

# Correlation between MI Effect and Transverse Anisotropy in Stress-Annealed Nanocrystalline Alloys: A Mössbauer Effect Study

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In order to understand the correlation between the large and sensitive magneto-impedance (MI) effect in the tensile stress-annealed Fe-based nanocrystalline alloy  $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  and the transverse magnetic anisotropy on a microscopic scale, a Mössbauer effect study has been carried out. A series of ribbon samples were subject to thermal annealing with tensile stress  $\sigma$  up to 60 MPa. MI measurements showed that the effective field of transverse magnetic anisotropy  $H_k$  increases with increasing  $\sigma$ . Analysis of Mössbauer spectra provided direct evidence that there is an increasing transverse magnetic structure with increasing  $\sigma$  in these tensile stress-annealed samples. This work confirms, on the microscopic scale, that the transverse magnetic anisotropy arises from stress-annealing and is essential for the giant magneto-impedance effect.

## 1. Introduction

Fe-based nanocrystalline alloys have been intensively investigated due to their excellent soft magnetic properties [1, 2]. The magnetic softness of these materials is associated with their novel microstructure composed of grains of  $\alpha$ -Fe(Si) embedded in an amorphous matrix. However, heat treatments can induce magnetic anisotropy, thus changing its soft magnetic properties. Recently, with the application of a tensile stress or a transverse magnetic field during annealing, several Fe-based nanocrystalline alloys, such as  $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$ ,  $\text{Fe}_{73}\text{Cu}_1\text{Nb}_{1.5}\text{V}_2\text{Si}_{13.5}\text{B}_9$  and  $\text{Fe}_{73}\text{Cu}_1\text{Nb}_{1.5}\text{Mo}_2\text{Si}_{13.5}\text{B}_9$ , have been found capable of obtaining not only a transverse magnetic structure but also a large magneto-impedance (MI) effect [3–6]. The fact that the transverse magnetic anisotropic is an essential factor for large MI effect has been explained theoretically by domain wall movements and moment rotations [4–7]. It has also been experimentally demonstrated by MI effect measurements [3, 5–7], and the transverse domain structure has been directly observed by a combination of transmission electron microscopy, electron diffraction, and magneto-optical Kerr effect microscopy [4]. However, clarifying the relevant mechanism, especially on a microscopic scale, still needs further investigation.

In this work, a series of nanocrystalline  $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  samples were prepared by annealing at the optimal temperature, each with a different longitudinal tensile

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stress up to 60 MPa. Their MI effect was measured and the effective field of transverse magnetic anisotropy  $H_k$  for each sample was evaluated. In order to obtain information on the distribution of magnetic moment orientations in these stress-annealed samples, a pair of Mössbauer spectra were recorded from each sample in two distinct orientations, one with the incident  $\gamma$ -rays perpendicular to the ribbon surface, and the other with  $\gamma$ -rays making an angle of  $30^\circ$  with the normal of the ribbon surface. By analyzing the intensity ratios of the second and third lines in the sextets, a transverse magnetic moment was revealed that increases with the increasing longitudinal tensile stress applied during annealing. Because Mössbauer effect uses  $^{57}\text{Fe}$  nuclei as probes for local magnetic moments, this work provides microscopic evidence for the transverse magnetic structure in the stress-annealed nanocrystalline alloys.

## 2. Experimental

$\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  amorphous ribbons were prepared by the single-roller melt-spinning method. Subsequently, three ribbon samples (each with 8 mm in width and  $25\ \mu\text{m}$  in thickness) were annealed in a nitrogen atmosphere for 30 min at the same temperature of  $540\ ^\circ\text{C}$  but under different longitudinal tensile stresses of 24, 48 and 60 MPa, respectively. All samples were cooled down to room temperature with the stress applied.

Magneto-impedance (MI) effect was measuring using a home-made computer-controlled system, in which the sample was placed at the center of a pair of Helmholtz coils which produced a dc magnetic field up to  $\pm 40$  Oe, along the longitudinal axis of the samples. The entire assembly was oriented such that this field was perpendicular to the Earth's magnetic field [3]. Impedance data were measured with an alternating current of  $I_{\text{rms}} = 1$  mA and  $f = 2$  MHz.

$^{57}\text{Fe}$  Mössbauer spectra were recorded at room temperature using a constant acceleration spectrometer with a  $9.2 \times 10^8$  Bq  $^{57}\text{Co}(\text{Pd})$  source in transmission geometry. Spectral parameters were obtained from computer-fitting and the isomer shift was calculated with respect to an  $\alpha$ -Fe foil of  $25\ \mu\text{m}$  thickness. For each sample, two Mössbauer spectra were obtained with the ribbon sample in two distinct orientations: 1. with the incident  $\gamma$ -rays along the normal direction of the ribbon plane, i.e. along the  $z$ -axis as shown in Fig. 1; and 2. with the incident  $\gamma$ -rays making an angle of  $30^\circ$  with the normal of the ribbon surface. The  $x$ -axis is defined to be along the longitudinal direction of the ribbon, and the  $y$ -axis along the transverse direction.

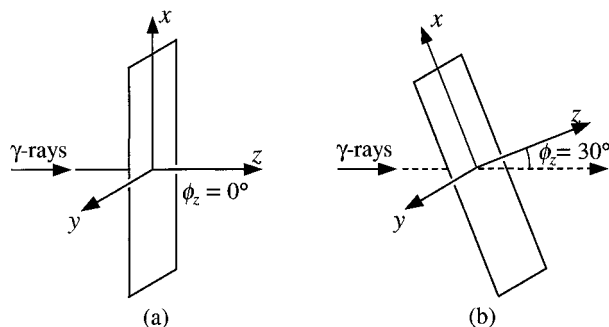


Fig. 1. Two orientations of a ribbon sample relative to the incident  $\gamma$ -ray direction for Mössbauer effect measurement a)  $\phi_z = 0^\circ$ , b)  $\phi_z = 30^\circ$

### 3. Result and Discussion

Figure 2 shows the MI ratio as a function of the external magnetic field  $H_{\text{ex}}$  for the as-cast sample ( $\sigma = 0$ ) and three stress-annealed samples with tensile stress values of 24, 48 and 60 MPa, respectively. The MI ratio is defined as  $\Delta Z/Z = [Z(H_{\text{ex}}) - Z(0)]/Z(0)$ . For the as-cast sample, the MI ratio declines with increasing  $H_{\text{ex}}$  and no positive MI effect was observed. For the stress-annealed samples with a nanocrystalline structure, the MI ratio vs.  $H_{\text{ex}}$  curve shows a positive MI effect with a clear maximum. These typical positive MI curves have been observed arising from all the tensile stress-annealed samples which feature a transverse magnetic anisotropy [5, 7]. The  $H_{\text{k}}$  values for the three stress-annealed samples were measured from the peak locations of MI curves [3] and plotted in Fig. 3a against the stress values. Within experimental uncertainty, the relationship between the effective field of transverse anisotropy  $H_{\text{k}}$  and the tensile stress  $\sigma$  is essentially linear.

In our previous work [5], longitudinal dynamic hysteresis loops indicated the existence of a transverse magnetic structure in  $\text{Fe}_{73}\text{Cu}_1\text{Nb}_{1.5}\text{V}_2\text{Si}_{13.5}\text{B}_9$  after longitudinal tensile stress-annealing. But hysteresis measurements only provided information on a macroscopic scale. In this work, Mössbauer spectroscopy is applied for a microscopic study on the transverse magnetic structure.

It is well known that the intensity ratio between the second and third peaks in a Mössbauer sextet contains information on the orientation of the magnetic hyperfine field at the Fe nuclei with respect to the  $\gamma$ -ray propagation direction. In the Fe-based nanocrystalline materials, the magnetic moments are in the direction opposite to the magnetic hyperfine field. If the  $\gamma$ -rays make an angle of  $\theta$  with the local magnetic moment, the angular factors for the second peak and the third peak are  $S_2 = \sin^2 \theta$  and  $S_3 = (1 + \cos^2 \theta)/4$ , respectively [8]. In our nanocrystalline materials, there is a distribution of magnetic moment orientations. If we assume that the  $i$ -th group of magnetic moments has an orientation that makes an angle of  $\theta_i$  with the  $\gamma$ -rays, and the probability of a magnetic moment belong to this group is  $P_i$  then the resulting angular

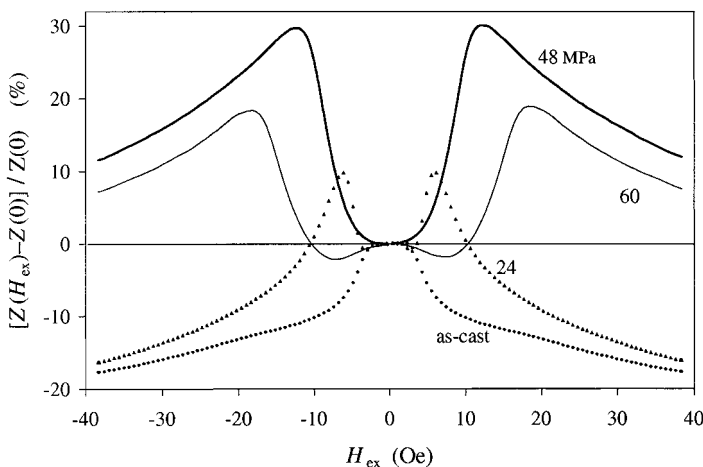


Fig. 2. Magneto-impedance (MI) ratio curves for four samples of increasing tensile stress values. The transverse anisotropic field  $H_{\text{k}}$  is the  $H_{\text{ex}}$  value that corresponds to the peak MI ratio

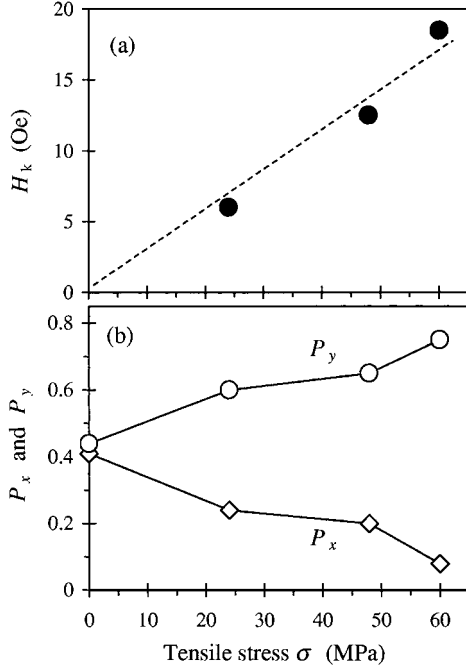


Fig. 3. a) Effective field of transverse magnetic anisotropy  $H_k$  as a function of tensile stress  $\sigma$ . b) Longitudinal and transverse axial parameters ( $P_x$  and  $P_y$ ) of the preferential degree of the orientation of magnetic moments as functions of the tensile stress  $\sigma$ . The induced transverse magnetic moment increases at the expense of the longitudinal component

factors are

$$S_2 = \sum P_i \sin^2 \theta_i, \quad (1)$$

$$S_3 = \sum P_i (1 + \cos^2 \theta_i)/4, \quad (2)$$

and their ratio is

$$A = \frac{\sum P_i \sin^2 \theta_i}{\sum P_i (1 + \cos^2 \theta_i)/4}. \quad (3)$$

When a Mössbauer spectrum is analyzed, we can obtain the area fraction of each subspectrum (equivalent to  $P_i$ ) and the second and third peak angular factors  $\sin^2 \theta_i$  and  $(1 + \cos^2 \theta_i)/4$ . Therefore,  $S_2$

and  $S_3$  can be determined experimentally, and the angular factor ratio  $A$  can be evaluated from each spectrum.

In order to describe an overall preferred direction of magnetic moments, we will use the following definition for a preferential degree of orientation of magnetic moments [9]:

$$P_0 = \sum P_i \cos^2 \theta_i. \quad (4)$$

Since  $\sum P_i = 1$ , this definition allows a simple relation between  $P_0$  and  $A$ ,

$$\begin{aligned} A &= \frac{\sum P_i \sin^2 \theta_i}{\sum P_i (1 + \cos^2 \theta_i)/4} = 4 \frac{\sum P_i (1 - \cos^2 \theta_i)}{\sum P_i (1 + \cos^2 \theta_i)} = 4 \frac{\sum P_i - \sum P_i \cos^2 \theta_i}{\sum P_i + \sum P_i \cos^2 \theta_i} \\ &= 4 \frac{1 - P_0}{1 + P_0}. \end{aligned}$$

Solving for  $P_0$ , we obtain

$$P_0 = \frac{4 - A}{4 + A}. \quad (5)$$

If a principal coordinate system is chosen and the magnetic moment distribution is symmetric about the principal axes (e.g., the  $x$ ,  $y$ , and  $z$ -axes as defined in Fig. 1),  $P_0$  is also related to two sets of physically meaningful quantities [9],

$$P_0 = P_x \cos^2 \phi_x + P_y \cos^2 \phi_y + P_z \cos^2 \phi_z \quad (6)$$

where  $P_x$ ,  $P_y$ , and  $P_z$  are known as the three axial parameters of the preferential degree of orientation, while  $\phi_x$ ,  $\phi_y$ , and  $\phi_z$  are the angles that the  $\gamma$ -ray direction makes with the  $x$ ,  $y$ , and  $z$ -axes, respectively. We can control the angles  $\phi_x$ ,  $\phi_y$ , and  $\phi_z$

by properly orienting the sample in the Mössbauer experiment, therefore, these are known geometric quantities. The axial parameters ( $P_x$ ,  $P_y$ , and  $P_z$ ) actually represent the probabilities of the overall magnetic moment along each of the axis,

$$P_x + P_y + P_z = 1. \quad (7)$$

For a ribbon sample,  $P_x$  is a measure of the longitudinal component of the magnetic moment,  $P_y$  is a measure of the transverse component, and  $P_z$  is a measure of the normal component. It becomes clear that these parameters indicate the distribution of magnetic texture and thus quantify the nature of magnetic anisotropy.

To calculate the three axial parameters  $P_x$ ,  $P_y$ , and  $P_z$ , a minimum of two independent equations in the form of Eq. (6) are necessary. Accordingly, for each sample, our Mössbauer experiments were performed such that in the first sample orientation (Fig. 1a),  $\phi_x = \phi_y = 90^\circ$  and  $\phi_z = 0^\circ$ . Spectral analysis gave an angular factor ratio  $A_1$  evaluated according to Eq. (3). Combining (5) and (6),

$$\frac{4 - A_1}{4 + A_1} = P_z \quad (\text{for } \phi_z = 0^\circ). \quad (8)$$

In the second sample orientation (Fig. 1b),  $\phi_x = 120^\circ$ ,  $\phi_y = 90^\circ$ , and  $\phi_z = 30^\circ$ . The spectrum gave a different angular factor ratio  $A_2$ , and when (5) and (6) were combined for this case,

$$\frac{4 - A_2}{4 + A_2} = P_x \cos^2 120^\circ + P_z \cos^2 30^\circ \quad (\text{for } \phi_z = 30^\circ). \quad (9)$$

From Eqs. (7), (8), and (9), the three parameters  $P_x$ ,  $P_y$ , and  $P_z$  were easily calculated.

Figure 4 shows representative Mössbauer spectra corresponding to  $\phi_z = 0^\circ$  and  $30^\circ$ , from each of the samples (as-cast and annealed with  $\sigma = 48$  MPa). All spectra were fitted using four sub-spectra resulting from the nanocrystalline  $\alpha$ -Fe(Si) phase (see Ref. 10) and one broad sextet corresponding to the residual amorphous phase.

Table 1 shows a complete list of the hyperfine parameters that produced the best fits of the spectra and the angular factor ratios  $A = S_2/S_3$  for the four samples of  $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  with  $\sigma = 0, 24, 48, 60$  MPa. The isomer shift values  $\delta$  and magnetic hyperfine field values  $H$  for the first four sub-spectra are consistent with the data from a classic Mössbauer study of FeSi alloys by Stearns [10]. The fifth subspectrum represents  $^{57}\text{Fe}$  in the amorphous phase. The data in the last column of Table 1 were calculated according to Eq. (3).

Table 2 summarizes the experimental and calculated data from both the MI measurements and Mössbauer spectra. The transverse anisotropy fields  $H_k$ , obtained from the peak locations of the MI ratio curves in Fig. 1, increases with increasing tensile stress (see also Fig. 3a). The angular factors  $A_1$  and  $A_2$  are from the last column in Table 1, and the axial parameters  $P_x$ ,  $P_y$ , and  $P_z$  were calculated using Eqs. (7), (8) and (9). Because  $P_x$ ,  $P_y$ , and  $P_z$  represent a degree of preferred orientation of the magnetic moments along the coordinate axes, they provide microscopic information on the magnetic structure. It is clear that as the tensile stress increases,  $P_x$  (longitudinal component) decreases and  $P_y$  (transverse component) increases, directly confirming that stress-annealing induces transverse anisotropy. Figure 3b shows the evolution of  $P_x$  and  $P_y$  with increasing stress  $\sigma$ , and demonstrates that the transverse magnetic moment is induced and increases at the expense of the longitudinal component. The normal com-

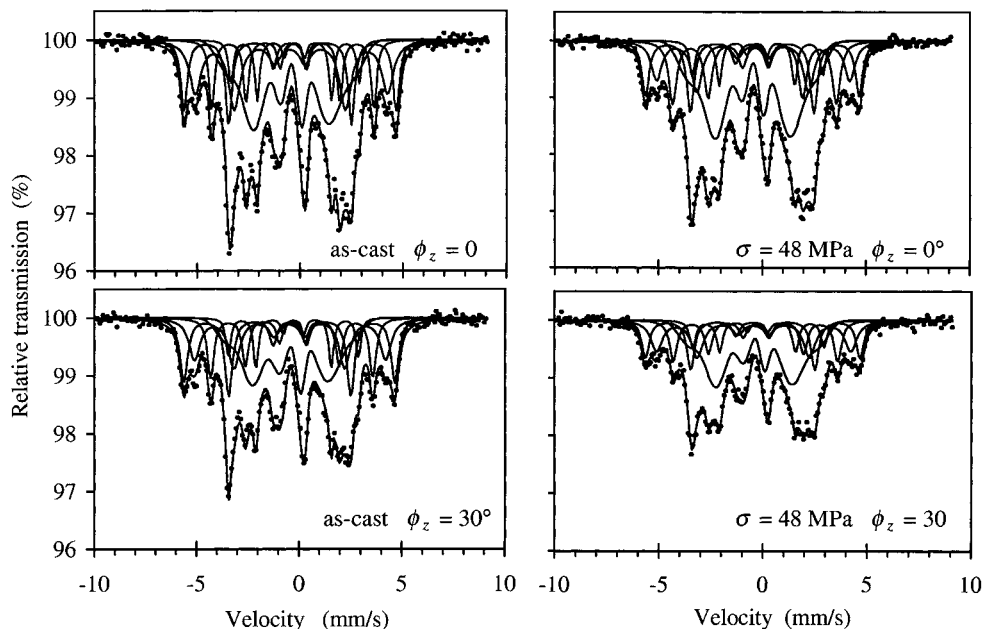


Fig. 4. Representative Mössbauer spectra from  $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  ribbon samples. Left side: from the as-cast sample; right side: from the sample annealed at  $540^\circ\text{C}$  under  $\sigma = 48\text{ MPa}$ . Top parts: first orientation,  $\phi_z = 0^\circ$ ; bottom parts: second orientation,  $\phi_z = 30^\circ$

ponent  $P_z$  is constant (see Table 2), but non-zero. This indicates that, although the “intrinsic” magnetic moment vector may not be completely within the ribbon surface, the induced transverse magnetic moment is strictly along the width of the ribbon. These results demonstrate that a large number of magnetic moments turn to the direction perpendicular to the ribbon axis within the ribbon plane and a transverse magnetic anisotropy is developed after annealing under a tensile stress.

Table 1

A complete list of hyperfine parameters that were used for fitting the Mössbauer spectra from the four samples, each in two different orientations. Isomer shift  $\delta$  is in mm/s and the magnetic hyperfine field  $H$  is in the kOe. The angular factor ratios of the second and third peaks  $A = S_2/S_3$  are also listed

Sample	sample orientation	subspectrum 1		subspectrum 2		subspectrum 3		subspectrum 4		subspectrum 5		$A = S_2/S_3$
		$\delta$	$H$	$\delta$	$H$	$\delta$	$H$	$\delta$	$H$	$\delta$	$H$	
as-cast	$0^\circ$	0.12	319.33	0.12	289.16	0.23	244.13	0.33	193.27	0.16	195.75	2.96
	$30^\circ$	0.11	318.51	0.12	288.54	0.24	244.62	0.31	196.44	0.16	195.75	2.58
$\sigma = 24\text{ MPa}$	$0^\circ$	0.10	320.84	0.11	292.73	0.23	243.10	0.34	193.61	0.12	193.42	2.89
	$30^\circ$	0.10	318.58	0.13	287.99	0.24	244.51	0.34	196.43	0.11	193.42	2.77
$\sigma = 48\text{ MPa}$	$0^\circ$	0.07	319.75	0.07	292.42	0.20	243.12	0.30	192.79	0.12	193.42	2.92
	$30^\circ$	0.09	318.90	0.10	288.00	0.24	243.47	0.33	193.71	0.12	193.42	2.86
$\sigma = 60\text{ MPa}$	$0^\circ$	0.10	318.51	0.11	287.45	0.23	243.54	0.33	194.40	0.12	193.42	2.81
	$30^\circ$	0.08	318.86	0.07	288.68	0.22	244.92	0.30	198.20	0.12	198.08	2.95

Table 2

A list of the transverse anisotropy field  $H_k$  obtained from the MI ratio curves, the angular factor ratios  $A_1$  and  $A_2$  for the two orientations, and the calculated axial parameters ( $P_x$ ,  $P_y$ , and  $P_z$ ) for the four  $\text{Fe}_{73.5}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  samples

Sample	$H_k$ (Oe)	$A_1$ ( $\phi_z = 0^\circ$ )	$A_2$ ( $\phi_z = 30^\circ$ )	$P_x$	$P_y$	$P_z$
as-cast		2.96	2.58	0.41	0.44	0.15
$\sigma = 24$ MPa	6.0	2.89	2.77	0.24	0.60	0.16
$\sigma = 48$ MPa	12.5	2.92	2.86	0.20	0.65	0.15
$\sigma = 60$ MPa	18.5	2.81	2.95	0.08	0.75	0.17

Our MI measurements and Mössbauer study have provided two aspects of the induced transverse magnetic anisotropy,  $H_k$  and  $P_y$ , both of which increase almost linearly with increasing longitudinal tensile stress, as shown in Fig. 3. In our previous work [3], an external transverse magnetic field was applied during sample annealing, resulting in decreasing  $P_x$  and increasing  $P_y$ , as expected. In this work, a similar change in the magnetic structure has been induced by longitudinal mechanical tensile stress, and quantitative data have been provided by a microscopic method. For Fe-based nanocrystalline alloys, although the residual amorphous matrix has a positive magnetostriction ( $\lambda > 0$ ) and the  $\alpha$ -Fe(Si) grains have a negative one ( $\lambda < 0$ ), the orientation of the induced moments is, however, transverse to the tensile stress direction. This fact has been explained by the atomic pair ordering process in the FeSi nanocrystals [11], and is experimentally verified in this work by both MI effect measurements and Mössbauer spectroscopy.

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