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^{125}Te-Mössbauer study of $Fe_{1.1}Te$ and $FeTe_{0.5}Se_{0.5}$ superconductor

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Abstract
A parent compound of iron-based superconductor, $Fe_{1.1}Te$, and a Se-doped superconductor, $FeTe_{0.5}Se_{0.5}$, were investigated using ^{125}Te-Mössbauer spectroscopy. By measuring precise temperature dependence of the spectra, discontinuous line broadening in the $Fe_{1.1}Te$ near the antiferromagnetic transition temperature (T_N) of Fe was observed. While the broadening of the line width were observed in both samples as the temperature decreases, the temperature dependences of the line width were well reproduced as that of the recoilless fraction, assuming the Debye model with a Debye temperature of about 200 K. The discontinuous additional line broadening in the $Fe_{1.1}Te$ can be considered as the internal magnetic fields of Te below T_N, since the temperature dependence of internal magnetic fields of Te is similar to that of Fe in ^{57}Fe-Mössbauer spectroscopy. Therefore, hybridization of the electronic orbital of Te and Fe was suggested and the transferred magnetic moment of Te was induced through the antiferromagnetic order of Fe at T_N.

Keywords ^{125}Te Mössbauer spectroscopy · Iron-based superconductor · Antiferromagnetic transition · Magnetic moment of Te

1 Introduction

The discoveries of Fe-oxipnictide superconductors, LaFePO [1] and $LaFeAsO_{1-x}F_x$ [2], have opened a new epoch of superconductivity. Since these discoveries, extensive

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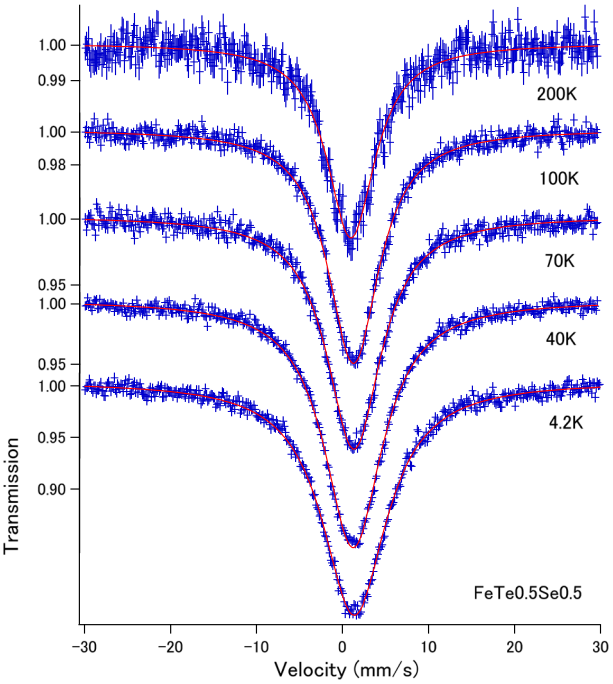

Fig. 1 Typical ^{125}Te-Mössbauer spectra of $FeTe_{0.5}Se_{0.5}$

researches have been performed to elucidate the mechanism of their superconductivity. Since it is believed that the Fe acts as a key element and its magnetism must relate to the superconductivity, Fe-Mössbauer spectroscopy is one of the most essential methods to investigate the nature of the superconductivity. Until today, a number of Mössbauer studies on these Fe-based superconductors have been carried out and revealed many facts, e.g., F-doping to the LaFeAsO suppresses the antiferromagnetic transition and leads the emergence of superconductivity in $LaFeAsO_{1-x}F_x$ [3]. The initial discoveries of so-called "1111" series of compounds gave rise to successive discoveries of several series of related compounds, "122" series such as $BaFe_2As_2$, "11" series such as FeSe [4], and so on. Among these Fe-based superconductors, "11" series of compounds have the simplest composition. The superconductivity is observed in $FeTe_{0.5}Se_{0.5}$ at a transition temperature (T_c) of about 15 K by Se doping to a non-superconducting parent compound, FeTe. The $FeTe_{0.5}Se_{0.5}$ does not show magnetic order even at low temperature, while FeTe shows antiferromagnetically-ordered phase below a Néel temperature (T_N) of about 65 K. However, the magnetic structure of FeTe, is rather complicated compared to other series of compounds, which has been observed by Fe-Mössbauer spectroscopy [5–7] and by neutron diffraction [8]. In addition, since in some "122" series of compounds the electronic nematic phase with anisotropic distortion in a-b plane above T_N is suggested [9], this phenomenon has to

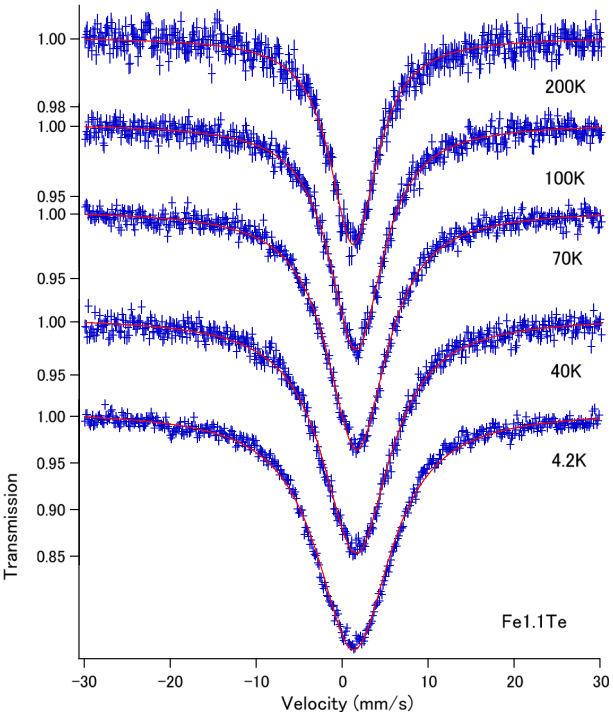

Fig. 2 Typical ^{125}Te-Mössbauer spectra of $Fe_{1.1}Te$

be investigated in the case of "11" series of compounds. To elucidate these features, it is important to investigate not only the electronic state of Fe but also that of Te, since the electronic bands of Fe and Te can affect each other. ^{125}Te is useful as another Mössbauer isotope to investigate FeTe compounds. In the ^{125}Te Mössbauer spectroscopy, the excited level with 35.5 keV and the half-life of 1.48 ns is used and the spins of the excited and ground state are 3/2 and 1/2, respectively. In fact, a former study using synchrotron-based ^{125}Te-Mössbauer spectroscopy suggested the existence of internal magnetic moment of Te in the magnetically-ordered phase in $Fe_{1.1}Te$ [10]. In this study, precise temperature dependences of ^{125}Te-Mössbauer spectroscopy was performed on a parent compound, $Fe_{1.1}Te$ and a superconductor, $FeTe_{0.5}Se_{0.5}$.

2 Experimental

Single crystals of $Fe_{1.1}Te$ and $FeTe_{0.5}Se_{0.5}$ were synthesized by self-flux method from stoichiometric powder of reduced Fe, distilled Te, and Se grains in a crucible of aluminum oxide sealed in a quartz tube. The Te was about 50% enriched by mixing a same amount of natural Te and ^{125}Te. The sealed tube was heated up to

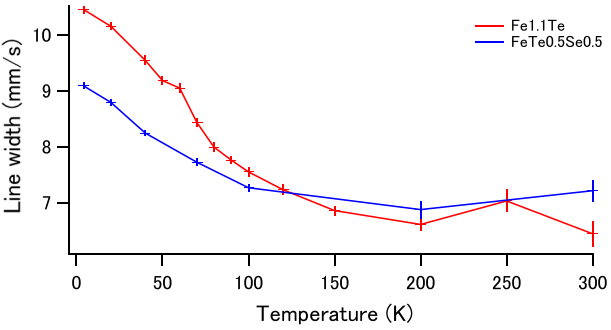

Fig. 3 Temperature dependences of line width obtained from ^{125}Te-Mössbauer spectra of $Fe_{1.1}Te$ and $FeTe_{0.5}Se_{0.5}$ by Lorentzian curve fit

1070 °C in 24 h and cooled down to 710 °C at a rate of 3 °C/h, and annealed at 400 °C for 10 days [11]. The obtained single crystal was characterized by x-ray diffraction. The T_N of 67 K in $Fe_{1.1}Te$ and the T_c of 14 K in $FeTe_{0.5}Se_{0.5}$ were evaluated by magnetic susceptibility measurements at Research Center for Low Temperature and Materials Sciences, Kyoto University. As for the ^{125m}Te Mössbauer source, neutron irradiation of $Mg_3{}^{124}TeO_6$ was performed at Kyoto University Reactor (KUR) for about 4 weeks followed by annealing at 700 °C. Typical obtained activity of ^{125m}Te was about 30 MBq. The Mössbauer spectra were measured using a pellet of 7 mm in diameter of ground powder of single crystals with about 5 mg. The 35-keV γ-ray from ^{125m}Te were measured by using a Ge solid state detector. Typical measurement time was about 12 h for each spectrum. The temperature of the sample was controlled in a liquid-He flow cryostat, while the $Mg_3{}^{125m}TeO_6$ source was set to the velocity transducer at room temperature. The velocity scale of ^{125}Te Mössbauer spectra are referenced to Mg_3TeO_6 at room temperature. Conventional ^{57}Fe-Mössbauer spectra were also measured using a ^{57}Co source with nominal activity of 1.85 GBq. The Mössbauer spectra were analyzed with MossWinn program.

3 Results and discussion

Precise temperature dependences of ^{125}Te-Mössbauer spectra of $FeTe_{0.5}Se_{0.5}$ and $Fe_{1.1}Te$ were measured. Typical ^{125}Te-Mössbauer spectra of $FeTe_{0.5}Se_{0.5}$ are shown in Fig. 1. The line widths of ^{125}Te-Mössbauer spectra were broadened as the temperature decreases. Typical ^{125}Te-Mössbauer spectra of $Fe_{1.1}Te$ were shown in Fig. 2. The line widths of the spectra were also broadened as the temperature decreases. The line widths of the spectra were firstly fit with Lorentzian curve as in Fig. 3. In the spectra of $Fe_{1.1}Te$, an additional discontinuous line broadening was observed below T_N of Fe. Since the line broadening was considered to be mainly

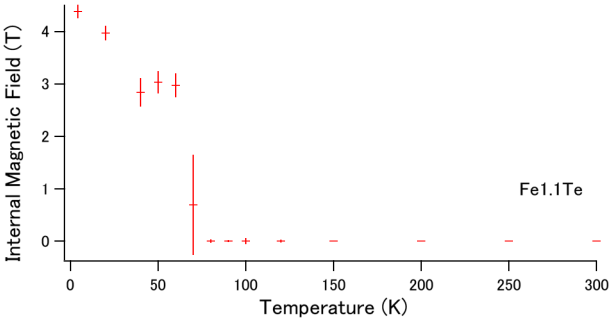

Fig. 4 Temperature dependence of internal magnetic fields from ^{125}Te-Mössbauer spectra of $Fe_{1.1}Te$

due to the temperature dependence of the recoilless fraction, we analysed the spectra with a transmission integral, assuming the Debye model with a Debye temperature of about 200 K [12]. The overall temperature dependences of the line width of the obtained spectra for both sample were well reproduced. The additional line broadening below T_N in $Fe_{1.1}Te$ is assumed to be due to the internal magnetic field of Te. Because of wide natural line width of the Te-Mössbauer spectroscopy, it is difficult to separate the contribution from internal magnetic fields and quadrupole splittings. In this analysis, the quadrupole splitting was included in an intrinsic line width of the sample from multiple contributions with a fixed value. The spectra are fitted only with free parameters of internal magnetic fields, isomer shift and back ground fraction. All other parameters were fixed. Thus, the temperature dependence of internal magnetic fields of Te was evaluated. In Fig. 4, the internal magnetic fields of Te in $Fe_{1.1}Te$ are shown. Typical value of internal magnetic field of Te in $Fe_{1.1}Te$ is about 4 T at 4.2 K. For comparison, ^{57}Fe-Mössbauer spectra of $Fe_{1.1}Te$ are also measured. The temperature dependence of averaged values of the distributed internal magnetic fields are shown in Fig. 5a. The results were similar to former reports [5–7]. Since the temperature dependence of the internal magnetic fields of Te was similar to that of Fe in ^{57}Fe-Mössbauer spectroscopy, the magnetic moments of Te were considered to be induced through the antiferromagnetic order of Fe at T_N. This fact implies a hybridization of the electronic orbitals between Te-5 *s* or Te-5*p* and Fe-3*d*. This result is also supported by the ab initio calculation, where the hybridization between Te-*p* and Fe-*d* orbitals is suggested [13]. Typical values of the isomer shifts of Te in $Fe_{1.1}Te$ and $FeTe_{0.5}Se_{0.5}$ were 1.3 and 1.4 mm/s at 4.2 K relative to Mg_3TeO_6, respectively. The temperature dependence of the isomer shift was not clearly observed. These results agreed well with the former results [10]. In the ^{57}Fe-Mössbauer spectra of $Fe_{1.1}Te$, the line width broadening was observed above T_N as shown in Fig.5b. This result is considered to be due to the electronic nematic phase [9]. It is suggested that a subtle quadrupole splitting may arise in the electronic nematic phase, which is observed at the temperature above T_N. The effect of electronic nematic phase in the ^{125}Te-Mössbauer spectra has not been well recognized because of wide natural line width.

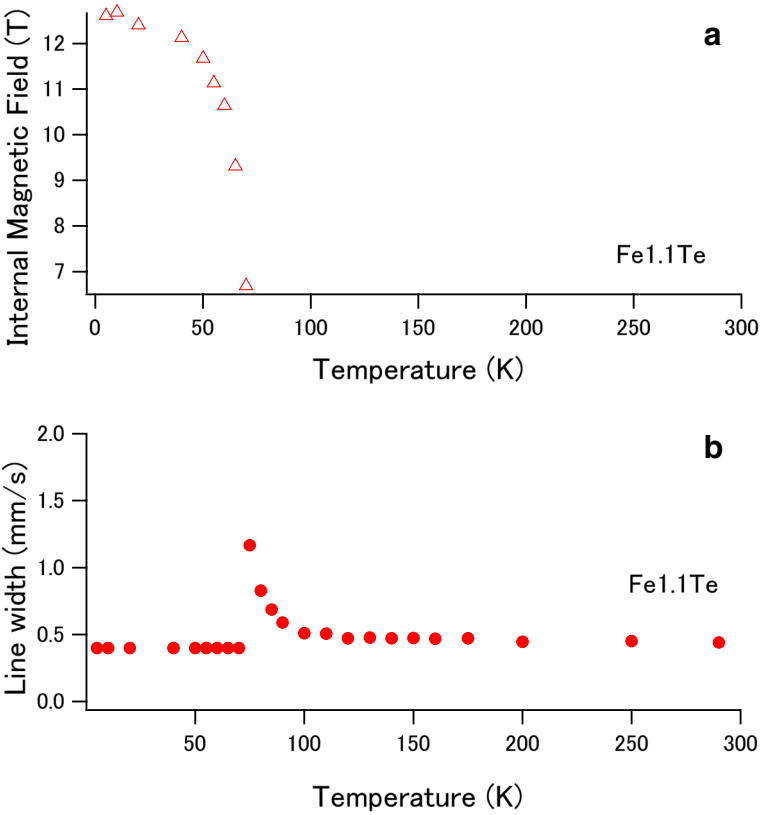

Fig. 5 Temperature dependence of (**a**) internal magnetic fields and (**b**) line widths from ^{57}Fe-Mössbauer spectra of $Fe_{1.1}Te$

4 Conclusion

In summary, we observed internal magnetic moment not only in Fe but also in Te below T_N in $Fe_{1.1}Te$ from the ^{125}Te-Mössbauer spectra. This internal magnetic moment of Te is considered to be due to transferred hyperfine fields through hybridization of Te and Fe orbitals. The overall temperature dependences of the line width of Te in $Fe_{1.1}Te$ and $FeTe_{0.5}Se_{0.5}$ were well reproduced by the change of recoilless fraction assumed as the Debye model.

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