Laser-Heated Diamond Anvil Cell Technique: Double-Sided Heating with Multimode Nd:YAG Laser

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Abstract A new laser heating system has been set up to minimize the sample temperature gradients both radially and axially in a diamond anvil cell. A multimode Nd:YAG laser with a relatively flat top of intensity profile allows us to reduce the radial temperature gradient; the axial temperature gradient is minimized by heating samples at both sides. Three dimensional uniform temperatures of 1200 - 4000 K are obtained at high pressures in samples with 20 - 50 μm (radial) diameter and 10 - 50 μm (axial) thickness. The radial and axial variations and temporal fluctuations of temperatures in specified volumes are of similar order and within 1-4%, depending on the temperature range with higher temperature in larger temperature variations. This technique provides a relatively large sample volume in a more homogenous temperature distribution relative to previous techniques. This is especially useful for *in situ* X-ray studies at simultaneous high pressures and high temperatures.

Introduction

Laser heated diamond anvil cell has been widely used to study the behavior of materials under the condition of the Earth's deep interior [e.g., Heinz and Jeanloz, 1987; Boehler, 1993; Saxena et al., 1993; Shen and Lazor, 1995; Fiquet et al., 1995]. Quantitative results with laser heated diamond anvil cells rely on accurate temperature determinations of heated samples. The accuracy of temperature determination has suffered from the large temperature gradients in both radial and axial directions that are resulted from the Gaussian intensity distribution of the laser beam and the high thermal conductivity of diamonds. The large temperature gradients in a small hot spot make it difficult to accurately characterize the temperature distribution due to the fact that the size is comparable to the resolution of the optical system. Furthermore, the three dimensional temperature gradients have limited the study of bulk physical properties of materials at high pressures and temperatures. We introduce a laser heating technique that produces a relatively large hot spot and minimizes the sample temperature gradients both radially and axially in diamond anvil cells.

The technique is based on the use of a high power multimode Nd:YAG laser, which has a relatively flat of intensity profile at its peak. The combination of boxcar power distribution and the laser's high power makes it possible to heat samples evenly in an area of about 20 - 50 μm in diameter to temperatures above 4000 K with a significantly smaller radial temperature gradient than in previous techniques. On both sides of the sample, the laser is focused simultaneously and temperatures are measured from both sides. This arrangement allows us to heat relatively thick samples thoroughly and to minimize the axial temperature gradient.

Experimental Apparatus

The arrangement consists of a 100 W cw Nd:YAG laser (Quantronix, 118) with multimode output, an optical system to heat samples on both sides in a diamond anvil cell, a spectroradiometric system to measure temperatures, and a feedback system to stabilize temperatures in a heating area. The heating laser operates in continuous-wave multimode specified with a beam profile of a relatively flat top in intensity (Figure 1). Since the radial temperature distribution is directly related to the intensity distribution of the laser beam, using the multimode laser reduces the radial temperature gradient in the central potion of the laser heated spot. Moreover, the high power (>100 W) of the laser is able to heat samples in a large area without rastering.

A schematic diagram of the optical system is shown in Figure 2. The two optical paths guiding the laser beams and collecting radiations from the heated sample are almost identical on each side of the diamond anvil cell. The laser beam is reflected by a dichroic mirror (M1) which reflects the 1064 nm laser light and transmits the visible portion of the spectrum. Apochromatic objective lens (L1, f=60 mm) is used to focus the beam on to the sample. The thermal radiation of the hot sample is collected by the same apochromat and focused with an achromatic lens (L2, f=1000 mm) to an imaging optical fiber (Schott Fiber Optics Inc.). L1 and L2 provide a magnification of about 17x. The other end of the imaging fiber is placed at the entrance of an imaging spectroradiometric system for temperature measurement. The fiber bundle is a square array of 250x250 fibers of 10 μm diameter, the image within a square of 2.5 mm on the side is preserved and transmitted from one end of the bundle to the other. The spectroradiometric system consists of a thermodynamically cooled CCD detector (Princeton Instruments, TE/CCD) and a 275 mm radius imaging spectrograph (Acton, SpectroPro-275). The CCD chip has 256x1024 pixels, each measuring 27 μm across. An input optics assembly (Acton, FC459) at the entrance of the spectrograph allows a linear image of the sample to be reproduced on the CCD in linear the direction perpendicular to dispersion.

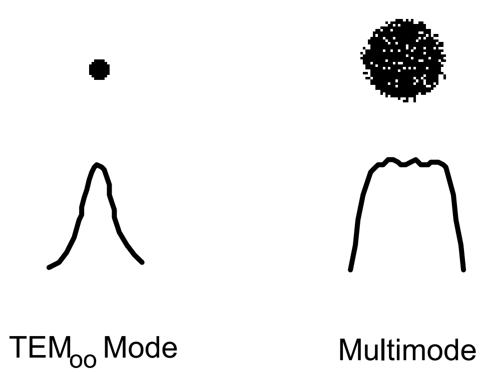

Figure 1. Upper: Beam cross section; middle: beam intensity profile; lower: laser mode. Laser beams with

TEM_{OO} mode are of Gaussian intensity profiles. A beam profile with a flat top in intensity is possible with multimode output.

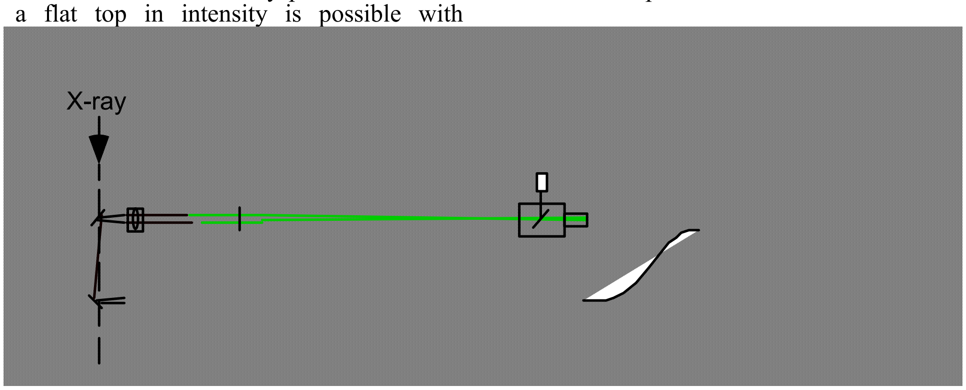

Figure 2. Diagram of the double sided laser heating system. L1: Apochromatic objective lens objective, 22 mm in diameter, 60 mm focal length; L2: achromatic lens, 50 mm in diameter, 1000 mm focal length; M1: dichroic mirror, 50 mm in diameter; M2: Gold coated mirror, 25 mm in diameter, 1 mm thick; F: shortpass filter, 25 mm in diameter; BS1: pellicle beamsplitter, 20% reflection; BS2: pellicle beamsplitter, 10% reflection. Feedback includes a photodiode and an electronic circuit connecting to the laser power supply. The image spectroradiometric system consists of an imaging optical fiber, imaging optics assembly, 275 mm spectrograph, two-dimensional CCD detector and computer that calculates temperatures from the spectra and stores the data. Synchrotron X-ray (dashed line) can be combined to make *in situ* X-ray diffraction measurements.

Because the intensity of the thermal radiation is proportional to the fourth power of the temperature, a fixed brightness indicates a steady temperature. Sample temperatures have been successfully controlled by stabilizing the light intensity from the heated sample [e.g., Heinz et al., 1991; Boehler and Chopelas, 1992]. We also chose to stabilize the intensity of the thermal radiation coming from the sample. A fraction (50%) of the thermal radiation is reflected to a photodiode (feedback in Figure 2) to control the laser power. The sample brightness can be maintained precisely for minutes with the system.

Heating and Temperature Measurement

Figure 3 shows the sample in a diamond cell being heated from both sides. The use of the heating laser with a boxcar power distribution reduces the radial temperature gradient over most of the heated area. When the temperatures of two sides are equalized, a uniform temperature distribution could be reached along the optical axis by the heat conduction in the sample. Polished single crystalline discs, compressed polycrystalline discs, and inert gases are used for medium/insulation materials. The discs with the same thickness are loaded at each side to assure a symmetric thermal conduction. Pressures are measured either by the ruby technique [Mao et al. 1978] or by P-V relation of standards when coupled with X-ray diffraction.

Factors affecting the efficiency of heating include the power density of the focused laser beam, the absorption of a sample, the thermal conductivity of the medium and the geometric dimensions of the medium and the sample. The multimode laser has a beam divergence of 5.5 mrad, which requires the focusing lens (f/2.7) to have a large numerical aperture to achieve a focused beam spot 50-70 μm in diameter. With this focus, the high power laser heats samples to more than 4000 K. Visibly opaque materials typically have strong absorption at 1064 nm. Silicates which do not contain transition metal ions are usually transparent at this wavelength. For these materials, it is necessary to mix absorbing materials, or to embed the silicate sample in two thin layers of metal foils such as platinum or rhenium. Sapphire, MgO, and alkali halide discs are good insulators to both protect the diamonds from damage and to reduce heat loss from the sample. Inert gases are of very low thermal conductivity, so that little laser power is needed to heat samples to sufficiently high temperatures.

In conventional one-sided heating experiments, it is usually necessary to load a thin sample to reduce the axial temperature gradient. Even with a thin sample the axial gradient is still not quantified or eliminated, so the relationship between sample thickness and axial temperature gradient is not known. With double sided heating, however, thick samples can be heated uniformly. The thickness of the sample is essentially controlled by the maximum pressure reached, as in ambient temperature studies. As an example, a polycrystalline disc (200x80 μm) of graphite was loaded in a diamond anvil cell at 15 - 18 GPa using thin MgO discs (< 5 μm thick) as the insulation material. When the heating laser was applied to both sides of the sample at the same time to about 2200 K, a transparent area of about 70 μm in diameter was immediately observed; on the other hand, the heating laser

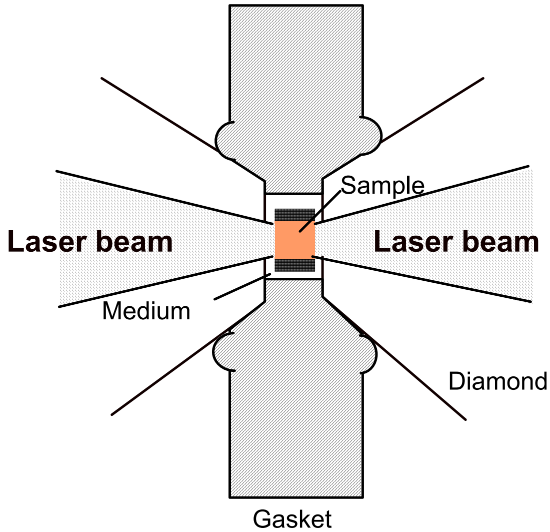

Figure 3. Detail of double sided laser heating of a sample in a diamond anvil cell.

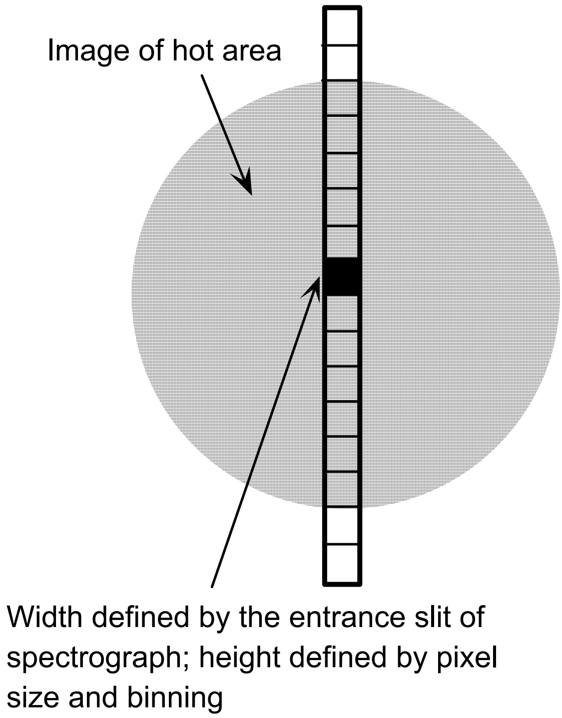

Figure 4. An image of a hot sample area on the two-dimensional CCD chip and the selection of regions to measure temperatures. A temperature profile in the vertical direction can be obtained for each measurement.

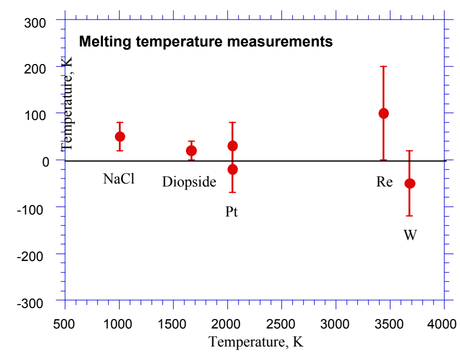

Figure 5. Melting temperature measurements of materials at ambient pressure with the spectroradiometric system. Sodium chloride (NaCl) and diopside samples were loaded in a hole with 1 mm in diameter of a flattened wire heater. For platinum (Pt), both wire heating and laser heating techniques were used. Melting temperatures by wire heating show slightly higher than those by laser heating. Rhenium (Re) was loaded in a diamond anvil cell with argon gas medium at 1 GPa. The melting was observed visually for all the above materials. For tungsten (W), tungsten halogen lamps were used; the highest recorded temperatures before filament breaking are presented. Error bars represent the scatter associated with repeated measurements.

was directed to only one side of the sample to the same temperatures, no visual change was observed. The X-ray diffraction and Raman spectra from the transparent sample area show that the graphite transformed to the diamond after heating. The thickness of the recovered sample disc was about 45 μm. This experiment qualitatively demonstrates that with the new technique, the axial temperature gradients can be efficiently reduced, and especially for thick samples.

Temperatures are determined by fitting the thermal radiation between 600 - 800 nm to the Planck radiation function, assuming constant emissivity with respect to wavelength. The imaging property of the system allows simultaneous measurements of all points on a temperature profile across a laser heated spot, rather than the measurement of only a small point, or an "average" temperature of a laser heated spot. As indicated in Figure 4, the entrance slit of the spectrograph is used to select a thin strip traversing the laser spot. Through the spectrograph, this linear strip is imaged onto the 256 (rows) x1024 (columns) pixels on the 2-dimensional CCD. Each row corresponds to a point of the strip with it thermal radiation wavelength spreading over each column. In this way the temperature profile across the hot spot is characterized at each measurement. Intensities measured by the CCD detector were calibrated with a tungsten ribbon lamp with known radiance, traceable to NIST standards.

A series of experiments was designed to verify the calibration of the measurement system over a wide range of temperatures. Temperature measurements were obtained at the melting points of a set of standard materials brought to melting with different means of heating. As shown in Figure 5, the measured melting temperatures are close to published values.

It is essential to reduce the chromatic aberration of the spectroradiometric system. However, because of the strong dispersion of the diamond windows, the optical system can not be totally free of chromatic aberration. The main source of error in the temperature estimate arises from the temperature gradient in a small hot spot. If temperature distribution is uniform, the uncertainty due to the chromatic aberration can be minimized by calibration. In order to check this, the melting temperature of platinum with wire heating was measured with a 6 mm thick silica glass plate between the platinum and the collecting lens. This glass created a severe chromatic aberration. The melting temperature was calculated to be 2380±25 K with a calibration without this 6 mm thick glass. However, when a calibration was made with the glass, this yielded a melting temperature of 2060±25 K, near the literature value. Unfortunately, It is difficult to locate standards with known temperature gradients comparable in degree and size with the laser heating spot. Without such a calibration, the temperature profile obtained with an existing temperature gradient may not represent true values due to the chromatic aberration and the possible lateral color effects. Therefore, it is important using multimode YAG laser that has a flat top of intensity beam profile to have a uniform heat on the sample and consequently to have an accurate temperature determination in laser heating experiments.

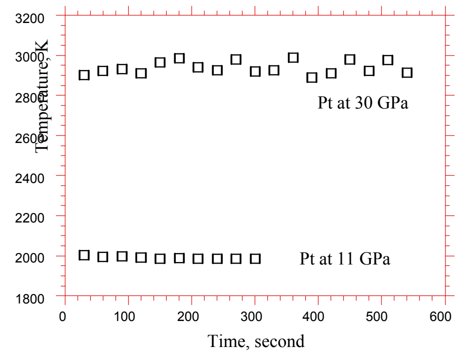

Figure 6. Temperatures with time measured on a platinum foil at high pressures with the feedback system. The integration time for each measurement is 1 and 0.2 second at 2000 K and 2950 K, respectively.

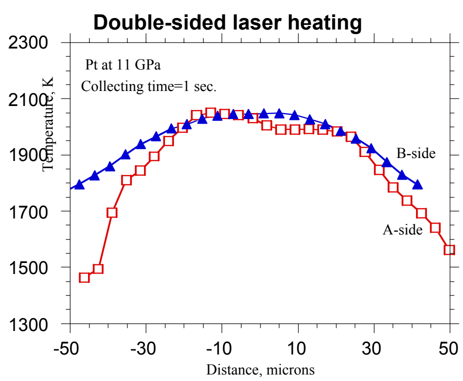

Figure 7. Temperature profiles measured from both sides of a Pt foil at 11 GPa. An even temperature distribution in a three dimensional volume (50 μm in diameter by about 30 μm thick) is obtained with the radial and axial temperature variations of less than 3%.

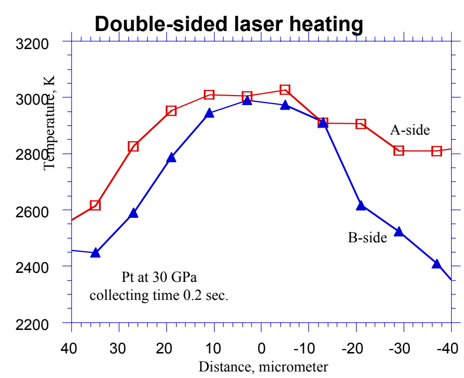

Figure 8. Temperature profiles measured from both sides of a Pt foil at 30 GPa. A volume of about 25 μm diameter and 15 μm thickness is heated to 3000 K with spatial temperature variations of about 3%.

As a consequence of slow laser power fluctuations (0.5 - 5 Hz) of up to 5%, it is necessary to maintain the sample brightness. To do this, signals from a photodiode which monitors the sample brightness are connected to an electronic feedback circuit which adjusts the laser power. The sample brightness is stabilized for minutes, sufficient time for temperature measurements and for synchrotron energy dispersive X-ray diffraction. Figure 6 shows the measured temperature as a function of time for a platinum sample at high pressures. At 2000 K, temperatures can be maintained with a constancy of 1% with respect to time. At 2950 K, this increases to about 3%.

The performance of the laser heating system was further tested by heating a platinum foil at high pressures in a cell. At temperatures around 2000 K, an area of about 50 μm in diameter could be created with a radial variation of less than 3% (Figure 7). To obtain higher temperatures at higher pressures, a laser beam with higher degree of focus is usually needed. For example, at around 3000 K, the radial temperature variation of less than 3% occurs in an area of about 25 μm in diameter (Figure 8). The axial temperature variation depends on the heating at each side. By regulating the input laser power, very close temperature values measured from both sides can be obtained (see Figures 7 and 8). The axial variation could be of similar magnitude as that in the radial direction. In this way, conditions approaching a uniform temperature in three dimensions is realized at high pressures in laser heated diamond anvil cells.

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