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 Y₃Al₅O₁₂, YVO₄, GdVO₄, stoichiometric and congruent LiTaO₃, 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. 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. 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 and modelled value of thermal conductivity (κ) on T.

In this work we proposed a novel model of thermal conductivity where it is very difficult to measure H in heated materials due to the heat losses from the periphery of the sample above RT. We developed quasi-one-dimensional flash method (Q1DFM) within temperature range from RT to 200°C besides previous studies on κ in laser media have been mainly about low temperature thermal conduction.

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 (Cᵥ), and H. While it is difficult to measure Cᵥ directly, the experimental evaluation of Cᵥ is quite easy by differential scanning calorimetry (DSC). κ can be expressed by the product of density (ρ), Cᵥ, and H, because Cᵥ is approximated to be a product of ρ and Cᵥ from Nernst-Lindemann’s law expressed by

\[ Cᵥ \propto \frac{\rho}{3} \alpha T \sqrt{\frac{E}{3\rho(1-2\nu)}} \]  

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

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

\[ Cᵥ = 3Nₐkₐ \frac{m}{M} f_D \left( \frac{T}{\Theta_D} \right) \]  

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

\[ f_D(x) = \frac{9x^2}{2} \int_x^\infty \left( e^{-y} - 1 \right) dy \]  

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 distribution is applicable only in cryogenic conditions, and the model where H inversely proportional to T is useful at higher than ΘD. In order to explain the geophysical issues, more detailed model containing three thermal parameters was given by

\[ H(T) = H(T₀) - H(T) \left( \frac{T}{T₀} \right)^n + Hₙ \]  

where T₀, H₀, and r are a representative temperature, a high temperature limit of H, and a constant depends on T₀ and phonon kinetics. However it has been difficult not only to define r and but also to confirm its validity. Because phonon
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 \( H_D \) (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

\[
H(T) = \frac{A}{T} + B
\]

(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, \( \kappa \) can be given by

\[
\kappa(T) = 3\rho N\kappa_s \frac{m}{M} \left[ \frac{A}{T} + B \right] \left( \frac{T}{\Theta_D} \right)
\]

(6).

There are only three material parameters of \( A \), \( B \), and \( \Theta_D \) 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 \( Y_3Al_5O_{12} \) (YAG, synthesized by Scientific Materials). (ii) Tetragonal materials of \( YVO_4 \) (ITI Electro-Optics) and \( GdVO_4 \) (Shandong Newphotonics). (iii) Rhombic materials of stoichiometric \( LiTaO_3 \) (SLT, OXIDE), and congruent \( LiTaO_3 \) (CLT, Yamaju ceramics). (iv) Amorphous of synthetic quartz (S-SiO_2). All crystal parameters are summarized in Table 1.

We measured \( C_P \) of YAG, SLT, CLT, YVO_4, and GdVO_4 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-SiO$_2$ 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, YVO$_4$, GdVO$_4$, and S-SiO$_2$ 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, 10×10×1mm$^3$, 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. $\Theta_D$ of each materials were calculated from $C_p$ at 25°C using eq.(2) except S-SiO$_2$ 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-SiO$_2$. The reason why an error in $C_p$ measurement of S-SiO$_2$ 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 by

$$H(T) = \frac{\Theta_D}{b} \exp\left(\frac{\Theta_D}{b T}\right)^{-1} - \frac{1}{e - 1}$$

(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 $\Theta_D$.

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 $1.1\times10^{-3}$ J/gK. It is less than 0.2% of $C_p$. This indicates that at least from RT to 200°C we can consider $\rho C_p$ to be $C_V$.

Because differences between reported $\Theta_D$ were very large: in the case of YAG from 505K$^{11}$ to 750K,$^{12}$ it is important that $\Theta_D$ should be determined from $C_p$ measurement. On the contrary our estimated $\Theta_D$ of YAG was 795K that was larger than these reported value. Larger $\Theta_D$ gives smaller $C_V$, which becomes one circumstantial evidence of experiment under adiabatic condition.

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 $\Theta_D$ of various materials are constant within the range of 25 to 200°C, and was also found that even in quite lower temperature than $\Theta_D$ eq.(5) gives adequate temperature dependence. The simulation of $\kappa$ 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 $\kappa$ of various optical materials as YAG, SLT, CLT, YVO$_4$, GdVO$_4$, and S-SiO$_2$ 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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