Development of *In Situ* Brillouin Spectroscopy at High Pressure and Temperature using Synchrotron Radiation and Infrared Laser Heating System: Application to the Earth's Deep Interior

One of the longstanding challenges in mineral physics has been the experimental determination of reliable acoustic wave velocities of deep-Earth materials under relevant high-pressure and high-temperature conditions, because the precise knowledge of sound velocity (elasticity) is essential for the interpretation of seismic observations and development of global seismological models of the Earth's interior. Considerable effort has thus been focused on the development of measurement techniques for use in high-pressure apparatuses to determine the sound velocities at high pressures and high temperatures. Recently developed and improved techniques include ultrasonic interferometry impulsive stimulated scattering, inelastic X-ray scattering and nuclear resonance inelastic X-ray scattering. However, there have been few studies on sound velocities under lower mantle conditions (above ~25 GPa in pressure and ~1900 K in temperature) because of the experimental difficulties.

A combined diamond anvil cell with Brillouin spectroscopy and synchrotron radiation X-ray diffraction enables the simultaneous measurements of sound velocities and materials density at a high pressure and temperature. This information allows us to determine elastic properties such as bulk and shear moduli, and their pressure and temperature derivatives. Simultaneous measurements under pressure and temperature conditions of the lower mantle and, in particular, achieving the high temperature, are, however, still a technical challenge. The application of resistive heating in a DAC, which is the conventionally used technique for the Brillouin method, normally works well below 10 GPa and 1000 K for simultaneous measurements [1]. A major disadvantage for resistive heating, which can produce temperatures of ~1500 K at most, is generating the higher temperatures of the lower mantle. An infrared laser heating technique, which can potentially generate temperatures over 3000 K at higher pressures, would be a more suitable alternative.

In order to meet all the requirements for simultaneous sound wave velocity measurements and sample characterization under lower mantle conditions, a Brillouin scattering measurement system that uses an infrared laser heating technique was recently installed at beamline **BL10XU** [2]. This system consists of three optical components used for Brillouin spectroscopy, X-ray diffraction and infrared laser heating (temperature measurement). For the simultaneous measurements, all optical probes for these three components must converge on the sample without optical and physical interference. The data obtained from each component is extracted by simultaneous and independent detector/analyzing systems. In this measurement system, a DAC mounted on a XYZ$\theta\chi\alpha\beta$ multiaxial stage is placed in the corner with a 150° angle of a pentagonal optical bench of a Brillouin measurement system (Fig. 1). An incident diode-pumped laser focused to ~20 μm in diameter is introduced into the sample and the scattered light is analyzed by a Fabry-Perot interferometer. A symmetric ~50° scattering angle is adopted in all experiments. The incident X-ray path in the experimental hutch is initially fixed. For simultaneous XRD measurements, the position of the DAC, which is aligned to the symmetric geometry in the optics for the Brillouin scattering measurements, must, therefore, be suitably adjusted to the X-ray independently of the Brillouin optics. For this purpose, the pentagonal optical bench used for Brillouin measurements is mounted on heavy-duty linear translation stages allowing vertical and horizontal motion (Figs. 1 and 2). This stage-scanning system enables a precise search of the exact position of the ~20 μm collimated X-ray beam and the adjustment of the sample to the X-ray position with a resolution

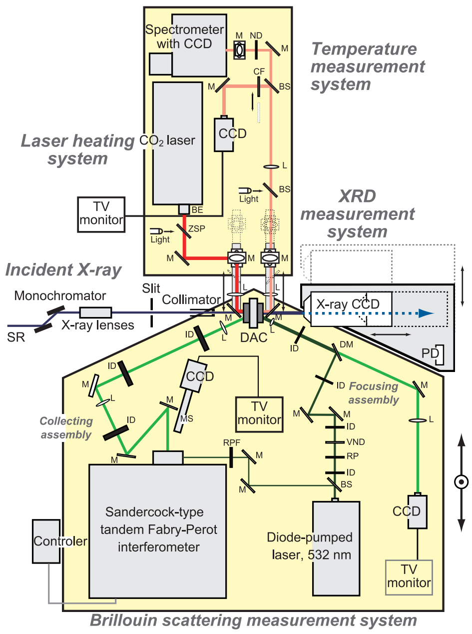

Fig. 1. Schematic layout of the Brillouin scattering measurement system combined with synchrotron radiation and infrared laser heating system of BL10XU beamline.

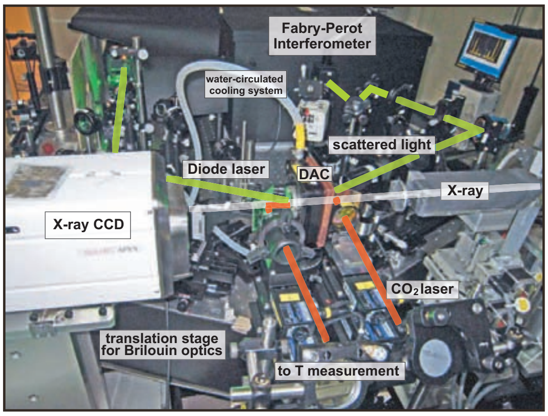

Fig. 2. View of the whole Brillouin scattering measurement system combined with synchrotron X-ray diffraction and laser heating systems at BL10XU. Green, white and red lines indicate the schematic optical paths for Brillouin scattering measurement, X-ray diffraction measurement and the laser heating system, respectively.

better than ~3 µm. A transparent sample is required for high-quality Brillouin spectra. Therefore, a carbon dioxide (CO_2) laser with a 10.59 µm wavelength is employed for the laser heating system, which can heat the transparent (colorless) oxide and silicate materials.

To evaluate the system performance and the potential capacity of this *in situ* Brillouin scattering measurement system, experiments were performed on polycrystalline, single-crystal and fluid-phase samples at high pressure and high temperature. Here we demonstrate experiments on polycrystalline MgO as a proxy of dominant mantle minerals. The experimental configuration of the DAC in this experiment is almost the same as that described in Ref. [3]. The sample mixture, placed in a rhenium gasket hole (100 µm in size), was compressed at a pressure of 45 GPa using 300 µm culet diamond anvils to, and then heated at ~2300 K for 2.5 hours to carry out Brillouin scattering measurements during heating. The sample position, once adjusted for the Brillouin scattering and X-ray diffraction measurements and CO_2 laser heating, was stable owing to the water-circulated cooling jacket used for the DAC. The size of the heating spot emitted by the visible radiation in the present experiment was a substantial proportion of the whole sample, as shown in Fig. 3. The Brillouin spectrum of polycrystalline MgO at 49 GPa and ~2300 K is shown in Fig. 4. We can recognize the shear acoustic modes of MgO and the longitudinal acoustic modes of NaCl together with the shear acoustic modes of diamond.

The infrared laser heating system is found to be highly suitable for use with this combined measurement system. Preliminary results in the present study indicate that a large variety of materials

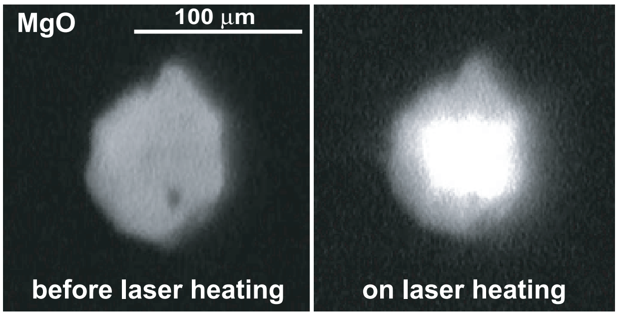

Fig. 3. Photomicrographs of polycrystalline MgO in a DAC before laser heating (at 45 GPa) and during laser heating (at ~2300 K).

such as oxides, glasses and fluids can be investigated regardless of their form (polycrystalline or single-crystal) at pressures up to 1 Mbar and temperatures up to 2300 K using our system. The present measurement system is thus a very powerful tool for providing direct information on acoustic sound velocity and elasticity data, and resolving remaining issues in mineral physics.

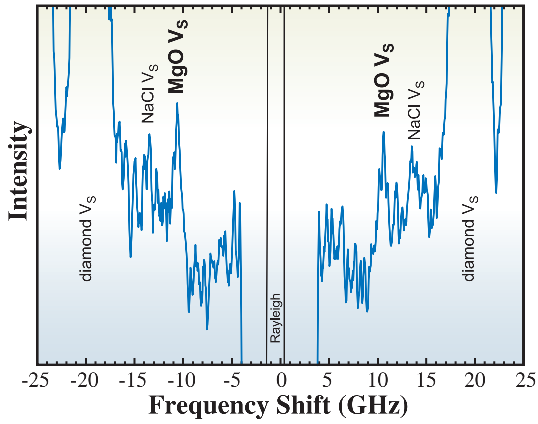

Fig. 4. Brillouin spectrum of polycrystalline MgO at 49 GPa and ~2300 K.

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References

[1] S.V. Sinogeikin *et al.*: Rev. Sci. Instrum. **77** (2006) 103905.
[2] *M. Murakami, Y. Asahara, Y. Ohishi, N. Hirao and K. Hirose: to be published in Phys. Earth Planet. Int. (2008).*
[3] M. Murakami *et al.*: Earth Planet. Sci. Lett. **259** (2007) 18.