

Hyperfine fields in UFe_5Sn compound

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Abstract. Local hyperfine interactions on Fe nuclei in UFe_5Sn compound were determined in Mössbauer experiment. The analysis of the Mössbauer spectra measured at room temperature reveals the presence of two doublets only. Absorption line width indicates that neither distribution of quadrupole splitting nor isomer shift exists in the alloy. The measurements at 13 K show the presence of four magnetic components. Because the magnetic dipole and electric quadrupole interactions are present, full Hamiltonian was used to determine the hyperfine interactions for each iron site. Electric field gradient determined at low temperatures agrees with quadrupole splitting observed at room temperature. Absorption areas of the components at low and at high temperatures correspond well to the occupation of the crystal sites by Fe atoms.

Key words: Mössbauer studies • uranium intermetallic • magnetic intermetallic

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Introduction

New intermetallic compound UFe_5Sn , discovered by A. P. Gonçalves *et al.* [3], crystallizes in orthorhombic CeCu_3Au -type structure (space group $Pnma$). The unit cell contains 4 U atoms located in $4c$ site, 4 Sn atoms in $4c$, 12 Fe atoms in $4c$ (Fe2, Fe3, Fe4) and 8 Fe atoms in $8d$ (Fe1) site [3]. The magnetization measurements carried out on a powder sample show a weak ferromagnetic behavior at room temperature and two magnetic anomalies at 248 K and 180 K [3]. The latter transition is magnetic field dependent and disappears at $B > 1$ T. Below 248 K magnetization strongly increases with decreasing temperature. More precise experiments on a single crystal [4] show that the transition at 248 K is associated with ferromagnetic ordering of iron magnetic moments along c -axis, while anomalies observed at 180 K with reorientation Fe moments towards to b -axis. This conclusion is deduced partially from Mössbauer measurements. However, the description of the spectra presented in [4] in our opinion does not fit the experimental data so well. We think that the main reason of not good agreement between the experimental and theoretical Mössbauer spectra is the use of the first order perturbation in the presence of mixed hyperfine interactions. Since the electric quadrupole interactions are not small with respect to the magnetic dipolar ones, a full Hamiltonian should be used for correct description of the Mössbauer spectra. The aim of our study is

determination of the hyperfine interactions of iron atoms located at different sites in the UFe_5Sn compound.

Sample preparation and data handling

The UFe_5Sn crystal was grown by the Czochralski method. Our sample crystallizes in the same structure as reported [3] and the lattice parameters, within the experimental errors, do not differ greatly from [3]. The powdered sample studied in the work was crushed from a single crystal, and was mixed with the LiCO_3 and epoxy glue. Such preparation results in the random distribution of the grains, so-called texture free sample. The sample thickness was $12 \text{ mg UFe}_5\text{Sn per cm}^2$.

The Mössbauer measurements at $T = 13 \text{ K}$ were carried out in a close cycle refrigerator equipped with an antivibration shroud. The source of radiation was ^{57}Co in Cr matrix. The velocity scale was calibrated with respect to $\alpha\text{-Fe}$ at room temperature.

All spectra were analysed by commercial Mössbauer package “Normos” using transmission integral [5]. Recoilless fraction of Fe atoms in UFe_5Sn was assumed to be the same as in $\alpha\text{-Fe}$ [2]. The resulting values of effective thickness of the absorber were estimated to be $\tau = 2.07 \pm 0.11$ at room temperature and $\tau = 2.41 \pm 0.11$ at $T = 13 \text{ K}$.

The spectra measured at $T = 13 \text{ K}$ were analyzed using full Hamiltonian approach for the texture free sample. Axial symmetry of the electric field gradient was assumed (asymmetry parameter $\eta = 0$) for all local environments of Fe atoms.

Results and discussion

The Mössbauer spectrum measured at room temperature was fitted by two doublets with the same values of width of the Lorentzian lines (D(1), D(2)). The result of the best fit is shown in Fig. 1 by the thick line, while parameters of the two components are listed in Table 1. The ratio of the areas of the two doublets is close to four, consistent with the assumption that doublet D(1) is created by 8 Fe atoms at $8d$ and 8 Fe atoms at $4c$

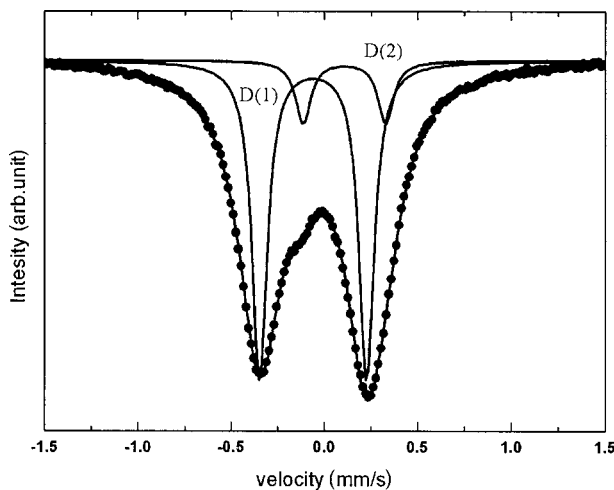


Fig. 1. Mössbauer spectrum measured at room temperature.

Table 1. The hyperfine parameters obtained from the spectrum measured at room temperature (eQ – electric quadrupole moment, V_{zz} – z component of electric field gradient, IS – isomer shift, I – intensities of the components and Γ_a – full width of the Lorentzian lines at half maximum. The width of the source was determined from calibration experiment $\Gamma_s = 0.17 \text{ mm/s}$)

	$ eQV_{zz}/2 $ (mm/s)	IS (mm/s)	I (%)	Γ_a (mm/s)
D(1)	0.54 ± 0.01	-0.06 ± 0.01	84 ± 4	0.10 ± 0.01
D(2)	0.44 ± 0.02	0.11 ± 0.01	16 ± 4	0.10 ± 0.01

sites, while D(2) doublet is created by 4 Fe atoms at $4c$ site. The difference in the isomer shift (IS) between D(2) and D(1) doublets is 0.18 mm/s . Because the Sn atoms located in the nearest neighborhood (NN) of Fe increase its IS [1], one may conclude that D(2) doublet results from the Fe2 site only, which has three Sn atoms, and being their NN located at distances ranging between $2.61\text{--}2.93 \text{ \AA}$ [3].

Despite four different local Fe sites in the unit cell, the room temperature spectra can be precisely fitted by two doublets only (see Fig. 1). The absorption lines are not broadened (their widths are equal to the widths of the lines in calibration spectrum of $\alpha\text{-Fe}$). We thus conclude that hyperfine interactions for the Fe1, Fe3 and Fe4 sites, which differ in local symmetry, result in the single doublet D(1).

The Mössbauer spectrum measured at $T = 13 \text{ K}$ is shown in Fig. 2. The shape of spectrum can be fitted by a sum of four magnetic components $S(n)$ ($n = 1, 2, 3, 4$) and a single doublet (D), all with very sharp lines ($\Gamma_a = 0.13 \pm 0.01 \text{ mm/s}$). Thick lines in Fig. 2 represent the best fit. The parameters obtained from the fit are presented in Table 2. Because the population of Fe at the $8d$ sites should be twice larger than that of Fe at each of remaining sites, one concludes that the most intense component $S(1)$ corresponds to the Fe1 site. It was observed in U-Fe-Al [7] and in Fe-Sn [6] alloys that hyperfine magnetic field (HMF) of Fe increases with a number of Fe NN. Assuming similar behavior in the studied compound one concludes that $S(2)$, $S(3)$ and

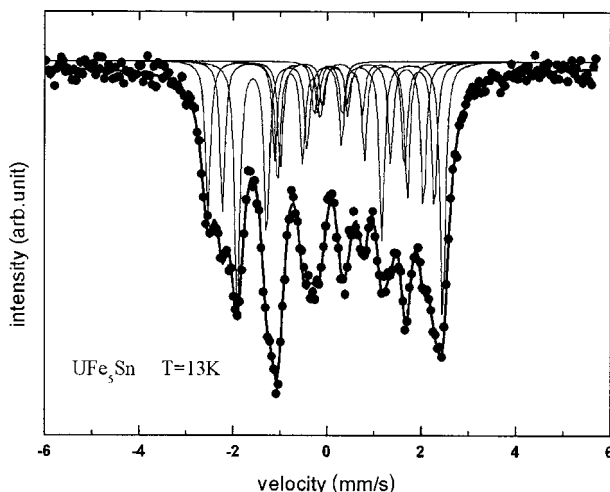


Fig. 2. Mössbauer spectrum measured at $T = 13 \text{ K}$.

Table 2. The hyperfine parameters obtained from the spectrum measured at $T = 13$ K for different local surroundings (eQ – electric quadrupole moment, V_{zz} – z component of electric field gradient, HMF – hyperfine magnetic field, I – intensities of the components)

		S(1)	S(4)	S(3)	S(2)	D
IS	(mm/s)	0.104 ± 0.005	0.094 ± 0.005	0.01 ± 0.03	0.23 ± 0.02	0.02 ± 0.02
$eQV_{zz}/2$	(mm/s)	0.46 ± 0.02	-0.63 ± 0.05	0.49 ± 0.10	0.56 ± 0.05	0.59 ± 0.04
HMF	(T)	13.4 ± 0.2	14.6 ± 0.2	13.0 ± 0.3	7.9 ± 0.2	0
I	(%)	36 ± 2	22 ± 2	20 ± 4	19 ± 4	3 ± 4

S(4) comes from Fe2, Fe3 and Fe4 sites, respectively, which is consistent with [3].

In contrast to the results of [3], relative area of the doublet D observed at low temperature is twice smaller than that of the sextet (HMF = 2T) [3]. The origin of this doublet is not clear because its relative area is small. Comparison of hyperfine parameters obtained here indicates that value of IS of the doublet D is close to that of S(3) component, which in turn indicates that this doublet may have Fe3 site origin, as suggested in [3]. However, it is not clear, why part of Fe iron atoms, located at the Fe3 site is still in the paramagnetic state at $T = 13$ K. Another possibility is that the considered doublet results from a small fraction of Fe atoms located at antisite positions, U or Sn atoms.

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