Temperature dependence of the high-field magnetization in Fe-Ag granular alloys and discontinuous multilayers

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Abstract. Fe-Ag co-deposited granular alloy and discontinuous multilayer samples with 10-36 at% Fe average compositions were prepared by vacuum evaporation onto Si(111) single crystal substrates at room temperature. The magnetic properties of the samples were studied by SQUID and in-field Mössbauer spectroscopy. Superparamagnetic behaviour was observed for all the samples with blocking temperatures ($T_B$) steeply increasing with increasing Fe concentration. The bulk magnetization ($M$) measured in 5 T shows linear temperature dependence for both types of samples, and the relative decrease with temperature is much larger for a granular alloy than for a multilayer sample of equal $T_B$. The Mössbauer spectroscopy results indicate that the faster decrease of the high field magnetization in granular alloys is explained by the larger amount of small grains that are not aligned by the 5 T field.

1. Introduction
Granular systems with grain sizes in the nanometer range and ultrathin multilayers are subject of intensive study. The magnetic properties show novel phenomena which are interesting both from theoretical and application points of view. In such systems the saturation magnetization (or the magnetization measured in high fields) is often a linear function of the temperature. In case of multilayers this behaviour is mostly explained (e.g. Fe/Cr superlattices [1], Cu/Fe multilayers [2], FeNiB/Ru multilayers [3]) by the reduced dimensions. On the other hand, the linear temperature dependence of the magnetization in high fields is a characteristic feature of superparamagnetic granular alloys [4]. In order to see which factor influences more significantly the temperature dependence of the high field magnetization, we compared the magnetic behaviour of co-deposited Fe-Ag granular alloys and ultrathin multilayers.

2. Experimental
Fe-Ag co-deposited and multilayer samples were prepared by evaporation from Knudsen cell in a molecular beam epitaxy (MBE) equipment and by vacuum evaporation using electron beam, respectively, onto Si(111) single crystal substrates at room temperature. The average composition ($x$) of the samples varies between 10 and 36 at% Fe. It was achieved with different evaporation rates of the sources during co-evaporation and with a varying nominal Fe layer thickness (between 0.2 and
The Ag layer thickness, 2.6 nm, was kept constant in case of the multilayer samples and since the ultrathin Fe layers are not continuous these samples will be referred to as discontinuous or granular multilayers. The multilayer and the granular films were covered by a 50 and 10 nm thick Ag layer, respectively, in order to prevent oxidization.

The magnetic properties of the Fe$_x$Ag$_{100-x}$ (10 ≤ $x$ ≤ 36 at %) samples were studied by a SQUID magnetometer (Quantum-Design MPMS-5S) in wide temperature (5-300 K) and magnetic-field range (0-5 T). The multilayer samples were removed from the substrate with a Scotch tape and four pieces of 4 x 4 mm$^2$ squares were stacked. The granular alloys were measured in the as-received form after co-evaporation together with the substrate and for $x = 10$ also after removing from the substrate, the results being rather similar. Moreover, in-field Mössbauer spectroscopy measurements were performed using a standard constant acceleration spectrometer and a Janis cryostat equipped with a 7T superconducting magnet supplying magnetic field parallel to the γ-ray direction. Here both types of samples were removed from the substrates with a Scotch tape and the removed pieces were cut into 8 x 8 mm$^2$ squares and stacked.

3. Results and discussion

In figure 1 the temperature dependence of the low-field magnetization (measured in $H = 100$ Oe after cooling in zero field, ZFC, and in $H = 100$ Oe, FC) is shown both for the multilayer with a nominal Fe thickness of 0.2 nm (corresponding to the average composition of Fe$_{10}$Ag$_{90}$) and for the co-evaporated granular alloy with $x = 10$. Both samples show a typical superparamagnetic behaviour with a similar blocking temperature ($T_B = 26$ and 32 K for the granular multilayer and the granular alloy, respectively). The blocking temperature moderately depends on the measuring field being $T_B = 40$ and 45 K in $H = 10$ Oe for the two alloys. It suggests that the average size of the magnetic clusters are rather similar (≈ 800 µB or 1.8 nm diameter if spherical granules are assumed [5]).

The superparamagnetic behaviour indicated by the magnetization measurements in low field was observed for all the Fe$_x$Ag$_{100-x}$ (10 ≤ $x$ ≤ 36 at %) samples (not shown here) with blocking temperatures steeply increasing with increasing Fe concentration.

The magnetization was also measured as a function of the magnetic field up to 5 T. The magnetization curves can be fitted satisfactorily with a single Langevin function for both types of samples when the Fe-content is low (Fe thicknesses of 0.2 and 0.4 nm for the granular multilayers and the corresponding average compositions $x = 10$ and 18 for the granular alloys). For both types of samples containing higher Fe concentrations a constant plus a Langevin term are necessary to fit satisfactorily the magnetization curves. This latter description is supported by the Mössbauer measurements which indicate that the $T_B$ distribution of these samples stretches above room temperature. Therefore, at room temperature or below, the blocked part of the clusters can be well described as a constant contribution to the magnetization.

![Figure 1. Temperature dependence of magnetization in small field ($H = 100$ Oe) after zero-field cooling (ZFC, thin lines) and field cooling (FC, thick lines) the Fe$_{10}$Ag$_{90}$ granular multilayer and granular alloy.](image-url)
Figure 2 shows the temperature dependence of the magnetization measured in 5 T external field and normalized to the magnetization measured at 5 K in 5 T for the Fe$_{10}$Ag$_{90}$ granular alloy and the respective multilayer sample. A linear temperature dependence of the high field magnetization was observed for both types of samples, the relative decrease with temperature gradually decreasing with increasing average Fe content, but being always larger for the granular alloy than for the respective multilayer sample.

![Figure 2. Temperature dependence of the normalized magnetization measured in 5 T (open symbols) and average hyperfine field (solid symbols) for the Fe$_{10}$Ag$_{90}$ granular multilayer (squares) and granular alloy (circles).](image)

The Mössbauer spectra measured at 100 and 150 K in 5 T for the Fe$_{10}$Ag$_{90}$ granular multilayer and granular alloy are shown in figure 3. The spectra were evaluated allowing two distributions of the hyperfine fields, also shown in figure 3, which were fitted according to the Hesse-Rübartsch method [6]. The distribution above 10 T could be fitted by hyperfine fields close to parallel with the applied field (close to 3:0:1:0:3:0 intensity ratios within the individual sextets), while a random distribution of the hyperfine fields (3:2:1:2:3 intensity ratios) was allowed for the distribution below 10 T. The ratio of the low field distribution is clearly larger for the granular alloy as compared to the granular multilayer and for both samples increases with the temperature. The average values calculated from the two distributions and normalized with the 4.2 K average values are shown in Fig. 2 with full symbols and agree fairly well with the magnetic data. Even though the spectra were fitted by a distribution of static hyperfine fields, the Mössbauer measurements make it clear that the linear temperature dependence of the magnetization is mainly explained by the superparamagnetic behaviour for both samples. The significance of superparamagnetic relaxation even in 5 T applied field is indicated both by the increase of the relative weight of the low field fraction with temperature and by the variation of the high field component when the external field is increased further. A further 2 T increase of the external field produces only about 1 T decrease of the average magnetic splitting, which clearly indicates that the magnetic moments are not yet fully aligned in 5 T field. The decrease of the magnetic splitting in applied field results from the opposite direction of the magnetization and the hyperfine field. At 4.2 K the 2 T decrease could in fact be observed.) It is important to note, however, that the larger temperature variation of the average value for the granular alloy is mainly the result of the larger variation in the weight of the low field fraction, i.e. the more rapid increase of the number of unblocked (fast relaxing) particles. It increases from 12 to 16% for the alloy and from 7 to 8% in the multilayer sample. The average hyperfine fields taken only over the high field (slow relaxation rate) component differ less significantly for the two samples. These observations indicate that the distribution of $T_B$ is narrower in the granular multilayer samples than in the co-evaporated granular alloy samples. The faster decrease of the magnetization and the average hyperfine field for the granular alloy sample is mainly explained by the larger amount of small grains that are not aligned by the 5 T field.
Figure 3. Mössbauer spectra measured in 5 T external field at elevated temperatures. The fitted hyperfine field distributions are shown in insets and the subspectra belonging to the low and high field part (red lines and blue dashed lines, respectively) are also shown. The scale of $B_{hf}$ is from 0 to 40 T and that of $p$ is from 0 to 0.1 (first and last ticks).

4. Conclusions
A linear temperature dependence of the high-field magnetization and the average hyperfine field were observed for Fe$_x$Ag$_{100-x}$ ($10 \leq x \leq 36$ at %) granular multilayers and granular alloys. The relative decrease is similar for both measured quantities and is much larger for the granular alloys than for the multilayers of similar average Fe content. The Mössbauer measurements clearly indicate the presence of magnetic relaxation even in 5 T applied field and the faster decrease of magnetization is explained by the wider distribution of the blocking temperatures in the granular alloys.

5. References
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