Thermal conductivity of spinels and olivines from vibrational spectroscopy: Ambient conditions

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ABSTRACT

The damped harmonic oscillator model for thermal conductivity of insulators is improved, leading to a formula that predicts thermal conductivity at ambient conditions ($k_0$) from various physical properties, most of which are commonly measured. Specifically, $k_0 = [\rho/(3ZM)] C_v [(u_p + u_s)/2]^2/\langle\Gamma\rangle$, where $\rho$ is density, $Z$ is the number of formula units in the primitive unit cell, $M$ is the molar weight, $C_v$ is heat capacity, $u$ is the sound speed ($P$ denotes compression; $S$ denotes shear), and $\langle\Gamma\rangle$ is the average of the damping coefficients determined from peak widths in infrared reflectivity spectra, or from suitable Raman and Brillouin spectra. The classical physics and quantum-mechanical basis for this model is discussed, with emphasis on the effect of phonon-phonon interactions on mode properties. The calculated values of $k_0$ all lie within the experimental uncertainty of the measurements for all samples with the spinel or olivine structure examined by Horai (1971) with known or approximately correct chemical compositions. Other divergent measurements of $k$ for MgAl$_2$O$_4$ are discounted for various reasons. Early studies of Fe-bearing spinels are not generally reliable, but rough estimates from the above equation are consistent with all data, and good agreement is obtained for samples such as Mg$_{0.8}$Fe$_{0.2}$Al$_2$O$_4$ and $\gamma$-Fe$_2$SiO$_4$ for which the previous authors obtained chemical data, and for which IR reflectivity data exist. The theory reproduces the measured dependence of $k_0$ on composition and structure. Anisotropy in $k_0$ results mainly from differences in lattice constants ($j$): the equation for olivine is $k_j/k_0 = [V(j)/j]^{0.73}$ which predicts the ratios within 3%. For solid solutions between Fe and Mg, the model provides a non-linear dependence of $k_0$ on mol% Fe, with the damping coefficient being the key factor producing non-linearity. Predicted ambient values are 11.3 $\pm$ 0.4 W/m-K for $\gamma$-Mg$_2$SiO$_4$, 6.5 $\pm$ 0.7 W/m-K for $\gamma$-Mg$_{0.7}$Fe$_{0.3}$SiO$_4$, and 6.9 $\pm$ 0.3 W/m-K for $\beta$-Mg$_2$SiO$_4$. The high $k_0$ for ringwoodite suggests that heat in Earth’s transition zone should be conducted twice as efficiently as in the adjacent upper and lower mantles: this discontinuous depth dependence of $k$ could impact thermal models of conduction in subducting slabs and of mantle convection.

INTRODUCTION

Thermal conductivity ($k$) is the key property in planetary heat transport. It not only governs conductive processes that occur in subducting slabs and in the lithosphere, but also plays a critical role in the time-dependence of mantle convection. Although the importance of variations in $k$ with temperature or pressure on convection was recognized prior to formulation of the paradigm of plate tectonics (MacDonald 1959; Lubimova 1958), inclusion of variable $k$ in convection models has been hampered by inconsistencies among measured values for thermal conductivity, by the scarcity of data, and by inaccuracies in modeling $k$ from other physical properties or from first principles. The goal of the present paper is to provide a useful relationship between thermal conductivity at ambient conditions ($k_0$) and easily measured physical properties, to establish the accuracy of this formula by testing it against olivine- and spinel-group minerals, and to predict $k_0$ for the high-pressure poly-morphs of olivine [(Mg,Fe)$_2$SiO$_4$] that are considered to be volumetrically important phases inside the Earth.

The following background material illustrates the problems encountered in experimental and theoretical studies of this physical property, and shows how this developing semi-empirical model for $k$ relates to geophysical studies.

Measurements involving physical contact of the sample and heater are problematic even at ambient conditions, with discrepancies in $k_0$ commonly exceeding 20% (Ross et al. 1984). For example, measured values of $k_0$ of MgAl$_2$O$_4$ (spinel) range from 9.4 W/m-K (Horai 1971) to 22 W/m-K (Slack 1962). Because of experimental difficulties, few thermal conductivity measurements have been made of high-pressure phases, e.g., $k$ has been determined for $\gamma$-Fe$_2$SiO$_4$ (Fujisawa et al. 1968) but not for the Mg-rich phase.

At high $T$, measurements differ greatly. The wide variation in the results for olivine and forsterite are shown in Figure 9 of Harrison (1998). For example, for the (001) faces of olivine with a composition ~Fe$_{0.9}$, reported values of $k$ at 1200 K vary from 3.0 to 5.6 W/m-K (cf., Kanamori et al. 1968; Schatz and...