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ScienceDirect
Physics Procedia 9 (2010) 32–35
Physics Procedia
www.elsevier.com/locate/procedia

12th International Conference on Magnetic Fluids

Ac-Susceptibility Study In Rare Earth Substituted Magnetite Ferrofluids.

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Abstract

It is shown that variation of the third order ac-susceptibility as a function of measuring field and frequency lead to distinguish between superparamagnetic and spin glass like ordering in the rare earth substituted magnetic ferrofluids. We observe the divergent behavior of the peak values of the third order susceptibility, as a function of measuring field and frequency, tends to zero is consistent with theoretical prediction for the ground state for a spin glass like system. This behavior is further substantiated by a linear dependence of log-log plots of peak of the third order susceptibility as a function of the measuring field and frequency.

Key Words: Ferrofluids; spin-glass; ac-susceptibility

1. Introduction

The discrimination between meta-stable magnetic systems in terms of superparamagnetic (SPM) or spin glass (SG) has a fundamental importance, though it is not easy to differentiate them experimentally. In the SPM, it is a interplay of thermal energy versus anisotropy energy which gives rise to metastable magnetism, whereas in SG, the cooperative freezing of spin or cluster of spin is responsible for metastable magnetism. Therefore to understand appropriately slow dynamics in nanomagnetic particle system, it is desirable to clarify the origin of slow dynamics i.e. SPM or SG. Bajpai and Banerjee [1] have shown that, only observation of frequency dependence in ac–susceptibility, particularly first order susceptibility(χ'_1) or bifurcation of field cooled (FC) and zero field cooled (ZFC) magnetization is not conclusive enough for exact determination of magnetic ground state in a disorder system. To differentiate among SPM and SG states in a given magnetic system measurement of higher order susceptibility becomes essential. The third order magnetic susceptibility (χ_3') shows a negative peak at temperature where χ'_1 shows a peak. This is also true for SG and SPM system. Therefore only on the basis of qualitative features of χ_3' it

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1875-3892
doi:10.1016/j.phpro.2010.11.009

is difficult to identify the exact nature of magnetic transition. However, it is known that for SG, χ_3' shows a negative divergence at glass transition temperature [2], when amplitude of measuring field (H) and frequency (f) tends to zero. This behavior has been observed for many SGs [3]. On the other hand, SPM is a progressive blocking of moments of the magnetic particles (in experimental time scale) which neither shows criticality in χ'_1 nor inχ_3' with H, f ,or T.

It is easier to study spin glass or SPM behavior in tailor-made nano particle systems as the properties within and among the particles can be easily tuned. This will help in settling various unsolved issues related to cluster spin glass or SPM behavior. The origin of spin glass behavior in such systems is considered to be arising from random dipolar interaction between the two particles. The detailed study on this system will throw more light on the origin of this random interaction required for formation of glassy system. In this paper, we report the measurements and analysis of higher order magnetic susceptibility of Ho-rare earth substituted magnetite ferrofluid. The study reveals that low temperature magnetic phase can be unambiguously concluded to be a spin glass like phase.

2. Experimental/Methodology

Experiments were performed on ferrofluid [4] sample consisting of 5% Ho substituted magnetite particles suspended in a hydrocarbon carrier, abbreviated as FH. Analysis of the x-ray diffraction pattern using Rietveld refinement program confirms the magnetic particles to be of single phase cubic spinel structure. The particle size was determined using Scherrer's formula for (311) reflection. The volume weighted average crystallite size (diameter D_{vol}) and the lattice parameter thus obtained are 11nm ± 1nm and 0.8372 ± 0.0001 nm, respectively. The size distribution histogram is constructed from TEM image by measuring the size of over 500 particles, which shows that more than 50% particles are having the size of ~ 10 (±1) nm while the rest are distributed over ± 4 nm from the average size of 10 nm[5]. Since the high magnetic field can smear the transition and mask the intrinsic signatures of the magnetic system, we performed only low field dc-magnetization and ac-susceptibility measurements using quantum design $MPMS_2$ SQUID magnetometer and the home built susceptometer, respectively.

3. Results and Discussion

Figure 1 shows the real part of χ'_1 for a diluted (volume fraction 0.2%, in order to stimulate the fluid with non-interacting particles) FH fluid sample measured at different frequencies. The bifurcation of ZFC and FC dc-susceptibility below the transition for this sample indicates the dependence of the field history on the measurement (Inset Figure 1 at 50 Oe) . The observed time and history dependent magnetization are generic features of the magnetic disordered systems showing metastable magnetism like spin-glass, cluster glasses, SPM and even inhomogeneous ferromagnetic [5]. To resolve this, higher order magnetic susceptibility measurements were carried out.

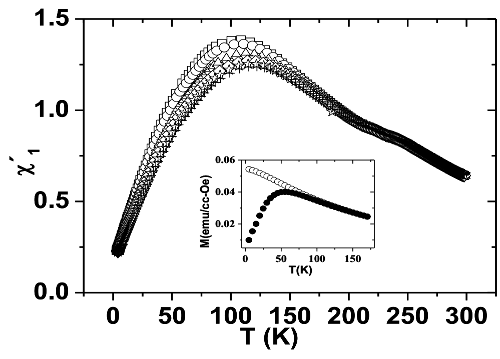

Figure.1 Frequency (for 67 Hz to 1.8 kHz) and temperature dependence of real part of the ac-susceptibility for the FH dilute magnetic fluid at 0.5 Oe. Inset shows FC(solid symbol) and ZFC (open symbol) dc-susceptibility measured at 50 Oe.

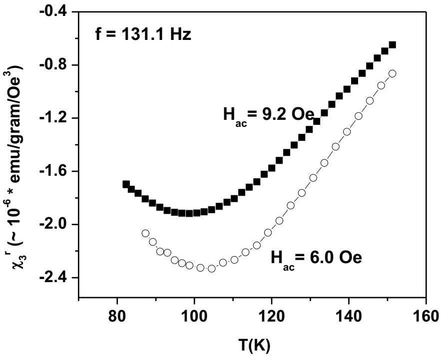

Figure. 2 The negative peak in the real part of the third harmonics as a function of applied magnetic field.

The third order magnetic susceptibility (χ_3') for two different ac-magnetic fields show negative peaks at the temperature where χ'_1 has shown a peak (Figure 2).The third order susceptibility is expected to show a negative peak as a function of temperature in both i.e. SPM and SG systems. In situation like this, when both χ'_1 and χ_3' show exactly similar temperature dependence around T_G or T_B for a SG and SPM phase, respectively, a detailed study of their corresponding field and frequency dependence can provide the missing link as guided by theories and experiments [5-9] . If ground state is a SG phase, χ_3' shows a divergence at the glass transition temperature, when amplitude of measuring field (H) and frequency (f) tends to zero. Whereas, in the case of SPM system, according to Wohlfarth's model χ'_1 shows 1/T dependence and χ_3' shows a $1/T^3$ dependence[3] Figure 3 shows the dependence of peak value of $|\chi_3^{max}|$ as a function of field , showing the divergent behavior as $H \rightarrow 0$. The value of χ'_1 derived from the χ_3' is shown in the Inset of Fig.3. It should be noted that while peak value of χ'_1 changes by 5% whereas the corresponding change in $|\chi_3^{max}|$ is more than 10 times, indicating the divergence behavior of χ_3 as $H \rightarrow 0$. This is also substantiated by linear dependence of log-log plot of $|\chi_3^{max}|$ versus H(Inset Fig.3).

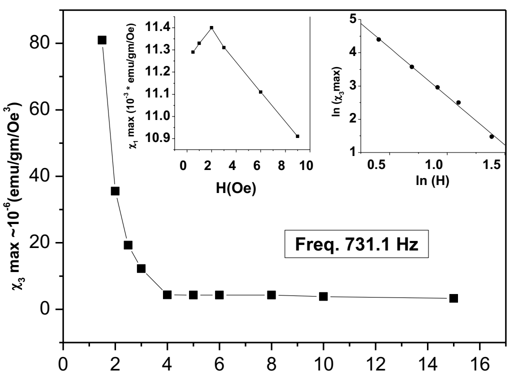

Figure. 3 The dependence of peak value of $|\chi_3^{max}|$ as a function of applied ac-field. The insets show variation of peak value of $\chi_1$and log-log plot of $|\chi_3^{max}|$ as a function of field.

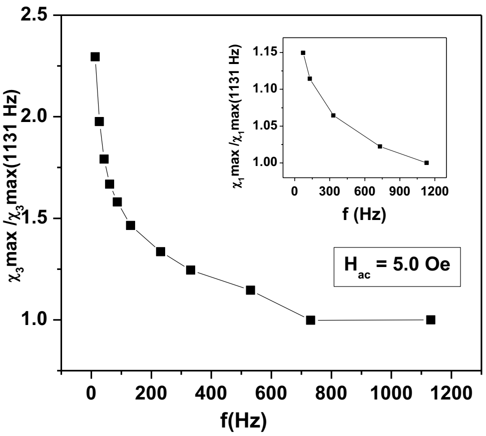

Figure 4. Normalized peak value of $|\chi_3^{max}|$ as a function of frequency. The inset shows similar variation for χ.

Figure 4 shows normalized peak value of χ_3' as a function of frequency. The inset shows the value for χ'_1. It can be seen that peak value of χ'_1 changes only by 15 percent with decrease in frequency, whereas corresponding change in peak value of χ_3 is about 230 percent. This indicates the divergent behavior of χ_3 as $f \rightarrow 0$. It should be noted that contrary to the canonical spin glass, in the present system it is possible to have internal spin dynamics which makes the frequency dependence rather complicated. In principle χ_3 is never affected neither by the amplitude nor by the frequency of the ac-field. Therefore, the observed divergence with lowering amplitude may be due to the contribution from higher order susceptibility and the observed divergence with lowering the frequency is attributed to the slowing down of the relaxation.

4. Conclusion

In this study Ho doped Fe_3O_4 nanoparticles system shows time and history dependent magnetization which are the characteristics of either SPM or spin glass. From the divergence of third order ac susceptibility as a function of magnetic field and frequency we conclude that such a metastable magnetization arises due to the spin glass like freezing, originating from inter particle random dipolar interaction. The study can be extended further by tuning this system to settle various fundamental problems related to critically slow dynamics.

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