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Super spin dimensionality of a mono-dispersed and densely packed magnetic nanoparticle system

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Abstract. The dynamics of a dense near mono-dispersed assembly of maghemite nanoparticles is investigated by measurements of the temperature dependence of the isothermal remnant magnetization induced by temporal application of weak magnetic fields at constant temperature. The results suggest that the dimensionality of the super spins of the particles is of Heisenberg character at high temperatures but crossover to become Ising like at lower temperatures.

1. Introduction

It has been demonstrated that dense magnetic nanoparticle systems attains spin glass like properties at low temperatures. [1, 2] However, the existence of a super spin glass temperature and conventional critical slowing down has only been evidenced in systems with narrow distributions of the particle sizes and provided that the Arrhenius behavior of the individual particle relaxation time is accounted for. [3, 4, 5]

Certain dynamic properties of atomic spin glasses have been found to depend on the spin dimensionality of the system, e.g. the temperature dependence of the weak field remnant magnetization attained in an isothermal protocol [6]. Here we employ this measurement protocol to investigate whether a densely packed system of near mono-dispersed maghemite nanoparticles [7] can be classified as consisting of Ising or Heisenberg super spins.

2. Experimental

The experiments were made in a Quantum Design MPMS SQUID magnetometer. Zero field cooled (ZFC) and field cooled (FC) magnetization were recorded as a function of temperature to characterize the sample and to ensure that weak enough fields were used to achieve linear response of the ZFC magnetization at low temperatures. The isothermal remnant magnetization (IRM) was recorded using the following protocol: The sample was cooled in zero field to a specific temperature T_h below the maximum in the ZFC magnetization. At this temperature, the sample was kept for a specific wait time, t_w. After the wait time the magnetic field was applied (1 mT) and kept constant during the hold time, t_h. The magnetic field was cut to

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zero and the sample was left with an IRM. The sample was then, immediately after the field had reached zero, cooled at the maximum cooling rate of the MPMS (10 K/min) to a lowest temperature (10 K). When reaching this temperature, the $IRM(T)$ was recorded on heating the sample at a constant reheating rate (2 K/min) to a temperature above the glass transition temperature.

Figure 1 a) and b) shows the results of measurements using this method on an Ising spin glass and a dilute magnetic alloy $Cu(Mn)$ representing a system with Heisenberg spins. There is a striking difference in between the behavior of the two model systems on approaching T_h; the $Cu(Mn)$ $IRM(T)$ curve shows a pronounced maximum just before the expected rapid decrease to zero just above T_h, such an anomalous maximum does not appear in the $IRM(T)$ curve for the Ising spin glass. (The temperature scale in figure 1 a) and b) has been normalized to the magnetization in the FC at T_h).

The maghemite particles of this study were synthesized by thermal decomposition [7]. The particles are single domain and have a narrow size distribution with a mean diameter of 8 nm. The sample used in this study was prepared by pressing the (γ-Fe_2O_3) nanoparticles into a compact disc. The filling factor of the sample is about 67% which is close to the value corresponding to random close packing. The spin glass temperature T_g for the sample was determined to 140 K [5, 8]. A dilute superparamagnetic reference sample prepared from the same batch of particles had a blocking temperature of 36 K, indicating that the strongly enhanced random dipolar interaction of the dense sample governs the dynamics at higher temperatures [5, 7].

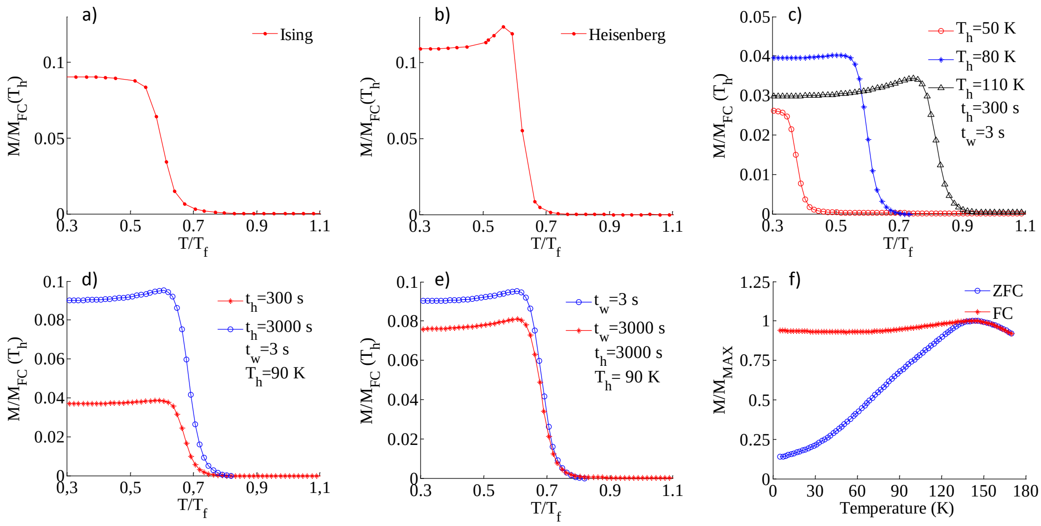

Figure 1. a) and b) show Typical $IRM(T)$ curves for an Ising ($Fe_{0.5}Mn_{0.5}TiO_3$) and a Heisenberg ($Cu(Mn)$) spin glass, adapted from Fig. 3 in ref. [6].) c) $IRM(T)$ for three different halting temperatures, T_h=50, 80 and 110 K. d) $IRM(T)$ at T_h=90 K with different t_h= 300 and 3000 s. e) $IRM(T)$ at T_h=90 K with different t_w= 3 s and 3000 s. f) ZFC and FC magnetization which have been normalized to the FC magnetization at 145 K. In a)-e), each $IRM(T)$ curve is normalized to the FC magnetization value at T_h. The applied field is in all experiments 1 mT.

3. Results and Discussion

Figure 1 shows the temperature dependence of the magnetization of the sample using the different protocols described above and applied fields of 1 mT. Figure 1 f) shows the ZFC

and FC magnetization curves as reference for the magnitude and temperature dependence of the magnetization. Figures 1 c-e) show different IRM-measurements where the magnetization is normalized to the value of the FC magnetization curve at T_h and the temperature is normalized to T_f. In the three experiments using different temperatures T_h, a pronounced peak is observed for the curve corresponding to the highest T_h, a weaker maximum for the intermediate T_h and for the lowest temperature no peak can be seen. Comparing these observations to those of the Ising and Heisenberg spin glasses of figure 1 a) and b), one is tempted to interpret the behavior as a crossover from essentially Ising to Heisenberg like super spins with increasing temperature. It can also be seen that the magnitude of the IRM for the curve corresponding to the middle halting temperature is larger than for both the lowest and the highest T_h. In figure 1 d), a plot at T_h/T_g=0.6 with two different values of t_h; 300 s and 3000 s and in figure 1 e) a plot using the same T_h and the same t_h=3000 s but different $t_w = 3$ s and 3000 s before the field was applied are shown. Both these figures confirm the expected behavior that the magnitude of IRM increases with t_h and that ageing slows down the relaxation, yielding a lower magnitude of the IRM for larger wait times.

Our experimental results are interpreted to indicate a crossover from Ising to Heisenberg character of the super spins of the particles on increasing temperature. This conclusion require certain caution: In atomic spin glasses, the spins are classified as being of Ising or Heisenberg character. The super spins of an super spin glass are on the other hand classical magnetic moments, i.e. the quantum mechanical concepts Ising and Heisenberg character are not applicable. However, at low temperature the magnetic moment of a particle is essentially directed along the magnetically easy axis in-between flips and may be considered as an Ising like super spin. At higher temperatures, the thermal fluctuations of the magnetic moment away from the easy direction become excessive in-between flips and the dimensional character of the super spin effectively crosses over to attain Heisenberg like character.

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