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

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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 Fe3+) 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 (Zn$_x$Fe$_{1-x}$) [Mg$_{1-x}$Fe$_x$]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 refer 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 ($H_{hyp}$), the magnetic phase diagram was established (Fig. 2); the re-entrant transition is detected by a kink [4] on ($H_{hyp}$ (T)) 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...
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 $90^\circ$, 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.