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# Magnetization distribution in nanocrystalline $Fe_{0.48}Al_{0.52}$

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Mössbauer polarimetry is used for investigating the orientation of Fe magnetic moments in the nanocrystalline $Fe_{0.48}Al_{0.52}$ disordered alloy prepared by mechanical grinding. Local Fe magnetic moments and their contributions to the net magnetization at selected external fields and temperatures were estimated. It was found that the components of the Fe magnetic moments parallel to the net magnetizations reduce their values much faster with an increasing number of neighbouring Al atoms than the total iron moments.

Key words: *Mössbauer polarimetry; magnetic moment; nanocrystalline* $Fe_{0.48}Al_{0.52}$

## 1. Introduction

Structurally ordered alloys with Al concentrations between 35% and 43% and with equiatomic composition have been suspected of the presence of antiferromagnetic order [1–5]. Quite recent neutron diffraction data for *bcc*-ordered Fe–Al with Al concentrations of 34–43 at. % [6] showed the presence of incommensurate spin density waves. It has been found earlier that ferromagnetic order can exist close to FeAl composition [7], and that supposedly antiferromagnetic or paramagnetic configurations can change to ferromagnetic upon simple filing [1, 8–11]. Ferromagnetism has also been detected in alloys with Al concentrations larger than 50 at. % [8, 12] and in the concentration range 40–60 at. % [13].

The ratio of hyperfine fields and magnetic moments (or magnetization) in many Fe-based ferromagnetic systems is in the range 10.0–15.0 T/$\mu_B$ [14–16]. Magnetization data for disordered Fe–Al alloys [11, 17], can be compared with the average hyperfine magnetic field (h.m.f.) obtained from Mössbauer experiments [8, 12, 17]. The ratio is an-

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omalously high, about 30 T/$\mu_B$ for concentrations close to equiatomic, while it is about 12.0–13.5 T/$\mu_B$ in ferromagnetic alloys with Al concentrations less than 40 at. %.

For systems with atomic and spin disorders – in particular for non-ordered metal systems with RKKY interaction – Mattis has considered a model in which spin–spin interactions were random in sign but did not result in frustrations [18]. The spin system was considered to be composed of two subsystems with randomly distributed antiparallel spins, which form the so-called Mattis phase. The magnetisation behaviour of the Fe–Al system and the ideas found in Refs. [18–20] indicate that concentrated Fe–Al alloys could exhibit the Mattis phase, no experimental support, however, has been reported so far.

Studies of Fe–Al alloys by the Monte Carlo techniques [21] in the Al concentration range $0.25 < x < 0.50$ showed that it was possible to qualitatively account for the transformation of the magnetic state from a ferromagnetic one to a certain spin glass state at about 34% Al using the indirect exchange model [1]. A peculiar feature of this spin-glass is the fact that frustrations arise from the positional disorder of Fe and Al atoms.

These controversial findings, and the apparent impact of various defects and inhomogeneities on the magnetic structure of bulk and nanocrystalline alloys, require more detailed investigations that will elucidate the nature of magnetism in this system. This paper reports the studies of nanocrystalline Fe–Al alloys in the concentration range close to the onset of ferromagnetism by the Mössbauer technique with circularly polarized monochromatic radiation. Polarized radiation has been widely used in investigating magnetically ordered systems, for an extended review see Ref. [22]. Circularly polarized radiation has the advantage of being sensitive to the sign of the h.m.f. Our observations strongly suggest that a non-collinear magnetic order must exist in the sample studied.

## 2. Sample

Fe–Al ingot was synthesized from high-purity components (99.99 % Fe and 99.99% Al) in an induction furnace in Ar atmosphere, then homogenized in a vacuum furnace at 1400 K for 6 h. The chemical analysis showed that the Al concentration was 52.0±0.5 at. %. The ingots were milled and a fraction with particle sizes lower than 300 μm was used for further mechanical treatment in a planetary ball mill with vials and balls made of tungsten carbide. Mechanical grinding was performed in an inert gas atmosphere. The milling time was adjusted to provide a disordered state in the Fe–Al alloys [17]. The obtained sample exhibited a single *bcc* phase with the lattice parameter 0.2918(4) nm. The X-ray diffraction pattern consists of the broadened peaks (110), (200), (211), and (220) of the *bcc* structure. The analysis of the shape of the diffraction lines using the harmonic analysis [23] lead to the conclusion that the mean grain size of the alloy is 4.0(3) nm.

## 3. Mössbauer polarimetric measurements

The absorbers were placed inside one of the rare earth magnets, producing an axially symmetric field perpendicular to the absorber surface and parallel to the gamma rays. The measurements were performed at temperatures of 13, 50, and 298 K and external fields between 0 and 1.1 T, see the example in Fig. 1.

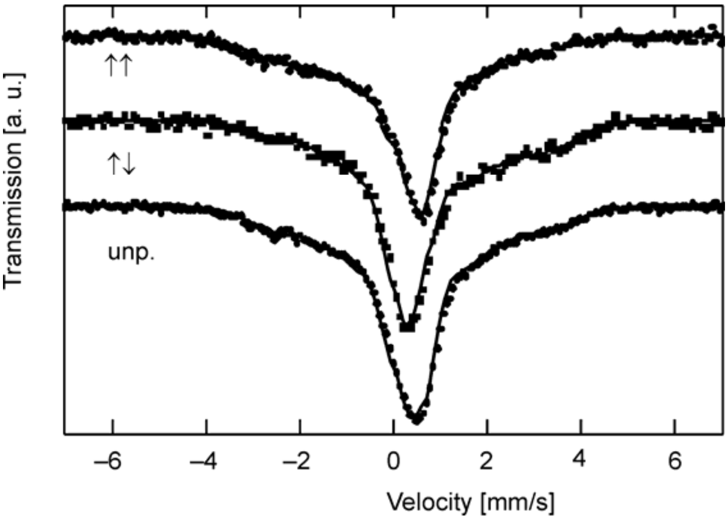

Fig. 1. An example of the Mössbauer spectra recorded at $T = 13$ K with polarized and unpolarized radiation in an axial, externally applied magnetic field of 1.1 T. The arrows ↑↑ and ↑↓ indicate two opposite circular polarizations. Solid lines represent the results of the simultaneous fitting the h.m.fs to all the recorded spectra

A full Hamiltonian was used for treating the mixed magnetic dipole and electric quadrupole interaction. The transmission integral was used to treat thickness effects, and the temperature dependence of the *f*-factor was taken into account. To analyse the low-temperature data, we assumed the same isomer shift and electric field gradient, as well as their probabilities determined at room temperature. We allowed only the second order Doppler shift and the appearance of the h.m.f. distribution. The principles of the adopted polarimetric method are given in Ref. [24], while details of the construction of the source of circularly polarized radiation in Refs. [25, 26].

An experiment with circularly polarized radiation is sensitive to the angular average $\langle \boldsymbol{\gamma}_r \cdot \mathbf{m} \rangle$, where **m** is a unit vector parallel to the local h.m.f., $\mathbf{B}_{hf}$, $\boldsymbol{\gamma}_r$ is the Cartesian vector ($r = x, y, z$) and the brackets $\langle\ \rangle$ denote angular averaging with the magnetic texture function $P(\Omega)$:

$$\langle \boldsymbol{\gamma}_r \cdot \mathbf{m} \rangle = \int_{4\pi} \boldsymbol{\gamma}_r \cdot \mathbf{m}\, P(\Omega)\, \mathrm{d}\Omega \tag{1}$$

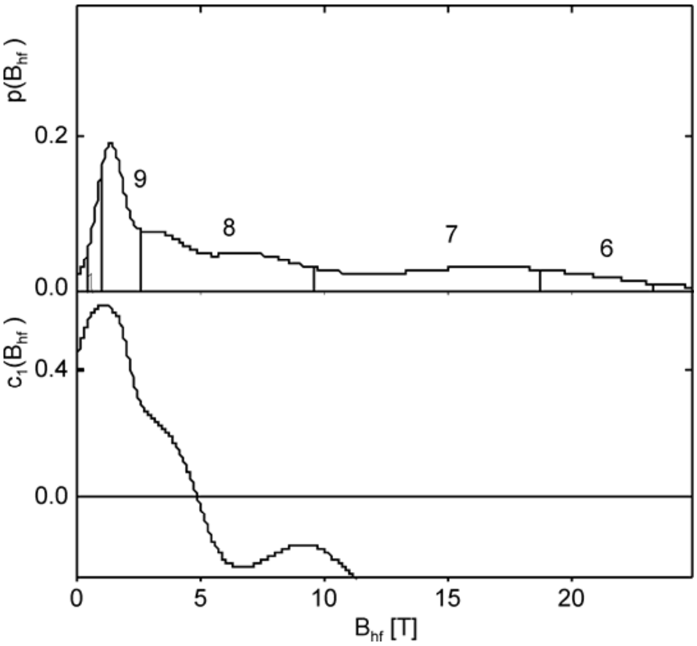

Fig. 2. An example of the distribution of h.m.f. obtained from experiment (up) and the h.m.f. dependence of the average cosine between the h.m.f. vector and magnetization (down) measured at $T = 13$ K and $B_{\mathrm{ext}} = 1.1$T. The vertical bars split the $p(B_{hf})$ distribution into sectors numbered by $k = \ldots, 9, 8, 7 \ldots$, for which the two first coordination shells are taken into account (cf. Eq. (2))

In particular, one can obtain the value of $c_1B_{hf}$, which is an average component of the h.m.f. along photon directions. In the case of α-Fe, the h.m.f. is antiparallel to the direction of the magnetic moment of iron atoms [27], so the $c_1$ parameter is negative. Negative values of $c_1$ for our sample are observed only for hyperfine fields larger than about 5 T (Fig. 2). Figure 2 also shows that a relatively large probability $p(B_{hf})$ is observed in the region close to $B_{hf} = 5$ T, much above the intensity of the external field. It is thus reasonable to suppose that $B_{hf}$ in this region corresponds to the Fe atoms with nonzero magnetic moments. If this is so, then a nearly zero value of the $c_1$ parameter in Fig. 2 indicates that these moments, on average, do not contribute to the total magnetization.

## 4. Discussion and conclusions

A considerable enhancement of the experimental value $B_{hf}/\sigma_{Fe}$ (attaining values close to 30 T/$\mu_B$) in the studied Fe–Al system with respect to the values calculated for collinear systems can be explained by a transformation from ferromagnetic to nonferromagnetic order. One should expect that the magnetization should be correlated with the $z$-th component of the h.m.f., $B_z$. This is indeed observed in our alloy $Fe_{0.48}Al_{0.52}$ (Fig. 3).

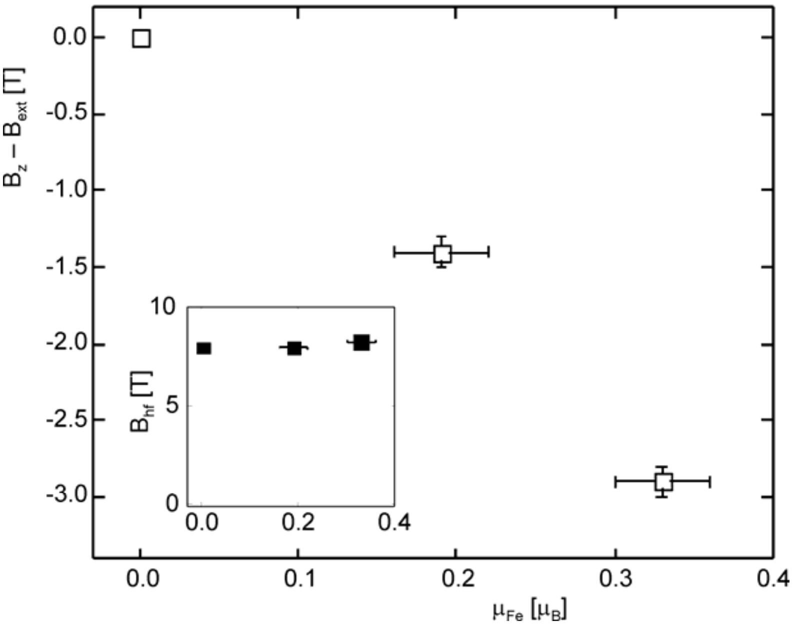

Fig. 3. Correlation between the average $z$-component of the h.m.f. (corrected to the external field) and the Fe magnetic moment (the inset shows a lack of correlation between the average h.m.f. and magnetic moment). Open and full symbols correspond to the $z$-component of the h.m.f. and the h.m.f. itself, respectively

It is reasonable to assume that the larger number of Fe atoms, the larger the observed $B_{hf}$. Next, it follows from EXAFS results [28] that the probabilities of finding the Fe atom in the first (I) and the second (II) coordination shells are given by $x_I = 0.10$ and $x_{II} = 0.883$, respectively. Assuming random distributions within the shells, the probability $P(k)$ that an iron atom is surrounded by $(14 - k)$ Fe and $k$ Al atoms in the two first coordination shells is:

$$P(k) = \sum_{\substack{i,j \\ i+j=k}} \binom{8}{j}\binom{6}{i} x_I^{\,8-j}(1-x_I)^j x_{II}^{\,6-i}(1-x_{II})^i \tag{2}$$

These assumptions allow one to divide $p(B_{hf})$ into a few sections, each having the area proportional to the probability $P(k)$. The sectioning is displayed in Fig. 2 by vertical lines. The average fields estimated for each section are displayed in Fig. 4 by full symbols, while the $z$-components of the fields by open symbols. As expected, an increase of the field from $B_{ext} = 0.2$ T to $B_{ext} = 1.1$ T results in an increase of $B_z$ and not $B_{hf}$ (see the open and the overlapping full points in Fig. 4). The average $\overline{B}(k)$ for $k = 6$ (Fig. 4) is slightly above the value related to ordered $Fe_3Al$ at low temperature [29].

It should be noted that Eq. (2) is valid under the assumption that the contributions of neighbouring atoms from the I and II coordination shells are the same, which is an approximation. The opposite approximation, in which the influence of the II coordination shell is neglected, can be made by using the binomial distribution instead of Eq. (2). The results of such treatment exhibit similar features and are not shown here (average $\overline{B}(k)$ for $k = 4$ is slightly below the value related to ordered $Fe_3Al$ reported in [29]).

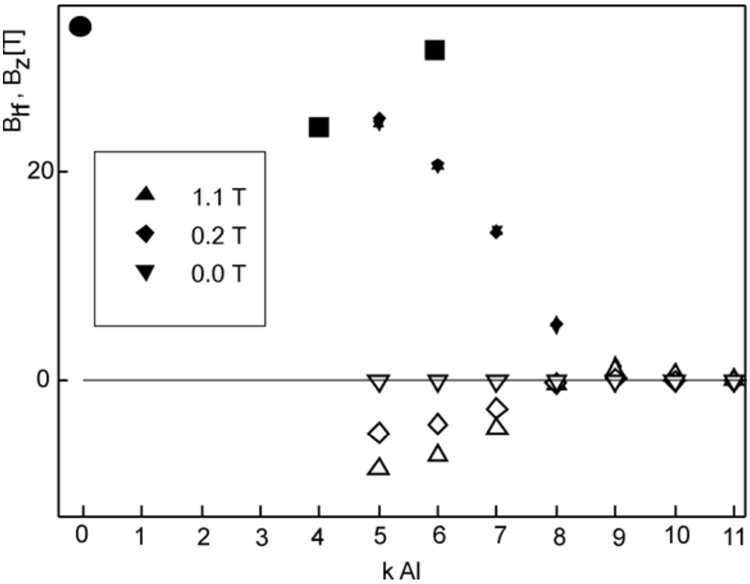

Fig. 4 Average h.m.f. and average *z*-th component of the field (not corrected for the external field) related to the sectors from Fig. 2, under the assumption that contributions from the first and the second coordination shells to the h.m.f. are the same. Full symbols correspond to the h.m.f., open symbols to the z-component of the h.m.f., • – Ref [16], ■ – Ref. [29]

The magnetization and Mössbauer data can be combined to obtain the local magnetic moments of Fe. To estimate the *z*-th component of the magnetic moment in the external magnetic field, we assume that it is proportional to $\overline{B}_z(k)$ and that the total contribution to the magnetization of all Fe atoms is equal to the measured magnetization. The two most important results are as follows.

An increase of the number of Al atoms in the two first coordination shells (*bcc* structure) causes a decrease of the magnetic moments and a much faster decrease of the *z*-component of the magnetic moments (local contribution to magnetization). Configuration (7, 1) was found as the most suitable for forming a non-collinear structure, where ($k_1$, $k_2$) denotes a Fe atom surrounded by $k_1$ Al atoms in the I coordination shell and $k_2$ Al atoms in the II coordination shell. It should be stressed that such a detailed description of the magnetic moment arrangement would hardly be possible if not for the extensive combination of experimental methods used in the present studies.

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