Mössbauer Study of Magnetic Texture of Finemet-Type Ribbons

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Magnetic texture of amorphous Finemet-type alloys substituted by Co, Ni, Mn, Ge and V was investigated by means of Mössbauer spectroscopy using various angular configurations. Samples in form of ribbons were manufactured by single-roller melt spinning technique. In most of investigated alloys some preference of alignment of atomic magnetic moments in-plane of sample is stated, except of some alloys substituted by germanium in which slight tendency to out-of-plane spin orientation is proved.

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1. Introduction

The subject of the present paper are magnetic materials belonging to an alloy group called Finemet. The first amorphous alloy from this family, of chemical composition Fe\textsubscript{73.5}Nb\textsubscript{2}Cu\textsubscript{1}B\textsubscript{3}Si\textsubscript{13.5}, was obtained in 1988 [1] by rapid quenching of melt and had the form of a ribbon. It is known from excellent soft magnetic properties [2] and also as a potential precursor of nanocrystalline alloys that can be manufactured through controlled annealing (under vacuum) of the initial amorphous material at suitably chosen temperature. Many attempts of improving the properties of classical Finemet have been done up to now. They consist in searching the optimal chemical composition [3–8] as well as manufacturing conditions [9] and also thermal treatment terms [10–11].

The most popular method of producing the amorphous alloys is melt spinning. In this method a hot melt of chosen composition is spilled on the surface of rotating copper drum. The process can be carried out in air or under protective argon atmosphere. The melt coagulates in the form of 20–30 µm thick ribbon. The cooling rate that can be achieved in this method is of order 10\textsuperscript{5} K/s. As seen, the specific manufacturing conditions cause an anisotropy of final product. We can expect that its in-plane and out-of-plane properties may be different and additionally a conspicuous axis of anisotropy may occur, for instance — along the ribbon.

In this paper, magnetic texture of various Finemet-like amorphous alloys is the main subject of interest and it is investigated by means of transmission Mössbauer spectroscopy (TMS) as well as conversion electron Mössbauer spectroscopy (CEMS). All the samples were manufactured using the same experimental system for melt-spinning, in similar conditions, therefore differences in their properties can be attributed mainly to their chemical composition.

2. Theoretical aspects

It is known that some information about alignment of atomic magnetic moments can be derived from Mössbauer spectra. In the case of unpolarized dipole radiation, relative intensities of lines 2, 5 in a Zeeman sextet are related to $\langle \cos^2 \psi_R \rangle$ — mean value of $\cos^2 \psi_R$, where $\psi_R$ signifies the angle between the incident gamma beam and the local magnetic hyperfine field, with the following relations [12]:

$$p = I_{2,5} / I_{3,4}, \quad \langle \cos^2 \psi_R \rangle = (4 - p)/(4 + p),$$

$$p = 4(1 - \langle \cos^2 \psi_R \rangle)/(1 + \langle \cos^2 \psi_R \rangle).$$

In the case of circularly polarized gamma radiation, when Mössbauer spectrum is asymmetric in respect of line intensities, the parameter $p$ can be derived from the formula:

$$p = (I_2 + I_3)/(I_3 + I_4).$$

One can increase the amount of information when some Mössbauer experiments in different geometrical arrangement are performed. Let us consider a magnetic material in form of ribbon and let us choose the coordination system shown in Fig. 1. According to the model presented by J-M Grenche et al. [13–14], parameters: $N_X$, $N_Y$, $N_Z$ can be introduced, called “populations”, in order to describe mean alignment of atomic magnetic moments. They denote relative numbers of spins directed along axes $OX$, $OY$, $OZ$, respectively, of a hypothetical discrete structure equivalent to that under

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examination, in which identical angular dependence of 
\(\langle \cos^2 \psi_R \rangle\) occurs. If the sample shows mirror symmetry 
towards 0X axis, the parameters can be derived experimentally by Mössbauer 
spectroscopy. They fulfill the following relations:

\[ N_X + N_Y + N_Z = 1, \]

\[ \langle \cos^2 \psi_R \rangle_X = N_X \sin^2 \theta \cos^2 \phi, \]

\[ \langle \cos^2 \psi_R \rangle_Y = N_Y \sin^2 \theta \sin^2 \phi, \]

\[ \langle \cos^2 \psi_R \rangle_Z = N_Z \cos^2 \theta, \]

\[ \langle \cos^2 \psi_R \rangle = N_X \sin^2 \theta \cos^2 \phi + N_Y \sin^2 \theta \sin^2 \phi + N_Z \cos^2 \theta, \]

where \(\langle \cos^2 \psi_R \rangle_i, i = X, Y, Z\) are contributions coming 
from axes OX, OY, OZ, respectively. In other words, we replace the real system by a hypothetical one, in which magnetic 
moements can lie only along the axes OX, OY, OZ and which gives the same Mössbauer spectrum as the real system.

\[ \phi = 90^\circ, \] respectively, and the “magic” angle \(\theta \approx 54.7^\circ\) 
that fulfill the relation:

\[ \cos^2 \theta = \frac{1}{3}, \]

where 1/3 is an average value of \(\cos^2 \theta\) for random angular 
distribution. From relations (4)–(7) one can obtain populations:

\[ N_Z = \langle \cos^2 \psi_R \rangle_A, \]

\[ N_Y = \frac{1}{2} \left( 1 - \langle \cos^2 \psi_R \rangle_A \right) + \frac{3}{4} \left( \langle \cos^2 \psi_R \rangle_B - \langle \cos^2 \psi_R \rangle_C \right), \]

\[ N_X = \frac{1}{2} \left( 1 - \langle \cos^2 \psi_R \rangle_A \right) - \frac{3}{4} \left( \langle \cos^2 \psi_R \rangle_B - \langle \cos^2 \psi_R \rangle_C \right), \]

where \(\langle \cos^2 \psi_R \rangle_K, K = A, B, C\) denote the values 
obtained from the measurement performed in corresponding 
configurations.

3. Experimental details

Fine-met-type alloys in which several atoms of iron or silicon were replaced by atoms of other elements: Co, Ni, Mn, V and Ge, were prepared by single-roller melt spinning technique. Transmission Mössbauer measurements were carried out by use of a conventional spectrometer arranged in vertical geometry with \(^{57}\)Co(Rh) source and a vibrator working in a constant acceleration mode. On the basis of investigations performed using standard configuration A, the population of magnetic moments lying in XOY plane: \(N_X + N_Y = 1 - N_Z\) was derived. For selected alloys the measurements were carried out also in configurations B and C by means of a specially projected sample holder which ensured its proper inclination. It enabled the evaluation of \(N_X\) and \(N_Y\) using relations (11), (12).

In order to compare the magnetic texture in bulk material and on the surface, conversion electron Mössbauer spectroscopy (CEMS) measurements were performed, using a gas-flow CEMS detector supplied with He+4%CH\(_4\) mixture.

4. Results

Mössbauer spectra of investigated alloys take a shape typical of the magnetic amorphous materials — a smeared Zeeman sextet, the line width of which increases with the distance of line from the centre of spectrum. The shape of spectrum indicates that it originates from atoms being subjected to distributed magnetic hyperfine fields (MHF). The spectra were fitted by means of programs NORMOS or MOSFIT [15] using histogram-like form of MHF distribution. In both procedures, a linear correlation between magnetic hyperfine field and isomer shift were assumed. Exemplary Mössbauer spectra of Fe\(_{72.5}\)Cu\(_{1}\)Nb\(_{3}\)Ge\(_{2}\)Si\(_{13.5}\)–\(_{2}\)B\(_{9}\) alloys are shown in Figs. 3a–

d, the values of \(p\) are also noticed.
Analogous measurements were carried out for alloys of composition: Fe$_{73.5-x}$Mn$_x$Cu$_1$Nb$_3$Si$_{13.5-x}$B$_9$, Fe$_{73.5-x}$Ni$_x$Cu$_1$Nb$_3$Si$_{13.5-x}$B$_9$, Fe$_{73.5-x}$Co$_x$Cu$_1$Nb$_3$Si$_{13.5-x}$B$_9$, Fe$_{73.5-x}$V$_x$Cu$_1$Nb$_3$Si$_{13.5-x}$B$_9$. The determined populations of magnetic moments derived from TMS measurements in Finemet alloys substituted by Ge seems to be strongly dependent on Ge concentration, which gives $N_X + N_Y = 0.69$, the value very close to that characteristic of random spin orientation. Since the alignment of magnetic moments in Finemet alloys substituted by Ge concentration, we attribute the difference to chemical content fluctuations. CEMS measurement of the former sample was also performed in order to investigate the spin orientation at the surface (Fig. 3e). It was found that $p = 3.7$, which is related to $N_X + N_Y = 0.95$. This means nearly full in-plane alignment of atomic magnetic moments at the surface of the ribbon.

### Table

<table>
<thead>
<tr>
<th>Alloy</th>
<th>$N_X$</th>
<th>$N_Y$</th>
<th>$N_Z$</th>
<th>$N_X + N_Y$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$_{73.5}$Cu$_1$Nb$<em>3$Ge$</em>{13.5}$B$_9$</td>
<td>0.433</td>
<td>0.163</td>
<td>0.404</td>
<td>0.596</td>
</tr>
<tr>
<td>Fe$<em>{51.5}$Co$</em>{22}$Cu$_1$Nb$<em>3$Si$</em>{13.5}$B$_9$</td>
<td>0.450</td>
<td>0.346</td>
<td>0.204</td>
<td>0.796</td>
</tr>
</tbody>
</table>

The alloys of composition Fe$_{73.5}$Cu$_1$Nb$_3$Ge$_{13.5}$B$_9$ and Fe$_{51.5}$Co$_{22}$Cu$_1$Nb$_3$Si$_{13.5}$B$_9$ were chosen for subsequent TMS measurements using configurations B, C. Mössbauer spectra of the former ribbon are shown in Fig. 5. The determined populations $N_X$, $N_Y$, $N_Z$ are presented in Table. It is worth reminding that for fully random orientation of magnetic moments each of the populations equals 1/3. The outcomes prove distinct preference of the
direction along the ribbon in both samples. Besides, the axis perpendicular to the surface of ribbon is preferred in Fe$_{73.5}$Cu$_{1}$Nb$_{3}$Ge$_{13.5}$B$_{9}$ alloy while in-plane, transverse direction is favored in Fe$_{51.5}$Co$_{22}$Cu$_{1}$Nb$_{3}$Si$_{13.5}$B$_{9}$.

5. Summary

TMS studies prove that in most of the Finemet-type ribbons the population of spin oriented parallel to the ribbon surface fulfills the condition: $0.75 < N_X + N_Y < 0.93$ and therefore atomic magnetic moments show strong preference to in-plane alignment. The alloys substituted with germanium seem to be the exception, since the observed tendency diminishes with growing germanium concentration and for 13.5 at% of Ge the ribbon exhibits nearly random orientation or slight preference to out-of plane spin alignment. As shown from CEMS investigations, even for this alloy nearly full in-plane alignment of atomic magnetic moments is observed at the surface of ribbon. The investigated alloys exhibit also the preferred anisotropy axis along the ribbon.

References