119\(^{\text{Sn}}\) Mössbauer Spectroscopy in the heavy – fermion ferrimagnet UCu\(_5\)Sn

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Abstract. 119\(^{\text{Sn}}\) Mössbauer studies of UCu\(_5\)Sn compound have been performed in a wide temperature range between 4.2 K and 78 K. Magnetic hyperfine split spectra observed below \(T = 54\) K indicate the onset of magnetic ordering. The temperature dependence of the average transferred effective magnetic hyperfine field \(< \text{H}_{\text{eff}} >\) at the tin site can be surprisingly well fitted using the Brillouin function for \(S = 1/2\). A rather small distribution of magnetic hyperfine fields \(\delta \text{H}\) obtained from the fitting procedure is in favour with the assumption that atomic mixing between Cu and Sn position can be neglected. The value of \(\theta = 10(2)^{\circ}\) shows that the direction of the effective magnetic hyperfine field \(\text{H}_{\text{eff}}\) is close to the c-axis being in accord with the uranium magnetic moment direction.

1. Introduction
The UCu\(_5\)Sn compound belongs to the family of moderate heavy-fermion systems \([1]\) being a ferrimagnet with the ordering temperature \(T_C = 54(1)\) K \([1-3]\). This compound exhibits the hexagonal CeNi\(_5\)Sn-type structure with space group \(P6_3/mmc\) and its magnetic structure below \(T_C\) consists of the ferromagnetic planes \((001)\) with the \(++−−\) stacking sequence, where the U magnetic moments are ordered along the hexagonal c-axis with two different values of magnetic moments at 2(a) and 2(c) U non-equivalent sites \([3]\).

The aim of this work is to present the results of detailed 119\(^{\text{Sn}}\) Mössbauer effect measurements performed in a wide temperature range below as well as above the magnetic transition temperature \(T_C\). Mössbauer Spectroscopy (MS) is well known as an extremely sensitive tool to study local electronic properties as it enables zero external magnetic field studies without disturbing of the investigated magnetic system. In particular, usage of the diamagnetic 119\(^{\text{Sn}}\) Mössbauer probe, being a natural constituent of the investigated sample, gives the unique opportunity to follow the temperature evolution of hyperfine parameters, which may be influenced by structural and magnetic phase transformations. It should be underlined that our Mössbauer studies were made on the same sample, which was used in the previous investigations of thermodynamic, transport, structural and magnetic properties of UCu\(_5\)Sn \([1-3]\).
2. Experimental
The $^{119}$Sn Mössbauer measurements have been performed using a triangular velocity drive of a Kankeleit type in transmission geometry. The $^{119}$Sn source in chemical form of Ba$^{119}$SnO$_3$ and strength of about 10 mCi was kept at temperatures close to 4.2 K and series of $^{119}$Sn Mössbauer spectra were recorded in a conventional variable temperature cryostat in the range of 4.2 K – 78 K. Absorber was prepared by grinding the sample to a fine powder and mixing it homogeneously with Al$_2$O$_3$ to ensure constant surface distribution over the absorber container with optimal thickness 26 mg cm$^{-2}$ of UCu$_5$Sn material. For detection of the resonance 23.875 keV gamma quanta a scintillation counter with a 3 cm NaI(Tl) crystal was applied. To reduce 25 keV Sn $K\alpha$ background the critical filter made of Pd-foil ca. 0.05 mm thick was used. The energy scale was calibrated by means of a $^{57}$Co(Cr) source and a metallic $\alpha$-Fe foil at room temperature. The spectra were analysed by least-squares fits to the experimental results within the Lorentz approximation. By means of the numerical diagonalization procedure of the hyperfine interaction Hamiltonian $H_{hf}$, the resonance-line positions and their relative intensities were calculated. The full refinements of the spectra lead to the results for the isomer shift $\delta_{is}$, the quadrupole interaction constant $\Delta Q = eQV_{zz}$, the effective magnetic hyperfine field $H_{\text{eff}}$ and for the polar angle $\theta$, that is, for the direction of the magnetic hyperfine field with respect to the z-axis of the EFG (electric field gradient) tensor. Czjzek and Berger [4] method was used to account for an observed small broadening of the low temperature spectra of UCu$_5$Sn sample caused by possible distribution in magnitudes of the effective magnetic hyperfine field $H_{\text{eff}}$. The average value $\langle H_{\text{eff}} \rangle$ and the magnetic hyperfine field distribution parameter defined as $\delta_H = (\langle H_{\text{eff}}^2 \rangle - \langle H_{\text{eff}} \rangle^2)^{1/2}/\langle H_{\text{eff}} \rangle$ (i.e. the ratio of the second moment $M_2(H_{\text{eff}})$ of the effective magnetic hyperfine field distribution to its average value) were fitted as two independent parameters. All spectra can be reproduced satisfactorily in this way with remarkable improvement of the fit quality.

3. Results
The resonance $^{119}$Sn Mössbauer spectra obtained for UCu$_5$Sn compound at $T = 78$ K and 4.2 K are shown in figures 1 and 2, respectively. Because of non-cubic local symmetry of the Sn sites the spectrum measured at $T = 78$ K (i.e. far above $T_C$) presents typical quadrupole doublet, and its slight asymmetry can be attributed to a small amount of an impurity of unknown origin (figure 1).

**Figure 1.** The $^{119}$Sn resonance spectrum of the UCu$_5$Sn sample obtained at $T = 78$ K, i.e. above the critical magnetic transition temperature $T_C = 54$ K. The continuous line represents the least-squares fit to the experimental points.

**Figure 2.** The $^{119}$Sn resonance spectrum of the UCu$_5$Sn sample obtained at $T = 4.2$ K, i.e. below the critical magnetic transition temperature $T_C = 54$ K. The continuous line represents the least-squares fit to the experimental points.
This impurity component, fitted as a single line, accounts for about 4% of the total absorption intensity and is almost temperature independent.

Below T_c all the recorded spectra can be effectively fitted as a sum of only one main magnetic hyperfine component with a slightly distributed magnitude of the hyperfine magnetic field (according to the Czjzek and Berger method [4], see Section 2) and the respective impurity contribution which starts to split below 8 K in a visible way. Since the value of ΔE_Q for the main component calculated as a free parameter was not sensitive to the temperature change, it was constraint during fitting of magnetically split spectra to the value obtained at 78 K (ΔE_Q = |1.22(2)| mm/s). It is worth noting that the best fits to the low temperature spectra were obtained for the positive value of the quadrupole interaction constant ΔE_Q conjugated simultaneously with the small value of θ angle equal to 10(2)°, although reasonable fits can also be obtained for the negative value of ΔE_Q associated with θ angle close to 90°. The spectrum obtained at T = 4.2 K is shown in figure 2 as a characteristic example. The hyperfine parameters obtained from the refinements at 78 K and 4.2 K are collected in table 1.

Table 1. The hyperfine interaction parameters obtained from the ^{119}Sn resonance spectra of the UCu_5Sn intermetallic compound at T = 78 K (i.e. above the critical temperature T_C = 54 K) and 4.2 K.

| T (K) | ∠H_{eff} (kOe) | ΔE_Q^a (mm/s\(^{-1}\)) | δ_is^b (mm/s\(^{-1}\)) | θ (deg) | Γ^c (mm/s\(^{-1}\)) | δ_H^d |
|---|---|---|---|---|---|---|
| 78 | - | | 1.57±0.01 | - | 0.82±0.01 | - |
| 4.2 | 67.1±0.1 | | 1.22^e | 1.58±0.01 | 10±2 | 0.82^e | 0.004±0.001 |

^a ΔE_Q = eQV_{zz}  
^b δ_is is isomer shift relative to the Ba^{119m}SnO\(_3\) source.  
^c Γ is half width of the resonance line.  
^d δ_H = (∠H_{eff}^2 - ∠H_{eff})^{1/2}/∠H_{eff} - the magnetic hyperfine field distribution parameter.  
^e parameter kept constant during the fit.

The temperature dependence of the average effective magnetic hyperfine field ∠H_{eff} can be well fitted by the Brillouin function for S = 1/2 (figure 3). The fit gives the expected value of the magnetic transition temperature T_C = 54(1) K as well as the average value of the saturation effective magnetic hyperfine field ∠H_{eff}(0) = 67.5(2) kOe.

Figure 3. Temperature evolution of the average effective magnetic hyperfine field at the Sn sites for UCu_5Sn. The continuous line represents the least-squares fit of the S = 1/2 Brillouin function for the average values of the effective magnetic hyperfine fields, ∠H_{eff}.
4. Discussion

The derived value of the isomer shift (please note that $\delta_{\text{is}}=1.57(1)$ mm/s obtained at room temperature is equal within the experimental error to $\delta_{\text{is}}=1.58(1)$ mm/s at liquid helium temperature) is typical for that of Sn in metallic systems. Almost the same values of isomer shifts were observed for example in hexagonal UCu$_2$Sn and UPt$_2$Sn compounds ($\delta_{\text{is}}=1.63(1)$ mm/s and $\delta_{\text{is}}=1.58(1)$ mm/s, respectively) [5]. It means that the s electron charge density at the tin nucleus is not much affected by the tin environment. The lack of a remarkable temperature dependence of the measured isomer shift leads to a conclusion that the influence of the lattice expansion on its value is compensated by the change of the second order Doppler shift.

Owing to the threefold symmetry axis at the Sn position which is directed parallel to the crystallographic c-axis the $V_{zz}$ axis of the EFG tensor coincides with this axis. Therefore the polar angle $\theta$ is the angle between the crystallographic c-axis and the direction of the magnetic hyperfine field $H_{\text{eff}}$ induced at the tin site by a transfer mechanism and dipole field due to dipole interaction of magnetic uranium moments. In other words, the value of $\theta$ angle gives a deviation of $H_{\text{eff}}$ vector from c-axis, which is the easy axis of magnetisation in our case. As this angle is only of about 10° it means that the direction of $H_{\text{eff}}$ is close to the c-axis. Since dipole field is usually small it can be neglected in a first approximation. Under assumption that transfer mechanism is of RKKY type the magnetic hyperfine field $H_{\text{eff}}$ at each Sn position is parallel and proportional to the magnetic uranium moments $\mu_U$, hence, as a result on the basis of MS measurements alone, we can deduce that direction of these moments is also close to the c-axis. In fact, neutron investigations confirm this result, which showed parallel alignment of the U moments to the c-axis. In turn, taking into account collinear ordering of the magnetic U moments along c-axis, their values and interionic distances, dipole field $H_d$ has been calculated. Using the value of experimentally derived $H_{\text{eff}}$ and calculated values of dipole contributions, one gets that the maximum deviation of $H_{\text{eff}}$ from the c-axis of about 11° is in a perfect agreement with the experimental $\theta$ value.

The estimated value of $T_c = 54(1)$ K is in a very good agreement with those obtained from magnetization and neutron diffraction measurements i.e. 54(1) K. One can stress here that fits to the experimentally obtained temperature dependence of the average effective magnetic hyperfine field $<H_{\text{eff}}(T)>$ using Brillouin functions with S higher than $\frac{1}{2}$ give a little higher estimates for the derived magnetic transition temperature $T_c$ than that observed by methods mentioned above. A rather small value of the derived magnetic hyperfine field distribution leads to the conclusion that there is not much atomic mixing between Cu and Sn positions. On the other hand such a mixing might change the local variation in the degree of hybridization of the U-5f electron states with the valence-band states of the ligands and in consequence causes a change of the local values of the U magnetic moment leading to the huge distribution of magnetic hyperfine fields as observed experimentally in UAuSn compound [6].

References

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