Low-energy properties of two-dimensional magnetic nanostructures: interparticle interactions and disorder effects

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Abstract. The low-energy properties of two-dimensional ensembles of dipole-coupled magnetic nanoparticles are studied as a function of structural disorder and particle coverage. Already small deviations from a square particle arrangement lift the degeneracies of the microvortex (MV) magnetic configuration and result in a strongly inhomogeneous magnetic order of the particle ensemble. The energy distribution of metastable states is determined. For a low degree of disorder a strongly asymmetric shape with a pronounced peak of the ground-state energy results. In contrast, for a strong disorder a Gaussian-like distribution is obtained. The average dipole energy \( \overline{E}_{\text{dip}} \) decreases with increasing structural disorder. Above a coverage-dependent degree of disorder \( \overline{E}_{\text{dip}} \) resembles the average dipole energy of a random particle set-up, for which a simple scaling behaviour is derived. The role of vacancies has been studied for a square particle array by determining the angular distribution of the preferred MV angle as a function of the vacancy concentration. Preferred angles along the axial as well as along the diagonal directions of the square array are obtained. A corresponding investigation for disturbed square arrays yields preferred MV angles only along the axial directions. The effect of dipole–quadrupole corrections resulting from the finite size of the particles is quantified.

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1. Introduction

Driven by their fundamental interest and technological perspectives, interacting magnetic nanostructured materials are currently the subject of intense research activity [1]. Numerous experimental and theoretical studies have been performed for various two-dimensional (2D) [2] and three-dimensional (3D) [3]–[8] arrangements of nanometre-size magnetic particles having different degrees of structural and magnetic disorder. The magnetic behaviour of these systems is determined by single-particle properties (e.g. particle moments, lattice and shape anisotropies, etc), by the composition and morphology of the nanostructure, and in particular by the nature of the dominant interparticle interactions. The latter comprises especially the magnetic dipole coupling, which will be addressed in the present study. Other interparticle interactions are, for example, the Rudermann–Kittel–Kasuya–Yosida (RKKY) indirect exchange mediated by the conduction electrons of a metallic substrate, or the short-range direct exchange in the case when the particles are in contact. The relative importance of single-particle versus interparticle contributions can be tuned experimentally, at least to some extent, by changing sample characteristics such as the particle-size distribution or the average interparticle distance. For low particle coverages the interactions can be treated as a perturbation to single-particle properties. However, for dense particle ensembles the interparticle interactions become increasingly important and eventually dominate. In this interesting case the single-particle approach is no longer applicable and an explicit treatment of the interactions is unavoidable [3]–[5]. The work presented refers to this strongly interacting case.

A fundamental question in this context is to identify and understand the collectively ordered magnetic states which are induced by the interactions in such particle ensembles. A few basic properties of strictly periodic dipole-coupled systems are summarized. The ordering of dipole-coupled magnetic moments is sensitive to the lattice structure. For instance, the ground state of a square lattice of equal-sized (monodispersed) particles is the so-called microvortex (MV) magnetic arrangement [9, 10]. This magnetic structure is characterized by the MV angle \( \phi_{\text{mv}} \), where the angles of the particle magnetizations of a plaquette of four neighbouring particles are given by \( \phi_1 = \phi_{\text{mv}} \), \( \phi_2 = -\phi_{\text{mv}} \), \( \phi_3 = 180^\circ + \phi_{\text{mv}} \) and \( \phi_4 = 180^\circ - \phi_{\text{mv}} \). The particular cases characterized by multiples of \( \phi_{\text{mv}} = 90^\circ \) represent columnar states, consisting of ferromagnetic (FM) rows or columns with alternating signs of magnetizations. Evidently, the MV state has a vanishing net magnetization. The parallel or FM state has a larger energy and is actually an unstable solution. Also, the honeycomb lattice has a ground state with a vanishing net magnetization which is similar to the MV state of the square lattice [10]. In contrast, for the hexagonal lattice the ground state is FM [11, 12].
Despite the fact that the dipole interaction is not rotationally invariant, the ground state of these dipole-coupled periodic particle arrays are continuously degenerate with respect to a rotation of the MV angle $\phi_{mv}$ or of the FM angle $\phi_{fm}$, respectively. This holds for classical spins at $T = 0$. Thermal fluctuations, quantum fluctuations or a structural disorder immediately lift these accidental ground-state degeneracies of the periodic structures. The energy lowering associated with the symmetry breaking stabilizes the system in some particular magnetic orientations. Therefore, the square and honeycomb lattices are said to show the so-called "order-by-disorder" effect [10, 13]. This implies that internal (nontrivial) degeneracies, e.g. the relative directions of magnetic sublattices, are lifted by the presence of disorder, and that particular directions of the sublattice magnetizations are preferred. For a dipole-coupled square lattice the fluctuations induce a fourfold in-plane magnetic anisotropy. As reported by Prakash and Henley [10], the preferred in-plane magnetic orientations for this lattice are the axial directions (columnar states) for thermal and quantum fluctuations and the diagonal directions for a structural disorder induced by a small amount of vacancies. The reason is that the magnetic excitations are not continuously degenerate [14]. Thus, one of the prerequisites of the Mermin–Wagner theorem, which excludes an ordered state for a continuously degenerate 2D system at finite temperatures [15], is not satisfied in this case. Notice that, in the presence of a dipole interaction, which decreases with distance $r$ as $r^{-3}$, already another prerequisite of this theorem is not fulfilled, namely a short-range interaction $\propto r^{-\alpha}$ with $\alpha \geq 2D$, where $D$ refers to the spatial dimension [16]. Therefore, 2D ferromagnets and antiferromagnets exhibit a collectively ordered state with a critical temperature of the order of the exchange coupling $J$, provided that, for example, dipole coupling is present even if this interaction is weak compared to $J$ [17]. As shown by Monte Carlo calculations and interacting spin wave theory, purely dipole-coupled magnets in 2D also exhibit collectively ordered states at finite temperatures, which correspond to the MV state for a square lattice and to the FM state for a hexagonal lattice [14].

Structural disorder, which is usually present in real magnetic nanostructures, results, for example, from the size and shape dispersion, positional disorder or random anisotropy axes. Due to the nonuniform and competing nature of the magnetic couplings the magnetic ordering in disordered particle ensembles is similar to that of a spin glass [18]. Thus, many different metastable states exist, which are characterized by complex magnetic structures. An intriguing question in this context is whether the dipole interaction in disordered planar particle ensembles results in a collectively ordered magnetic state. Several experiments on interacting, high-density ferrofluid systems indicate the onset of a collectively ordered state below a characteristic, concentration-dependent temperature. For example, at this temperature a ‘critical slowing down’ of the magnetic relaxation is observed [3, 4]. Furthermore, recent measurements on Co islands on Cu(001) also exhibit a magnetic hysteresis and remanence in the temperature range up to 150 K for coverages below the magnetic percolation threshold [19]. Due to the small size of the Co islands these findings could not be explained simply by single-particle blocking effects. Note that the experimental determination of the ordering temperature is difficult, since the relaxation times are often very long. Hence, it is of considerable interest to analyse how the magnetic order depends on the sample parameters which can be controlled in the experiment.

The purpose of this paper is the theoretical study of the low-energy properties of dipole-coupled magnetic particle ensembles. Inhomogeneous planar particle arrangements with various degrees of disorder ranging from a quasi-periodic square lattice to a random array are taken into account. The low symmetry of the system and the complicated nature of the interaction seem to preclude simple analytical approaches. Thus, numerical simulations are performed in order to
achieve a detailed microscopic description within the model. We focus on the strongly interacting case, neglecting single-particle anisotropies. Special attention is paid to the role of disorder and nonuniform arrangements of the particle magnetic moments. In particular, we determine the energy distribution of metastable states and the average magnetic dipole energy for different coverages and types of disorder. Furthermore, we investigate global and local order parameters as a function of disorder. Finally, angular histograms of nonrelaxed microvortex states for a square lattice with vacancies are determined. All calculations are performed at $T = 0$.

The rest of the paper is organized as follows. The theoretical methods are outlined in section 2. Representative results for the magnetic arrangements, as well as for various magnetic properties, are presented in section 3. A conclusion is given in section 4. In the appendix we report details for the extension of the dipole summation beyond the point-dipole approximation which takes into account effects resulting from the finite particle size.

2. Theory

We consider a 2D rectangular unit cell in the $xy$ plane with $n = n_x \times n_y$ non-overlapping, disc-shaped magnetic particles. Due to the strong direct exchange interaction and the small size of the particles under consideration, each particle $i$ can be viewed as a single magnetic domain (Stoner–Wohlfarth particle) [20]. Thus, a particle containing $N_i$ atoms carries a giant spin $M_i = N_i \mu_{at}$, where $\mu_{at}$ is the atomic magnetic moment. For simplicity, we restrict the particle magnetizations to be confined to the $xy$ plane. The planar rotator $M_i$ is then characterized by the in-plane angle $\phi_i$: $M_i = (M_i^x, M_i^y, M_i^z) = M_i(\cos \phi_i, \sin \phi_i, 0)$. In this study no size dispersion is considered, i.e. $N_i = N$. For disc-shaped particles the particle radius $r_0$ is given by $r_0 / a_0 = \sqrt{N / 2}$, where $a_0$ is the interatomic distance. Unless otherwise stated, the present results refer to a unit cell containing $n = 100$ particles with $N = 1000$ atoms each. The size of the unit cell is given by $L_x \times L_y = (n_x R_0) \times (n_y R_0)$, with $R_0$ the average interparticle distance. For a planar array of circular particles the overall surface coverage is $C = \pi (r_0 / R_0)^2$. Four different types of lateral particle arrangement are considered:

(i) a periodic square array, i.e. the particle centres are located on the sites of a square lattice with lattice constant $R_0$,

(ii) a disturbed (quasi-periodic) array for which the particle centres deviate randomly from the square array, using a Gaussian distribution $P(R)$ with positional standard deviation $\sigma_R$,

(iii) a diluted square particle lattice containing a number of vacancies with concentration $C_{\text{vac}}$, and

(iv) a fully random distribution of non-overlapping particles within the unit cell.

Periodic boundary conditions are introduced in order to describe an infinitely extended planar particle ensemble. In some cases a single finite unit cell is realized as well. Note that for finite cell calculations the truncation of long-range interactions could affect the resulting magnetic arrangement.

For such particle ensembles we consider the dipole–dipole interaction between magnetic moments $M_i$, which can also be expressed in terms of a dipole field $B_i^{\text{dip}}$ due to all other particle magnetic moments acting on $M_i$:

$$E_{\text{dip}} = \frac{\mu_0}{2} \sum_{i \neq j} \left[ M_i M_j r_{ij}^{-3} - 3 (r_{ij} M_i)(r_{ij} M_j) r_{ij}^{-5} \right] = -\frac{\mu_0}{2} \sum_i M_i B_i^{\text{dip}}, \quad (1)$$
where $r_{ij} = |r_i - r_j|$ is the distance between centres of particles $i$ and $j$ and $\mu_0$ is the vacuum permeability. 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 thin film [21]. In addition to the usual point-dipole sum we consider the leading correction resulting from the finite particle size (dipole–quadrupole interaction), which is outlined in detail in the appendix. This correction becomes comparable to the point-dipole sum for large particle coverages or small interparticle distances. The energy unit of the dipole coupling is given by $E_{\text{dip}}^0 = \mu_0^2/\alpha_0^3$. In this study we assume values appropriate to Fe, i.e. $\mu = 2.2 \mu_B$ and $\alpha_0 = 2.5 \text{ Å}$, yielding $E_{\text{dip}}^0 = 0.19 \text{ K}$. The effects of single-particle anisotropies resulting from the spin–orbit interaction, the dipole interaction among atomic magnetic moments within each particle (shape anisotropy), external magnetic fields, and finite temperatures are beyond the scope of the present study.

Starting from an arbitrary initial configuration $\{\phi_i^{\text{initial}}\}$ of the magnetic directions, the total magnetic energy $E_{\text{dip}}$ of the system is relaxed to the nearest local minimum by varying all angles $\phi_i$ of the particles using a conjugated gradient method [22]. For example, the experimental situation of a remanent state after removal of an external magnetic field is simulated by choosing a fully aligned initial arrangement along a certain direction. We emphasize that the applied procedure is not intended to search preferentially for the global energy minimum or the ground state, which is the equilibrium state at $T = 0$. Rather, at first we determine the energy distribution of metastable states for different degrees of structural disorder introduced in the planar particle array. A large number of arbitrary initial configurations is created, which can be a FM, a MV or a randomly chosen set of angles $\{\phi_i^{\text{initial}}\}$, and which are relaxed to a nearby local minimum. Each state is characterized by its energy $E_{\text{dip}}$ and its set of angles $\{\phi_i\}$. A twofold (uniaxial) symmetry is always present due to time inversion symmetry, i.e. the energy of a state does not change under the transformation $\phi_i \to \phi_i + 180^\circ$ performed simultaneously for all particles. These mirrored states are considered to be equivalent. The numbers of initial configurations yielding relaxed states with energies falling into given energy intervals are monitored, and the corresponding energy histogram is determined. Since the minimization procedure relaxes typically to the local energy minimum that is closest to the initial state, this frequency provides a measure of the catchment area, i.e. the area of the ‘energy valley’ belonging to that state in the high-dimensional configuration space.

In order to account for the large number of local energy minima occurring in particle arrangements with structural inhomogeneities we determine the average dipole energy $\bar{E}_{\text{dip}}$ of the relaxed states resulting from many initial configurations for the same particle array. For comparison, $\bar{E}_{\text{dip}}$ is also calculated for the same spatial set-up by assuming a FM state (i.e. $\phi_i = \phi_{\text{fm}}$) and by averaging over the FM angle $\phi_{\text{fm}}$. In the case of a disturbed (quasi-periodic) square lattice, i.e. if the particle numbering allows for the identification of a square plaquette of four neighbouring particles, we also determine the dipole energy for the MV magnetic arrangement, averaging over the MV angle $\phi_{\text{mv}}$. Note that for a nonuniform particle ensemble these FM and MV magnetic arrangements do not correspond in general to local energy minima.

In addition, a second averaging procedure for $\bar{E}_{\text{dip}}$ is physically necessary, namely the average over a number of different realizations of the unit cell in order to minimize spurious effects of the finite size of the unit cell. This is done by using the same global variables which characterize the particle ensemble, such as coverage $C$, standard deviation $\sigma_R$, etc. For a random set of angles $\{\phi_i\}$ one obtains $\bar{E}_{\text{dip}} = 0$ which constitutes the natural energy reference.
The deviations of the metastable low-energy magnetic arrangements from the MV state are quantified by the global and local MV order parameters $M_{\text{global}}(\sigma_R)$ and $M_{\text{local}}(\sigma_R)$ given by

$$M_{\text{global}}(\sigma_R) = \frac{1}{n} \left[ \left( \sum_{i} (-1)^{i_y} \cos \phi_i \right)^2 + \left( \sum_{i} (-1)^{i_y} \sin \phi_i \right)^2 \right]^{1/2},$$

and

$$M_{\text{local}}(\sigma_R) = \frac{1}{4} \left[ \left( \sum_{i} (-1)^{i_y} \cos \phi_i \right)^2 + \left( \sum_{i} (-1)^{i_y} \sin \phi_i \right)^2 \right]^{1/2},$$

where $i_x$ and $i_y$ denote the numbering of rows and columns corresponding to particle $i$. Clearly, a reference square lattice is a prerequisite. Hence, we restrict ourselves to a quasi-periodic particle array or to a square array with vacancies for which $i_x$ and $i_y$ can be uniquely defined. These two order parameters differ by the sum $i$ running either over all $n$ particles of the unit cell or over the four neighbouring particles within a square plaquette. The definitions given in equations (2) and (3) have a simple geometrical interpretation as the projection of the magnetic configuration on the two linear independent columnar states having $\phi_{\text{mv}} = 0^\circ$ and $90^\circ$, regardless of all possible rotations of the MV states within this plane. $M_{\text{global}}(\sigma_R)$ measures the MV order in the nanostructure, whereas $M_{\text{local}}(\sigma_R)$ refers to the short-range ordering. Both quantities are averaged over an appropriate number of initial configurations for the same particle arrangement, and over different realizations of the unit cell. In addition, $M_{\text{local}}(\sigma_R)$ is averaged over all four-particle plaquettes within the unit cell.

Finally, another quantity characterizing the magnetic properties of nanostructured particle arrangements is the distribution of the optimal MV angle $\phi_{\text{mv}}$, i.e. the lowest-energy MV state with respect to $\phi_{\text{mv}}$. Prakash and Henley [10] reported that the preferred MV angles for a square particle array with a small number of randomly distributed vacancies are the diagonal directions ($\phi_{\text{mv}} = 45^\circ$, $135^\circ$, etc). Note that in [10] only the dipole coupling between nearest neighbours is taken into account, and that some degree of relaxation of the magnetic arrangement is allowed.

In the present study we like to investigate how the angular distribution of the preferred MV angle depends on the type and degree of disorder. For each particle set-up the lowest-energy MV state is determined. By creating different realizations of the nanostructure the corresponding frequencies for the preferred $\phi_{\text{mv}}$ are monitored. We consider a disturbed square particle array with standard deviation $\sigma_R$ and a diluted square particle lattice with a concentration $C_{\text{vac}}$ of vacancies. It should be noted that for a small number of vacancies one obtains an appreciable dependence on the shape of the unit cell, i.e. the aspect ratio of the rectangle, since these vacancies are located effectively on a square or rectangular lattice due to the periodic boundary conditions. To avoid this problem we consider also a number of vacancies distributed randomly in a single circular unit cell. As before, the vacancies are introduced into a nonrelaxed MV magnetic arrangement. The corresponding angular distribution of the preferred MV angle is monitored by considering a large number of randomly chosen vacancy set-ups with the same $C_{\text{vac}}$.

3. Results

First, figure 1 illustrates some representative low-energy magnetic arrangements of disordered particle arrays. With increasing disorder, characterized by the positional standard deviation $\sigma_R$, the magnetic configurations of the relaxed solutions become increasingly complex. Examples are given for the magnetic states of a slightly disturbed ($\sigma_R/R_0 = 0.05$), a moderately disturbed ($\sigma_R/R_0 = 0.10$) and a strongly disturbed ($\sigma_R/R_0 = 0.50$) particle set-up having all the same...
coverage $C = 35\%$. In the left column we consider optimal MV states having the lowest energy with respect to $\phi_{\text{inv}}$. In the right column examples of fully relaxed states are shown. For $\sigma_R/R_0 = 0.05$ the MV arrangement resembles quite closely the true solution, see figure 1(a). However, as can be seen in figure 1(b), already a moderate amount of positional disorder destroys the MV state. This is physically reasonable since the MV order is tightly connected to the presence of a square-lattice symmetry of the particle ensemble. The degree of the MV ordering will be quantified below. For large $\sigma_R$ or for a random particle set-up the resulting magnetic arrangement is dominated by the formation of chains and loops of magnetic moments with a correlated 'head-to-tail' alignment of the particle magnetizations, see figure 1(c) [23]. This reflects the tendency of the dipole interaction to favour a locally demagnetized state with a vanishing or small net magnetization.

In figure 2 we present examples for the dipole energy distribution of metastable states of slightly and moderately disturbed particle set-ups. These were obtained by considering 20 000 random initial configurations $\{\phi_i^{\text{initial}}\}$ for the same realization of the unit cell, and by assigning the energies of the relaxed states to energy intervals $[E, E + \Delta E]$, with $\Delta E = 5 \times 10^{-3}$ K. Distinctly different energy distributions are observed for weak and strong disorder. For weak disorder with $\sigma_R/R_0 = 0.05$, see figure 2(a), an asymmetric energy distribution is found. The lowest-energy state is reached very often: in fact, for $\sigma_R/R_0 = 0.05$ about 30% of the initial configurations relax to that ground state, and about 50% for $\sigma_R/R_0 = 0.03$. Moreover, the ground-state configuration resembles closely the MV state with an almost vanishing net magnetization, see figure 1(a). In addition, a number of metastable states is obtained, which energies are distributed over a relatively broad range, and which are reached far less frequently. In other words, for weak disturbances the catchment area of the ground state is much larger than the ones of the higher-energy states. It is interesting to note that the energy density of metastable states is much smaller for the bottom than for the centre of the distribution. Furthermore, the higher-energy states often exhibit a finite net magnetization. Thus one could expect that, in the presence of an external magnetic field, these states become more favourable than the ones with a vanishing net magnetization.

Already for moderate disorder with $\sigma_R/R_0 = 0.10$ the character of the energy distribution changes strongly. An almost symmetric, Gaussian-like energy distribution is obtained around the average dipole energy $\bar{E}_{\text{dip}}$, see figure 2(b). The number of metastable states has increased markedly. In fact, considering 20 000 random initial configurations, no single state is reached twice after relaxation. This is particularly true for the low-energy states which are obtained with a very small frequency, see the circled region in figure 2(b). For an even stronger disorder this small peak in the frequency for the low-energy states disappears completely. Since the total number of metastable states increases strongly with increasing disorder, the corresponding catchment areas decrease dramatically. The large number of metastable obtained states is consistent with experiments on ferrofluids, which observe that, after the application and removal of an external magnetic field, the same magnetic arrangement is seldomly reached for a second time [24].

Furthermore, from figure 2 one observes that the average magnetic dipole energy $\bar{E}_{\text{dip}}$ decreases with increasing positional standard deviation $\sigma_R$. In figure 3 we present results for $\bar{E}_{\text{dip}}(\sigma_R, C)$ per particle as a function of $\sigma_R$ and for different particle coverages $C$. A square lattice corresponds to $\sigma_R = 0$. Results for random particle set-ups are also shown. $\bar{E}_{\text{dip}}(\sigma_R)$ is calculated for the relaxed solutions averaged over a number of initial spin configurations, as well as for the FM and MV magnetic states averaged over the corresponding angles $\phi_{\text{fm}}$ and $\phi_{\text{mv}}$. The minimum values of $\bar{E}_{\text{dip}}(\sigma_R)$ obtained are also displayed in order to illustrate the spread of the
Figure 1. Illustrations of the magnetic arrangements in 2D nanostructures. The particle positions are scattered around the lattice sites of a square array with positional standard deviations (a) $\sigma_R/R_0 = 0.05$, (b) $\sigma_R/R_0 = 0.10$ and (c) $\sigma_R/R_0 = 0.50$, where $R_0$ denotes the average interparticle distance. The particle coverage amounts to $C = 35\%$. The left columns refer to the lowest-energy MV state with respect to the MV angle. The right columns show examples of relaxed magnetic states, which were generated in this case by using the MV arrangements shown in the left columns as starting configurations.
Figure 2. Energy distribution of local energy minima for two disturbed square particle arrays using different positional standard deviations (a) $\sigma_R/R_0 = 0.05$ and (b) $\sigma_R/R_0 = 0.10$. The particle coverage amounts to $C = 35\%$. The distributions of the metastable states are obtained from 20 000 randomly chosen initial configurations and sampled into energy intervals with width $\Delta E = 5 \times 10^{-3}$ K. Also indicated is the resulting average dipole energy $\bar{E}_{\text{dip}}(\sigma_R)$.

In figure 3(a), $\bar{E}_{\text{dip}}(\sigma_R)$ is shown for a particle coverage $C = 35\%$. With increasing disorder $\sigma_R$ the average dipole energy decreases [25]. For $\sigma_R/R_0 \gtrsim 0.5$, $\bar{E}_{\text{dip}}(\sigma_R)$ approaches a constant value which corresponds, within the numerical dispersion of the data, to the average dipole energy for a random particle array. The decrease of the average energy is caused by the
Figure 3. (a) Average dipole energy $\overline{E}_{\text{dip}}(\sigma_R)$ per particle of a planar array of magnetic particles as a function of the positional standard deviation $\sigma_R/R_0$. $R_0$ refers to the average interparticle distance and the particle coverage is $C = 35\%$. Displayed are $\overline{E}_{\text{dip}}(\sigma_R)$ for the relaxed magnetic states, averaged over 40 different initial conditions (full curves, circles), and the minimum energies (broken curves, squares) which indicate the dispersion of the data. Results are also given for the FM and MV magnetic arrangements averaged over the corresponding angles. The dotted lines denote $\overline{E}_{\text{dip}}$ for a random particle set-up with the same particle sizes and coverages. All results correspond to the average over 20 different realizations of the unit cell. (b) $\overline{E}_{\text{dip}}(\sigma_R, C)$ of relaxed magnetic arrangements as a function of $\sigma_R$ for different particle coverages $C$ as indicated. The dotted lines denote $\overline{E}_{\text{dip}}(C)$ for random particle set-ups. (c) $\overline{E}_{\text{dip}}(\sigma_R, C)$ of relaxed magnetic arrangements as a function of the particle coverage $C$ for different $\sigma_R$ as indicated. We show $\overline{E}_{\text{dip}}(C)$ as a function of $C^{-3/2}$ which should yield a linear behaviour from a simple scaling estimate.

nonlinear dependence of the dipole interaction with respect to the interparticle distance [25]. In fact, once disorder is introduced, the increase of $\overline{E}_{\text{dip}}$ for enlarged distances between some particle pairs is more than counterbalanced by a corresponding decrease for smaller distances between other pairs of particles. A similar behaviour is obtained for the FM arrangement, albeit with a larger $\overline{E}_{\text{dip}}(\sigma_R)$. In contrast, for the MV state the average energy exhibits a minimum as
a function of $\sigma_R$ at $\sigma_R/R_0 \sim 0.15$, and approaches $\bar{E}_{\text{dip}} = 0$ with increasing $\sigma_R$. Relaxation in a disordered particle arrangement is thus crucial to the disorder-induced reduction of $\bar{E}_{\text{dip}}(\sigma_R)$. Let us recall that, for $\sigma_R > 0$, the FM and MV arrangements usually do not correspond to local energy minima.

In figure 3(b) we show $\bar{E}_{\text{dip}}(\sigma_R)$ of the relaxed solutions and for different particle coverages $C$. First of all one observes that the overall dependence of $\bar{E}_{\text{dip}}$ on $\sigma_R$ is not significantly affected by $C$. Increasing the mean interparticle spacing $R_0$ decreases the magnitude of the average dipole energy, which should scale as $\bar{E}_{\text{dip}} \propto R_0^{-3} \propto C^{3/2}$. The dependence of $\bar{E}_{\text{dip}}$ on $C$ is depicted in figure 3(c) for the relaxed solutions. Indeed, the expected behaviour $\bar{E}_{\text{dip}} \propto C^{3/2}$ is obtained for weak positional disorder $\sigma_R$. However, for strong disorder and for small coverages a different concentration dependence is observed. It seems that the strong magnetic noncollinearities of the magnetic configurations render the simple scaling expectation no longer applicable. It would therefore be interesting to investigate in detail the energy scaling as a function of coverage $C$, especially in the limit of strongly disturbed arrays of magnetic particles.

Note that the magnitude of the dipole energy is comparably small. This is a consequence of the disc-shaped particles assumed in our calculations. For spherical-shaped particles with the same radius $r_0$ as for disc-shaped ones, thus yielding the same particle coverage $C$, the corresponding dipole energy will be significantly enhanced by the factor 550 due to the larger number of atoms $N = (r_0/a_0)^3$ per spherical particle. Moreover, similar to the positional disorder, a particle-size dispersion yields nonuniform magnetic arrangements even if the particle centres form a periodic lattice. However, one observes that $\bar{E}_{\text{dip}}$ does not vary strongly with increasing size dispersion [26].

Furthermore, we discuss the crossover from a quasi-periodic to a random particle ensemble. In figure 4 we show the coverage dependence of the positional standard deviation $\bar{\sigma}_R$, above which the average energy $\bar{E}_{\text{dip}}(\sigma_R)$ of a quasi-periodic particle arrangement reaches the limiting value of a random set-up. One observes that $\bar{\sigma}_R$ decreases with increasing coverage $C$, as can qualitatively be explained by the following simple scaling consideration. The particles are scattered within a certain distance around the sites of the square lattice and the average scattering radius $R_{\sigma_R}$ is estimated by $R_{\sigma_R} \simeq \sigma_R R_0$. For a given coverage $C$ the average dipole energy for a random particle set-up is approximately reached for the crossover standard deviation $\bar{\sigma}_R$ for which $R_{\sigma_R}$ plus the particle radius equals half of the average interparticle distance:

$$R_{\sigma_R} + r_0 \simeq R_0/2.$$  (4)

Using $C = \pi (r_0/R_0)^2$, one obtains

$$\bar{\sigma}_R(C) \simeq \frac{1}{2} - \sqrt{\frac{C}{\pi}}.$$  (5)

In figure 4, $\bar{\sigma}_R(C)$ is compared with the inflection points of $\bar{E}_{\text{dip}}(\sigma_R)$ extracted from figure 3(b) for various coverages $C$. One observes that, except for the largest considered coverage $C = 35\%$, a satisfactory agreement is obtained, which supports the validity of the previous scaling considerations.

Results for the averages of global and local MV order parameters $M_{\text{mv}}^{\text{global}}(\sigma_R)$ and $M_{\text{mv}}^{\text{local}}(\sigma_R)$ are presented in figure 5, assuming two different types of disorder. In figure 5(a) we consider a disturbed square particle lattice as a function of the positional standard deviation $\sigma_R$ for a particle coverage $C = 35\%$. In figure 5(b) the concentration of vacancies $C_{\text{vac}}$ in an otherwise periodic array is varied. The rather large dispersions of the MV order parameters result from many

New Journal of Physics 5 (2003) 68.1–68.22 (http://www.njp.org/)
different realizations of the unit cell. Locally the MV order is preserved even for strong disorder, regardless of its type. For example, for $\sigma_R/R_0 = 0.5$, which refers to an almost random particle array, we obtain nevertheless $M_{\text{mv}}^{\text{local}} \sim 0.5$. Also, for a diluted particle lattice with $C_{\text{vac}} = 20\%$ we yield $M_{\text{mv}}^{\text{local}} \sim 0.7$. In contrast, the global MV order parameter $M_{\text{mv}}^{\text{global}}$ is affected more strongly by positional disturbances than by vacancies, since the former type of disorder quickly destroys the long-range MV ordering in the square lattice. For instance, for a particle coverage $C = 35\%$ this occurs already for $\sigma_R/R_0 \gtrsim 0.10$, which is consistent with the minimum of $\bar{E}_{\text{dip}}(\sigma_R)$ of the MV state, see figure 3(a).

Finally, we present results for the angular distribution of the optimal MV angle $\phi_{\text{mv}}$ for different degrees of disorder. Note that for this investigation only nonrelaxed MV magnetic arrangements are taken into account, i.e. $\phi_{\text{mv}}$ is varied while keeping the perfect MV correlations between all spins, even for those spins close to defects. We study first the effect of vacancies in an otherwise periodic square particle lattice, as previously considered by Prakash and Henley [10]. The angular distributions determined for a unit cell with periodic boundary conditions are shown in figure 6, assuming different numbers of vacancies in the unit cell. For vacancy concentrations $C_{\text{vac}} = 2.0$ and 3.9% a strong frequency of both the diagonal states with MV angles $\phi_{\text{mv}} = 45^\circ$ and $135^\circ$, as well as the columnar states with $\phi_{\text{mv}} = 0^\circ$, $90^\circ$, etc, is obtained. For $C_{\text{vac}} \gtrsim 2.0\%$ the frequencies of the diagonal states decrease with increasing vacancy concentration and vanish for $C_{\text{vac}} \gtrsim 6–7\%$, see figure 6(c). The columnar states are present for all considered $C_{\text{vac}}$. Note that in [10] the preferred MV angles correspond to diagonal directions, which were obtained by allowing some degree of relaxation.

We have investigated whether the angular distributions of the optimal MV angle, in particular the pronounced frequencies of the columnar states, are affected by the size and orientation of the
Figure 5. Averages of the global and local MV order parameters $M_{\text{global}}(\sigma_R)$ and $M_{\text{local}}(\sigma_R)$ as a function of the distortion of the particle array assuming (a) a disturbed square lattice with positional standard deviation $\sigma_R$ and particle coverage $C = 35\%$, and (b) a square array containing randomly distributed vacancies with concentration $C_{\text{vac}}$. These order parameters are averaged over many different initial configurations and different realizations of the unit cell. In addition $M_{\text{local}}(\sigma_R)$ is averaged over all four-particle plaquettes within the unit cell. The calculated data points are connected by straight lines. The resulting standard deviation is indicated by error bars.

unit cell. In fact, calculations varying the aspect ratio of the rectangular unit cell, as well as by rotating the unit cell with respect to the axes of the particle lattice, show no significant change in angular distribution. This problem has been further studied by considering a number of vacancies distributed randomly in a single circular unit cell, i.e. without periodic boundary conditions. Such an approach allows the consideration of a much wider range of vacancy concentrations, and is free from spurious effects resulting from periodically arranged vacancies in different unit cells.
Figure 6. Angular distribution of the preferred MV angle $\phi_{mv}$ for different vacancy concentrations $C_{\text{vac}}$ on a square lattice. (a) $C_{\text{vac}} = 2.0\%$ (5 vacancies per unit cell), (b) $C_{\text{vac}} = 3.9\%$ (10 vacancies) and (c) $C_{\text{vac}} = 5.9\%$ (15 vacancies). The size of the unit cell is $16 \times 16$ particles and 1000 different realizations of the unit cell are performed. Here only nonrelaxed MV states are considered. Due to the mirror symmetry we show the angular distribution for $-20^\circ \leq \phi_{mv} \leq 200^\circ$.

First of all, one can show by simple calculations that for a single isolated vacancy the energy of the MV state is independent of $\phi_{mv}$. Moreover, for two vacancies half of the possible set-ups of this vacancy pair yields the lowest energy for the columnar state ($\phi_{mv} = 0^\circ, 90^\circ$). For the other half of the set-ups all $\phi_{mv}$ are equally probable, since the optimal $\phi_{mv}$ is determined by the position vector connecting the randomly chosen vacancy pair. For more than two vacancies
one has to perform numerical simulations. By considering 25 vacancies we found different types of MV angular distributions as a function of the vacancy concentration. For a very small $C_{\text{vac}}$ only the columnar states exhibit a pronounced frequency. With increasing $C_{\text{vac}}$, typically for $C_{\text{vac}} \gtrsim 10^{-3}\%$, peaks in the angular distribution appear also for $\phi_{\text{mv}} = 45^\circ$ and $135^\circ$, corresponding to diagonal states. However, by a further increase of $C_{\text{vac}}$ the frequency peaks near these states become wider, and finally disappear for a large $C_{\text{vac}} \sim 10\%$. Therefore, a pronounced frequency of diagonal MV states is present only for intermediate $C_{\text{vac}}$ and seems to be related to the interactions between vacancies. For the investigated concentration range $0 < C_{\text{vac}} < 10\%$ the frequency peaks of the columnar states become reduced with increasing $C_{\text{vac}}$, but are always pronounced. Thus, the results of the angular distributions of the preferred MV angle for a single unit cell confirms the conclusion derived for periodically arranged unit cells shown in figure 6. Note that for periodic boundary conditions the finite size of the unit cell precludes the consideration of very small $C_{\text{vac}}$.

Moreover, a disturbed square lattice characterized by the positional standard deviation $\sigma_R$ is considered. Assuming nonrelaxed MV arrangements, we observe no particular preference of the diagonal states. For this type of disorder only the columnar states exhibit a pronounced frequency in the angular distribution of the optimal MV angle. The frequency peaks for these states decrease for increasing disorder and vanish above $\sigma_R / R_0 \sim 0.20$ for the particle coverage $C = 35\%$. A marked dependence of the angular distribution on the size and shape of the unit cell is also not observed for this type of disorder. To conclude this discussion, we would like to emphasize that the angular distributions presented in figure 6 were all obtained for a nonrelaxed MV state. We have also performed corresponding calculations for the relaxed solutions, and have determined the MV states which are closest to the relaxed magnetic arrangements. In contrast to the results shown in figure 6, the corresponding angular histograms obtained by this procedure exhibit a strong dependence on the size and shape of the unit cell. Therefore, further investigations would be necessary in order to clarify this matter.

4. Conclusion

In the present study we have investigated the low-energy properties of disordered planar arrays of magnetic nanoparticles interacting by dipole coupling. Different kinds of disorder have been considered. Already small deviations from a square particle lattice lift the continuous degeneracy of the MV ground state. A strongly nonuniform magnetic order appears in the nanostructure which destroys the MV arrangement, as can be seen from the corresponding order parameter. Positional disorder has a stronger effect on the MV order than the presence of vacancies. We have shown that, with an increasing disorder, the energy distribution of metastable states changes qualitatively. Our results indicate that it should be quite difficult to identify the ground-state energy and its magnetic configuration for strongly inhomogeneous particle arrangements. This behaviour is typical for spin glass systems or random magnets. The energy distribution of states deserves to be further investigated by considering different types of disorder and the effect of finite temperatures.

We have shown that the average magnetic dipole energy $\bar{E}_{\text{dip}}$ of an ensemble of magnetic particles decreases with increasing positional disorder. Moreover, $\bar{E}_{\text{dip}}$ of a quasi-periodic particle arrangement resembles that of a random particle set-up above the coverage-dependent crossover standard deviation $\tilde{\sigma}_R(C)$. A simple scaling behaviour of $\tilde{\sigma}_R(C)$ has been derived, which reproduces quite accurately the coverage dependence of $\tilde{\sigma}_R(C)$. Structural disorder and
strong magnetic noncollinearity effects result in a deviation from the straightforward scaling \( E_{\text{dip}} \propto C^{3/2} \) of the average dipole energy as a function of the coverage.

For nonrelaxed MV states in a square particle lattice with a number of vacancies we found pronounced frequencies of the preferred MV angle \( \phi_{\text{mv}} \) referring to columnar states in the whole investigated vacancy concentration range. On the other hand, the diagonal states exhibit a strong frequency only for intermediate \( C_{\text{vac}} \), which vanish for \( C_{\text{vac}} \to 0 \) and for large \( C_{\text{vac}} > 6\text{--}7\% \). These results have been obtained for unit cells with free and with periodic boundary conditions. In contrast to the case of vacancy-induced disorder, for a disturbed square particle array characterized by the positional standard deviation \( \sigma_R \) a pronounced frequency is found only for the columnar, but not for the diagonal, states.

The determination of \( E_{\text{dip}} \) allows us to distinguish whether a magnetic particle ensemble is considered as a weakly or a strongly interacting system. In the first case the interactions can be treated as perturbations to the single-particle couplings, whereas in the latter case they have to be considered explicitly. The present study can be easily extended in order to take into account single-particle anisotropies with distributions of their magnitudes and easy axes. The importance of collectively ordered magnetic states for strongly interacting particle systems has been pointed out. An increasing average binding energy |\( E_{\text{dip}} \)| favours magnetic ordering and should cause an increase of its critical spin glass temperature. A detailed investigation of the spin glass behaviour of the nanostructure is certainly worthwhile. This would require the consideration of additional properties such as the nonlinear susceptibility, the magnetic relaxation and time-dependent correlation functions [3, 18]. Finite temperature effects can be introduced in the framework of a mean field approximation or by performing Monte Carlo simulations. Magnetic hysteresis loops and susceptibilities can be derived by applying magnetic fields with different directions and strengths.

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Appendix. Dipole interaction between magnetic particles with a finite size

In this appendix we determine the leading correction to the magnetic dipole interaction, see equation (1), beyond the point-dipole sum. The finite extension of the particles for the most general case of arbitrary sizes and shapes is taken into account. For a hexagonal lattice the finite particle size has been considered already by Politi and Pini [12]. First, the interaction between a particle pair is calculated. Then a particle ensemble with an infinite lateral extension is modelled by means of a rectangular unit cell with periodic boundary conditions.

The dipole–quadrupole correction

Consider a particle \( i \) containing \( N_i \) atoms on lattice sites \( k \) with position vectors \( \mathbf{r}_{ik} = \mathbf{r}_{i0} + \mathbf{r}_k \). The centre of gravity of this particle is given by

\[
\mathbf{r}_{i0} = (x_{i0}, y_{i0}, z_{i0}) = \frac{1}{N_i} \sum_{k \in \ell} \mathbf{r}_{ik} = \frac{1}{N_i} \sum_{k \in \ell} (x_{ik}, y_{ik}, z_{ik}).
\] (A.1)
The finite size of particle $i$ is taken into account by the quadratic deviations

$$
\langle x_i^2 \rangle = \frac{1}{N_i} \sum_{k \in i} x_k^2 = \frac{1}{N_i} \sum_{k \in i} (x_{i0} - x_{ik})^2,
$$

(A.2)

and similarly for $\langle y_i^2 \rangle$ and $\langle z_i^2 \rangle$. Furthermore, we define $\varepsilon_{ik}$ by

$$
r_{ik}^2 = (x_{i0} + x_k)^2 + (y_{i0} + y_k)^2 + (z_{i0} + z_k)^2 = r_{i0}^2(1 + \varepsilon_{ik}),
$$

(A.3)

and hence

$$
\varepsilon_{ik} = \frac{1}{r_{i0}^2} (2x_{i0}x_k + 2y_{i0}y_k + 2z_{i0}z_k + x_k^2 + y_k^2 + z_k^2),
$$

(A.4)

with $r_{i0}^2 = x_{i0}^2 + y_{i0}^2 + z_{i0}^2$. The factor $(1 + \varepsilon_{ik})$ appearing in the denominator of the dipole interaction energy is expanded to second order in $x_k$, $y_k$, and $z_k$ as

$$
\frac{1}{(1 + \varepsilon_{ik})^{5/2}} \simeq 1 - \frac{5}{2} \varepsilon_{ik} + \frac{35}{8} \varepsilon_{ik}^2 \simeq 1 - \frac{5}{2r_{i0}^2} (2x_{i0}x_k + 2y_{i0}y_k + 2z_{i0}z_k + x_k^2 + y_k^2 + z_k^2) + \frac{35}{2r_{i0}^4} (x_{i0}x_k + y_{i0}y_k + z_{i0}z_k)^2.
$$

(A.5)

Now all different sums over the atomic sites $k$ of particle $i$ are performed up to this order. The non-vanishing terms are listed in the following equations (A.6)–(A.15):

$$
\sum_{k \in i} \frac{1}{r_{ik}^2} = \sum_{k \in i} \frac{1}{r_{i0}^2(1 + \varepsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{i0}^5} \left[ 1 + \frac{5}{2r_{i0}^2} \left( \frac{7y_{i0}^2}{r_{i0}^2} - 1 \right) \langle x_i^2 \rangle + \frac{5}{2r_{i0}^4} \left( \frac{7y_{i0}^2}{r_{i0}^2} - 1 \right) \langle y_i^2 \rangle \right],
$$

(A.6)

$$
\sum_{k \in i} \frac{x_{i0} + x_k}{r_{ik}^2} = \sum_{k \in i} \frac{x_{i0} + x_k}{r_{i0}^2(1 + \varepsilon_{ik})^{5/2}} \simeq \frac{N_i x_{i0}}{r_{i0}^5} \left[ 1 + \frac{5}{2r_{i0}^2} \left( \frac{7x_{i0}^2}{r_{i0}^2} - 3 \right) \langle x_i^2 \rangle + \frac{5}{2r_{i0}^4} \left( \frac{7x_{i0}^2}{r_{i0}^2} - 1 \right) \langle y_i^2 \rangle \right],
$$

(A.7)

$$
\sum_{k \in i} \frac{y_{i0} + y_k}{r_{ik}^2} = \sum_{k \in i} \frac{y_{i0} + y_k}{r_{i0}^2(1 + \varepsilon_{ik})^{5/2}} \simeq \frac{N_i y_{i0}}{r_{i0}^5} \left[ 1 + \frac{5}{2r_{i0}^2} \left( \frac{7y_{i0}^2}{r_{i0}^2} - 3 \right) \langle y_i^2 \rangle + \frac{5}{2r_{i0}^4} \left( \frac{7y_{i0}^2}{r_{i0}^2} - 1 \right) \langle z_i^2 \rangle \right],
$$

(A.8)

$$
\sum_{k \in i} \frac{z_{i0} + z_k}{r_{ik}^2} = \sum_{k \in i} \frac{z_{i0} + z_k}{r_{i0}^2(1 + \varepsilon_{ik})^{5/2}} \simeq \frac{N_i z_{i0}}{r_{i0}^5} \left[ 1 + \frac{5}{2r_{i0}^2} \left( \frac{7z_{i0}^2}{r_{i0}^2} - 3 \right) \langle z_i^2 \rangle + \frac{5}{2r_{i0}^4} \left( \frac{7z_{i0}^2}{r_{i0}^2} - 1 \right) \langle x_i^2 \rangle \right],
$$

(A.9)

$$
\sum_{k \in i} \frac{x_k^2}{r_{ik}^2} = \sum_{k \in i} \frac{x_k^2 + 2x_{i0}x_k + x_{i0}^2}{r_{i0}^2(1 + \varepsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{i0}^5} \left[ \langle x_i^2 \rangle + \frac{5}{2r_{i0}^2} \left( \frac{7x_{i0}^2}{r_{i0}^2} - 5 \right) \langle x_i^2 \rangle + \frac{5}{2r_{i0}^4} \left( \frac{7x_{i0}^2}{r_{i0}^2} - 1 \right) \langle y_i^2 \rangle \right] + \frac{5}{2r_{i0}^4} \left( \frac{7x_{i0}^2}{r_{i0}^2} - 1 \right) \langle z_i^2 \rangle,
$$

(A.10)
\[ \sum_{k \in i} \frac{y_{ik}^2}{r_{ik}^5} = \sum_{k \in i} \frac{y_{ik}^2 + 2y_{ik}y_j + y_{jik}^2}{r_{ik}^5(1 + \epsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{ik}^5}(y_{ik}^2 + \langle y_{ik} \rangle^2) + \frac{5N_i}{2r_{ik}^{7/2}} \left[ \left( 7\frac{x_{ik}^2}{r_{ik}^2} - 1 \right) \langle x_{ik}^2 \rangle + \left( 7\frac{y_{ik}^2}{r_{ik}^2} - 5 \right) \langle y_{ik}^2 \rangle + \left( 7\frac{z_{ik}^2}{r_{ik}^2} - 1 \right) \langle z_{ik}^2 \rangle \right], \]  

(A.11)

\[ \sum_{k \in i} \frac{z_{ik}^2}{r_{ik}^5} = \sum_{k \in i} \frac{z_{ik}^2 + 2z_{ik}z_j + z_{jik}^2}{r_{ik}^5(1 + \epsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{ik}^5}(z_{ik}^2 + \langle z_{ik} \rangle^2) + \frac{5N_i}{2r_{ik}^{7/2}} \left[ \left( 7\frac{x_{ik}^2}{r_{ik}^2} - 1 \right) \langle x_{ik}^2 \rangle + \left( 7\frac{y_{ik}^2}{r_{ik}^2} - 5 \right) \langle y_{ik}^2 \rangle + \left( 7\frac{z_{ik}^2}{r_{ik}^2} - 1 \right) \langle z_{ik}^2 \rangle \right], \]  

(A.12)

\[ \sum_{k \in i} \frac{x_{ik}y_{ik}}{r_{ik}^5} = \sum_{k \in i} \frac{x_{ik}y_{ik} + x_{ik}y_j + x_{ik}y_{jik}}{r_{ik}^5(1 + \epsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{ik}^5}(x_{ik}y_{ik} + \langle x_{ik} \rangle \langle y_{ik} \rangle) + \frac{5}{2r_{ik}^{5/2}} \left( 7\frac{x_{ik}^2}{r_{ik}^2} - 3 \right) \langle x_{ik}^2 \rangle + \frac{5}{2r_{ik}^{5/2}} \left( 7\frac{y_{ik}^2}{r_{ik}^2} - 1 \right) \langle z_{ik}^2 \rangle \]  

(A.13)

\[ \sum_{k \in i} \frac{x_{ik}z_{ik}}{r_{ik}^5} = \sum_{k \in i} \frac{x_{ik}z_{ik} + x_{ik}z_j + x_{ik}z_{jik}}{r_{ik}^5(1 + \epsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{ik}^5}(x_{ik}z_{ik} + \langle x_{ik} \rangle \langle z_{ik} \rangle) + \frac{5}{2r_{ik}^{5/2}} \left( 7\frac{x_{ik}^2}{r_{ik}^2} - 3 \right) \langle x_{ik}^2 \rangle + \frac{5}{2r_{ik}^{5/2}} \left( 7\frac{y_{ik}^2}{r_{ik}^2} - 1 \right) \langle z_{ik}^2 \rangle \]  

(A.14)

\[ \sum_{k \in i} \frac{y_{ik}z_{ik}}{r_{ik}^5} = \sum_{k \in i} \frac{y_{ik}z_{ik} + y_{ik}z_j + y_{ik}z_{jik}}{r_{ik}^5(1 + \epsilon_{ik})^{5/2}} \simeq \frac{N_i}{r_{ik}^5}(y_{ik}z_{ik} + \langle y_{ik} \rangle \langle z_{ik} \rangle) + \frac{5}{2r_{ik}^{5/2}} \left( 7\frac{x_{ik}^2}{r_{ik}^2} - 3 \right) \langle x_{ik}^2 \rangle + \frac{5}{2r_{ik}^{5/2}} \left( 7\frac{y_{ik}^2}{r_{ik}^2} - 1 \right) \langle z_{ik}^2 \rangle \]  

(A.15)

A simple extension of the previous considerations allows us to calculate to the same order the corresponding sums involved in the interaction between two extended particles \( i \) and \( j \) with sizes \( N_i \) and \( N_j \). Let \( r_{i,j} = (x_{i,j0}, y_{i,j0}, z_{i,j0}) \) denote the position vector between the particle centres. The non-vanishing sums in the interaction energy are given by equations (A.6)–(A.15) after performing the replacements \( N_j \rightarrow N_i/N_j, r_{j0} \rightarrow r_{i,j0}, x_j \rightarrow x_{i,j0}, \langle x_j^2 \rangle \rightarrow \langle x_{i,j0}^2 \rangle + \langle x_j^2 \rangle, \) etc. Taking into account the summation over all atoms \( k' \) of particle \( j \), one obtains, for example, for equation (A.6),

\[ \sum_{k \in i} \sum_{k' \in j} \frac{1}{|r_{ik} - r_{jk'}|^3} \simeq \frac{N_iN_j}{r_{ik}^5} \left[ 1 + \frac{5}{2r_{i,j0}^{3/2}} \left( 7\frac{x_{i,j0}^2}{r_{i,j0}^2} - 1 \right) (\langle x_{i,j0}^2 \rangle + \langle x_j^2 \rangle) \right. \]  

\[ + \frac{5}{2r_{i,j0}^{3/2}} \left( 7\frac{y_{i,j0}^2}{r_{i,j0}^2} - 1 \right) (\langle y_{i,j0}^2 \rangle + \langle y_j^2 \rangle) \]  

\[ + \frac{5}{2r_{i,j0}^{3/2}} \left( 7\frac{z_{i,j0}^2}{r_{i,j0}^2} - 1 \right) (\langle z_{i,j0}^2 \rangle + \langle z_j^2 \rangle) \right]. \]  

(A.16)

Thus, within this expansion the dipole interaction between particles \( i \) and \( j \) is expressed in terms of the distance \( r_{i,j0} \) between their centres and the quadratic deviations \( \langle x_j^2 \rangle, \langle x_j^2 \rangle, \) etc., characterizing the sizes and shapes of the two particles. The point-dipole sum is recovered by setting \( \langle x_j^2 \rangle = 0, \) etc. The correction to the point-dipole sum is a dipole–quadrupole interaction, being of the order \( (\langle x_j^2 \rangle + \langle x_j^2 \rangle)/r_{i,j0}^2 \), i.e. the ratio of the particle extensions and the square of the interparticle distance. Evidently, the effect of finite particle sizes becomes more important the closer the particles are located. This is the case, for example, for a densely packed 3D ferrofluid or for a layered nanostructured particle ensemble with a large surface coverage.
The extended planar system

Let us now consider a planar particle ensemble infinitely extended in the \(xy\) plane. The unit cell has the size \(L_x \times L_y\) and consists of \(n\) particles. Periodic boundary conditions are applied laterally, whereas the vertical extension along the \(z\) direction is finite. The dipole interaction sum runs over all particle pairs within the same and between different unit cells. The position vectors connecting two particle centres are given by \(r_{ij0} = (x_{i0} + l_x L_x, y_{i0} + l_y L_y, z_{j0})\), with \(l_x\) and \(l_y\) integers. For such a periodic planar system an Ewald summation technique can be applied by using a rapidly convergent 2D lattice summation [21]. The following general sums over the
unit cells need to be considered:

\[ T^{\alpha\beta\gamma}_{ij} = \sum_{l_i, l_j = -\infty}^{\infty} \frac{x_{ij0}^{\alpha} y_{ij0}^{\beta} y_{ij0}^{\gamma}}{r_{ij0}^{\alpha}}. \]  
(A.17)

The prime on the sum indicates that the term with \( r_{ij0} = 0 \) is omitted. The lattice sums given by equations (A.6)–(A.15) are obtained by an appropriate choice of the positive integers \( \alpha, \beta \) and \( \gamma \). In general one has to distinguish between the cases \( z_{ij0} = 0 \) and \( z_{ij0} \neq 0 \).

The dipole field \( B_i^{\text{dip}} \) of particle \( i \), see equation (1), has the components

\[ B_i^{x,\text{dip}} = \mu_\text{at} \sum_j N_j \sum_{l_i, l_j = -\infty}^{\infty} \frac{1}{r_{ij}^2} \{(2x_{ij0}^2 - y_{ij0}^2 - z_{ij0}^2)m_i^x + 3x_{ij0}(y_{ij0}m_i^y + z_{ij0}m_i^z)\}, \]  
(A.18)

\[ B_i^{y,\text{dip}} = \mu_\text{at} \sum_j N_j \sum_{l_i, l_j = -\infty}^{\infty} \frac{1}{r_{ij}^2} \{(2y_{ij0}^2 - x_{ij0}^2 - z_{ij0}^2)m_i^y + 3y_{ij0}(x_{ij0}m_i^x + z_{ij0}m_i^z)\}, \]  
(A.19)

\[ B_i^{z,\text{dip}} = \mu_\text{at} \sum_j N_j \sum_{l_i, l_j = -\infty}^{\infty} \frac{1}{r_{ij}^2} \{(2z_{ij0}^2 - x_{ij0}^2 - y_{ij0}^2)m_i^z + 3z_{ij0}(x_{ij0}m_i^x + y_{ij0}m_i^y)\}. \]  
(A.20)

The unit vector \( m_j = M_j/N_j = (m_j^x, m_j^y, m_j^z) = (\sin \theta_j \cos \phi_j, \sin \theta_j \sin \phi_j, \cos \theta_j) \) determines the direction of the particle magnetic moment, with \( \theta_j \) and \( \phi_j \) as the polar and azimuthal angles.

Introducing now the lattice sums \( T^{\alpha\beta\gamma}_{ij} \), after some algebra one obtains for the three components of the dipole field

\[ B_i^{x,\text{dip}} = \mu_\text{at} \sum_j N_j \{2T_{ij}^{s20} - T_{ij}^{s02} - z_{ij0}T_{ij}^{s00}\}m_j^x + 3T_{ij}^{s11}m_j^y + 3z_{ij0}T_{ij}^{s10}m_j^z \]

\[ + \frac{5}{2}[(14T_{ij}^{r40} - 10T_{ij}^{r20} - 77T_{ij}^{r92} + T_{ij}^{r70} + T_{ij}^{r70} - 7T_{ij}^{r92} + 3x_{ij0}(T_{ij}^{r70} - 7T_{ij}^{r92}) + \frac{4}{5}T_{ij}^{r500})m_j^x \]

\[ + (2T_{ij}^{r31} - 9T_{ij}^{r711})m_j^y + z_{ij0}T_{ij}^{r90} - 9T_{ij}^{r710}m_j^z]((x_j^2) + (x_j^2)) \]

\[ + \frac{5}{2}[(14T_{ij}^{r922} - 2T_{ij}^{r720} - 77T_{ij}^{r920} + 3T_{ij}^{r70} + 3T_{ij}^{r70} - 7T_{ij}^{r922} + \frac{2}{5}T_{ij}^{r500})m_j^x \]

\[ + (2T_{ij}^{r713} - 9T_{ij}^{r711})m_j^y + z_{ij0}T_{ij}^{r90} - 9T_{ij}^{r710}m_j^z]((y_j^2) + (y_j^2)) \]

\[ + \frac{5}{2}[(14z_{ij0}T_{ij}^{r920} - 2T_{ij}^{r720} - 7z_{ij0}T_{ij}^{r920} + 3T_{ij}^{r70} + 3T_{ij}^{r70} - 7z_{ij0}T_{ij}^{r920} + \frac{2}{5}T_{ij}^{r500})m_j^x \]

\[ + z_{ij0}(21z_{ij0}T_{ij}^{r911} - 3T_{ij}^{r711})m_j^y + z_{ij0}(21z_{ij0}T_{ij}^{r910} - 9T_{ij}^{r710})m_j^z]((z_j^2) + (z_j^2)). \]  
(A.21)

\[ B_i^{y,\text{dip}} = \mu_\text{at} \sum_j N_j \{3T_{ij}^{s511}m_j^x + (2T_{ij}^{s502} - T_{ij}^{s520} - z_{ij0}T_{ij}^{s500})m_j^y + 3z_{ij0}T_{ij}^{s501}m_j^z \]

\[ + \frac{5}{2}[(14T_{ij}^{r922} - 2T_{ij}^{r720} - 77T_{ij}^{r920} + 5T_{ij}^{r70} + z_{ij0}(T_{ij}^{r70} - 7T_{ij}^{r920}) - \frac{2}{5}T_{ij}^{r500})m_j^x \]

\[ + (2T_{ij}^{r931} - 9T_{ij}^{r711})m_j^y + z_{ij0}(21T_{ij}^{r911} - 3T_{ij}^{r711})m_j^z]((x_j^2) + (x_j^2)) \]

\[ + \frac{5}{2}[(14T_{ij}^{r904} - 10T_{ij}^{r720} - 77T_{ij}^{r920} + 5T_{ij}^{r70} + z_{ij0}(T_{ij}^{r70} - 7T_{ij}^{r920}) + \frac{2}{5}T_{ij}^{r500})m_j^x \]

\[ + (2T_{ij}^{r913} - 9T_{ij}^{r711})m_j^y + z_{ij0}(21T_{ij}^{r910} - 9T_{ij}^{r710})m_j^z]((y_j^2) + (y_j^2)) \]

\[ + \frac{5}{2}[(14z_{ij0}T_{ij}^{r920} - 2T_{ij}^{r720} - 7z_{ij0}T_{ij}^{r920} + 5T_{ij}^{r70} + z_{ij0}(T_{ij}^{r70} - 7T_{ij}^{r920}) + \frac{2}{5}T_{ij}^{r500})m_j^x \]

\[ + z_{ij0}(21z_{ij0}T_{ij}^{r911} - 3T_{ij}^{r711})m_j^y + z_{ij0}(21z_{ij0}T_{ij}^{r910} - 9T_{ij}^{r710})m_j^z]((z_j^2) + (z_j^2)). \]  
(A.22)
The lateral extensions $L_x$ and $L_y$ of the unit cell and the components $x_{ij0}$ and $y_{ij0}$ of the interparticle distances within the unit cell are involved in a complicated manner in the lattice sums $T_{ij}^{aply}$, whereas the dependence on the vertical distances $z_{ij0}$ appears explicitly. Note that the contributions from mirror particles $i = j$ located in different unit cells are taken into account. The expressions for the dipole field components, equations (A.21)–(A.23), are simplified appreciably if all particles are located in the same plane ($z_{ij0} = 0$). In this case $B_{z\text{,dip}} \propto m_z^0$, and hence the $z$ component of the dipole field vanishes for an in-plane magnetization ($m_z^0 = 0$). As before, the point-dipole sum is recovered by setting $\langle x_i^2 \rangle = 0$, etc. The dipole energy $E_{\text{dip}}$ per unit cell is obtained from equation (1) by performing the sum over all particles $i = 1 \ldots n$ of the unit cell.

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