



## Radiative heat transfer in a hydrous mantle transition zone

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### ABSTRACT

The structure and dynamics of Earth's interior depend crucially upon heat flow and thus upon the thermal conductivity of its constituents. We measured optical absorbance spectra of hydrous wadsleyite and hydrous ringwoodite at simultaneous high-pressure and high-temperature conditions up to 26 GPa and 823 K in order to determine their radiative conductivities and to study the potential influence of hydration in the transition zone on thermal conductivity of the mantle. We report radiative thermal conductivities of  $1.5 \pm 0.2 \text{ Wm}^{-1} \text{ K}^{-1}$  for hydrous wadsleyite and  $1.2 \pm 0.1 \text{ Wm}^{-1} \text{ K}^{-1}$  for hydrous ringwoodite at transition zone conditions. The analytically derived radiative thermal conductivities of anhydrous wadsleyite and ringwoodite are 40% and 33% higher, respectively. The total thermal conductivities, calculated from temperature- and pressure-dependent optical absorption measurements, maintain an energy transmission window in the infrared and visible spectral range at high pressures and temperatures. The results indicate that the mantle transition zone may contribute significantly to heat transfer in the mantle and demonstrate the importance of radiative heat transfer in controlling geodynamic processes in Earth's mantle.

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### 1. Introduction

Heat in Earth's interior is transported by convection and conduction. Temperature- and pressure-dependent thermal conductivity of mantle materials is an important parameter for geodynamic models of mantle convection (e.g., Dubuffet et al., 2000, 2002; Matyska et al., 1994; van den Berg et al., 2001). The bulk thermal conductivity has two components: lattice conductivity ( $k_{\text{lat}}$ ) and radiative conductivity ( $k_{\text{rad}}$ ) (e.g., Clark, 1957; Schatz and Simmons, 1972). Whereas lattice conductivity is governed by phonon propagation, radiative conductivity arises from heat transport by emission and absorption of photons (e.g., Hofmeister, 2005, 2010). The latter, therefore, can be indirectly measured by analyzing the visible and infrared (vis-IR) regions of a material's optical absorption spectrum because absorption bands provide a mechanism for diffusive radiative transfer (Hofmeister, 2004). Thermal conductivity in the mantle is controlled by temperature, pressure, the electronic structure and concentration of transition metal ions (such as iron), and the water content of the material (Clark, 1957; Hofmeister, 2004).

Until recently, radiative heat transfer was considered relatively unimportant in the mantle. Earlier experimental work suggested that most ferromagnesian mantle minerals become opaque in the

vis-IR range at high-pressure due to intensification and red-shift of  $\text{Fe}^{2+}-\text{Fe}^{3+}$  charge-transfer bands (Mao, 1976; Mao and Bell, 1972). More recent studies show some mantle minerals such as ringwoodite remain transparent at high pressure (Keppler and Smyth, 2005), and thus radiative conductivity could contribute to the total heat flux of the Earth's mantle (Dubuffet et al., 2000, 2002). The total thermal conductivity, including  $k_{\text{lat}}$  and  $k_{\text{rad}}$ , and its dependence upon temperature, pressure, and composition, is essential for accurate models of heat flow in Earth (Dubuffet et al., 2000, 2002; Hofmeister, 2004). For example, Hofmeister (2004) showed that structurally incorporated  $(\text{OH})^-$  in nominally anhydrous minerals can contribute significantly to their total thermal conductivity by providing a mechanism for radiative diffusion of heat. At 1500 K, Hofmeister (2004) reports for sub-cm-sized olivine with  $\sim 10\text{--}100 \text{ wt ppm H}_2\text{O}$  a magnitude of  $k_{\text{rad}}$  of  $\sim 1/8$  of the lattice contribution to  $k_{\text{tot}}$  of olivine. The presence of  $\text{H}_2\text{O}$  enhances diffusion rates, promotes the growth of large grains, allows steeper temperature gradients, affects flow and fabric and can be invoked to explain slab behaviour (Hofmeister, 2004; Jung and Karato, 2001). Models suggest that a larger  $k_{\text{rad}}$  may stabilize planforms of mantle convection (Dubuffet et al., 2000, 2002) and allow greater heat loss and faster cooling of material. Other theoretical work shows that without the addition of radiative conductivities to geodynamic models of Earth's lower mantle, geophysical inferences of megaplumes are difficult to explain (Matyska et al., 1994; van den Berg et al., 2001). However, experimental results on temperature effects upon radiative heat transfer were not available, and absolute

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$k_{\text{rad}}$  values were uncertain. Instead, olivine spectra were used to model mantle heat transfer and it was assumed that denser phases have similar spectral features (e.g., Hofmeister, 2005).

Most previous work focused on Earth's upper and lower mantle and assumed grain sizes of 0.1–1 cm (e.g., Fukao et al., 1968; Goncharov et al., 2006, 2010; Keppler et al., 2008; Shankland et al., 1979). Experimentally derived thermal conductivity values for the lower mantle are inconclusive and vary by an order of magnitude (Goncharov et al., 2006; Hofmeister, 2010; Keppler et al., 2008). Recent experiments suggest a lower mantle thermal conductivity of 8–9 Wm<sup>-1</sup> K<sup>-1</sup> (Manthilake et al., 2011). Little is known about the mantle transition zone at 410–660 km depth, which constitutes about 7.5% of Earth's mass. The transition zone is thought to consist mainly of high-pressure polymorphs of olivine, wadsleyite and ringwoodite, along with garnet–majorite solid solutions (Ringwood, 1991). The thermal conductivity of the transition zone contributes to the regulation of heat flux between lower and upper mantle. The transition zone may be critical to the dynamics of the entire mantle because many subducted slabs appear to stagnate there, and because the transition zone minerals wadsleyite and ringwoodite have the highest H<sub>2</sub>O storage capacity among mantle minerals.

Previous measurements of thermal conductivity in mantle minerals include work at high pressures and room temperature (Goncharov et al., 2006, 2010; Keppler et al., 2008; Keppler and Smyth, 2005), at high temperature and room pressure (Shankland et al., 1979), and at simultaneous *P*–*T* conditions, to 20 GPa and 1373 K (Xu et al., 2004). Manthilake et al. (2011) measured  $k_{\text{lat}}$  of Al- and Fe-bearing silicate perovskite up to 26 GPa and 1073 K, and ferropericlase with a range of Fe-contents up to 14 GPa and 1273 K. These studies have led to implications for radiative heat transfer and estimates of the lattice thermal conductivity of mantle material. However, there is a lack of data about the radiative conductivity of transition zone minerals, especially under hydrous conditions and at simultaneous high pressures and temperatures. Earlier studies on radiative thermal conductivity in wadsleyite and majorite (Keppler and McCammon, 1996; Ross, 1997) concluded that those minerals are relatively unimportant for heat transfer. Other studies (Keppler and Smyth, 2005) showed that ringwoodite might contribute to radiative heat transfer in the mantle, but the effect of temperature at high pressure has not been determined. Theoretical models have estimated temperature–pressure variation of thermal conductivity (e.g., Stackhouse and Stixrude, 2010), without, however, accounting for radiative contributions. Future models can benefit from experiments at conditions relevant to Earth's mantle. Here, we studied the effect of pressure, temperature and hydration on the optical absorption spectrum of wadsleyite and ringwoodite at mantle conditions.

## 2. Material and methods

### 2.1. Sample syntheses and characterisation

Gem-quality single crystals of Fo<sub>90</sub>-composition hydrous wadsleyite and hydrous ringwoodite were synthesized at 18 GPa and 1400 °C in the 5000-ton multi-anvil apparatus at the Bayerisches Geoinstitut, Bayreuth, Germany by Smyth et al. (2003) (ringwoodite, run SZ0104) and by Jacobsen (wadsleyite, run Z570, see Mao et al., 2011). The ringwoodite crystals from run SZ0104 are reported to contain 1.07 wt% H<sub>2</sub>O by FTIR (Smyth et al., 2003), in good agreement with 1.11 wt% H<sub>2</sub>O measured by secondary ion mass spectrometry (SIMS) (Mao et al., 2011) and 0.9 wt% H<sub>2</sub>O measured by Raman spectroscopy using the technique from Thomas et al. (2009). The average ferric iron content of the hydrous ringwoodite is about 10% (Smyth et al., 2003). Hydrous Fo<sub>90</sub>-wadsleyite crystals came from run Z570 and contain

1.9(2) wt% H<sub>2</sub>O as determined by SIMS (Mao et al., 2011), which is in fairly good agreement with a value of 1.4 wt% H<sub>2</sub>O determined by FTIR using the absorption coefficient established in Deon et al. (2010). Mössbauer spectroscopy shows the wadsleyite also contains about 10% ferric iron (Mao et al., 2011).

### 2.2. Optical absorption spectroscopy

For optical absorption measurements in the IR–vis–UV spectral range (400–50000 cm<sup>-1</sup>), 50 to 60 μm-sized optically clear single-crystals of ringwoodite (blue) and wadsleyite (green) were doubly polished to thicknesses of 13 μm, and 18 and 19 μm, respectively. Samples were loaded in resistively heated diamond-anvil cells with type IIa diamonds (300 μm culet size) and argon as the pressure medium (Figs. S1–S3). Several ruby spheres were placed next to the sample, and pressure was determined using the pressure-dependent energy shift of the R<sub>1</sub> ruby fluorescence line (Mao et al., 1986; Goncharov et al., 2005). Additional experimental details can be found in the supplementary material.

UV–vis spectra were acquired using the 300 grooves/mm grating of an Acton Research Corporation Spectra Pro 500-i spectrometer with a 0.5 m triple grating monochromator and a system of all-reflecting relay optics with double confocal geometry. A combined deuterium/halogen lamp served as the light source, focused to ~20 μm diameter at the sample. Unpolarised IR-spectra were obtained with a Varian Resolution Pro 670-IR spectrometer. For each spectrum, 1024 scans were accumulated in the range of 400–10000 cm<sup>-1</sup>, using a quartz (2800–10000 cm<sup>-1</sup>) or KBr (400–6000 cm<sup>-1</sup>) beam splitter with 4 cm<sup>-1</sup> resolution. After taking measurements at high pressure and room temperature (Figs. S4 and S5), ringwoodite was studied at 26 GPa up to 650 °C, and wadsleyite spectra were recorded at 15 GPa up to 450 °C. At each pressure and temperature, spectra through the sample and the reference spectrum through diamonds and pressure medium were obtained for the flat-field correction.

For data analysis the spectra from three (UV–vis–IR) spectral ranges were merged after correction for reflection losses and sample thickness to produce the final spectrum. The typical photon path length estimated from optical absorption spectra is on the order of ~200 μm. This allows a proper description of the radiative part of heat conduction in the deep Earth, as it is smaller than the grain size in the Earth's mantle 0.1–1 cm (Keppler et al., 2008). Spectra were fitted using Peakfit software of Jandel Scientific. A linear baseline subtraction was applied. The linear baseline subtraction was only used to characterize the absorption bands and determine band positions. The basis for thermal conductivity calculations used measured spectra corrected exclusively for thickness and reflection losses.

### 2.3. Correction for reflection losses in measured spectra

Corrections for reflection losses were carried out according to Keppler et al. (2008). The absorbance  $A_R$  due to reflection was subtracted from the measured absorbance.  $A_R$  was calculated from the reflectivity given by the Fresnel formula

$$A_R = -2\log(1-R)$$

$$R = (n_{\text{sample}} - n_{\text{Ar}})^2 / (n_{\text{sample}} + n_{\text{Ar}})^2$$

where  $n_{\text{sample}}$  and  $n_{\text{Ar}}$  are the refractive indices of sample and argon, respectively. For argon a pressure-dependent refractive index was utilised (Grimsditch et al., 1986). We used a constant refractive index of 1.76 for wadsleyite and 1.77 for ringwoodite (Anthony et al., 1995). The net result of this approximation on the measured absorption spectra is negligible (<0.007 absorbance units). Moreover, the correction for reflection losses affects the results very moderately (<0.007 absorbance units), which is within the experimental

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