Novel Model of Thermal Conductivity for Optical Materials

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We have proposed a novel model of thermal conductivity in various optical materials. This numerical model for thermal conductivity requires one material parameter for isovolumic specific heat and two parameters for thermal diffusivity in the calculation of each optical material. The validity of our model was demonstrated and it was adequately able to present thermal conductivities between 25°C and 200°C in $\text{Y}_3\text{Al}_5\text{O}_{12}$, YVO_4 , GdVO_4 , stoichiometric and congruent LiTaO_3 , and synthetic quartz.

Key Words: Laser materials, Thermal conductivity, Thermal diffusivity, Specific heat

1. Introduction

Recent progress in the “Giant Micro Photonics” enables an extremely high photon-energy emission from the microscopic volume.¹⁾ An optical energy density of extracted laser output from a laser material has increased every year, and microchip based active mirror laser allows efficient heat removing for the highly density laser operation up to 200 kW/cm³.²⁾ Although it would be significant to scale up the brightness of laser output, severe thermal problems prevent further power scaling of laser oscillators. In the case of a high power microchip laser reported in Ref. 2, temperature (T) of laser crystal rose up to 200°C due to an excessive heat from optical pumping. Therefore it is necessary for designing of high-power laser cavities to avoid thermal problems by careful heat treatment with precise thermal conductivity (κ). Dependence of κ on T from room temperature (RT, =25°C) to 200°C should be also included into this thermal management. Although models that can explain temperature dependence already exist,³⁾ a novel model that can give isopiestic specific heat for unit mass (C_p) and thermal diffusivity (H) separately is necessary for developing laser cavities that have various boundary condition of laser head.

There is no conventional expression of H under temperature range between RT and 200°C besides empirical $1/T$ -law above Debye temperature (Θ_D),⁴⁾ moreover it is very difficult to measure precise H in heated materials due to the heat losses from the periphery of the sample above RT.⁵⁾ We developed quasi-one-dimensional flash method (Q1DFM) within temperature range from RT to 200°C,⁶⁾ besides previous studies on κ in laser media have been mainly about low temperature thermal conduction.^{7,8)}

In this work we proposed a novel model of κ for optical materials, which can describe κ by three material parameters. We also showed correspondence between the measured value and modelled value of κ in optical materials that have cubic, tetragonal, rhombic, and amorphous structures.

2. Numerical Model

The value of κ is a product of isovolumic specific heat for unit volume (C_V), and H . While it is difficult to measure C_V directly, the experimental evaluation of C_p is quite easy by

differential scanning calorimetry (DSC). κ can be expressed by the product of density (ρ), C_p , and H , because C_V is approximated to be a product of ρ and C_p from Nernst-Lindemann’s law expressed by

$$C_p - \frac{C_V}{\rho} = \frac{T\alpha^2 E}{3\rho(1-2\nu)} \quad (1),$$

where α , E , and ν are thermal expansion coefficient, modulus of elasticity, and Poisson ratio, respectively.

From Debye model of phonon in dielectric materials, C_p can be expressed by molecular mass (M), ion number in a molecule (m), and Debye temperature (Θ_D) as

$$C_p = 3N_A k_B \frac{m}{M} f_D \left(\frac{T}{\Theta_D} \right) \quad (2),$$

where T , N_A , and k_B are material temperature, Avogadro’s constant, and Boltzmann’s constant, respectively. Here $f_D(x)$ is Debye function defined by

$$f_D(x) = 3x^3 \int_0^{\frac{1}{x}} \frac{z^3 e^z}{(e^z - 1)^2} dz \quad (3).$$

In the case of H there are many reports on the numerical model, and unfortunately they are useful only in limited conditions. The model where H depends on Plank’s distribution⁸⁾ is applicable only in cryogenic conditions, and the model where H inversely proportional to T ⁴⁾ is useful at higher than Θ_D . In order to explain the geophysical issues, more detailed model containing three thermal parameters was given by⁹⁾

$$H(T) = [H(T_0) - H_H] \left(\frac{T_0}{T} \right)^r + H_H \quad (4),$$

where T_0 , H_H , and r are a representative temperature, a high temperature limit of H , and a constant depends on T_0 and phonon kinetics. However it has been difficult not only to define r and but also to confirm its validity. Because phonon

Table.1 Crystal parameters of YAG, SLT, CLT, YVO₄, GdVO₄, and S-SiO₂.

	ρ (g/cm ³)	m	M
Y ₃ Al ₅ O ₁₂	4.55	20	593.613
YVO ₄	4.22	6	203.843
GdVO ₄	5.47	6	272.188
SLT	7.44	5	236.059
CLT	7.46	5	236.059
S-SiO ₂	2.22	3	60.083

Table.2 Thermal parameters of YAG, SLT, CLT, YVO₄, GdVO₄, and S-SiO₂.

	Θ_D (K)	A (mm ² K/s)	B (mm ² /s)
Y ₃ Al ₅ O ₁₂	795	1309.65	-0.700365
YVO ₄ a-axis	718	1255.98	-0.46298
YVO ₄ c-axis	718	1670.15	-0.455651
GdVO ₄ a-axis	677	1261.40	-0.525256
GdVO ₄ c-axis	677	1468.06	-0.466545
SLT c-axis	750	929.534	-0.511528
CLT c-axis	750	349.311	0.225441
S-SiO ₂	990	55.7963	0.597186

kinetics are dependent not only on crystal structures but also on crystal qualities, r is a peculiar value in each sample. Moreover an experimental estimation of r requires precise H under wide temperature range from RT to quite high temperature near H_H (generally over 1000°C)

In order to obtain a convenient and generalized model for analysis thermal condition of optical materials, we have tried to develop a novel model for H by both fixing r to 1 for all materials and limiting the applicable temperature range. In this assumption H is given by

$$H(T) = \frac{A}{T} + B \quad (5)$$

where A and B are fitting parameters. Equation (5) is the same as the first two terms of eq.(4) expanded by Taylor series. This means that assuming r to be 1 is the reason why applicable temperature range in eq.(5) is limited between RT to 200°C. B is a time-independent component of H , and sometimes B become negative as a result of the nonlinear temperature dependence of H . Finally, κ can be given by

$$\kappa(T) = 3\rho N_A k_B \frac{m}{M} \left(\frac{A}{T} + B \right) f_D \left(\frac{T}{\Theta_D} \right) \quad (6)$$

There are only three material parameters of A , B , and Θ_D in eq.(6) except what are determined from the crystal structure.

3. Experimental Setup

C_p and H was measured with following samples. (i) Cubic material of Y₃Al₅O₁₂ (YAG, synthesized by Scientific Materials). (ii) Tetragonal materials of YVO₄ (ITI Electro-Optics) and GdVO₄ (Shandong Newphotonics). (iii)

Fig.1 Measured C_p of various optical materials (marker) and simulations by eq.(2) (line). Θ_D are shown in Table 1, 2.

Fig.2 Measured H of various optical materials (marker) and simulations by eq.(5) (line). Fitting parameters are shown in Table 1, 2.

Rhombic materials of stoichiometric LiTaO₃ (SLT, OXIDE), and congruent LiTaO₃ (CLT, Yamaju ceramics). (iv) Amorphous of synthetic quartz (S-SiO₂). All crystal parameters are summarized in Table 1.

We measured C_p of YAG, SLT, CLT, YVO₄, and GdVO₄ relatively to sapphire standard within the range from 0 to 300°C at a heating of 10K/min by DSC (DSC 204 F1, Netzsch). The sample size is 5.0 mm in diameter and 1.0 mm

in thickness. C_p of S-SiO₂ with the size of 12.7-mm in diameter and 1.0-mm thickness was evaluated from 25 to 200°C relatively to YAG by flash method (LFA 447 nanoflash, Netzsch). Error under C_p measurement was below 2% except by flash method of 7% from SI-trace.

The measurement of H was carried out by the Q1DFM from 25 to 200°C with samples of YAG, SLT, CLT, YVO₄, GdVO₄, and S-SiO₂ by flash method (LFA 447 nanoflash, Netzsch). The sample size of cubic, rhombic, tetragonal, and amorphous materials were Ø-12.7 mm×t-1.0mm, 10×10×1mm³, Ø-8.0 mm×t-1.0mm, and Ø-12.7 mm×t-1.0mm, respectively. Reproducibility of Q1DFM was below 1%.

4. Results

Figures 1 and 2 show the result of the measured C_p and H , respectively. Θ_D of each materials were calculated from C_p at 25°C using eq.(2) except S-SiO₂ at 125°C, and parameters of A and B were determined by minimum square fitting by eq. (5). All fitting parameters shown in Table 2 can reproduce measured C_p and H with the error below 0.5% except C_p of S-SiO₂. The reason why an error in C_p measurement of S-SiO₂ reached 5% is due to influences of its inhomogeneous structure.

5. Discussions

The model proposed in this work is applicable for only dielectric materials because eq.(2) is based on an assumption where main heat carrier is limited to only lattice phonons. Moreover eq.(2) is not appropriate for opaque materials. Since radiated infra-red photons from a measured sample under high temperature conditions is self-absorbed, photon in opaque samples can be one of heat carriers.

Figure 3 shows the reported value of H in YAG under various temperature range.⁶⁻⁸⁾ Only under cryogenic conditions H was well fitted by the model given by⁸⁾

$$H(T) = H\left(\frac{\Theta_D}{b}\right) \frac{\exp\left(\frac{\Theta_D}{bT}\right) - 1}{e - 1} \quad (7),$$

where b is a fitting parameter. High-order phonon-phonon interaction occurs under higher temperature than RT, therefore b become larger than that of under cryogenic condition. It is thought that an increase of energy dissipation due to high-order phonon-phonon interaction causes temperature dependence of H in optical materials to the empirical $1/T$ -law at quite lower temperature that is far lower than Θ_D .

In order to confirm a validity of eq.(2), it is necessary to evaluate the right side of eq.(1) that is a difference between C_V and C_p . For example by using of the most common material YAG,¹⁰⁾ this difference was calculated as 1.1×10^{-3} J/gK. It is less than 0.2% of C_p . This indicates that at least from RT to 200°C we can consider ρC_p to be C_V .

Because differences between reported Θ_D were very large: in the case of YAG from 505K¹¹⁾ to 750K,¹²⁾ it is important that Θ_D should be determined from C_p measurement. On the contrary our estimated Θ_D of YAG was 795K that was larger than these reported value. Larger Θ_D gives smaller C_V , which becomes one circumstantial evidence of experiment under adiabatic condition.

Fig.3 H of YAG under various temperature range⁶⁻⁸⁾ (marker) and simulations by eq.(7) (line). Fitting parameter b was 2.14.

Fig.4 Measured κ of various optical materials (marker) and simulations by eq.(6) (line). Fitting parameters are shown in Table 1, 2.

It is enough for optical materials to consider thermal condition from RT to 200°C,²⁾ and according to our result this temperature relates a quite narrow range within $0.30\Theta_D$ and $0.70\Theta_D$, which guarantees the validity of eq.(5).

It was experimentally found that Θ_D of various materials are constant within the range of 25 to 200°C, and was also found that even in quite lower temperature than Θ_D eq.(5) gives adequate temperature dependence. The simulation of κ by eq.(6) can reproduce a good agreement with the

experimentally obtained value as shown in Fig. 4, which indicates that our model proposed in this work is quite useful for the optical design of photonic devices.

6. Conclusion

The novel model of a thermal conductivity for optical materials was proposed. We demonstrated that the model containing a few material parameters was adequately able to reproduce κ of various optical materials as YAG, SLT, CLT, YVO₄, GdVO₄, and S-SiO₂ within a temperature range between 25°C and 200°C. This numerical model is quite useful for not only thermal analysis in laser cavities or optical waveguides but also the evaluation of physical properties in various transparent materials.

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