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Analysis and direct calculation of the energy migration interaction parameter in high concentration Nd^{3+} doped YAG laser crystal

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Abstract: The energy migration interaction parameter C_{DD} in high concentration Nd^{3+} doped YAG laser crystal was estimated using the Yokota-Tanimoto (Y-T) model and the spectral overlap model (SOM) of Kushida, respectively. Firstly, the experimental luminescence decay curves of $^4F_{3/2}$ state of Nd^{3+} ions in YAG laser crystal at room temperature for 2.0at.% and 3.0at.% Nd^{3+} concentrations reported by Mao were fitted successfully using the Y-T model, and the parameter C_{DD} was obtained to be 1.50×10^{-39} cm^6/s. Secondly, the parameter C_{DD} was directly calculated using the spectral overlap model (SOM) of Kushida; C_{DD} was calculated to be 2.73×10^{-39} cm^6/s. By comparing the energy migration interaction parameter C_{DD} with the energy transfer interaction parameter C_{DA} (1.794×10^{-40} cm^6/s), it was concluded that the energy migration rate between Nd^{3+} ions in YAG laser crystal was about 11 times larger than the energy transfer rate, and that energy migration played a very important role in high concentration Nd^{3+} -doped YAG laser crystal.

Keywords: energy transfer; energy migration; Y-T model; spectral overlap model; rare earths

Diode-pumped solid-state (DPSS) lasers have gained high prominence as compact high quality infra red sources. Neodymium-doped $Y_3Al_5O_{12}$ (Nd:YAG) has been the predominant solid-state laser material used, although in recent years, other materials such as Nd:YVO_4 and Nd:$GdVO_4$ have attracted considerable interest owing to their strong absorption for diode pump radiation at 808 nm and higher stimulated emission cross-section at 1064 nm. Nd:YAG displays better thermal and mechanical properties than Nd:YVO_4 and Nd:$GdVO_4$ and is therefore better suited for high power laser operation. It also exhibits a longer upper state lifetime, and hence higher energy storage capacity, making it suitable for high pulse energy using quasi-continous wave (QCW) diode-pumping and Q-switched operation[1].

A key difficulty for Nd:YAG is its low doping limitation. Standard Nd doping is typically 1at.% and such crystals can be grown with excellent optical quality. One of the best known growth techniques is the Czochralski (CZ) method, but until recently, it was not possible to produce Nd:YAG with doping concentration larger than 1.4at.%. In the recent years, several other techniques to grow Nd:YAG to higher concentrations have become available. The flux growth method can produce crystals with up to 4.3at.% doping, although the growth is very slow and produces only small samples[2,3]. Another technique to produce highly doped Nd:YAG in large samples is the temperature gradient technique (TGT), which has achieved doping concentration up to 3%[4–6]. Finally, polycrystalline ceramic Nd:YAG can be doped with up to 8.2at.%Nd concentration in large samples[7]. These recent results testify to the improvement in the efficiency of the DPSS Nd:YAG laser with the use of highly Nd doped crystals.

It is well known that the optimum doping level is in the vicinity of 1% replacement of Y with Nd in YAG crystal. However, recently, this doping limit was relatively irrelevant Sauder et al[1] and Mao et al[6] successfully obtained high efficient laser operation of 2at.% and 3at.% doped crystalline Nd:YAG. The existence of fluorescence quenching in Nd:YAG has been known for a long time, and the energy transfer micro-parameter C_{DA} was obtained to be 1.80×10^{-40} cm^6/s[8]. However, to the best of our knowledge, another important parameter-the energy migration parameter C_{DD} in Nd:YAG has not been obtained for lack of high- quality high concentration Nd^{3+} doped YAG single crystal. This article focuses on analyzing and calculating the energy migration interaction parameter C_{DD} between Nd^{3+} ions in highly doped YAG laser crystal, using Y-T model and SOM, respectively.

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1 Theory

It is well known that when the acceptor ion concentration is very low, the interaction between donors (D) and acceptors (A) may be neglected and the donor luminescence decay curve exhibits a single exponential behavior. As the acceptor concentration increases, the D–A energy transfer becomes important and the donor luminescence decay curve shows non-exponential behavior. By analyzing the donor luminescence decay curves, we studied the mechanism of the D–A energy transfer process and derived the energy transfer/migration rate. Here we only introduced the Y-T model and Kushida's SOM, which will be used in section 3.

1.1 Y-T model

When the donor concentration is increased, the D–D interaction becomes important and the energy migration among donors should be considered. In 1967, Yokota and Tanimoto presented a general solution for the donor luminescence decay function[9], including the D–D energy migration and the D–A energy transfer via dipole-dipole coupling, and treating the D–D energy migration as a diffusion process, as follows

$$I(t)=I(0)\exp\left[-\frac{t}{\tau}-\frac{4\pi^{3/2}R_0^3N_A}{3}\left(\frac{t}{\tau}\right)^{\frac{1}{2}}\left(\frac{1+10.87z+15.50z^2}{1+8.743z}\right)^{\frac{3}{4}}\right] \tag{1}$$

$$z=\frac{D\tau}{R_0^2}\left(\frac{t}{\tau}\right)^{\frac{2}{3}} \tag{2}$$

$$D=\frac{1}{2}\left(\frac{4\pi N_D}{3}\right)^{\frac{4}{3}}C_{DD} \tag{3}$$

where, τ is the donor intrinsic lifetime, N_A is the acceptor concentration in ions/cm^3, D is the diffusion coefficient, N_D is the donor concentration in ions/cm^3, C_{DD} is the D–D energy migration interaction parameter, which is connected with the energy migration rate P_{DD} by the following equation $C_{DD}=P_{DD}\cdot R^6_{DD}$, and C_{DA} is the D–A energy transfer interaction parameter, which is connected with the critical distance R_0 by $C_{DA}=R_0^6/\tau$.

1.2 Kushida's SOM

With the help of two approximations (the Judd-Ofelt closure approximation, and the Dexter's average of the $\left|\left\langle a_i b_j \left| H_{AB} \right| a_i' b_j' \right\rangle\right|^2$ overall possible orientations of $\overrightarrow{R_{AB}}$), Kushida obtained the average transfer rate of the dipole-dipole process as follows [10]:

$$\overline{P}_{AB}^{(ED\text{-}ED)}=\frac{1}{(2J_a+1)(2J_b+1)}\left(\frac{2}{3}\right)\left(\frac{2\pi}{\hbar}\right)\left(\frac{e^4}{R^6}\right)\times\left[\sum_\lambda \Omega_{A\lambda}\left|\left\langle J_a\left\|U^{(\lambda)}\right\|J_{a'}\right\rangle\right|^2\right]\left[\sum_\lambda \Omega_{B\lambda}\left|\left\langle J_b\left\|U^{(\lambda)}\right\|J_{b'}\right\rangle\right|^2\right]S_{AB} \tag{4}$$

where, λ=2, 4, 6, and Ω_λ indicates the phenomenological Judd-Ofelt intensity parameters, which depends on both the host lattice and the species of the rare earth ion. $\left|\left\langle J_a\left\|U^{(\lambda)}\right\|J_{a'}\right\rangle\right|^2$ is the reduced matrix of λ orders unit tensor operator $U^{(\lambda)}$. S_{AB} is the average overlap integral defined by $S_{AB}=\int G_{aa'}(E)G_{bb'}(E)\,\mathrm{d}E$, therefore, if

$\left[\sum_\lambda \Omega_{A\lambda}\langle J_a \| U^{(\lambda)} \| J_{a'}\rangle^2\right]$ and $\left[\sum_\lambda \Omega_{B\lambda}\langle J_b \| U^{(\lambda)} \| J_{b'}\rangle^2\right]$

are obtained, the average transfer rate of the dipole-dipole process can be estimated.

2 Results and discussion

In 2002, Mao et al[4] measured the emission decays of $^4F_{3/2}$ level of Nd^{3+} in highly doped YAG laser crystals grown by the temperature gradient technique (TGT) by pumping into the $^4G_{5/2}$ level at temperature ranging from 6 to 300K for 2.0at.% and 3.0at.% Nd^{3+} dopant concentrations. They found that there was strong concentration quench, which was owing to the cross-relaxation energy transfer $^4F_{3/2}$+ $^4I_{9/2}\rightarrow{}^4I_{15/2}$ + $^4I_{15/2}$ assisted by energy migration $^4F_{3/2}$ (Nd1)+ $^4I_{9/2}$ (Nd2)$\rightarrow{}^4I_{9/2}$ (Nd1)+$^4F_{3/2}$ (Nd2) (Fig.1).

The luminescence lifetime of isolated ions for $^4F_{3/2}$ level of Nd^{3+} in YAG laser crystal is 260 μs at 300 K[11] and can be considered as the intrinsic lifetime τ of $^4F_{3/2}$ level at room temperature. For 2.0at.% and 3.0at.% Nd^{3+} highly doped YAG laser crystals, the luminescence decay curves show a net departure from a single exponential decay at the initial short times[4]. We fitted these two luminescence decay curves by means of the Y-T model in order to obtain the pa-

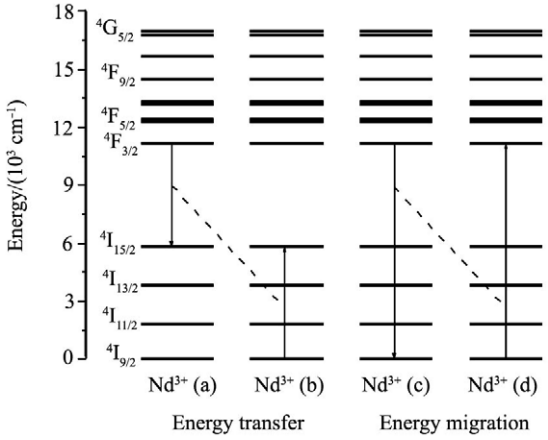

Fig.1 Cross-relaxation energy transfer and energy migration between Nd^{3+} ions in YAG laser crystal

rameters R_0 and the energy migration interaction parameter C_{DD}. The energy transfer/migration between Nd^{3+} ions is electronic dipole-electronic dipole (ED-ED) interaction[11]. Fig.2 presents the fitted decay curves by the Y-T model, and the parameters R_0 and C_{DD} are derived to be 0.5 nm and 1.50×10^{-39} cm^6/s, respectively. C_{DA} is calculated to be 1.794×10^{-40} cm^6/s by $C_{DA}=R_0^6/\tau$.

In addition, the energy migration interaction parameter C_{DD} was also directly calculated using the spectral overlap model (SOM) of Kushida. In the calculation, the Judd-Ofelt parameters Ω_2, Ω_4, and Ω_6 were 0.32×10^{-20}, 3.0×10^{-20}, and 5.16×10^{-20} cm^2, respectively[12], and the matrix elements $|\langle {}^4F_{3/2}\| U^{(\lambda)} \|{}^4I_{9/2}\rangle|^2$ were 0.0, 0.2283 and 0.0554 for λ=2, 4, 6, respectively[13]. The overlap integral of S_{DD} for energy migration between donors is obtained to be 1.09×10^{-3} cm by analyzing the absorption ($^4I_{9/2}\rightarrow{}^4F_{3/2}$) and emission ($^4F_{3/2}\rightarrow{}^4I_{9/2}$) spectrum (Fig.3)[14–16]. The value of C_{DD} is calculated to be 2.73×10^{-39} cm^6/s using Eq.(4).

It is obvious that the calculated value of C_{DD} using SOM of Kushida is in good agreement with that obtained by the Y-T model mentioned above. By comparing C_{DD} and C_{DA}, it is concluded that the energy migration rate between Nd^{3+} ions in YAG laser crystal is about 11 times larger than the energy transfer rate, and that energy migration plays a very important role in high concentration Nd^{3+}-doped YAG laser crystal.

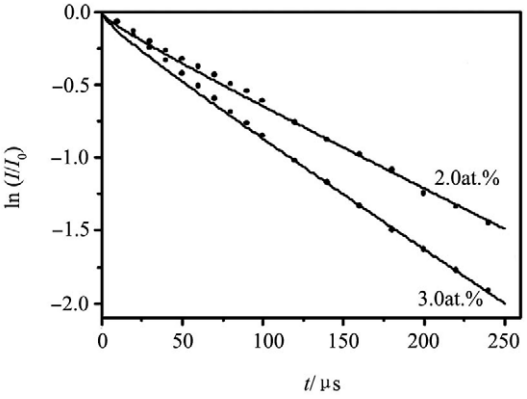

Fig.2 Luminescence decay curves of $^4F_{3/2}$ level of Nd^{3+} in 2.0at.% and 3.0at.% doped YAG laser crystal at room temperature (The dots show the experimental results; the solid lines are the best fits obtained by application of the Y-T model for electronic dipole-electronic dipole interactions)

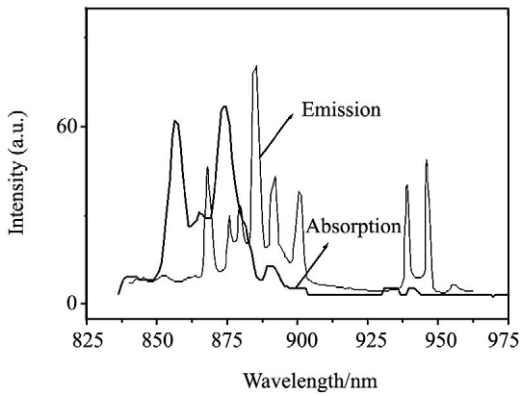

Fig.3 Absorption and emission spectra for $^4I_{9/2}\leftrightarrow{}^4F_{3/2}$ transition of Nd^{3+} in YAG laser crystal at room temperature

3 Conclusion

The energy migration interaction parameter C_{DD} in high concentration Nd^{3+} doped YAG laser crystal was analyzed and calculated by two means. One was to fit the experimental luminescence decay curves of $^4F_{3/2}$ state of Nd^{3+} ions in YAG laser crystal at room temperature for two different concentrations reported by Mao using the Y-T model. It was found that the donor-donor interaction parameter C_{DD} was 1.50×10^{-39} cm^6/s. The other was to directly calculate the energy migration interaction parameter by means of the spectra overlap model of Kushida; C_{DD} was calculated to be 2.73×10^{-39} cm^6/s, which is in good agreement with the Y-T model. All the analysis and calculation showed that the energy migration rate between Nd^{3+} ions in YAG laser crystal is about 11 times larger than the energy transfer rate, and that energy migration plays a very important role in high concentration Nd^{3+} -doped YAG laser crystal.

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