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Thermal diffusivity measurement in a diamond anvil cell using a light pulse thermoreflectance technique

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Abstract
Apparatus to measure the thermal diffusivity of a plate between diamond anvils at high pressure was developed. The Mg(OH)2 plates coated with Pt films on both sides were used for testing the apparatus. One side of the Pt film played the role of an absorber of a laser pulse, and the other side was used for the detection of the temperature change. The sample was loaded into a hole drilled in a rhenium foil gasket together with the insulation layers of NaCl as a pressure medium. Subsequently, the sample was compressed with 150 μm culet beveled diamond anvils. The pressure in this study reached 57 GPa. Then, a front face of the sample was heated by laser pulses with a duration of 2 ns and a wavelength of 1064 nm. To probe the temperature change at the back face of the sample, a continuous wave laser beam with a wavelength of 782 nm was irradiated. Using the reflected intensity of the continuous wave laser beam, a transient temperature curve was measured when the heat transferred across Pt/Mg(OH)2/Pt layers. The thermal diffusivity of Mg(OH)2 was calculated by analyzing the transient temperature curves. The thermal diffusivity of Mg(OH)2 is 5.9 × 10−6 m2 s−1 at 57 GPa, which is larger than that at ambient pressure by a factor of ≈6.

Keywords: thermal diffusivity, diamond anvil, high pressure, thermoreflectance, pulse heating

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Thermal transport properties for the crust and mantle are of essential importance to reveal dynamics within the Earth. The pressure of the Earth’s interior drastically increases with the depth from the surface of the Earth: several hundred MPa at the region of the crust, ~20 GPa at the upper mantle and ~130 GPa at the lower mantle [1]. Precise data of thermal conductivity and/or thermal diffusivity for minerals at elevated pressure make it possible to estimate the heat budget in the Earth. Ross et al [2] have reviewed an abundance of thermal conductivity data in conjunction with pressure for metals, insulators and liquids. However, few measurements have been made for thermal conductivity over 10 GPa. In order to supplement the lack of experimental data at higher pressure, it is usual to use extrapolations and theoretical estimations [3–7] based on the lower pressure data. Recently, the high-spin to low-spin transition of iron [8, 9] has been discovered to occur in both perovskite and ferropericlase under the lower mantle conditions at 70 GPa and 120 GPa, respectively. Furthermore, the principal lower mantle phase of perovskite undergoes a phase transition to post-perovskite [10] in the lowermost mantle at 120 GPa. It is expected that discontinuity of physical properties [11–15] should arise from...
such transitions. Accordingly, experimental measurement techniques for thermal transport properties at higher pressure are crucial.

A diamond anvil cell (DAC) technique is required to obtain static pressure higher than 20 GPa. Using a time-resolved radiometry technique, Beck et al [16] have measured the temperature decay of the sample loaded into a DAC after flash-heating. They have analyzed the temperature decay at the heated surface of the sample and reported the thermal diffusivities of MgO, NaCl and KCl up to 32 GPa. They have shown the possibility of the in situ observation of thermal transport inside the DAC. However, there remain difficulties for quantitative measurement using DAC, associated with the small aperture of the anvil and small sample size. The former causes difficulty in gaining enough radiation from the aperture of the DAC. Regarding the latter, small sample size leads to an intensively shorter time-scale of heat diffusion. Therefore, a measurement technique designed for DAC must allow sufficient sensitivities of both time resolution and temperature change.

More recently, Hsieh et al [17] have measured the thermal conductivity of muscovite crystal up to 24 GPa using a time-domain thermoreflectance technique. They have loaded a muscovite sheet (20 μm thick) coated with an aluminum film (~80 nm thick) into a DAC. A laser pulse was irradiated on the aluminum film, and then the temperature decay of the heated area was observed using a probe laser pulse. Taketoshi et al [18–20] have developed a pulse heating thermoreflectance technique and measured the thermal diffusivity for metal thin films, where a transient temperature change below 1 K in the below/above ns time range can be observed. In this technique, a short light pulse heated the film surface and the temperature rise of the opposite surface was observed, which is similar to the configuration of the laser flash method [21]. In addition, the thermoreflectance techniques have also been applied to small size samples [22, 23].

Consequently, the primary objective of this study is to develop an instrument adapted to a DAC for measuring thermal diffusivity using a pulse heating thermoreflectance technique with a laser flash-like configuration. Furthermore, an analysis procedure for a transient temperature curve obtained by the developed instrument is proposed. To validate the measurement and analysis technique, we demonstrate the thermal diffusivity measurement for Mg(OH)2 over 50 GPa at ambient temperature.

2. Experimental details

2.1. Sample preparation

Mg(OH)2 powder was pressed to obtain thin plates. The Mg(OH)2 plate (1.5–2 μm in thickness) was carefully formed into a small round shape with a diameter of 50 μm. Each side of the surface of the Mg(OH)2 plate was coated with Pt film (0.5–0.8 μm in thickness) by means of a physical vapor deposition process. One side of the Pt surface plays the role of an absorber for a heating laser pulse, and the other side is used for the detection of temperature change. The coated plate was loaded into a hole drilled in a rhenium foil gasket together with insulation layers of NaCl as a pressure medium. After being assembled, the coated plate was compressed with 150 μm culet beveled diamond anvils. The schematic illustration of the assembled DAC is shown in figure 1.

2.2. Measurement apparatus

An apparatus using a light pulse heating thermoreflectance technique designed for the DAC was developed to measure the thermal diffusivity of the Mg(OH)2 plate. Figure 2 shows a block diagram of the apparatus. The pulse heating on the sample was carried out using a pulsed YAG laser with a wavelength of 1064 nm, pulse duration of 2 ns and pulse energy of 0.5 μJ. The heating laser beam was focused on one side of the Pt film surface using a ×15 cassegrain mirror with a spot diameter of 50 μm. A linearly polarized continuous-wave (CW) diode laser beam (782 nm in wavelength and 4.8 mW in averaged power in front of the DAC) was focused on the opposite side of the Pt film surface to measure the temperature rise using a ×10 microscope objective lens with a spot diameter of 50 μm. In order to introduce a probe laser beam into the diamond anvil, an anti-reflection coating was applied to the surface of the diamond anvil exposed to air. The coating reduces reflection at the air/diamond interface to 0.3% at the wavelength of the probe laser. The reflected probe laser beam was separated from the incident beam by a polarized beam splitter (PBS), and its intensity was detected by a high-speed photo detector with a bandwidth of 1.4 GHz. The typical power of the reflected probe beam in front of the detector was about 0.1 mW, which contains small amount of fluctuation because the reflectivity of the Pt film slightly changes due to its temperature change. The measured signal was averaged by an oscilloscope (500 MHz in bandwidth), and the transient temperature change of the sample by the pulse heating was recorded.

Two specimens (cells 1 and 2) were prepared for the thermal diffusivity measurement. All the measurements were carried out by means of a decompression process. After the measurements, the anvils were removed from the samples, and the thicknesses of the Mg(OH)2 plates and Pt films were measured. In addition, the thermal diffusivity of Mg(OH)2 at the ambient pressure was also measured by means of the
Figure 2. Schematic diagram of the pulse heating thermoreflectance apparatus for thermal diffusivity measurement at high pressure.

Figure 3. Scanning electron microscope image of the specimen after being removed from the diamond anvil.

3. Results

3.1. Specimen structure

Figure 3 shows a photograph of a scanning electron microscope (SEM) image of the sample after being removed from the anvil. The conical hollow is the cubic-BN gasket and the bottom of the hollow is the pressure medium (NaCl). In order to observe the cross-sectional layered structure of the specimen, a square hole was formed at the center of the hollow using a focused ion beam (FIB) method. Figure 4 shows the cross-sectional view of the sample. Two white lines are the Pt films, and the Mg(OH)$_2$ plate is sandwiched between those Pt films. Since the plate does not seem to be perfectly flat, averaged thicknesses were estimated from the cross-sectional observation. As a result, the thicknesses of the Mg(OH)$_2$ plate and the Pt films of cell 1 are 1.6 $\mu$m $\pm$ 0.22 $\mu$m and 0.85 $\mu$m $\pm$ 0.06 $\mu$m, respectively. Those of cell 2 are 2.3 $\mu$m $\pm$ 0.44 $\mu$m and 0.47 $\mu$m $\pm$ 0.08 $\mu$m, respectively.

3.2. The transient temperature curve of a coated Mg(OH)$_2$ plate

Figure 5 shows the transient temperature curves for the samples in the pressure range from 10 to 57 GPa along with the calculated thicknesses of Pt/Mg(OH)$_2$/Pt; the calculation is described in section 4.2. It can be clearly seen that the speed of the temperature rise becomes faster as the attained pressure increases. This indicates that the thermal diffusivity of the sample increases with increasing pressure. Also the thickness of the sample slightly changes due to pressure. However, it can be considered that this compressibility of the thickness is a minor effect on the transient temperature curves.

4. Discussions

4.1. Analyzing transient temperature curves

The transient temperature curves in figure 5 demonstrate heat transfer from the surface of the front Pt film, where the
Figure 4. Scanning electron microscope image of the cross section of the specimen.

Figure 5. Transient temperature curves for the coated Mg(OH)$_2$ plates after pulse heating. The attained pressures and calculated thicknesses of Pt/Mg(OH)$_2$/Pt at respective pressures are shown in the figure.

Figure 6. Schematic illustration of the areal heat diffusion time. The transient temperature curve, when the periphery of the sample is adiabatic, is normalized; the initial temperature is 0 and the maximum temperature rise is 1. Areal heat diffusion time is defined as the area represented by $A$, which is bordered by the maximum temperature rise and the normalized temperature curve.

Pulse heating was provided, to that of the back Pt film via the Mg(OH)$_2$ plate. The transient temperature curves were analyzed based on the three-layered model of Pt/Mg(OH)$_2$/Pt, because the thickness of the Pt films deposited on Mg(OH)$_2$ is not negligible in comparison with the total thickness of the sample. Baba [24] has reported the areal heat diffusion time (AHDT) method to analyze a transient temperature curve for the multi-layered system observed by a laser flash measurement. The definition of the AHDT is described below. The transient temperature curve for a layered sample is normalized as the initial temperature is 0 and the maximum temperature rise is 1. Then, the area bordered by a horizontal line at maximum temperature rise and the normalized transient temperature curve is derived as shown in figure 6. This area is determined by the thermophysical properties of the layers which heat passes through. The mathematical solution of the AHDT for the three-layer system has been described in [24], when the boundary condition at the periphery of the sample is adiabatic. With respect to the experimental condition in this study, the samples directly contacted the pressure medium (NaCl), and heat effusion from the sample to the NaCl cannot be ignored, as shown in the lower curve in figure 6. Accordingly, before applying the AHDT method, the effect of the heat effusion should be compensated. Thus, to apply the AHDT method of the three-layer model, we separately analyzed the transient temperature curves using the following steps.

1. An actual heat diffusion model of the sample can be described as a five-layered structure that consists of the NaCl/ Pt/Mg(OH)$_2$/Pt/NaCl layers as shown in figure 7(a). Here, a pulse heating was provided at the surface of the left-hand Pt layer. The temperature was measured at the surface of the right-hand Pt layer.

2. The part of Pt/Mg(OH)$_2$/Pt is supposed to be a homogeneous plate where the plate has the sum of the thicknesses of the Pt films and Mg(OH)$_2$. Thus, the five-layered model can be simplified as a three-layered model such as NaCl/plate/NaCl (figure 7(b)). Using an analytical solution of the three-layered model in figure 7(b), the heat diffusion time of that plate (Pt/Mg(OH)$_2$/Pt) was obtained.

3. Using the value of the heat diffusion time obtained at the second step, the value of the AHDT, when the periphery of the sample was adiabatic, was deduced. Then, the AHDT method was applied to the three-layered model of Pt/Mg(OH)$_2$/Pt (figure 7(c)) in order to obtain the thermal diffusivity of Mg(OH)$_2$.

4.1.1. An analytical solution of the NaCl/coated plate/NaCl model. First, the five-layer model (figure 7(a)) is simplified as the three-layered model of NaCl/plate/NaCl (layer 1)/plate (layer 2)/NaCl (layer 3) as shown in figure 7(b). Based on this model, an analytical solution is derived to decide the thermal diffusion time of the plate containing two Pt layers and a Mg(OH)$_2$ layer. Here, the following assumptions are considered:

1. layer 2 represents Pt/Mg(OH)$_2$/Pt;
2. the heating laser pulse is absorbed at $x = 0$;
3. heat diffusion parallel to each layer can be neglected;
4. layers 1 and 3 are semi-infinite;
5. boundary thermal resistances between layers are negligible.
Figure 7. The heat diffusion models of the sample, where $\alpha$, $b$, $d$, $\tau$, $k$ and $C$ represent the thermal diffusivity, thermal effusivity, thickness, heat diffusion time, thermal conductivity and heat capacity per unit volume, respectively. $Q$ is the absorbed heat flux by the heating laser pulse and $\delta(t)$ is the delta function. The five-layered model (a) fully describes the sample structure in this study. In order to simplify the model (a), two kinds of three-layered models are introduced. For the three-layered model (b), Pt/Mg(OH)$_2$/Pt is supposed to be a homogeneous plate. Using the model (b), the heat diffusion time $\tau$ of Pt/Mg(OH)$_2$/Pt can be obtained. Then, the value of $\tau$ is analyzed using the AHDT method based upon the three-layered model (c) of Pt/Mg(OH)$_2$/Pt under the adiabatic condition.

Within the layers, the temperature $T(x, t)$, where $t$ is the time and $x$ is the position, can be expressed using one-dimensional heat diffusion equations normal to the respective layers as follows:

\[
\begin{align*}
C_1 \frac{\partial T_1(x, t)}{\partial t} &= k_1 \frac{\partial^2 T_1(x, t)}{\partial x^2} \quad (x < 0) \quad (1) \\
C_2 \frac{\partial T_2(x, t)}{\partial t} &= k_2 \frac{\partial^2 T_2(x, t)}{\partial x^2} \quad (0 \leq x < d) \quad (2) \\
C_3 \frac{\partial T_3(x, t)}{\partial t} &= k_3 \frac{\partial^2 T_3(x, t)}{\partial x^2} \quad (x > d), \quad (3)
\end{align*}
\]

where $C$ and $k$ are the heat capacity per unit volume and the thermal conductivity, respectively. In addition, the subscript numbers represent the layer to which the parameter belongs. The boundary conditions at $x = -\infty$, 0, $d$ and $\infty$ are as follows:

\[
\begin{align*}
T_1(-\infty, t) &= T_1(\infty, t) = 0, \quad (4) \\
T_1(x, t)|_{x \to 0} &= T_2(x, t)|_{x \to 0}, \quad (5) \\
Q \delta(t) &= -k_2 \frac{\partial T_2(x, t)}{\partial x} \bigg|_{x \to 0} + k_1 \frac{\partial T_1(x, t)}{\partial x} \bigg|_{x \to 0}, \quad (6)
\end{align*}
\]

where $Q$ is the absorbed heat flux and $\delta(t)$ is the delta function:

\[
\begin{align*}
T_2(x, t)|_{x \to d-0} &= T_1(x, t)|_{x \to d-0}, \quad (7) \\
k_2 \frac{\partial T_2(x, t)}{\partial x} \bigg|_{x \to d-0} &= k_1 \frac{\partial T_1(x, t)}{\partial x} \bigg|_{x \to d}. \quad (8)
\end{align*}
\]

By applying the Laplace transformation to equations (1)–(8), equations (1)–(3) can be solved. The matrix equation [24] is described as

\[
\begin{bmatrix}
b_2 \sqrt{p} T_{1}(p) \\
T_{1}(p)
\end{bmatrix} = \begin{bmatrix}
\cosh \sqrt{p} + \beta \sinh \sqrt{p} & -b_2 \sqrt{p} \sinh \sqrt{p} \\
-b_2 \sqrt{p} \cosh \sqrt{p} & \cosh \sqrt{p} - \beta \sinh \sqrt{p}
\end{bmatrix} \times \begin{bmatrix}
Q \\
\bar{T}_1(p)
\end{bmatrix}.
\]

where $p$ is variable in the Laplace space, $b = \sqrt{Ck}$ is the thermal effusivity, and $T_1(p)$ and $\bar{T}_1(p)$ are the Laplace transforms of $T_1(0, t)$ and $T_2(x, t)$, respectively. In addition, the heat diffusion time, $\tau$, is described using the thickness, $d$, and the thermal diffusivity, $\alpha$, of the sample as follows:

\[
\tau = \frac{d^2}{\alpha}.
\]

Equation (9) can be simplified as

\[
\begin{bmatrix}
b_2 \sqrt{p} (\sinh \sqrt{p} \tau + \beta \cosh \sqrt{p} \tau) & b_2 \beta \sqrt{p} \\
-b_2 \beta \sqrt{p} & \cosh \sqrt{p} \tau - \beta \sinh \sqrt{p} \tau
\end{bmatrix} \begin{bmatrix}
\tilde{T}_1(p) \\
\bar{T}_1(p)
\end{bmatrix} = Q \begin{bmatrix}
\cosh \beta \sqrt{p} \tau \\
\sinh \beta \sqrt{p} \tau
\end{bmatrix}.
\]

\[
\gamma = \frac{1 - \beta}{1 + \beta}. \quad (13)
\]

Equation (12) can be solved and the inverse Laplace transform of equation (12) gives the temperature at $x = d$ is

\[
\tilde{T}_1(p) = \frac{2Q}{b_2 \sqrt{p}(1 + \beta)^2} \sum_{n=0}^{\infty} \gamma^{2n} \exp[-(2n + 1) \sqrt{p} \tau].
\]

Equation (14) describes the transient temperature curve at the rear face of the Pt/Mg(OH)$_2$/Pt plate embedded between the NaCl layers when the front face of the plate is heated by a laser pulse. Figure 8 shows that equation (14) was fitted to the transient temperature curve of cell 2 at 47 GPa. The decrease in the temperature behind the maximum temperature rise is attributed to the heat effusion from the Pt/Mg(OH)$_2$/Pt plate to the NaCl layers. The fitting curves can well reproduce the observed transient temperature curves. The heat diffusion times of the Pt/Mg(OH)$_2$/Pt plates were analyzed using equation (14) and listed in table 1.
4.1.2. An areal heat diffusion time method of the Pt/Mg(OH)$_2$/Pt three-layer model. Subsequently, the thermal diffusivity of Mg(OH)$_2$ was analyzed based on the AHDT method for the three-layered model of Pt (layer i)/Mg(OH)$_2$ (layer j)/Pt (layer k) as shown in figure 7(c). The definition of the AHDT is described in figure 6. Here, the transient temperature curve is normalized as the maximum temperature rise becomes 1. The AHDT of three layers, $A_3$, can be expressed as [24]

$$A_3 = \lim_{\rho \to 0} \frac{1}{P} \left[ b_1 \sqrt{\tau_1} + b_2 \sqrt{\tau_2} + b_3 \sqrt{\tau_3} \right] T_i(\rho)$$

$$= \frac{b_1 \sqrt{\tau_1(\frac{2+\sqrt{5}}{2})} + b_2 \sqrt{\tau_2(\frac{2+\sqrt{5}}{2})} + b_3 \sqrt{\tau_3(\frac{2+\sqrt{5}}{2})}}{b_1 \sqrt{\tau_1} + b_2 \sqrt{\tau_2} + b_3 \sqrt{\tau_3}} \left( \sqrt{\tau_1(\frac{2+\sqrt{5}}{2})} + \sqrt{\tau_2(\frac{2+\sqrt{5}}{2})} + \sqrt{\tau_3(\frac{2+\sqrt{5}}{2})} \right),$$

(15)

where i, j and k represent the respective layers. Considering the symmetry of the layered structure, subscripts are changed: i = k = Pt, j = Mg(OH)$_2$.

$$A_3 = \left\{ \frac{4}{3} C_{Pd} d_P + \frac{C_{Mg(OH)_2} d_{Mg(OH)_2}}{6} \right\} \frac{1}{\tau_P}$$

$$+ \left( \frac{C_{Pd} d_P}{\alpha_{Mg(OH)_2}} \right)^2 + \frac{C_{Mg(OH)_2} d_{Mg(OH)_2}}{6} \right\} \frac{1}{\tau_P}$$

(16)

The value of $A_3$ is obtained, the thermal diffusivity of Mg(OH)$_2$ can be systematically analyzed. However, the area of $A_3$ cannot be directly determined from the experimental curves since the boundary condition at the Pt film face of the NaCl side was not adiabatic in the experiment. Therefore, the value of $A_3$ is deduced from the heat diffusion time, $\tau$, of Pt/Mg(OH)$_2$/Pt, which is regarded as a homogeneous plate. Baba has also reported the AHDT for the homogeneous plate, $A_1 = \tau / 6$ [24]. If substituting $A_1$ for $A_3$ in equation (16) is acceptable, equation (16) can be converted as follows:

$$\alpha_{Mg(OH)_2} = \frac{\frac{\tau}{6} + 1}{(G + 2)A_3 - (G + \frac{1}{2}) \frac{\partial C_{Pd}}{\partial P}} d^2_{Mg(OH)_2},$$

(18)

$$\Gamma = \frac{\frac{\tau}{6} + 1}{(G + 2)A_3 - (G + \frac{1}{2}) \frac{\partial C_{Pd}}{\partial P}} \frac{d^2_{Mg(OH)_2}}{C_{Pd} d_P}.$$  

(19)

The values for $\tau$ of Pt/Mg(OH)$_2$/Pt regarded as a homogeneous plate are listed in table 1. Consequently, if the values of the heat capacities per unit volume of Mg(OH)$_2$ and Pt, the thermal diffusivity of Mg(OH)$_2$ can be derived from equation (18) using the value of $\tau$.

4.2. Thermal diffusivity of Mg(OH)$_2$

In order to calculate the thermal diffusivities of Mg(OH)$_2$ using equation (18), thermophysical and structural properties needed

<table>
<thead>
<tr>
<th>Specimen</th>
<th>$P$ (GPa)</th>
<th>$d_{Mg(OH)_2}$ (µm)</th>
<th>$d_P$ (µm)</th>
<th>$\alpha_P$ (m² s⁻¹)</th>
<th>$\tau$ (10⁻⁶ s)</th>
<th>$\alpha_{Mg(OH)_2}$ (10⁻⁶ m² s⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 cm² plate</td>
<td>0</td>
<td>–</td>
<td>–</td>
<td>2.5 x 10⁻⁵</td>
<td>–</td>
<td>0.9</td>
</tr>
<tr>
<td>1</td>
<td>10</td>
<td>1.5</td>
<td>0.84</td>
<td>2.6 x 10⁻⁵</td>
<td>6.2</td>
<td>1.1 ± 0.39</td>
</tr>
<tr>
<td>2</td>
<td>28</td>
<td>2.0</td>
<td>0.46</td>
<td>2.8 x 10⁻⁵</td>
<td>4.8</td>
<td>1.5 ± 0.45</td>
</tr>
<tr>
<td>2</td>
<td>40</td>
<td>2.0</td>
<td>0.45</td>
<td>2.9 x 10⁻⁵</td>
<td>1.7</td>
<td>4.2 ± 1.6</td>
</tr>
<tr>
<td>2</td>
<td>47</td>
<td>2.0</td>
<td>0.45</td>
<td>3.0 x 10⁻⁵</td>
<td>1.7</td>
<td>4.1 ± 1.2</td>
</tr>
<tr>
<td>2</td>
<td>57</td>
<td>1.9</td>
<td>0.45</td>
<td>3.1 x 10⁻⁵</td>
<td>1.2</td>
<td>5.9 ± 1.8</td>
</tr>
</tbody>
</table>

* Averaged thicknesses of the front and back Pt films are shown.

$b$ The value is calculated using the thermal conductivity, the density and the specific heat capacity of Pt from [31].
for the calculation are estimated below. The thicknesses of the Mg(OH)$_2$ plates at high pressure were calculated based upon the reported compressibility data. Xia et al. [25] have reported the compressibility of the volume up to 10 GPa for the Mg(OH)$_2$ crystal using a Birch–Murnaghan equation [26]. We extrapolated their equation up to 57 GPa, and the compressibility of the thickness was estimated as one-third of that of the volume. With respect to the Pt films, the compressibility of the thickness was estimated as one-third −2370 kg m$^{-1}$ for the specific heat capacity and 21 400 kg m$^{-1}$ mol$^{-1}$.

We supposed that the values of $C_p$ would also behave the same as the above metals. Thus, it is roughly estimated to be twice the uncertainty of the thickness with the data used for the calculations. The uncertainty is of the same order of magnitude and of opposite sign for $(\partial \ln \rho / \partial P)_T$ [2], Ross et al. [2] have summarized that the derivatives of Al, Ag, Au, Cu, Fe and Ni are all about 4%/GPa for thermal conductivity as well as thermal diffusivity. We assumed that Pt should increase at the rate of $4\%$/GPa.

As a result, the thermal diffusivities of Mg(OH)$_2$ are calculated using equation (18) and listed in table 1 along with the data used for the calculations. The uncertainty is roughly estimated to be twice the uncertainty of the thickness measurement as mentioned in section 3.1. The thermal diffusivity of Mg(OH)$_2$ increases with the increase in pressure. The thermal diffusivity of Mg(OH)$_2$ at 57 GPa becomes $5.9 \times 10^{-6}$ m$^2$ s$^{-1}$ that is about six times larger than that at ambient pressure.

### 4.3. Verification of analyzed data

In order to verify the analysis procedure of the thermal diffusivity described in section 4.1, the transient temperature curve at 57 GPa was simulated using a finite element method. In the simulation, the experimental setup of the sample was simplified as a one-dimensional heat transfer model such as heatbath/NaCl/Pt/Mg(OH)$_2$/Pt/NaCl/heatbath (figure 9). A horizontal bold line shows the modeling of the sample, and

![Figure 9](image-url)
**Figure 10.** Temperature distribution of the model described in figure 9. At \( t = 0 \) and \( x = 0 \), impulse heating was applied. In the experiment, the temperature at the Pt/NaCl interface (\( x = 2.84 \) μm) was measured.

**Figure 11.** Measured transient temperature curve obtained at 57 GPa is compared with the simulation using the finite element method. The broken lines in the figure also show the simulation results when the uncertainty of thermal diffusivity for Mg(OH)\(_2\) is considered.

### 4.4. Pressure dependence of thermal conductivity for Mg(OH)\(_2\)

It is well known that thermal transport properties for insulator at elevated pressure directly relate to the change of the state of phonon. Hofmeister [4] has proposed the model describing the effect of pressure and temperature on lattice thermal conductivity, \( k(P, T) \) as follows:

\[
k(P, T) = k(298)\left(\frac{298}{T}\right)^a \times \exp\left[-\left(4\gamma_G + \frac{1}{2}\right)\int_{298}^T \nu(\theta) \, d\theta\right] \times \left(1 + \frac{K'_0}{K_0}P\right) + f(T),
\]

(20)

where \( a \) is the fitting parameter, \( \gamma_G \) is the Grüneisen parameter, \( \nu \) is the thermal expansion, \( K_0 \) is the bulk modulus at 0 GPa and \( K'_0 \) is the pressure derivative of the bulk modulus. All of our experiments were carried out at room temperature. Thus, equation (20) becomes

\[
k(P) = k_{P=0}\left(1 + \frac{K'_0}{K_0}P\right),
\]

(21)

where \( k_{P=0} \) is the thermal conductivity at the ambient pressure.

The Mg(OH)\(_2\) lattice structure is tetragonal; the \( a \) axis consists of the Mg–O layer and in contrast to that, the \( c \) axis contains a weak –O–H H–O– bond. Therefore, there is large anisotropy in the pressure dependence of the unit cell parameters \( a \) and \( c \). Xia et al [25] have reported that the equation of state for Mg(OH)\(_2\) is described by not only the unit cell volume but also the volume of \( a^3 \) and \( c^3 \). They have shown that there is a significant difference among the values of \( K_0 \) and \( K'_0 \) for the volume of the unit cell, \( a^3 \) and \( c^3 \) such as 39.6 GPa and 6.7, 79.1 GPa and 3.7, 18.24 GPa and 6.27, respectively.

Figure 12 shows that the experimental thermal conductivities, \( k \), are compared to equation (21), which takes into account the variation of \( K_0 \) and \( K'_0 \) for the volume of the unit cell, \( a^3 \) and \( c^3 \). Here the experimental thermal conductivities are derived from \( k = \alpha C \). In figure 12, the model based on the \( a \) axis well reproduces the experimental results by 28 GPa. Xia et al have reported that the linear compressibility of the \( a \) and \( c \) axes becomes almost unity [25] by 10 GPa. In addition, their data suggest that the compressibility of the \( c \) axis converges along with that of the \( a \) axis with the increase of pressure. For this reason, it is considered that the thermal conductivity of Mg(OH)\(_2\) should reflect the compressibility of the \( a \) axis rather than that of the \( c \) axis.

### 4.5. Temperature increase by pulse heating

Since the thermal diffusivity is affected not only by pressure but also by temperature, it is of importance to estimate the increase of the temperature for Mg(OH)\(_2\) by the pulse heating.

The coated Mg(OH)\(_2\) plates were surrounded by the pressure medium, but to simplify the estimation the thermal effusion from the coated Mg(OH)\(_2\) plate to the pressure medium is ignored here. The increase of temperature by a laser pulse, \( \Delta T \), is expressed using provided heat per unit area by a single laser pulse, \( q \), averaged heat capacity of unit volume for the Pt/Mg(OH)\(_2\)/Pt-layered structure, \( \bar{C} \), and total thickness of...
with the pulse heating are estimated to be 8–9 K. It is expected that such a temperature increase slightly affects Mg(OH)$_2$ for the thermal diffusivity, because the temperature dependence on insulators for thermal diffusivity is generally proportional to the inverse of absolute temperature at near room temperature.

5. Conclusions

The thermoreflectance apparatus adapted to a DAC was developed to obtain the thermal diffusivity at high pressure. We observed transient temperature curves when the heat diffuses across the plate samples in the DAC. The analysis procedure for the transient temperature curve was described and the thermal diffusivities of Mg(OH)$_2$ up to 57 GPa were obtained. The thermal diffusivity of Mg(OH)$_2$ increases as the attained pressure increases. At 57 GPa, the thermal diffusivity of Mg(OH)$_2$ is $5.9 \times 10^{-6}$ m$^2$ s$^{-1}$, which is larger than that of ambient pressure by a factor of $\approx 6$.

References

[4] Hofmeister A M 1999 Mantle values of thermal conductivity and the geotherm from phonon lifetimes mantle values of thermal conductivity and the geotherm from phonon lifetimes Science 283 12
[14] Goncharov A F, Struzhkin V V and Jacobsen S D 2006 Reduced radiative conductivity of low-spin (Mg,Fe)O in the lower mantle Science 312 1205

Table 2. Physical values for the estimation of the temperature increase by pulse heating.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Cell 2</th>
<th>$P$ (GPa)</th>
<th>$d_{Mg(OH)2}$ ($10^{-6}$ m)</th>
<th>$d_{NaCl}$ ($10^{-6}$ m)</th>
<th>$d_{total}$ ($10^{-6}$ m)</th>
<th>$C_{Mg(OH)2}$ ($10^6$ J m$^{-3}$ K$^{-1}$)</th>
<th>$C_{NaCl}$ ($10^6$ J m$^{-3}$ K$^{-1}$)</th>
<th>$R_{NaCl}$/dia</th>
<th>$R_{NaCl}$/Pt</th>
<th>$R_{dia}$/NaCl</th>
<th>$I$ ($10^{-7}$ J)</th>
<th>$r$ ($10^{-6}$ m)</th>
<th>$\Delta T$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>57</td>
<td>1.9</td>
<td>0.45</td>
<td>2.8</td>
<td>3.1</td>
<td>2.85</td>
<td>0.17</td>
<td>0.05</td>
<td>0.65</td>
<td>5</td>
<td>25</td>
<td>8.3</td>
</tr>
</tbody>
</table>

Figure 12. Thermal conductivity of Mg(OH)$_2$ in relation to pressure along with the calculated curves. The calculated curves were derived from equation (21) using bulk modulus and its pressure derivative reported by Xia et al [25]; $K_0 = 79.1$ GPa and $K'_0 = 3.7$ for the a axis, $K_0 = 18.24$ GPa and $K'_0 = 6.27$ for the c axis, $K_0 = 39.6$ GPa and $K'_0 = 6.7$ for the unit cell.