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INHOMOGENEOUS PROPERTIES OF IONIC SEMI-SPIN-GLASSES STUDIED BY MÖSSBAUER SPECTROSCOPY

M. Zemirli ⁽¹⁾ ¹, J. M. Grenèche ⁽¹⁾, F. Varret ⁽¹⁾, M. Lenglet ⁽²⁾ and J. Teillet ⁽²⁾

⁽¹⁾ UA-CNRS n° 807, Université du Maine, 72017 Le Mans Cedex, France

⁽²⁾ INSA de Rouen, LPCM, B.P. 8, 76131 Mont Saint Aignan, France

Abstract. – We have analysed in terms of static hyperfine field distributions the Mössbauer spectra of mixed Zn-Mg ferrites. Typical features of semi-spin glasses are observed both in average and locally: in each sample, the relevant data are smeared out in a way which reveals the behavior of the series.

1. Introduction

Mössbauer spectroscopy of ferrites provides the advantage of weak quadrupole interactions (high-spin Fe^{III}) and well-defined saturation value of the hyperfine field (H_{hyp}); this allows an accurate analysis of H_{hyp} distributions, easily revealing the variation of the magnetic moments with temperature or applied magnetic field. On the other hand, short-range magnetic interactions lead to highly inhomogeneous magnetic properties, for which the description in terms of magnetic cluster glass [2] may be adequate.

We have studied the mixed spinels $(\text{Zn}_x\text{Fe}_{1-x})[\text{Mg}_{1-x}\text{Fe}_{1-x}]\text{O}_4$, where () and [] respectively stand for A and B sites. The available compositions, $x = 0.6, 0.7, 0.8, 0.9$, are close to the percolation threshold of A-B interactions [1], where a re-entrant spin glass behavior is expected.

We summarize here the results [2] of a static analysis of Mössbauer spectra, in terms of H_{hyp} -distribution, depicting the inhomogeneous behavior of weakly coupled magnetic clusters [3]. These clusters originate from the introduction of strong interactions of A-B type, diluted into a matrix with much weaker (and frustrated) B-B interactions. The relevance of such static analysis is supported by the observation that Mössbauer spectra (Fig. 1) in a large range of temperatures consist of superimposed quadrupole and magnetically-split contributions, so that only few iron atoms have their magnetic relaxation time in the Mössbauer window ($\approx 10^{-8} - 10^{-9}$ s). This discussion, together with the detailed magnetic study (high-field Mössbauer spectroscopy and magnetization measurements) will be developed in forthcoming papers.

2. Average Mössbauer data

Since A and B sites are not resolved in the Mössbauer spectra out of field, the following data re-

Fig. 1. – Typical Mössbauer spectra fitted by a static discrete distribution of hyperfine fields and a quadrupole doublet.

fer to “average” sites, which are predominantly B sites (from 80 % to 95 % for x ranging from 0.6 to 0.9, respectively).

From the thermal variation of the measured average hyperfine field $\langle H_{\text{hyp}} \rangle$, the magnetic phase diagram was established (Fig. 2); the re-entrant transition is detected by a kink [4] on $\langle H_{\text{hyp}}(T) \rangle$ curve. The disappearance of the magnetic pattern is interpreted, as usual, rather in terms of clusters blocking than in terms of a true Curie transition.

3. Inhomogeneous analysis

Here we follow a static analysis previously described [5], for which each cluster type is characterized by the thermal variation of a hyperfine field H_n . Provided the curves $H_n(T)$ do not intersect, each cluster can be characterized by the probability $Q_T(H_n)$ cumulated from 0 to H_n .

Sections of the surface $Q = f(T, H_n)$ by the planes $Q = Q_i$ yield the thermal variations of the hyperfine

¹ Present address: INES Génie Civil, Université de Tizi-Ouzou, 15000 Algeria.

Fig. 2. - Magnetic phase diagram obtained from average Mössbauer data (dashed line) and from inhomogeneous analysis (full line) of sample $x = 0.6$.

Fig. 3. - Set of curves $H_n(T)$ for various values of the labelling parameter Q . The determination of the canting angle is shown for $Q = 0.4$: $|\cos \theta| = H_{0.4} / H_0$.

fields of the clusters labelled by the Q_i (Fig. 3). The data treated this way show the presence of a kink on

each curve; the canting angles are obtained from the ratio $H_i / H_0 = |\cos \theta|$ (according to [4]) where H_i and H_0 are extrapolated values at 0 K of the hyperfine field, in the high and low temperature range, respectively. They vary from almost 0 to 90°, following the labeling parameter Q . Also, the re-entrance temperatures vary according to Q .

It is likely that the labeling parameter follows the strength of the magnetic coupling of the various clusters, which depends on the local density of strong A-B interactions, i.e. the local density of tetrahedral irons. We have calculated, as a function of x , the correlation $Q - y$, where y is a "local" zinc content deduced from a binomial analysis of the magnetic first-neighbours [2] so that the phase diagram could be built up again, in the $T - y$ plane, from the data of a single sample. This is done in figure 2 for the data of $x = 0.6$ (full line); it roughly agrees with the former (dashed line).

High field Mössbauer study [2] confirms this inhomogeneous magnetic properties, with a spread of canting angles similarly correlated to the labeling parameter Q .

The sensitivity of the magnetic structure to local composition (and to external field [2]) probably originates from the frustration of the B sub-lattice.

The compositions near the antiferromagnetic structure ($0.8 < x < 1$) are under study.

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