Ultrahigh-pressure Synchrotron Radiation ^{57}Fe-Mössbauer Spectroscopy using Single-line Pure Nuclear Bragg Reflection

Pure nuclear Bragg reflection of a $^{57}FeBO_3$ single crystal at the Néel temperature can select single-line ^{57}Fe Mössbauer radiation from synchrotron radiation (SR) [1,2]. This fully recoilless single-line is a very attractive probe beam for various applied researches using energy-domain SR Mössbauer spectroscopy because it has high directivity, small beam size, and pure linear polarization. In particular, SR Mössbauer spectroscopy using single-line pure nuclear Bragg reflection combined with a focusing X-ray optics has enabled us to easily achieve micron-scale small-target researches. As a typical example of such experiment, the Mössbauer transmission spectra were observed for polycrystalline iron metal and hematite under multimegabar pressures (> 200 GPa) in a diamond anvil cell (DAC) for the first time [3].

The nuclear diffraction optics for ultrahigh-pressure SR Mössbauer spectroscopy is shown in Fig. 1. The experiments were performed at beamline **BL11XU** (JAEA). A σ-polarized X-rays with an energy width of 2.5 meV at 14.4 keV nuclear resonance of ^{57}Fe were produced by a liquid-nitrogen-cooled Si (111) double-crystal pre-monochromator and a high-resolution monochromator (HRM). A bent elliptical multilayer mirror focused the incidence X-rays were horizontally with a focal size of 400 × 36 μm^2 at the sample position. Downstream of a Doppler vibrating sample mounted on a velocity transducer, the transmission X-rays were ultimately monochromatized into a 15.4 neV bandwidth by the electronically forbidden (333) pure nuclear Bragg reflection of a $^{57}FeBO_3$ single crystal near the Néel temperature (75.8°C) in a 150 Oe external magnetic field [2]. Behind a slit, the nuclear Bragg diffracted X-rays were detected by a NaI(Tl) scintillation detector. The peak photon counting rate of $^{57}FeBO_3$ (333) reflection was 2.4 × 10^3 cps, and the noise level was below 8.0%. In this optics, the Mössbauer spectrum was measured by counting the intensity of single-line nuclear Bragg reflection as a function of velocity. The measurement scheme is shown in Fig. 2.

As the first demonstration of ultrahigh pressure SR Mössbauer spectroscopy, a micron-sized polycrystalline iron metal (^{57}Fe 95%) placed in a DAC was measured in the multimegabar range. The specimen and small platinum (Pt) chip were enclosed in a hole of 18 μm diameter in a rhenium (Re) gasket between the flat parallel faces of two oppositely facing diamond anvils, without a pressure medium. The pressure was estimated at 252 GPa from the X-ray diffraction profile of Pt. The Mössbauer spectrum was observed using a vibrating DAC mounted on a velocity transducer at sample position of Fig. 1. The result is shown in Fig. 3.

The statistically sufficient spectrum of a polycrystalline iron metal under the ultrahigh pressure of 252 GPa was obtained in a short measurement time of 2.0 h. It clearly showed a single-line absorption profile, which was a typical Mössbauer spectrum of paramagnetic α-Fe. As for the quadrupole interaction of α-Fe, no significant change was observed in the spectrum. However, the isomer

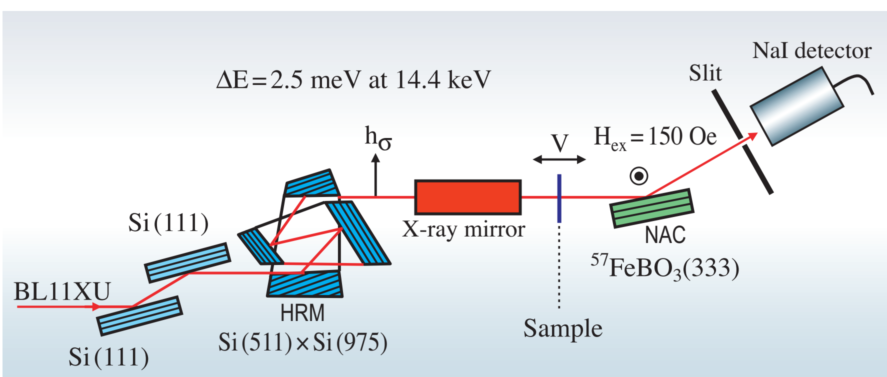

Fig .1. Optics for the energy domain SR Mössbauer spectroscopy using single-line pure nuclear Bragg scattering. (HRM: high-energy-resolution monochromator; NAC: nuclear analyzer crystal).

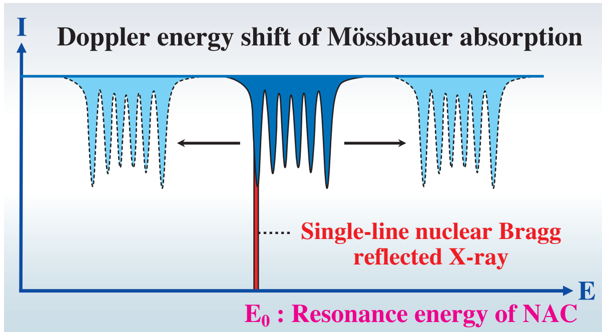

Fig. 2. Conceptual diagram for the energy domain SR Mössbauer spectroscopy with single-line pure nuclear Bragg scattering.

shift showed a marked energy shift (-0.85 mm/s) at 252 GPa. This is attributed to the considerable increase in the s-electron charge density at the ^{57}Fe nucleus owing to the decrease in atomic volume at 252 GPa.

Figure 4 shows the high-pressure Mössbauer spectra of a hematite (^{57}Fe 95%) polycrystal measured in the multimegabar range. The measured spectra show the clear dependence of hyperfine magnetic field and electric quadrupole splitting under pressure. This method will become a powerful tool for high-pressure science and geophysics.

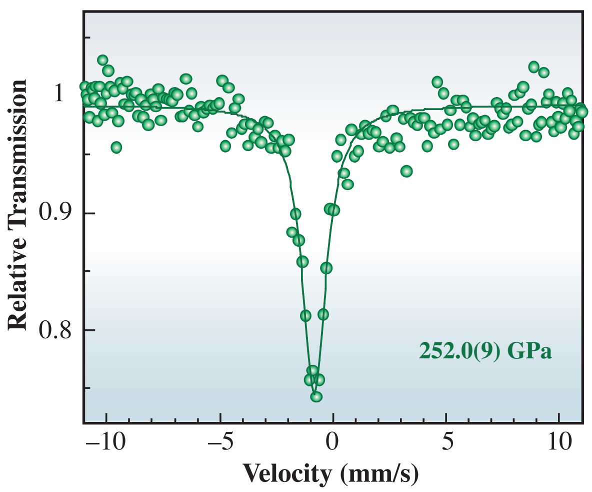

Fig. 3. SR Mössbauer spectrum of a polycrystalline iron metal (^{57}Fe 95%) in DAC at ultrahigh pressure of 252 GPa. The solid line is fit with Lorenzian line.

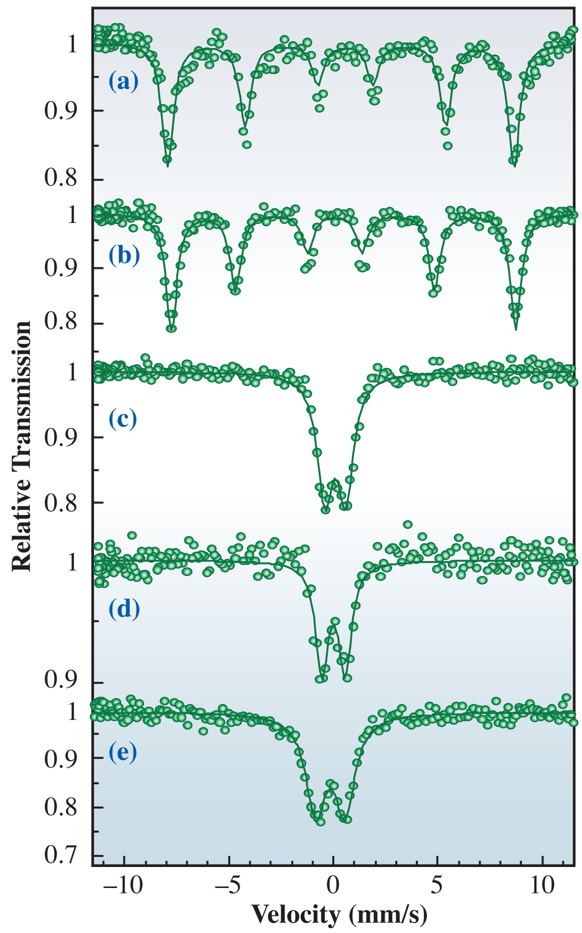

Fig. 4. SR Mössbauer spectra of $^{57}Fe_2O_3$ at different pressure conditions. **(a)** 0 GPa, **(b)** 43 GPa, **(c)** 91 GPa, **(d)** 121 GPa and **(e)** 204 GPa. The solid lines are fits with Lorenzian lines.

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