Response of magnetic nanoparticle assemblies

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Abstract. Properties of magnetic nanoparticle assemblies are analysed by employing the randomly jumping interacting moments model including quantum fluctuations, ferromagnetic inter-particle coupling and disorder. Using spatially uniform external field we find the respective magnetic state equation and phase diagram indicating an existence of spinodal regions and critical points. Possibilities to use such systems as magnetoresistive sensors are considered.

1. Introduction

Supermagnetism (SM) was invoked to specify the properties of a system containing quantum confined objects, e.g., quantum dots, atomic clusters and/or magnetic nanoparticles, see [1-5] and refs. therein. In particular, an assembly of magnetic nanoparticles (MNPs) displays ferromagnetic (FM) long range order at sufficiently large density. Such a property requires to consider the inter-dot magnetic interaction beyond dipolar components [1-5], like quadrupole, superexchange, strain fields etc. Especially, the superferromagnetic (SFM) structures with certain electronic coherent states [1] provide a system exhibiting giant magnetoresistance (GMR) [6,7] and can be employed as magnetoresistive (MR) sensors. The SFM response in external magnetic fields represents an important relevant issue.

In this contribution we use the recently developed randomly jumping interacting moments (RJIM) model, see [2,3] and refs. therein, to analyse SFM magnetic reactivity, on examples of (i) spatially uniform field changing slowly in time and (ii) spatially local perturbation, i.e., a sensor mode for detection of, e.g., a single magnetic particle. In Sect.2 we briefly recall the RJIM model and the Monte Carlo simulation technique, cf., [2,3], used to describe the system dynamics. Optimized serial algorithms and parallel CUDA programming on graphic processing units (GPU) were developed to provide acceptable level of calculation rates. Arrays of nanoparticles with multiple magnetic response anomalies are demonstrated to display strong dependence on system disorder. Possible implications of peculiarities in magnetic planar array response as sensors are discussed in Sect 2. Conclusions are given in Sect. 3.

2. SFM structure of magnetic nanoparticle assemblies with disorder

Overall magnetisation of a system composed from $\Pi$ elements with moments $m_i$ can be represented as $P = \sum_i m_i / V = \langle m \rangle / V_D$ with total volume $V$ and an area occupied by $i$th nanoparticle.
For SFM with ferromagnetic coupling of a strength $J$ between the nearest neighbor (nn) nanoparticles the respective Ising term $-J \sum_{ij} m_i m_j$ contributes to the Hamiltonian. Here the sum runs over the nn elements. Besides, inhomogeneity and disorder in the form of defects, grain boundaries, impurities lead to random crystalline anisotropy and varying interaction strengths in the super-crystalline heterostructure. Such effects can be accounted for by the random fields $h_i$. We point out also dynamical components of $h_i$ due to inexactness of the model description with nn interaction. The central limit theorem suggests, thereby, that the random fields obey the Gaussian distribution $\mathcal{W}(h) = \exp\left\{-h^2 / R^2 \right\} / R \sqrt{\pi}$ of a width $R$, which we call the disorder. The total Hamiltonian $H$ of an array in a field $H$ can be expressed through an interaction of the nanoparticle magnetic moment $m_i$ with local fields

$$b_i = H(t) + J V_0 \sum_{j=\text{nn}} P_j + h_i$$

as $H = -\sum_j m_i b_i$. We refer for this model as randomly jumping interacting moments (RJIM) model [2,3]. Hereafter, we consider an array of nanoparticles with single discontinuity in magnetic moment response, $m_i = m \text{sign}(b_i)$. Due to the ferromagnetic interaction a jumping moment can cause some of the nearest neighbours to jump, which may in turn trigger some of their neighbours, and so on, generating, thereby, (re)magnetizing avalanche. As a consequence some sharp stepwise discontinuity arises on magnetization curves. Such sharp changes of magnetic induction result in a release of magnetic energy with noise signals proportional to avalanche size, similarly to the well-known Barkhausen effect.

![Magnetic state equation and phase diagram](image)

**Figure 1.** Magnetic state equation (Panel A) and phase diagram (Panel B) for SFM composed from MNPs with single jump at $b=0$ and disorder $R$. The values $H$, $b$ and $R$ are measured in units of $J$. 

### 2.1. Magnetic state equation and phase diagram of SFM

The basic features of the nonequilibrium system corresponding to the Hamiltonian Eq. (1) can be analysed by employing the mean-field approach, in which one assumes an equal interaction strength between MNPs with the coupling constant $J_{ij} = J / \Pi$. The local field in Eq. (1) can be then simplified to the form $b_i^{mf} = H(t) + J P + h_i$ with an averaged over a sample magnetization $P$, see above. We see, therefore, that random fields can be viewed as mean-field fluctuations (cf, e.g., [2,3,8]).

In the thermodynamic limit $\Pi \rightarrow \infty$ for spatially uniform field $H$ we calculate the magnetic state equation (MSE) $P = \int dh \mathcal{W}(h)m(b)$ and find for the magnetic susceptibility
\[ \chi = -dP / dH = \chi^{-1}_{NI} - J^{-1} \]  \hspace{1cm} (2)

with \( \chi_{NI} = W(b) \) representing the susceptibility of an array without inter-particle interaction (i.e., \( J=0 \)). The case of negatively defined susceptibility, see Eq. (2), defines spinodal region for an array. Figure 1 shows MSE and phase diagram. For coupled MNPs the value \( J \chi_{NI} \) measures an average number of induced jumps per single jumping moment. When the number of induced moment jumps exceeds 1 the system favours to evolve in an avalanche spanning almost entire sample with a macro-(de)magnetization discontinuity. Evidently, the relation \( \chi_{NI} \geq J^{-1} \) corresponds to the instability condition. Such spinodal regions are located on \{H,R\}-plane between the low and up critical fields as indicated in Fig. 1B. The field-lines meet at the critical point \( \{ H_c = b_0 = 0, R_c = J / \sqrt{\pi} \} \). The difference between the number of induced jumps and 1, \( d = J \chi_{NI} - 1 \), provides, therefore, a measure for a vicinity of self-organized criticality.

**Figure 2.** Size distribution of avalanches induced by local fields for \( 2 \times 10^5 \) randomly distributed positions (see text) at different disorders \( R= 2 \) (line), 8(open circles), 16(solid circles). Solid line shows the fit according to Eq. (3).

2.2. Sensing magnetic nanoparticles by SFM structures

To analyse possibilities for using the realistic SFM systems as MR sensors for detecting, e.g., a single magnetic particle we consider spatially local field inducing jumps of several assembly elements. Such an event creates an avalanche of moment jumps due to ferromagnetic coupling and the GMR signal proportional to a number \( S \) of particles in an avalanche. We perform the simulations for SFM of simple quadric planar lattice of a size \((1000)^2\) with random distribution of such avalanche grains. As seen in Fig. 2 the avalanche size distribution \( D(S) \) shows nearly exponential behaviour with increasing width at growing disorder. To understand such a trend we employ the mean-field arguments. Since
within the mean-field treatment an average number of the moments induced to jump by a single jumping seed is site independent for the avalanche size distribution at $1 \ll S \ll \Pi$ one obtains [3]

$$D_{mf}(S) \sim S^{-3/2} \exp\{-Sd^2/2\}$$

(3)

For growing disorder in the considered range the parameter $d$ decreases. Therefore, the distribution $D(S)$ becomes wider according to Eq. (3). As a consequence, such conditions correspond to stronger MR signal of a sensor.

### 3. Conclusions

As seen above the magnetic nanoparticle assemblies display the SFM structure effects at sufficiently dense packing. Magnetic state equation and phase diagram of SFM are demonstrated to exhibit spinodal regions on \{disorder, magnetic field\}-plane and the critical points. We argue that such SFM features are well suited for magnetoresistive sensors. The disorder in magnetic nanoparticle assemblies improves the detection ability at considered parameters. We note that quantum fluctuations due to the nanoparticle discrete levels can bring additional anomalies of magnetic response [9] and new phases [3]. Such properties could provide further possibilities for sensors.

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