Spectral Hole-Burning of Eu^{3+}:Y_2SiO_5 Crystal at 16 K *

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(Received 10 November 2006)

By using an Ar^+ ion laser, a tunable Rh 6 G dye laser (linewidth 0.5 cm^{-1}) pumped by the second harmonic of a YAG:Nd laser and a Coherent 899-21 dye laser as light sources and using a monochromator, a phase-locking amplifier and a computer as the data detecting system, we detect the optical properties of Eu^{3+}-doped Y_2SiO_5 crystal. Persistent spectral hole burning (PSHB) are observed in the Eu^{3+} ions spectral lines ($^5D_0-^7F_0$ transition) in the crystal at the temperature of 16 K. For 15 mW dye laser burning the crystal for 0.1 s spectral holes with hole width about 80 MHz both at 579.62 nm and at 579.82 nm are detected and the holes can remain for a long time, more than 10 h.

PACS: 78. 55. −m, 78. 20. −e, 42. 70. Ln

Study on persistent spectral hole burning has grown in importance over the past dozen years, both as a powerful method for the study on the dynamics of the amorphous state, energy transfer, and low-temperature photoeffects in solids and on surfaces and also as a possible method to achieve high-performance optical storage and parallel optical processing. Trivalent europium ions doped in crystals are often used for the study of high-resolution nonlinear spectroscopy because Eu^{3+} ions doped in crystals have a very long optical dephasing time T_2. A material with long dephasing time T_2 can be used for frequency-selective optical memory, both in the frequency domain (persistent spectral hole burning) and the time domain (photon echo memory).[1] The value of T_2 of impurity ions in solids mainly depends on (1) guest-ion/phonon interaction, (2) guest-ion/guest-ion interaction, and (3) guest-ion/host-ion interaction. Because the effect of phonons becomes smaller at low temperature the value of T_2 can become temperature independent by lowering temperature. The mutual interaction between guest ions can be decreased by doping crystals with low rare earth ion concentrations. The interaction between guest ions and host ions may be reduced by choosing appropriate host crystals composed of elements with small nuclear moment.

Eu^{3+}:Y_2SiO_5 crystal is a new luminescent material. Because the nuclear moments of the composite elements are small in this crystal, the spin–spin interaction between the guest ion and the host ion is minimized. The guest–host interaction does not cause large additional line broadening, therefore this crystal is a good optical data storage material. Spectral hole burning of Eu^{3+}:Y_2SiO_5 crystal usually is researched at liquid helium temperature. The spectral hole burning of the crystal is more difficult at the higher temperature. The spectral hole-burning of Eu^{3+}:Y_2SiO_5 crystal is investigated at higher temperature (16 K) in this study. The spectral hole can be also kept for a long time more than 10 h at 16 K in our experiment, while it can be kept for a long time more than 24 h at liquid helium temperature. This means that our crystal is of good quality.

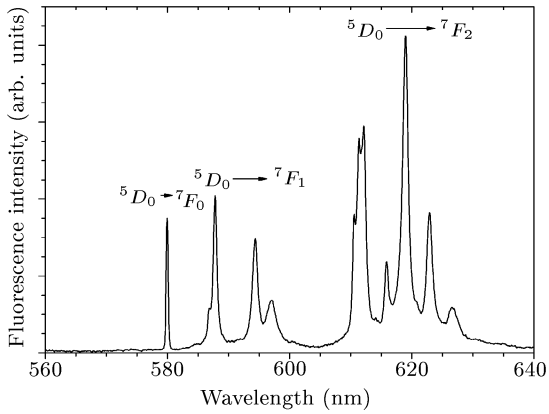


Fig. 1. $^5D_0 \rightarrow {}^7F_{0,1,2}$ fluorescence spectrum of Eu^{3+} in Y_2SiO_5 at 293 K (λ_{ex} = 514.5 nm).

The crystal was grown by Shanghai Institute of Optics and Fine Mechanics. Eu^{3+} ions-doped concentration was 0.15 wt.% in our sample. The emission spectra of the Eu^{3+}-doped Y_2SiO_5 crystal from

* Supported by the National Natural Science Foundation of China under Grant Nos 69807001 and 19834030.

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560 nm to 720 nm were measured using a Coherent Innova 10 argon ion laser (line 514.5 nm) excitation at 293 K. All fluorescence spectra were recorded with a Jobin-Ybon HR-1500 model monochromator and detected by an SR565 phase-locking amplifier. Figure 1 shows the fluorescence spectra of $^5D_0 \rightarrow {}^7F_{0,1,2}$ transitions. Figure 2 shows the fluorescence spectra of $^5D_0 \rightarrow {}^7F_{3,4}$ transitions.

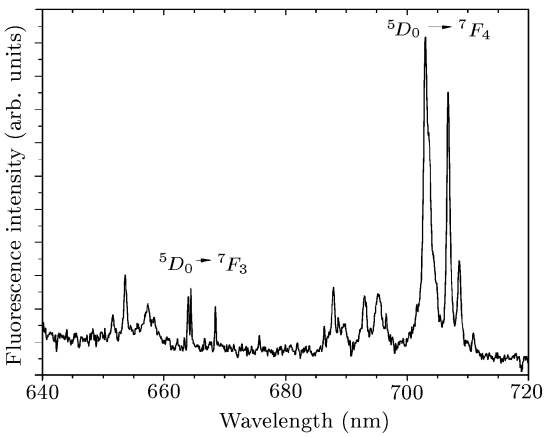


Fig. 2. $^5D_0 \rightarrow {}^7F_{3,4}$ fluorescence spectrum of Eu^{3+} in Y_2SiO_5 at 293 K ($\lambda_{ex} = 514.5$ nm).

The photoluminescence excitation spectrum excited by a Q-switched Nd^{3+}:YAG laser pumped tunable rhodamine 6G dye laser with 0.5 cm^{-1} linewidth was studied by monitoring fluorescence of $^5D_0 \rightarrow {}^7F_2$ transition whose main peak wavelength is about 618 nm. Eu^{3+} ions occupy two optical sites whose spectral lines of the $^7F_0 \leftrightarrow {}^5D_0$ transition are about 0.2 nm away from each other at room temperature and both the sites are the luminescence centres with the C_1 symmetry. The site selective fluorescence spectra of the Eu^{3+} $^5D_0 \rightarrow {}^7F_0$ transition were detected by exciting every site respectively.

A tunable Rh 6 G dye laser with 0.5 cm^{-1} spectral linewidth pumped by the second harmonic of a pulsed Nd^{3+}:YAG laser was used as the excitation light source; a prism monochromator, a boxcar and a computer were used as the data detecting system. The photoluminescence excitation spectrum in the Eu^{3+}: $^7F_0 - {}^5D_0$ region (579 nm) was detected by monitoring the $^5D_0 \rightarrow {}^7F_2$ fluorescence centred at 618 nm with spectral range from 610.5 nm to 625.5 nm at 293 K.

Figure 3 shows the two spectral peaks (site 1: 579.62 nm and site 2: 579.82 nm), suggesting that Eu^{3+} ions occupy sites of two Y^{3+} ions in the Y_2SiO_5 crystal were shown in the excitation spectrum. Site selective fluorescence spectra were also detected using the 579.62 nm and 579.82 nm excitation wavelengths of a Coherent 899-21 dye laser at 77 K in our experiments, respectively. The line-width of the Coherent 899-21 dye laser was 1 MHz full width at half maximum (FWHM). They were displayed in Fig. 4. Fluorescence spectra of $^5D_0 \rightarrow {}^7F_{1,2,3,4}$ transitions can clearly be seen in the site selective fluorescence spectra. By studying the selective fluorescence spectra we found that the positions of spectral peaks in Fig. 4(a) were different from them in Fig. 4(b). The fluorescence spectra of different sites do not overlap each other.

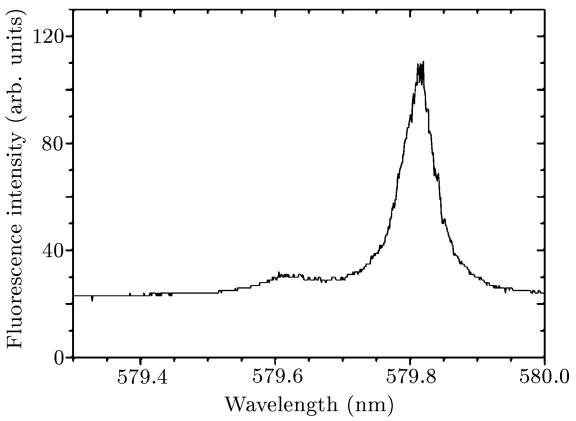


Fig. 3. Photoluminescence excitation spectrum in the $^5D_0 \rightarrow {}^7F_0$ region of Eu^{3+}:Y_2SiO_5 crystal by monitoring the $^5D_0 \rightarrow {}^7F_2$ fluorescence centred at 618 nm with spectral range from 610.5 nm to 625.5 nm at 293 K ($\lambda_{em} = 618$ nm, $\Delta\lambda_{em} \approx 15$ nm).

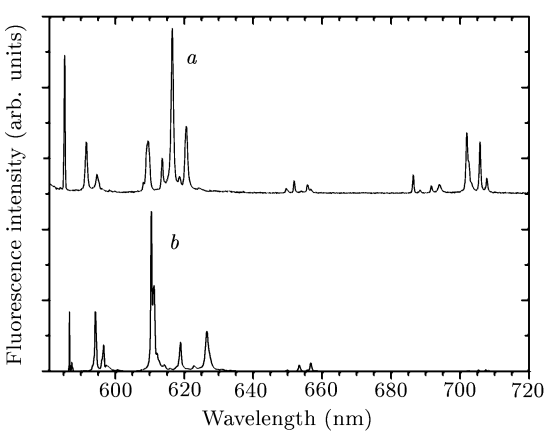


Fig. 4. (a) Site selective fluorescence spectra of site 1 at the temperature of 77 K ($\lambda_{ex} = 579.62$ nm); (b) site selective fluorescence spectra of site 2 at the temperature of 77 K ($\lambda_{ex} = 579.82$ nm).

Y_2SiO_5 belongs to the monoclinic biaxial crystal class $2/m$ (space group $C2/c$).[2] The lattice parameters were given in Refs. [3,4]. Because of the biaxial character of the system three principal axes of polarization can be defined: (1) the ⟨010⟩ direction which is a two-fold symmetry axis of the crystal, and (2) the $D1$ and $D2$ directions perpendicular to each other and to the ⟨010⟩ direction, which correspond to extinction directions[4] when the sample is viewed in the ⟨010⟩ direction between crossed polarizers. These directions were determined exactly with respect to the crystal-

lographic axis by x-ray analysis and shown in Ref. [4].

Figure 5 shows the XRD patterns of the crystal which contain more than ninety diffraction peaks. Table 1 displays the lattice constants a, b, c and β calculated by

$$\frac{1}{d^2} = \frac{1}{\sin^2\beta}\left(\frac{h^2}{a^2} + \frac{k^2\sin^2\beta}{b^2} + \frac{l^2}{c^2} - \frac{2hl\cos\beta}{ac}\right),$$

where d is the interplanar distance, and h, k, l are the lattice indices.

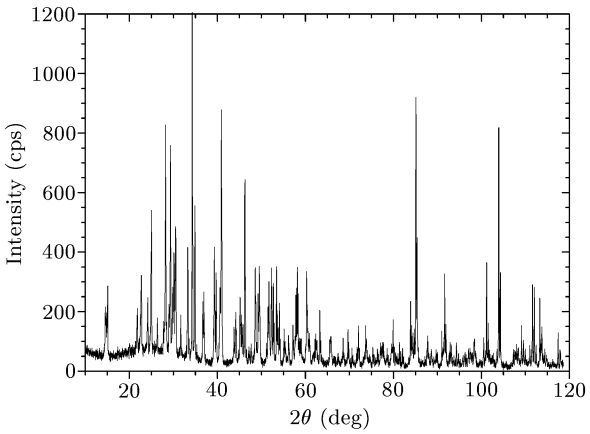


Fig. 5. Pattern of x-ray diffraction of Eu^{3+}: Y_2SiO_5 crystal.

Table 1. Crystal lattice constants of Eu^{3+}:Y_2SiO_5.

	Doped	Undoped (Ref. [3])
a (nm)	1.2533 (85)	1.25013 (17)
b (nm)	0.6735 (36)	0.67282 (8)
c (nm)	1.0435 (80)	1.04217 (13)
β (deg)	102.58 (72)	102.682 (11)

Spectral hole was burnt on the spectral line of the $^7F_0 - {}^5D_0$ transition of the Eu^{3+} ions. For hole burning of Eu-doped crystals, the spectral resolution of laser is very important because the homogeneous linewidths of Eu-doped crystals are of the order of a kilohertz or less. However, it is in practice difficult for a commercial dye laser to achieve that narrow spectral resolution. In our experiment, a Coherent 899-21 dye laser with 1 MHz spectral linewidth pumped by an Ar^+ ion laser was used for spectral hole-burning of Eu^{3+}:Y_2SiO_5 crystal. The dye laser was tuned to the $^7F_0 - {}^5D_0$ transition of site 1 (579.62 nm) and site 2 (579.82 nm), respectively. The laser beam was then focused on the sample at 16 K in a cryostat with a power of 15 mW. A CTI-based 10 K closed cycle refrigerator systems was used for low temperature experiments. The system of a Jobin-Yvon HR-1500 model monochromator, an SR565 phase-locking amplifier and a computer was used for receiving and printing transmission spectra. Figure 6 shows the experimental setup of spectral hole burning.

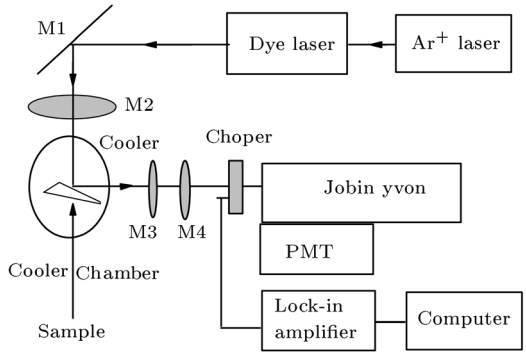


Fig. 6. Experimental setup of spectral hole burning.

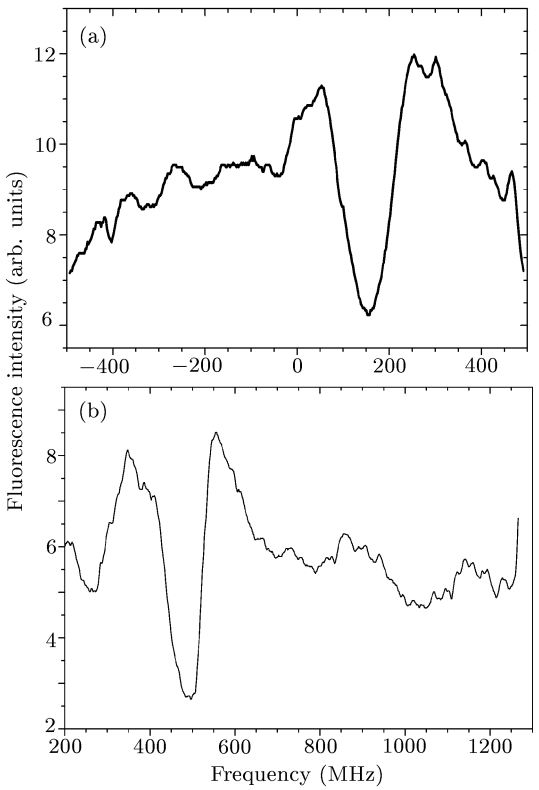


Fig. 7. (a) Single hole in the absorption line of $^5D_0 - {}^7F_0$ transition of site 1 at temperature 16 K. The linewidth of scanning dye laser is 1 MHz, the hole burning laser wavelength is 579.62 nm, the power of hole-burning laser is P_h = 15 mW, the power of scanning laser is P_d = 1 mW, and the receiving wavelength of monochromator is λ_{em} = 618 nm. (b) Single hole in the absorption line of the $^5D_0 - {}^7F_0$ transition of site 2 at the temperature of 16 K. The linewidth of scanning dye laser is 1 MHz, the hole burning laser wavelength is 579.82 nm, the power of hole-burning laser is P_h = 15 mW, the power of scanning laser is P_d = 1 mW, and the receiving wavelength of monochromator is λ_{em} = 611 nm.

Spectral holes with about 80 MHz hole width both at 579.62 nm and at 579.82 nm are observed after 15 mW dye laser burns Eu^{3+}:Y_2SiO_5 crystal for 0.1 s at the temperature of 16 K. Hole spectra were recorded by scanning the dye laser near the hole-burning laser wavelength (579.62 nm for site 1 and 579.82 nm for site

2, respectively) while monitoring the fluorescence of the $^5D_0 \rightarrow {}^7F_2$ transition (at 618 nm for site 1 and at 616 nm for site 2, respectively), as shown in Figs. 7(a) and 7(b). The laser power for scanning was 1 mW. Spectral holes can last for a long time more than 10 h at the temperature of 16 K.

In summary, we have presented optical properties of Eu^{3+}:Y_2SiO_5 crystal. Two absorption peaks in photoluminescence excitation spectrum of the $^5D_0 \rightarrow {}^7F_0$ transition show that Eu^{3+} ions occupy two Y^{3+} ion sites in the Y_2SiO_5 crystal. Spectral hole is burnt at higher temperature (16 K) while it is usually burnt at 4.3 K[1] in Eu^{3+}:Y_2SiO_5 crystal. Spectral hole remain for a long time more than 10 h in Eu^{3+}:Y_2SiO_5 crystal at 16 K.

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