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Wavelength- and angle-selective properties of optical memory effect by interference of multiple-scattered light in Sm-doped ZnS nanocrystals

Atusi Kurita,[1] Yasuo Kanematsu,[1] Masahito Watanabe,[1] Kuniko Hirata,[1] Takashi Kushida[2]

[1] *Department of Physics, Osaka University, Toyonaka, Osaka 560-0043, Japan*
[2] *Graduate School of Materials Science, Nara Institute of Science and Technology, Ikoma, Nara 630-0101, Japan*

Abstract

Our recent experiments have shown that both the wavelength and the angle of incident light are memorized in Sm-doped ZnS nanocrystals by photobleaching coupled with interference of the multiple-scattered light. We measured the angle-selective properties by scanning the incident angle, as well as the wavelength-selective properties, and discuss the density of data storage that can be derived from this effect.

Keywords : optical memory effect; interference; multiple scattering; samarium

Recently we have discovered a new kind of optical memory effect in Sm-doped ZnS nanocrystals[1]. The effect was observed in the excitation spectrum of fluorescence, and a 'hole' was found to persist in the excitation spectrum after irradiation of monochromatic light. Thus this effect appears to be similar to the persistent spectral hole-burning effect. However, the hole was only observed when not only the wavelength but also the incident angle of the probe light coincided with those of the recording light. This observation indicates that this effect differs from the conventional persistent hole burning in kind. We have shown that the recording process of this effect is that the interference pattern of multiple-scattered light, which is dependent on both the wavelength and the incident angle of the recording light, is registered by photobleaching of Sm as a spatial modulation of the absorbance. The optical inhomogeneity of the nanocrystal sample probably makes it a multiple-scattering medium. In the reading process, the intensity of the fluorescence from the sample is proportional to the overlap integral between the spatially modulated absorbance and the interference pattern formed by the reading, or exciting, light. Thus, the fluorescence intensity shows a dip when both the wavelength and the incident angle of the reading light coincide with those of the recording light. In this paper, we report angle selective properties, which are most characteristic of this effect, measured by continuously scanning the incident angle of the light beam and compare them with the wavelength selective properties.

The samples employed were Sm-doped ZnS nanocrystals coated by pentafluorothiophenol. Sample preparation procedure and fundamental optical properties are described in a separate paper[2]. The pentafluorothiophenol coating prevents the nanocrystals from aggregation and makes them soluble in alcohol while insoluble in water. The sample was obtained from a ethanol solution as powder by evaporating the solvent. Each powder particle was made up of many nanocrystals and appeared milky with slightly pink color. The powder was ground and sorted by particle size by using a sequence of sieves with the mesh sizes of 125, 75 and 45 μm. We also prepared a sample by drying precipitates in water. This type of sample was obtained as a rather solid lump. The sample was placed on a stage equipped with a computer-controlled micrometer, which rotates the stage around an axis lying in its surface to scan the angle of the incident beam relative to the surface normal. The minimum step of the rotating angle was 0.0028°. The powder particles were dispersed on the stage sparsely enough so as not to touch each other. The sample was irradiated by attenuated light from a cw dye laser with a linewidth of 0.2 cm^{-1}, and the laser wavelength ($\sim$580 nm) or the incident angle was scanned while fluorescence around 630 nm was detected; the same laser beam was used for burning the

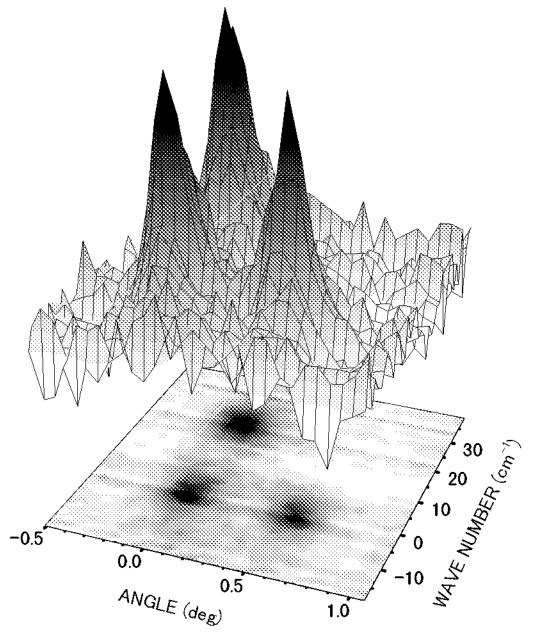

Fig. 1. Profile of three holes in the two-dimensional space of the wave number and the incident angle. The vertical axis represents decrease of the emission intensity. Burning time and power were 15 s and 0.6 mW/mm^2, respectively, for each hole. Sample was Sm-doped ZnS nanocrystals with the diameter larger than 125 μm.

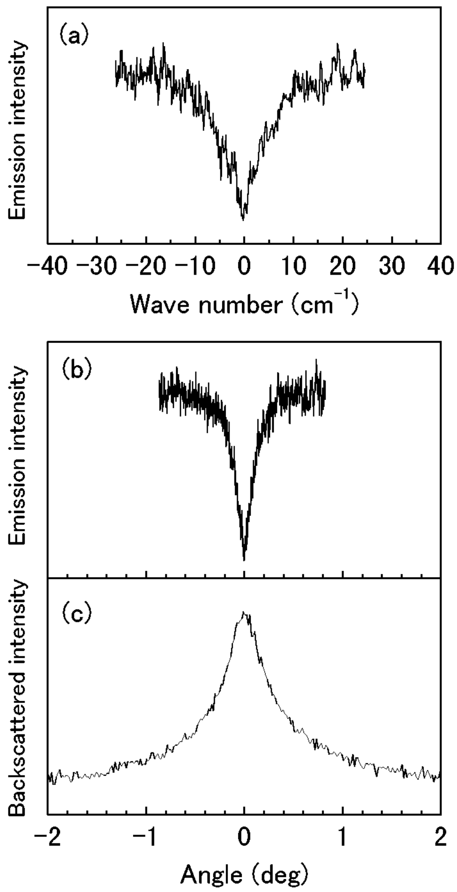

Fig. 2. Hole profile as a function of the wave number (a), hole profile as a function of the incident angle (b), and coherent backscattering cone (c) of particles of Sm-doped ZnS nanocrystals with the diameter between 75 and 125 μm.

holes, but the intensity was about 10^4 times higher than that for the fluorescence measurements.

Fig. 1 shows a profile of three holes measured by raster-scanning in the two-dimensional space of the wave number and the incident angle. It is evident that the hole memorizes both the wave number and the incident angle of the burning light, and the hole profiles along the wave-number axis and the angle axis look very similar. Holes are well isolated and there is no crosstalk between the holes within the measured region.

We compared the hole profile in sample particles with different sizes. The size of samples sorted by sieves was, (a) smaller than 45 μm, (b) between 45 and 75 μm, (c) between 75 and 125 μm, and (d) larger than 125 μm. In addition to these powder samples, (e) lump sample from precipitates, which looked whiter than the particles of the powder samples but looked uniform when it was observed by an optical microscope, was also measured. We burned a hole and measured its profile as functions of two kinds of variable, *i.e.*, by scanning the wavelength of the measuring laser while the incident angle fixed at the burning angle, and by scanning the incident angle while the wavelength fixed at the burning wavelength. We also measured the angle dependence of the coherent backscattered light from each sample. In multiple-scattering media, a constructive interference of time-reversed counter-propagating waves is known to enhance the intensity of the scattered light in the exact backward direction. Fig. 2 shows a result of the three kinds of measurement in sample (c). In Table 1, the width of the hole profile as a function of the wave number, the width of the hole profile as a function of the angle, and the width of the coherent backscattering cone of the samples with differing sizes are summarized. All of the three kinds of width narrow in order of increasing particle size.

The interference pattern that causes this memory effect results from waves which are scattered many times after impinging upon the sample at different points in the incident plane and encounter at a point within the sample. Therefore, the width of the hole profile as a function of the incident angle is determined by the distance l between the impinging points of the interfering waves, and is approximately given by λ/l, where λ is

Table 1. Widths of, the hole profile as a function of the wave number, the hole profile as a function of the incident angle, and the coherent backscattering cone.

Sample size (μm)	Width (FWHM)		
	Hole (cm^{-1})	Hole (deg)	Backscattering (deg)
(a) <45	18.5	0.35	0.88
(b) 45~75	12.9	0.31	0.74
(c) 75~125	10.6	0.22	0.60
(d) >125	6.3	0.17	0.59
(e) lump	8.6	0.44	1.15

the light wavelength. The distance l is limited by the particle size, which accounts for the particle-size dependence of the angular width of the hole shown in Table 1. The width is comparable to the width of Fraunhofer diffraction by a circular aperture, which is estimated at 0.27° (FWHM) when the diameter is 125 μm and λ is 0.58μm. In Table 1, the width of the backscattering cone is also larger for smaller particles. However, if it is compared with the angular width of the hole of the sample with the same particle size, we notice that the backscattering cone is broader than the hole. The backscattering cone stems from the interference of time-reversed counter-propagating waves which are scattered many times after impinging upon the sample and then exit from it. Thus the width of the backscattering cone is determined by the distance between the impinging point and the exit point[3]. This distance is also limited by the particle size. However, probably it is shorter than l, and the width is larger than that of the hole, which explains the discrepancy between the two kinds of angular width. As to sample (e), both the width of the hole profile as a function of the incident angle and the width of the backscattering cone are larger than those of sample (a). This result indicates that, although sample (e) is not small particles, diffusion length of the scattered light within the sample is less than 45 μm probably because of stronger scattering in sample (e) compared with other samples.

The width of the hole profile as a function of the wavelength, on the other hand, is determined by the difference of the optical pathlength between the interfering waves. This difference also is probably smaller for smaller particle, although it is not directly bounded by the particle size, because the light path is folded many times in the particle. Let us estimate the pathlength from the measured hole width by comparing it with the hole width that is obtained by an interference fringe in a uniform non-scattering medium with a mirror at one end of it[1]. If the hole width 10.6 cm^{-1} of sample (c), for example, was obtained from a fringe in a uniform medium with the refractive index 2.37 (of ZnS), the length of the medium would be 120 μm. Thus, in the powder samples, the total length of the folded path is comparable to the particle size. The hole width determines the density of data when this effect is utilized for optical data storage. The hole width of the uniform medium with a mirror corresponds to the storage density of conventional methods which is limited by the wavelength of light. The above comparison suggests that the density of data that can be stored by using the present effect in the powder samples might be nothing more than that with conventional methods. However, the hole of sample (e) is much narrower than the hole of sample (a), although the effective volume for the recording is smaller than sample (a), as is known from the angular width of the hole and the backscattering cone. This high storage density surmounting the wavelength limit presumably comes from the strong scattering of light in sample (e), where weak localization of light probably occurs[4]. If light is weakly localized and stays around the incident point for a long time, the path difference, or the time difference, between the interfering waves can be very long, and the hole becomes narrow.

In conclusion, we have compared the angle selective properties and the wavelength selective properties of the optical memory effect that is due to interference of multiple-scattered light in Sm-doped ZnS nanocrystals. The hole width has given information about scattered waves in the samples. The density of data that can be stored by utilizing this effect is possibly very high in strongly scattering media. Further studies are necessary for quantitative understanding of this effect.

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