Tailoring heat dissipation in linear arrays of dipolar interacting magnetic nanoparticles

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Abstract. We perform numerical simulations to analyse the effect of dipolar interaction, particle size $D$ and thermal fluctuations on the magnetic hysteresis in the linear array of magnetic nanoparticles (MNPs). The shape of the hysteresis curve is that of Stoner and Wohlfarth particle for non-interacting MNPs and temperature $T = 0$ K. The area under the magnetic hysteresis curve is minimal with $D \approx 8–16$ nm and $T = 300$ K, indicating the predominance of superparamagnetic character. Interestingly, the dipolar interaction of sufficient strength moves the nanoparticles from superparamagnetic to a blocked state even at $T = 300$ K, resulting in an enhanced hysteresis loop area in such cases. Even with negligible dipolar interaction strength ($\lambda \approx 0.0$) and $T = 300$ K, the hysteresis loop area is appreciable for ferromagnetic particles ($D > 16$ nm). The coercive field $\mu_0 H_c$ and the blocking temperature $T_B$ also depend strongly on the dipolar interaction. They are found to increase with dipolar interaction strength $\lambda$ and particle size. Our extensive simulations also reveal a significant deviation of $\mu_0 H_c$ from $T^{3/4}$ dependence because of dipolar interaction. There is a rapid fall in the amount of heat dissipated $E_H$ and coercive field with temperature for superparamagnetic nanoparticles ($D \approx 8–16$ nm) and small dipolar interaction strength ($\lambda \leq 0.6$). In contrast, they are significantly large and depend weakly on thermal fluctuations for $D > 16$ nm. Therefore, observations made in the present work can help experimentalists to choose precise values of system parameters to obtain desired hysteresis properties, essential for diverse technological applications such as magnetic hyperthermia, data storage, etc.

Keywords. Superparamagnetism; dipolar interaction; hysteresis; magnetic hyperthermia.

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1. Introduction

Recent years have seen a surge in magnetic nanoparticles (MNPs)-based research due to their numerous technological applications [1–6]. These include digital data recording, spintronics devices, drug delivery, biosensors, etc. [7–10]. MNPs have also received significant attention because of their usages in magnetic hyperthermia [11,12]. In this application, nanoparticles are directed near the malignant cells, and an oscillating magnetic field is applied. The heat release due to the hysteresis kills the malignant cells [12]. The heating capability of these nanosystems depends strongly on the particle size distribution, anisotropy strength, dipolar interaction, frequency, the strength of an external magnetic field, etc. [13–15]. Therefore, there is a growing interest in understanding how nanoparticles’ heat release can be enhanced and manipulated in a more controlled manner.

Various aspects of magnetic hyperthermia are well understood for the dilute assembly of MNPs [16,17]. However, MNPs interact due to dipolar interaction. Therefore, their heating capability is drastically modified in such cases [18]. The dipolar interaction can favour ferromagnetic or antiferromagnetic interaction depending on the relative orientation and position of the particles [19]. It is because of the anisotropic nature of dipolar interaction [19]. Consequently, it has a far-reaching effect on various magnetic properties of crucial importance, such as modified energy barriers, frustrations of spins, complex magnetisation dynamics, unique morphology, etc. [20–23]. For instance, the dipolar interaction promotes spin glasses characteristics in the assembly of randomly distributed MNPs [24]. In other cases, the dipolar interaction promotes ferromagnetic coupling in a linear chain of MNPs [25]. Numerous works have shown that dipolar interaction strongly affects various magnetic properties in MNPs assembly
systems using experiments and analytical methods [29]. Dipolar interaction is found to dictate the hysteresis characteristics in the high-temperature limit [29]. Tan et al. observed spatial distribution of heat dissipation because of the dipolar interaction in a three-dimensional assembly of MNPs [30]. Fu et al. also studied the effect of dipolar interaction on the heating efficiency of MNPs using computer simulations and experiments [31]. The heat dissipation decreases as the dipolar interaction strength is enhanced in the isotropic assembly of MNPs. Generally, superparamagnetic nanoparticles are used for magnetic hyperthermia because of their easy controllability, superior magnetic and physicochemical properties [32,33]. Although, the major disadvantage associated with such particles is their aggregation, which reduces their dispersibility and their intrinsic stability for a more extended period [34,35]. The amount of heat dissipation is significantly reduced due to the dipolar interaction in the isotropic assembly of these ultrafine nanoparticles [31,36]. Usov et al. [37] and Guibert et al. [38] also found the detrimental effect of magnetic interaction on the heating efficacy. In contrast, the dipolar interaction improves the heating capability in systems with substantial shape anisotropy such as chain-like and columnar arrangement [39–42].

The arrangement of MNPs in a particular shape depends on various factors such as dipolar interaction, an external magnetic field strength, temperature, etc. [43,44]. Therefore, we should find a way to maximise the heat dissipation and reduce the adverse effect of dipolar interaction even in the isotropic assembly. Various works advocate that ferromagnetic nanoparticles can be excellent alternatives to the superparamagnetic counterpart as the former has a higher heating capability [45–49]. For instance, Mehdaoui et al. probed the heating performance in MNPs assembly using experiments and numerical simulations. Ferromagnetic particles were found to be the best candidates for hyperthermia as they displayed the maximum specific losses reported in the literature [47]. Carrey et al. also obtained a more substantial heating efficiency with ferromagnetic nanoparticles [48]. Chang et al. studied the heating efficiency in human-like collagen protein-coated MNPs using experiments [49]. They also found that ferromagnetic nanoparticles are better-suited candidates for magnetic hyperthermia. Thus motivated, we extensively investigate the hysteresis mechanism in ordered arrays of nanoparticles of various sizes ranging from superparamagnetic to ferromagnetic using kinetic Monte Carlo (kMC) simulation. We also probe the dependence of hysteresis on the long-ranged dipolar interaction and thermal fluctuations. The one-dimensional array is chosen as it is the building block of various possible geometries. We assume the anisotropy of the MNPs to be randomly oriented to be close to the real system. The KMC simulation is extensively utilised to investigate a variety of time-dependent properties in MNP assembly [18,30,50].

The rest of the paper is organised as follows. In §2, we present the model and describe the various energy terms. In §3, we present the numerical results. Finally, in §4 we provide the summary of the present work.

2. Model

We consider $n$ spherical and monodisperse nanoparticles arranged in a linear array, as shown in figure 1a. Let the particle diameter be $D$, MNP volume be $V$, magnetocrystalline anisotropy strength be $K_{eff}$ and saturation magnetisation be $M_s$. The magnetic moment of each particle $\mu = M_s V$. We introduce a control parameter $\lambda = D/d$ to manipulate dipolar interaction strength, $d$ being the distance between the consecutive MNPs (see figure 1a). Therefore, $\lambda = 0$ captures the physics of the non-interacting case while $\lambda = 1.0$ mimics the strongest dipolar interacting system.
The anisotropy energy of such a nanoparticle can be given by the following relation [30,48,50]:

\[ E_K = K_{\text{eff}} V \sin^2 \Phi, \]  

(1)

where the magnetic moment is inclined at an angle \( \Phi \) with respect to the easy axis. The anisotropy energy [eq. (1)] has two minima at \( \Phi = 0 \) and \( \pi \), respectively. An energy maximum, also termed as the energy barrier, of strength \( K_{\text{eff}} V \) separates them. The magnetisation reversal occurs from one energy minimum to another and vice versa if thermal fluctuations are comparable or more significant than \( K_{\text{eff}} V \). The average time taken in such a reversal of magnetisation is known as the Néel relaxation time \( \tau_N \), which is expressed as [48,50]

\[ \tau_N = \tau_0 \exp(K_{\text{eff}} V/k_B T), \]  

(2)

where \( \tau_0 = (2v_0)^{-1} \) with \( v_0 = 10^{10} \text{ s}^{-1} \), \( k_B \) is the Boltzmann constant and \( T \) is the temperature. When \( \tau_N \) becomes comparable to the experimental measuring time \( \tau_m \), i.e., \( \tau_N \approx \tau_m \), the nanoparticle is said to be in the blocked state. In such a case, the magnetic behaviour of the MNPs is distinguished by the blocking temperature \( T_B \) below which the magnetic moments appear frozen. It is approximated as [51]

\[ T_B \approx \frac{K_{\text{eff}} V}{k_B \ln(\tau_m/\tau_0)}, \]  

(3)

where \( \tau_m \approx (2\pi v)^{-1} \). Equation (3) is valid for single nanoparticle or dilute assembly of MNPs.

The single-particle energy barrier gets modified in a dense assembly of magnetically interacting MNPs. We can write the expression of the corresponding dipolar field as [30,50]

\[ \mu_0 \tilde{H}_{\text{dip}} = \frac{\mu_0}{4\pi} \sum_{j \neq i} \frac{3(\hat{\mu}_j \cdot \hat{r}_{ij})\hat{r}_{ij} - \hat{\mu}_j}{r_{ij}^3}. \]  

(4)

Here \( \mu_0 \) is the permeability of free space, the \( j \)th magnetic moment has a unit vector \( \hat{\mu}_j \), \( r_{ij} \) is the separation between two nanoparticles at the \( i \)th and \( j \)th positions in the system and \( \hat{r}_{ij} \) is the unit vector corresponding to \( r_{ij} \). We apply the external oscillating magnetic field along the linear array of nanoparticles or the \( z \)-axis to investigate the hysteresis response of the underlying system. The following relation gives the corresponding magnetic field:

\[ \mu_0 H = \mu_0 H_0 \cos 2\pi vt, \]  

(5)

where \( \mu_0 H_0 \) and \( v \) are the amplitude of the magnetic field and frequency, respectively. Therefore, the energy of the \( i \)th nanoparticle in such a case takes the following form [30,50]

\[ E(\Phi_i, \phi_i) = K_{\text{eff}} V \sin^2 \Phi_i \]

\[ -\mu_0 H_{\text{total}} \cos(\Phi_i - \phi_i). \]  

(6)

The anisotropy field of the \( i \)th nanoparticle is inclined at an angle \( \phi_i \) with respect to the total field \( H_{\text{total}} \) (dipolar and external magnetic fields).

The KMC simulation is used to investigate the effect of dipolar interaction on the hysteresis characteristics with particle sizes ranging from superparamagnetic to ferromagnetic. The KMC method is more efficient in probing the time-dependent magnetic properties with better precision compared to the Metropolis Monte Carlo simulation technique.

The KMC algorithm implemented in the present work is described in refs [30,42,50]. We, therefore, provide brief details of the KMC algorithm used in the present article. The total simulation time is divided into time steps \( t_{\text{step}} \) during which the magnetic field is kept constant. The hysteresis is discretised into 2000 equal time steps. We now briefly discuss the algorithm implemented in the main loop of the KMC code:

(i) The direction of magnetisation is taken in three-dimensional space.

(ii) The total magnetic field experienced by each magnetic nanoparticle is then calculated using eqs (4) and (5).

(iii) We then divide the \( E(\Phi) \) function into 200 points to evaluate the energy extremum of a nanoparticle. We assume \( \Phi_1, \Phi_2 \) and \( \Phi_3 \) to be the angles of two energy minima and lower energy maximum with energies \( E_1, E_2 \) and \( E_3 \), respectively. We evaluated them using Newton’s method by taking the first derivative of eq. (6).

(iv) The magnetisation switching rate \( v_1 \), which is a quantity to change the orientation of magnetisation from \( \Phi_1 \) to \( \Phi_2 \), is now calculated as [52]

\[ v_1 = v_1^0 \exp \left(-\frac{E_3 - E_1}{k_B T}\right). \]  

(7)

Similarly, the flipping rate \( v_2 \) from \( \Phi_2 \) to \( \Phi_1 \) can be evaluated as follows [52]:

\[ v_2 = v_2^0 \exp \left(-\frac{E_3 - E_2}{k_B T}\right). \]  

(8)

Here \( v_1^0 = v_2^0 \) are attempt frequencies, which equals \( 10^{10} \text{ Hz} \) [30]. We then estimate the probability of finding the magnetisation at \( \Phi_1 \) after a time \( t_{\text{step}} \) if it is in \( \Phi_2 \) using the following expression [53]:

\[ P(t_{\text{step}}) = \frac{v_2}{v_1 + v_2} [1 - \exp(1 - (v_1 + v_2)t_{\text{step}})]. \]  

(9)
(v) A random number between 0 and 1 is then generated. The switching of magnetisation is accepted depending on whether the random number is greater or lower than $P \langle t_{\text{step}} \rangle$.

(vi) The reiteration starts from step (i).

We can evaluate the total heat release $E_H$ because of hysteresis using the following expression [48,54]:

$$E_H = \int_{-\mu_0 H_0}^{\mu_0 H_0} M(H) \, dH,$$

where $M(H)$ is the magnetisation of the system at magnetic field $H$.

3. Simulation results

The values of system parameters considered are: $n = 100$, $D = 8–64$ nm, $K_{\text{eff}} = 13 \times 10^3$ J m$^{-3}$, $M_s = 4.77 \times 10^5$ A m$^{-1}$, $\nu = 10^3$ Hz and $T = 0–300$ K. We have also taken three values of magnetic field strength $\mu_0 H_0 = 0.05, 0.075$ and 0.1 T, which are about 0.92, 1.38 and 1.84 times the anisotropy field $H_K = 2K_{\text{eff}}/M_s$, respectively. These magnetic field and frequency values meet the criteria $\mu_0 H_0 \nu < 5 \times 10^9$ A m$^{-1}$ s$^{-1}$, a safer limit suited for magnetic hyperthermia applications [55]. We have varied $\lambda$ between 0 and 1.0. $K_{\text{eff}}$ and $M_s$ values are considered for bulk magnetite (Fe$_3$O$_4$), which is one of the most promising candidates for magnetic hyperthermia applications due to its bio-compatibility and driving accumulation features [56].

We first study the hysteresis behaviour of the superparamagnetic nanoparticles as a function of dipolar interaction. Figure 1 shows the magnetic hysteresis curves for five typical values of $\lambda$: 0.0, 0.4, 0.6, 0.8 and 1.0 with $T = 0$ and 300 K and $D = 8$ nm. The external magnetic field axis (x-axis) is re-scaled by the single-particle anisotropy field $H_K$ and the magnetisation axis (y-axis) by $M_s$ in all the hysteresis curves. In the absence of temperature ($T = 0$ K) and dipolar interaction ($\lambda = 0.0$), the hysteresis curve resembles the Stoner and Wohlfarth model [57]. In this case, coercive field $\mu_0 H_c \approx 0.479 H_K$ and remanent magnetisation $M_r$ is close to 0.5, as expected for Stoner and Wohlfarth particle with randomly oriented anisotropy axis [57]. The hysteresis loop area is extremely small for weakly or non-interacting MNPs with $T = 300$ K indicating superparamagnetic behaviour. We can explain it using the fact that particle size being in the superparamagnetic regime, the blocking temperature $T_B$ is minimal ($\approx 16.86$ K) [using eq. (3)] compared to simulation temperature $T = 300$ K. Therefore, thermal energy overcomes the energy barrier, instigating the magnetisation to change its directions randomly, resulting in extremely negligible hysteresis. By solving the Fokker–Planck equation, Usov and Grebenschikov also obtained minimal hysteresis loop area for cobalt particles in the superparamagnetic regime at $T = 250$ K [58]. The hysteresis loop area gets enhanced as dipolar interaction strength $\lambda$ is increased, irrespective of thermal fluctuation strength $T$. We can explain it by the fact the ferromagnetic coupling increases with $\lambda$ in the linear arrangement of nanoparticles. Mohapatra et al also obtained higher hysteresis loss for the linear array compared to isotropic assembly [59]. Next, we probe the magnetic hysteresis as a function of the size of MNPs ranging from superparamagnetic to ferromagnetic with various values of $\lambda$. In figure 2, we plot hysteresis curves with $D = 8, 12, 16, 24, 32$ and 64 nm at $T = 0$ K. We have also taken four typical values of dipolar interaction strength $\lambda$: 0.0, 0.6, 0.8 and 1.0. The hysteresis follows the Stoner–Wohlfarth particle with $\lambda = 0.0$, as expected [57]. As thermal fluctuations are absent in the present case, the system behaves like a blocked state, irrespective of $D$. Therefore, the hysteresis loop area is significant even for superparamagnetic nanoparticles with negligible dipolar interaction. The hysteresis loop increases with $\lambda$ because of an enhancement in ferromagnetic interaction.

Figure 3 shows hysteresis curves with large thermal fluctuations ($T = 300$ K). All the other simulation parameters are the same as for figure 2. There is a negligibly small hysteresis loop area for $D \approx 8–16$ nm and $\lambda = 0.0$, indicating the dominance of the superparamagnetic character. As far as the role of blocking temperature is concerned for such a behaviour, $T_B$ lies in the range $\approx 17–135$ K for $D = 8–16$ nm, which is well...
below the simulation temperature \( T = 300 \) K. As a consequence, thermal fluctuations dictate the hysteresis of superparamagnetic nanoparticles in the non-interacting case, resulting in extremely weak hysteresis. Our observation for \( D = 8 \) nm is in qualitative agreement with the numerical work of Usov and Grebenshchikov [58]. We could not compare the entire particle size \( D \) range as they are concentrated on \( D = 8 \) nm. The hysteresis loop area is significant for \( D > 16 \) nm even with \( \lambda = 0.0 \). It means that particles with sizes greater than 16 nm show a ferromagnetic character. It is further strengthened by the fact that \( T_B \) is close to 300 K for \( D > 16 \) nm. Therefore, we observe Stoner–Wohlfarth-like behaviour even with \( T = 300 \) K and negligible dipolar interaction (\( \lambda = 0.0 \)). Li et al also obtained a large hysteresis loop area for ferromagnetic nanoparticles even in the non-interacting case using experiments [60]. We could not compare our results in the presence of dipolar interaction and for the whole particle size range as they studied with a single value of particle size and \( \lambda \approx 0.0 \) only. So our results are more generic and can be used as a benchmark in this context. The dipolar interaction increases the hysteresis loop area as it induces ferromagnetic interaction in the system. For the closest packing (\( \lambda = 1.0 \)), there is less dependence of hysteresis on \( D \) because of huge ferromagnetic coupling as dipolar interaction is the maximum. These results suggest that ferromagnetic nanoparticles could be one of the best candidates to achieve significant heat dissipation, essential for magnetic hyperthermia applications. The significant hysteresis for \( \lambda \approx 0.0 \) and \( D > 16 \) nm suggests that heating efficiency can be much improved with ferromagnetic nanoparticles even in the isotropic assembly of MNPs.

It is equally important to probe the effect of external magnetic field strength \( \mu_0 H_0 \) on hysteresis. Figure 4 shows the hysteresis curves for three values of \( \mu_0 H_0(= 0.05, 0.075, 0.1 \) T) with various values of \( D \) and \( \lambda \) at \( T = 300 \) K. The hysteresis loop area is minimal for \( D \leq 16 \) nm and \( \lambda = 0.0 \), irrespective of the external magnetic field strength, indicating superparamagnetic behaviour. There is a significant hysteresis loop area for the ferromagnetic particle (\( D > 16 \) nm) even with small \( \lambda \), which is robust with respect to the field strength. The area under the hysteresis loop also increases with the external magnetic field strength. It is in qualitative agreement with the work of Avugadda et al [61] and Kita et al [62]. The hysteresis loop area increases with \( \lambda \) and \( D \). These results also indicate that we can have considerable heat dissipation with ferromagnetic nanoparticles even in an isotropic assembly and \( \lambda = 0.0 \). So, it can be used for the efficient application of magnetic hyperthermia. These results should also help choose specific magnetic field strength, dipolar interaction and particle size to optimise the heat dissipation, essential for magnetic hyperthermia application.

We then probed the thermal effect on the hysteresis behaviour with the ferromagnetic nanoparticle to probe further. Figure 5 presents the simulated hysteresis curves for \( D = 24 \) nm with six representative values of \( T = 0, 20, 40, 100, 200 \) and 300 K. We have also considered four distinct values of \( \lambda = 0.0, 0.6, 0.8 \) and 1.0. As
It also depends weakly on $T$ with considerable fluctuation. Due to the rise in ferromagnetic coupling with temperature for large $T$, the hysteresis loop area always increases with dipolar interaction strength. These observations also strengthen the fact that the ferromagnetic nanoparticles can have significant heating efficiency, irrespective of the shape of the assembly. To quantify the hysteresis mechanism, we then analyse the coercive field $\mu_0 H_c$ as a function of temperature, particle size $D$ and $\lambda$. In figure 6, we plot the simulated coercive field $\mu_0 H_c$ as a function of $T$ for three values of $D$ ($= 8, 12$ and $16$ nm) and three values of $\lambda$ ($= 0.0, 0.3$ and $0.5$). Large particle sizes $D (> 16$ nm) are not considered, as $\mu_0 H_c$ depends weakly on $T$ in these cases due to the dominance of ferromagnetic character for larger particle sizes. We have also considered only those values of $T$ for a fixed particle size for which $\mu_0 H_c$ is non-zero. It is evident that $\mu_0 H_c$ decreases to zero rapidly with $T$ for very small and negligible dipolar interaction strength. The rapid decrease of $\mu_0 H_c$ with $T$ also agrees well with the work of Usov and Grebenschikov [58]. The comparison is not possible for the whole range of $D$ and $\lambda$ as they have shown results only for $D = 8$ nm with the dilute assembly of MNPs. For moderate strength of magnetic interaction $\lambda$, the $\mu_0 H_c$ decreases very slowly.

It is also quite evident that $T_B$ is a valuable quantity in determining the dependence of hysteresis response on particle size $D$ and dipolar interaction strength $\lambda$.

$T_B \approx 455$ K exceeds the maximum simulation temperature $T = 300$ K considered in the present work. we observe significant hysteresis for relatively smaller $\lambda$ even with a large temperature ($T = 300$ K). The area under the hysteresis curve decreases sharply with $T$ for $\lambda \leq 0.6$. The hysteresis loop area depends weakly on temperature for large $\lambda$, which can be attributed to the enhanced $T_B$ because of long-ranged dipolar interaction. Due to the rise in ferromagnetic coupling with $\lambda$, the hysteresis loop area always increases with dipolar interaction strength. These observations also strengthen the fact that the ferromagnetic nanoparticles can have significant heating efficiency, irrespective of the shape of the assembly. To quantify the hysteresis mechanism, we then analyse the coercive field $\mu_0 H_c$ as a function of temperature, particle size $D$ and $\lambda$. In figure 6, we plot the simulated coercive field $\mu_0 H_c$ as a function of $T$ for three values of $D$ ($= 8, 12$ and $16$ nm) and three values of $\lambda$ ($= 0.0, 0.3$ and $0.5$). Large particle sizes $D (> 16$ nm) are not considered, as $\mu_0 H_c$ depends weakly on $T$ in these cases due to the dominance of ferromagnetic character for larger particle sizes. We have also considered only those values of $T$ for a fixed particle size for which $\mu_0 H_c$ is non-zero. It is evident that $\mu_0 H_c$ decreases to zero rapidly with $T$ for very small and negligible dipolar interaction strength. The rapid decrease of $\mu_0 H_c$ with $T$ also agrees well with the work of Usov and Grebenschikov [58]. The comparison is not possible for the whole range of $D$ and $\lambda$ as they have shown results only for $D = 8$ nm with the dilute assembly of MNPs. For moderate strength of magnetic interaction $\lambda$, the $\mu_0 H_c$ decreases very slowly.

It is also quite evident that $T_B$ is a valuable quantity in determining the dependence of hysteresis response on particle size $D$ and dipolar interaction strength $\lambda$.
al also increases with \( \lambda \) and \( D \). Researchers used eq. (11) with the constant value of \( \alpha \) even for strongly interacting MNPs \([64,67,68]\). Our results suggest that \( \alpha \) should not be taken as constant; instead, it should also be considered a fitting parameter; otherwise, one may estimate incorrect values of various parameters of interest, such as \( T_B \) and \( K_{\text{eff}} \).

Finally, we analyse the variation of \( E_H \) with particle size \( D \) and temperature \( T \) in figure 7. \( D \) has been varied between 8 and 64 nm, and \( T \) has been changed in the range 0–300 K. For weak and negligible \( \lambda \), \( E_H \) decreases rapidly with \( T \) for the superparamagnetic nanoparticle \( (D \approx 8–16 \text{ nm}) \). In contrast, \( E_H \) is enormous when \( D > 16 \text{ nm} \), and it also depends very weakly on \( T \), even in the non-interacting case. It can be explained by the fact that \( T_B \) is comparable to or greater than the maximum simulation temperature \( T = 300 \text{ K} \). As a consequence, MNPs remain blocked even at \( T = 300 \text{ K} \), which results in a large hysteresis loop area and hence \( E_H \) is significantly high. As the dipolar interaction induces ferromagnetic coupling in an anisotropic assembly of MNPs, \( T_B \) increases with \( \lambda \). Therefore, \( E_H \) increases with \( \lambda \) and has a very weak dependence on \( T \). Zubarev et al also found an increase in heat dissipation because of dipolar interaction with ferromagnetic nanoparticles \([69]\). The rapid decrease of \( E_H \) with \( T \) for superparamagnetic nanoparticles with \( \lambda \approx 0 \) also agrees well with the work of Carrey et al \([48]\). We could not compare the non-zero value of dipolar interaction strength \( \lambda \) as they have presented results with \( \lambda = 0.0 \) only. Therefore, the results of our simulations are more generic in this regard. The decrease of hysteresis loop area with temperature for superparamagnetic nanoparticles is also in qualitative agreement with the work of Hu et al \([70]\). Similarly, the variation of \( E_H \) with \( D \) for \( T = 300 \text{ K} \) and \( \lambda = 0.0 \) also agrees with the work of Carrey et al \([48]\). We could not compare our results for the entire range of \( T \) and \( \lambda \) because they presented results with \( T = 300 \text{ K} \) and \( \lambda = 0.0 \) only. The variation of coercive field \( \mu_0H_c \) with \( D \) and \( T \) is the same as the variation of \( E_H \). So, we have not shown the curves for \( \mu_0H_c \) variation to avoid duplications. These observations also agree with the work of Carrey et al for \( \lambda = 0.0 \) \([48]\). These observations are instrumental in choosing correct values of various system parameters to get the desired heat release, which is one of the essential aspects of hyperthermia applications.

### 4. Summary and conclusion

In summary, the magnetic hysteresis obeys the Stoner and Wohlfarth model without thermal fluctuations and magnetic interaction, irrespective of particle size \([57]\). The perfect agreement between simulation and theoretical model also validates the KMC method used. In the presence of sufficient thermal energy, we observe a minimal hysteresis loop area for the smaller particle \( (D \approx 8–16 \text{ nm}) \) and negligible dipolar interaction, indicating the dominance of superparamagnetic character. Remarkably, the hysteresis loop area is significant with ferromagnetic particles \( (D > 16 \text{ nm}) \) and is the same as that of Stoner and Wohlfarth particles even at a high value of temperature and weak or negligible dipolar interaction. Baker et al also observed more substantial hysteresis loss for ferromagnetic nanoparticles of Fe\(_2\)O\(_3\) using experiments \([71]\). Our findings also agree well with the experimental work of Hergt et al \([72]\). The dipolar interaction induces ferromagnetic coupling between the magnetic moments even at a sufficiently high-temperature value. Consequently, the hysteresis loop increases with \( \lambda \) even at \( T = 300 \text{ K} \), irrespective of particle size. These observations are also in perfect agreement with the work of Zubarev \([73]\). The hysteresis loop area is greatly hampered by the thermal fluctuations, which is quite evident with superparamagnetic particles and negligible dipolar interaction. The hysteresis depends weakly on temperature with ferromagnetic nanoparticles even without magnetic interaction. The same trend is observed with superparamagnetic nanoparticles and sufficient dipolar interaction.
The coercive field $\mu_0 H_c$ is an essential quantifier to understand the hysteresis response of these nanosystems. $\mu_0 H_c$ decreases rapidly with temperature for $\lambda = 0$ and smaller particle size $D$. It is due to the rapid transition from ferromagnetic to superparamagnetic state with an increase in thermal fluctuations. Pauly et al. also found similar behaviour of $\mu_0 H_c$ with $T$ in experiments [74]. Because of significant ferromagnetic coupling with considerable magnetic interaction strength, MNPs remain in the blocked state even at tremendous $T$ values for appreciable $\lambda$. Consequently, $\mu_0 H_c$ decays very slowly with thermal fluctuations $T$ for considerable interaction strength. The same trend is observed with ferromagnetic particles, even in the absence of magnetic interaction. In addition to $\mu_0 H_c$, blocking temperature $T_B$ is also a valuable quantifier to analyse the hysteresis characteristics with particle size and dipolar interaction. Therefore, we extracted $T_B$ from the simulated hysteresis curves using Garcia-Otero et al. model [63]. The so-obtained $T_B$ increases with particle size and dipolar interaction strength. Pauly et al. also observed an enhancement in $T_B$ with particle size [74]. Remarkably, $T_B$ exceeds the highest simulation temperature considered in the present work ($T = 300$ K) for ferromagnetic nanoparticles. Consequently, we observe the Stoner and Wohlfarth-like behaviour even at $T = 300$ K and negligible dipolar interaction for such nanoparticles. In general, two distinct regimes are observed depending on the relative strength of $T_B$ and simulation temperature $T$. For $T < T_B$, the energy barriers trap the magnetic moments in metastable orientation, resulting in a blocked state. On the contrary, the thermal energy $k_B T$ overcomes the energy barrier for $T > T_B$ resulting in the well-known superparamagnetic regime. The fitting of simulated values of coercive field $\mu_0 H_c$ with the Garcia-Otero et al. model also suggests that $T^\alpha$ dependence of coercive field with $\alpha = 0.75$ deviates significantly as a function of particle size and dipolar interaction strength. The exponent $\alpha$ is found to increase with an increase in $\lambda$ and $D$. Therefore, our numerical result suggests that $\alpha$ should not be considered constant; otherwise, one may estimate incorrect values of $T_B$ and $K_{eff}$.

To conclude, we have systematically analysed the particle size dependence of magnetic hysteresis in ordered arrays of magnetic nanoparticles using extensive computer simulations. We have also investigated the dependence of hysteresis response on dipolar interaction and temperature. Our results suggest that the hysteresis response of the underlying system depends critically on these parameters. Our results strengthen the fact that ferromagnetic nanoparticles can be one of the best candidates for magnetic hyperthermia irrespective of the shape of the assembly. In these contexts, we have also explained a variety of experimental studies. So, the results presented in this work should also help experimentalists in choosing suitable value of various parameters such as particle size, dipolar interaction strength and magnetic field to obtain desired heating for different applications such as magnetic hyperthermia, drug delivery, etc.

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