Variable-bandwidth ^{57}Fe Synchrotron Mössbauer Source

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A synchrotron Mössbauer source (SMS) is a powerful tool for the local analysis of iron-based materials. The SMS filters the single-line ^{57}Fe-Mössbauer radiation from synchrotron radiation (SR) using pure nuclear Bragg reflection (PNBR) of the $^{57}FeBO_3$ crystal near the Néel point (T_N), and it allows us to conduct conventional ^{57}Fe Mössbauer spectroscopy, whereby information on the local magnetic state, valence, and electron density can be obtained via the electron nuclear hyperfine couplings. Additionally, the high degree of polarization, low divergence, and small beam size of SR enables an advanced Mössbauer experiment [1]. Moreover, SMS has great potential to improve the radiation properties because the magnetic control of $^{57}FeBO_3$ crystal significantly affects the PNBR. As a first attempt, we have developed a new method of varying the bandwidth of SMS by controlling the magneto-acoustic vibrations in a $^{57}FeBO_3$ crystal [2]. The scheme is as follows. Below the T_N, nuclear Zeeman splitting is observed in $^{57}FeBO_3$. In this case, the PNBR has a multiline resonance structure, corresponding to the four absorption lines of $\Delta m = \pm 1$ nuclear transitions. At room temperature, the resonance energies spread in the region of ~10^{-6}eV, and the photon flux is more than 10 times larger than that of single-line PNBR from the $^{57}FeBO_3$ crystal near the T_N. However, multiline PNBR is not suitable as a probe in spectroscopy. To solve this problem, a radio frequency magnetic field (H_{RF}) is applied to the temperature-controlled $^{57}FeBO_3$ crystal to excite magneto-acoustic vibrations. In this case, the random vibrations of ^{57}Fe atoms are excited in the crystal via magneto-elastic coupling [3], and single-line PNBR below the T_N is obtained by the collapse of nuclear Zeeman splitting in $^{57}FeBO_3$. The linewidth is controllable in the range of 10^{-8} to 10^{-6}eV by adjusting the H_{RF} and the crystal temperature. Figure 1(a) shows the optical system of variable bandwidth ^{57}Fe-SMS. The σ-polarized X-rays were monochromatized to a bandwidth of 2.5 meV at the ^{57}Fe nuclear resonance energy (14.4keV) by a high-resolution monochromator consisting of Si(511) and (975) crystals. It was incident on the RF vibrating $^{57}FeBO_3$ (95% enriched in^{57}Fe): the magneto-acoustic vibrations were excited by the H_{RF} of f= 8.0 MHz and of variable amplitude (0–3.6 Oe); the H_{RF} was generated by a Helmholtz coil (ϕ20mm); the crystal temperature was controlled in the range of 25–76 °C. ^{57}Fe Mössbauer radiation was filtered from the SR by the $^{57}FeBO_3$ (333) PNBR and the bandwidth (energy distribution) of the PNBR was evaluated by measuring the Mössbauer spectra of a Doppler-vibrating 90% ^{57}Fe-enriched 2μm-thick stainless-steel (SS) foil.

The observed spectra are shown in Figure 1(b). Ata glance, one can see that the results give clear evidence that single-line absorption profiles with different linewidths are obtained by adjusting the H_{RF} and the crystal temperature: the bandwidths of SMS show continuous variation in the range of 10^{-8} to 10^{-6} eV, depending on H_{RF} and T; the photon flux markedly increases with linewidth-broadening. Table I summarizes the linewidth and beam flux of the RF collapsed ^{57}Fe-SMS.

The variable-bandwidth ^{57}Fe-SMS provides new application

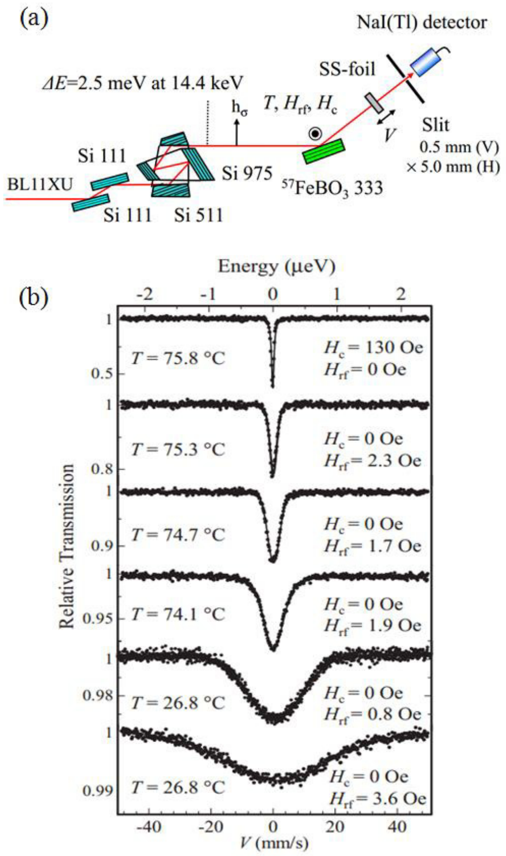

Figure.1. Variable-bandwidth ^{57}Fe-SMS.
(a) Optical system. (b) Energy distribution of ^{57}Fe-SMS under RF field at various temperatures.

possibilities of SR-Mössbauer experiments. As the first feasible application, quasielastic γ-ray scattering of liquid glycerol was measured. Figure 2(a) shows the experimental setup: the H_{RF} (0.75Oe,8 MHz) was applied to the $^{57}FeBO_3$ crystal at T= 25°C. The glycerol was placed on the Cu-stage in a LN_2-cryostat; the^{57}Fe-SR-Mössbauerradiation, whose bandwidth was ~1μeV, was incident on the sample and a Mössbauer reference absorber (MRA), whose absorption linewidth was 50neV, was used to detect the linewidth broadening of the scattered radiation.

Initially, to obtain an instrumental function, the absorption

Table I. Linewidth vs beam flux of the RF collapsed ^{57}Fe-SMS.

T (°C)	H_c (Oe)	H_{RF} (Oe)	Linewidth (neV)	Beam flux (cps)
75.8	130	0	15.1(1)	1.8×10^4
75.3	0	2.3	97.4(3)	5.0×10^4
74.7	0	1.7	187.9(4)	1.1×10^5
74.1	0	1.9	344.9(6)	1.2×10^5
26.8	0	0.8	$0.99(1) \times 10^3$	1.6×10^5
26.8	0	3.6	$1.83(3) \times 10^3$	1.6×10^5

spectrum of an MRA placed in the forward direction was measured with no sample. As shown in the upper part of Figure2(b), the spectrum was a single-line profile and was well fitted by the Voigt function with a linewidth of ~1μeV. Subsequently, the quasielastic γ-ray scattering of glycerol was observed at −123 and 25°C. The former was below the glass-transition temperature T_g~115°C and the latter was over the melting point T_m~17°C. The quasielastic γ-ray photons scattered by the sample were transmitted through the MRA placed at the scattering angle of 12°. The results in Figure 3(b) show that the spectrum of glycerol at −123 °C is almost the same as that of the instrumental function, meaning that the line broadening of the quasielastic scattering of polycrystallized glycerol at a low temperature (T < T_g) is too small to be measured with the resolution of this spectrometer. In contrast, the spectrum at T= 25°C shows a marked linewidth broadening. Considering the linewidth of the instrumental function, the line broadening due to atomic vibration is estimated to be ~1μeV.

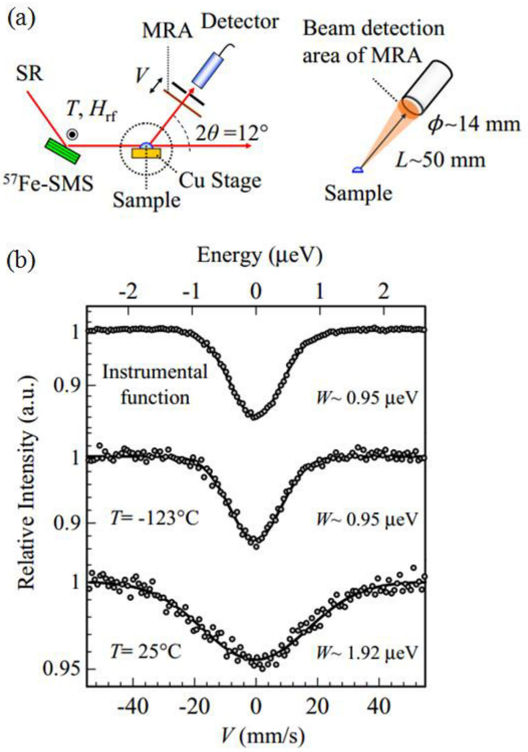

Figure.2. Quasielastic γ-ray scattering of liquid glycerol. (a) Experimental setup. (b) Velocity spectra displaying quasielastic line broadening in scattering from glycerol.

This result shows that the variable-bandwidth^{57}Fe-SMS has a high potential to determine the energy transfer of soft condensed matter (e.g., proteins, colloids, or polymers). The bandwidth-controllable single-line ^{57}Fe-SMS will open new possibilities of SR-based Mössbauer studies with an ultrahigh resolution of 10^{-8} to 10^{-6} eV.

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