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Mössbauer study of ε-Fe under an external magnetic field

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Abstract
Using a diamond anvil cell, ^{57}Fe Mössbauer measurements of the high-pressure phase of iron, ε-Fe at 20 GPa, have been performed at 4.5 K under external magnetic fields up to 7 T. The magnitudes of the hyperfine magnetic fields depend linearly on the external magnetic fields, H_{ext}. This implies that there is no induced hyperfine field due to the local magnetic moment and ε-Fe under 20 GPa at 4.5 K is determined as a Pauli paramagnet.

1. Introduction

It is well recognized that the high-pressure phase of iron, ε-Fe, does not show any magnetic order down to 30 mK [1]. Whether the Fe atoms in ε-Fe have a local moment showing a magnetic order below 30 mK or Fe has no magnetic moment at all is an important question, because non-magnetic Fe might show superconductivity [2]. ^{57}Fe Mössbauer spectroscopy is a convenient tools for investigating magnetic properties of iron and materials containing Mössbauer probe iron atoms even under very high pressure using a diamond anvil cell (DAC), because the hyperfine magnetic field at the ^{57}Fe nucleus reflects the magnetic properties of the material and the diamond anvil is rather transparent to the Mössbauer γ-rays. In this investigation, we performed again ^{57}Fe Mössbauer measurements on ε-Fe at 20 GPa under externally applied magnetic fields up to 7 T at 4.5 K in order to examine the existence or non-existence of the local magnetic moment of Fe in ε-Fe. In this investigation, we have not observed any induced hyperfine magnetic fields larger than 0.5 T at the ^{57}Fe nucleus in ε-Fe, which means that Fe in ε-Fe has no local magnetic moment larger than 0.05 μ_B at 4.5 K and 20 GPa.

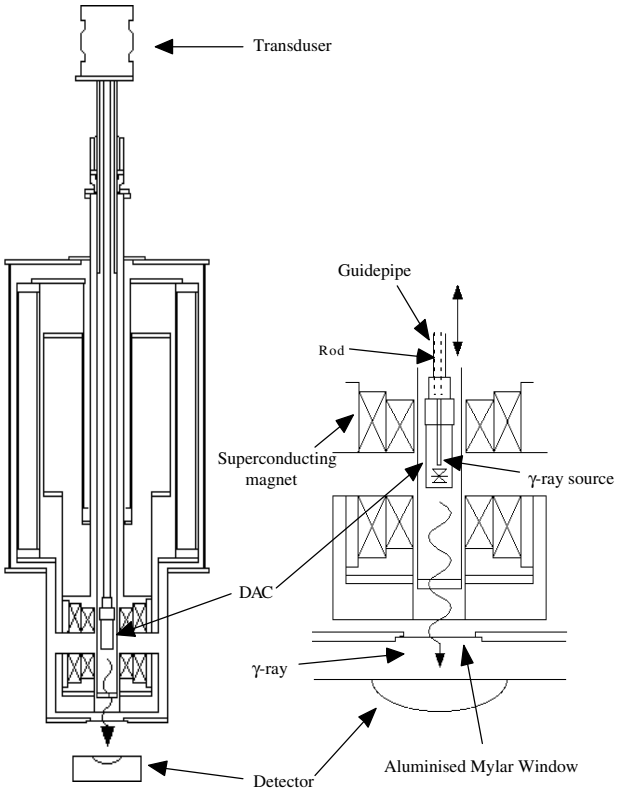

Figure 1. A schematic drawing of the high-pressure Mössbauer cryostat with a velocity transducer and the gamma-ray detector.

2. Experimental details

^{57}Fe Mössbauer measurements were performed in the transmission geometry using ^{57}Fe-enriched iron foil placed in a DAC and a γ-ray source of ^{57}Co in a Rh matrix. A Basset-type DAC made of Cu–Be alloy was used. As the metal gasket, a Re sheet was used. The size of the culet plane of the diamond is 600 μm and the diameter of the gasket hole that is the specimen chamber is 300 μm. The specimen chamber was filled with a mixture of methanol and ethanol as the pressure-transmitting medium. Ruby manometry was used to determine the pressure at 300 and 77 K. The pressure at 4.5 K is assumed to be the same as the pressure at 77 K. A 975 MBq ^{57}Co-in-Rh matrix and a 95.5% enriched ^{57}Fe foil whose thickness is 2 μm were used as the γ-ray source and absorber specimen. Figure 1 shows a schematic drawing of the high-pressure Mössbauer cryostat with superconducting solenoids, Mössbauer velocity transducer and Xe-gas-filled proportional counter for the detection of the γ-rays. As shown in figure 1, the direction of the magnetic field is parallel to the direction of the γ-rays. High magnetic fields were supplied by using a superconducting magnet on the absorber specimen and also on the γ-ray source, ^{57}Co in Rh. Since the Fe-in-Rh matrix has a weak local magnetic moment, although its spin-glass behaviour requires rather large concentration of magnetic impurity [3], one can expect induced hyperfine fields at the ^{57}Fe nucleus in Rh. In this investigation we determined the line-shapes of the ^{57}Fe Mössbauer spectra arising from the ^{57}Co-in-Rh source (975 MBq, 4 mm $\times$ 4 mm $\times$ 6 μm) experimentally at 4.5 K under external magnetic fields.

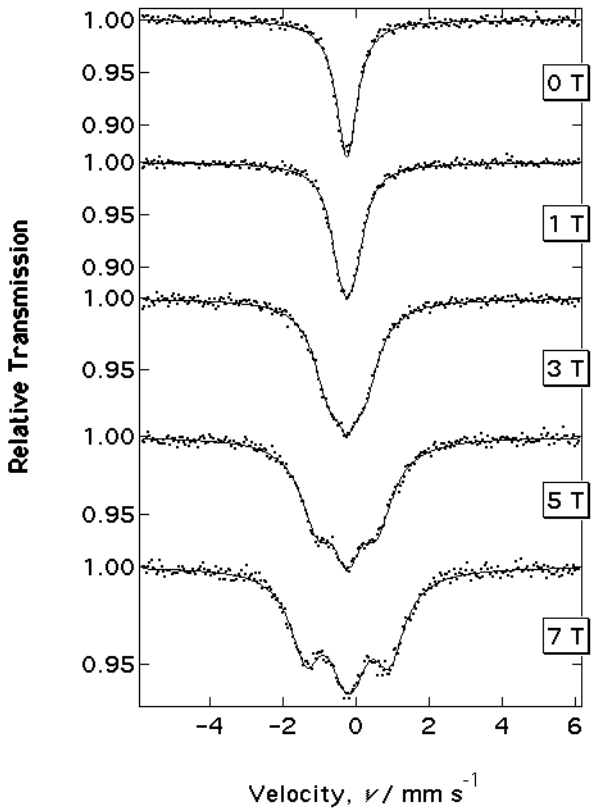

Figure 2. Typical ^{57}Fe Mössbauer spectra obtained from ε-Fe under 20 GPa at 4.5 K as a function of longitudinally applied external magnetic fields. The ^{57}Co in Rh is also at 4.5 K under the same external magnetic fields. Velocity is relative to α-Fe at 300 K.

Indeed, the induced hyperfine fields at the ^{57}Fe-in-Rh source at 4.5 K were observed and determined as a function of H_{ext} by means of measurements using a Pd–2% Fe single-line absorber placed outside the cryostat.

In these measurements, the induced hyperfine fields at the ^{57}Fe nucleus in Rh have nearly same magnitude as the external magnetic field with opposite direction. Consequently the observed total hyperfine fields cancel each other and rather single-line-like Mössbauer absorption spectra have been observed in the combination with the Pd–2% Fe absorber placed outside of the cryostat. The width of the broad single line due to the induced hyperfine field at the Fe in Rh has been determined as a function of the external magnetic fields, H_{ext}. Using these values, it is possible to determine the magnitude of the field at the ^{57}Fe nucleus in ε-Fe.

3. Results and discussion

From precise spectral analysis, the ^{57}Fe Mössbauer absorption spectrum of ε-Fe without an external magnetic field is known to be a non-resolved doublet caused by the electronic quadrupole interaction due to the hexagonal structure [4]. The magnitude of the quadrupole interaction at 20 GPa and 4.5 K is determined as 0.14 mm s^{-1} and the centre shift is -0.23 mm s^{-1} relative to α-Fe at 300 K. Typical ^{57}Fe Mössbauer spectra obtained from ε-Fe at 4.5 K under longitudinally applied magnetic fields up to 7 T are shown in figure 2.

The Mössbauer spectrum without an external magnetic field, as shown in figure 2, is a broad unresolved doublet at 4.5 K. The centre shift and the magnitude of the electric quadrupole interaction were determined as -0.23 mm s^{-1} relative to room temperature bcc Fe and 0.14 mm s^{-1}, respectively. The quadrupole interaction at 4.5 K is nearly equal to the value at 300 K. As an increase of the externally applied longitudinal magnetic field, the spectra became broad and above 3 T the spectra became hyperfine split patterns: nearly just four lines. At external fields of 5 and 7 T a slight asymmetry in the spectrum became visible because of the mixed hyperfine interaction with the electric quadrupole and magnetic dipole interactions. Dots are experimental data and the solid curves shown in figure 2 are results from least-squares fits between the experimental data and the calculated spectrum with a hyperfine field, quadrupole splitting and centre shift. We used two methods to analyse the spectrum. One is an exact Hamiltonian method for the mixed hyperfine interactions and the other is a simple method using four shifted absorption lines, each of which is a convolution with the emission line obtained experimentally with the ^{57}Co-in-Rh source. The two methods gave nearly identical results; this implies that the electric quadrupole interaction is negative and the asymmetry parameter is nearly zero. The most important result is that the magnitudes of the magnetic dipole interaction are nearly the same for the two methods within their experimental errors (the maximum error was 0.4 T at $H_{ext} = 3$ T). The magnitudes of the hyperfine magnetic fields have been determined as a function of H_{ext} and are nearly equal to the magnitudes of the external magnetic fields. If the Fe in ε-Fe has a weak magnetic moment like Fe in Rh, the induced hyperfine field can be estimated from the hyperfine coupling constant. Assuming the local moment of Fe in ε-Fe is of the order of 0.1 μ_B and the hyperfine coupling constant is of the same order as that of the ^{57}Fe in the bcc ferromagnetic Fe, namely 11 T/μ_B, we can expect an induced hyperfine magnetic field of the order of 1 T, a value easily distinguishable from the Mössbauer measurement. As shown in figure 2, the ^{57}Fe Mössbauer spectra under external fields show no shrinkage or expansion due to the extra fields and the spectral shapes are well described by a shape without $\Delta m = 0$ lines. Figure 3 shows the results obtained for the centre shifts and the observed hyperfine magnetic fields as a function of H_{ext}. The observed hyperfine fields depend linearly on the external magnetic fields. The centre shift values do not depend on the external magnetic field, which has no physical meaning but shows the correctness of the method used to analyse the spectral shape. From the above results, we can conclude that Fe in ε-Fe has no local moment larger than 0.05 μ_B and the magnetism of ε-Fe is most probably Pauli paramagnetism at 4.5 K under 20 GPa.

Quite recently Shimizu *et al* [5] found superconductivity of ε-Fe, whose maximum superconducting critical temperature is 2 K at around 20 GPa. One could speculate that the origin of the superconductivity of ε-Fe may be magnetic like in the cases of UGe_2 [6] and $ZrZn_2$ [7]. However, the present results show that there is no local magnetic moment larger than 0.05 μ_B at Fe in ε-Fe at 4.5 K and 20 GPa and one might expect a different mechanism of superconductivity for ε-Fe from the cases for the above two compounds. Further investigation in the search for the magnetism of ε-Fe is necessary to understand the superconductivity of ε-Fe—for example, at lower temperatures with larger external magnetic fields and investigations at various pressures.

4. Summary

We re-examined the ^{57}Fe Mössbauer spectroscopy of ε-Fe at 20 GPa under externally applied magnetic fields up to 7 T at 4.5 K in order to clarify the existence or non-existence of the local magnetic moment of Fe in ε-Fe. In this investigation, we have not observed any induced hyperfine magnetic fields at the ^{57}Fe nucleus in ε-Fe larger than 0.5 T, which means that Fe in ε-Fe has no local magnetic moment larger than 0.05 μ_B at 4.5 K and 20 GPa.

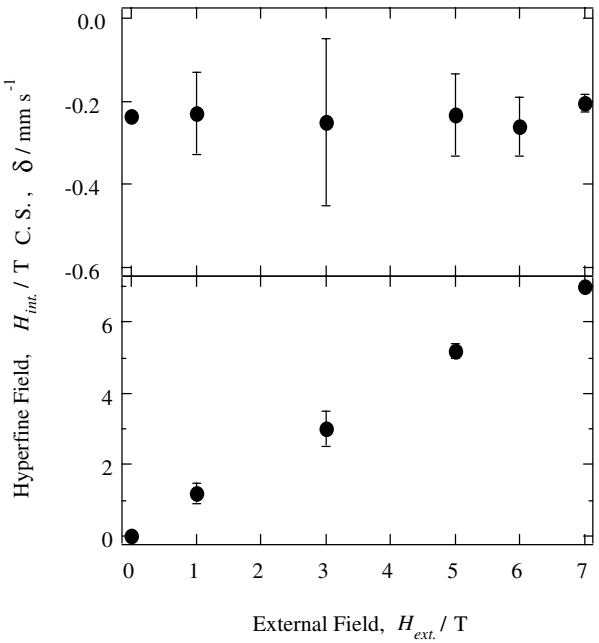

Figure 3. Centre shifts and observed hyperfine magnetic fields as a function of the externally applied longitudinal magnetic field H_{ext} at ^{57}Fe in ε-Fe at 4.5 K and 20 GPa. The centre shift values are relative to α-Fe at room temperature. The observed hyperfine fields fit well to the straight line up to 7 T.

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