Measuring magnetic correlations in nanoparticle assemblies

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Measuring magnetic correlations in nanoparticle assemblies

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Abstract. We illustrate how to extract correlations between magnetic moments in assemblies of nanoparticles from, e.g., electron holography data providing the combined knowledge of particle size distribution, inter-particle distances, and magnitude and orientation of each magnetic moment within a nanoparticle superstructure, We show, based on simulated data, how to build a radial/angular pair distribution function $f(r,\theta)$ encoding the spatial and angular difference between every pair of magnetic moments. A scatter-plot of $f(r,\theta)$ reveals the degree of structural and magnetic order present, and hence provides a measure of the strength and range of magnetic correlations.

[1] Introduction

Assemblies (or superstructures) of magnetic nanoparticles arranged over a superlattice represent a novel class of magnetic material where inter-particle exchange interactions may be negligible, and, hence, dipolar inter-particle interactions tend to dominate. As a result, magnetic superstructures behave differently than conventional magnets and their properties may be controlled and tuned by selecting the spacing and compositions of their constituents. Recently [1], we have studied dipolar interactions in self-assembled 15 nm cobalt particle structures using off-axis electron holography [2]. This technique enables the determination of orientation and magnitude of the magnetic moment of each nanoparticle [3] coupled with the structural ordering (particle arrangement and spacing) of the superlattice. Our study revealed that dipolar interactions are sufficiently strong to support long-range ferromagnetic order in quasi-2D superstructures, even when the lattice of nanoparticles is highly disordered. This observation supports the possibility of creating amorphous dipolar magnets, in contrast to the expectation that a disordered dipolar system may lead to spin-glass-like behaviour.

In order to quantify dipolar ferromagnetic order and reveal magnetic correlations, the first step is to extract the magnitude and orientation of each magnetic moment present in the structure. Considering that particles may be in the 10-20 nm size range, only a few techniques have the necessary single-particle spatial resolution and sensitivity to nanoscale magnetic fields. Electron holography [2] is one of such techniques: by creating an interference pattern between the electron wave scattered magnetically from the sample and a planar reference wave, it is possible to obtain a quantitative map of the projected local magnetic field within and around every nanoparticle in the system. The magnitude

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and orientation of each nanoparticle's (in-plane) magnetic moment is then measured from the local projected magnetic field, corrected with a proper demagnetization factor to accommodate the particles' shapes. Location, size and shape of each particle can be extracted e.g. from conventional bright field micrographs and particle-size analysis. Combining the information on particles arrangement with their overall magnetic state gives us access to the strength and range of magnetic correlations, as will be illustrated in the following section by the analysis of simulated data.

[2] The radial/angular pair distribution function

Consider a finite superstructure made of spherical single-domain nanoparticles with negligible magnetocrystalline anisotropy arranged over a planar close-packed lattice. Let the number of particles along the two axes be N and M, so that the total number of particles is $n=NM$; each particle, of radius R_i sits at location $\mathbf{r}_i$ and has a magnetic moment $\mathbf{m}_i$, with $i=1..n$. The quantities R_i, $\mathbf{r}_i$, $\mathbf{m}_i$ are assumed to be known; for example they can be measured from electron microscopy data as explained above, or by any other technique capable of providing the required information. We build the radial/angular distribution function $f(r,\theta)$ by calculating $r=|\mathbf{r}_i-\mathbf{r}_j|$ and $\theta=\arccos[\mathbf{m}_i\cdot\mathbf{m}_j/(m_i m_j)]$ for every pair of particles. If the lattice is regular, r can only assume discrete values; with disorder, r varies continuously but will peak at every nearest neighbor distances, with peak widths proportional to the degree of disorder. We define the degree of lattice disorder σ as the ratio between the first neighbor peak half-width and the first neighbor distance.

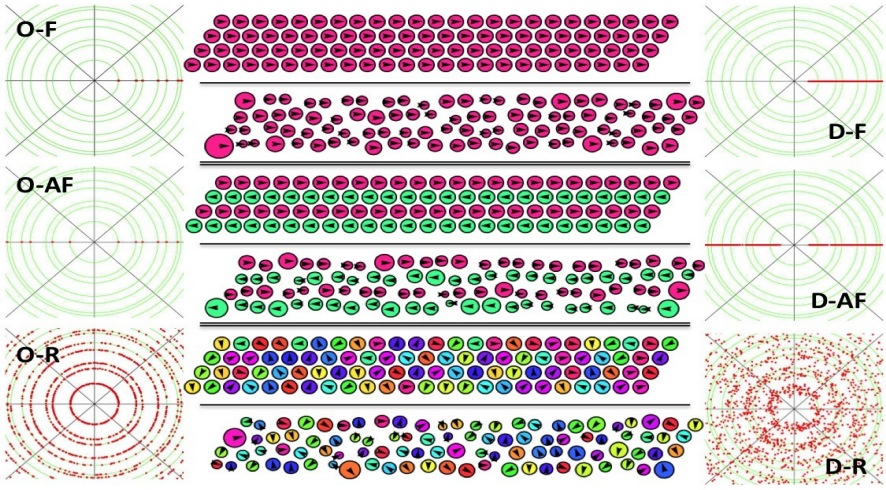

Figure 1. Radial/angular correlations (left, right) of ordered (O) and disordered (D) nanoparticle structures with imposed ferromagnetic (F), antiferromagnetic (A) and random (R) order.

The angular part reflects magnetic correlations: if the magnetic state is purely ferromagnetic, then $\theta=0$, independently of range; if the magnetic state is ferromagnetic only at short-ranges, then θ will be small for the first few nearest neighbor peaks, and then spread out at larger distances. To illustrate the behavior of $f(r,\theta)$ in a few representative scenarios, we consider a $N=24$, $M=4$ close-packed superstructure, in the outer shape of a sheared rectangle, structurally ordered (O, $\sigma=0$) and disordered

(D, σ=0.4), and impose three particular magnetic states upon it: ferromagnetic (F), antiferromagnetic (AF), random (R). As shown in Figure 1, the O-F system is characterized by clustering of the θ=0 (full alignment) points at the exact n^{th}-nearest neighbor distances (indicated with green circles), highlighting that magnetic correlations are present at long range; in contrast, the D-F system maintains the θ=0 independent on *r*, but the points are dispersed along the *r*-axis, broadening the possible neighbor distances distribution. The O-AF and D-AF systems behave similarly, except now we have two values of θ: θ=0, and θ=π; note, however, the absence of the θ=π point on the 3rd-neighbor circle due to the particular configuration (no 3rd-neighbour pairs are antiparallel). The O-R shows clearly the clustering of θ points over the neighbor distance circles, with random spreading over the angle, indicating total loss of magnetic correlations; the D-R system shows θ points spread randomly through the angle and the distance, although some radial oscillations remain (barely) visible.

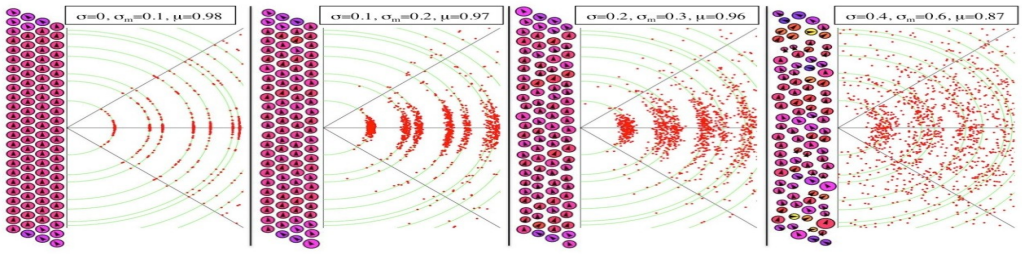

Figure 2. Superstructure of magnetic particles (*N*=24, *M*=4) with a mean diameter of 15 nm, a mean magnetic moment of 2.7 10^5 Bohr magnetons in a close-packed arrangement with lattice parameter of 20 nm and varying degree of structural disorder (σ=0,0.1,0.2,0.4 from left to right).

[3] Magnetic correlations as a function of lattice disorder

Allowing magnetic relaxation (at a temperature low enough to bypass superparamagnetic issues), the system evolves towards some energy minimum, reachable from the chosen initial condition. If we select relaxation from an initial F state, a situation representing the removal of a strong applied field and the relaxation to remanence, we can examine how magnetic correlations vary as a function of lattice disorder. Figure 2 shows four 24x4 systems, with σ=0,0.1,0.2,0.4; here we also measure the total net moment μ of the structure, normalized to its maximal value at the initial condition (saturation). The σ=0 case differs from the D-F system analyzed earlier in that now edge effects are playing a role introducing a number of high-θ points. As disorder increases, so does the angular spread of the magnetic moments, although even in the σ=0.4 case, where the lattice is barely distinguishable from amorphous, ferromagnetic correlations are still present: the angular difference near the nominal first neighbor distance has a standard deviation of about σ_m=0.6 rad=30°, meaning that at short-range most moments are aligned within 60°, and the assembly maintains a net remanent moment of 0.87.

In conclusion, we have illustrated how to measure magnetic correlations, and examined the radial/angular pair distribution function for 24x4 close-packed planar superstructures with varying disorder parameter, highlighting the role of dipolar interactions in maintaining long-range ferromagnetic order in quasi-2D assemblies of nanoparticles

References

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