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Single domain magnetic arrays: role of disorder and interactions

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Abstract. The hysteresis of an array of interacting single domain magnetic particles is studied, where the particles interact via exchange and dipolar interactions. The dependence of the magnetic properties on the distribution of grain sizes, the density of the grains, the anisotropy energy and the exchange interactions in the array is investigated through Monte Carlo simulations for $\gamma - Fe_2O_3$ nanoparticle systems. We also present some experimental results on the $\gamma - Fe_2O_3$-polypyrrole nanocomposite system which agree with the trends observed in our simulations.

PACS. 75.75.+a Magnetic properties of nanostructures – 75.50.Lk Spin glasses and other random magnets – 75.40.Mg Numerical simulation studies – 75.50.Tt Fine-particle systems; nanocrystalline materials

1 Introduction

The magnetic properties of ultra fine and nanosized magnetic particles show significant variations from their bulk values. This has led to considerable interest on both the technological and fundamental level. On the technological front, these materials are used in magnetic recording media, ferrofluids and as catalysts etc. [1]. Their fine structure also exhibits electron interference, oscillatory magnetic coupling, superparamagnetism and low dimensional magnetism [2]. From the point of view of industrial application it is essential to obtain single domain magnetic fine particle arrays with negligible size distribution of the particles, which are well separated and non-interacting. However the standard techniques of synthesis results in clustering, a non-negligible size distribution, as well a variation in shape of the particles. Recent experiments show that most of the magnetic properties, namely the magnetization, coercivity and Curie temperature of these magnetic systems are very sensitive to the above mentioned parameters [3].

A general description of the magnetization of fine magnetic particles is based on the theory of superparamagnetism, in which it is assumed that the magnetic particle is actually a single domain entity in which the atomic magnetic moments rotate coherently, such that the magnetization of the particle may be represented by a single magnetization vector with a large magnitude. The important feature of the system is its anisotropy. Uniaxial anisotropy, as often seen in transition metal oxides, leads to two equivalent magnetization states on each particle. The relevant time scale in the system is the relaxation time, which is the time taken to reverse the magnetization of the particle from one equilibrium magnetization state to the other. The relaxation time τ depends on the anisotropy constant K and the volume V of the particle as $\tau^{-1} \propto e^{-KV/k_BT}$ where T is the temperature of the system. The magnetic behavior observed in the system at different temperatures however depends both on the experimental time scale t_S for the observation being done and the relaxation time τ. For those temperatures where $\tau > t_S$, the system appears like a ferromagnet and for temperatures at which $\tau < t_S$ the system appears superparamagnetic. The blocking temperature is determined by $\tau = t_S$ and separates the two regimes.

The magnitude of the magnetization on a single domain magnetic nanoparticle is in itself a complicated problem which depends explicitly on the size and shape of the particle and several studies of the magnetization properties of single domain particle are available in literature [4–7] which are based either on micromagnetic modeling or Monte Carlo simulations. From these simulations as well it is established that the magnetic properties of mono domain magnetic particles are strongly influenced by finite size and surface effects, these effects becoming more important as the size of the particles becomes smaller.

For an array of single domain magnetic nanoparticles numerical studies are further complicated by the inherent disorder in the sample, as well as the interparticle interactions. The disorder in the system may be classified as (i) the spatial disorder in the actual positions of the grains in the array and (ii) the possible randomness in the orientation of the easy axes of the different particles, depending on the shape and size of the particles. The relevant interactions are primarily the long range dipolar interactions, and since in magnetic nanomaterials the particles shows a strong tendency to agglomerate one has to include

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the short range exchange interactions. Numerical studies of the magnetic properties of single domain magnetic arrays are based either on Langevin Dynamics or Monte Carlo methods. Using these methods hysteresis, magnetization and magnetic susceptibility are calculated and simulated [8–13] to study magnetization reversal and blocking temperatures. Most simulations in these systems however include only the randomness in the localized magnetic moment, and the spatial disorder is ignored. Garcia-Oterro et al. have reported the field cooled and zero field cooled susceptibility for a system of single domain ultrafine ferromagnetic particles using Monte Carlo simulations [13]. However their simulation neglects the effect of exchange interactions.

$\gamma - Fe_2O_3$ which is ferrimagnetic is an important material for magnetic storage. Several investigations of these materials both in its pure form and fine $\gamma - Fe_2O_3$ embedded in a host matrix has been investigated in some detail using Mossbauer, ac-dc susceptibility, etc. [14–17]. Morup and Tronc have investigated superparamagnetic relaxation in weakly interacting $\gamma - Fe_2O_3$ system [20]. In this paper we report the results of our simulations for a model system in which we incorporate disorder (due to shape and size distribution of the particles and their random positions in the array) and exchange interactions, using the parameters for a $\gamma - Fe_2O_3$ system. To our knowledge this is the first systematic numerical investigation in which the effects of disorder, short range exchange, long range dipolar and anisotropy effects have been studied simultaneously. We also present some ac susceptibility results performed on $\gamma - Fe_2O_3$-polypyrrole nanocomposites [14,18,19], to elucidate the role of interactions in these type of systems.

In Section 2 we discuss the model Hamiltonian used for the simulation, in Section 3 we describe the details of the simulation, in Section 4 we present our results and discussions and in Section 5 we present our conclusions.

2 Model

The model Hamiltonian for a system of interacting single domain magnetic particles [8–13,21], each having a magnetic moment vector $\vec{\mu}_i$ is written as

$$\begin{aligned} H = & -K\sum_i V_i \frac{(\vec{\mu}_i \cdot \vec{n}_i)^2}{|\vec{\mu}_i|^2} - \sum_{\langle i \neq j \rangle} J_{ij} \vec{\mu}_i \cdot \vec{\mu}_j \\ & - \mu_0 \sum_{\langle i \neq j \rangle} \frac{3(\vec{\mu}_i \cdot \vec{e}_{ij})(\vec{\mu}_j \cdot \vec{e}_{ij}) - \vec{\mu}_i \cdot \vec{\mu}_j}{r_{ij}^3} \\ & - \mu_0 \sum_j \vec{H} \cdot \vec{\mu}_j \end{aligned} \quad (1)$$

where the first term is the anisotropy energy, with K as the anisotropy constant. The anisotropy energy associated with each particle depends on the volume of the particle V_i and the angle between its magnetic moment vector $\vec{\mu}_i$ and $\vec{n_i}$ is the unit vector along the easy axis direction of the particle. The second term is the exchange energy, J_{ij} is the ferromagnetic exchange interaction between two particles with localized magnetic moment vectors $\vec{\mu}_i$ and $\vec{\mu}_j$ respectively, which for the purpose of simulation is assumed to have a site independent constant value J. The third term is the dipolar interaction between the i and jth particles, with r_{ij} as the distance between the particles. $\vec{e}_{ij}$ is the unit vector pointing along r_{ij}. The last term is the energy of the particles due to an externally applied magnetic field of magnitude H. It is assumed that the magnetic moment vector for a single particle has a temperature independent constant value $\vec{\mu}_i = V_i M_S \vec{\sigma}_i$ where M_S is the saturation magnetization of the particle and $\vec{\sigma}_i$ with coordinates $(\sigma_i^x, \sigma_i^y, \sigma_i^z)$ is the unit vector along the direction of magnetization vector. Several estimates of the actual value of M_S and the anisotropy constant K for a variety of nanomagnetic materials are to be found in literature [22]. It is also possible to make a reasonable estimate of the average size as well as the shape and size variation of the particles in an actual sample from various characterization methods like TEM, SEM etc. However there seem to be no definite estimates for the exchange interaction parameter J.

Disorder is manifested in the system in the following ways (a) the single domain grains have a random distribution of shape and size and (b) the magnetic grains are positioned randomly in the array. Because of the shape and size anisotropy in the sample there is a random distribution of the magnitude and direction of the localized magnetic moment and also in the direction of the easy axis of magnetization on the different particles in the sample. The positional disorder and the particle concentration in the sample affects the dipolar interaction between two magnetic particles which depends explicitly on the distance between the particles.

3 Details of simulation

We work with a basic simulation cell which is a cube of side $4L$. The cube contains N magnetic particles, the i th particle having a volume V_i (equivalent to a sphere of radius r_i) and L is the lattice spacing. The minimum allowed distance between any two particles in the array is $r_{min} \sim \alpha L$, where α can assume a value between 0 to 1. For the following simulation we have set $\alpha = 0.5$. The center of the ith particle has the coordinates x_i, y_i and z_i, where each of the coordinates is a random number between βL and L where β is a very small cutoff $\simeq 0.001$. The distance between any two sites i and j in the array is $r_{ij} = (x_{ij}^2 + y_{ij}^2 + z_{ij}^2)^{1/2}$. The coordinates of the magnetization direction vector for each particle i.e. $(\sigma_i^x, \sigma_i^y, \sigma_i^z)$, are picked randomly with $(\sigma_i^{x\,2} + \sigma_i^{y\,2} + \sigma_i^{z\,2}) = 1$. If the systems are polydisperse the volumes of the particles in the array are picked from a Gaussian distribution of width t which is defined by

$$P(V)dV = \frac{1}{(2\pi t)^{1/2}} \exp\left(-\frac{(V - V_0)^2}{2t^2}\right) \quad (2)$$

where V_0 is the mean volume of the particles. The coordinates of the easy axis $\vec{n}_i$ for each particle (n_i^x, n_i^y, n_i^z) are

also chosen randomly. The easy axes, site positions, magnitude of the magnetic moment at each site $|\vec{\mu}_i|$ and the volumes of the particles are kept constant for the entire simulation. The effect of disorder, size and shape distribution is assumed to be encapsulated in this random distribution of the positions, volume, magnetization direction and easy axes of the particles in the sample. It is well-known that for realistic simulation of the nanomagnetic system the calculation of the dipole dipole interaction energy of the sample is computationally expensive since the interaction is of a long range nature. According to current belief the best results for the dipole interaction energy are obtained by summing the energy between each pair of particles over periodic repeats of the basic simulation cell. We perform the summation over units cells using a variation of the Lekner summation method developed for electrical dipole energies which we find convenient for a positionally disordered array of particles [23].

We study the hysteresis curves for this sample by the Monte Carlo simulation technique using the standard Metropolis algorithm [24]. For each hysteresis curve the temperature, anisotropy energy, exchange interaction and the volume are kept fixed. The simulation is started at a very low field along the z-axis of the simulation cube, using an ensemble of magnetization vectors for the array, which gives net magnetization zero along the z-direction. The system is saturated by gradually increasing the magnetic field up to a very high field which is sufficiently higher than its anisotropy field. Then the magnetic hysteresis loop is simulated. For each value of the magnetic field 10000 Monte Carlo steps are used for the thermalization of the system and the calculation of the net magnetization along the field direction is made over the next 10000 Monte Carlo steps. A single Monte Carlo step is one in which an attempt is made to change the orientation of the magnetization vector on each site of the basic simulation cell. A change in the orientation of the magnetization vector on the site i from σ_i to σ_i' is done in the following fashion,

$$\begin{aligned}
\sigma_i'^x &= \frac{\sigma_i^x \pm \delta x \cdot \Delta}{\sqrt{\{}\sigma_i^{x2} + \sigma_i^{y2} + \sigma_i^{z2}\}} \\
\sigma_i'^y &= \frac{\sigma_i^y \pm \delta y \cdot \Delta}{\sqrt{\{}\sigma_i^{x2} + \sigma_i^{y2} + \sigma_i^{z2}\}} \\
\sigma_i'^z &= \frac{\sigma_i^z \pm \delta z \cdot \Delta}{\sqrt{\{}\sigma_i^{x2} + \sigma_i^{y2} + \sigma_i^{z2}\}}. \qquad (3)
\end{aligned}$$

For this attempt to change the direction of the magnetic moment on the ith particle the energy required is given by,

$$\begin{aligned}
\Delta E_i = &-KV_i\left[\frac{(\vec{\mu}_i'\cdot\vec{n}_i)^2}{|\vec{\mu}_i'|^2} - \frac{(\vec{\mu}_i\cdot\vec{n}_i)^2}{|\vec{\mu}_i|^2}\right] \\
&- J\sum_{nn}\left(\vec{\mu}_i'\cdot\vec{\mu}_j' - \vec{\mu}_i\cdot\vec{\mu}_j\right) - \mu_0(\vec{\mu}_i' - \vec{\mu}_i)\cdot\vec{H} \\
&+ \sum_j\left[\frac{(\vec{\mu}_i' - \vec{\mu}_i)\cdot\vec{\mu}_j}{r_{ij}^3} - \frac{3((\vec{\mu}_i' - \vec{\mu}_i).\vec{e}_{ij})(\vec{\mu}_j\cdot\vec{e}_{ij})}{r_{ij}^3}\right] \qquad (4)
\end{aligned}$$

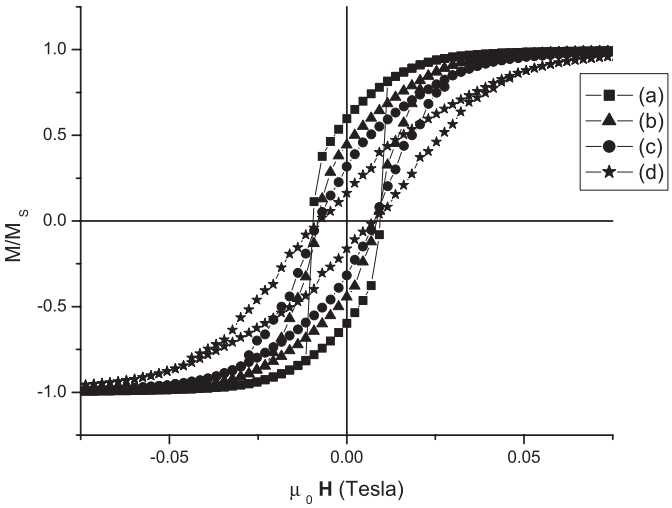

Fig. 1. The simulated hysteresis loops for $K = 0.046 \times 10^5$ J/m^3, $J_{eff} = 0.1E_A$ and uniform particle volume equivalent to a sphere of diameter 10 nm at a temperature of 5 K for increasing iron oxide particle concentration from (a–d), where (d) is the most concentrated system.

where $\vec{\mu}_i' = M_S V_i \vec{\sigma}_i'$. The sum over the nearest neighbors nn involves a summation over all sites within unit distance from i. The move is accepted with a probability $\exp(-\beta E_i)$ for $\Delta E_i > 0$ and 1 for $\Delta E_i < 0$ where $\beta = 1/k_B T$. δx, δy and δz are picked randomly but it is important to choose Δ such that the acceptance rate of our moves remains the same at all fields to ensure a constant rate of motion through the phase space [10,12]. Our results are averaged over 10000 MC steps, five different initial spin configurations and five different positional configurations. In the following section we discuss the results for various sets of parameters, which are K, $J_{eff} = M_s^2 V_0^2 J$, particle volume V_i and particle density, which is varied by varying the length of the simulation cell L.

4 Results and discussions

The studies depend strongly on the parameters which are M_S and K. For most of the simulation we work with the reported values of the parameters for the $\gamma - Fe_2O_3$ fine particle system which are (a) K is 0.046×10^5 J/m^3 and (b) $M_S = 4 \times 10^5$ A/m [22]. For the first set of simulations we study a monodisperse system where the volume of the particle is equivalent to that of a spherical particle of diameter 10 nm. The exchange interaction is written as a fraction of the average effective anisotropy energy for a spherical particle of diameter 10 nm which is $E_A = KV_0$.

In Figure 1 we plot the hysteresis curves for the above mentioned sample at a fixed value of J_{eff} which is approximately $0.1E_A$ at four different values of the particle concentration. The temperature is kept fixed at 5.0 K which is much smaller than the expected blocking temperature of

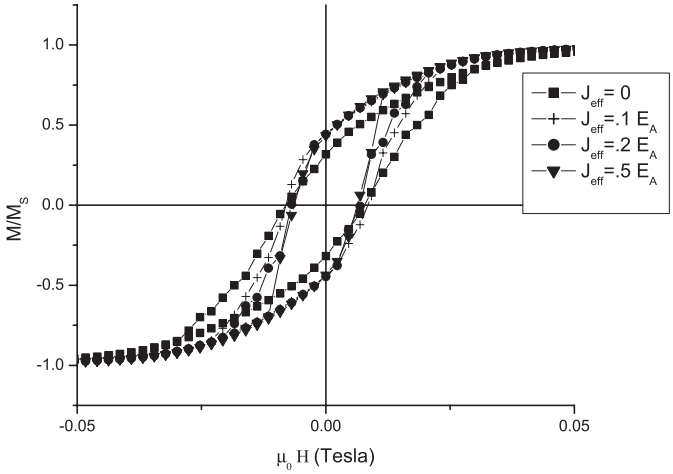

Fig. 2. The simulated hysteresis loops for $K = 0.046 \times 10^5$ J/m^3, fixed particle concentration and a uniform particle volume equivalent to a sphere of diameter 10 nm at a temperature of 5 K for 4 different values of J_{eff} which are 0, $0.1E_A$, $0.2E_A$ and $0.5E_A$.

the system. The system response is expected to be ferromagnetic for all the concentrations studied. With increasing particle concentration, the following effects are noted: (i) the magnetization at high fields becomes progressively lower, in fact the relative magnetization at 0.05 T reduces from 1.0 for the most dilute system (curve (a)) to 0.7 for the most concentrated system (curve (d)), (ii) the coercively of the sample shows hardly any change, which is expected, because below the blocking temperature the coercivity depends only on the magnetic anisotropy and (iii) the remanence decreases strongly with the increase of particle density, since the effective demagnetization due to dipolar interactions also increases. The different behaviors obtained can only be due to the stronger dipolar interactions because of larger concentrations.

In the next figure, Figure 2 we study the hysteresis loop at a fixed particle concentration and a temperature of 5.0 K for four different strengths of the effective exchange interaction. We observe that (i) for $J_{eff} = 0$, i.e. for a system in which the exchange interactions are negligible the magnetization and the remanence is smaller than for systems with a finite J_{eff}, (ii) for a finite value of J_{eff}, which is less than the anisotropy field, the remanence is almost a constant which seems to suggest that the remanence below the blocking temperature depends mostly on the particle concentration (iii) even for large values of the exchange interaction the coercivity of the sample does not show any remarkable change as it is mostly controlled by the anisotropy constant and (iv) the magnetization increases but the knee in the hysteresis curves appears earlier with increasing J_{eff}.

In Figures 3a and 3b we investigate the temperature changes in the hysteresis loop for a system with $J_{eff} = 0$ in Figure 3a and $J_{eff} = 0.2E_A$ in Figure 3b at the same value of the particle concentration as in Figure 2. In Figure 3a we see a clear transition from ferromagnetic phase at $T = 5$ K and 126 K to a superparamagnetic phase at

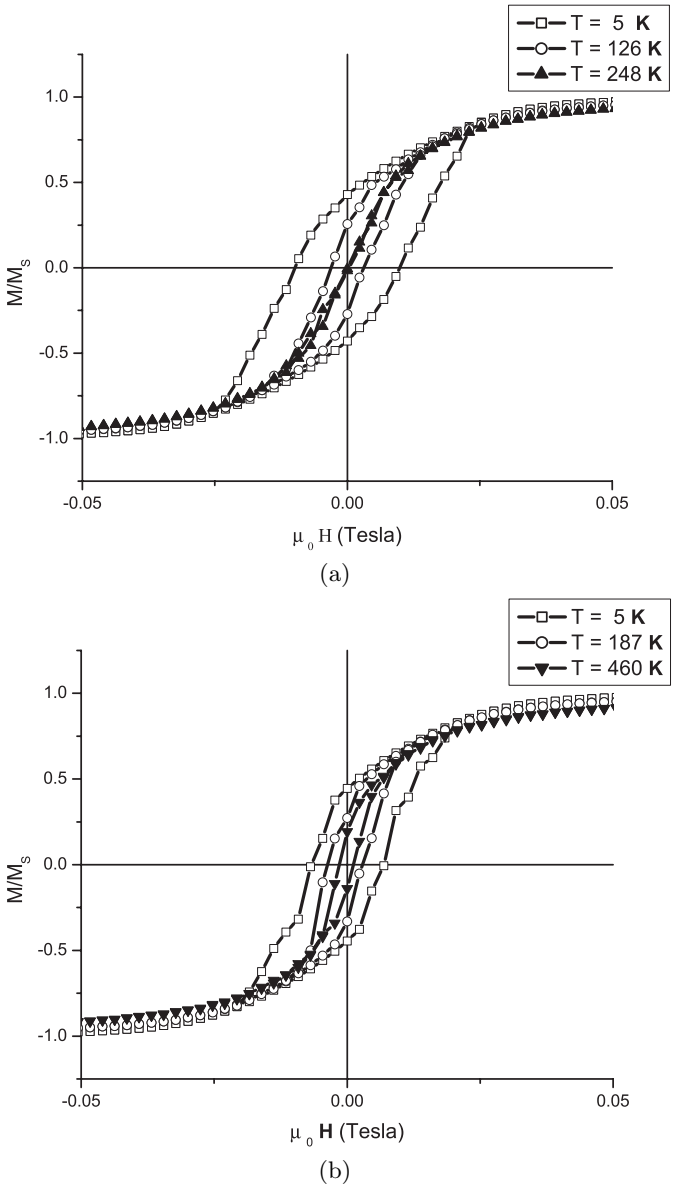

Fig. 3. The simulated hysteresis loops for $K = 0.046 \times 10^5$ J/m^3, fixed particle concentration and a uniform particle volume equivalent to a sphere of diameter 10 nm at different temperatures for (a) $J_{eff} = 0$ at 5, 126 and 248 K and (b) $J_{eff} = 0.2E_A$ at 5, 187 and 460 K.

248 K, with both the remanence and coercivity zero. Obviously for the monodisperse system the blocking temperature is between 126 and 248 K, in the absence of exchange interactions. For finite J_{eff} in Figure 3b we find that (i) the remanence is larger but the coercivity is smaller than the corresponding values for $J_{eff} = 0$ (Fig. 3a) and (ii) both the remanence and the coercivity decrease very slowly with temperature and retains a finite value even at very high temperatures, $T = 460$ K. Thus the superparamagnetic transition seems to take place at very high temperatures in the presence of exchange interactions.

The role of the change in anisotropy energy on the hysteresis was investigated and is shown in Figures 4a

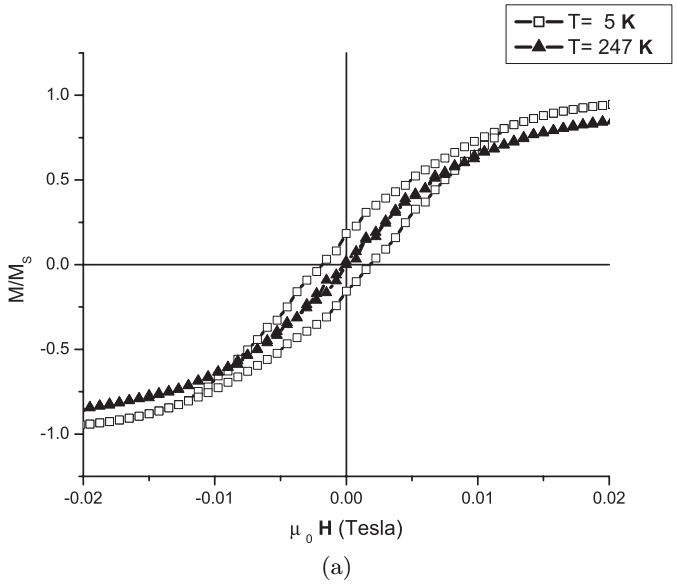

Fig. 4. The simulated hysteresis loops for $J_{eff} = 0.2E_A$, fixed particle concentration and a uniform particle volume equivalent to a sphere of diameter 10 nm for (a) $K = 0.015\times10^5$ J/m^3 at 5 and 247 K and (b)$K = 0.065\times10^5$ J/m^3, at temperatures 5 and 460 K.

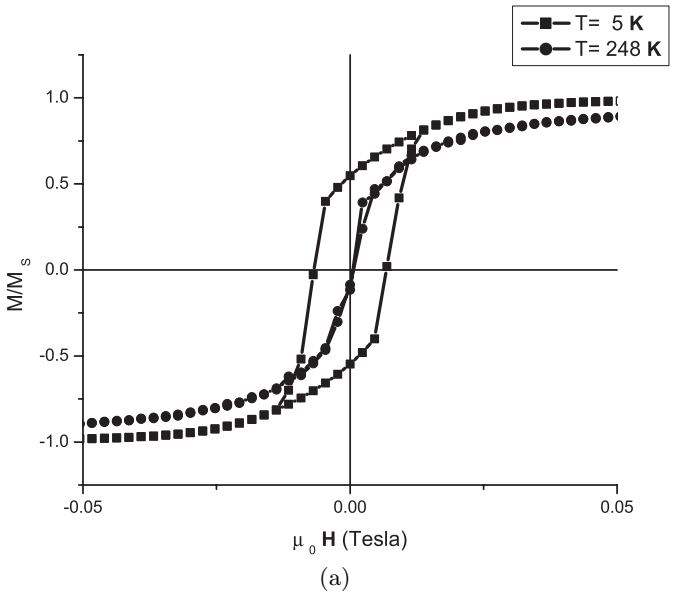

Fig. 5. The simulated hysteresis loops for $K = 0.046 \times 10^5$ J/m^3, $J_{eff} = 0.2E_A$, and a fixed particle concentration at 5 and 248 K for (a) uniform particle volume equivalent to a sphere of diameter 7 nm and (b) uniform particle volume equivalent to a sphere of diameter 13 nm.

and 4b. The particle size and the particle concentration remains the same as in Figures 2 and 3. In Figure 4a we plot the hysteresis curve for a low value of K i.e. $K = 0.015 \times 10^5$ J/m^3 (which is lower than the value reported for the $\gamma - Fe_2O_3$ system), with $J_{eff} = 0.2E_A$. On comparing with Figure 2, we can see that both the remanence and the coercivity of the system at 5 K is much less than that for a system with $K = 0.046 \times 10^5$ J/m^3. The low field magnetization is also larger for the system with lower anisotropy. Also for the same ratio of J_{eff}/E_A the system with a lower value of K seems to be superparamagnetic at 247 K (Fig. 4a) whereas the system with higher K does not exhibit superparamagnetic behavior even at 460 K as seen from Figure 3b. In Figure 4b the value of K is 0.065×10^5 J/m^3. This system is slow to saturate, and has larger coercivity and remanence than the system with lower K as seen from Figures 4a or 3b. Also the system is not superparamagnetic even up to 460 K.

In Figures 5a and 5b we compare the hysteresis of two monodisperse system where the particle volumes are equivalent to spheres of diameters 7 nm and 13 nm respectively, at 5 K and 248 K. The particle concentration is kept the same as in the earlier studies with $K = 0.046 \times 10^5$ J/m^3 and $J_{eff} = 0.2E_A$. By comparing their hysteresis curves at 5 K we see that the system with

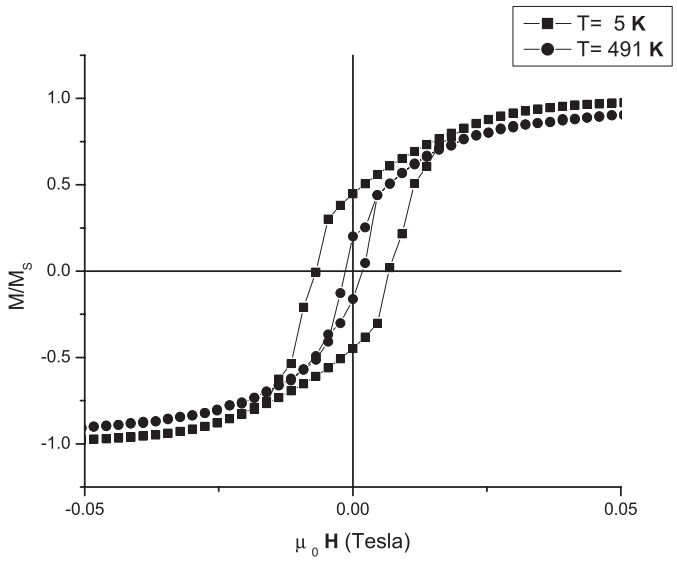

Fig. 6. The simulated hysteresis loops for $K = 0.046 \times 10^5$ J/m^3, $J_{eff} = 0.2E_A$, and a fixed particle concentration at different temperatures (5 and 491 K) for a system of particles which have their sizes distributed in a Gaussian of width $t = 0.2$ about a mean volume equivalent to a sphere of diameter 10 nm.

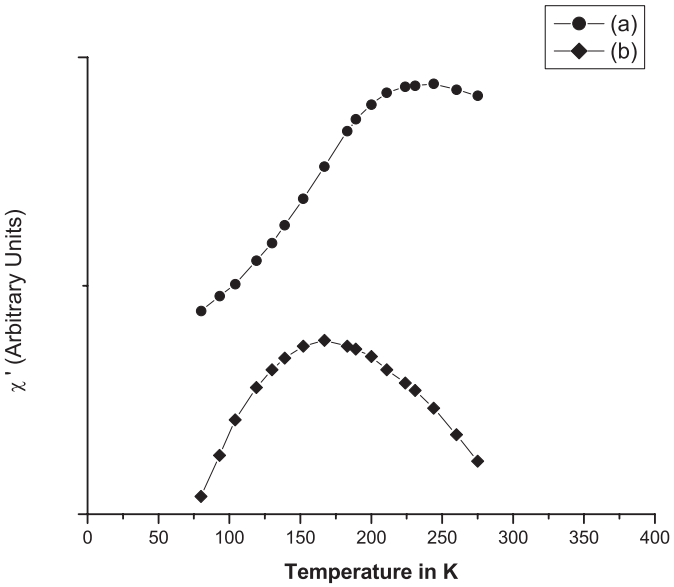

Fig. 7. The variation of ac susceptibility with temperature for the $\gamma - Fe_2O_3$ polypyrrole nanocomposite for (a) 5% and (b) 15% pyrrole.

smaller sized particles shows a larger remanence. It is expected that the blocking temperature is lower for smaller particle size and indeed we see from Figure 5a that this system is superparamagnetic at 248 K, whereas the larger sized particle (Fig. 5b) is not.

In Figure 6 we investigate a polydisperse system for which the volumes of the spherical particles are picked from a Gaussian distributed about a mean volume V_0 which is that of a sphere of 10 nm diameter. The width of the distribution is taken to be 0.2. The value of $K = 0.046 \times 10^5$ J/m^3 and $J_{eff} = 0.2E_A$. The hysteresis loops are plotted at two different temperatures. We find that this system is not completely superparamagnetic even at 491 K. On comparing with Figure 3b which is exactly the same as this system except than for 3b all the particles have the same volume equivalent to the that of a sphere of diameter 10 nm, we find that at 5 K the uniformly sized system has larger remanence and coercivity. The superparamagnetic behavior of the array sets in only when the blocking condition is satisfied for all the particles. Since in a polydisperse systems the blocking temperature is different for different sized particles, the hysteresis loops still retain their remanence and coercivity up to very high temperatures.

For comparison we report the results of the ac susceptibility measurements carried out on $\gamma - Fe_20_3$ / polypyrrole nanocomposites prepared by simultaneous gelation-polymerization process for different concentrations of pyrrole [14]. In Figure 7 we plot the thermal variation of the ac susceptibility [19] at 16 Hz for (a) 5% and (b) 15% pyrrole. Here the grain sizes were estimated to be in the range 10 to 20 nm. In (a) χ'_{ac} increases up to 250 K after which a tendency to decrease is observed, indicating that the blocking temperature is ∼250 K. In (b), the nanocomposite with 15% pyrrole, there is a fairly sharp transition to a superparamagnetic phase at ∼150 K. In the case of the nanocomposite containing 5% of pyrrole the iron oxide particles show a tendency to cluster, resulting in stronger exchange interactions, whereas in the system with larger pyrrole concentration the iron oxide particles are relatively well separated, resulting in a lower blocking temperature. This compares well with our simulation results.

Several other experimental results on blocking temperatures indicate the effect of inter particle interactions. Morup and Tronc [20] report the blocking temperature as a function of the average particle volume obtained from Mossbauer measurements. The blocking temperature was found to be high (∼300 K) for particles even as small as 800 nm^3. This has been attributed to the strength of the interaction between the particles. From ZFC-FC studies on RE-Fe based alloys, Wang et al. [10] report blocking temperatures varying in the range of 50 to ∼400 K and compare it favorably with Monte Carlo simulations on a cluster system with strong interactions and random anisotropy.

5 Conclusions

The entire thrust in the area of magnetic nanomaterials as in most other nanomaterials is driven by the possibility of industrial application. However the existing techniques like spin coating, sputtering, CVD etc. which are used for preparing these materials, result in films having an inherent disorder not only in terms of shape and size of the magnetic grains but also in their spatial arrangement in the array. In addition since most magnetic materials show a tendency to agglomerate, short range exchange interactions are also present in the system. The purpose of the

simulation was to investigate in a systematic fashion the role of various factors like particle concentration (which affects both the dipolar interaction and the exchange) and the particle size (which affects the anisotropy). Our Monte Carlo studies show that the ferromagnetic signature persists in the hysteresis loops up to very high temperatures in the presence of exchange interactions. The results indicate that the blocking temperatures can be very high in the presence of large exchange and low dipolar interactions, which would be favorable for recording media. This implies that it is essential to tailor make materials with well separated interacting magnetic single domains which may not be possible by conventional methods like sol-gel, sputtering, etc. Self assembling of these magnetic particles using bio molecules have been suggested as an alternative method of preparation.

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