Ultra-high-pressure thermal-conductivity measurements of griceite and corundum

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Abstract: Time-dependent interface temperatures were measured for 500 Å to 1-μm iron and stainless steel films encapsulated within LiF and Al₂O₃ transparent anvils and shocked to between 200 and 315 GPa (Gallagher et al. 1994). The corresponding anvil shock pressures are 140–170 GPa in LiF and 112–242 GPa in Al₂O₃. Thermal conductivity, k, is related to measured diffusivity, ν = k/ρCₚ, where ρ and Cₚ are the density and specific heat of the shocked state. Existing models predict that the temperature dependence of conductivity may be sensitive to defect concentration, making existing theories inappropriate.

Key words: Griceite, Corundum. Thermal conductivity. Shock temperature. Iron

1. Introduction

The experimental determination of thermal conductivities of dielectrics such as griceite (LiF) and corundum (Al₂O₃) at ultra-high pressures is important because of the use of these materials in experiments to obtain shock temperatures for metals. Those experiments observe the greybody radiation emitted from the surface of the metal through a transparent anvil. The shocked metal’s Planck radiation peaks are in the visible wavelengths because the samples are 10¹⁰ K, and metals are opaque at those wavelengths so their interiors cannot be optically observed. In order to keep the surface of the metal as high pressure after the shock wave has reached it, the metal is put in contact with a transparent window such as griceite or corundum. The internal temperature of the shocked metal is then obtained from the measured metal-window interface temperature and the contrast in thermal conductivities of the two materials.
Measurements of conductivities of dielectrics at high pressures and comparison with theory are also of intrinsic interest. In this paper, we summarize the theory of the effects of temperature and pressure on $k$ for dielectrics and describe our initial experimental results.

2. Theoretical

2.1. Thermal conductivity

Roufossé & Jesuik (1983) produced a theory which Bae et al. (1987) used to calculate the thermal conductivity of dielectric windows used in shock-temperature experiments on iron samples. They assumed a temperature dependence and density dependence of $k$ as follows:

$$k_T = A + B/T$$  (1)

$$k_P = k_0 \left( \frac{P}{P_0} \right)^{n-1.3}$$  (2)

where $A_{Al_2O_3} = -2.5 \text{ W/mK}$, $B_{Al_2O_3} = 1.2 \times 10^8 \text{ W/m}$, $A_{SiO_2} = -0.2 \text{ W/mK}$, and $B_{SiO_2} = 3.7 \times 10^8 \text{ W/m}$ are measured at ambient pressure and high temperature and $\gamma_1$, $\rho_0$, and $\rho$ are the Grüneisen parameter and the initial and compressed densities of the material. Tang (1994) gives a model for conductivity with a theoretical dependence on $T$ and $P$, originally developed by Leibfried.

![Figure 1. Comparison of thermal conductivity of granite and corundum calculated along their Hugoniot with the Roufossé & Jesuik (1983) and Tang (1994) methods. For granite, both theories predict a much higher conductivity than is measured, and for corundum the Tang theory is generally closer to the present data.](image)
Thermal-conductivity measurements

\[ \frac{k}{k_0} = \left( \frac{\rho}{\rho_0} \right)^{1/3} \exp \left[ \frac{\rho_0}{\rho} \left( 1 - \frac{\rho}{\rho_0} \right) \frac{T_f}{T} \right] \]  

(3)

Equation (3) agrees with the Eqs. (1) and (2) in both slope and amplitude at low pressures, but at higher pressures the ther conductivities are lower (Fig. 1).

2.2. Radiative transfer

During the conductivity experiments, the temperature of the metal that is in contact with our dielectric window can be as high as 10^6 K. Thus, it is unclear how much of the observed decay in temperature results from radiative heat transfer as opposed to thermal conduction. However, if the total radiative energy that can be lost from the metal via thermal radiation, \( E_{\text{rad}} \), during the 250 ns of the experiment is compared to the total energy loss implied by the decrease in temperature in our sample, \( \Delta E_{\text{rad}} \), we can determine if the energy from radiative transfer is significant in our experiment. For example, for shot 266, the peak temperature is 10,200 K, so the total greybody radiation energy is at most 170 J/m^2 (Eq. 4). The temperature decrease of the sample is 2000 K in 250 ns, so the energy is 184 J/m^2 (Eq. 5). Thus the ratio \( E_{\text{rad}}/\Delta E_{\text{rad}} \) is 1/11, so to first order, radiative heat transfer may be ignored.

\[ E_{\text{rad}} = \int_{T_f}^{T} \sigma T^4 \, dt \]  

(4)

\[ \Delta E_{\text{rad}} = \rho C_v \Delta T \]  

(5)

where \( \sigma \) is 5.670 x 10^{-8} W/m^2K^4, \( T_f \) is the measured time-dependent temperature of the radiating surface, \( \Delta T \) is the change in temperature of the sample with density \( \rho \), thickness \( t \), and heat capacity \( C_v \).

2.3. Heat flow

To measure thermal conductivity of the window material, a sample configuration in which a thin metal film is sandwiched between two dielectric anvils was used. Symmetric heat flow from the metal film into the anvils occurs upon shock compression of the metal while the shock wave is traveling through the two anvils. Previous work on the shock temperatures of metals (Bass et al. 1987; Ahrens et al. 1990) used a thick metal film with an anvil on one side and a metal driver plate on the other. Thus the heat-flow configuration was approximated as an infinite half-space. Unlike the half-space configuration, the thin-film configuration does not have a simple analytic solution to the coupled thermal conduction equations for the anvil and film, thus finite-difference calculations were performed using the 'CochMan' code (King et al. 1989) (Fig. 2).
solution is symmetric across the center plane of the thin film. Shown are four plots for differing values of the ratio of thermal diffusivities, $R = \kappa_{\text{metal}}/\kappa_{\text{film}}$. For higher values of $R$, the interface temperature is closer to the interior temperature of the film, and it changes more slowly with time.

3. Experimental

Time-resolved radiance from the samples was measured at six wavelengths (e.g., Fig. 3). This is compared to predictions of interface temperature from the model shown in Fig. 2 for various values of $R$. We conclude that the best-fit value of $R = 100$. The $\kappa_{\text{metal}}$ calculated via the Wiedemann-Franz law, was assumed to be valid, whereas the $\kappa_{\text{film}}$ was considered to be the unknown quantity. The measurement of $R$ together with the separately measured complete equations of state of the metal and anvil materials can be used to calculate a value for $\kappa_{\text{metal}}$ in the shocked state. The thermal conductivities determined from these data are shown in Table 1. The corundum anvil did not behave consistently when shocked above 220 GPa. However, the radiative signals from experiments with Al$_2$O$_3$ are usable up to at least 245 GPa.
4. Results

The thermal conductivities measured are reported in Table 1. For LIF the Rouxset and Jeandel theory overpredicts the conductivity by a factor of 10^9 and the Tang theory overpredicts it by a factor of 10^2. For Al₂O₃ the Rouxset and Jeandel theory overpredicts experiment by a factor of 10^2, as does the Tang theory. If we use the metal film’s peak temperature instead of its average temperature we get a lower conductivity, and in fact in the case of corundum the conductivity predicted by Tang agrees with experiment. However this is less physically meaningful because the temperature is very high for a short time and most of the thermal decay occurs while the sample is at a lower temperature.

<table>
<thead>
<tr>
<th>Shot #</th>
<th>Anvil</th>
<th>Fp (GPa)</th>
<th>Ts (K)</th>
<th>k_{TP} (W/mK) calc</th>
<th>k_{TP} (W/mK) calc</th>
<th>k_{TP} (W/mK) mean</th>
</tr>
</thead>
<tbody>
<tr>
<td>265</td>
<td>Al₂O₃</td>
<td>222</td>
<td>2753</td>
<td>1.7</td>
<td>18</td>
<td>3.02</td>
</tr>
<tr>
<td>266</td>
<td>LIF</td>
<td>266</td>
<td>2100</td>
<td>3.5</td>
<td>24.2</td>
<td>0.02</td>
</tr>
<tr>
<td>267</td>
<td>LIF</td>
<td>165</td>
<td>2087</td>
<td>3.6</td>
<td>24.1</td>
<td>0.01−0.002</td>
</tr>
</tbody>
</table>
5. Discussion and conclusions

We conclude that the thermal conductivities of diopside shocked to 150-250 GPa are much lower than has been previously calculated. These low conductivities can be used in data analysis for shock-temperature experiments on metals. A low window conductivity implies a smaller correction from the metal's observed interface temperature to its internal temperature. The reason why theory overpredicts the measured values for thermal conductivity is poorly understood. It may be that the defects in the shocked state inhibit phonon transport. An example of this is that k is too much lower for amorphous solids than for crystalline solids of the same composition. More research on this question is required.

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References


