Scaling behavior of the dipole coupling energy in two-dimensional disordered magnetic nanostructures

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Numerical calculations of the average dipole-coupling energy $\mathcal{E}_{dip}$ in two-dimensional disordered magnetic nanostructures are performed as function of the particle coverage $C$. We observe that $\mathcal{E}_{dip}$ scales as $\mathcal{E}_{dip} \propto C^{\alpha}$ with an unusually small exponent $\alpha^* \approx 0.8-1.0$ for coverages $C \lesssim 20\%$. This behavior is shown to be primarily given by the contributions of particle pairs at short distances, which is intrinsically related to the presence of an appreciable degree of disorder. The value of $\alpha^*$ is found to be sensitive to the magnetic arrangement within the nanostructure and to the degree of disorder. For large coverages $C \gtrsim 20\%$ we obtain $\mathcal{E}_{dip} \propto C^{\alpha}$ with $\alpha = 3/2$, in agreement with the straightforward scaling of the dipole coupling as in a periodic particle setup. Taking into account the effect of single-particle anisotropies, we show that the scaling exponent can be used as a criterion to distinguish between weakly interacting ($\alpha^* \approx 1.0$) and strongly interacting ($\alpha^* \approx 0.8$) particle ensembles as function of coverage.

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I. INTRODUCTION

Nanostructured materials consisting of interacting magnetic particles are currently the subject of an intense research activity, driven by their fundamental interest and technological perspectives. Numerous experimental and theoretical studies have been performed for various two-dimensional (2D) and three-dimensional (3D) arrangements having different degrees of structural and magnetic disorder. The magnetic behavior of these systems is determined both by single-particle properties, like lattice or shape anisotropies, and by interparticle interactions, like magnetic dipole coupling or indirect exchange. The relative importance of these two types of contributions can be tuned experimentally, at least to some extent, by varying the particle-size distribution, the nanostructure morphology, and the average interparticle distance, for example. For the weakly interacting regime the single-particle properties dominate, and the interactions can be handled as perturbations. However, in the most interesting case of strongly interacting systems, where the interparticle couplings are stronger than the single-particle energies, an explicit treatment of the interactions is crucial. Taking into account the complexity of the problem, particularly in the presence of disorder, it is a challenge to derive simple trends for the dependence of the interaction energy on the main nanostructure characteristics.

In this study we focus on the dipole coupling energy $E_{dip}$ for magnetic particle ensembles with varying particle coverages $C$ and different kinds of nanostructures. Since the dipole interaction energy between two particles scales with the average interparticle distance $R$ as $R^{-3}$, and since $C$ is proportional to $R^{-D}$, $D$ being the spatial dimension, one could in principle expect that the average dipole energy $\mathcal{E}_{dip}(C)$ should be proportional to $C^{\alpha}$, with $\alpha = 3/D$. However, several previous studies indicate a different scaling behavior for disordered dipole-coupled systems. For example, a weaker decrease of $\mathcal{E}_{dip}$ characterized by an effective exponent $\alpha^* = 0.8 \pm 0.1$ has been found experimentally upon thinning of ferrofluids ($D=3$). In addition, the strength of the dipole coupling seems to be underestimated as compared to measurements, when one calculates its effect on the energy barrier for single-particle magnetic reversals. Moreover, numerical results for $\mathcal{E}_{dip}$ of a ferrofluid monolayer ($D=2$) with different particle coverages reveal significant deviations from the $\alpha = 3/2$ scaling. These experimental and theoretical findings suggest that the effective dipole energy in disordered systems decreases more slowly with decreasing coverage than originally expected.

The purpose of this study is to determine the role of global nanostructure characteristics, such as coverage and degree of disorder, on the average dipole-coupling energy of 2D particle ensembles. In particular we show that the scaling exponent $\alpha$, $E_{dip} \propto C^\alpha$, allows to discern whether $E_{dip}$ is dominated by the average interparticle distance $R$, as in a periodic particle setup, or by short-distance contributions. Moreover, taking into account single-particle anisotropies we demonstrate that the value of $\alpha$ can be used as a criterion to distinguish between weakly and strongly interacting disordered particle ensembles.

II. MODEL SIMULATIONS

The 2D nanostructure is modeled using a square unit cell containing non-overlapping, sphere-shaped magnetic particles, and periodic boundary conditions. We consider disordered particle ensembles with a fully random distribution within the unit cell, as well as disturbed square arrays where the particles are scattered around periodic lattice sites by applying a Gaussian distribution $P(r)$ with standard deviation $\sigma_R$. For such planar particle
ensembles the overall coverage $C$ is related to the average interparticle distance $R > 2 r_0$ by $C = \pi (r_0/R)^2$. A uniform-particle size distribution with $N \simeq 24000$ atoms per particle is assumed, which corresponds to a particle radius $r_0 \simeq 16 a_0$, where $a_0$ is the nearest-neighbor interatomic distance. Due to the strong interatomic exchange interaction and for particle diameters smaller than typical magnetic domain wall widths, each particle can be regarded as a single-domain Stoner-Wohlfarth magnet carrying a giant spin $M \simeq N \mu_{at}$, where $\mu_{at}$ is the magnetic moment per atom. For simplicity, the particle magnetizations $M_i$ are restricted to be within the plane of the nanostructure. The magnetic configuration is thus characterized by the set of in-plane angles $\{\phi_i\}$, with $i = 1, \ldots, N$. For each given particle distribution and magnetic configuration we determine the magnetic energy

$$E = E_{\text{anis}} + E_{\text{dip}} = -K_2 \sum_i (M_i \cdot n_i)^2$$

$$+ \frac{1}{2} \sum_{i,j} [M_i \cdot M_j - 3 (r_{ij} \cdot M_i) (r_{ij} \cdot M_j)] (r_{ij} \cdot r_{ij}^{-5}) \ . \ (1)$$

The first term represents the single-particle anisotropy energy of particle $i$, resulting from spin-orbit interactions and the shape anisotropy. For simplicity, a uniaxial anisotropy is assumed with strength $K_2$ per atom and easy axis directed along a given unit vector $n_i$. The second term of Eq. (1) is the dipole-dipole interaction between magnetic moments $M_i$, where $r_{ij} = |r_{ij}| = |r_i - r_j|$ is the distance between the centers of particles $i$ and $j$. The infinite range of the dipole interaction is taken into account by applying an Ewald-type summation over all periodically arranged unit cells of the extended planar system. In addition, we also include the leading correction resulting from the dipole-quadrupole interaction, which takes into account the effect of finite particle sizes.

In order to determine the average magnetic energy $E = \bar{E}_{\text{anis}} + \bar{E}_{\text{dip}}$ we start from an arbitrary initial guess of the angles $\phi_i$ of each particle, and relax the magnetic configuration to a nearby local minimum of the total energy, using a conjugated gradient method. The resulting relaxed states depend in general on the initial configurations and show strong magnetic noncollinearities. The large diversity of metastable states which is characteristic of disordered particle arrangements is taken into account by averaging $E$ over many different random initial guesses (typically $\sim 100$) for the same spatial arrangement of the particles. Notice that this procedure is not intended to search preferently for the global energy minimum. For comparison, $E$ is also calculated for the same particle setup but assuming a parallel alignment of all particle magnetizations, i.e., a ferromagnetic state with $\phi_i = \phi_{\text{fm}}$ and averaging over the common magnetization angle $\phi_{\text{fm}}$. Note that for a disordered particle ensemble such a ferromagnetic state does not usually correspond to a local energy minimum. In addition, a second average over the spatial configuration of the nanostructure is necessary in order to minimize spurious effects of the finite size of the unit cell. For this purpose, an appropriate number (typically $\sim 200$) of different realizations of the unit cell is considered, using the same global variables which characterize the particle ensemble. In the following calculations we use parameters appropriate for Fe, i.e., $\mu_{at} = 2.2 \mu_B$ and $a_0 = 2.5 \AA$, yielding the dipole energy unit $E_{\text{dip}}^0 = \mu_{at}^2/a_0^3 = 0.19$ K. Concerning the strength $K_2$ of the uniaxial anisotropy we consider values appropriate for films and particles of 3d transition metals ($K_2 = 0.1 - 1$ K/atom).

III. RESULTS AND DISCUSSION

First we focus on strongly interacting 2D systems of magnetic particles, neglecting the single-particle contribution ($K_2 = 0$). In Fig.1 results are given for the absolute value of the average dipole energy per particle $|\bar{E}_{\text{dip}}(C)|$ as function of particle coverage $C$ for a fully random particle ensemble. $|\bar{E}_{\text{dip}}(C)|$ is shown for the relaxed magnetic arrangements as well as for the ferromagnetic ones. From the log-log-plot of $|\bar{E}_{\text{dip}}(C)|$ one observes that the exponent $\alpha = 3/2$, corresponding to the straightforward scaling of the dipole interaction for $D = 2$, is approximately valid only for large coverages $C \gtrsim 20\%$. For smaller $C$, in the investigated range $0.1\% \leq C \leq 20\%$, one finds a different dependence with clearly smaller effective exponents. For the ferromagnetic arrangement we obtain $\alpha_{\text{fm}}^* \approx 1.0$ and, quite interestingly, for the relaxed solution an even smaller value $\alpha_{\text{rf}}^* \approx 0.8 - 0.9$. The numerical accuracy of the calculated exponents $\alpha_{\text{fm}}^*$ and $\alpha_{\text{rf}}^*$ is estimated to be about $\pm 0.02$.

These remarkable results can be qualitatively ex-
plained by the following simple analysis. Consider two non-overlapping particles with radius \( r_0 \) in a \( D \)-dimensional space coupled by an interaction \( E_n(r) \propto r^{-n} \). Let the first particle be located at \( r = 0 \), and let \( P(r) \) be the probability to find the second one between \( 2r_0 \) and a maximal distance \( R_{\text{max}} \). The upper bound \( R_{\text{max}} \) is related to the particle concentration by \( C \propto (r_0/R_{\text{max}})^D \), whereas the lower bound \( 2r_0 \) results from the hard core (excluded volume) of the non-overlapping particles. The average interaction energy \( \langle E_n \rangle \) of such a particle pair is proportional to the average

\[
\langle r^{-n} \rangle = \int_{2r_0}^{R_{\text{max}}} dr \frac{r^{D-1}}{r^{D-1}} P(r) r^{-n} \int_{2r_0}^{R_{\text{max}}} dr \frac{r^{D-1}}{r^{D-1}} P(r).
\]

(2)

For a random distribution corresponding to a constant \( P(r) \) one obtains

\[
\langle r^{-n} \rangle = \frac{D}{n-D} \frac{R_{\text{max}}^{n-D} - (2r_0)^{n-D}}{(2r_0 R_{\text{max}})^{n-D} [R_{\text{max}}^{n-D} - (2r_0)^{n-D}]} \quad \text{(3)}
\]

for \( n > D \), and

\[
\langle r^{-n} \rangle = \frac{D \ln(R_{\text{max}}/2r_0)}{R_{\text{max}}^{n-D} - (2r_0)^{n-D}} \quad \text{(4)}
\]

for \( n = D \). In the limit of low coverages, \( 2r_0 \ll R_{\text{max}} \to \infty \), this yields

\[
\langle r^{-n} \rangle \approx \frac{D}{n-D} \frac{2^D}{(2r_0)^n} \left( \frac{r_0}{R_{\text{max}}} \right)^D \propto (2r_0)^{-n} C, \quad \text{(5)}
\]

for \( n > D \), and a similar limit for \( n = D \). For \( n < D \) one finds \( \langle r^{-n} \rangle \propto C^{n/D} \), which coincides with straightforward scaling. Notice that for \( n \geq D \) the average interaction energy \( \langle E_n \rangle \) for \( R_{\text{max}} \to \infty \) is no longer proportional to \( R_{\text{max}}^{n-D} \). Instead, an effective exponent \( n^* = D \) is obtained. Consequently, as function of concentration one obtains \( \langle E_n \rangle \propto C^{n/D} \) with \( n^* = 1 \). The physical reason behind this behavior is that only the upper bound \( R_{\text{max}} \) scales with \( C \), since it is proportional to the average interparticle distance, but not the lower bound \( 2r_0 \). Hence, the average energy is dominated by the interactions between particle pairs at distances of the order of the particle diameter, which is proportional to \( (2r_0)^{-n} \), multiplied by the probability to find such a particle pair, which is proportional to \( C \). In other words, as a consequence of the relatively strong distance dependence of the interaction for \( n \geq D \), in a diluted random system the average energy \( \langle E_n \rangle \) is dominated by the short-distance contributions, and not by the energy corresponding to the average interparticle distance. Notice that if \( n \geq D \), \( \langle E_n \rangle \propto C \) is independent of the particular kind of interaction characterized by the number \( n \). Moreover, this behavior of a random particle ensemble is also obtained for an exponentially decreasing interaction \( E \propto \exp(-\lambda r) \) for all \( D \).

In the context of the present study corresponding to \( D = 2 \) and \( n = 3 \), one concludes that a 2D random ensemble of magnetic nanoparticles exhibits a stronger than expected average dipole interaction, which scales with the particle coverage as \( \langle E_{\text{dip}} \rangle(C) \propto C^{\alpha^*} \) with \( \alpha^* \approx 1.0 \). In contrast, if one varies the interparticle distance in a periodic array the smallest distance between particle pairs also scales with coverage, yielding therefore the straightforward scaling behavior with \( \alpha = 3/D \). Thus, by analyzing the exponent \( \alpha \) one can distinguish whether the average dipole energy is dominated by short-distance contributions or by the energy corresponding to the average interparticle distance. The difference between \( \alpha \) and \( \alpha^* \) is significant for 1D and 2D systems, whereas for 3D systems one expects \( \alpha \approx \alpha^* \approx 1 \).

Another interesting effect is the difference in the exponents \( \alpha^*_n \) and \( \alpha^*_m \) obtained for the relaxed and ferromagnetic arrangements. This seems to be an indication of the formation of particle chains with correlated ‘head-to-tail’ alignments of the magnetizations of close-by particles. Such magnetic rearrangements are possible only by relaxing the angles \( \phi_i \). In fact, recent calculations have shown that \( \langle E_{\text{dip}} \rangle \) increases with increasing positional disorder as a result of magnetic relaxations. Moreover, reduced values of \( \alpha^* = 0.8 \pm 0.1 \) have also been observed in experiments on frozen high-density ferrofluids.

By closer inspection of Fig.1 one observes that the exponent \( \alpha^*_n \) is not exactly the same for all coverages, since a slight change of slope appears in the curve of the relaxed solution. Moreover, differences between \( \alpha^*_n \) and \( \alpha^*_m \) are found in the coverage range where magnetic relaxation driven by the dipole coupling is significant, i.e., in the strongly interacting regime. This can be demonstrated by considering in addition to the dipole coupling a single-particle uniaxial anisotropy \( K_2 \) with randomly distributed directions \( \mathbf{n}_i \) of the easy axes. Unless the average anisotropy energy \( \langle E_{\text{anis}} \rangle \) is so strong that it dominates over \( \langle E_{\text{dip}} \rangle \) even for large coverages, a crossover from the strongly interacting regime (\( \langle E_{\text{anis}} \rangle < \langle E_{\text{dip}} \rangle \)) to the weakly interacting regime (\( \langle E_{\text{anis}} \rangle > \langle E_{\text{dip}} \rangle \)) takes place with decreasing \( C \) at the crossover coverage \( C(K_2) \). This is shown in Fig.2 where, as in Fig.1, \( \langle E_{\text{dip}} \rangle \) and \( \langle E_{\text{anis}} \rangle \) are given as function of \( C \) for different values of \( K_2 \). We have determined the relaxed magnetic arrangements in presence of local anisotropies, and have derived the scaling exponents \( \alpha^*_n \) corresponding to the dipole contribution of the magnetic energy. First of all, one observes that the scaling behavior, as estimated by Eq.(5) and depicted in Fig.1, is also valid in presence of single-particle anisotropies. In the coverage range where \( \langle E_{\text{dip}} \rangle \) is dominated by short-distance contributions, i.e., for \( C \lessgtr 20\% \), and in the strongly interacting regime \( C > \tilde{C} \) where \( \langle E_{\text{dip}} \rangle > \langle E_{\text{anis}} \rangle \), a reduced value of \( \alpha^*_n < 1 \) is clearly observed. As \( C \) is further decreased, \( \alpha^*_n \) changes gradually to \( \alpha^*_n \approx 1.0 \) below the crossover coverage \( \tilde{C} \). This can be understood by recalling that magnetic relaxation driven by dipole coupling is significant only in the strongly interacting regime. In the opposite case of weak interactions \( C \lessgtr \tilde{C} \) relaxation due to dipole coupling is negligible, thus resulting in \( \alpha^*_n \approx \alpha^*_m \approx 1.0 \).
Note that for the largest considered value of $K_2$ the crossover happens directly from $\alpha = 3/2$ to $\alpha^*_r \simeq 1.0$, since here $\bar{C}(K_2) \simeq 20\%$. Consequently, the determination of the scaling exponent $\alpha^*_r$ allows one to discern between strongly and weakly interacting ensembles.

The average anisotropy energy $|\overline{E}_{\text{anis}}|$, is of course independent of coverage $C$ in the absence of dipole interactions and other magnetic couplings. A dependence on $C$ appears only if the interactions force the particle magnetizations $\mathbf{M}_i$ to deviate from the local easy axes $\mathbf{n}_i$, cf. Eq.1. Moreover, the exponent $\alpha^*_\text{fm}$ for a ferromagnetic state is not affected at all by single-particle anisotropies. Since for such an arrangement a magnetic relaxation is not effective, the single-particle energy yields a contribution to the total magnetic energy which is independent of the coverage. For anisotropies with aligned easy axes directions we obtain the same behavior of $\alpha^*_r$ as for randomly distributed ones, since the dipole-coupling-induced relaxation of the particle magnetizations is not much affected by the easy axes distribution.

In order to study the differences between $\alpha^*_r$ and $\alpha^*_\text{fm}$, in further detail we have also considered 1D systems of dipole-coupled particles. In this case the particle magnetizations are aligned parallel to the chain axes in the most stable state. A numerical investigation of such 1D systems with randomly assembled particles yields effective exponents $\alpha^*_\text{fm} \simeq \alpha^*_r \simeq 1.0$. This is quite reasonable since in this case the relaxed and ferromagnetic solutions coincide. In the case of 1D stripes with finite widths, we observe a smooth crossover from $\alpha^*_r \simeq 1$ to $\alpha^*_r \simeq 0.8$ with increasing stripe width in the investigated coverage range $0.1\% \leq C \leq 20\%$. These findings confirm the idea that the reduction of $\alpha^*_r$ with respect to $\alpha^*_\text{fm}$ in 2D and 3D nanostructures is the result of magnetic relaxations.

To quantify the role of disorder on the coverage dependence of $\overline{E}_{\text{dip}}$, we have studied the crossover between periodic and fully random particle setups by considering systems with intermediate degrees of positional disorder. Here the single-particle anisotropy is neglected for simplicity ($K_2 = 0$). Two-dimensional particle ensembles have been constructed with a quasi-periodic arrangement in which the particles are scattered around the sites of a square lattice following a Gaussian-like distribution with standard deviation $\sigma_R$. A disturbed square particle array is assumed, neglecting single-particle anisotropies ($K_2 = 0$). The exponent corresponds to the coverage range $0.1\% < C < 20\%$. The dashed curve refers to $\alpha^*$ as derived from Eq.3. The horizontal lines indicate $\alpha^*_r$ and $\alpha^*_\text{fm}$ calculated for a fully random particle ensemble.

These numerical results are corrob-
rated by the simple analytical estimate given by Eq. (3), using for $P(r)$ a Gaussian distribution with mean $R_{\text{max}}/2$ and standard deviation $\sigma_R$. The different scaling properties of ordered and disordered nanostructures confirm the importance of the sample characteristics on the effective scaling exponents.

Finally, several other effects have been investigated which could in principle modify the values of the exponents $\alpha^\ast$. We have considered (i) unit cells with different numbers of particles, (ii) particle-size dispersions with Gaussian or log-normal distributions, (iii) disk-shaped particles with the same particle radius $r_0$ as the previously considered sphere-shaped particles, and (iv) the role of dipole-quadrupole corrections to the point-dipole sums. While the magnitude of the importance of the sample characteristics on the effective scaling exponents $\alpha$ and standard deviation $\sigma$ and $\alpha^\ast$ remain unchanged within numerical accuracy. These results confirm the validity of the previously discussed trends and reinforce the idea that $\alpha^\ast$ can be used as a parameter to characterize some aspects of the magnetic behavior of interacting magnetic nanostructures with varying degrees of disorder.

IV. CONCLUSION

The average dipole energy $E_{\text{dip}}$ of two-dimensional disordered magnetic nanostructures has been determined by using numerical simulations and simple analytical estimates. A dependence of the form $|E_{\text{dip}}(C)| \propto C^{\alpha^\ast}$ is obtained with nearly constant $\alpha^\ast$ over a wide coverage range. For $0.1% < C < 20\%$ the exponent $\alpha^\ast \approx 0.8$–1.0 is significantly smaller than the value $\alpha = n/D = 3/2$ expected from a straightforward scaling of the dipole interaction. This scaling results from dominating short-distance contributions, and is intrinsically related to the presence of significant disorder. In contrast, for $C \gtrsim 20\%$ the behavior of $E_{\text{dip}}(C)$ is determined mainly by the average interparticle distance, which depends on coverage as $R \propto C^{-1/D}$, and therefore yields $\alpha = 3/2$, for a periodic particle setup. A smooth transition between these two scaling regimes is observed with increasing degree of disorder.

Taking into account the effect of single-particle anisotropies, we have shown that the value of the scaling exponent $\alpha^\ast$ allows to distinguish between strongly and weakly interacting particle ensembles, since $\alpha^\ast$ is sensitive to the magnetic arrangement within the nanostructure. Indeed, one observes a crossover from $\alpha^\ast \approx 0.8$, when magnetic relaxations driven by dipole couplings dominate, to $\alpha^\ast \approx 1.0$, when the directions of the particle moments are determined mainly by local anisotropies. It would be very interesting to extend these investigations to 3D nanostructures in view of the numerous experimental results that are available for such systems. In addition, the scaling behavior should be analyzed as function of temperature, since it is likely to be affected by thermally induced crossings of energy barriers between different magnetic arrangements.

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