Cooperative Down-Conversion Luminescence in Tb3+/Yb3+ Co-Doped LiYF4 Single Crystals (Postprint)

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Abstract

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Full Text

Preamble

Cooperative Down-Conversion Luminescence in Tb^3 /Yb^3 Co-Doped LiYF Single Crystals

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Abstract: Cooperative down-conversion (DC) with emission of two near-infrared photons for each blue photon absorbed was achieved in Tb^{3} /Yb^{3} co-doped yttrium lithium fluoride single crystals grown by an improved Bridgman method. The luminescent properties of the crystals were investigated through photoluminescence excitation, emission spectra, and decay curves. With excitation of Tb^{3} ions by 486 nm light, emission between 980 and 1030 nm from the Yb^{3} : ^{2}F / → ^{2}F / transition was observed, and this emission originated from DC between Tb^{3} and Yb^{3} ions. The energy transfer processes were studied based on the Inokuti-Hirayama model, and the interaction between Tb^{3} and Yb^{3} was confirmed to be electric dipole-dipole. The large quantum cutting efficiency approaches up to 166.7% for 0.32 mol% Tb^{3} and 7.98 mol% Yb^{3} co-doped LiYF , which is potentially useful as a DC layer in silicon-based solar cells.

Index Terms: Tb^{3} , Yb^{3} , LiYF single crystal, down-conversion, energy transfer, I-H model.

1. Introduction

Nowadays there is great interest in solar power due to its green, abundant, and renewable nature [1]. The capacity to convert sunlight into electricity via photovoltaic silicon solar cells is a major approach for humanity to use solar energy to solve current energy problems [2], [3]. However, the energy conversion rate of crystalline silicon solar cells in the market is mostly about 15%, which is far lower than the 29% predicted by classical theory. This difference is due to spectral mismatch that results in the major part of energy losses in solar cells. In order to increase the conversion efficiency of sunlight to electricity, two different approaches can be considered. One is using multi-junction tandem cell III-V solar cells [4]. Recent works on InGaAsN-containing quantum wells grown on GaAs have led to high-performance and very low threshold lasers [5]-[8], and these advances in dilute-nitride materials have led to promising results with more than 43.5% solar conversion efficiency [9]. The other approach is using spectral modification [2], [10]. The main methods of spectral modification include up-conversion, photoluminescence, and down-conversion (DC) [11].

[Figure 1: see original paper] Tb^{3} /Yb^{3} co-doped LiYF single crystal. (a) As-grown crystal. (b) Polished crystal.

Recently, cooperative down-conversion has attracted much more attention for its potential applications in solar cells, plasma displays, and other fields. This method is based on the principle that two near-infrared (NIR) photons are generated when each incident high-energy ultraviolet (UV) or visible (VIS) photon is absorbed. Through such spectral modification, the energy conversion rate of crystalline silicon solar cells can be enhanced significantly because the most effective absorption band of crystalline Si for sunlight is around the near-infrared wavelength of 1000 nm.

DC has been realized from Tb^{3} /Yb^{3} [12]-[15], [21], Pr^{3} /Yb^{3} [15], [16], Tm^{3} /Yb^{3} [15], [17], Ce^{3} /Yb^{3} [18], [19], and Nd^{3} /Yb^{3} [20] couples co-doped in micro-crystals or glasses for converting visible photons into near-infrared [12]-[20]. In previous investigations, most reported materials are in the forms of powders [12], [15], [18], [21] and glasses [13], [14], [16], [17], [19], [20]. Generally, powders have strong scattering and low transmission for light [1], [22], and some glasses have suboptimal mechanical, thermal, and chemical properties, as well as low luminescent efficiency for rare earth ions located in the glass network [23], [24]. These drawbacks limit their effective application in solar cells and other optical devices.

In fact, many single crystals exhibit better properties in terms of high optical transmission and luminescent efficiency for solar cell applications [1]. However, there are few reports on rare earth doped single crystals for DC. LiYF crystal doped with rare-earth ions has been proven to be an efficient all-solid-state laser medium because of its good chemical durability and thermal stability, high optical transparency in a wide wavelength range from infrared to ultraviolet, and lower phonon energy. These characteristics also render LiYF single crystals extremely suitable for quantum cutting. More recently, we have presented experimental evidence for cooperative energy transfer (ET) from Pr^{3} to two Yb^{3} ions in Pr^{3} /Yb^{3} co-doped LiYF single crystals for potential application in solar cells [1].

In the Tb^{3} /Yb^{3} system, the ^{2}F / → ^{2}F / single transition of Yb^{3} is located at approximately half the energy of the Tb^{3} : D → F transition, and the emission of Yb^{3} ions by the ^{2}F / → ^{2}F / transition is located in the range between 950 and 1050 nm, which matches the energy gap [21]. DC has been experimentally realized in Tb^{3} /Yb^{3} co-doped Y Yb PO :Tb^{3} compounds [13], in which the theoretical maximum quantum efficiency can reach nearly 200% [15]. In this paper, we measured the fluorescence spectra of Tb^{3} /Yb^{3} co-doped LiYF single crystals prepared by a Bridgman method using a sealed platinum crucible in air atmosphere. The cooperative ET from Tb^{3} to Yb^{3} ions in Tb^{3} /Yb^{3} co-doped LiYF single crystals will be discussed.

2. Experimental Details

The Tb^{3} singly doped, Yb^{3} singly doped, and Tb^{3} /Yb^{3} doubly doped LiYF single crystals were grown using an improved Bridgman method. Y^{3} ions were substituted by Tb^{3} and Yb^{3} ions. To ensure crystallization of the desired phase, feed materials for crystal growth were prepared with a slight excess of LiF. The raw materials (LiF, YF , TbF , YbF powders) with 99.999% purity were weighed according to the formulas LiY Tb F , LiY Yb F , and LiY Tb Yb F , respectively. The detailed growth process and schematic of the apparatus used for the Bridgman method have been reported in our previous work [25].

The grown Tb^{3}/Yb^{3} co-doped LiYF single crystal with the 100 direction is shown in Fig. 1. It was light yellow and transparent. The color changed gradually along the growth direction. The top of the crystal is opaque with a segregation layer comprised of redundant LiF and other impurities. The crystal was cut into small pieces and well polished on both sides to about 2 mm thickness for optical measurements.

The X-ray diffraction (XRD) was measured using an XD-98X diffractometer (XD-3, Beijing). The absorption spectra were recorded with a Cary 500 UV/VIS/NIR spectrophotometer (Agilent Co., America). The excitation and emission spectra and decay curves were obtained with an FLSP 920 spectrometer (Edinburgh Co., England). The Tb^{3} and Yb^{3} concentrations in LiYF single crystals were measured by inductively coupled plasma atomic emission spectroscopy (ICP-AES, PerkinElmer Inc., Optima 3000). All measurements were performed at room temperature. Table 1 shows the concentrations of Tb^{3} and Yb^{3} in some samples.

Concentration of Tb^{3} and Yb^{3} ions in the crystals (mol%)

3. Results and Discussion

The X-ray diffraction pattern for Tb^{3}/Yb^{3} co-doped LiYF crystal is shown in Fig. 2(a). By comparing the peak positions with those in JCPD 77-8016 for LiYF displayed in Fig. 2(b), one can confirm that the crystal is of pure orthorhombic phase and the current doping levels do not cause any obvious peak shift or second phase. Similar XRD patterns obtained from different Tb^{3} and Yb^{3} doping concentrations verify that all samples have crystallized into the pure orthorhombic phase.

[Figure 2: see original paper] (a) XRD pattern of LiYF :Tb^{3}/Yb^{3}; (b) the standard line pattern of the orthorhombic phase LiYF (JCPD 77-0816).

The absorption spectra of samples a1, a2, and undoped LiYF crystals at room temperature in the region of 300–1100 nm are shown in Fig. 3. No visible absorption bands were observed in undoped LiYF crystal, and an obvious band at 980 nm attributed to Yb^{3} : ^{2}F / → ^{2}F / was observed in Yb^{3} singly doped crystal. However, a strong absorption band at 980 nm (Yb^{3} : ^{2}F / → ^{2}F /) and another at 486 nm (Tb^{3} : F → D) were observed in Tb^{3}/Yb^{3} co-doped crystal. This indicates that the crystal can be pumped efficiently by 486 nm light. Meanwhile, some bands centered at 340, 350, 365, 378, and 486 nm appeared, corresponding to the energy level transitions of Tb^{3} from F to L (j = 7, 8, and 9), D , and D , respectively.

[Figure 3: see original paper] Absorption spectra of the undoped (dash-dot line), Yb^{3} doped (dash line), and Tb^{3}/Yb^{3} co-doped LiYF crystals (solid line).

The excitation (a) and emission (b) spectra of samples a0, a1, and a2 are depicted in Fig. 4(a) and (b), respectively. The energy level diagram of Tb^3 and Yb^3 in LiYF crystals is also shown in Fig. 4(c). An intense excitation band at 486 nm can be observed by monitoring emissions from both the Tb^3 : D → F transition at 544 nm and the Yb^3 : ^{2}F / → ^{2}F / transition at 980 nm in Tb^3 /Yb^3 co-doped LiYF crystal (sample a2) in Fig. 4(a). However, the band at 486 nm can be observed only by monitoring the Tb^3 : D → F transition at 544 nm in Tb^3 singly doped crystal, and the absorption bands in Yb^3 singly doped crystal are very weak, as shown in Fig. 4(a).

Three emission peaks in the visible range at 544, 587, and 620 nm are observed in Tb^3 singly doped LiYF crystal (sample a0) in Fig. 4(b) under excitation at 486 nm. They correspond to the transitions of D → F (j = 5, 4, and 3) [12]-[15], [21] as shown in Fig. 4(c). Multiple sharp peaks around these emission bands can be observed. These sharp peaks may be attributed to energy splitting in the crystal. It can also be found from Fig. 4(b) that the 544, 587, and 620 nm emission intensities decrease when Yb^3 ions are co-doped into Tb^3 -LiYF crystal (sample a2), and an intense new emission peak around 1000 nm appears, which is attributed to transitions from the Stark level ^{2}F / multiplet of Yb^3 to the Stark level ^{2}F / multiplet. The emission around 1000 nm is very weak under excitation at 486 nm for Yb^3 singly doped LiYF (sample a1) in Fig. 4(b), and no absorption exists at 486 nm for Yb^3 singly doped LiYF in Fig. 3. These facts indicate that a possible energy transfer from Tb^3 to Yb^3 exists in Tb^3 /Yb^3 co-doped LiYF crystal.

[Figure 4: see original paper] (a) Excitation and (b) emission spectra of Tb^3 doped, Yb^3 doped, and Tb^3 /Yb^3 co-doped LiYF crystals. (c) Energy level diagram of Tb^3 and Yb^3 ions. The ET processes are indicated by dashed lines and excitation and emission transitions are indicated by solid lines.

The possible mechanisms of the ET processes between Tb^3 and Yb^3 ions have been reported by Vergeer et al. [13]. In this paper, we attempt to quantitatively investigate the energy transfers between Tb^3 and Yb^3 . It can be seen from the energy level diagram in Fig. 4(c) that the D level of Tb^3 is located just above twice the energy of the ^{2}F / level of Yb^3 . One Tb^3 ion transfers its energy to two Yb^3 ions or radiates to lower energy states associated with emissions at 544, 587, and 620 nm, which are assigned to transitions from D to F (j = 5, 4, and 3) levels [12]-[15], [21], respectively, under irradiation at 486 nm. However, resonant ET from the Tb^3 : D state to one Yb^3 ion is not possible because Yb^3 has only one excited state around 1000 nm and no transition from Tb^3 : D is suitable to match the gap of Yb^3 . The mismatch between the D state of Tb^3 and the ^{2}F / state of Yb^3 is about 10,372 cm ^{1}, and the phonon energy in LiYF crystal is about 425 cm ^{1} from our measured Raman spectrum in Tb^3 /Yb^3 co-doped LiYF crystal, which almost excludes the phonon-assisted ET process. Therefore, it is reasonable to propose a cooperative DC process from one Tb^3 ion to two Yb^3 ions to explain this ET process, as shown in Fig. 4(c). After excitation of the Tb^3 : D level by 486 nm light, the de-excitation pathway of the ET process is the cooperative down-conversion mechanism: D (Tb^{3}) → ^{2}F / (Yb^{3}) + ^{2}F / (Yb^{3}). Since the energy of an absorbed visible photon is more than twice that of a NIR photon emission at 980 nm, it is theoretically possible to achieve two NIR emission photons for every incident visible photon with quantum efficiency up to 200%.

[Figure 5: see original paper] Visible-NIR emission spectra of various Tb^{3} /Yb^{3} co-doped LiYF crystals upon excitation at 486 nm.

Fig. 5 presents the photoluminescence spectra for x mol% Tb^{3} /y mol% Yb^{3} samples upon light excitation at 486 nm corresponding to the Tb^{3} : F → D transition, where x = 0.32, 0.32, 0.33, 0.32, and y = 0, 1.98, 4.97, and 7.98, respectively. The characteristics of emission bands are similar to those shown in Fig. 4(b). It can be seen from Fig. 5 that when the Tb^{3} concentration is held at about 0.32 mol%, the intensity of the green emission at 544 nm decreases and the NIR emission at 980–1030 nm increases monotonically as the Yb^{3} concentration increases from 0 to 7.98 mol%. The NIR emission band at 980–1030 nm reaches a maximum when the Yb^{3} concentration is 7.98 mol%. However, there is no emission in the NIR in the Tb^{3} singly doped LiYF sample.

[Figure 6: see original paper] Decay curves of the Tb^{3} /Yb^{3} ions co-doped LiYF crystal samples surveyed at 544 nm under 486 nm excitation. The black solid lines are the stretched exponential fits for the decays.

The decay curves of the Tb^{3} : D → F luminescence at 544 nm for Tb^{3} /Yb^{3} co-doped LiYF samples under 486 nm light excitation are shown in Fig. 6. For 0 mol% Yb^{3} concentration, the decay curve of the 0.32 mol% Tb^{3} singly doped LiYF sample is close to single exponential with a lifetime of 4.68 ms. When Yb^{3} ions are introduced, energy transfer between Tb^{3} and Yb^{3} occurs. The decay lifetime decreases rapidly with increasing Yb^{3} ion concentration from 0 mol% to 7.98 mol% in the co-doped samples, and the decay curves become non-exponential with the average lifetime decreasing from 4.68 ms to 1.56 ms. One can evaluate the average experimental lifetime for the non-exponential nature of the emission decay of these samples by the following equation [26]:

$$\tau_{avg} = \frac{\int I(t)t\,dt}{\int I(t)\,dt}$$

where $I(t)$ is the luminescence intensity as a function of time t. The results are listed in Table 2.

The stretched exponential function shown below best describes the decay curves in Fig. 6 [27]:

$$I(t) = I_0 \exp\left[-\left(\frac{t}{\tau}\right)^{\beta}\right]$$

where $0 < \beta \leq 1$.

Luminescence decay parameters, average decay lifetime, ETE and QE for x mol% Tb³ and y mol% Yb³ co-doped in LiYF samples

Here, $I(t)$ and I_0 represent the luminescence intensity during decay and at $t = 0$, respectively. The parameters τ and β depend on the material and other conditions such as temperature [28]. The parameter τ is a characteristic lifetime for the decay of Tb³ excited states, and the parameter β, a dispersion factor ranging from 0 to 1, represents the degree to which the measured decay differs from purely exponential decay. Fig. 6 displays the stretched exponential fit of the luminescence decay curves of various Tb³ /Yb³ doped samples. The obtained parameters τ and β are listed in Table 2. One can note that the parameter β was reduced with increasing Yb³ concentration when the concentration of Tb³ was held constant at 0.32 mol%. The lifetime τ clearly decreases with increasing Yb³ concentration in LiYF , indicating the existence of ET from Tb³ ions to Yb³ ions in the samples.

It is well known that the non-exponential fluorescent decay follows the Inokuti-Hirayama model [1], [17]. The fluorescent decay curves can then be expressed as:

$$I(t) = I(0) \exp\left[-\frac{t}{\tau_0} - \Gamma\left(1 - \frac{3}{s}\right)\frac{N}{N_0}\left(\frac{t}{\tau_0}\right)^{3/s}\right]$$

where $I(t)$ and $I(0)$ are the luminescent intensity during decay and at $t = 0$, respectively. Here, s is a parameter describing the distance dependence of the cross-relaxation process. The values $s = 6, 8$, and 10 denote electric dipole-dipole, dipole-quadrupole, and quadrupole-quadrupole interactions between luminescent centers, respectively. R_0 is the critical transfer distance, N is the concentration, τ_0 is the intrinsic radiation lifetime, and $\Gamma(1-(3/s))$ is a Gamma function. The fitted curves are shown in Fig. 7 as solid lines. The value of s derived from the fitting process is about 6.00. The results for τ_0 from the data fitting with $s = 6$ are also summarized in Table 2. This indicates an energy transfer of electric dipole-dipole interaction between Tb³ and Yb³ . The value of τ_0 decreases rapidly with increasing Yb³ concentration in LiYF , implying fast cross-relaxation from Tb³ to Yb³ since the interaction between Yb³ and Yb³ changes from long to short distance.

[Figure 7: see original paper] Decay curves of the Tb³ -Yb³ ions co-doped LiYF crystal samples surveyed at 544 nm under 486 nm excitation. The black solid lines are the I-H model fits for the decays.

The energy transfer efficiency (ETE) η_{ETE} and the total quantum efficiency (QE) η_{QE} can be determined from the luminescence decay curves. The energy transfer efficiency is defined as the ratio of Yb³ ions depopulated by ET to the total number of Tb^3 ions excited. η_{ETE} is obtained as a function of Yb^3 concentration as follows [1], [17]:

$$\eta_{ETE} = \eta_{x\%Yb} = 1 - \frac{\int I_{x\%Yb}\, dt}{\int I_{0\%Yb}\, dt}$$

where I stands for the intensity at 544 nm at time t and $x\%Yb$ represents the Yb^3 concentration.

Additionally, the relation between the energy transfer efficiency η_{ETE} and the quantum efficiency η_{QE} is linear and is defined as:

$$\eta_{QE} = \eta_{Tb}(1 - \eta_{ETE}) + 2\eta_{ETE}$$

where η_{Tb} is the quantum efficiency for the Tb^3 ions. Ignoring nonradiative energy loss by defects and impurities, the quantum efficiency for Tb^3 ions η_{Tb} is set to 1 [13], [21]. The values of η_{ETE} and η_{QE} are also summarized in Table 2.

We can confirm from Table 2 that when the concentration of Yb^3 is held at 1.97 mol%, the η_{ETE} reduces from 19.8% to 15.9% with increasing Tb^3 concentration from 0.32 mol% to 1.05 mol%. At the same 0.32 mol% Tb^3 concentration, the η_{ETE} increases monotonically from 19.8% to 66.7% with increasing Yb^3 concentration from 1.98 mol% to 7.98 mol%. The maximum quantum efficiency value is 166.7% for the 0.32 mol% Tb^3 /7.98 mol% Yb^3 co-doped LiYF sample.

4. Conclusion

In summary, LiYF samples doped with various Tb^3 and Yb^3 concentrations were synthesized by the Bridgman method. Absorption, excitation, and emission spectral studies proved the occurrence of cooperative ET from Tb^3 to Yb^3 ions. With excitation of Tb^3 ions by a blue photon at 486 nm, Yb^3 ions emit two NIR photons at 980 nm through cross-relaxation ET from Tb^3 to Yb^3, with a maximum QE as high as 166.7% for the LiYF :Tb^3 (0.32 mol%)/Yb^3 (7.98 mol%) sample. Because of the advantages of efficient NIR quantum cutting and good chemical and optical properties, these LiYF single crystals are promising materials for applications in solar cells.

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Note: Figure translations are in progress. See original paper for figures.