

Mössbauer Study of Cubic Phase in the Mn–Sb System

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Mössbauer spectroscopy was applied to investigate the structural and magnetic parameters of cubic phase in the Mn–Sb system. The validity of the model of triangular magnetic structure in Mn₃Sb has been confirmed.

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1. Introduction

One of the methods to produce novel materials is high temperature and high pressure treatment. In such a manner in the Mn–Sb system the cubic modification of Mn₃Sb could be produced [1, 2]. This compound has the *L*₁₂-type of crystal structure (Cu₃Au) with the lattice parameter $a = 0.400$ nm. The Mn atoms occupy 3c positions with the coordinates $(0, 1/2, 1/2)$, the Sb atoms — the positions 1a $(0, 0, 0)$, Fig. 1. In normal conditions, the compound exists in a thermodynamically non-equilibrium state and decomposes at 420 K on the Mn₂Sb + Mn phases. The magnetic measurements of the Mn₃Sb were performed by the Faraday method in the field 8.6 kOe and the temperature region from the liquid nitrogen to the temperature of phase decomposition. The derived value of specific magnetization is very small, ≈ 1.0 A m²/kg [2]. The neutron diffraction analysis for the Mn₃Sb was performed as well [3]. The data obtained were interpreted in the model of the ferrimagnetic ordering of Mn magnetic moments. The magnetic moments of Mn atoms are supposed to be antiparallel and have different values initially taking into account the similar local environment of Mn atoms in both tetragonal Mn₂Sb and cubic Mn₃Sb. However, it is possible to interpret the neutron diffraction data in a different way, using the model of triangular magnetic ordering of Mn magnetic moments. In contrast to previous one, in this model the Mn atoms are considered to have equal magnetic moments ordered in the (111) plane and forming an equilateral triangle with each moment pointing to the center of the triangle. The similar model was used in [4] to describe the magnetic ordering of Mn in Mn₃Pt with the same *L*₁₂ type of structure below 400 K.

Thus the model of the magnetic ordering of Mn atoms in the cubic Mn₃Sb is ambiguous. Therefore the purpose of this study was to get Mössbauer information on the number of magnetically nonequivalent states of Mn atoms and to clarify a type of the magnetic ordering in Mn₃Sb.

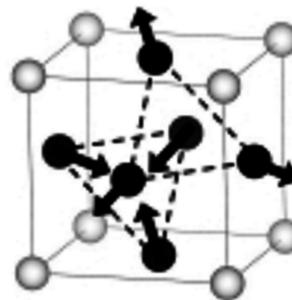


Fig. 1. The *L*₁₂-type of crystal structure and the triangular magnetic ordering of Mn atoms in Mn₃Sb (Mn atom — black circle, Sb atom — open circle).

2. Experimental details

The Mössbauer investigations were performed in a usual transmission geometry and in a constant acceleration regime. The gamma-resonance source was ⁵⁷Co/Rh with the FWHM equal to 0.11 mm/s.

All samples were doped with 2 atomic percent of pure ⁵⁷Fe substituted for the Mn that provided the possibility to implement the Mössbauer experiment. The sample Mn_{2.98}Fe_{0.02}Sb with cubic structure for the Mössbauer study was synthesized in two stages:

1) First, the sample was obtained by usual hetero phase reaction in the mixture of powdered components — the “green alloy”. The calculation of the component weights was made under the assumption of chemical formula Mn_{2.98}Fe_{0.02}Sb,

2) In the second stage the “green alloy” was treated by high temperature and pressure, 2500 K and 7 GPa, respectively. Thus the cubic Mn_{2.98}Fe_{0.02}Sb sample has been obtained.

X-ray pattern of the green alloy shows the coexistence of two phases, namely Mn₂Sb and Mn. No additional reflexes were detected in the diffraction pattern of the cubic sample.

3. Results and discussion

The Mössbauer spectra of the “green alloy” as well as the cubic manganese antimonide are presented in

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Fig. 2a,b. The parameters for the cubic $\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ spectrum were refined by Recoil program (see Table).

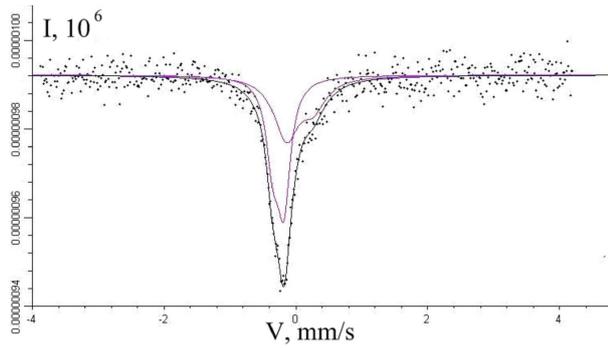


Fig. 2. ^{57}Fe Mössbauer spectra of $\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ “green alloy” (a) and cubic $\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ (b) measured at room temperature.

TABLE

The hyperfine parameters for samples determined for room temperature estimated from spectra.

Sample/parameters	IS [mm/s]	QS [mm/s]	H [T]	W [mm/s]
$\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ “green alloy”	-0.26 0.06	0.16 0.41	– –	0.12 0.22
$\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ cubic	0.46	-0.13	17.3	0.20
$\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ “green alloy”	0.49 0.52	0.40 -0.06	– 7.9	0.21 0.10
$\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ treated	0.51	0.41	–	0.15

The spectrum of the cubic sample $\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ is presented in Fig. 2b. The single sextet suggests that the alloy is a monophasic magnetic one. No different hyperfine magnetic field values at Mn nuclei have been detected. It is known that the hyperfine magnetic field at each nucleus is a sum of three contributions: $H_{\text{eff}} = H_{\text{p}} + H_{\mu} + H_{\text{str}}$, where the first term is created by the self-polarised s -electrons of the atom, H_{μ} — by the own magnetic moment of the atom, and H_{str} — by the magnetic moments of the nearest neighboring atoms in the structure. Therefore this last term for the cubic crystals is equal to zero. The H_{eff} at the Mn nucleus in the cubic manganese antimonide is the result of its own spin and the orbital magnetic moment. The Mössbauer data indicate that all Mn atoms are situated in the same magnetic state. Thus the data correspond to the triangular model of magnetic ordering, in which the magnetic moments of Mn atoms have the same values.

The lines of sextet are slightly broadened (relative to the source line width), that testifies to the high regularity of the alloy. The value of the hyperfine magnetic field at the Mn atoms is $H_{\text{eff}} = 17.3$ T, that corresponds to the nuclei magnetic moment $\mu \approx 1.22 \mu_{\text{B}}$.

In order to make sure that there is no phase with a hexagonal (NiAs) type of crystal structure in the cubic

sample, we should compare the Mössbauer data for cubic $\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ and hexagonal Mn_{1+x}Sb . It is known that the x value can be $0 \leq x \leq 0.22$ [5] for the Mn_{1+x}Sb obtained by usual heterophase reaction and $0 \leq x \leq 0.5$ [6] for the sample prepared under the high temperature and pressure treatment. The Mössbauer data for the samples with $0 \leq x \leq 0.22$ were obtained earlier [7]. It has been shown that the hyperfine magnetic field value at Mn nuclei does not exceed 8 T and differs from the value $H_{\text{eff}} = 17.3$ T for the cubic manganese antimonide mentioned above.

For the purpose of this work we prepared the $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ in the same two-stage manner as the cubic sample. X-ray analysis has shown two-phase composition of the “green alloy”, the phases are hexagonal (NiAs-type) MnSb and tetragonal Mn_2Sb . The diffraction pattern of the treated $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ fits to the hexagonal phase.

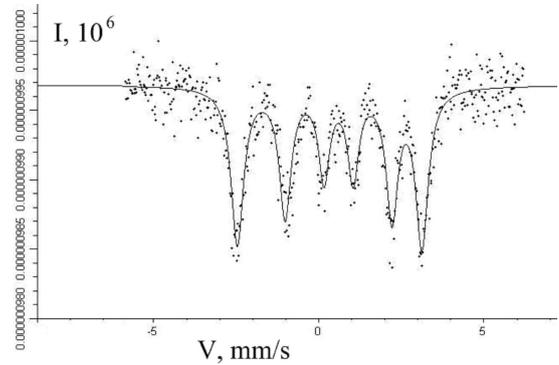


Fig. 3. ^{57}Fe Mössbauer spectra of $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ “green alloy” (a) and treated $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ (b) measured at room temperature.

The Mössbauer spectra of the green alloy $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ and the sample obtained under the high temperature and pressure treatment are presented in Fig. 3a and b, respectively. The spectrum of the NiAs-type green alloy has a magnetic subspectrum with the maximal hyperfine field value about 8 T. There is no magnetic part in the spectrum of $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ obtained under high temperature and pressure. From the comparison of $\text{Mn}_{1.48}\text{Fe}_{0.02}\text{Sb}$ and $\text{Mn}_{2.98}\text{Fe}_{0.02}\text{Sb}$ spectra (Fig. 2b and Fig. 3b) it is clear that the hexagonal NiAs-phase is absent in the cubic manganese antimonide under the study.

4. Conclusions

The Mössbauer spectroscopy technique was used to study the cubic manganese antimonide Mn_3Sb . It was shown that all the Mn atoms have identical magnetic state. The adaptability of the model of triangular magnetic ordering of Mn atom magnetic moments in Mn_3Sb was confirmed.

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