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Spatial manipulation of valence states of samarium ions by the interference of multiply scattering light in random media

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Spatial manipulation of valence states of samarium ions has been demonstrated in dielectrically disordered media consisting of glass powders. When the photoionization of Sm^{2+} is combined with multiple light scattering in glass powders, holes are burned in frequency and wave-vector domains. The hole profile strongly depends on the amount of optical absorption, and the variation of the hole profile can be explained based on the diffusion approximation. We also demonstrate the dependence of hole formation on the polarization direction of laser beam.
Key words: glass powders, samarium ions, photoionization, multiple light scattering, interference

1. INTRODUCTION

The propagation of light waves is strongly modified by the spatial modulation of the dielectric constant in a medium. The field of such photonic materials has attracted considerable attention because of the tremendously broad range of applicability of optical systems [1]. Also, the photonic systems exhibit a fascinating fundamental phenomenon such as light localization.

Recently, it has been reported that a new type of optical memory effect takes place in a photoreactive and dielectrically disordered material [2]. Irradiation of such a material with a monochromatic light causes the interference of multiply scattering light. As a result of the spatial modulation of optical absorbance through photobleaching, a dip or a hole is observed in the frequency and wave-vector domains, since the interference effect depends on the wavelength and incident angle of the monochromatic light. The selectivity comes from the morphologically derived resonance on a macroscopic length scale, which comprises grating due to the interference of multiply scattering light. Consequently, by combining dielectrically disordered structures with photobleaching, the manipulation of the chemical and/or physical state of ions or molecules is possible inside a small spatial volume. This phenomenon can be a promising candidate for high-density optical storage; information is recorded as three-dimensional random interference pattern.

Various properties concerning the hole production were extensively studied from both experimental and theoretical viewpoints, and significant progress has been made accordingly [3-7]. The previous studies revealed that the amount of optical absorption and the efficiency of scattering have significant effects on hole profile.

Table I. Volume fraction of mixtures (%) and their abbreviated representations.

Notation	Sm^{2+}-doped glass powder *	Sm^{2+}-undoped glass powder **
100SM	100	0
50SM-50G	50	50
33SM-67G	33.3	66.7
20SM-80G	20	80
10SM-90G	10	90

* The composition is $15Na_2O \cdot 85B_2O_3 \cdot 1.0SmO$ (mol%).
** The composition is $15Na_2O \cdot 85B_2O_3$ (mol%).

In the present investigation, we examine the polarization dependence of hole formation in dielectrically disordered media consisting of Sm^{2+}-doped glass powders.

2. EXPERIMENTAL

Sm^{2+}-doped glass powders were prepared by grinding the bulk glass [7]. The nominal composition was $15Na_2O \cdot 85B_2O_3 \cdot 1.0SmO$ (molar ratio). We also made the Sm^{2+}-undoped glass powders with the composition of $15Na_2O \cdot 85B_2O_3$ (molar ratio). According to scanning electron microscope images, the average size of finely grained powders was 1~3μm. The volume fraction of mixtures and their notations were described in Table I.

A linear polarized beam from a tunable cw dye laser (Rhodamine 6G, linewidth~$1.3 cm^{-1}$) was used to burn and subsequently to probe the hole. The beam diameter on the sample was about 3 mm. First, the sample was irradiated with the writing laser beam for 60 s to burn the hole through the photobleaching of Sm^{2+}. The burning power was 30 mW. Then, the laser beam was attenuated by a factor of 10^3 and used

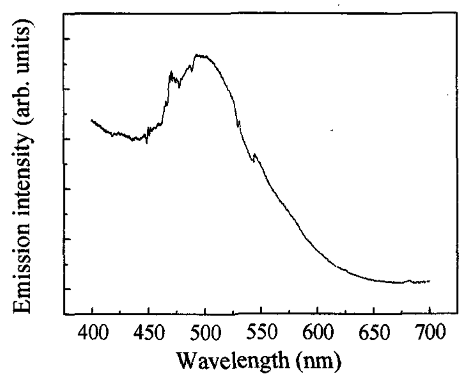

Fig. 1. Excitation spectrum at room temperature for 100SM obtained by monitoring the fluorescence due to the $^5D_0 \rightarrow ^7F_2$ transition of Sm^{2+} around 720nm.

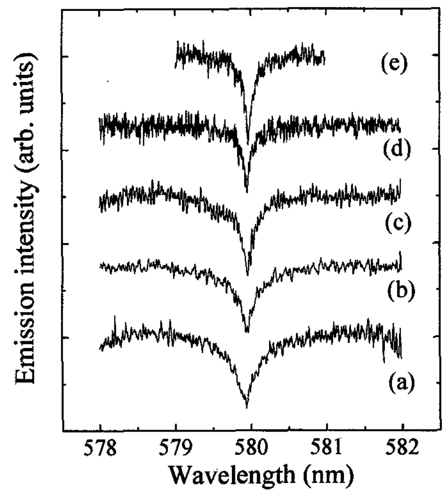

Fig. 2. Hole profiles for (a)100SM, (b)50SM-50G, (c)33SM-67G, (d)20SM-80G, and (e)10SM-90G normalized to hole depths as a function of the frequency of laser light. The burning of hole was carried out at the wavelength of 580 nm.

as the reading beam to probe the hole; the wavelength or incident angle of the attenuated laser beam was scanned while the fluorescence of Sm^{2+} due to $^5D_0 - ^7F_{0,1,2}$ transitions in the ranges of 680 to 720 nm was detected in the reflection geometry. For the measurement of angular dependence, the sample was rotated around an axis perpendicular to the incident plane, so that the angle of incidence was varied.

3. RESULTS AND DISCUSSION

Figure 1 shows the excitation spectrum of Sm^{2+}-doped glass powders. The measurement was performed monitoring the fluorescence due to the $^5D_0 \rightarrow ^7F_2$ transition of Sm^{2+} around 720nm. Intense absorption bands ranging 450 to 650nm are ascribable to the $4f^6 \rightarrow 4f^55d$ transition of Sm^{2+}. Excitation into the $4f^6 \rightarrow 4f^55d$ transition of Sm^{2+} causes the photobleaching as a result of the photoionization of Sm^{2+} to Sm^{3+} [8]

When the photoionization of Sm^{2+} is combined with multiple light scattering in glass powders, holes are burned in the wide wavelength ranges of visible spectrum [7]. Figures 2 and 3 show hole profiles for 100SM, 50SM-50G, 33SM-67G, 20SM-80G, and 10SM-90G. The burning of hole was performed at the wavelength of 580 nm. The fluorescence intensity is shown in Fig. 2 as a function of laser frequency. As the volume fraction of Sm^{2+}-undoped glass powders increases, the hole profile becomes narrow. The hole width of 10SM10-90G becomes smaller by approximately a factor of 3 than that of the 100SM. In Fig. 3 is shown the fluorescence intensity as a function of the incident angle of laser beam. The hole width slightly decreases with increasing the volume fraction of Sm^{2+}-undoped glass powders.

The mechanism of hole formation is described as follows. When a writing laser beam with frequency ν_1 and wave vector k_1 propagates through glass powder, a random interference

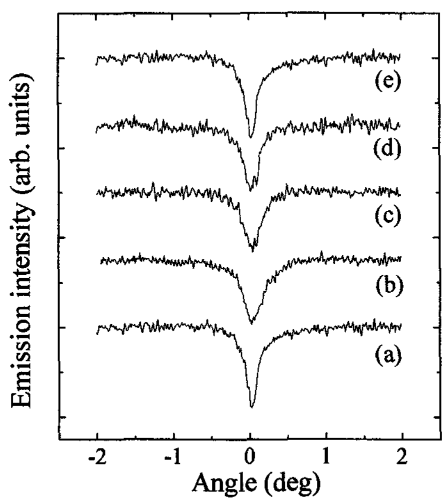

Fig. 3. Hole profiles for (a)100SM, (b)50SM-50G, (c)33SM-67G, (d)20SM-80G, and (e)10SM-90G normalized to hole depths as a function of the incident angle of laser light. The burning of hole was carried out at the wavelength of 580 nm

pattern, known as volume speckle, is formed and recorded inside the medium through the photoionization of Sm^{2+}. The reading laser beam with frequency ν_2 and wave vector k_2 induces the fluorescence due to Sm^{2+} as a function of $\Delta\nu = \nu_2 - \nu_1$ and $\Delta k = k_1 - k_2$. Under the conditions of $\nu_2 = \nu_1$ and $k_1 = k_2$, the interference pattern created by the reading beam coincides with that by the writing beam. Then, the reading beam passes through only the photobleaching paths in the medium and brings about the decrease in fluorescence intensity. A hole is thus burned in both frequency and wave-vector domains.

Since the frequency correlation of fluctuations caused by writing and reading beams is inversely proportional to the average travel time of photons through the sample [9, 10], it is anticipated that the hole width in the frequency domain is of the order of inverse of photon lifetime in the medium. On the other hand, it is considered that the hole width in the wave-vector domain is inversely proportional to the lateral spread of photons in the medium.

When Sm^{2+}-doped glass powders are mixed with Sm^{2+}-undoped glass powders, the scattering strength does not vary so significantly, as we previously revealed from coherent backscattering experiments [6]. The sample dependences of hole profiles, which are shown in Figs. 2 and 3, can be thus explained only by the variation in the amount of Sm^{2+} absorption. When diffusive absorption length L_a is smaller than the sample thickness, absorption removes light paths longer than L_a. The decrease in Sm^{2+} absorption brings about the increase in L_a, or light paths. Owing to the increased light paths, the average length of time that a photon stays inside the medium becomes long, and also photons spatially spread within the medium, sharpening both the spectral and angular widths of hole profiles. Because the scattering in glass powders is less efficient, the effect of Sm^{2+} absorption on the hole profile

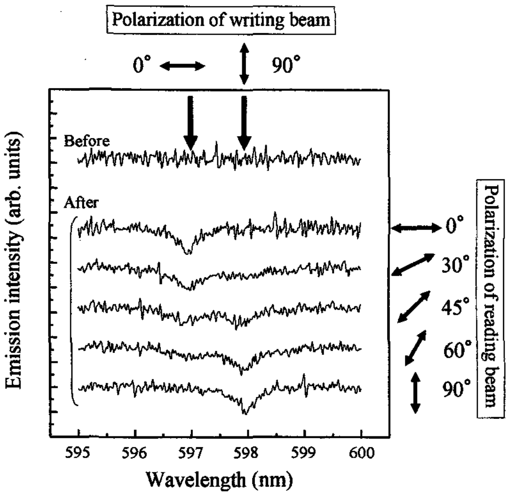

Fig. 4. Excitation spectra for 100SM before and after the irradiation with writing laser beams at the wavelengths of 597 and 598 nm. These writing lights correspond to the polarization directions of 0° and 90°, respectively. The readout results for two holes are shown for the polarization angles of 0°, 30°, 45°, 60°, and 90°.

can be quantitatively interpreted in terms of the intensity correlation of fluctuations based on the diffusion approximation, as we described previously [6].

Also, it is expected that the hole formation depends on the polarization of laser beam, because this phenomenon is based on the interference of light waves. Figure 4 shows the readout results for two holes in frequency domain, which are recorded at the wavelengths at 597 and 598 nm with the polarization angle of 0° to 90°, respectively. The readout was carried out with a polarization angle of 0°, 30°, 45°, 60°, and 90°. Holes are most clearly observed when the polarization directions of writing and reading laser beams are parallel to each other. As the polarization direction of reading beam is rotated with respect to that of the writing laser beam, the hole formation becomes ambiguous. The hole disappears completely when the polarization direction of reading beam is perpendicular to that of writing beam. This result confirms that in addition to the frequency and wave vector of monochromatic light, the information on the polarization direction is registered in the medium.

4. CONCULUSIONS

We have demonstrated the spatial manipulation of valence states of samarium ions using the interference of multiply scattering light in glass powders. Holes are burned in frequency and wave-vector domains, and the hole profile can be controlled by the amount of optical absorption. The hole formation depends on the polarization directions of writing and reading laser beam; the hole is most clearly detected when the frequency, the wave vector, and the polalization of reading laser beam coincide with those of writing laser beam.

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