STUDIES ON MATERIALS UNDER ULTRAHIGH P-T AT THE ADVANCED PHOTON SOURCE

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ABSTRACT

The Advanced Photon Source (APS) is a third-generation synchrotron now in operation at Argonne National Laboratory. A national user facility for research in the Earth sciences is being constructed by the GeoSoilEnviro group of the Consortium for Advanced Radiation Sources (GSECARS). One component of this effort is the program for diamond anvil cell (DAC) research, to study materials at ultrahigh pressure and temperature conditions using a variety of techniques.

In this paper, we describe the DAC facility that is being developed at the GSECARS. Recent progress in developing a laser heating system is presented.

INTRODUCTION

In situ measurements with the intense, small x-ray beam of synchrotron sources provide definitive information on such fundamental properties of materials at extreme P-T conditions as structure, melting, and elasticity. Since ultrahigh pressures in the diamond anvil cell are achieved at the expense of reducing sample volume, characterization capability with high spatial resolution is fundamental for probing microscopic samples at high pressure and temperature and for minimizing the effect of pressure and temperature gradients. High pressure DAC experiments using x-ray techniques require the highest possible x-ray brilliance. APS, the new third generation synchrotron source, is thus ideal for micro-sized samples at high P-T. We describe the DAC program at GSECARS at the APS and report recent progress and developments. The high P-T experiments include:

- diffraction at high pressures and high temperatures with resistance and laser heating,
- single crystal and polycrystalline x-ray diffraction,
- elastic strain measurements,
- high resolution x-ray diffraction,
- hydrothermal reaction measurements,
- measurements of amorphous materials,
- three dimensional structural microprobe,
- time resolved x-ray diffraction,
- x-ray absorption spectroscopy,
- x-ray fluorescence microprobe, and
- x-ray Raman and inelastic scattering.

FACILITIES AND TECHNIQUES

Beamline components

The GSECARS sector consists of an undulator (13-ID) and a bending magnet (13-BM) beamline. Each beamline has two experimental stations, each of which is designed to be able to receive all kinds of beams (white, monochromatic, focused, and unfocused). The undulator is the standard APS undulator A which is a 3.3-cm-period device and is ideal for monochromatic diffraction experiments. When in tapered mode, the spectrum is smooth above 25 keV, and the on-axis brightness is greater than that derived from the standard APS wiggler to energies beyond 100 keV. This makes the tapered undulator an excellent source for energy dispersive (ED) diffraction experiments.

To exploit the new third generation source, several kinds of focusing mirrors are employed. A pair of bent flat mirrors in a Kirkpatrick-Baez (refereed to as 7:1KB) geometry will be installed, which demagnify the source by 10:1 to 3:1 depending on location in the experimental stations. When a beam size greater than 10 microns is acceptable, the 7:1KB mirror produces the advantage of larger flux. For smaller beam sizes, microfocusing KB optics will be employed. The small KB mirror system consists of 100-mm long Pt- or Rh- or Au-coated glass bent to an elliptical shape with a two-moment bender. In initial tests, a 70x70 μm collection area was focused to 3x9 μm focal spot (demagnification of 180:1) with an efficiency of greater than 90% for energies below 70 keV [1].

The incident beam is controlled by a pair of adjustable slits made from WC cubes. Beam sizes down to 10 μm can be obtained in this way. The automated slit system also serves to control the beam size delivered to the small focusing mirrors with which beams are focused down to less than 10 μm. A "clean-up" slit system is introduced to produce the "clean" x-ray beam required for very high pressure, low-Z materials, and amorphous scattering experiments.

<u>DAC stage</u>

The DAC stage is designed to have excellent stability and good resolution of motion. It is based on a two-circle horizontal diffractometer (Huber 440 and 480, 0.001^{o} angular resolution) mounted on a lift table which is adjustable in position, height and tilt with position accuracy of 0.5 μm (Figure 1). The lift table enables users to easily move to the different positions and directions required when the beam is changed from white to monochromatic or from focused to unfocused. The sample stage has motorized x-y-z translations and ω and χ rotations. The transitions have position resolution of 0.1 μm, and rotations have angular resolution of 0.001^{o}.

<u>Detectors</u>

Both energy dispersive (ED) and angle dispersive (AD) techniques are employed. An ED diffractometer for DAC experiments has been constructed [2]. ED is a well-established technique, and is fast and able to achieve excellent collimation and spatial selection. The latter is especially important to have good signal-to-noise ratios in high pressure studies to minimize background due to scattering from the diamond anvils and gaskets. In addition, ED allows use of white beam with the highest possible flux with focusing mirrors. These make ED suitable for exploring ultra high pressures, to study low-Z materials, and to perform fast collection and time resolved measurements. The excellent spatial selection also makes a three dimensional structural microprobe possible (e.g., to study small inclusions in diamonds). Although ED is limited by the intrinsically low energy resolution of the solid state detector ($\Delta E/E \sim 1\%$ FWHM), the peak positions are typically determined to an accuracy of $\Delta d/d<0.1\%$. Therefore, ED has been successfully used for determination of equations of state of known structures [3,4]. Peak broadness is a drawback in resolving overlapping peaks in polycrystalline diffraction. However, this is comparable or even smaller than the broadening from other sources such as nonhydrostatic stress and P-T gradients.

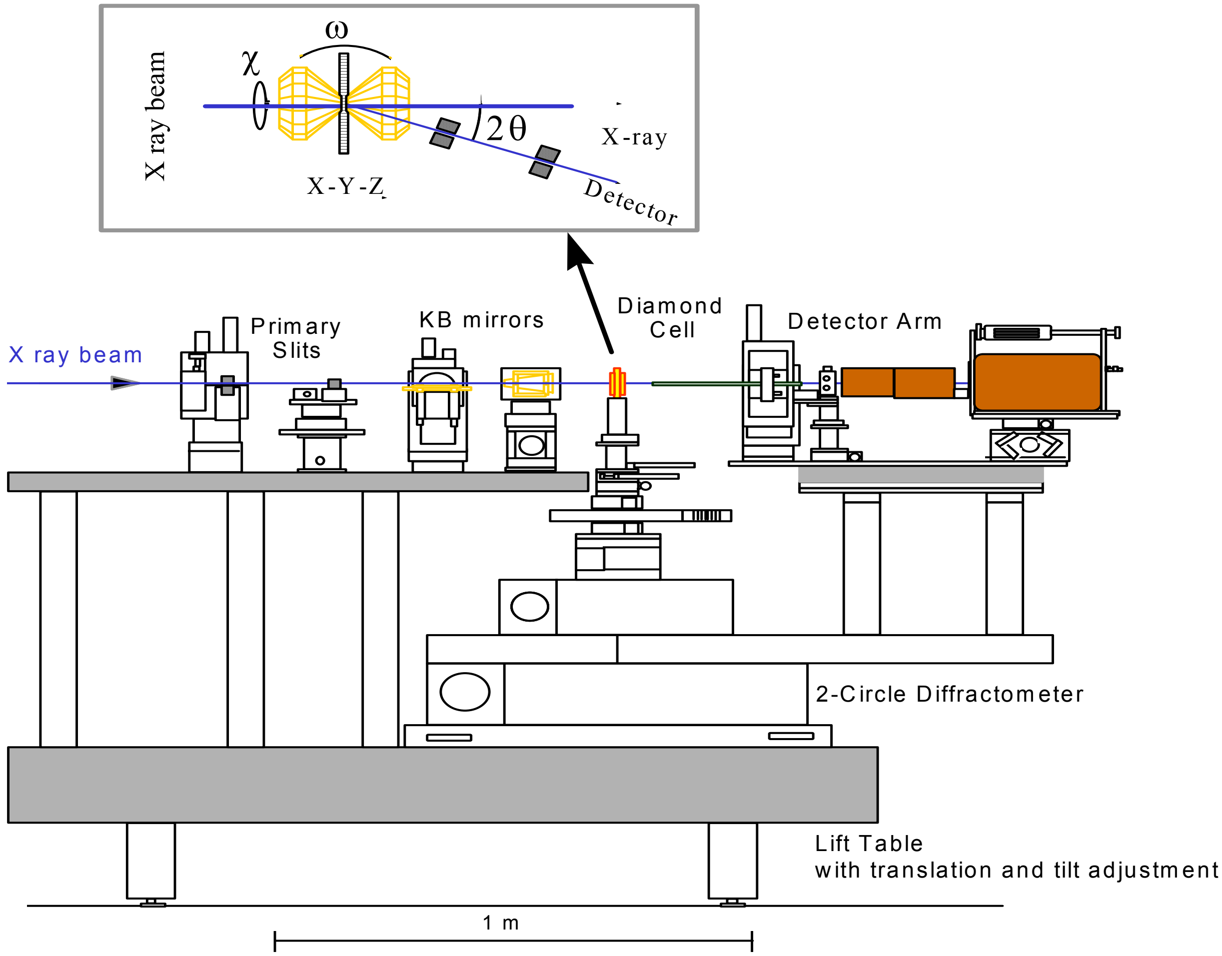


Figure 1. Setup of two circle diffractometer for diamond anvil cell experiments.

High resolution technique (AD) is only used in the cases where it is beneficial with the intensity information and/or the intrinsic resolution. The most reliable intensity data so far is from the area detector, as demonstrated by the imaging plate (IP) techniques [6]. Improving signal-to-noise ratio is a challenge, however, with area detecting techniques, especially for materials with low scattering power or at ultra high P-T conditions.

Conical diffraction geometry has been used with ED for DAC experiments [7] and it is well matched to the DAC geometry and synchrotron radiation conditions. Combining the conical diffraction geometry with a multi-element detector allows measurement of x-ray diffraction at different directions relative to the loading axis simultaneously, thus making it possible to monitor the elastic strain of a sample under uniaxial stress conditions without a need to rotate the cell to collect each direction. The conical geometry can also be combined with area detectors to reduce the background and improve the signal-to-noise ratio.

Laser heated DAC technique

DAC as a high pressure tool has been successfully coupled with different temperature techniques, with temperatures ranging from a few Kelvin to 6000 K with cryostat, external heating, internal wire heating, and laser heating techniques. At GSECARS, all these temperature techniques will be employed. A laser heated diamond cell system in connection with synchrotron x-ray has been recently set up and used successfully. The system is based on double-sided laser heating technique (Figure 2) and consists of two Nd:YLF lasers, optics to heat the sample from both sides, and spectroradiometric temperature measurement system.

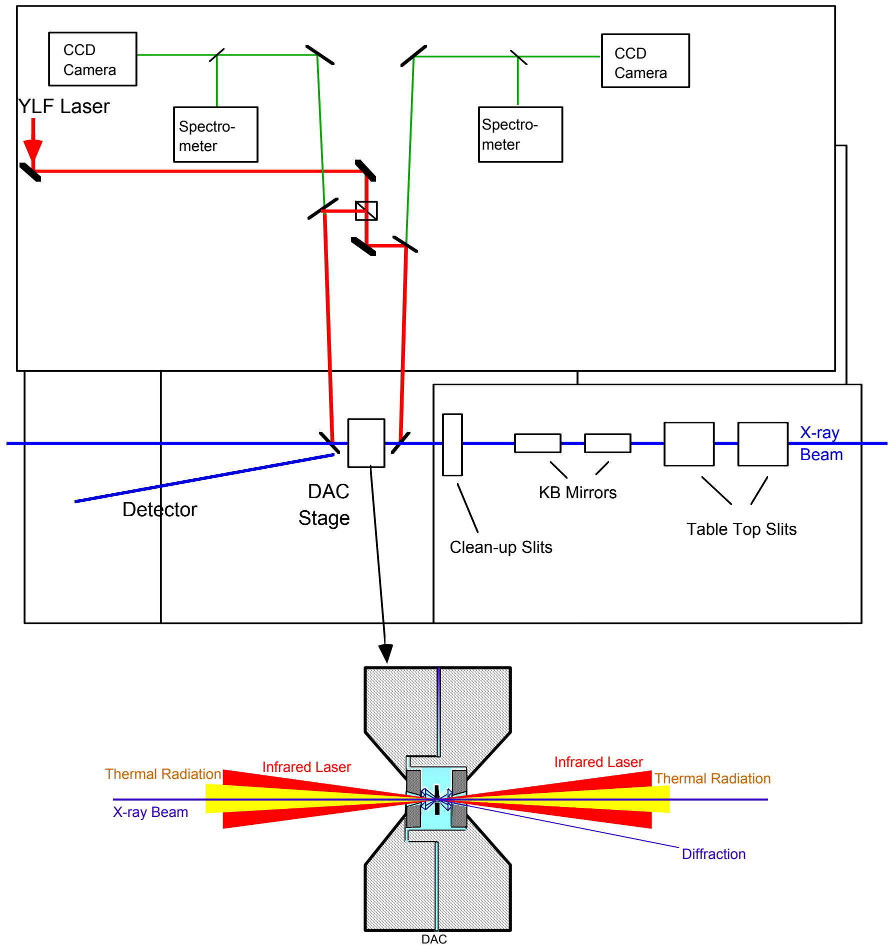
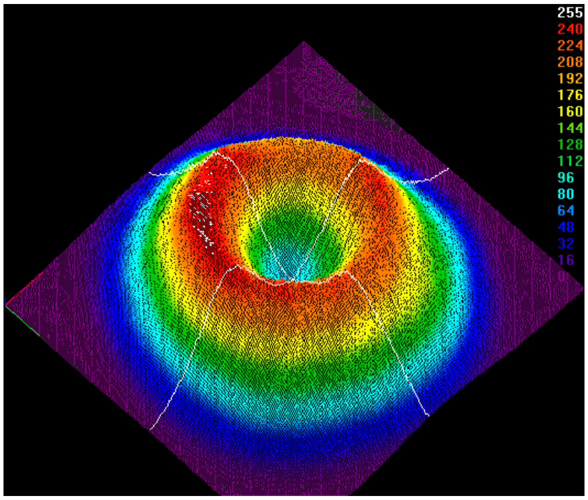


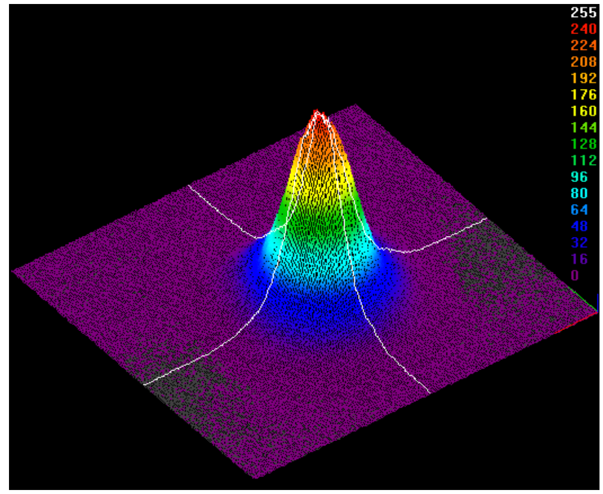
Figure 2. Schematic illustration of double-sided laser heating with diamond anvil cell. A symmetric cell is suitable for this application.

The CW Nd:YLF lasers can operate in multimode, TEM_{01}* (donut) mode, and TEM_{00} mode with output power of 92 W, 65 W and 55 W, respectively. The different beam modes make the system flexible in experimental setup. For instance, to achieve a uniform temperature distribution in a heated area, we combine two laser beam together with TEM_{01}* beam and TEM_{00} beam. Then the combined beam is split into two providing heating from two sides (Figure 2). As shown in Figure 3, an uniform and stable flat top in beam intensity can be reached in this way. The combination ratio of the two modes are adjusted according to the temperature profile measurements across the heated area in diamond anvil cells.

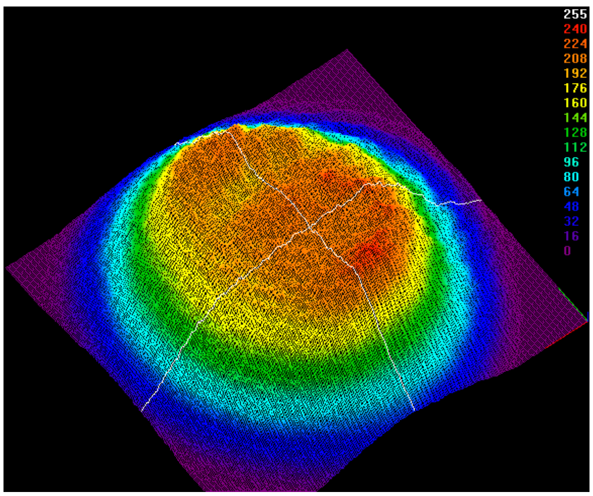
Optics and spectroradiometric temperature measurement system are based on the design of Shen et al [8]. The double sided heating technique (Figure 2) minimizes the axial temperature gradient, which is essential for reliable in situ structure determination. The whole system is on a 3'x6' breadboard. The breadboard is equipped with a kinematic base system, which allows us to move the whole laser heating system between the experimental station and the laboratory with minimal re-alignment.



(a)



(b)



(c)

Figure 3. Beam profiles in three dimension of Nd:YLF lasers at APS. (a) TEM_{01}* (donut) mode; (b) TEM_{00} Gaussian Mode; (c) TEM_{00} + TEM_{01}*.

<u>Support laboratory</u>

In additions to experimental stations, a high pressure support laboratory is being constructed with the sector laboratory space. This laboratory contains a variety of DACs, sample preparation and characterization facilities. Major facilities include a micro-Raman spectrometer, gas loading system, glove box, mechanical microdrill and an electric discharge machine. The existence of the laboratory makes it possible to carry out complete high pressure experiments at the APS, rather than relying on samples prepared at the home institution.

GSECARS

GSECARS sector is a national facility. The program is operated and enriched by the large user community. Beam time at the GSECARS sector will be assigned on the basis of competitive proposals and those proposals that take advantage of the unique characteristics of the third generation sources will receive the highest priority.

ACKNOWLEDGMENT

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