Low temperature superspin glass behavior in a Co/Ag multilayer

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ABSTRACT

We report on the low temperature magnetic behavior of an epitaxially grown multilayer formed by 32 repetitions of a nominal period corresponding to 1 monolayer (ML) Co and 16 ML Ag. The study of the magnetic properties was based on the measurement of the temperature dependencies of the dc magnetization upon field cooling (FC) and zero field cooling (ZFC) and of the ac field real and imaginary parts of the susceptibility. From our results we conclude about the occurrence of i) a well-defined bilayers stacking sequence matching the nominal one, ii) a discontinuous growth in the Co layers resulting on close-to-monodisperse, spherical Co nanoparticles having an average diameter of 1.6 nm, iii) a frequency dependent peak in the temperature dependence of the real part of the ac susceptibility exhibiting a per decade relative temperature variation of 4.5 x 10^{-2}, iv) an applied dc field, H_{dc}, variation of the temperature at which the irreversibility is detected in the FC/ZFC curves corresponding to the Almeida-Thouless prediction, and v) a critical behavior characterized by a glass-transition temperature slightly below the peak temperatures observed at low frequency in the temperature dependence of the ac susceptibility and a dynamic scaling exponent in the range of the values usually obtained for spin glass systems. From our results we conclude that i) our sample experiences a superspin-glass/paramagnetic phase transition, ii) the interactions mediating the spin glass freezing process are the dipolar ones taking place among the Co particles (creating fields at the average interparticle distance of the order of 8 x 10^5 A/m) which provide competitiveness that combined with the reduced amount of disorder built-in the Co layers results on frustration.

INTRODUCTION

The phenomenology and the theoretical understanding of the spin glass phases have continued to be an active topic of research after the initial works on these phases were published at the beginning of the 1970's decade.[1] This is so due to several reasons among which it is worth to notice both the complexity of the experimental behavior,[2,3] which, as of today, still incorporates new phenomena[4] and the limited suitability of some of the (both standard and specifically developed) statistical mechanical tools that have been considered in order to theoretically analyze the experimental data (the experiments could, in many cases, be explained in the classical statistical mechanics framework if the difficulties linked to the built-in randomness could be tackled and overcome).

Also, and relevantly, from the structurally simple canonical spin glass metallic systems where indirect exchange between localized atomic moments is on the ground of the interactions originating the freezing behavior,[1] many different materials involving different crystallinities, morphologies, anisotropies, inter-moments interaction types and, above all, quenched disorder types and magnitudes have been identified and continue to be identified[5–9] as exhibiting spin glass phenomenologies. This includes many different two-phase, 3-D dispersed systems with magnetic atoms concentrations below the magnetic percolation limit, and involves from the materials with short-range coupled pairs and triplets of magnetic moments to the so-called cluster (superspins) glasses.[7]

Our work is framed into the latter research scenario,[10] since we aim at identifying, from its low temperature magnetic behavior, the actual occurrence of a spin glass freezing transition in a sample characterized by the restriction of the occurrence of disorder exclusively to well stacked magnetic layers separated by a non-magnetic spacer.

SAMPLE GROWTH AND EXPERIMENTAL TECHNIQUES

Our sample was grown by molecular beam epitaxy on a clean MgO (001) surface, by alternating Ag and Co deposition up to complete 32 periods. It was covered by a 3 nm Ag capping layer, with a nominal thickness per period of the Co and Ag layers corresponding to 1 and 16 monolayers (ML), respectively. The structural and morphologic characterization of the sample was carried out by means of X-rays reflectivity (XRR), and diffraction (XRD), and transmission electron microscopy (TEM), respectively. The magnetic characterization included the use of vibrating sample (VSM) and SQUID magnetometers (measuring temperatures from 2 K up to 290 K and fields of up to 9 T) and that of an ac susceptometer (measuring temperatures from 2 K up to 290 K and ac field frequencies from 10^{-2} up to 10^{4} Hz).

RESULTS AND DISCUSSION

The XRD measurements that were acquired in the sample clearly evidenced the achievement of a well-defined superlattice periodicity. The TEM imaging of the sample showed the fact that the Co layers did not grow continuously but with a morphology corresponding to a close-to-monodisperse distribution of Co particles having a lognormal diameter distribution with an average diameter of 1.6 ± 1 nm and embedded in a single-crystal Ag(001) matrix.

We have measured the low field dependence of the sample magnetization upon zero field-cooling (ZFC) and field-cooling (FC) it from 290 K down to 5 K. Our results show the occurrence of an irreversibility at T_{irr} = 14 K. As for the behavior of the sample at temperatures above its T_{irr}, we have fitted (in the temperature range from 30 K up to 90 K) our experimental data for the thermal dependencies of the ZFC, low field dc susceptibility, χ, to a Curie-Weiss law, $1/\chi = (T - \theta)/C$ (where θ is the ordering temperature and C is the Curie constant, given by $C = M_s V/3\, k_B$; where $M_s V$ is the average magnetic moment of the magnetic entities present in the sample, and k_B is the Boltzman constant[11]). The value obtained from the fit for the average moment of the magnetic entities is 4 x 10^{-15} Am^2, which is much larger than the fcc Co atomic moment (1.6 x 10^{-17} Am^2) and indicates that the paramagnetic behavior is related to the aggregation of ca. 200 individual Co moments having fcc co-ordination. From the TEM results, evidencing the presence in the sample of periodical particulate Co layers, it is straightforward to associate these large Co aggregates identified from the χ data to the Co particles observed by microscopy. The point is supported by the average paramagnetic moment of the sample obtained from the Curie-Weiss law data, which considering the bulk Co spontaneous magnetization value (1.42 MA/m[11]) corresponds to a diameter of the (assumed spherical) paramagnetic clusters responsible for the Curie-Weiss behavior of 1.9 ± 0.05 nm. This value is in good agreement with the TEM evaluation of the same parameter (1.6 ± 0.1 nm), which supports the identification of the Co particles as those responsible for the paramagnetic phenomenology.

Although the described dc magnetic behavior is compatible with the occurrence of a magnetic order phase transition, at temperatures of the order of the T_{irr}, from these results it is not possible to exclude the occurrence of blocking processes of the superparamagnetic type in the Co nanoparticles. Thus, and aiming at elucidating the actual occurrence of a magnetic order phase transition (and, if appropriate, at identifying the nature of that transition) we have measured and analyzed information about the ac low frequency, dynamic behavior of the sample. Figures 1 a) and a') show the temperature dependencies of the real, χ', and imaginary, χ", parts of the ac susceptibility[7] measured in the sample at applied ac field frequencies, *f*, in the range from 10^{-1} Hz up to 10 kHz. Maxima on the measured quantities are observed at temperatures of the order of the T_{irr} obtained in the dc measurements. The temperatures and peak magnitudes associated to the χ' maxima increase and decrease, respectively, with the frequency increase whereas in the case of the χ" maxima both the peak temperature and its magnitude increase with the frequency increase.

In order to detailedly describe the real part of the susceptibility dependency on temperature, we have fitted our χ' vs. T data to lognormal functions. The variation of the peaks temperatures T_{p_ac}, obtained from the fits, with the ac field frequency f is parameterized, by considering the relative increase of the peak temperature per decade, throughout $\Gamma = (\Delta\, T_{p\_ac}/T_{p\_ac})/\Delta\, log_{10} f$,[7] ($\Delta\, T_{p\_ac}/T_{p\_ac}$ *and* $\Delta\, log_{10}\, f$ are evaluated taking as reference f = 1 Hz). The experimental Γ value obtained is 0.05, unambiguously lower than those associated to superparamagnetic relaxation processes (in a large majority of materials, above 0.2[7]). In comparison to the Γ values measured in well identified spin glasses, our sample exhibits an ac field behavior corresponding to the upper limit of the Γ range commonly associated to the former phases (between 0.006 and 0.08[7]).

With the purpose of gaining a deeper insight on the actual occurrence in the sample of a collective, glassy-like behavior at temperatures below T_{irr} we have fitted our experimental data for the frequency variation of the thermal dependence of the χ' peaks to the Vogel-Fulcher (V-F) model.[12] This model has been widely contemplated for describing the temperature dependence of the characteristic times of the cooperative dynamics taking place in the freezing processes of spin glass-like systems (and more generally in glassy systems as, for instance, supercooled liquids.[13] The explicit form of the V-F law we used for the fits of the variation of T_{p_ac} with the frequency was $\omega = \omega_0\, exp\, [- E_a/k_B\, (T_{p\_ac} - T_0)]$, where ω is the ac field frequency and the fitting parameters ω_0, E_a and T_0 are the try frequency, the activation energy for the relaxation, and the magnitude in temperature units of the interactions present in the system, respectively. In Fig. 1 b) we have plotted the fit of our data to the V-F law evidencing a reasonable agreement with the model. The obtained fitting parameters are: try frequency, 1.8 x 10^{8} s^{-1}; activation energy reduced to the Boltzman constant, 46 K; and interaction temperature, 10 K. In summary, these results clearly endorse the

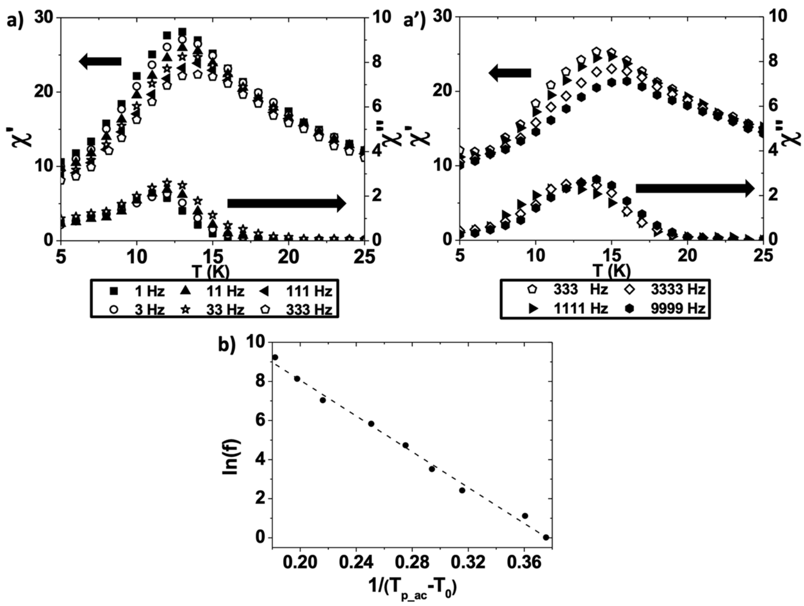

FIG. 1. Temperature dependencies for the real and imaginary parts of the ac susceptibility measured in the ac frequencies range from a) 1 Hz up to 33 Hz, a') 333 Hz up to 9999 Hz; b) variation of the logarithm of the measuring frequency with the reciprocal of the temperature at which the ac susceptibility peak was observed for each measuring frequency.

occurrence of spin glass-like collectiveness in our sample at temperatures below T_{p_ac} although they do not positively prove the occurrence at that temperature of a spin glass-like magnetic order phase transition.

Pursuing the elucidation of the actual occurrence of that phase transition we have measured the applied dc field, H_{dc}, variation of the temperature at which the irreversibility is detected in the FC/ZFC curves. Our purpose was the comparison of the experimental data with the Almeida and Thouless (A-T) analytic evaluation,[14] obtained for temperatures close to the freezing ones, of the dependence on H_{dc} of the stability of a spin glass phase against a field induced spin glass-to-ferromagnetic transition. According to the A-T predictions the temperatures at which a spin glass phase would exhibit the occurrence of irreversibility (that we will identify with T_{irr}) should depend on H_{dc} according to $T_{irr} \propto H_{dc}^{2/3}$. In Fig. 2 we have plotted the ZFC and FC curves measured in our sample by applying different fields in the range from 8 x 10^2 A/m up to 4 x 10^5 A/m. Figure 3 a) shows the results of the fit of T_{irr} to $H_{dc}^{2/3}$. As it is apparent from this figure, a close-to-linear behavior is obtained for the variation of the irreversibility temperature with $H_{dc}^{2/3}$ which validates the presence in the sample of an A-T spin glass stability line. According to the results of this fit, our sample should experience a field induced transition from the spin glass order to ferromagnetism at 0 K under a field H_{dc} = 4.72 x 10^5 A/m. This point is experimentally confirmed through the data in the inset on Fig. 2 where the absence of irreversibility for an applied field H_{dc} = 6 x 10^5 A/m is shown.

Once the agreement of the experimental data with the A-T forecasts has been shown, we can proceed to explicitly examine the critical behavior taking place at the transition temperature (that in the following we will identify as a spin glass freezing one). In particular, we will consider the dynamical scaling relating the relaxation time at the phase transition (the critical relaxation time, τ) to the correlation length associated to the interactions underlying the phase transition, ξ. According to the standard theory of the critical behavior, Refs. 3 and 15, these two quantities are related through $\tau \approx \xi^{Z}$. The correlation length diverges at the transition temperature following $\xi \approx [T/(T-T_g)]^{\nu}$ (where T_g is the so-called glass transition temperature, measuring the temperature at which the transition takes place in absence of dynamic effects) and the critical relaxation time diverges with temperature according to the law $\tau = \tau_0\ [T/(T-T_g)]^{Z\nu}$ (the product $Z\nu$ is the so-called dynamic scaling exponent). Figure 3 b) shows our best fit of the experimental data to the latter expression, which corresponds to a glass transition temperature, T_g = 12 K, a dynamic scaling exponent $Z\nu$ = 6.7 and a pre-potentialτ_0 value of

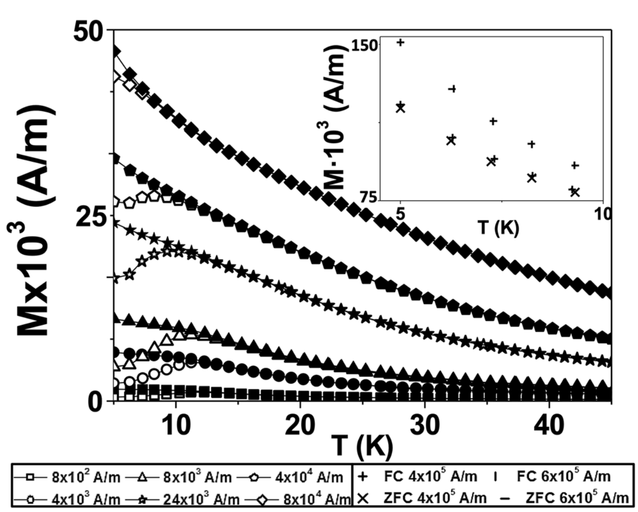

FIG. 2. Temperature dependence of the ZFC-FC curves measured under high fields.

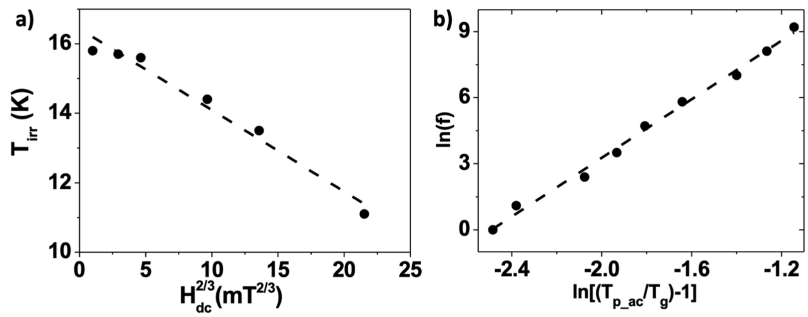

FIG. 3. a) variation of the irreversibility temperature with the dc field value used in the FC curves; b) fit to the critical power law (see text) of the ac susceptibility maxima in Figure 1.

10^{-8} s. The glass transition temperature is slightly lower than the measured T_{irr} value, whereas the pre-potential parameter τ_0 and the dynamic scaling exponent are in the range of the values usually obtained for spin glass systems. This is especially relevant in the case of the dynamic scaling exponent since it falls within the range from 4 to 12 that corresponds to the critical behavior of well-identified spin glass systems.[7]

CONCLUSIONS

From our data we can conclude about the occurrence in our sample of a spin glass transition which does not correspond to the canonical type. This is so due to the fact that the entities freezing at T_f are the Co nanoparticles we identified by TEM. Our sample falls into the commonly called super-spin glass systems class for which the magnetic moments experiencing disorder and competitive interactions (and from them frustration) are not atomic moments but moments associated to larger entities as clusters or, as in our case, nanoparticles. Regarding the latter basic ingredients for the glassy state we can outline that the disorder in our sample is restricted to the 2D position of the Co nanoparticles within the nominal Co layers. As for the competitive interactions, we consider that the dipolar ones should play a major role in our sample. This is so due to i) the large size of the nanoparticles moments coupling (originating large dipolar fields, of the order of 8 x 10^5 A/m, at first neighbors inter-particle distances) and ii) the order of magnitude of the inter-nanoparticles distances being large for the occurrence of substantial RKKY exchange coupling. Also, the dipolar interactions incorporate competitiveness since they are highly anisotropic both in magnitude and sign.

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