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Effects of rare-earth doping on thermal conductivity in $Y_3Al_5O_{12}$ crystals

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ABSTRACT

We have proposed a new model of thermal conductivity, which is applicable for both single-crystalline and poly-crystalline $Y_3Al_5O_{12}$. This model can explain the temperature dependence of thermal conductivity from room temperature to 200 °C. By comparing the measured value of thermal conductivity in Nd^{3+} and Yb^{3+}-doped $Y_3Al_5O_{12}$, we can involve effects of rare-earth concentration into our model. It has been confirmed that there is no difference in characteristics of thermal conduction between single- and poly-crystalline $Y_3Al_5O_{12}$. While the influence of grain-boundary in poly-crystals has been considered to be effective only in low temperature condition (below 100 K), we confirmed that the difference between thermal conductivity of single- and poly-crystalline $Y_3Al_5O_{12}$ remains several percent even at room temperature when the grain size is enough small. It has been demonstrated that our model is quite useful for thermal analysis of laser cavities in high-power density by using a single-crystalline and poly-crystalline $Y_3Al_5O_{12}$.

1. Introduction

Recent progress in the "Giant Micro Photonics" has enabled to extract an extremely high photon-energy emission from the microscopic volume [1], and an optical energy density of the extracted laser output from a laser material has increased every year. Ceramic lasers with poly-crystalline laser media are very useful for high-power lasers due to their toughness against thermal stress [2], and maximum laser energy density reached up to 200 kW/cm^3 by using a poly-crystalline composite device [3]. Although it would be significant to scale up the brightness of the laser output, severe thermal problems have prevented further power scaling of ceramic lasers. For example, temperature (T) of the laser crystal in high-power microchip lasers rose up to 200 °C due to an excessive heat from optical pumping [2]. Therefore it is important to include careful heat management for designing of high-power laser cavities. Considering influences of rare-earth (RE) doping should be also required for this thermal management, because solid-state laser media are often doped with some luminous ions. This is the reason why precise thermal conductivity (κ) dependence on both T and doping concentration (C) is desired. In order to treat various boundary conditions, the model that can give isopiestic specific heat for unit mass (C_P) and thermal diffusivity (D) separately is more useful. The temperature range should covers at least from room temperature (RT = 25 °C) to 200 °C especially for developing high power laser cavities.

At low temperature range (from several 10 K to 250 K) the model based on Boltzmann distribution of phonon gives adequate D [4]. It is also well known that there is empirical $1/T$-law above Debye temperature (Θ_D) [5] which is often used in analyses of thermophysical characteristics in minerals [6]. However there has been no enoughly useful model of D around RT to design laser cavities, and it is very difficult to measure precise D in heated materials because of the heat losses from the periphery of the sample above RT [7]. Attempts to obtain D around RT by extrapolation from high or low temperature have not given satisfactory results [8]. Although the expression that shows D in $Y_3Al_5O_{12}$ (YAG) around RT exists [9], it gives neither a correct D in RE-doped YAG or D in other materials.

Influences of RE-doping on D was already examined [10], and it is well known that reduction of D due to RE-doping can be estimated from irregularity of crystal density [11,12]. However, doping concentration of RE (C_{RE}) is more desirable for a design parameter of laser rather than the irregularity in crystal mass when we want to calculate κ in RE-doped materials. Moreover, these discussions are not enough quantitative for thermal design of laser cavities because they were based on conventional measurement of κ that contains large experimental errors. Therefore in order to discuss this influence due to RE-doping, the novel measurement that can detect a faint change in κ is necessary.

We developed quasi-one-dimensional flash method (Q1DFM) within temperature range from RT to 200 °C [13] aiming to enable a precise measurement of κ. We can detect a minute deviation of κ due to RE-doping that is not able to be measured by conventional method thanks to a small the reproducible error margin below 1%

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in Q1DFM. As a result Q1DFM made it clear that empirical $1/T$-law in D is not enough correct around RT (see Appendix), and enhanced the motivation to construct novel model of κ in laser media that can explain the dependence on T and C_{RE} precisely.

In this work we proposed a new model that can give temperature-dependent κ where one material parameter for the specific heat and two parameters for the thermal diffusivity are required. This model was proved experimentally by using of single-crystalline and poly-crystalline YAG. Dependences on rare-earth concentration in this model have been developed by comparing the measured value of κ in Nd^{3+}-doped YAG (Nd:YAG) and Yb^{3+}-doped YAG (Yb:YAG). The influence of grain-boundary was also discussed.

2. Experimental setup

Measurements of D were carried by Q1DFM (LFA 447 nanoflash, Netzsch) from 25 to 200 °C. Samples of Nd^{3+}- and Yb^{3+}-doped YAG single crystals with a diameter of 12.7 mm and a thickness of 1.0 mm were purchased from Scientific Materials Corp. (SMC). World Lab. Co., Ltd. (WLC), and Konoshima Chemicals Co., Ltd. (KCC), synthesized poly-crystalline Nd^{3+}- and Yb^{3+}-doped YAG ceramic samples. The mean grain size (g), diameter, and thickness of YAG ceramics from WLC were ca. 60 μm, 8.0 mm, and 1.0 mm, respectively, while YAG ceramics from KCC were ca. 3 μm, 12.7 mm, and 1.0 mm, respectively. Aperture sizes in Q1DFM were 4.5 mm for measurements with YAG ceramics from WLC, and 7.0 mm for other samples. Reproducibility of Q1DFM was below 1%.

In order to find out the influence of dilute doping of rare-earth trivalent on C_P, we measured C_P of undoped and Nd^{3+}-doped single-crystalline YAG relatively to sapphire standard within the range from 0 to 200 °C at a heating of 10 K/min by differential scanning calorimetry (DSC) (DSC 204 F1, Netzsch). Samples of single-crystalline YAG were grown by SMC and Shandong Newphotons Science and Technology Co., Ltd. (SNC), and were formed to have a size of 5.0 mm in diameter and 1.0 mm in thickness. We also evaluated C_P of poly-crystalline undoped YAG ceramics sintered by WLC and KCC with a view to check the influence of grain-boundary on C_P. Reproducibility of C_P measurement by DSC was below 2%.

Differences of C_P between undoped YAG specimens and samples heavily doped with Yb^{3+} were evaluated relatively from comparing of heat-up of specimens during the Q1DFM, where reproducibility of measured C_P was below 7%.

3. Results

Fig. 1 shows the measured κ in single- and poly-crystalline Nd:YAG by SMC and WLC. The decline of D seemed to be linear to concentration of Nd^{3+} (C_{Nd}) as −0.4%/at.% at least in the case that C_{Nd} was below 1.3at.%. Although H decreased nonlinearly to C_{Nd} in heavily doped condition, measured D in poly-crystalline Nd:YAG almost coincided to D in single crystals with same C_{Nd}. Measured κ in single- and poly-crystalline Yb:YAG by SMC and WLC are shown in Fig. 2. Similarly in the case of Nd^{3+}-doping, D depended nonlinearly on doping concentration of Yb^{3+} (C_{Yb}). The difference between D in poly-crystalline Yb:YAG fabricated by WLC and single crystals grown by SMC with same C_{Yb} was also negligible. The difference of D in YAG between single-crystals and poly-crystals can be observed when poly-crystals have smaller g, as shown in Fig. 3. D in poly-crystalline YAG sintered by KCC was ca. 5–7% smaller than single crystals.

Fig. 4 shows the C_P of undoped and Nd^{3+}-doped single-crystalline YAG, where there is no significant difference between these samples. On the other hand, heavy Yb^{3+}-doping into YAG caused to reduce C_P as shown in Fig. 5. As a result, κ in YbAG was almost the same as 5at.% Yb:YAG, while D in YbAG was the same as 10at.%.

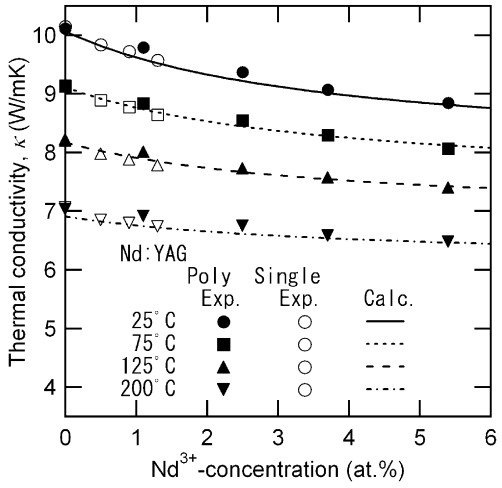

Fig. 1. Thermal conductivity in single-crystalline Nd:YAG with C_{Nd} from 0 to 1.3 at.%, and in poly-crystalline YAG with C_{Nd} from 0 to 5.4 at.% sintered by WLC. Lines are calculated value from Eq. (1) and Table 1. The difference between measured and simulated κ are within ±2%.

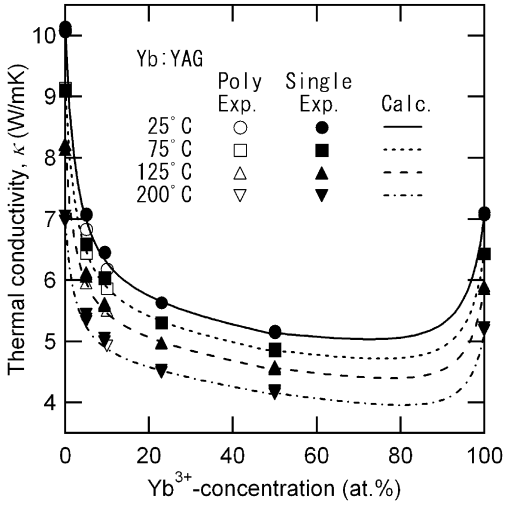

Fig. 2. Thermal conductivity in single-crystalline Yb:YAG with C_{Yb} from 0 to 100 at.%, and in poly-crystalline Yb:YAG with C_{Yb} from 0 to 10 at.% sintered by WLC. Lines are calculated value from Eq. (1) and Table 1. The difference between measured and simulated κ are within ±2%.

4. Discussions

We have introduced a numerical model on κ depending on T and a concentration of rare-earth trivalent (C_{RE}) as

$$\kappa(T, C_{RE}) = \rho C_P(T, C_{RE}) D(T, C_{RE}), \tag{1}$$

where ρ is density. C_P and D are given by

$$C_P(T, C_{RE}) = 3N_A k_B \frac{m}{M} f_D\left(\frac{T}{\Theta_D(C_{RE})}\right), \tag{2}$$

$$D(T, C_{RE}) = \frac{A(C_{RE})}{T} + B(C_{RE}). \tag{3}$$