Precise Determination of Elastic Constants by Inelastic X-Ray Scattering

Precise measurement of the elastic properties of materials in extreme conditions is crucial for both materials and earth sciences. However, it is difficult to measure elastic velocities on small and/or optically opaque materials, especially when they are contained within complex sample environments. In earth science, for example, seismic wave observations are an important source of information about the earth's interior: in principle, the thermal and chemical structure of the earth may be investigated by comparing measured seismic velocities with the elastic properties of candidate materials measured in the lab in controlled environments. However, conventional techniques such as ultrasound interferometry (UI) and Brillouin light scattering (BLS) are not easily applied, as UI usually requires large sample volumes and BLS requires optically transparent samples.

High resolution inelastic X-ray scattering (IXS) is a technique that may be used to determine elastic

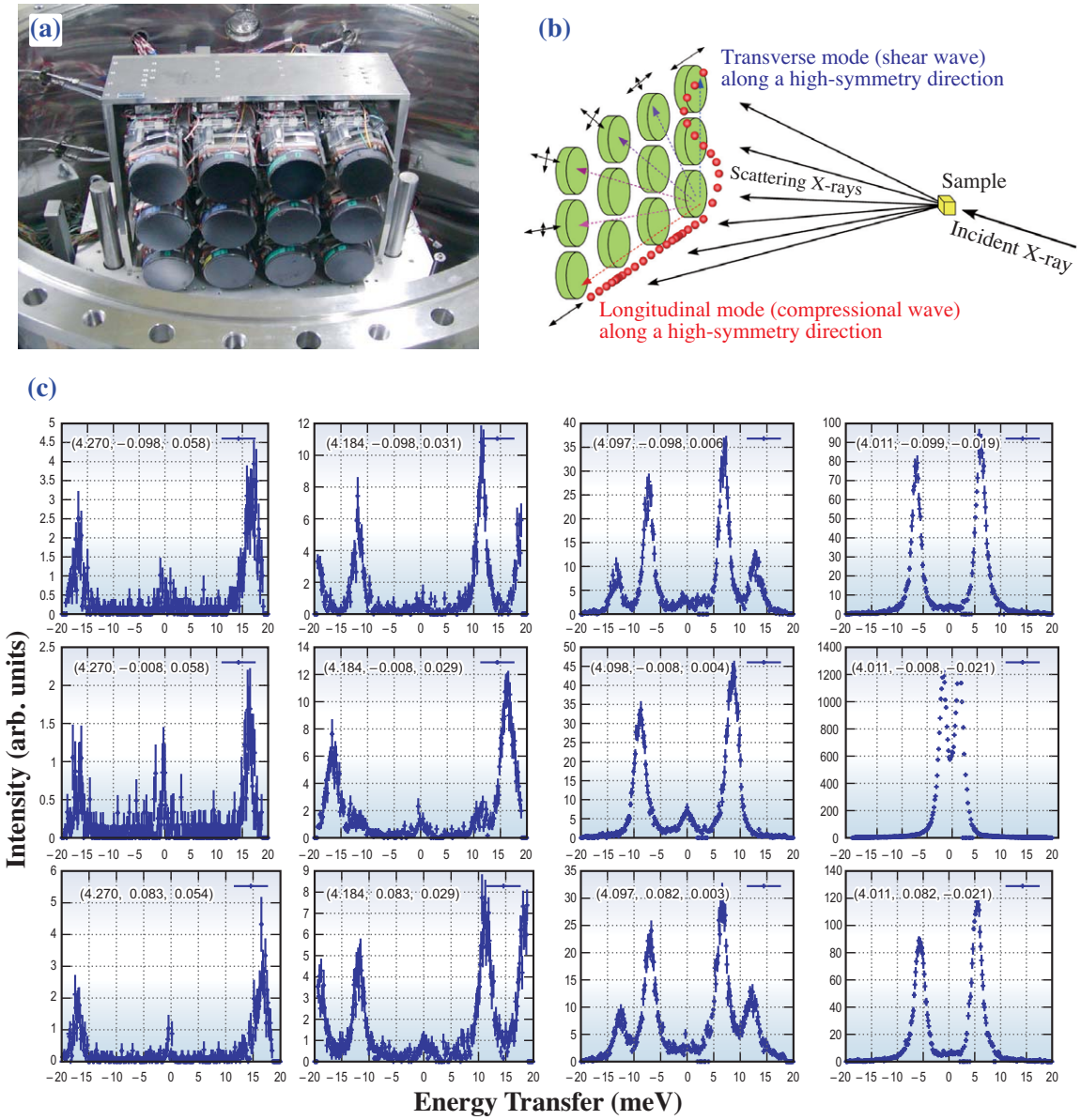

Fig. 1. **(a)** Photograph of the two-dimensional analyzer array. **(b)** Schematic drawing of detectable phonons at one scattering position. Colored broken arrows indicate phonon propagation direction and thin black arrows the detectable phonon polarizations. **(c)** Representative IXS spectra obtained using the 12-analyzer array. The reciprocal lattice coordinate for each analyzer is shown in the figure, and the Bragg point would be just off the figure on the right-hand side.

constants from small opaque samples under extreme conditions: the measured acoustic phonon velocity, as wavenumber approaches zero, corresponds to that of the elastic wave. As the ~20 keV X-rays used in IXS are penetrating and can be focused to a few micron size, they are an interesting probe of elastic constants. This has been demonstrated by several groups with the uncertainties around 3% in bulk modulus. However, to maximize utility, the precision of the determination should be improved. In the case of (Mg,Fe)O, for example, uncertainties of 3% in sound velocity and bulk modulus corresponds to uncertainties of 450 K in temperature and 17% in iron content, respectively. We have achieved the precision of 0.4% in bulk modulus using special characteristics of beamline **BL35XU** and a new method of analysis [1].

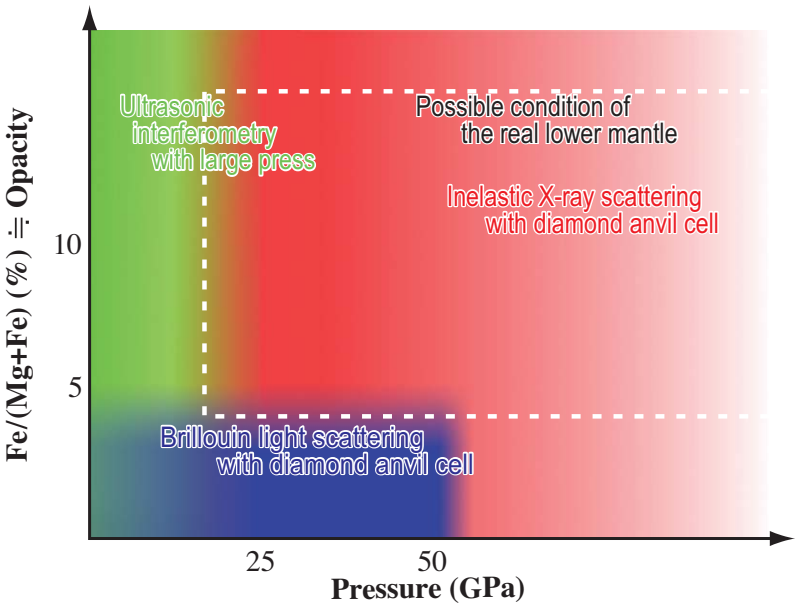

Fig. 2. Regions in pressure-opacity (iron content) space where elastic constants of a single crystal can be measured by UI and large press with a solid pressure medium (green), BLS and DAC (blue), and IXS and DAC (red). White broken lines indicate possible conditions of the real lower mantle. The pressure region for UI with large press would be limited because a single crystal may be broken by a buffer rod and a pressure medium at higher pressure. In the case of BLS with DAC, signals from longitudinal velocities might be covered by that from diamond anvils at higher pressure. This technique is difficult to apply to opaque materials. Since IXS with DAC does not suffer from those, the region is potentially the widest.

A single crystal of magnesium oxide, a known material, was used to characterize our method. MgO, being cubic, has only 3 independent elastic constants, and therefore it is sufficient, in principle, to measure sound velocities along limited high symmetry directions, e.g. [1 0 0] (longitudinal only) and [1 1 0] directions (longitudinal and transverse). However, redundancy is highly desirable to reduce errors. The unique two-dimensional analyzer array at BL35XU [2] allows simultaneous collection of data at twelve *k* points, clustered around a central momentum transfer, making the system well matched to this problem (Fig. 1). Elastic constants were obtained by fitting Christoffel's equation [3] to measured phonon energies at *arbitrary k* points. The redundancy and quality of the data allowed isolation of a correction term to the alignment of $(\Delta H\ \Delta K\ \Delta L) = (0.0056 \pm 0.0005, -0.0080 \pm 0.0007, 0.0043 \pm 0.0012)$ – the importance of this correction (neglecting it leads to an error of 1.9% in bulk modulus) demonstrates the sensitivity of the method. The elastic constants determined in this analysis were 293.9±1.0, 95.2±0.2, and 154.9±0.2 GPa for C_{11}, C_{12}, and C_{44} respectively, consistent with those measured by other techniques. The uncertainties are much better than 1%, and about one order of magnitude better than previously published IXS work.

This technique is now being applied to materials under extreme conditions. Here, the combination of IXS and diamond anvil cells (DACs) can cover the widest area for single crystal samples in pressure-composition (or opacity) space (See Fig. 2). However, one should note that the sample thickness in a DAC is only a few percent of the 1 mm thickness used in the present test. Thus the efficiency and redundancy of the multi-analyzer array becomes crucial for high pressures, and efforts should be made to maximize the flux onto the sample.

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