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Persistent spectral hole burning and optical properties of Sm^{2+} doped into $Mg_{0.5}Sr_{0.5}FCl_{0.5}Br_{0.5}$ mixed crystal

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Abstract

We measured the temperature dependence of the fluorescence of Sm^{2+} ions doped into a $Mg_{0.5}Sr_{0.5}FCl_{0.5}Br_{0.5}$ mixed crystal and observed the persistent spectral hole burning at 90 K and 300 K. The electrons appearing at the $^5D_0 \rightarrow {}^7F_0$ transition mostly come via the 4f5d $\rightarrow {}^5D_1 \rightarrow {}^5D_0 \rightarrow {}^7F_0$ route but some of the electrons come directly from the 4f5d band. At 90 K, persistent spectral hole burning was achieved much more efficiently by the photon-gated spectral hole-burning process than by the one-photon hole-burning process, while the one-photon hole-burning process is much more efficient at 300 K.

1. Introduction

Persistent spectral hole burning (PSHB) has attracted considerable interest because of its potentiality for use in a high-density optical memory system or an all-optical data processor such as a packet switch. A drastic increase in data storage density using PSHB is indicated by the ratio $\Gamma_{inh}/\Gamma_{hole}$, where Γ_{inh} is the inhomogeneous linewidth of the doped ions, and Γ_{hole} is the width of the spectral hole. For practical use, PSHB at room temperature is required as well as the stability of the spectral hole. If reading photons can also burn holes, they destroy the information during the process of reading. However, hole burning takes place with the presence of two photons in photon-gated hole burning, while reading requires only one photon. Thus, PSHB achieved by photon-gated spectral hole burning at room temperature is an important candidate for producing high-density optical memory in the frequency domain because it does not destroy the information during the reading process.

Jaaniso and Bill reported room temperature PSHB for the Sm^{2+}-doped BaFCl crystal for the first time [1]. Since then, room temperature PSHB for Sm^{2+} doped into several different

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host materials has been reported [2–4]. The optical properties of the Sm^{2+} ion have also been widely studied [5–9] because it is one of the most important candidates for producing high-temperature PSHB materials. PSHB mechanisms are closely related to the optical properties of the ions doped into the crystals or glasses. This paper presents the optical properties and the results of PSHB for Sm^{2+} ions doped into $Mg_{0.5}Sr_{0.5}FCl_{0.5}Br_{0.5}$ mixed crystal at two different temperatures, 90 K and 300 K.

2. Experiment

The Sm^{2+}-containing $Mg_{0.5}Sr_{0.5}FCl_{0.5}Br_{0.5}$ mixed crystal was prepared by mixing stoichiometric amounts of MgF_2, $SrCl_2{\cdot}6H_2O$, $BrCl_2{\cdot}6H_2O$ and Sm_2O_3 and then reacting the mixture with H_2 gas for one hour at 1100 °C in an alumina tube.

In the measurements of the temperature dependence of the ${}^5D_J \rightarrow {}^7F_0$ ($J = 0, 1, 2$) (refer to figure 1) fluorescence intensity of the Sm^{2+} ion, the sample was put into a helium gas closed-temperature-cycle cryostat, in which the temperature was varied from 9 to 300 K. The 355 nm third-harmonic-generation (THG) light of a Nd:YAG pulsed laser was used for excitation. The fluorescence intensity, due to the ${}^5D_J \rightarrow {}^7F_0$ transition, was obtained using a spectrometer (1702 Spex) equipped with a photomultiplier tube (Hamamatsu R298).

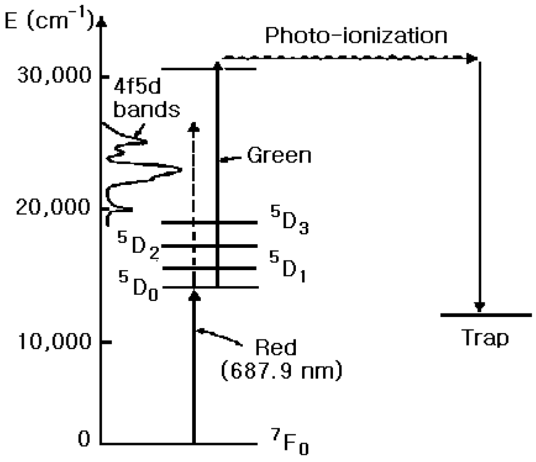

Figure 1. Schematic energy levels for Sm^{2+} doped into BaFCl crystal (reference [9]).

The PSHB was also observed on the excitation of the ${}^7F_0 \rightarrow {}^5D_1$ transition of the Sm^{2+} ion at two different temperatures, 90 K and 300 K, because the sample was prepared in a powder state. The linewidth of the tunable dye laser (DCM) used in this experiment is $\sim$1.0 cm^{-1} full width at half-maximum. The excitation spectrum was obtained by scanning the output of an Ar^+-ion laser-pumped tunable dye laser from 15 885 to 15 772 cm^{-1} while monitoring the ${}^5D_0 \rightarrow {}^7F_0$ emission at 14 477 cm^{-1}. The PSHB was performed by two different hole-burning processes: the photon-gated hole-burning process at 90 K and the one-photon hole-burning process at 300 K. The PSHB was achieved by using overlapped 514.5 nm (1.04 W cm^{-2}) beams from an argon-ion laser and a dye laser (5.4 W cm^{-2}) at 90 K.

In the photon-gated hole-burning process, the gated laser beam was blocked after a certain time of irradiation and the excitation spectrum was measured as described above. However, the PSHB at 300 K was observed by using a tunable dye laser operating only at 5.4 W cm^{-2}, because the one-photon hole-burning process is more efficient than the photon-gated hole-burning process for burning a hole. The scanning laser power was attenuated by the neutral density filter to less than 10% of the burning laser power in both hole-burning processes.

3. Results and discussion

Figure 2(a) shows the temperature dependence of the $^5D_J \to {}^7F_0$ ($J = 0, 1, 2$) fluorescence intensity of Sm^{2+} ions doped into $Mg_{0.5}Sr_{0.5}FCl_{0.5}Br_{0.5}$ mixed crystal. The fluorescence intensity of the $^5D_0 \to {}^7F_0$ transition increases with increasing temperature, while the fluorescence intensity of the $^5D_2 \to {}^7F_0$ transition decreases. However, the fluorescence intensity of the $^5D_1 \to {}^7F_0$ transition increases with temperature below 150 K then decreases with temperature above 150 K. According to the multi-phonon relaxation theory, one expects an increase in the non-radiative transition probability of the $^5D_1 \to {}^5D_0$ transition as the temperature increases. This explains why the $^5D_0 \to {}^7F_0$ fluorescence intensity increases while that of the $^5D_1 \to {}^7F_0$ transition decreases with increasing temperature.

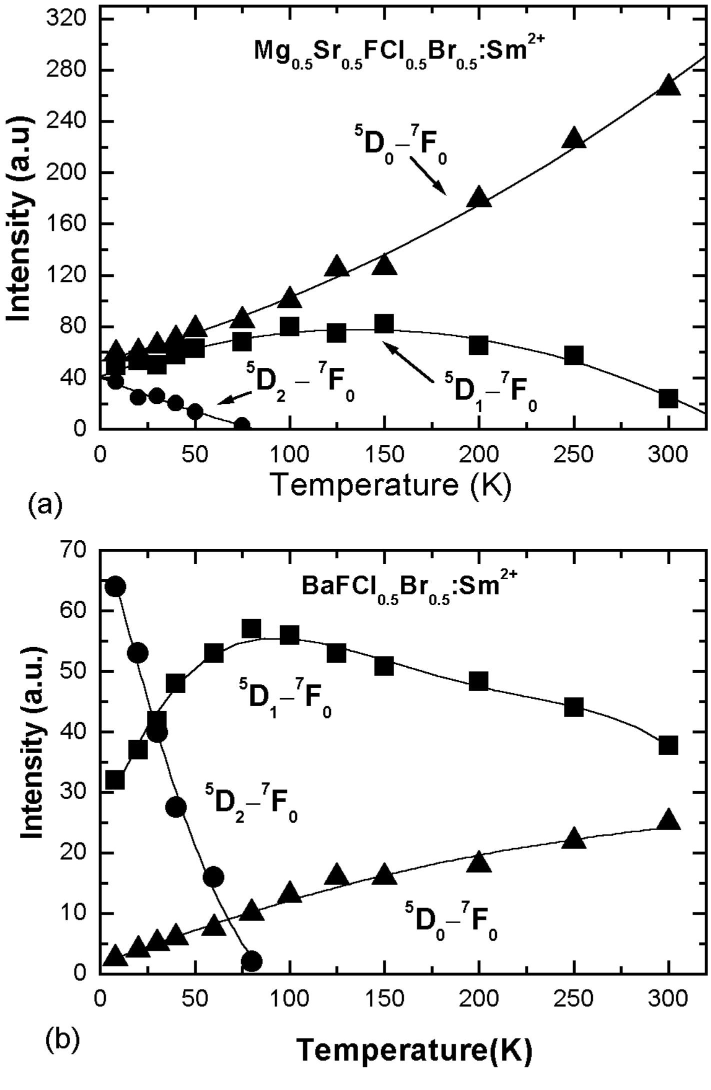

Figure 2. The temperature dependence of the $^5D_J \to {}^7F_0$ fluorescence intensity of Sm^{2+} doped into (a) $Mg_{0.5}Sr_{0.5}FCl_{0.5}Br_{0.5}$ mixed crystal, (b) $BaFCl_{0.5}Br_{0.5}$ single crystal (reference [10]).

Song *et al* reported the temperature dependence of the $^5D_J \rightarrow {}^7F_0$ ($J = 0, 1, 2$) fluorescence intensity of Sm^{2+} ions doped into $BaFCl_{0.5}Br_{0.5}$ single crystal [10]. They showed that the fluorescence intensity of the $^5D_2 \rightarrow {}^7F_0$ transition is greater than that of the $^5D_1 \rightarrow {}^7F_0$ and the $^5D_0 \rightarrow {}^7F_0$ transitions below 20 K. They also showed that the fluorescence intensity of the $^5D_0 \rightarrow {}^7F_0$ transition is weaker than that of the $^5D_1 \rightarrow {}^7F_0$ transition over all temperature ranges studied. However, our results show that the fluorescence intensity of the $^5D_0 \rightarrow {}^7F_0$ transition is stronger than those of other transitions. This probably means that some of the electrons in the $^5D_0$ level come directly from the 4f5d band through the non-radiative relaxation process and some of them come from the 4f5d band via the $^5D_1$ level. This direct transition from the 4f5d band to the $^5D_0$ level may be due to the phonon relaxation since the phonon density of states in the mixed crystal studied may be different from that of single crystal. Song *et al* also showed that the temperature dependence of the $^5D_J \rightarrow {}^7F_0$ transition of Sm^{2+} ions doped into glass is quite different from that of Sm^{2+} doped into single crystal and that most electrons in the $^5D_0$ level come directly from the 4f5d band through the non-radiative relaxation process [10].

Figure 3(a) shows the hole burned around the peak position of the $^7F_0 \rightarrow {}^5D_1$ transition by using a tunable dye laser only at 300 K. It is also possible to achieve PSHB by using the photon-gated spectral hole-burning process at 300 K. However, the one-photon hole-burning process is much more efficient than the photon-gated hole-burning process at 300 K. The hole obtained was very stable at room temperature and its hole depth had not changed even 24 hours later without any irradiation. However, it was easily erased by irradiation with an argon-ion laser at 514.5 nm within a few seconds. Figure 3(b) shows the PSHB achieved at different wavelengths of the $^7F_0 \rightarrow {}^5D_1$ transition by the photon-gated hole-burning process at 90 K. The holes obtained were made by irradiating with the 514.5 nm beam of an argon-ion laser and the dye laser output for 15 seconds simultaneously. Subsequent hole burning at different wavelengths partially refills the holes burnt earlier. Such behaviour may be a weak point of the multiplex PSHB. This is apparent on comparing hole 2 and hole 3, because they are positioned at the same height on the excitation spectrum. The holes observed in the photon-gated hole-burning process were also easily erased by irradiation with an argon-ion laser at 514.5 nm within a few seconds. However, they were very stable for several hours without any laser irradiation. Accordingly, we do not know the actual hole lifetime because the hole was stable during the experiment. At 90 K, PSHB was also observed by using a tunable dye laser only, but it was much less efficient than that achieved by the photon-gated hole-burning process. It is quite interesting that the photon-gated spectral hole-burning process is much more efficient than the one-photon hole-burning process at 90 K, while the latter process is more efficient than the former process at 300 K. This probably means that there could be two different hole-burning processes depending on the temperature.

The hole-burning processes of Sm^{2+} ions doped into host materials have been described in two categories: photon-gated PSHB processes in which the electron traps are mainly attributed to the Sm^{3+} ions and one-photon two-step hole-burning processes where the traps are considered not to be Sm^{3+} ions. For example, the electrons of Sm^{2+} ions doped into fluoride single crystals are usually ionized through the excited state into the conduction band by the photon-gated PSHB [9, 11]. This photo-ionization of Sm^{2+} ions is considered to be the main reaction for persistent spectral hole burning [9, 12, 13]. However, it was also reported that holes are usually formed by one-photon excitation of Sm^{2+} ions doped into fluoride and borate glasses [14]. The electrons in the excited states can also be gated into a conduction band by the burning light itself; this is called the one-photon two-step hole-burning process. Recently, Nogami *et al* also reported PSHB in the $^5D_0 \rightarrow {}^7F_0$ transition of Sm^{2+} ions doped into glasses with and without OH bonds prepared by the sol–gel method and explained that PSHB was achieved by both the

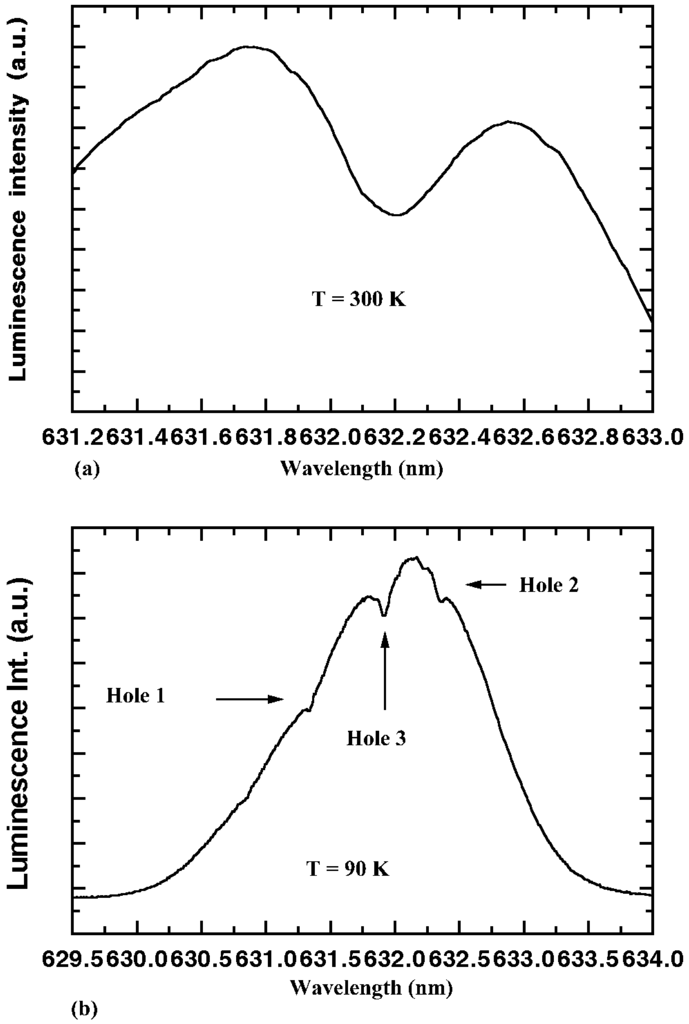

Figure 3. Persistent spectral holes in the $^7F_0 \leftrightarrow {}^5D_1$ transition of Sm^{2+} in Mg$_{0.5}$Sr$_{0.5}$FCl$_{0.5}$Br$_{0.5}$ mixed crystal (a) at 300 K and (b) at 90 K. The hole number represents the order in which the holes were burned.

photo-induced rearrangement of the OH bonds and the photo-ionization of Sm^{2+} ions [15]. However, to the best of our knowledge, there has been no report that hole-burning processes depend on temperature. At present, we do not fully understand why the photon-gated spectral hole-burning process is more efficient than the one-photon hole-burning process at 90 K, while the latter process is more efficient at 300 K. Theoretical and experimental work is thus needed to understand the detailed physics of this phenomenon.

4. Conclusions

We measured the temperature dependence of the $^5D_J \rightarrow {}^7F_0$ ($J = 0, 1, 2$) fluorescence of Sm^{2+} ions doped into a Mg$_{0.5}$Sr$_{0.5}$FCl$_{0.5}$Br$_{0.5}$ mixed crystal. The fluorescence intensity of the $^5D_0 \rightarrow {}^7F_0$ transition is stronger than that of any other transition over all of the temperature ranges studied. This means that electrons appearing at the $^5D_0 \rightarrow {}^7F_0$ transition mostly come from the 4f5d band via the $^5D_1$ level, while some electrons come directly from the 4f5d band through non-radiative relaxation.

We also performed PSHB experiments at two different temperatures, 90 K and 300 K. The photon-gated hole-burning process is much more efficient than the one-photon hole-burning process at 90 K, while the latter is more efficient than the former at 300 K. It is also possible to achieve a persistent spectral hole by using the former or the latter process only at both temperatures, but this has low efficiency. This probably indicates that two kinds of hole-burning mechanism operate in the mixed crystal studied.

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