Dynamics of magnetic nano-particle assembly

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Abstract. Ferromagnetically coupled nano-particle assembly is analyzed accounting for inter- and intra-particle electronic structures within the randomly jumping interacting moments model including quantum fluctuations due to the discrete levels and disorder. At the magnetic jump anomalies caused by quantization the magnetic state equation and phase diagram are found to indicate an existence of spinodal regions and critical points. Arrays of magnetized nano-particles with multiple magnetic response anomalies are predicted to display some specific features. In a case of weak coupling such arrays exhibit the well-separated instability regions surrounding the anomaly positions. With increasing coupling we observe further structure modification, plausibly, of bifurcation type. At strong coupling the dynamical instability region become wide while the stable regime arises as a narrow islands at small disorders. It is shown that exploring correlations of magnetic noise amplitudes represents convenient analytical tool for quantitative definition, description and study of supermagnetism, as well as self-organized criticality.

1. Introduction
The assemblies of magnetic nano-particles provide an opportunity to develop new materials with characteristics beyond traditional solids and bring plausible benefits in 'figures of merits' for advanced technology and therapy. In particular, several laboratory tasks, e.g., injection, sample preparation, manipulation, reaction control, detection, separation etc, can be integrated in a single nano-magnet assembly, i.e., 'lab on a chip' system, cf. [1, 2] and refs. therein. In this contribution we consider analytical tools employed to specify, quantify and analyze such devices in respect with magnetodynamics.

At sufficiently high density of nano-magnets the assembly structure changes from superparamagnetism (SPM) to superferromagnetism (SFM) [1-4] inducing, thereby, jerky magnetodynamics with sharp discontinuities in the array magnetization process. The electronic spatial quantization is well-known to represent specific nano-particle feature bringing the sharp step-like discontinuities of the magnetic response in varying fields due to the Zeeman splitting [4, 5]. Familiar examples are related to the singlet-triplet transitions (cf., e.g., [4] and refs. therein) in semiconductor quantum dots and carbon nanotubes in magnetic field. We compare in this contribution different cases and demonstrate that this particular feature makes the system of nano-particles significantly different from traditional ensemble of magnetic elements with constant spins. In particular, we demonstrate that an effect of multiple discontinuity anomalies can bring in SFM structure some new phases related to self-organized (SO) criticality.
2. Superferromagnetism with disorder
Recent theoretical studies [3, 4] of realistic supercrystalline objects show, that the relationship between inter-particle magnetic interaction and the disorder substantially determines the magnetic structure and dynamics. Then an occurrence of SO criticality represents, perhaps, one of the most interesting phenomenon. At such a regime magnetic induction of nano-particle arrays displays erratic stochastic discontinuities with rather wide distribution of jump amplitudes.

2.1. Mean-field approach to ensemble of interacting discrete moments at a disorder
At a modeling of the nano-particle assembly magnetism we employ very general form for nano-particle magnetic moment
\[ m_n = \mu \sum_n \nu_n \theta(b - b_n) \]
with moment unit \( \mu \) and step function \( \theta(x) \) depending on local magnetic field \( b \). The set of quantities \( \{\nu_n, b_n\} \) determines the value of magnetic moment directed along the field vector (for more details see [4] and refs. therein). The conditions \( \nu_n \neq 0 \) simulate the discrete anomalies of moment jumps. The field independent spin \( 1/2 \) corresponds to \( \{\nu_n, b_n\} = \{g, 0\} \) at \( n \neq 0, 1 \). The particles with a singlet-triplet transition at a field \( B_{st} \) are associated with the non-zero parameters \( \{\nu_n, b_n\} = \{-g, -\infty\}, \{\nu_n, b_n\} = \{g, -B_{st}\}, \{\nu_n, b_n\} = \{g, B_{st}\} \). In addition, we consider the particle array with 5 such jumps.

Magnetisation of a system composed from \( \Pi \) elements with moments \( m_i \) can be represented as
\[ P = \sum_i m_i/V = \langle m \rangle/V_0 \]
with total volume \( V \) and an area occupied by \( i \)th particle \( V_0 = V/\Pi \). For SFM with ferromagnetic coupling of a strength \( J \) between the nearest neighbor (nn) particles the respective Ising term
\[ \sum_{ij} J m_i m_j \]
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 \( h_i \). Considering also dynamical components of \( h_i \) allows improve inexactness of the model description with nn interaction. The central limit theorem suggests, thereby, that the random fields obey the Gaussian distribution
\[ \exp\left\{ -h^2/R^2 \right\}/R\sqrt{\pi} \]
of a width \( R \), which we call the disorder. The total Hamiltonian \( H \) of an array in a field \( b \) can be expressed through an interaction of the particle magnetic moment \( m_i \) with local fields \( b_i \)
\[ b_i = H(t) + J V D \sum_{j=\pm m} P_j + h_i, \quad H = -\sum_i m_i b_i \]  
(1)

We refer for this model as randomly jumping interacting moments (RJIM) model [3, 4].

2.2. SFM structure within the mean-field approximation
The nonequilibrium system corresponding to the Hamiltonian Eq. (1) can be analysed by employing the mean-field approach implying an equal inter-particle interaction strength with a coupling constant \( J / \Pi \). The local field in Eq. (1) is then simplified to the form
\[ b_i^{mf} = H(t) + JP + h_i \]
with an averaged over a sample magnetization \( P \), see above. The random fields can be viewed, therefore, as mean-field fluctuations (cf, e.g., [3, 4, 6]). In the thermodynamic limit \( \Pi \rightarrow \infty \) we calculate the magnetic state equation (MSE)
\[ P = \int dh W(h) m(H + JP + h) \]  
(2)

and the magnetic susceptibility \( \chi = \left[ k_B^{-1} - J \right]^{-1} \) with \( \chi_{mf} = \Sigma_{W}(b-b_0) \) representing the susceptibility of an array without inter-particle interaction (i.e. \( J=0 \)). Figure 1 shows the dependence of magnetisation \( P \) for such non-interacting particle assembly on the disorder \( R \) and magnetic field \( b \). As is seen
magnetic jump anomalies induced by quantization result in discontinuities in MSE. At increasing disorder such stepwise behaviour is smeared out.

It is worthy to notice here, that each particle jump anomaly brings two MSE discontinuities, see Fig. 1B. Therefore, Fig. 1C shows part of 6 jumps from the total 10 discontinuities on magnetization curve.

**Figure 1.** Magnetic state equation, MSE, of an assembly of nano-particles with constant spin (panel A), singlet-triplet transition (panel B) and 5 jump anomalies (panel C).

### 2.3. Magnetic phase diagram of SFM

Ferromagnetic inter-particle coupling brings the hysteresis loops in magnetization curves at the external field strengths corresponding to discontinuity anomalies. Such a behaviour can be understood in terms of avalanche propagation. When the local field $b_i$ of some $i$-th lattice element passes through the value $b_n$ the moment changes step-wise. Due to the ferromagnetic interaction a jumping moment can cause some of the nearest neighbors to jump, which may in turn trigger some of their neighbors, and so on, generating, thereby, magnetic avalanche, cf., [3, 4]. As a consequence some sharp stepwise discontinuity arises on magnetization curves. Within the mean-field approach MSE is given by the solution of an equation $P=P[H+J\chi]$, cf., Eq. (2). Therefore, the value $J\chi_{NJ}$ represents an average number of induced jumps per single jumping moment and the negatively defined susceptibility yields adiabatic spinodal region for SFM, cf. Eq. (2). Since the number of induced moment jumps exceeds 1 the system favours to evolve in an avalanche spanning almost entire sample with a macro-magnetization discontinuity. Such spinodal regions are located insight of contour lines on $\{B,R\}$-plane as is indicated in Fig. 2. The upper and lower field-lines meet at the critical point. As is seen in Fig. 2B and Fig. 2C for the case of multiple jumps in particle magnetization the array with weak coupling experience well isolated spinodal regions. The respective magnetodynamics is, therefore, quite similar to the case of a single jump. However, for strong coupling the region of instability is very spread so that we observe a confined isolated stability domain at small disorders.

**Figure 2.** The magnetic susceptibility for an array of non-interacting nano-magnets with constant spin (panel A), singlet-triplet transition at $B_{st}=1$ (Panel B) and 5 jump anomalies (Panel C) at disorder $R$.

### 2.4. SFM structure in magnetodynamics

As is seen above from the mean-field analysis the SFM objects display a spinodal regions at small disorder and approaches stable behaviour at large values $R$. Such a structure implies pronounced hysteresis loop at the instability region. Such a loop vanishes to a nearly smooth magnetization curve at increasing disorder. The intermediate regime matches a vicinity of SO criticality measured for as a difference between the number of induced jumps and 1, $d = J\chi_{NJ} - 1$. Since at such conditions the
mean number of induced jumps shows an extremum approaching 1, the mean linear size $l_b$ of the biggest avalanche $S_b$ can be estimated as $l_{\text{mf}}^b \approx (1 + d) / 2$ of the total linear size.

Since within the mean-field treatment an average number of the moments induced to jump by a single jumping moment is site independent for the noise size distribution one obtains

$$D_{\text{mf}}(S) \sim S^{-3/2} \exp \{-Sd^2 / 2\}$$  \hspace{1cm} (3)

We perform the simulations for SFM of simple cubic lattice of a size $(30)^3$. The cumulative size distribution $C(S) = \sum_{N \geq S} D(N)$ allows one to reduce statistical errors in the data analysis. Figure 3A represents distributions $C(S)$ integrated over the magnetization reversal hysteresis loop and averaged over 100 events of, e.g., array samples. One sees a clear transition from the 'U' shape distribution at small disorders to an abrupt exponential suppression of large size avalanches at large disorders. At transitional values $R$ the distribution shows behaviour close to the power law dependence $C \sim S^{-\tau}$ with an exponent $\tau \approx 0.85$ corresponding to 1.85 for $D(S)$ and being slightly different from the mean-field estimate.

Figure 3. Panel A - Cumulative avalanche size distributions are compared to the power law with an exponent $\tau = 0.85$. Results of the RJIM model for $(30)^3$ simple cubic lattice are shown for disorders $R=1$ - solid circles, 1.6 - solid triangles, 3 - open circles. Panel B - Mean avalanche size versus the linear size of the biggest avalanche in units of array length. Results of RJIM model at various disorders are shown by particles while dashed-dotted line joins respective average values for each disorder. Solid line displays the prediction of the mean-field approximation in the thermodynamic limit.

Making use of the analytical form Eq. (3) we analyze some analytical tools which might be employed in order to specify and analyze SFM systems. Similarly to methods of high energy physics (cf, e.g., [7,8] and refs. therein) the correlations of avalanche size distribution might provide the criticality signals and a tool specifying and quantifying magnetic structure. For certain $i$th (re)magnetization event we define the mean noise signal

$$<p> = \sum_S S D(S) / \sum_S D(S) = (N_{\text{tot}} - 1) / \Pi - S_b$$  \hspace{1cm} (4)

where the sum runs over the avalanche sizes $S$ excluding the biggest one, the quantity $N_{\text{tot}}$ gives the total number of noisy jumps, i.e. avalanches. Substituting Eq. (3) into Eq. (4) the mean avalanche size, i.e. mean value for magnetic emission signals, is evaluated to be $<p>_{\text{mf}} \approx [d]^{-3} + \text{const}(d)$, and diverges at critical conditions, i.e. $d \to 0$, in the thermodynamic limit $\Pi \to \infty$.

If the system undergoes a critical behaviour being a precursor of SO criticality in some particular (re)magnetization events, strong correlations will appear in magnetic noise. For instance, in case of magnetodynamics we can study correlations between the strongest signal (i.e., the largest avalanche $S_b$) and the mean signal value $<p>$ for remaining avalanches in this particular event, e.g., from
numerical model or experimental data. These correlations are similar to the Campi scatter plots [7,8]. In Fig. 3B we plot the mean avalanche size versus the length of largest avalanche for certain event. The data in Fig. 3B were obtained from RJIM simulations assuming various disorders as partially presented in Fig. 3A. In Fig. 3B we can clearly distinguish two branches corresponding to under-critical, i.e., large size of the biggest jump amplitude and small mean value, and over-critical, i.e. small size of the biggest avalanche and small average values. The right branch consists mainly of events with small disorders, while the left branch originates from events having large $R$. The set of two branches meet in the critical region. The results of the mean field approximation in the thermodynamic limit are in a reasonable agreement with numerical data for overcritical disorders. At sub-critical conditions the mean field approach reproduces only qualitatively the RJIM model simulations.

3. Conclusions

We considered the magnetic structure of nano-particle assemblies accounting for inter- and intra-particle properties. Spatial quantization due to intra-particle confinement brings sharp step-wise anomalies for magnetic moments in external field. As was shown in [3,4] such moment jumps in conjunction with ferromagnetic inter-particle coupling induce jerky magnetodynamics with sharp steps in the array (de)magnetization process. Making use of the RJIM model [3,4] we account for quantum effects due to the particle discrete levels, inter-particle coupling and disorder. MSE and phase diagram of such system are demonstrated to exhibit spinodal regions on {R,B}-plane. In a case of weak coupling these regions of instability are well separated and surround the positions of the particle magnetic anomalies. With increasing coupling we observe further structure modification, plausibly, of bifurcation type. At strong coupling the instability region become wide while the stable regimes are reduced to narrow islands at small disorders. We consider analytical tools exploring correlations of magnetic noise amplitudes and demonstrate an application for quantitative definition, description and study of SFM structure and origin, as well as self-organized criticality. Further analysis based on numerical simulations within RJIM model and implications of proposed tools will shed a light on the origin and properties of this transition.

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