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Growth and characterization of ^{167}Er doped-Y_2SiO_5 single crystal

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1. Introduction

The hyperfine structure of the optical transition in rare-earth-doped crystal offers suitable three-level schemes for efficient electromagnetically induced transparency (EIT) and stimulated Raman adiabatic passage (STIRAP) in solid state materials. The use of the hyperfine structure of nuclear spin of isotopically purified rare-earth dopant in Y_2SiO_5 single crystal is promising and indeed ^{151}Eu (I=5/2) in Y_2SiO_5 single crystal has been grown and optically addressable nuclear spins are realized with six hours coherence time [1]. To realize long-distance quantum optical communications through optical fiber (e.g. 1000 Km), however, it is very important to realize optical addressable nuclear spins with long coherence time at telecommunications wavelength of 1.5 um.

Er has optical transitions at 1.5 um and six different types of isotopes which are 162, 164, 166, 167, 168, 170 [2]. Among them, only ^{167}Er (natural abundance is 23 %) has nuclear spin I=7/2 and its hyperfine energy levels are separated without and with external magnetic fields [3]. For purpose of realizing EIT in three levels of nuclear spins in ^{167}Er, Baldit *et al.*, have obtained the hole burning spectrum from isotopically purified ^{167}Er (0.005 %) in Y_2SiO_5 single crystal [4,5]. Hashimoto *et al.*, on the other hand, have shown using coherent Raman beat measurements that the coherence time of nuclear spin (T_2) of ^{167}Er in Y_2SiO_5 (0.001 %) is 10 μs, and they have also predicted that T_2 can be increased up to 50 μs when all 0.001% Er ions are replaced with ^{167}Er [6]. To realize this prediction in the three-levels of the nuclear spins of ^{167}Er ions, we have to originally grow high-quality Y_2SiO_5 single crystals doped with isotopically purified ^{167}Er.

Here we report the crystal growth of only ^{167}Er doped-Y_2SiO_5 single crystal and its structural and optical characterizations. We found the crystal quality and optical quality are comparable to those of the commercially available non-isotope controlled Y_2SiO_5 single crystal.

2. Experiments

Single crystals of Er doped-Y_2SO_5 were grown by the Czochralski (Cz) method by the OXIDE Corporation. A commercially available isotope ^{167}Er metal (ATOX Corporation) was mixed into high-purity powders of SiO_2 and Y_2O_3 before the Cz growth so that the concentration of ^{167}Er in Y_2SiO_5 would be 0.001 %. The Cz growth was repeated several times to obtain a high-quality ingot. The ingot is 5 cm in diameter and 15 cm in length (**Fig. 1**). After the ingot had been cut into a cylindrical shape, XRD measurements were performed with monochromated Cu $K\alpha_1$ X-rays to find the best portions in it.

Figure 1: ^{167}Er doped-Y_2SiO_5 ingot grown by Cz method

We further cut the cylindrical ingot to obtain a single crystal for quantum optics experiments. The crystal is 5-mm square and 6-mm long on b-axis of Y_2SiO_5. The surfaces were polished and anti-reflection coating was applied. Raman spectroscopy was performed with a Renishaw system. Secondary ion mass spectroscopy (SIMS) was also performed. Photoluminescence (PL) and PL excitation (PLE) measurements were performed with high-resolution spectroscopy. Furthermore, we prepared a natural Er (^{167}Er: 23 %) doped-Y_2SiO_5 single crystal (0.001 %) grown by the Cz method in the Scientific Materials Corporation as a reference.

3. Results and discussion

Table 1 summarizes of the full width at half maximum (FWHM) of the (080) X-ray diffraction peaks in omega scans at $2\theta = 133.55^\circ$ from several areas of cylindrical ingot. The average FWHM is 0.0116° and the sharpest peak (FWHM=0.0107°) is obtained from the central area of the ingot. On the basis of this result, we cut out the single crystal from the central area of the ingot for quantum optics measurements.

Position of cylindrical ingot	FWHM (deg.)
Central area	0.0107
Side area 1	0.0108
Side area 2	0.0116
Side area 3	0.0108
Side area 4	0.0109

Table 1: Summary of peak widths of X-ray diffractions from different areas of the cylindrical ^{167}Er dpoped-Y_2SiO_5 ingot.

Figure 2 shows Raman spectra obtained from the ^{167}Er doped-Y_2SiO_5 single crystal (0.001 %) cut from the central area of the cylindrical ingot and from the Er doped-Y_2SiO_5 crystal as a reference. The Raman spectra almost match each other, indicating that the crystal quality of the ^{167}Er-doped Y_2SiO_5 single crystal is comparable to that of Er doped-Y_2SiO_5 single crystal.

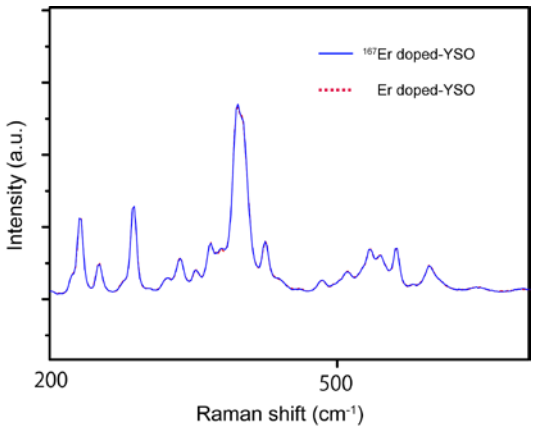

Figure 2: Raman spectra from ^{167}Er doped-Y_2SiO_5 (YSO) and Er doped-Y_2SiO_5 single crystals.

Figure 3 shows SIMS depth profiles of Er isotopes obtained from the ^{167}Er doped-Y_2SiO_5 single crystal. The profile of ^{167}Er is the most intense over the whole depth range, confirming that the majority of the isotope in the Y_2SiO_5 single crystal is ^{167}Er as designed, whereas it is the third in the natural abundance. Note here that natural abundances of Er are 0.14 % for ^{162}Er, 1.6% for ^{164}Er,

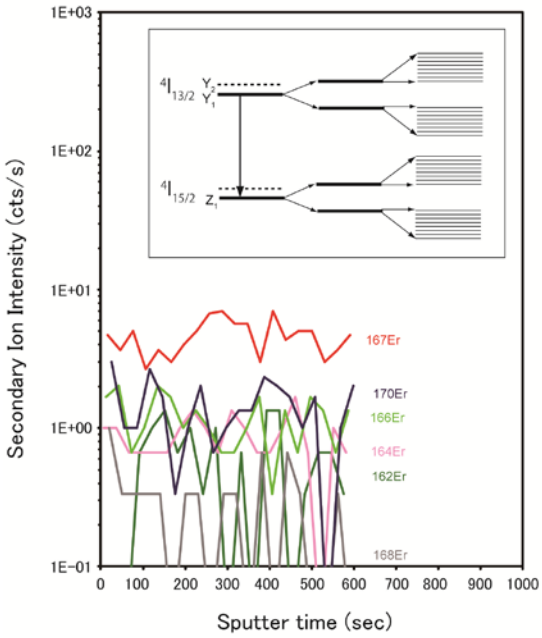

Figure 3: SIMS depth profiles from the ^{167}Er doped-Y_2SiO_5 single crystal. Inset shows the hyperfine structure of an individual Er^{3+} ion.

33.5 % for ^{166}Er, 22.9 % for ^{167}Er, 27 % for ^{168}Er, and 14.9 % for ^{170}Er.

Figure 4 shows PLE color mapping obtained from the ^{167}Er doped-Y_2SiO_5 (a) and Er doped-Y_2SiO_5 (b) at 4 K. All peaks can be clearly assigned to the optical transitions of Er^{3+} ions located at Y_1 and Y_2 sites of Y_2SiO_5 single crystal in both crystals. The PL peak width of ^{167}Er doped- Y_2SiO_5 is almost the same as that of Er doped-Y_2SiO_5. The energy levels of the optical transitions (Y_1-Z_1)**,** including hyperfine energy levels of Er^{3+} ions, are schematically shown in the inset of **Fig. 3**.

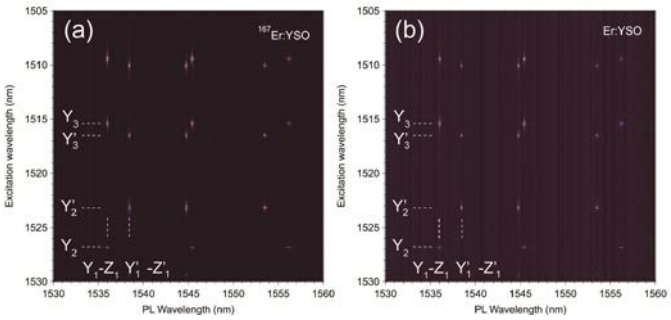

Figure 4: PLE color mapping from ^{167}Er doped-Y_2SiO_5 (a) and Er doped-Y_2SiO_5 (b) at 4 K.

4. Conclusion

Isotopically purified ^{167}Er doped-Y_2SiO_5 single crystals were grown by the Cz method. We confirmed that the quality of the originally made ^{167}Er doped-Y_2SiO_5 single crystals is comparable to that of commercially available Er doped-Y_2SiO_5 in structure and optical properties. We are planning to examine hole burning from the ^{167}Er doped-Y_2SiO_5 single crystal at low temperature.

Acknowledgement

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