GEOPHYSICAL RESEARCH LETTERS, VOL. 23, NO. 25, PAGES 3775-3778, DECEMBER 15, 1996

Temperature distribution in the laser-heated diamond cell with external heating, and implications for the thermal conductivity of perovskite

Xiaoyuan Li

Department of Geology and Geophysics, University of California, Berkeley

Michael Manga

Department of Geological Sciences, University of Oregon

Jeffrey H. Nguyen

Department of Physics, University of California, Berkeley

Raymond Jeanloz

Department of Geology and Geophysics, University of California, Berkeley

Abstract. Applying external heating reduces temperature gradients in the laser-heated diamond cell by roughly a factor of two as the external temperature is raised by 300 K. A heat conduction model is presented which can account for most of the reduction. In addition, the temperature distribution measured in a $(Mg_{0.86}Fe_{0.14})SiO_3$ perovskite sample at 56 GPa is compatible the expected inverse temperature-dependence of thermal conductivity.

Introduction

The laser-heated diamond cell is an important tool for experimentally reproducing the pressures and temperatures of the Earth's deep interior. The temperature distribution that is achieved in the diamond cell depends both on the experimental conditions and on the physical properties of the sample, however. In the present study, we combine external heating with laser heating, and measure the temperature distribution inside the laser-heated diamond cell at different background temperatures. Our primary objective is to develop techniques to reduce temperature gradients within laser-heated samples. A thorough understanding of the temperatures inside the laser-heated diamond cell can help improve the use of this instrument, as well as yielding quantitative information about material properties at high pressures and temperatures.

Experimental Methods

The diamond cell used in this study is of a lever-type described by *Ming et al.* [1987]. The diamond anvils are glued with high-temperature epoxy onto WC rockers which in turn are fixed onto a piston and inside a cylinder. A resistance heater made of Pt wire is placed around the anvils, hence is external to the sample, and heats the diamonds, gasket and sample. The piston and cylinder are made of a high-temperature alloy (Inconel 718) in order to minimize temperature-induced deformation of the cell during prolonged heating. In addition, circulating water keeps the body of the cell cool, hence further stabilizing the sample pressure. The background temperature is monitored with a Pt/Pt-Rh thermocouple attached to one of the diamond anvils. This thermocouple is independently calibrated against an internal thermocouple embedded inside the sample chamber, as described by *Ming et al.* [1983] and *Li and Jeanloz* [1987].

The laser-heating system consists of a 25 W continuous-wave Nd:YAG laser operated in TEM_{00} mode, as described by *Heinz and Jeanloz* [1987a]. The thermal radiation of the sample is measured using a CCD detector over the wavelength range of 400 - 900 nm, and the radial temperature distribution is obtained using an inverse Abel transform combined with a gray-body fit to the thermal radiation [*Heinz and Jeanloz*, 1987a; *Jeanloz and Kavner*, 1996].

The starting sample is a natural enstatite of composition $(Mg_{0.86}Fe_{0.14})SiO_3$ from Bamble, Norway. The finely ground sample was contained in a spring-steel gasket having a hole $\sim$120 μm in diameter, along with a thin layer of ruby powder placed on top of the sample for pressure calibration. For the present experiments, the sample pressure was 56($\pm$10) GPa. The starting enstatite is first converted to the high-pressure perovskite phase by laser heating, as described by *Li and Jeanloz* [1990]. Then background temperatures are varied using the external heater, and the sample is further heated at different background temperatures by means of the Nd:YAG laser.

Paper number 96GL03648.
0094-8534/96/96GL-03648$05.00

Results

We measured the temperature distribution within the laser-heated sample at background temperatures, T_0, ranging from 295 K (room temperature) to 605 K. At higher temperatures we risked damaging the diamonds. A selection of the measured thermal-radiation profiles obtained at different background temperatures, T_0, is shown in Fig. 1a. As is evident from the raw data, the width of the thermal-radiation profiles increases rapidly with increasing T_0. Assuming constant emissivity of the sample, the corresponding temperature profiles also become wider as T_0 increases (Fig. 1b). The broadening of the intensity and temperature distributions with increasing T_0 is relatively independent of the power of the focussed laser beam. Thus, for fixed T_0, the width of the hot spot also remains unchanged even when the laser power is changed considerably: see the intensity profiles at 590 and 597 K in Fig. 1a (changes in laser power are reflected by changes in peak amplitude). In contrast, when T_0 is changed at constant laser power, the width of the hot spot changes correspondingly (e.g., profiles at 510 and 590 K in Fig. 1a).

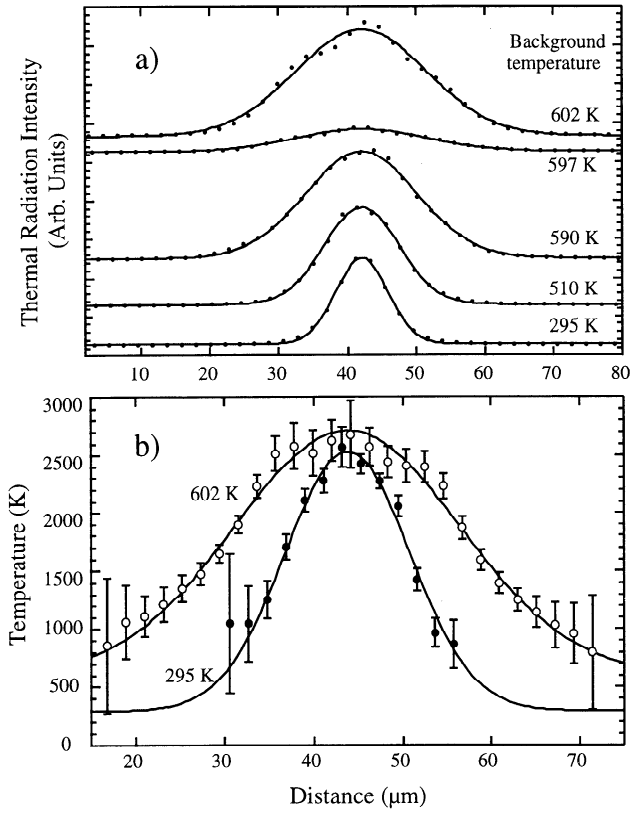

Figure 1. (a) Measured intensity of the emitted thermal-radiation across the laser-heated spot, $I(\lambda, x)$ [see *Jeanloz and Kavner*, 1996]; background temperature achieved by external heating is indicated on the right. For clarity measurements are shown only for the wavelength with maximum intensity at the peak temperature. All profiles are plotted at the same scale, and offset from each other to avoid overlap. (b) Corresponding temperature distributions, $T(r)$, for background temperatures of 295 and 602 K. Temperatures are obtained by inverse Abel transform of intensity measurements [*Heinz and Jeanloz*, 1987a]. Curves in (a) and (b) are Gaussian fits to the data.

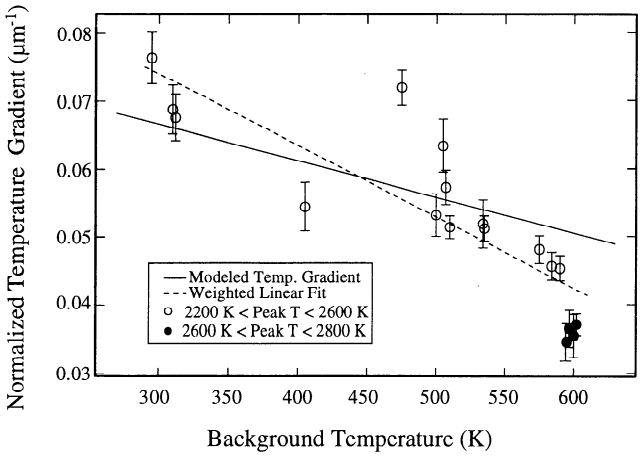

Figure 2. Maximum temperature gradient (see Fig. 1b) normalized by the peak sample temperature as a function of background temperature. Dashed curve is a least-squares fit to all the data. Solid curve is the prediction of the heat conduction model with $k \propto 1/T$ (see text).

All the temperature profiles were fit with Gaussian functions (e.g. Fig. 1b). In order to quantify the effect of external heating, we determine the maximum temperature gradient (calculated from these Gaussian functions and normalized by the peak sample temperature) as a function of T_0 (Fig. 2), for profiles with peak temperatures ranging from 2200 to 2800 K. Temperature gradients are reduced by nearly a factor of 2 as the background temperature increases from 300 to 600 K.

Discussion

The temperature distribution within the laser-heated sample can be modeled based on the thermal diffusion equation [e.g., *Bodea and Jeanloz*, 1989],

$$\rho C \frac{\partial T}{\partial t} = \nabla \cdot k \nabla T + A, \tag{1}$$

where T, ρ, C and k are temperature, density, specific heat and thermal conductivity of the sample. The absorbed power density is described by A. Since the laser-beam power in TEM_{00} mode is Gaussian, and dielectric samples are typically optically thin, A can be approximated as

$$A(r) = A_0 e^{-(r/R)^2}, \tag{2}$$

where $R = 16(\pm 1)\mu$m is the radius of the focussed laser beam [*Williams et al.*, 1991].

Assuming a static solution with axial symmetry, (1) becomes

$$\frac{1}{r}\frac{\partial}{\partial r} r k \frac{\partial T}{\partial r} + \frac{\partial}{\partial z} k \frac{\partial T}{\partial z} = -A(r), \tag{3}$$

where r and z are the radial and axial directions, respec-

tively. As the radial dimension of the sample is much greater than its thickness, the axial portion of equation (3) can be separated out to yield a one-dimensional conduction equation:

$$\frac{\partial}{\partial z}k\frac{\partial T}{\partial z} = -A(r). \quad (4)$$

Due to the high thermal conductivity of the diamonds, the sample-diamond interface should be approximately at the background temperature [*Bodea and Jeanloz*, 1989; *Manga and Jeanloz*, 1996], so that

$$T = T_0 \text{ at } z = \pm L \quad (5)$$

where L represents the half-thickness of the sample. For hot spots with peak temperatures less than 4000 K, the integrated measurement of temperature across the thickness of the sample is within 10% of the maximum temperature between the anvils, even if the emissivity is strongly temperature-dependent [*Manga and Jeanloz*, 1996]. For simplicity, we assume the measured temperature is exactly equal to the maximum value between the anvils at $z = 0$.

We consider three models for the thermal conductivity of the perovskite sample: k = constant, $k \propto T^3$ which corresponds to radiative heat transfer in the diffusion limit [e.g., *Zel'dovich and Raizer*, 1967], and $k \propto 1/T$ which describes heat transfer by phonons at high temperatures [e.g., *Berman*, 1976]. For all three models, analytical solutions can be obtained; for example, if $k \propto 1/T$, the temperature distribution is given by

$$T = T_0 \left(\frac{T_m}{T_0}\right)^{\exp[-(r/R)^2]} \quad (6)$$

where T_m is the peak temperature in the sample.

In Fig. 3 we compare model predictions and observations at a background temperature of 295 K. Contrasting results from the three conductivity models, it is clear that the data are best matched by assuming a thermal conductivity varying inversely with temperature, as expected in non-metals in which thermal conduction is due to phonon transport [*Berman*, 1976]. The temperature-dependence of A can further modulate the temperature distribution. If A increases with increasing temperature [e.g., *Fukao et al.*, 1968; *Mitra*, 1985], the temperature profile becomes more sharply peaked; in fact, a model with k = constant and $A \propto T$ provides an equally good fit to the data in Figs. 2 and 3.

It should be noted that the phonon scattering processes determining thermal conduction at temperatures greater than the Debye temperature (T_D), with $k \propto 1/T$, are different from those acting at low temperatures; the latter result in $\log(k) \propto T_D/T$, and $k \propto T^3$ at still lower temperatures [*Berman*, 1976, pages 48 and 56-58]. *Osako and Ito* [1991] reported measurements of the thermal diffusivity of $MgSiO_3$ perovskite in the range 160-340 K, a temperature range far below the Debye temperature of 1030 K [*Akaogi and Ito*, 1993].

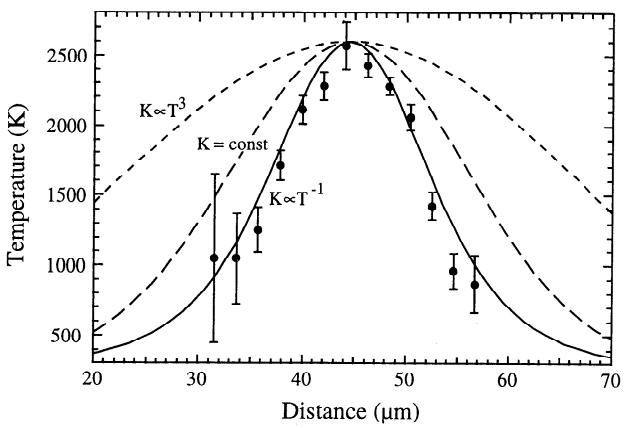

Figure 3. Experimental (solid circles) and theoretical (curves) temperature distributions across the laser-heated spot at a background temperature equal to room temperature. The solid curve is calculated for $k \propto 1/T$; the dashed curve is for k = constant; the dotted curve is for $k \propto T^3$. The curves are not fit to the data, but are calculated independently.

Their diffusivity data, combined with the specific heat data of *Akaogi and Ito* [1993], imply a thermal conductivity that is nearly constant over the temperature range of their study. This can be understood in terms of the phonon mean free path, which gives a measure of the distance over which energy is exchanged between phonon modes. At temperatures below a few hundred K, the mean-free path of phonons can be several mm for non-metallic crystals, so that in small samples, the mean free path of phonons, and consequently k, approach a constant determined by the sample size and geometry [*Berman*, 1976, pages 13-14]. Indeed, the size of the polycrystalline sample studied by *Osako and Ito* [1991] was about 1 mm. Therefore, the constant thermal conductivity derived from the measurements of *Osako and Ito* [1991] and *Akaogi and Ito* [1993] is consistent with an extrinsic value that is governed by the sample size, rather than reflecting intrinsic properties of perovskite.

The model described by (6) predicts that profiles become broader as T_0 increases (Fig. 4). The model is also in reasonable agreement with the experimental observations summarized in Fig. 2 (model predictions are shown with a solid curve for $T_m = 2500$ K), which further supports $k \propto 1/T$; for example, if k and A are constant, then the maximum temperature gradient in Fig. 2 is independent of T_0. The model tends to underestimate the reduction of temperature gradients, in particular, for the four measurements with the highest background and peak temperatures in Fig. 2 (solid points). Thus, external heating may be more effective at reducing temperature gradients than anticipated from the model. Alternatively, temperature profiles can become wider if absorption decreases significantly at the center of the hotspot (in this case for temperatures greater than about 2600 K). For example, because absorption depends strongly on iron content, these results are com-

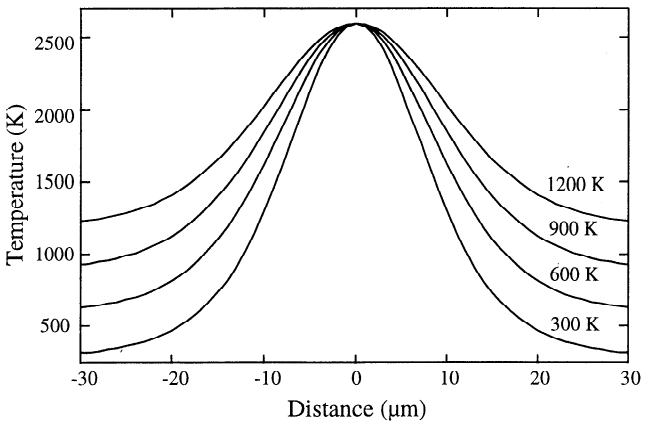

Figure 4. Temperature distribution across the hot spot for different background temperatures using the thermal conductivity model $k \propto 1/T$.

patible with Soret diffusion of iron from the center of the hotspot to colder regions of the sample as the melting temperature is approached. Soret diffusion is sometimes observed in laser-heated perovskite samples [e.g., *Heinz and Jeanloz*, 1987b], although no visible signs of iron migration were detected in the current experiments.

Concluding remarks

The use of an external heater can reduce temperature gradients by up to a factor of two for only modest (300 K) increases in the background temperature, and thus may improve the accuracy of temperature measurements. The shape of measured temperature profiles and the reduced temperature gradients is compatible with the thermal conductivity being inversely proportional to temperature, as expected in non-metallic materials at high temperatures.

Acknowledgments. We thank L.-C. Ming (Hawaii) and A. Kavner for assistance with the experiments, and three reviewers for comments and suggestions. This work was supported by NASA, NSF and the Miller Institute.

References

Akaogi, M., and E. Ito, Heat capacity of $MgSiO_3$ perovskite, *Geophys. Res. Lett., 22,* 105-108, 1993.

Berman, B. R., *Thermal Conduction in Solids*, Oxford University Press, New York, 1976.

Bodea, S., and R. Jeanloz, Model calculations of the temperature distribution in the laser-heated diamond cell, *J. Appl. Phys., 65,* 4688-4692, 1989.

Fukao, Y., H. Mizutani, and S. Uyeda, Optical absorption spectra at high temperatures and radiative thermal conductivity of olivines, *Phys. Earth Planet. Int., 1,* 57-62, 1968.

Heinz, D., and R. Jeanloz, Temperature Measurements in the Laser-Heated Diamond Cell, in *High-Pressure Research in Mineral Physics*, edited by M. H. Manghnani and Y. Syono, Am. Geophys. Union, Washington, DC, 113-127, 1987a.

Heinz, D., and R. Jeanloz, Measurement of the melting curve of $(Mg,Fe)SiO_3$ at lower mantle conditions and its geophysical implications, *J. Geophys. Res., 92,* 11,437-11,444, 1987b.

Jeanloz, R., and A. Kavner, Melting criteria and imaging spectroradiometry in laser-heated diamond-cell experiments, *Phil. Trans. Roy. Soc. Lond., 354,* 1279-1305, 1996.

Li, X., and R. Jeanloz, Measurement of the B1-B2 transition pressure in NaCl at high temperatures, *Phys. Rev. B, 36,* 474-479, 1987.

Li, X., and R. Jeanloz, Laboratory studies of the electrical conductivity of silicate perovskites at high pressures and temperatures, *J. Geophys. Res., 95,* 5067-5078, 1990.

Manga, M., and R. Jeanloz, Vertical temperature gradients in the laser-heated diamond cell, *Geophys. Res. Lett., 23,* 1845-1848, 1996.

Ming, L. C., M. H. Manghnani, S. B. Qadri, E. F. Skelton, J. C. Jamieson, and J. Balogh, Gold as a reliable internal pressure calibrant at high temperatures, *J. Appl. Phys, 54,* 4390-4397, 1983.

Ming, L. C., M. H. Manghnani and J. Balogh, Resistive heating in the diamond-anvil cell under vacuum conditions, in *High-Pressure Research in Mineral Physics,* edited by M. H. Manghnani and Y. Syono, Am. Geophys.Union, Washington, DC, 69-74, 1987.

Mitra, S.S., Optical properties of nonmetallic solids for photon energies below the fundamental band gap, in *Handbook of Optical Constants of Solids II,* edited by E.D. Palik, Academic Press, New York, 213-270, 1985.

Osako, M., and E. Ito, Thermal diffusivity of MgSiO3 perovskite, *Geophys. Res. Lett., 18,* 239-242, 1991.

Williams, Q., E. Knittle and R. Jeanloz, A high-pressure melting curve of iron: a technical discussion, *J. Geophys. Res., 96,* 2171-2184, 1991.

Zel'dovich, Y. B., and Y. P. Raizer, *Physics of Shock Waves and High-Temperature Hydrodynamic Phenomena*, Vol. 1, Academic Press, New York, 1967.

Xiaoyuan Li, Department of Geology and Geophysics, University of California, Berkeley, CA 94720

Michael Manga, Department of Geological Sciences, University of Oregon, Eugene, OR 97403

Jeffrey H. Nguyen, Department of Physics, University of California, Berkeley, CA 94720

Raymond Jeanloz, Department of Geology and Geophysics, University of California, Berkeley, CA 94720

(Received February 26, 1996; revised July 8, 1996; accepted August 15, 1996.)