Hyperfine Interact
DOI 10.1007/s10751-011-0488-0

Synchrotron radiation ^{57}Fe-Mössbauer spectroscopy using nuclear monochromator

Takaya Mitsui · Ryo Masuda · Naohisa Hirao · Ko Mibu · Makoto Seto

Abstract A synchrotron radiation (SR) Mössbauer spectrometer has been developed by using a nuclear Bragg monochromator (NBM). It enables energy-domain ^{57}Fe SR-Mössbauer spectroscopy (SRMS). Doppler-shifted SR-Mössbauer radiation is produced by pure nuclear Bragg reflection (PNBR) from an oscillating $^{57}FeBO_3$ crystal near the Néel point. In this paper, we describe the optics and its applications, including high-pressure SRMS, grazing incidence SRMS and γ-ray diffraction studies with the Rayleigh scattering of Mössbauer radiation (RSMR) method.

Keywords Synchrotron radiation · Mössbauer effect · Nuclear resonant scattering · Bragg scattering

1 Introduction

To bring a marked progress in Mössbauer spectroscopy (MS), we have successfully developed a SR-^{57}Fe-Mössbauer spectrometer with a NBM, which yields high-brilliant SR-^{57}Fe-Mössbauer radiation of about three natural linewidths (See Fig. 1) [1–3]. This paper reports on the optics and applications.

T. Mitsui (✉) · R. Masuda · M. Seto
Japan Atomic Energy Agency, Sayo-cho, Sayo-gun, Hyogo 679–5148, Japan
e-mail: taka@spring8.or.jp

N. Hirao
Japan Synchrotron Radiation Research Institute, 1–1–1 Kouto, Sayo-cho, Sayo-gun, Hyogo 679–5198, Japan

K. Mibu
Nagoya Institute of Technology, Gokiso-cho, Showa-ku, Nagoya, Aichi 466–8555, Japan

Makoto Seto
Research Reactor Institute, Kyoto University, Kumatori, Osaka 590–0494, Japan

Published online: 07 December 2011

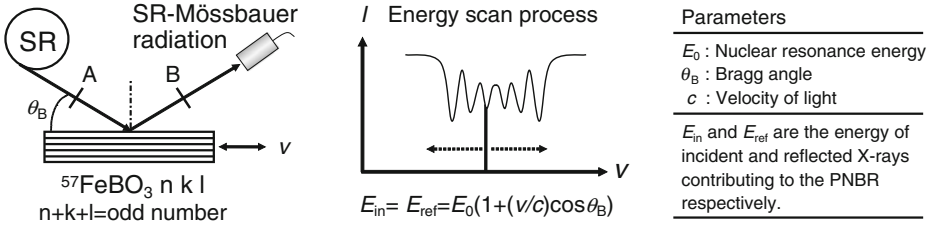

Fig. 1 Experimental scheme of ^{57}Fe-SRMS with single-line pure nuclear Bragg reflection. A PNBR from $^{57}FeBO_3$ (95% ^{57}Fe), oscillating parallel to the reflection plane, filters a Doppler-shifted ^{57}Fe-Mössbauer radiation from SR X-rays and emits at a fixed beam position. An extremely narrow-bandwidth (∼neV) is achieved by PNBR near the Néel point. Then, if a sample is placed at position A or B, the Mössbauer spectrum is measured by counting the reflected radiation as a function of velocity (v)

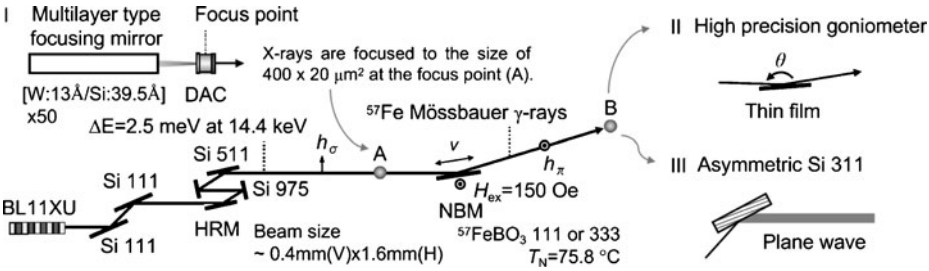

Fig. 2 Optical system for SR-^{57}Fe-Mössbauer spectrometer at SPring-8 (BL11XU). Optical devices (I, II and III) are used for high-pressure study with a diamond anvil cell (DAC), grazing incidence MS (GIMS) and γ-ray diffraction study, respectively. In this optics, NBM converts the σ-polarized SR X-rays into the π-polarized γ-rays

2 Optics and applications

Figure 2 shows the optics of the ^{57}Fe-SRMS spectrometer at SPring-8 (BL11XU) [3]. X-rays with a bandwidth of 2.5 meV at 14.4 keV, produced by a high resolution monochromator (HRM), are ultrafinely monochromatized to a 15.4 neV bandwidth by PNBR from a heated $^{57}FeBO_3$ crystal in $H_{ex} = 150$ Oe. The resonance energy is varied as shown in Fig. 1. The counting rate is $\sim 2.0 \times 10^4$ Hz for $^{57}FeBO_3$ 111 ($\sim 1.2 \times 10^4$ Hz for $^{57}FeBO_3$ 333).

As a transmission experiment, the SRMS of FeH was studied under high pressures up to 65 GPa at 300 K [4]. A tiny polycrystalline iron metal (^{57}Fe 95%, ϕ <50 μm) and ruby pressure markers were enclosed in a DAC filled with liq. H_2. By using a focusing optics (Fig. 2), a good quality spectrum was obtained in a rather short time (2–5 h), whose results proved a great advantage in high pressure SRMS using a NBM in comparison with conventional MS using a ^{57}Co source (See Fig. 3). The spectra show that ferromagnetic dhcp-FeH, formed at $P > 3.5$ GPa, changes into a non-magnetic high-pressure phase ($P > 32.0$ GPa). The origin is the pressure-induced 3d band broadening; it reduces the density of states at the Fermi level $D(E_f)$ so that the Stoner criterion for ferromagnetism is unsatisfied.

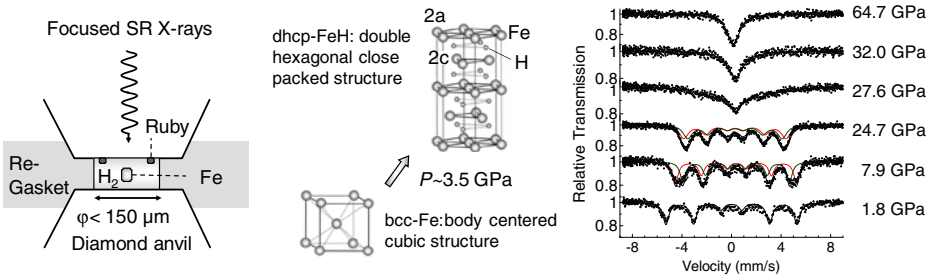

Fig. 3 Hydrogenation of Fe in a DAC and the MS spectra of FeH at high pressures up to 65 GPa. At $P = 7.9$ GPa, the spectrum shows two magnetic sextets due to two different Fe sites (2a, 2c) in a dhcp-FeH. The origin of the ferromagnetic state is the volume expansion and increase in $3d$ electron occupation due to the H_2 absorption, leading to the increase in $D(E_f)$ to satisfy the Stoner criterion. In contrast, at $P > 32.0$ GPa, a ferromagnetic-nonmagnetic transition is caused by the volume reduction, leading to the decrease in $D(E_f)$

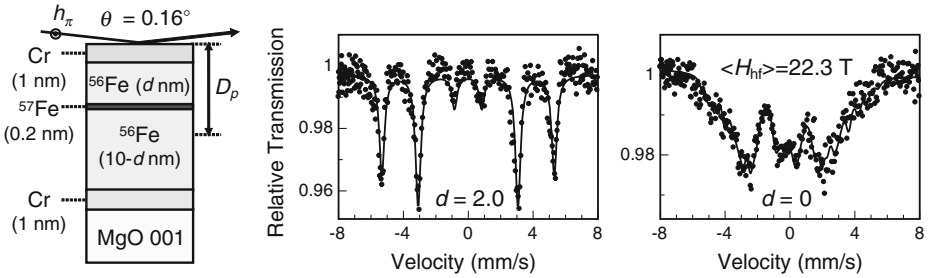

Fig. 4 A principal structure of the prepared Cr/Fe thin films and the observed GIMS spectra. Under the conditions of electronic total external reflection, the penetration depth D_p of X-rays is a few nm, which is deeper than the ^{57}Fe probe layer position in the films. Then, the reflectivity is dominated by the contribution of much larger electronic scattering; the nuclear scattering contribution becomes negligible due to the relatively low ^{57}Fe content. As the result, the GIMS spectrum shows a MS absorption profile

In a grazing-incidence experiment, SRMS of a Cr/Fe film, with 1 monolayer (ML) ^{57}Fe probe atoms, was performed at 300 K without external field. Figure 4 shows the prepared Cr/Fe multilayer structures. The spectra of two different probe layer depths d (2.0 and 0.0 nm) were measured with a data acquisition time of 4.0 h. The probe beam was incident on the films at an angle $\theta = 0.16°$, below the critical angle of electronic total reflection. Then, the GIMS spectrum gives a usual absorption profile as explained in Fig. 4. In fact, the spectrum for $d = 2.0$ nm shows a magnetic sextet of bulk α-Fe (H_{hf} ~33T). In contrast, the result for $d = 0$ nm shows a broad hyperfine field distribution, whose average value $< H_{hf} >$ is less than that of $d = 2.0$ nm; the Cr/Fe interface is composed of flat regions separated by step sites on a scale of a few ML so that the magnetic hyperfine field at the ^{57}Fe nuclei is reduced upon an increase of neighboring Cr atoms [5]. The result verifies that the GIMS, combined with the probe layer method, is a useful tool for thin film study with atomic-scale resolution.

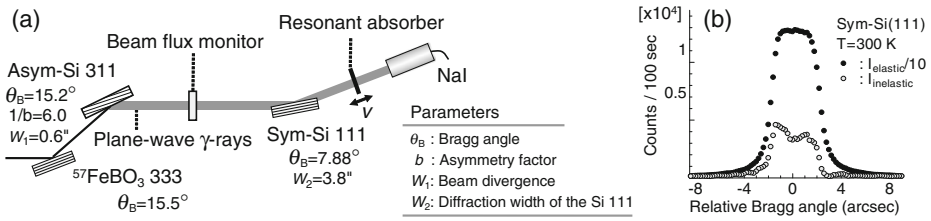

Fig. 5 **a** Experimental setup. **b** Elastic and inelastic scattering at a Si 111 reflection. The beam flux is monitored by a transparent type avalanche photo diode detector (transmittance ∼90%). The resonant absorber of a mixture of fluoferrates exhibits a wide line of about 50 neV

As a γ-ray diffraction study, the thermal diffuse scattering (TDS) was measured at a Si Bragg reflection. The plane-wave γ-rays were produced by asymmetric Si 311, forming a parallel (±) setting with $^{57}FeBO_3$ 333. The beam divergence $W_1 = 0.6''$ was much smaller than the diffraction width $W_2 = 3.8''$ of symmetric Si 111. In Fig. 5a, the Si 111 rocking curves were measured with on and off resonant conditions by a "Black" resonant absorber with a line width of ∼50 neV. The elastic and inelastic (TDS) parts were determined according to ref. [6]. In Fig. 5b, the TDS curve, increasing near the Bragg angle, shows a small dip in the total reflection region for dynamical Bragg diffraction. The dip is a result of energy conservation; the reflected beam is quite strong and consequently little energy enters the crystal to produce TDS [7]. The result proves that the SR-based RSMR enables the TDS study at the Bragg peaks in a high angular resolution ($\sim 10^{-6}$ rad).

3 Conclusion

Today high-brilliant ^{57}Fe γ-rays are available at any bunch-mode of SR. This contrasts with time domain ^{57}Fe-SRMS, requiring a bunch-period over 100 ns. Energy domain ^{57}Fe-SRMS enters a new stage in both basic and practical studies.

Acknowledgements This work was supported by JST/CREST and partially by NEDO (Hydro-Star).

References

1. Chumakov, A.I., Zelepukhin, M. V. , Smirnov, G. V. , van Bürck, U., Rüffer, R., Hollatz, R., Rüter, H.D., Gerdau, E.: Phys. Rev. B **41**, 9545–1547 (1990)
2. Smirnov, G.V., van Bürck, U., Chumakov, A.I., Baron, A.Q.R., Rüffer, R.: Phys. Rev. B **55**, 5811–5815 (1997)
3. Mitsui, T., Hirao, N., Ohishi, Y., Masuda, R., Nakamura, Y., Enoki, H., Sakaki, K., Seto, M.: J. Synchrotron Radiat. **16**, 723–729 (2009)
4. Mitsui, T., Hirao, N.: Mater. Res. Soc. Symp. Proc. **1262**, W06–09 (2010)
5. Landes, J., Sauer, Ch., Brand, R.A., Zinn, W., Mantl, S., Kajcsos, Zs.: J. Magn. Magn. Mater. **86**, 71–77 (1990)
6. O'Connor, D.A., Butt, N.M.: Phys. Lett. **7**, 233–235 (1963)
7. Annaka, S., Kikuta, S., Kohra, K.: J. Phys. Soc. Jpn. **21**, 1559–1564 (1966)