Dipolar interaction and sample shape effects on the hysteresis properties of 2\textit{d} array of magnetic nanoparticles

MANISH ANAND

Department of Physics, Bihar National College, Patna University, Patna 800 004, India

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Abstract. We study the ground-state and magnetic hysteresis properties of 2\textit{d} arrays (\(L_x \times L_y\)) of dipolar interacting magnetic nanoparticles (MNPs) by performing micromagnetic simulations. Our primary interest is to understand the effect of sample shape, \(\Theta\), the ratio of the dipolar strength to the anisotropy strength and the direction of the applied field \(\vec{H} = H_0 \hat{e}_H\) on the ground state and the magnetic hysteresis in an array of MNPs. To study the effect of the shape of the sample, we have varied the aspect ratio \(A_r = L_y/L_x\), which in turn, is found to induce shape anisotropy in the system. Our main observations are: (a) When the dipolar interaction is strong (\(\Theta > 1\)), the ground-state morphology has an in-plane ordering of magnetic moments, (b) the ground-state morphology has randomly oriented magnetic moments that are robust regarding system sizes and \(A_r\) for weakly interacting MNPs (\(\Theta < 1\)), (c) micromagnetic simulations suggest that the dipolar interaction decreases the coercive field \(H_c\), (d) the remanence magnetisation \(M_r\) is found to be strongly dependent not only on the strength of dipolar interaction but also on the shape of the sample and (e) due to the anisotropic nature of dipolar interaction, a strong shape anisotropy effect is observed when the field is applied along the long axis of the sample. In such a case, the dipolar interaction induces an effective ferromagnetic coupling when the aspect ratio is enormous. These results are of vital importance in high-density recording systems, magneto-impedance sensors, etc.

Keywords. Self-assembled arrays; dipolar interaction; hysteresis; micromagnetic simulations; ferromagnetism.

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1. Introduction

Magnetic nanoparticle (MNP) arrays are of profound importance because of their exciting physics and their numerous technological applications [1–12]. For instance, a two-dimensional array of MNPs is suitable for high-density digital storage and perpendicular recording media [13–15]. The magnetic properties in such a case depend strongly on the shape, size, geometry of the systems and the magnetic interactions. The primary magnetic interactions in such systems are dipole–dipole interactions. The dipolar interaction are long-ranged and favours ferromagnetic as well as anti-ferromagnetic couplings. As a result, unconventional morphologies are observed when the size of the system is comparable to the range of dipolar interactions. Due to the anisotropic nature of dipolar interactions, there have also been observations of ferromagnetic, striped, checkerboard patterns, vortices, etc., depending on the geometry of the lattice [16–21]. The magnetic hysteresis properties in such systems are found to be significantly influenced by the dipolar interactions [22–25].

Several studies in the literature make implicit and explicit references to the ubiquitous dipolar interactions and anisotropy in ordered arrays of MNPs. We summarise a few of them, relevant to our present work, as follows: (a) Li \textit{et al} studied the ground-state, magnetic-specific and magnetic hysteresis properties for three types of closely spaced nanomagnet arrays using Monte Carlo simulation [26]. They observed the vortex state due to the dipolar interactions in these arrays. For face-centred cubic nanomagnet arrays, a slight jump occurs in the hysteresis curve. (b) Yang \textit{et al} studied the magnetic properties in two-dimensional arrays of MNPs using the micromagnetic simulation [27]. They found that coercivity increases with arrays disorder. (c) Using the Landau–Lifshitz–Gilbert approach, Morales-Meza \textit{et al} studied the magnetisation reversal in a two-dimensional array of MNPs [28]. They showed that coercivity is reduced even if the particle position in the
array is random. (d) Chinni et al performed experiments and micromagnetic simulation using object-oriented micromagnetic framework (OOMMF) to study magnetic properties in nanocomposite films [29]. They observed an unexpected hysteretic behaviour characterised by in-plane anisotropy and crossed branches in the hysteresis curves measured along the hard direction. (e) Faure et al studied the magnetic properties of ordered arrays of MNPs using experiment and Monte Carlo simulation [30]. The dipolar interaction induces a ferromagnetic coupling that increases in strength with decreasing thickness of the array. (f) Xue et al studied the magnetic hysteresis properties of the two-dimensional hexagonal array of MNPs using theoretical calculations [15]. The hysteresis curves are found to vary their shapes from a rectangle to a non-hysteresis straight line through a set of complicated loops, in accordance with the magnetisation reversal process.

Although the aforementioned studies imply that dipolar interaction and geometrical arrangement of MNPs affect the magnetic properties of an ordered array of MNPs, a systematic study as a function of dipolar interaction strength, shape anisotropy of the system and direction of the applied field is still missing. In this work, we attempt to understand the effect of dipolar interaction manipulated by changing the interparticle separation, the direction of applied magnetic field and shape anisotropy induced by varying the aspect ratio on the magnetic properties of two-dimensional \( (2d) \) arrays of MNPs. The main questions which we have attempted to address are: (1) What are the consequences of dipolar interaction on ground-state organisations of MNPs? (2) How are the magnetic hysteresis properties modified due to the dipolar interactions, shape anisotropy of the sample and direction of the applied magnetic field? To answer these questions, we consider \( 2d \) \( (L_x \times L_y) \) arrays of cubical shaped Fe\(_3\)O\(_4\) MNPs. It is a well-known fact that uniformly magnetised cubical MNPs do not possess shape anisotropy [31]. But we have been able to induce it in the system by just varying the sample shape. It will be shown that the latter has a drastic effect on hysteresis behaviour. These ordered arrays of MNPs have many interesting properties due to structural order, well-defined interparticle interactions and geometry confinement [32]. In such a case, MNPs are coated with an inorganic surfactant to prevent agglomeration during self-assembling. Due to this, MNPs are at a reasonable distance beyond the range of exchange interaction. As a consequence, the predominant interaction in such arrays is dipolar [33]. Therefore, the energy of such an assembly is given by the sum of the anisotropy energy and the dipolar interaction energy.

To vary the relative dipolar interaction strength with respect to anisotropy strength, we have defined a ratio \( \Theta = D/KV \) where \( D \) is the dipolar interaction strength, \( K \) is the anisotropy constant and \( V \) is the volume of the nanoparticle. The value of \( \Theta \) for various interparticle separation \( a \) for Fe\(_3\)O\(_4\) is given in table 1. We refer to \( \Theta > 1 \) as the strong dipolar interaction regime and \( \Theta < 1 \) as the weak dipolar interaction regime. We have performed micromagnetic simulation using OOMMF code from NIST [34]. In the OOMMF code, the finite difference method is employed, which requires discretisation of a chosen geometry over a grid of identical prism cells. The magnetisation is supposed to be uniform in each cell. In the implementation of OOMMF, the continuous magnetic material is divided into discrete cubes (i.e. grid cells), which are in geometrical contact. This discretisation scheme suits a continuous magnetic film rather than an assembly of non-touching nanoparticles. Due to this reason, this type of modelling does not discriminate between the magnetic behaviour of \( 2d \) continuous films and \( 2d \) nanoparticle arrays. To overcome this, we have put exchange interaction to zero to mimic the separated MNP arrays magnetically. The absence of exchange interaction is also needed for the MNPs where they are coated with a surfactant to avoid agglomeration. We have successfully implemented it in our earlier studies [35,36]. We have used the Landau–Lifshitz–Gilbert (LLG) equation, which is used to describe the precessional motion of a moment in the magnetic field at \( T = 0 \text{ K} \). We have solved the coupled equation of motion for the given lattice to obtain the minimum energy configurations. To study hysteresis, we have applied a DC magnetic field \( \mu_0\bar{H} = H_0\hat{e}_H \), where \( \hat{e}_H = \hat{x}, \hat{y} \) and \( \hat{z} \) is the direction of the applied field along the \( x-, y- \) and \( z- \) axes respectively. We also study the role of aspect ratio \( A_r = L_y/L_x \) to induce shape anisotropy in the system and the direction of the field \( \hat{e}_H \).

The interplay of anisotropy, dipolar energy and aspect ratio creates unusual morphologies that profoundly affect magnetic properties. Our work, therefore, throws light on this subject from a microscopic picture and provides a basis for results obtained in experimental and theoretical studies. This paper is organised as follows: First, §2 introduces the model for an array of MNPs, the LLG equation, which provides the prototypical ground state (GS) morphologies and equilibrium morphologies. Then, in §3, we present our numerical results and discuss the dependence of the magnetic hysteresis on \( \Theta \), aspect ratio \( A_r = L_y/L_x \) and the direction of the applied field \( \hat{e}_H \). Finally, a conclusion of our results is provided in §4.
Table 1. Evaluation of the ratio $\Theta = D/KV$ for Fe$_3$O$_4$ as a function of interparticle separation $a$ for cubical shaped particle. The lateral dimension of the particle is $l = 10$ nm.

| $a$ (nm) | $\Theta = D/KV$ |
|---------|------------------|
| 10      | 1.75             |
| 12      | 1.01             |
| 16      | 0.42             |
| 20      | 0.22             |
| 30      | 0.06             |
| 40      | 0.03             |

2. Model and methodology

2.1 Model for dipolar interacting arrays of MNPs

We consider a self-assembled 2d array ($L_x \times L_y$) of cubical shaped MNPs in the xy-plane. The total number of the MNPs in the assembly are $N = L_x/a \times L_y/a$, where $l$ is the length of an edge of the particle and $a$ is the lattice spacing. The magnetic moment of each MNP is $\mu_i = \mu \hat{e}_i$, $i = 1, 2, \ldots, N$. The magnitude of the magnetic moment $\mu = M_s V$ where $M_s$ is the saturation magnetisation and $V = l^3$ is the volume of the MNP. The MNPs are assumed to have uniaxial anisotropy $K = K \hat{k}_i$, where $K$ is the anisotropy constant and $\hat{k}_i$ is the direction of anisotropy. Generally, MNPs are coated with a surfactant to prevent agglomeration, which suppresses the exchange interactions. The energy of such a system, therefore, includes contributions from anisotropy energy $E_a$ and dipolar interaction energy $E_d$ [37,38]:

$$E = E_a + E_d$$

$$= -K V \sum_i (\hat{k}_i \cdot \hat{e}_i)^2$$

$$- D \sum_{i,j} \frac{3(\hat{e}_i \cdot \hat{r}_{ij})(\hat{e}_j \cdot \hat{r}_{ij}) - (\hat{e}_i \cdot \hat{e}_j)}{(r_{ij}/a)^3},$$

(1)

where $\mu_0$ is the permeability of the free space, $\hat{e}_i$ is the unit vector along the magnetic moment, $\hat{r}_{ij}$ is the distance between particles $i$ and $j$ and $\hat{r}_{ij}$ is the unit vector along with it. The $1/r_{ij}^3$ dependence means that the dipolar interaction is long-ranged in nature. The analytical calculation of dipolar interaction between the cubical shaped MNPs can be found in ref. [39]. It is well-known that self-assembled MNPs usually form 2d close-packed hexagonal lattices, but magnetic properties are nearly independent of the lattice structure [40]. Here we define the dipolar interaction strength $D = \mu_0 \mu^2 / 4\pi a^3$. As magnetic properties of such an assembly are governed by the relative strength of the anisotropy and dipolar energy, we have defined a ratio $\Theta = D/KV$.

Although dipolar interactions and anisotropy energy depend on various system parameters, the behaviour of the assembly will be dictated by $\Theta$ rather than by the precise values of parameters such as $a$, $V$, $\mu$ and $K$. When $D$ is more significant than $KV$, the dipolar interaction is stronger than anisotropy, i.e., $\Theta > 1$. Similarly, $\Theta < 1$ can be termed as the weak dipolar regime.

In the presence of an external magnetic field $\mu_0 \vec{H}$, there is an additional contribution to energy $E$ given by [38]

$$E_H = -H_0 \sum_{i=1}^{N} \mu_i \cdot \hat{e}_H,$$

(2)

where $H_0$ is the magnitude of the field and $\hat{e}_H$ is the unit vector in the direction of the applied field.

2.2 Landau–Lifshitz–Gilbert equation

The precessional motion of the magnetic moment $\mu_i$ in a magnetic field can be described by the LLG equation [41]:

$$\frac{d\mu_i}{dt} = -\gamma \frac{\mu_i \times \vec{H}^e}{\mu_0} - \lambda (\mu_i \times (\mu_i \times \vec{H}^e)),$$

(3)

where $\gamma$ is the electron gyromagnetic ratio, $\lambda = \gamma \alpha / M_s$ is a phenomenological dimensionless damping factor and $\vec{H}^e = -\partial E_T / \partial \mu_i$ is the effective field experienced by the magnetic moment, where $E_T = E + E_H$. While studying ground-state properties, $E_H$ is taken to be zero. The first term in eq. (3) takes care of the precession of $\mu_i$ around $\vec{H}^e$. The second term is due to a phenomenological dissipative motion: the magnetic moment $\mu_i$ precesses around $\vec{H}^e$. The solution of these coupled differential equations yields the GS configuration $[\mu_i]$, of the assembly.

The entire system is discretised into cells to perform micromagnetic simulation using OOMMF code, where the lateral dimension of each cell is $l$. Each cell in such a case represents a magnetic moment $\mu = M_s V$. The centre-to-centre separation between moments is, therefore, $l \equiv a_0$. By eq. (1), the strength of the dipolar interaction can be manipulated by varying the centre-to-centre separation $a$ of the MNPs. However, with the protocol implemented in OOMMF, a change in the centre-to-centre separation from $a_0$ to $a_\beta$, changes the cell volume from $V \equiv V_0$ $(=a_0^3)$ to $V_\beta$ $(=a_{\beta}^3)$. Consequently, the magnetic moment gets altered to $\mu_\beta = M_s V_\beta$, which ultimately modifies the magnetic properties of the particles under study. This undesirable artefact in the simulation needs to be overcome. To overcome
this artefact, we formulated a rescaling method for saturation magnetisation:

\[ M^\beta_s = M^0_s \frac{V_0}{V_\beta} \]  
\( \text{(4)} \)

where \( M^0_s \) is the saturation magnetisation for nanoparticle of volume \( V_0 = a_0^3 \). It is easy to see that now \( \mu = M^\beta_s V_\beta = M^0_s V_0 \) as desired. Corresponding changes need to be incorporated in other related variables of interest such as the coercive field \( H_c \) and the anisotropy field \( H_K = 2K/M_s \) for non-interacting or weakly interacting MNPs, which plays an important role in hysteresis:

\[ H_c^\beta = H_c^0 \frac{V_\beta}{V_0}, \]  
\( \text{(5)} \)

\[ H_K^\beta = H_K^0 \frac{V_\beta}{V_0}. \]  
\( \text{(6)} \)

We have successfully implemented this scaling procedure to study the heat dissipation and spin transport properties in the assembly of dipolar interacting MNPs in our earlier work [35,36].

3. Numerical results

We consider cubical shaped MNPs of Fe3O4 arranged on a 2d \((L_x \times L_y)\) lattice. The particle size is chosen to be \( l = 10 \) nm. We have used anisotropy constant \( K = 13 \times 10^3 \) Jm\(^{-3}\) and saturation magnetisation \( M_s = 4.77 \times 10^2 \) A/m for numerical evaluations. Six values of interparticle separation \( a \) are considered for magnetic hysteresis study: \( a_0 = 10 \) nm, \( a_1 = 12 \) nm, \( a_2 = 16 \) nm, \( a_3 = 20 \) nm, \( a_4 = 30 \) nm and \( a_5 = 40 \) nm. Table 1 provides the values of \( \Theta \) for these interparticle separations. The initial condition that we choose for the assembly of MNPs is random orientations of magnetic moments and anisotropy axes. All the data obtained using the simulations are averaged over 50 sets of initial conditions. The ground-state morphologies are obtained by solving the LLG equation using the OOMMF code without an external magnetic field. To study the magnetic hysteresis properties, we apply a DC magnetic field of strength \(-150\) mT to \(150\) mT for \( a = 10 \) nm. For other interparticle separations, the scaled saturation magnetisation \( M^\beta_s \) was obtained using eq. (4), corresponding \( H_c^\beta \) and \( H_K^\beta \) were calculated using eqs (5) and (6), respectively. We have chosen the magnetic field to be large enough compared to \( H_c \) to achieve saturation. The ramping up or slowing down has been appropriately fine-tuned to capture the magnetic properties near \( H_c \). We study the dependence of magnetic hysteresis on \( \Theta \), aspect ratio \( A_r = L_y/L_x \) and the direction of the applied field \( \hat{e}_H \). In our simulations, \( L_x = 120 \) nm; \( A_r = 1, 2, 4, 8, 16 \) and 32; \( \Theta = 1.75, 1.01, 0.42, 0.22, 0.06 \) and 0.03; \( \hat{e}_H = \hat{x}, \hat{y} \) and \( \hat{z} \).

3.1 Ground-state (GS) morphologies

We first study the effect of \( \Theta \) and \( A_r \) on ground-state spin morphologies. For this purpose, we took square samples, i.e. \( A_r = 1.0 \), such that \( L_x \times L_y \equiv L_x \times L_y \). Figure 1 depicts GS morphologies for \( L_x = 120 \) nm. The interparticle separation \( a \) is chosen to be 10 nm (see figure 1a) and 20 nm (see figure 1b). These correspond to \( \Theta = 1.75 \) and 0.22, respectively. So the number of spins in figure 1a is \( 12 \times 12 \) and \( 6 \times 6 \) in figure 1b. We depict the spins with non-zero \( z \)-component by the green cone. Those lying in the \( xy \) plane with a positive \( x \)-component have been indicated by the red coloured cone, while those with negative \( x \)-component by the blue coloured cone. It is quite evident that for strongly interacting MNPs (\( \Theta = 1.75 \)), all the spins are in the \( xy \)-plane (see figure 1a). They exhibit locally ordered regions. On the other hand, when magnetic interactions are weak (\( \Theta = 0.22 \)), the moments are randomly oriented, signifying a lack of magnetic order (see figure 1b). Magnetic moments also tend to align normal to the plane of the sample. It means that dipolar interaction favours in-plane ordering. Then, we study the effect of aspect ratio \( A_r \) on the spin morphologies. For \( \Theta < 1 \), the features are unchanged as \( A_r \) is increased to 2, 3, 4, etc. and are prototypically represented by figure 1b. We do not show them to avoid repetition. In figure 2, we depict morphologies corresponding to the strongly dipolar interacting MNPs (\( a = 10 \) nm, \( \Theta = 1.75 \)) for \( L_x = 120 \) nm and two aspect ratios (a) \( A_r = 2 \) (see figure 2a) and (b) \( A_r = 4 \) (see figure 2b). In both cases, the MNPs exhibit local order and prefer to lie in the \( xy \)-plane. The morphology of the magnetic moments near the sample edges is very distinct from that in bulk. The moments tend to align along the edges as ferromagnetic chains. These observations can be explained as follows. As the anisotropy axes are assumed to have random orientations in three-dimensional space in the present work, the magnetic moments tend to follow the anisotropy directions for small dipolar interaction strength, i.e. \( \Theta < 1 \). Consequently, there is no preferential direction of magnetic moments for weakly interacting MNPs, as evident in figure 1b. It is also well-known that the dipolar interaction favours in-plane ordering of magnetic moments as the energy cost is very high for the orientations normal to the sample plane [43]. As a result, there is an in-plane ordering of magnetic moments for large \( \Theta \) (see figure 1a). Interesting physics emerges in the highly anisotropic system (huge aspect ratio \( A_r \)). In such a case, the dipolar interaction of enough strength induces shape
anisotropy in the system [44]. In other words, the dipolar interaction promotes ferromagnetic coupling among the magnetic moments in such a case [44]. As a consequence, magnetic moments tend to form chain-like configurations for large $\Theta$, as depicted in figure 2.

3.2 Magnetic hysteresis study

Next, we study the effect of dipolar strength $\Theta$, shape anisotropy of the sample and direction of the applied field $\vec{e}_H$ on the magnetic hysteresis in a systematic manner. To vary the dipolar interaction strength, we have varied the interparticle separation. To induce shape anisotropy in the system, we have changed the aspect ratio $A_r$ of the sample. In figure 3, we have plotted magnetic hysteresis curves for the square sample, i.e., $L_x = L_y = 1.0$ as a function of $\Theta$ and $\vec{e}_H$. We have considered six values of dipolar strength $\Theta (= 1.75, 1.01, 0.42, 0.22, 0.06$ and $0.03)$ in each case. The magnetic field axis ($x$-axis) has been scaled by $H_K$, the anisotropy field (for non-interacting MNPs with randomly oriented anisotropy axes). The direction of the external field is (a) $\mu_0\vec{H} = H_0\hat{x}$ (see figure 3a), (b) $\mu_0\vec{H} = H_0\hat{y}$ (see figure 3b) and (c) $\mu_0\vec{H} = H_0\hat{z}$ (see figure 3c). It is quite evident that when the field is applied in the plane of the sample, i.e., either along the $x$- or $y$-direction, the coercive field $H_c$ decreases with an increase in the strength of dipolar interaction. As a consequence, the area under the hysteresis curve diminishes. When the field is applied normal to the sample ($z$-direction) plane, the magnetic moment ceases to follow the applied field for $\Theta > 1$. As a result, non-hysteresis is observed, and so $H_c$ and $M_r$ tend to zero for strongly interacting MNPs. It is clearly seen that the coercive field $H_c \approx 0.48H_K$ and $M_r \approx 0.5$ for weak interaction or non-interacting MNPs ($\Theta < 1$) irrespective of the direction of the applied field. These values of $H_c$ and $M_r$ correspond to single-particle hysteresis with the randomly oriented anisotropy axes [42,45]. It means that the magnetic hysteresis follows the Stoner–Wohlfarth model when the interaction among the MNPs is weak as expected [42,45].

To study the dependence of shape anisotropy on the magnetic properties, we study the magnetic hysteresis as a function of aspect ratio $A_r$ for various values of $\Theta$ and three directions of the applied field, i.e., along $x$-, $y$- and $z$-axes, respectively. We have considered six values of aspect ratio $A_r (= 1.0, 2.0, 4.0, 8.0, 16.0$ and $32.0)$ in each case. In figure 4, the direction of the applied field is in the plane of the sample [$\mu_0\vec{H} = H_0\hat{x}$ and $\mu_0\vec{H} = H_0\hat{y}$]. The values of $\Theta$ are (a) $\Theta = 1.75$ (see figures 4a and 4e), (b) $\Theta = 1.01$ (see figures 4b and 4f), (c) $\Theta = 0.42$ (see figures 4c and 4g) and (d) $\Theta = 0.22$ (see figures 4d and 4h). The direction of the applied
Figure 3. Magnetic hysteresis behaviour as a function of dipolar interaction strength and direction of the applied field for square sample, i.e., $A_y = 1.0$ ($L_x = L_y = 120$ nm). (a) The external field $\mu_0 \hat{H}$ is applied along the $x$-direction, (b) $\mu_0 \hat{H} = H_0 \hat{y}$ and (c) $\mu_0 \hat{H} = H_0 \hat{z}$. The strength of dipolar interaction has been varied from a very large value of 1.75–0.03. When the field is applied in the sample plane, coercive field $H_c$ and remanent magnetisation decrease with an increase in the strength of the dipolar interaction. When the field applied is normal to the sample plane, $H_c$ and $M_r$ decrease to zero for large dipolar strength. The coercive field $H_c \approx 0.48 H_K$ and $M_r \approx 0.5$ are observed for weakly interacting MNPs irrespective of the direction of the applied field as expected (signature of the Stoner–Wohlfarth model).

field is normal to the plane of the sample (along the $z$-axis) in figures 5. The other parameters, $A_r$ and $\Theta$ in figures 5, remain the same as that of figures 4. When the applied field is along the shorter axis (along the $x$-axis), dipolar interaction decreases the value of $H_c$ and $M_r$ (see figures 4a–4d). There is a weak dependence of $H_c$ on $A_r$. For weakly interacting MNPs, hysteresis curves follow the Stoner–Wohlfarth model [45]. A strong effect of shape anisotropy is observed when the field is applied along the increasing length of the sample, i.e., along the $y$-axis. The remanent magnetisation $M_r$ increases with an increase in $A_r$, and it reaches 0.9 for strongly interacting MNPs (see figure 4e). In this case also, the larger is the strength of dipolar interaction, the smaller is the value of $H_c$ (see figures 4c–4h). The magnetic moments cease to follow the applied magnetic field when the field is applied along the $z$-axis for strongly interacting MNPs. As a consequence, almost no hysteresis is observed for strongly interacting MNPs with applied field normal to the plane of the sample (see figures 5a and 5b). Like the other two cases stated already, weakly interacting MNPs follow the Stoner–Wohlfarth model irrespective of $A_r$.

3.3 Characterisation of magnetic hysteresis curves

Finally, we study the variation of the coercive field $H_c$ and $M_r$ as a function of $\Theta$, $A_r$ and $\hat{e}_H$ by extracting their values from the simulated magnetic hysteresis curves. Figure 6 shows the variation of $H_c$ (scaled by $H_K$) and $M_r$ as a function of dipolar strength $\Theta$ for six values of
Figure 5. Magnetic hysteresis as a function of dipolar interaction strength $\Theta$ and aspect ratio $A_r$ when the field is applied normal to the plane of the sample $\mu_0 \vec{H} = H_0 \hat{z}$. Four values of $\Theta$ (= 1.75, 1.01, 0.42 and 0.22) have been considered. The aspect ratio $A_r$ is varied from 1.0 to 32.0 in each case. For large dipolar interaction strength, the magnetic moments cease to align normal to the sample and as a result, non-hysteresis is observed. The coercive field $H_c$ and $M_r$ decrease down to zero in this case. For weak dipolar interaction, i.e., $\Theta = 0.22$, magnetic hysteresis follows the Stoner–Wohlfarth model ($H_c \approx 0.48H_K$ and $M_r \approx 0.5$) irrespective of the aspect ratio $A_r$.

These results can be explained by probing the effect of dipolar interaction and shape anisotropy of the sample induced by varying the aspect ratios. It is evident from the ground-state morphologies that the dipolar interactions favour the in-plane ordering of magnetic moments. When $A_r$ is increased, the dipolar interaction favours ferromagnetic coupling between the magnetic moments. The demagnetisation field decreases as shape anisotropy is increased ($A_r$ increases in our case), which is also reported by Wysin [46]. Consequently, magnetic moments tend to align along the long axis of the sample as it costs less energy compared to other configurations. It has also been reported by Jordanovic et al [9]. This ferromagnetic coupling for strongly interacting MNPs can also be explained by the fact that the exchange interaction between the MNPs is assumed to be zero in the present work. Due to the absence of exchange interaction, the energetics of domain walls are entirely controlled by the dipolar interaction. It makes it energetically favourable to form domain walls along the increasing length of the sample. When an external field is applied to this system, the response of this system not only depends on the dipolar strength but also on the shape anisotropy of the sample, which has been induced by increasing $A_r$ in our work. When the field is applied along the shorter axis of the sample, i.e., along the $x$-axis, the natural tendency to get aligned along the long axis of the sample is hindered. As a result, magnetic moment ceases to follow the external field, which is reflected in a decrease in the value of $M_r$ ($\approx 0.1$) and $H_c$ to $0.1H_K$ for strongly interacting MNPs with $A_r = 32.0$. Magnetic moments are found to follow the field when the field is applied along the long axis of the sample, but due to the increase in the strength of dipolar interaction as well, $H_c$ decreases, but its value remains slightly higher than that of the previous situation. Due to the same reason, $M_r$ increases with $A_r$ for $\Theta > 1$. This increase of $M_r$ and decrease of $H_c$ when the field is applied along the long axis of the sample is in qualitative agreement with the work done by García-Arribas et al [47]. They have studied the shape anisotropy effect in thin-film permalloy microstrips.

When we force the magnetic moments to get aligned normal to the plane of the sample by applying the field along the $z$-axis, they oppose strongly for a large value of $\Theta$ and $A_r$. This happens due to the fact that the natural tendency of the magnetic moments is to get organised in the plane of the sample ($xy$-plane in our case). As a result, non-hysteresis is observed in this case, which is reflected in the zero value of $H_c$ and $M_r$. This non-hysteresis behaviour has also been reported by Xue et al [15]. They have studied the hysteresis properties in a two-dimensional hexagonal array with aligned uniaxial anisotropy. From all the cases, it is quite
evident that dipolar interaction always decreases the coercive field \( H_c \) because the dipolar interaction causes a collective reversal of the magnetic moments under an applied magnetic field. Consequently, the coercive field decreases with an increase in dipolar interaction strength.

4. Conclusion

To conclude, we have studied the effect of dipolar interactions and shape of the sample on the magnetic properties of the 2d \((L_x \times L_y)\) array of cubical shaped magnetic nanoparticles (MNP) by performing micromagnetic simulation using OOMMF code [34]. It is well-known that uniformly magnetised cubical shaped particles do not possess shape anisotropy [31]. Still, we have been able to induce the latter in the system by varying the aspect ratio \( A_r \). Our primary aim was to study ground-state (GS) morphologies and understand the magnetic hysteresis properties as a function of \( \Theta \), shape anisotropy induced by varying the aspect ratio \( A_r (= L_y/L_x) \) and the direction of the applied field \( \mu_0 \hat{H} = H_0 \hat{e}_H \). Our observations are as follows: (a) For weakly interacting MNP (\( \Theta < 1 \)), the magnetic moments are randomly oriented, and the morphology is unaffected by \( A_r \). MNP also tend to align normal to the plane of the sample. (b) For strong dipolar strengths (\( \Theta > 1 \)), magnetic moments prefer to orient in the sample plane. The morphology, in this case, comprises regions of correlated moments. (c) Magnetic moment favours ferromagnetic coupling along the longer axis of the sample when \( A_r \) is increased provided \( \Theta > 1 \). (d) When the dipolar interaction is weak, magnetic hysteresis curves follow the Stoner–Wohlfarth model \( H_c \approx 0.48 H_K \) and \( M_r \approx 0.5 \) irrespective of the direction of the applied field \( \hat{e}_H \) and aspect ratio \( A_r \). (e) A strong effect of shape anisotropy is observed when the field is applied along the increasing length of the sample, reflected in the very large value of remanent magnetisation \( M_r \approx 0.9 \). (f) The dipolar interaction always decreases the coercive field \( H_c \). (g) Thus, for strongly interacting MNP, \( H_c \) and \( M_r \) tend to zero when the field is applied normal to the sample plane.

We have succeeded in obtaining guidelines regarding how magnetic hysteresis properties may change in the dipolar interacting MNP arrays. The obtained results contribute to expanding the fundamental comprehension of two-dimensional dipolar interacting systems. Furthermore, they offer exciting implications for creating self-assembled arrays of magnetic nanoparticles of desired magnetic response. The micromagnetic simulation showed that the dipolar interaction induces an effective ferromagnetic coupling in thin arrays (when the aspect ratio is enormous). It means that one can modulate the magnetic properties of the ordered arrays of MNPs by varying the strength of shape anisotropy of the system. The latter can be changed by varying the width and length of the system. Our results are also relevant for the experimental samples, which can be divided into various categories depending on how they behave when an external magnetic field is applied to them.

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