and hole burning mechanism were studied.

INTRODUCTION

The sharp morphology-dependent-resonances (MDR'S) of spherical dielectric microparticles have attracted substantial interest for several potential applications, such as low-threshold microlasers, intracavity optical nonlinearity, optoelectronics and integrated optics. In particular, it has been suggested to apply the MDR's for frequency domain optical storage at room temperature by using a collection of microparticles as hole burning medium.[1-2]

MDR modes correspond to photons being strongly confined within the microsphere by repeated total internal reflection from the inner surface at grazing incidence. The resonant field satisfies a phase condition after circling around the particle perimeter. MDR modes are characterized by $x_{n,l}$, where $x=2\pi a / \lambda$ is the size parameter of the particle. a is the particle radius and λ the wavelength. n and l are integers termed mode number and order number respectively, corresponding to the number of poles of internal field distribution in the angular and radial directions. [3]

MDR modes are homogenous. Linewidth associated with the leakage loss of the spherical microcavity is $\gamma_h=\langle k\rangle/Q$,where $\langle k\rangle$ is the average wave vector of the radiation near a resonance. Since the quality factor Q higher than 10^8 has been confirmed by particle-fiber coupling measurement,[4] linewidth narrower than 10^{-5} nm

can be achieved. The modes shift in wavelength as the particle size changes. Therefore, a collection of microparticles generates inhomogeneously broadened MDR lines. The inhomogeneous width γ_{in} is proportional to the width of size distribution, $\gamma_{in}=\sigma_a\langle k\rangle/\langle a\rangle$, where $\langle a\rangle$ is the mean radius of the particles and σ_a is the standard deviation of radius. The great advantage of using microparticles as hole burning medium is that both γ_h and γ_{in} are insensitive to temperature and can be controlled independently. This ensures the necessary condition $\gamma_h \ll \gamma_{in}$ for the realization of hole burning at room temperature and will produce high capacity of storage in frequency domain. We report here an investigation on multiple persistent spectral hole burning with Nile blue molecules-doped polymeric microparticles. Hole parameters, sidehole generation, dynamical process of hole formation and hole burning mechanism were studied.

EXPERIMENTAL

Microparticles of polystyrene (Ps) and polymethylmethacrylate (PMMA) prepared by dispersion polymerization method were dyed with Nile blue (NB) chromophone molecules in the hydrosol state. A drop of hydrosol containing so-dyed particles was placed on a cover glass plate to form a two dimensional distribution. The sample was air dried before used as hole burning medium. Mean radius and standard deviation of the particle size were typically $\langle a\rangle$=3.00 μm and σ_a=0.18, corresponding to an inhomogenous width of 37 nm in red region.

An Ar^+ laser-pumped tunable Rh-B dye laser (CR-599-21) was utilized as the light source for hole burning and detection. The line width of the laser used was 0.01nm. Laser beam was slightly focused to a ~1mm^2 spot on the particle sample plate for the hole burning. Fluorescence excitation spectra were measured before and after hole burning. Fluorescence emission was dispersed with a monochromator, detected by a photomultiplier and recorded by a microcomputer after lock-in amplification.

RESULTS AND DISCUSSIONS

Room temperature persistent spectral hole burning for both NB/Ps and NB/PMMA systems have been realized. We shall show some examples measured on NB/Ps, the results are similar for the other system. Fig.1 shows a deep hole burnt for 13 min at 610 nm with a burning laser power 150 mW. The hole width (FWHM) was 2.2 nm and its depth ($\Delta I/I$) was 66%. It was found out that higher power and longer burning

time led to deeper and broader holes. By choosing proper laser power and burning time, multiple holes were recorded. Fig. 2 shows 6 holes burnt at 607, 613, 619, 625, 631 and 637 nm. The powers used were 100, 100, 100, 106, 96 and 86 mW respectively and the burning times were 40 s for the former 4 holes and 45 s for the rest 2 holes. Hole widths were reduced to ~ 1 nm. During the process of multiple hole burning , we noticed that the previously burnt holes could be partially erased by the subsequent burning. The reading also had some erasing effect on the written holes.

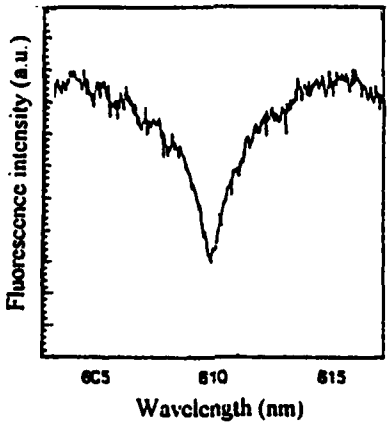

FIGURE 1 Fluorescence excitation spectrum revealing a deep hole burnt at 610 nm for 13min with a burning power of 150 mW.

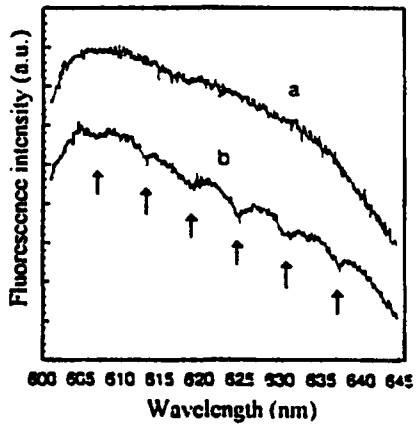

FIGURE 2 Fluorescence excitation spectra before hole burning (a) and after 6 holes burnt successively (b).

The holes observed were much broader than expected. This is mainly attributed to the absorption of Nile blue molecules within the particles, although power broadening also affects on hole width. For the internal resonant field, the existence of absorption within the particle causes internal loss in addition to the light leakage, giving rise to the decrease of the effective quality factor of the particle resonator and the broadening of MDR line. In this case , the homogeneous line width of a resonance is expressed by $\gamma_h = \gamma_o + \gamma_a$, where γ_o is the intrinsic part caused by light leakage and γ_a is the absorption part depending on the ratio of the imaginary part to the real part of the complex reflective index of the particle.

Time evolution of hole formation was observed by monitoring the fluorescence intensity during the burning . Fig.3 shows the process, where the burning power used was 150 mW. As the burning laser was set on , the fluorescence intensity rose up rapidly to a peak and then fell exponentially until finally reached a steady level . From

which, we recognized that the transient hole formation rate is at the highest near the beginning of hole burning . Therefore, it's favorable and sufficient to choose shorter burning time in order to gain the multiplicity of recording and to reduce the energy broadening as was done in the measurement of Fig.2. Data of the trailing edge in Fig.3 was fitted to a formula $I = c_1(1+c_2e^{-c_3t})$ from which three constants were obtained: c_1=1.390 × 10^{-2} , c_2=1.618 , c_3=6.676 × 10^{-3} s^{-1}. Among them the parameter c_3 characterizes the effective rate of hole formation which is useful for material evaluation. Table 1 lists the parameter c_3 and the depth of saturated holes measured directly from the time evolution curves for different burning powers.

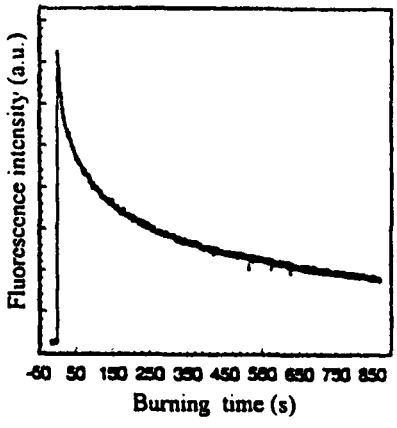

FIGURE 3 Time evolution of hole formation measured by monitoring the fluorescence intensity during the hole burning.

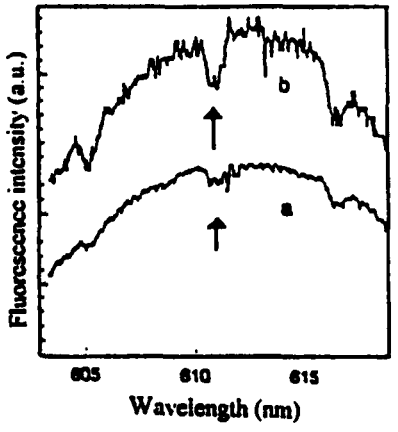

FIGURE 4 An original hole burnt at 610 nm for 30 s (a) and 80 s (b), and two sideholes appeared symmetrically on both sides of it.

TABLE 1 Hole parameters for different burning powers

Intensity of burning laser (mW)	c_3 ($10^{-3}s^{-1}$)	Depth of saturated hole (%)
150	6.676	71
50	4.039	63
20	3.721	55

The existence of a series of resonances for a special particle size may result in complex structure in hole spectrum. The burning at one resonance also reduces the fluorescence at other resonances when probed. It would be interesting to observe sideholes stimulated by the write beam. Fig.4 gives an example of this kind measured for a highly monodispersive collection of microparticles, where the original hole indicated by an arrow was burnt by the laser at 610 nm for 30 s (curve a) and 80 s (curve b) and two sideholes appear symmetrically on both sides of it. The sideholes were not probed in the hole spectra in Fig.1 and Fig.2. This may be explained by the existence of several subsets of particles in resonance with the write wavelength for the sample with relatively broad particle size distribution function. The original hole was contributed by several particle subsets in resonance with the same write wavelength and thus became strong enough to be detected, while the sideholes located at different wavelength positions for different particle sizes and, as a result, became less distinct and were in fact undetectable.

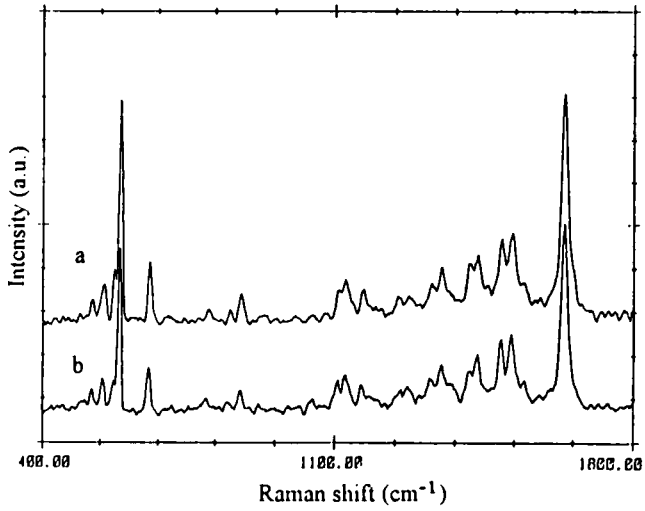

FIGURE 5 Raman scattering spectra measured on Nile blue molecules embedded in KBr sheet before (a) and after (b) laser irradiation.

Hole burning mechanism of the dye-molecules-doped microparticles may be understood as MDR- enhanced photochemical reaction and selective bleaching of resonant subsets of particles. In order to locate the chemical reaction, Raman scattering spectra and Fourier transform infrared (FTIR) transmission spectra were measured before and after laser irradiation for Nile blue molecules imbedded in KBr sheet. The host material KBr has no transition in the wavelength range of measurement. In the Raman spectra shown in Fig.5, the evident decrease of the

demonstrates again overall decrease of Nile blue absorption after irradiation. Besides, the relative decrease of the peak at 1275 cm^{-1} and the increase of the 828 cm^{-1} peak tends to suggest the C-N bond breaking on the heterocyclic ring in Nile blue molecular structure.

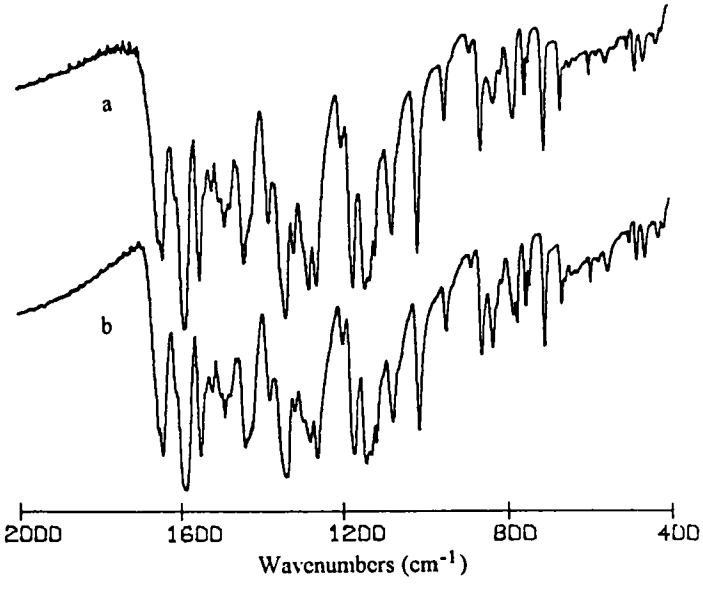

FIGURE 6 Fourier transform infrared transmission spectra measured on the same sample as Figure 5 before (a) and after (b) laser irradiation.

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