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Down-conversion near infrared emission in Pr³⁺, Yb³⁺ co-doped Y₂O₃ transparent ceramics

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ABSTRACT

Pr³⁺, Yb³⁺ co-doped Y₂O₃ transparent ceramics have been prepared by the solid state reaction and vacuum sintering. Down-conversion near infrared emission has been demonstrated upon a 482 nm excitation. The energy of the 482 nm blue photon was first absorbed by Pr³⁺ and then delivered to Yb³⁺. Possible energy transfer mechanisms from Pr³⁺ to Yb³⁺ have been discussed. Under the 482 nm excitation, the Pr⁴⁺–Yb²⁺ charge transfer state would not seriously influence the energy transfer process. The dominant one should be either the cooperative down-conversion or the two-step photon emission. The efficient down-conversion near infrared emission has potential application in enhancing the conversion efficiency of crystalline silicon solar cells.

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1. Introduction

Recently efficient down-conversion near-infrared (NIR) emission, that is, converting one UV or blue photon into two or more NIR photons was intensively studied. This efficient energy conversion process was also called “NIR quantum cutting” (NIR QC). Its promising potential application is to reduce the charge carrier thermalization in the present commercial crystalline silicon solar cells [1–4]. So far, the NIR QC process was achieved in different kinds of host materials (including powders [5–9], glasses [10,11], glass ceramics [12–17] and thin films [18]) by co-doping a donor ion like Tb³⁺, Tm³⁺, Pr³⁺, Eu²⁺ or Ce³⁺, and Yb³⁺ as the acceptor. Recently, the QC process in transparent ceramic materials has been reported in Refs. [19,20]. The advantages of transparent ceramic hosts lie in their high transparency and their good physical, chemical and ultraviolet radio-resistance stability. Among the different kinds of transparent ceramic materials, Y₂O₃ is a commonly investigated one. The benefits of the Y₂O₃ host are its high thermal conductivity for efficient thermal load removal, high transparency from ultraviolet to mid-infrared, good physical and chemical stability and its low phonon energy among the oxide materials. [21] So here in this paper we report the down-conversion NIR emission in Pr³⁺, Yb³⁺ co-doped Y₂O₃ transparent ceramics. The energy transfer mechanisms from Pr³⁺ to Yb³⁺ in the Y₂O₃ host under the 482 nm excitation were discussed.

2. Experimental

Firstly, Pr₆O₁₁ (99.999%), Yb₂O₃ (99.999%) and Y₂O₃ (99.999%) powders according to the designed (Pr_{0.01}Y_{0.99-x}Yb_x)₂O₃ (x=0, 0.02, 0.05 and 0.1; named as P0, P1, YP2 and P3, respectively) formula with 0.5wt% ethyl orthosilicate were mixed by ball milling in ethanol for 12 h. After drying, the powders were first uniaxially pressed into plates at 10 MPa and then cold isostatically pressed at 200 MPa. The plates were sintered at 1850 °C under a base pressure of $\sim 1.0 \times 10^{-3}$ Pa for 20 h. The as-sintered samples were annealed in the air at 1450 °C to eliminate the possible introduction of Yb²⁺ or O²⁻ vacancies during the vacuum sintering. Then every sample was double-side polished to 1 mm thickness (shown in Fig. 1). The optical transmittance and absorption spectra of the samples were measured on a V-570-type ultraviolet/visible/near-IR spectrophotometer (JASCO, Japan). JSM 6360-LA scanning electron microscopy (JEOL, Kyoto, Japan) was used to analyze the microstructure of ceramics. The excitation, emission spectra and the Pr³⁺ 511 nm emission decay curves were measured on an FLS920 fluorescence spectrometer (Edinburgh Instruments, Britain). All the characterizations were performed at room temperature.

3. Results and discussion

Fig. 1 presents a picture of the single Pr³⁺ doped, and the Pr³⁺, Yb³⁺ co-doped Y₂O₃ transparent ceramic samples after double-side mirror polishing. The characters behind the samples could be

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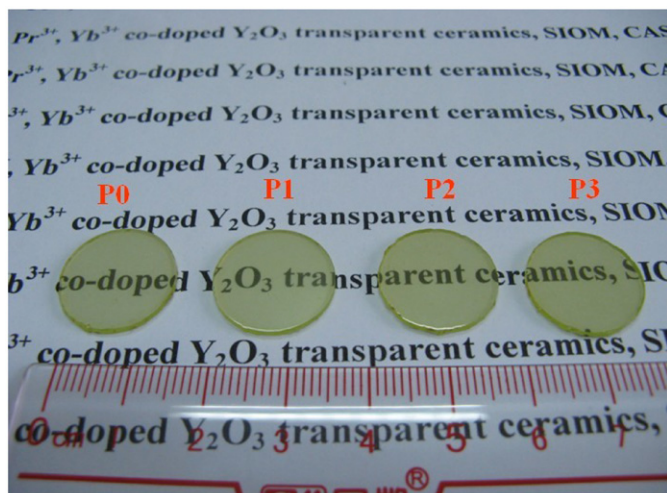


Fig. 1. Picture of the Pr^{3+} , Yb^{3+} co-doped Y_2O_3 ceramic samples.

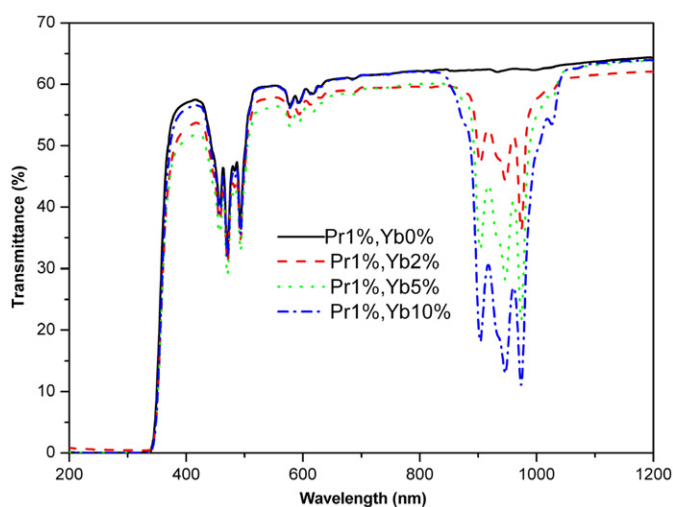


Fig. 2. Optical transmittance of the Pr^{3+} , Yb^{3+} co-doped Y_2O_3 transparent ceramics.

clearly seen. X-ray diffraction θ - 2θ scans (not given here) show that all the four samples were of the cubic Y_2O_3 structure (JCPDS41-1105).

Fig. 2 shows the transmittance of the four transparent ceramic samples. For P0 only the Pr^{3+} : $^3\text{H}_4 \rightarrow ^3\text{P}_2$, $^3\text{P}_1 + ^1\text{I}_6$ transitions could be seen at 457, 470 and 493 nm [14], while for P1–P3, besides the absorption of Pr^{3+} , the Yb^{3+} : $^2\text{F}_{7/2} \rightarrow ^2\text{F}_{5/2}$ transition between different Stark energy levels can be indentified around 1000 nm. The transmittance from 450 to 1200 nm for all the samples was $\geq 60\%$ except for the absorption bands. The transparency of the ceramic samples could be further enhanced by optimizing the preparation process, for instance, to improve the temperature schedule, to select a proper sintering aid, etc.

The microstructures of ceramic samples were investigated by scanning electron microscopy (SEM). The SEM morphology of P0 is presented in Fig. 3. Pores could be observed in the grains. The average grain size of the samples was $\sim 30 \mu\text{m}$.

Excitation spectra of P0 and P2 were carried out to demonstrate the $\text{Pr}^{3+} \rightarrow \text{Yb}^{3+}$ ET process (see Fig. 4(a)). With the Yb^{3+} 1030 nm emission monitored, the excitation band from 400 to 500 nm for P2 is in good agreement with that for P0 by monitoring the Pr^{3+} 511 nm emission.

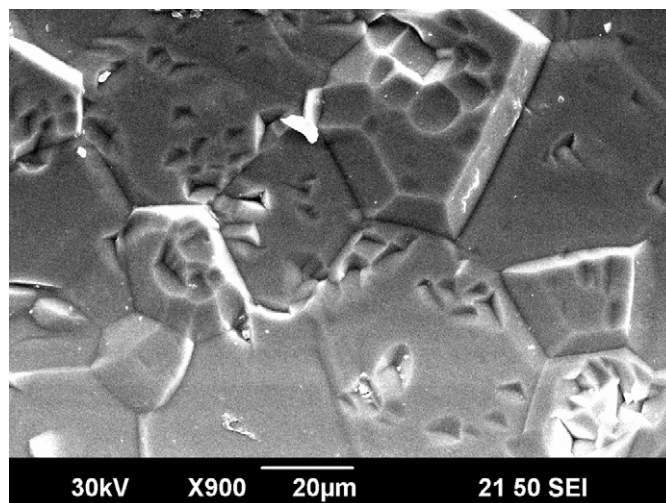


Fig. 3. SEM morphology of the microstructure of Sample P0.

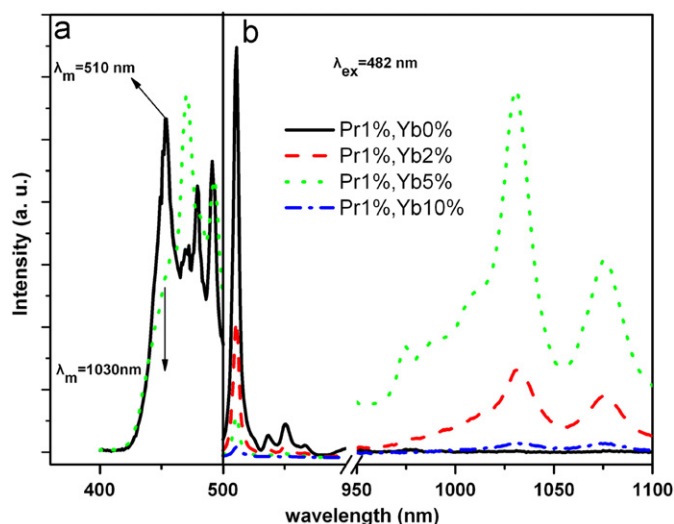


Fig. 4. (a) Solid line: excitation spectrum of Sample P0, $\lambda_{em}=511 \text{ nm}$; dotted line: excitation spectrum of Sample P2, $\lambda_{em}=1030 \text{ nm}$; (b) emission spectra of the four transparent ceramic samples, $\lambda_{ex}=482 \text{ nm}$.

In the emission spectra (Fig. 4(b)), for the Pr^{3+} single doped sample P0, under the 482 nm excitation, emission due to the Pr^{3+} : $^3\text{P}_0 \rightarrow ^3\text{H}_5$, $^3\text{H}_6$ and $^3\text{F}_{3,4}$ transitions could be observed. There was no emission detected in the 950–1100 nm range, while for P1, P2 and P3, besides the emission of Pr^{3+} , the Yb^{3+} emission around 950–1100 nm could also be observed. This clearly demonstrated the ET from Pr^{3+} to Yb^{3+} . The emission of Pr^{3+} turned weaker with higher Yb^{3+} concentration, while the Yb^{3+} emission intensity reached a maximum for P2. The weakest Yb^{3+} emission in P3 indicated that severe concentration quenching of Yb^{3+} occurred.

The decay curves of the Pr^{3+} : 511 nm emissions under the 482 nm excitation for all the samples exhibited a non-exponential characteristic, as shown in Fig. 5. The mean lifetime (τ_m) can be calculated by [13]

$$\tau_m = \int_{t_0}^{\infty} [I(t)/I_0] dt \quad (1)$$

Table 1 gives the lifetime of the Pr^{3+} : 511 nm emissions for P0–P3. The higher the Yb^{3+} doping concentration, the shorter the lifetime of Pr^{3+} : 511 nm emissions. Since the concentration of

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