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Temperature dependent emission cross-section and fluorescence lifetime of Cr,Yb:YAG crystals

Jun Dong^{a,*}, Peizhen Deng^b

^aSchool of Optics and Center for Research and Education in Optics and Lasers (CREOL), University of Central Florida, Orlando, FL 32816-2700, USA

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Abstract

The Cr,Yb:YAG single crystals with 10 at.% Yb and different Cr^{4+} concentrations were grown by the Czochralski method and their optical properties are presented. The effect of Cr^{4+} concentration and of the temperature on emission cross-section, and fluorescence lifetime of Cr,Yb:YAG crystals were presented. With the increase of Cr concentration in Cr,Yb:YAG crystals, the emission cross-section increases dramatically and fluorescence lifetime decreases under different temperature. With the decrease of temperature, not only the emission cross-section increases and lifetime decreases, but the line-shape of fluorescence spectra is changed. Also the potential of Cr,Yb:YAG as a self-Q-switched laser crystal was discussed. © 2003 Elsevier Science Ltd. All rights reserved.

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1. Introduction

In recent years, Cr^{4+} -doped crystals have attracted a great deal of attention as passive Q-switches [1–4]. These Cr^{4+} -doped crystals include Cr^{4+} :YAG [1,2], Cr^{4+} :GSGG [3], Cr^{4+} :YSO [4], etc. They have a large absorption cross-section and low saturable intensity at the laser wavelength. In comparison with previously used saturable absorbers such as dyes [5] and $LiF: F_2^-$ [6] color center crystals, Cr^{4+} -doped crystals are more photo-chemically and thermally stable and have a higher damage threshold. They can be used as Q-switches for both pulsed lasers [1,3,4] and continuously pumped lasers [2]. Moreover, Cr^{4+} :YAG can be co-doped with amplifying medium in a monolithic structure to form self-Q-switched laser [2]. As a result of the above mentioned advantages, Cr^{4+} -doped crystals become the most promising saturable absorbers for passively

Q-switched lasers of stability, low cost, reliability, long life, compactness, and simplicity.

Recent advances in high performance strained layer diode lasers with wavelengths between 0.9 and 1.1 µm have stimulated interest in diode pumped Yb3+ laser [7,8]. Furthermore, as there are no additional 4f energy levels in Yb³⁺ ions, like in other trivalent rare earths, complications in laser media which result from concentration quenching, up-conversion and excited state absorption are not anticipated to affect laser performance. At the same time, the broad emission band centered at 1.03 µm of Yb3+ couples well with the saturation absorption of Cr4+:YAG, and passively Q-switched Yb3+ lasers using Cr4+:YAG as saturable absorber have been demonstrated [9-11]. Self-Q-switched Cr,Yb:YAG laser has also been demonstrated [12]. Compared to Nd ions in laser crystals, the Yb ion matches diode pumping ideally since it has a very simple energy level scheme with desirable properties for a laser system. And Yb:YAG has a long storage lifetime (951 µs) [13] and a very low quantum defect (8.6%), resulting in three times less heat generation during lasing than comparable Nd-based laser systems [14]. In addition,

^bShanghai Institute of Optics and Fine mechanics, Chinese Academy of Sciences, Shanghai 201800, People's Republic of China

^{*} Corresponding author. Tel.: +1-407-823-5009; fax: +1-407-823-6880.

E-mail address: jundong_99@yahoo.com, jundong@mail.ucf. edu (J. Dong).

the absorption at 940 nm makes this material highly suitable for diode pumping using InGaAs diodes which are potentially more robust than AlGaAs diodes used to excite Nd:YAG at 808 nm [8]. Another advantage of using Yb:YAG is that the 940 nm absorption feature is approximately five times broader than the 808 nm absorption feature in Nd:YAG and therefore the Yb:YAG system is less sensitive to diode wavelength specifications [15]. So Cr–Yb-codoped Cr,Yb:YAG self-Q-switched laser crystal has the advantage over Cr,Nd:YAG, it will be a potential self-Q-switched laser material used for generating sub ns laser pulses.

In the present paper, we present the room temperature absorption spectra of Cr,Yb:YAG crystals with different Cr concentrations, and temperature dependent emission spectra and fluorescence lifetime. The variation of emission cross-section and fluorescence lifetime under different temperatures was presented. And the concentration quenching of Cr,Yb:YAG crystals under different temperature was observed with different Cr concentration in Cr,Yb:YAG crystals. The study of the concentration quenching in Cr,Yb:YAG crystals under different temperatures will have great benefit to the development of the high quality, suitable Cr/Yb ratio Cr,Yb:YAG self-Q-switched laser crystals.

2. Experiments

Yb:YAG crystal doped with 10 at.%Yb, Cr,Yb:YAG crystals co-doped with 10 at % Yb and 0.025 at.%Cr, 0.1 at. % Cr were grown by the Czochralski method. The detailed growth parameters are shown in Ref. [16]. Samples for spectroscopic and lifetime measurements were cut out of boules and the surfaces perpendicular to the $\langle 111 \rangle$ growth axis were polished. The thicknesses of the samples are 1 mm.

The absorption spectra were measured using a Cary 500 Scan UV-Vis-NIR spectrophotometer. Emission spectra were measured between 950 and 1100 nm. The excitation source was a diode laser operating at 943 nm. Yb³⁺ ions were pumped into their ${}^2F_{5/2}$ states and then relax nonradiatively to the ground ${}^2F_{7/2}$ state. The excitation signal was monitored during the experiment with a silicon (Si) detector. An indium gallium arsenide (InGaAs) detector located at the output slit of a 25 cm focal length Jarell-Ash monochromator was used to detect the fluorescence emission intensity. With 50 µm slits the resolution of this detection system was about 0.4 nm. Prior to use in calculating the stimulated emission cross-section, the recorded spectra were corrected for the spectral response of the detector and of the monochromator grating. The calibration of the detection system was achieved by recording its response to the light from a tungsten-iodine white light source that had been calibrated at the National Institute for Standards and Testing (NIST).

And the excitation of the Cr,Yb:YAG samples for the emission lifetimes measurements was at 941 nm by a tunable optical parametric oscillator (Quanta Ray MOPO-SL) which is pumped by the third harmonic of a Q-switched Nd:YAG laser. The OPO's linewidth amount to 0.2 cm^{-1} with a pulse duration of $\sim 5 \text{ ns}$. The energy of the idler pulse delivered by the OPO at 941 nm is less than 2 mJ. The fluorescence was collected with a f = 5 cm lens and dispersed with a 25 cm focal length Jarell-Ash monochromator. An InGaAs photodiode connected to a preamplifier was used to detect the fluorescence emission intensity. Decay curves were recorded using a Tektronix 2440 500 Ms/s digital oscilloscope with data acquisition on a computer. The measurements were carried out at temperature from 70 to 300. A temperature controlled compressed helium cryostat was used to cool samples to temperatures below room temperature.

The ytterbium emission from both YAG and ${\rm Cr}^{4+}$ co-doped YAG is not polarization dependent, so the emission spectra obtained could be used to calculate the effective stimulated emission cross-section of a Yb³⁺ ion from the manifold ${}^2F_{5/2} \rightarrow {}^2F_{7/2}$ transitions in the Yb:YAG and Cr-Yb-codoped Cr,Yb:YAG by applying the F-L formula. The fundamental relationship between spontaneous emission distribution

$$E_{(\lambda)} = \frac{I(\lambda)}{\int I(\lambda) d\lambda}$$

(integrating over all ${}^2F_{5/2} \rightarrow {}^2F_{7/2}$ transitions) and the stimulated emission cross-section distribution $\sigma_{\rm em}(\lambda)$ is

$$\sigma_{\rm em}(\lambda) = \frac{1}{8\pi} \cdot \frac{\lambda^5}{n^2 c \tau} \cdot \frac{I(\lambda)}{\int I(\lambda) \lambda d\lambda}$$
 (1)

where τ is the radiative lifetime of the upper laser level, c is the light velocity in vacuum, and n is the refractive index at the emission wavelength, the refractive index at 1030 nm is approximately 1.82. $I(\lambda)$ is the emission spectral intensity at different wavelength of Yb ions.

3. Results and discussion

The absorption spectra of Yb:YAG with 10 at.% Yb and Cr,Yb:YAG crystals with 10 at.% Yb and different Cr concentrations at room temperature (293 K) are shown in Fig. 1. Maximum absorption in the 900–1200 nm wavelength range is observed at $\lambda = 941$ nm where commercial laser diodes are available. With the increase of Cr concentration in Cr,Yb:YAG crystals, the absorption coefficient centered at 1.03 μ m increases from 1.87 cm⁻¹ for 10 at.% Yb:YAG to 4.26 cm⁻¹ for 0.1 at.% Cr and 10 at.% Yb-co-doped Cr,Yb:YAG. There is self-absorption at the lasing wavelength of 1.03 μ m for Yb:YAG crystal;

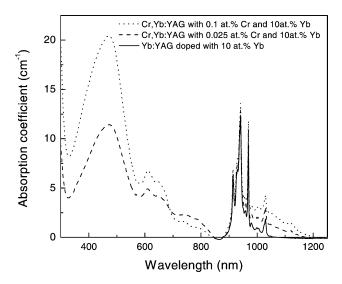


Fig. 1. The absorption spectra of Yb:YAG and Cr,Yb:YAG crystals with different Cr concentration.

the saturable absorption coefficient can be calculated by subtracting the absorption coefficient of 10 at.% Yb:YAG from the absorption coefficient of Cr,Yb:YAG at the lasing wavelength of 1.03 μ m. Thus the absorption coefficients at 1.03 μ m of Cr⁴⁺ in two different Cr,Yb:YAG crystal are 1.21 cm⁻¹ for 0.025 at.% Cr and 10 at.% Yb co-doped Cr,Yb:YAG crystal and 2.40 cm⁻¹ for 0.1 at.% Cr and 10 at.% Yb co-doped Cr,Yb:YAG crystal, respectively.

Fig. 2 shows the fluorescence spectra of Cr,Yb:YAG crystals with different Cr concentrations at room temperature (293 K) and 70 K. There is a broad fluorescence spectrum from 955 to 1080 nm centered at 1.03 and 1.05 μm in Cr,Yb:YAG at room temperature. The strongest emission peak is centered at 1.03 μm and the emission cross-section calculated by F–L formula is $4.52\times10^{-20}\,\mathrm{cm^2}$ for 0.025 at.% Cr and 10 at.% Yb codoped Cr,Yb:YAG and $8.12\times10^{-20}\,\mathrm{cm^2}$ for 0.1 at.% Cr and 10 at.% Yb:YAG, and the FWHM of the peak linewidth near 1.03 μm is about 9.7 nm. With the decrease in temperature, the emission spectra changes a lot, the main emission band centered near 1030 nm narrowed, is about only 1.5 nm, and the emission cross-section increases dramatically, as shown in Fig. 2(b).

The main disadvantage of Yb:YAG as a quasi-three system is re-absorption loss due to significant population of the lower laser level at room temperature. For such systems the effect of radiation trapping by single centers or pairs takes place and can lead to a considerable increase in the measured fluorescence lifetime [13]. This is very important because, according to the Fuchtbauer–Ladenburg formula Eq. (1), the effective emission cross-section value is inversely proportional to the radiative lifetime. In our fluorescence lifetime measurements special measurements were taken in order to eliminate this effect. The measured samples were 1 mm thickness plates with ground facets; and

aperture was placed at the Cr,Yb:YAG samples to limit transverse sample area seen by the detector. The fluorescence lifetime curves for all samples are nearly characterized by a single exponential decay behavior. Fig. 3 shows the relationship of lifetime of Cr,Yb:YAG crystals with temperature. The emission lifetime is 520 µs for low Cr concentration Cr, Yb: YAG and 280 µs for high Cr concentration Cr,Yb:YAG at room temperature. The lifetime is shortened to 190 and 90 µs for both samples at temperature of 70 K. With the decrease in temperature, the lifetime was shortened, there are three stages of the changing of lifetime with temperature, the lifetime decreases slowly with the decrease in temperature at the temperature range of 200-300 K, then lifetime decreases linearly with the decrease in temperature at the temperature range of 100-200 K, at the low temperature (below 100 K), the lifetime changes little and nearly keeps constant with the decrease in temperature. With the increase in Cr concentration in Cr,Yb:YAG crystals, at each temperature, the lifetime of high Cr concentration is shorter than that of low Cr concentration Cr, Yb: YAG. We fit the lifetime experimental data of certain Cr concentration Cr,Yb:YAG using formula of

$$\tau = \frac{A_1 - A_2}{1 + e^{(T - T_0)/dT}} + A_2,$$

where A_1 , A_2 , T_0 and dT are fitting parameters. The fitting results are shown in solid line for 0.025 at.% Cr and 10 at.% Yb co-doped Cr,Yb:YAG crystals and dotted line for 0.01 at.% Cr and 10 at.% Yb co-doped Cr,Yb:YAG crystals in Fig. 3.

With the measured lifetime of Cr,Yb:YAG crystals at different temperatures, we calculated the effective emission cross-section of Cr,Yb:YAG crystals using Fuchtbauer-Ladenburg formula Eq. (1), the relationship

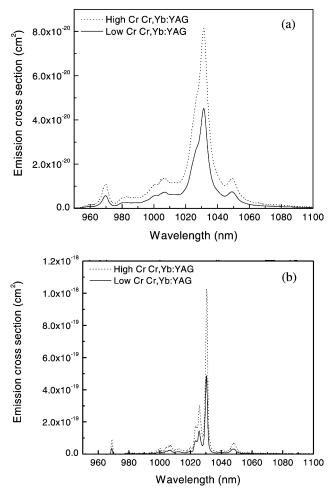
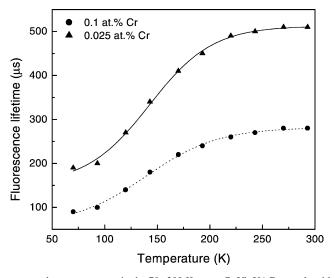


Fig. 2. Fluorescence spectra of Yb:YAG and Cr,Yb:YAG crystals with different Cr concentrations at room temperature (a) and 70 K (b).



 $Fig. \ 3. \ The \ emission \ lifetime \ measured \ at \ temperatures \ in \ the \ 70-300 \ K \ range \ Cr, Yb: YAG \ crystals \ with \ different \ Cr \ concentrations.$

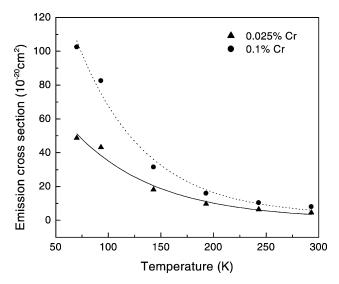


Fig. 4. Peak emission cross-section at 1.03 µm from Cr,Yb:YAG crystals with different Cr concentration as a function of temperature.

of effective emission cross-section of Cr,Yb:YAG crystals with temperature are shown in Fig. 4. The emission cross-section is $4.52\times 10^{-20}\,\mathrm{cm^2}$ for low Cr concentration Cr,Yb:YAG and $8.13\times 10^{-20}\,\mathrm{cm^2}$ for high Cr concentration Cr,Yb:YAG at room temperature, the emission cross-sections of both samples are higher than that of 10 at.% Yb:YAG crystal (usually $2.3\times 10^{-20}\,\mathrm{cm^2}$). The emission cross-section near 1030 nm increases dramatically at low temperature, and the emission cross-section reaches to $48.7\times 10^{-20}\,\mathrm{cm^2}$ for low Cr concentration Cr,Yb:YAG and $102.6\times 10^{-20}\,\mathrm{cm^2}$ for high Cr concentration Cr,Yb:YAG, even excess the emission cross-section of Nd:YAG (usually $2-7\times 10^{-19}\,\mathrm{cm^2}$). In the temperature range between 50 and 200 K, the emission cross-section decreases dramatically with the increase in temperature for both of Cr,Yb:YAG

crystals, when the temperature is above 200 K, the emission cross-section decreases slowly with the increase in temperature. The relationship of emission cross-section of Cr,Yb:YAG crystals with temperature is nearly exponential decay as shown in dotted line for 0.1 at.% Cr and 10 at.% Yb co-doped Cr,Yb:YAG and 0.025 at.% Cr and 10 at.% Yb co-doped Cr,Yb:YAG crystal in Fig. 4.

From measured lifetime τ and emission cross-section σ at 1030 nm at different temperature, we can calculate the $\sigma\tau$ product at the peak lasing wavelength at different temperatures. The measured $\sigma\tau$ product at peak lasing wavelength of the different Cr concentrations Cr,Yb:YAG crystals is shown in Fig. 5 as a function of temperature. From Fig. 5, we can see that $\sigma\tau$ product is nearly independent of Cr concentration at each temperature. We can also see that $\sigma\tau$

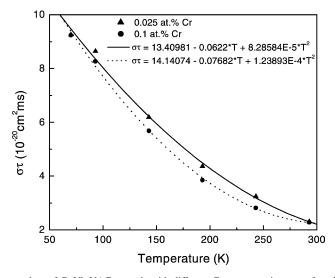


Fig. 5. Measured $\sigma\tau$ product of Cr,Yb:YAG crystals with different Cr concentrations as a function of temperature.

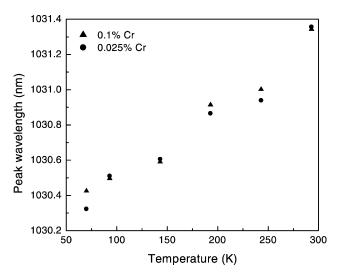


Fig. 6. Peak wavelength of Cr,Yb:YAG crystals with different Cr concentration as a function of temperature.

product increases as the temperature decreases from $2.3\times10^{-20}~\text{cm}^2~\text{ms}$ at room temperature (293 K) to $9.3\times10^{-20}~\text{cm}^2~\text{ms}$ at 70 K.

These $\sigma\tau$ product data of Cr,Yb:YAG crystals with different Cr concentrations benefit the design of thin disk high-power self-Q-switched lasers chosen to minimize thermal gradient induced stresses. Lasing properties are determined by the $\sigma\tau$ product and the ability to select high Yb concentration without sacrificing lasing performance may enable efficient absorption of pump power by thin disk Cr,Yb:YAG lasers. Higher concentration material has higher absorption and so pump light can be deposited in the two passes through a coated thin disk without using complex multiple reflecting mirrors. The most interesting thing is that the $\sigma\tau$ product is large at liquid nitrogen temperature, about four times of that at room temperature.

Thus, if the thin disk laser is operated at liquid nitrogen temperature, the threshold pump power will be significantly reduced and the saturation intensity lowered so that power can be extracted efficiently.

The temperature dependence of the peak emission wavelength and the peak linewidth (FWHM) near 1030 nm were shown in Figs. 6 and 7. For both samples, with the increase in temperature from 70 K to room temperature, the peak linewidth (FWHM) and the peak wavelength increases, the peak wavelength shifts to long wavelength.

For both the samples investigated, temperature has a great influence on the fluorescence spectral properties such as fluorescence spectral line shape, emission cross-section, peak wavelength, linewidth (FWHM) and fluorescence lifetime. Compared to the lifetime of Yb:YAG crystal

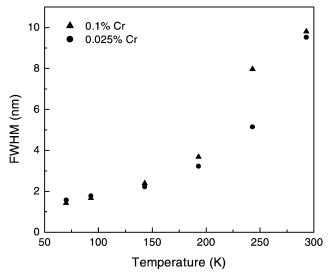


Fig. 7. The linewidth (FWHM) centered at peak wavelength of Cr, Yb: YAG crystals with different Cr concentration as a function of temperature.

(951 µs) at room temperature, introduction of Cr⁴⁺ into Yb:YAG crystal shortens the lifetime of Cr,Yb:YAG dramatically. We assume that the total decay rate (an inverse of the measured lifetime) consists of radiative decay rate w_r and nonradiative decay rate w_{nr} (first of all due to the multiphonon relaxation), and contains also the ion-ion interaction decay rate $(w_{tot} = w_r + w_{nr} + w_{int})$. Owing to the nature of quasi three-level laser, the difficulty in the estimation of actual decay rates is the influence of the radiation trapping effect on measured lifetimes of emission. It is a common effect [13] that when investigation a transition from a metastable state to a ground state, spontaneously emitted photons are reabsorbed by neighboring ions in the ground state. Repetition of this process over the volume of the sample brings about an increase in the measured emission lifetime relative to the actual one of the 'isolated' ions. But fortunately, there are no obvious radiation trapping effect in Cr,Yb:YAG owing to the effect of Cr concentration. The ${}^2F_{5/2}$ emission lifetime for both Cr,Yb:YAG samples is practically independent of the temperature up to 100 K. The increase in the lifetime values at higher temperature is most probably due to the rise of reabsorption of emission, as pertinent absorption and emission spectra overlap to a greater extent at higher temperatures. The ${}^2F_{5/2}$ emission lifetime of a more Cr concentrated Cr,Yb:YAG sample is significantly shorter at every temperature.

The energy transfer efficiencies of co-doped solid-state laser gain medium were calculated by equation [17]

$$\eta = 1 - \tau/\tau_0 \tag{2}$$

where τ is the Yb³⁺ donor's lifetime for Yb³⁺ and Cr⁴⁺ co-doped Cr,Yb:YAG, and τ_0 is the lifetime of Yb³⁺ in Yb:YAG crystal.

From Fig. 3, we can see that the fluorescence lifetime reduces dramatically with the increase in the concentration

of Cr4+ in Cr,Yb:YAG crystals at every temperature, the energy transfer efficiencies from Yb3+ to Cr4+ are very efficient, especially at low temperature. Usually, there are two ways to realize energy transfer in co-doped solid-state laser gain medium: direct energy transfer, which is a one-step process involving resonant energy transfer between donors and acceptors, and energy migration to acceptors. The characteristics of donor system can be discussed in terms of three limiting cases [18]: (1) direct relaxation—no diffusion, (2) fast diffusion and (3) diffusion-limited relaxation. When the acceptor concentration is low, as in Cr and Yb do-doped Cr, Yb: YAG crystal, only a small fraction of the total number of excited donors, Yb³⁺ are within the critical transfer distance of an acceptor, Cr⁴⁺. Therefore, the donor decay will be governed principally by intrinsic relaxation and by diffusion-limited relaxation to acceptors. The fluorescence lifetime of Yb³⁺ in Cr,Yb:YAG crystals is related to concentrations of Yb3+ and Cr⁴⁺ by equation [19]

$$\frac{1}{\tau} - \frac{1}{\tau_0} = K_{\rm D} N_{\rm Yb^{3+}} N_{\rm Cr^{4+}} \tag{3}$$

with

$$K_{\rm D} = \frac{1.48 \times 10^{-35} + 3.36 \times 10^{-36} e^{(T-115)/24}}{1 + e^{(T-115)/24}}$$
(4)

where τ is the fluorescence lifetime of Yb³⁺ in Cr,Yb:YAG crystal, τ_0 is the radiative lifetime of Yb³⁺ doped crystal without other impurity ions. T is the temperature of crystal, $N_{\text{Yb}^{3+}}$ and $N_{\text{Cr}^{4+}}$ are the concentrations of Yb³⁺ and Cr⁴⁺ ions for Cr–Yb co-doped Cr,Yb:YAG crystals (in ions/cm³).

Temperature and concentration dependent on the quenching rate $\tau^{-1} - \tau_0^{-1}$ mainly attributed to introduction of ${\rm Cr}^{4+}$ ions in Cr,Yb:YAG crystal as a function of temperature is plotted in Fig. 8. $K_{\rm D}$ is a temperature

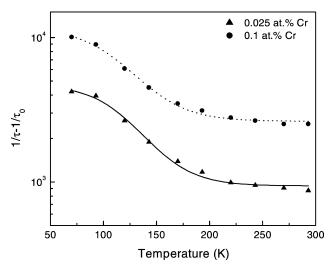


Fig. 8. Temperature dependent quenching rate $\tau^{-1} - \tau_0^{-1}$ as a function of temperature of Cr,Yb:YAG crystals.

dependent parameter, from Eq. (4), we can see that K_D becomes larger with the decrease of temperature, so from Eq. (3), we can see that the lifetime of Cr,Yb:YAG crystals at low temperature is shorter than that of higher temperature, which agrees with our temperature dependent lifetime measurements of Cr,Yb:YAG. Also from Eq. (3), we can see that the lifetime is shorter for high Cr concentration Cr,Yb:YAG than that of low Cr concentration Cr,Yb:YAG crystal, this is in good agreement with our experimental measured lifetime results (Fig. 8).

Other impurities such as Ho³⁺, Er³⁺ ions inevitably exist in Cr,Yb:YAG crystals. Since the integral spectra between Yb³⁺ emission and Ho³⁺ absorption or Er³⁺ absorption are very small, the energy transfer from Yb³⁺ ions to Ho³⁺ or Er³⁺ ions is accomplished with the photon assistance. The integral spectra between Yb³⁺ emission and Cr⁴⁺ absorption is very large, the energy can be transferred directly from Yb³⁺ ions to Cr⁴⁺ without the assistance of photon. Therefore, the influence of Cr⁴⁺ on the fluorescence lifetime of Cr,Yb:YAG is very significant.

4. Conclusions

The room temperature absorption spectra, the temperature dependent emission cross-sections and fluorescence lifetimes of Cr and Yb co-doped Cr,Yb:YAG crystals were measured, with the increase in the Cr concentration in Cr,Yb:YAG crystals, the emission crosssections of Cr,Yb:YAG increase and the fluorescence lifetimes decrease. The emission cross-section and emission lifetime of Cr,Yb:YAG crystals were influenced by the temperature greatly, the peak stimulated-emission cross-sections have been determined to be $8.13 \times 10^{-20} \,\mathrm{cm}^2$ for high Cr concentration Cr,Yb:YAG and $4.25 \times 10^{-20} \,\mathrm{cm}^2$ for low Cr concentration Cr,Yb:YAG at room temperature; the peak emission crosssection at 1030 nm increases dramatically at nitrogen temperature $(48.7 \times 10^{-20} \text{ cm}^2 \text{ for low Cr concentration Cr,Yb:YAG and } 102.6 \times 10^{-20} \text{ cm}^2 \text{ for high Cr concen$ tration Cr,Yb:YAG, are higher than that of Nd:YAG), although the emission lifetime is shortened. At the same time with the decrease in temperature, the peak wavelength shifts to short wavelength and also the linewidth center near 1030 nm is narrowed. The luminescence lifetimes of Cr,Yb:YAG crystals at room temperature are 280 µs for high Cr concentration Cr,Yb:YAG and 520 µs low Cr concentration Cr,Yb:YAG, respectively. With the introduction of Cr in Cr,Yb:YAG crystals, the energy transfer efficiency increases dramatically, especially at low temperature. These temperature dependent emission properties of Cr,Yb:YAG crystal will make it a more potential self-Q-switched laser material compared to Cr,Nd:YAG.

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