Development of ^{125}Te Synchrotron-Radiation-Based Mössbauer Spectroscopy

**Masayuki Kurokuzu · Shinji Kitao ·
Yasuhiro Kobayashi · Makina Saito ·
Ryo Masuda · Takaya Mitsui · Yoshitaka
Yoda · Makoto Seto**

Abstract The Mössbauer spectra of the 35.49 keV excited state of ^{125}Te were observed in the energy domain using synchrotron radiation for the first time. The obtained isomer shifts and Debye temperature agree with previous experiments. These results show that this method will be applicable to various compounds.

1 Introduction

The Mössbauer effect has been observed for approximately 100 nuclear transitions of nearly 50 elements. It has been used in various fields such as chemical, biological, earth, and fundamental physical sciences. However, except for some elements such as ^{57}Fe, ^{119}Sn, and ^{151}Eu, it is not easy to obtain a radioactive isotope source for each nuclear transition. Energy-tunable, highly-bright, and polarized synchrotron radiation (SR) offers an alternative and is an easily accessible choice as a source of Mössbauer spectroscopy. Usually, Mössbauer effect measurements using SR have been performed in the nuclear forward scattering that gives the time domain spectra. However, these spectra are sometimes difficult to analyze,

M. Kurokuzu · S. Kitao · Y. Kobayashi · R. Masuda · M. Seto
Research Reactor Institute, Kyoto University, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan
E-mail: kurokuzu@post3.rri.kyoto-u.ac.jp

T. Mitsui · M. Seto
Japan Atomic Energy Agency, 1-1-1 Koto, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan

Y. Yoda
Japan Synchrotron Radiation Research Institute, 1-1-1 Koto, Sayo-cho, Sayo-gun, Hyogo 679-5198, Japan

M. Saito
Sincrotrone Trieste, SS 14 - km 163, 5 Basovizza, Trieste, Italy

M. Kurokuzu · S. Kitao · Y. Kobayashi · M. Saito · R. Masuda · T. Mitsui · Y. Yoda · M. Seto
CREST, Japan Science and Technology Agency, Honcho Kawaguchi, Saitama 332-0012, Japan

especially when the materials under study have multiple sites. Alternatively, energy domain methods using SR that afford absorption type Mössbauer spectra are similar to the spectra obtained with radioactive sources and complicated spectra are slightly easier to analyze[1]. Recently, the magnetic and vibration properties of Te in $Fe_{1+y}Te$ have been studied to elucidate the superconducting mechanism of iron chalcogenide superconductors[2]. and ^{125}Te Mössbauer spectroscopy is a suitable technique for this study. We have developed a system for ^{125}Te SR-based Mössbauer spectroscopy and show results confirming its feasibility for materials science research.

2 Experiment

^{125}Te SR-based Mössbauer Spectroscopy was performed at the Nuclear Resonant Scattering beam line (BL09XU) of SPring-8. The storage ring was operated at the special timing mode (4/58-filling + 53 bunches) and a ring current of 100 mA by top-up injections. This mode contained 53 isolated bunches, each interval was 82.6 ns, and a 4/58-filled 2-ns-spaced bunch train. The experimental setup is shown in Fig.1. A liquid-nitrogen cooled pre-monochromator was used to handle
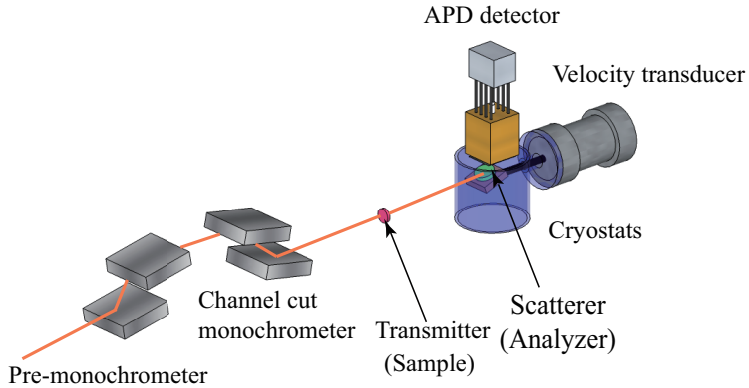

Fig. 1 Schematic of experimental setup. The glancing angle of the incident radiation with respect to the surface of the scatterer is approximately $7°$ and delayed emission from the irradiated surface of the scatterer was measured using a multielement Si-APD detector (4×2 APDs elements, with a detection area of $3 \times 5mm^2$ each).

the high-heat load of the undulator radiation of SPring-8. An Si (1060) asymmetric channel-cut monochromator was then used to select SR with the nuclear resonance energy of 35.49 keV. For this method, two targets, a transmitter and a scatterer containing the same ^{125}Te Mössbauer nuclei are used; the scatterer is placed behind the transmitter on the path of the synchrotron beam. The scatterer was moved using a Mössbauer velocity transducer and was cooled by connecting it to the cold head of a He flow cryostat with Cu springs. The thickness of the springs was chosen to achieve a balance between the smooth motion of the scatterer and

the cooling power. The delayed scattering from the scatterer was measured using a multielement Si-avalanche photodiode (APD) detector placed above the scatterer; wherein, the scatterer was inclined by approximately 7° against the beam direction. It contains 4×2 arrays of APD elements with a detection area of 3 × 5 mm^2 each. Fluorescent X-rays (K$_\alpha$: 27.4 keV, K$_\beta$: 31.0 keV) accompanied by the de-excitation were detected, adding to the γ-rays directly emitted from the excited state (35.49 keV). The detection efficiencies of the APD detector were estimated to be 0.05, 0.04, and 0.03 for 27.4, 31.0, and 35.49 keV X-rays, respectively. The time resolution of the detector was around 1 ns. The velocity-dependency of the scattering intensity yielded a spectrum in the energy domain. We used a Mössbauer calibrator consisting of a laser interferometer to determine the velocity of the transducer.

In the case of SR-based Mössbauer spectroscopy, a reference sample as either the scatterer or transmitter is a required addition to the target sample. We used ^{125}Te-enriched Mg$_3$TeO$_6$ for both target and reference samples because it shows a single line Mössbauer absorption spectrum and has a high recoilless fraction even at room temperature[3]. The enriched Mg$_3$TeO$_6$ was synthesized by the solid reaction method described in Ref.[3]. To optimize the effective thickness, the enriched Mg$_3$TeO$_6$ powder was mixed with boron nitride and polyethylene powders and pressed to form pellets with a diameter of 10 mm. The pellets with a thickness of 0.5 mg ^{125}Te/cm^2 and 5.0 mg ^{125}Te/cm^2 were used as the scatterer and the transmitter, respectively.

3 Result and discussion

A typical count rate of prompt electronic scattering was approximately 9.0×10^6 counts per second (cps). The delayed scattering signals within a time window from 8 to 14 ns were counted after each prompt signal, and it's typical count rate was approximately 3 cps. The ^{125}Te Mössbauer spectrum of the Mg$_3$TeO$_6$ transmitter at 300 K and the scatterer at 60 K is shown in Fig.2(a). The total acquisition time was about 8 hours. The spectrum showed a single line as reported in Ref.[3]. For simplicity, a Lorentzian line shape was used to fit the spectrum. The isomer shift obtained by fitting the observed spectrum is 0.1(2) mm/s and is consistent with the value of 0.01(2) mm/s obtained by conventional Mössbauer spectroscopy[3]. Although the difference of second-order Doppler shifts between 300 and 78 K is reported to be 0.01(2) mm/s[3], the apparent difference of the isomer shifts was within experimental errors and was not observed. Additionally, the ^{125}Te Mössbauer spectrum of Mg$_3$TeO$_6$ at 300 K with the scatterer at 300 K is shown in Fig.2(b). The total acquisition time was about 12 hours. The obtained isomer shift was 0.3(3) mm/s at 300 K. This measurement proves that Mg$_3$TeO$_6$ can be used as the scatterer at room temperature and that it reduces the experimental difficulty by removing the cryostat apparatus of the scatterer. We also evaluated Debye temperature θ_D from the temperature dependence of the normalized absorption area in the spectra using the Debye model. Obtained θ_D was about 340 K, and this value is consistent with the values obtained by conventional methods, 390(2) K [3] and 320 K [4].

Although we measured the ^{125}Te Mössbauer spectra at the special timing mode (82.6 ns bunch interval) of the storage ring, appropriate bunch intervals, such as
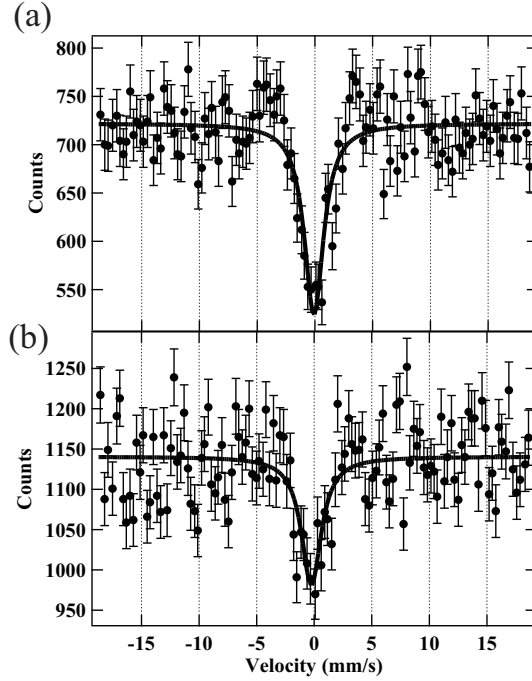
Fig. 2 ^{125}Te Mössbauer spectra measured by using SR. As a transmitter, Mg_3TeO_6 with a thickness of 5.0 mg ^{125}Te /cm^2 was used at 300 K. As a scatterer, Mg_3TeO_6 with a thickness of 0.5 mg ^{125}Te /cm^2 was used at (a) 60 and (b) 300 K. The solid line shows fit spectra with Lorentzian line shapes.

the 203 bunch mode with 23.6 ns bunch intervals, improves the count rate because the half-life of the ^{125}Te (35.49 keV first excited state) is 1.48 ns. Furthermore, if the time resolution of the APD detector is improved, a measurement with an earlier timing gate after the prompt pulse is possible. For example, if the delayed scattering signals within a time window from 7 to 14 ns can be measured as in Ref.[1], we can obtain signal approximately 1.8 times larger than those from 8 to 14 ns. Finally, the detection of internal conversion electrons, well known in conventional Mössbauer spectroscopy and previously measured by nuclear resonance scattering of SR by ^{197}Au[5], is expected to substantially improve efficiency due to the internal conversion process, which is dominant in ^{125}Te (the internal conversion coefficient is 13.6). Therefore, ^{125}Te SR-based Mössbauer spectroscopy is promising for many fields of investigations on various Te-containing compounds, including the superconductor $Fe_{1+y}Te$.

4 Summary

The ^{125} Mössbauer spectra were measured using SR for the first time. It was found that Mg_3TeO_6 can be used as a scatterer at room temperaturefor ^{125}Te SR-based Mössbauer spectroscopy. The isomer shifts and θ_D of Mg_3TeO_6 obtained

by SR-based Mössbauer spectroscopy are consistent with the values reported in conventional methods. These results show that this spectroscopic method can be applied to various Te-containing compounds. There are possibilities to enhance the effectiveness of this method through the optimization of bunch mode, improvement of time resolution in the APD detector, and detection of the internal conversion electrons.

References

1. M. Seto, R. Masuda, S. Higashitaniguchi, S. Kitao, Y. Kobayashi, C. Inaba, T. Mitsui, Y. Yoda, Phys. Rev. Lett. **102**(21), 217602 (2009)
2. F.C. Hsu, J.Y. Luo, K.W. Yeh, T.K. Chen, T.W. Huang, P.M. Wu, Y.C. Lee, Y.L. Huang, Y.Y. Chu, D.C. Yan, et al., Proc. Natl. Acad. Sci. USA **105**(38), 14262 (2008)
3. W. Bresser, M. Zhang, L. Koudelka, J. Wells, P. Boolchand, G. Ehrhart, P. Miller, Phys. Rev. B **47**(18), 11663 (1993)
4. M. Pasternak, M. Van der Heyden, G. Langouche, Nucl. Instrum. Methods, B **4**(1), 152 (1984)
5. S. Kishimoto, Y. Yoda, M. Seto, Y. Kobayashi, S. Kitao, R. Haruki, T. Kawauchi, K. Fukutani, T. Okano, Phys. Rev. Lett. **85**(9), 1831 (2000)