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Magnetization Dynamics in Arrays of Quantum Dots

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<http://dx.doi.org/10.5772/intechopen.73008>

Abstract

The possibility of preparing materials based on quantum dots with fine-tuned magnetic properties has opened up the door for designing new and more efficient devices where the interplay of different microscopic phenomena balances out in useful ways. Nevertheless, our knowledge of the precise interaction of complex objects built from a great number of such nanometric magnetic components is still limited. The investigation of the spin or magnetization dynamics in such materials represents an important opportunity to better comprehend and predict some missing pieces for the advancement of a great deal of promising technologies.

Keywords: magnetism, quantum dots, magnetization dynamics, arrays, macrospin

1. Introduction

Since quantum dots were first discovered [1] and later fabricated, they have attracted a great deal of attention given how, just as single atoms or simple molecules, they depict quantum behavior at the level of their electronic and optical properties, but at the same time allow their tuning as a function of their shape, size, and composition—reason that has led some to refer to them as *artificial atoms* [2].

Depending on whether quantum dots are made of a semiconductor, a metal, or another material, different intrinsic properties of a system based on them can be tailored to exhibit specific values or signatures. By themselves, single quantum dots are of prominent relevance and have found applications in diverse scientific and technological fields [3]. Nonetheless, it is really the properties of arrays of these particles which outline a unique landscape in the design space of new materials. Arrays of regularly ordered *magnetic quantum dots* (MQDs) provide an opportunity to develop materials with characteristics different from those exhibited by traditional solid-state systems.

Arrays—and more generally assemblies—of MQDs comprise all of those systems in which the magnetic nanoparticles are embedded in, dispersed into, or arranged over a different nonmagnetic medium—either a liquid or a solid, e.g., some polymeric material. More concisely, all of them could be simply classified as magnetic nanocomposite or hybrid materials, which depending on the spatial relative placement of the MQDs can form one-, two-, or three-dimensional architectures [4, 5].

Assemblies of MQDs are attractive for their rich spectra of potential technological applications that range from biomedical uses such as magnetic resolution imaging, magnetic hyperthermia, and drug delivery [3–7] to magnetic refrigeration and energy-harvesting devices [8]. Notwithstanding, the major driving forces behind the study of arrays of MQDs are related to the area of information storage technologies [9, 10]. In particular, there is a great hope of achieving through these magnetic storage media of ultrahigh densities [11–13], nonvolatile magnetic random-access memories (MRAM) [14, 15], and logic devices [16–18]. Moreover, the importance of these systems does not stop here; arrays of MQDs are also of special interest as model systems for better understanding interactions and transport processes of magnetic materials in general [19–22].

In this chapter, we present the results from several—mainly theoretical and numerical—works focused on the analysis of magnetization dynamics and interactions of a representative variety of two-dimensional systems of MQDs arrays. Throughout each of the sections, we will review some of the physical models that are used to study, predict, and understand the dynamics and how interparticle interactions affect the overall behavior of these systems. We will also take the opportunity for introducing some of the limitations that have hindered progress on the realization of some of the applications mentioned above and will discuss based on the aforementioned works what possible strategies could be followed in order to steer away the status quo in the field.

While it is not the goal of the current exposition to dive into the experimental aspects that entail the fabrication of arrays of MQDs, it is worth mentioning that numerous efforts have been devoted to the development of different synthetic pathways for MQD arrays, as it can represent a highly challenging task to prepare some of these systems in such a way that the targeted properties of the system are obtained within a desired precision in a predictable fashion. Indeed, at the characteristic length scale of MQDs, i.e., down to less than ten to a few hundreds of nanometers, variations on the shape, size, and distribution of MQDs can significantly impact the magnetic behavior of the whole array.

2. Theoretical modeling of the magnetization dynamics

In general, the starting point for the description of the magnetization dynamics of arrays of MQDs is to consider the relevant fields that modify the magnetization of each individual QD. If we denote the total effective field “felt” at every point in space as $\vec{H}_{eff}(\vec{r})$, then the equation of motion of the magnetization $\vec{m}(\vec{r})$ is the Landau-Lifshitz-Gilbert (LLG) equation [23, 24]:

$$\frac{d\vec{m}(\vec{r})}{dt} = -\gamma\vec{m}(\vec{r}) \times \vec{H}_{eff}(\vec{r}) + \frac{\alpha}{M_s} \left(\vec{m}(\vec{r}) \times \frac{d\vec{m}(\vec{r})}{dt} \right), \quad (1)$$

where γ is the gyromagnetic ratio, M_s is the saturation magnetization of the material, and α is the viscous damping coefficient [25]. Here, the magnetization $\vec{m}(\vec{r})$ is normalized over M_s .

Equation (1) can be solved analytically in very few cases and needs, in general, to be integrated numerically, which is usually done by recasting it into the following form:

$$\frac{d\vec{m}(\vec{r})}{dt} = -\frac{\gamma}{1+\alpha^2} \left\{ \vec{m}(\vec{r}) \times \vec{H}_{eff}(\vec{r}) + \frac{\alpha}{M_s} \left[\left(\vec{m}(\vec{r}) \cdot \vec{H}_{eff}(\vec{r}) \right) \vec{m}(\vec{r}) - \vec{H}_{eff}(\vec{r}) \right] \right\}, \quad (2)$$

also referred to as the Landau-Lifshitz equation or explicit form of LLG equation.

The total effective field $\vec{H}_{eff}(\vec{r})$ should be specified for each system of interest, but for most of the systems, it includes magnetic anisotropy contributions, interparticle or substrate-particle interactions, and external field terms.

Depending on the nature, shape, and size of the MQDs, there are two general approaches to solve Eq. (2). The first one, *micromagnetics*, [24] describes the spatial magnetization distribution in the regime where the MQDs are large enough for domain walls and vertices to appear. In this scenario, the interactions between pairs of intra-dot domains render quite involved and time-consuming the computational task of solving the LLG equation, requiring highly technical numerical expertise or specialized software to handle the computational complexity. The second approach, called *macrospin model* [24, 26], is appropriate to describe the magnetization time evolution of assemblies of small nanoparticles, such that each dot magnetization can be described by a single magnetic moment within the QD volume. What we mean by small is unfortunately system dependent, but some known data for spherical MQDs can be found in **Figure 1** as reference.

In the macrospin limit, the magnetization distribution of an array of MQDs becomes $\vec{m}(\vec{r}) = \sum_i \vec{m}_i \delta(\vec{r} - \vec{r}_i)$, where \vec{m}_i denotes the monodomain magnetization of the i th particle in the array and $\delta(\vec{r} - \vec{r}_i)$ is the Dirac delta function. Plugging the above expression for $\vec{m}(\vec{r})$ into Eq. (2), we obtain

$$\frac{d\vec{m}_i}{dt} = -\frac{\gamma}{1+\alpha^2} \left\{ \vec{m}_i \times \vec{H}_{eff}(\vec{r}_i) + \frac{\alpha}{M_s} \left[\left(\vec{m}_i \cdot \vec{H}_{eff}(\vec{r}_i) \right) \vec{m}_i - \vec{H}_{eff}(\vec{r}_i) \right] \right\}, \quad (3)$$

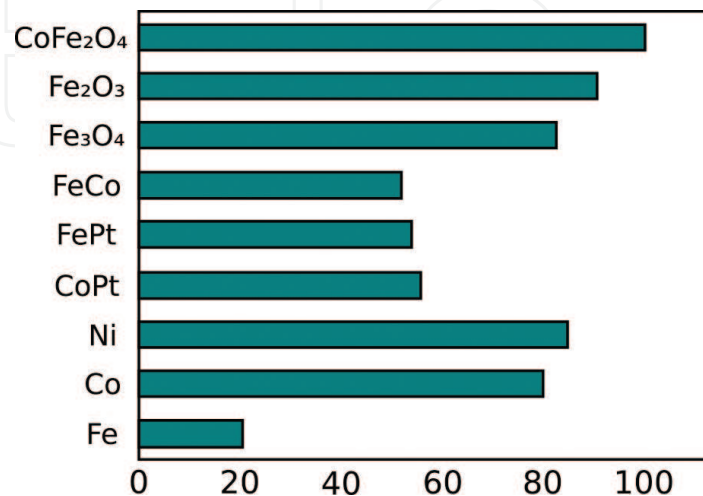


Figure 1. Threshold diameters for maximum monodomain size for spherical nanoparticles. Adapted from Majetich et al. [27].

which is an i -dimensional nonlinear system of ODEs whose solutions can be approximated by traditional numerical integration schemes, e.g., Heun's or Runge-Kutta methods.

Within the same limit, the total effective field \vec{H}_{eff} can be usually written as

$$\vec{H}_{eff} = \vec{H}_{ani} + \vec{H}_{dem} + \vec{H}_{int} + \vec{H}_{ext} \quad (4)$$

that is, the total field is the sum of the contributions from the magnetocrystalline anisotropy \vec{H}_{ani} , the demagnetizing field \vec{H}_{dem} , the particle interaction term \vec{H}_{int} , and the applied external magnetic field \vec{H}_{ext} .

Depending on the crystal structure of a ferromagnetic material, one or more privileged axes—known as *easy axes*—that energetically favor the alignment of the magnetization along their direction may exist. This energy contribution results from spin-orbit interactions and is referred to as magnetocrystalline anisotropy energy.

The demagnetization energy is the energy of the magnetization in the magnetic field created by the magnetization itself. This means that this energy contribution accounts for the dipole-dipole interaction of the elementary magnets.

For MQDs modeled as macrospins and in the absence of other interactions, the field \vec{H}_{int} is simply the dipolar field induced by all of the particles, that is

$$\vec{H}_{int}(\vec{r}_i) = -\frac{VM_s}{4\pi} \sum_{j \neq i} \left(\frac{\vec{m}_j}{r_{ij}^3} - 3 \frac{(\vec{m}_j \cdot \vec{r}_{ji}) \vec{r}_{ji}}{r_{ji}^5} \right). \quad (5)$$

For the micromagnetic description of a system, Eq. (4) should also include the quantum-mechanical exchange interaction.

3. The effect of the geometry of MQD arrays

At this point, it should be clear that when analyzing the magnetic behavior of MQD arrays, one must take into account a lot of different factors. Just to mention a few, let us consider both the chemical composition and the crystalline structure of the nanoparticles themselves, as these factors determine—among other things—the magnetic anisotropy of each MQD; the shape of a single QD gives rise to a demagnetizing field; the spatial distribution among particles in an array establishes how particles will interact via pairs of dipole-dipole potentials; and, equally, any external factor that modifies the array, e.g., some applied external field, will impact the time response of the system.

Given the wide variety of contributing factors to the global magnetic properties of an array of MQDs, let's restrict our attention first to just one of them: the geometric distribution of particles in a two-dimensional assembly.

3.1. Single-domain limit

The possibility to synchronously manipulate the motion of all the magnetic moments in a cluster of magnetic nanoparticles in order to attain the fastest dynamic response in the presence

of an external field while achieving some stability for the magnetic moment at the same time has motivated the study of the influence of the geometrical parameters—such as the inter-dot separation and crystal structure of arrays of MQDs—on the magnetic collective behavior of such systems.

In 2014, Meza et al. [28] reported an analysis of the coercivity fields for two-dimensional clusters of ellipsoidal cobalt nanoparticles in two different crystalline configurations (square and hexagonal) as a function of the inter-dot spacing and the frequency of a switching continuous external applied field. For this work oblate nanoparticles of semiaxis lengths of $3 \text{ nm} \times 3 \text{ nm} \times 1.5 \text{ nm}$ were chosen. The easy axis of the particles, oriented in plane, was chosen parallel to the boundary of the cluster, and the external field was applied along the same direction. Given that these MQDs lie in the monodomain regime for cobalt, it is safe to assume that they can be properly described by the macrospin model.

By simulating hysteresis cycles for small clusters of cobalt nanoparticles (3×3 , 5×5 , and 10×10) at different frequencies for the applied oscillating external field (**Figure 2**), it was found that the hexagonal configuration stabilizes the magnetization reversal and narrows the coercivity with respect to the square crystal structure; this is clearly a consequence of the interaction strength promoted by the greater number of the nearest neighbors.

For both crystal systems, it was also observed that as the cluster size gets bigger the coercivity narrows at all frequencies and it is much lower than the coercivity for a single MQD. This is

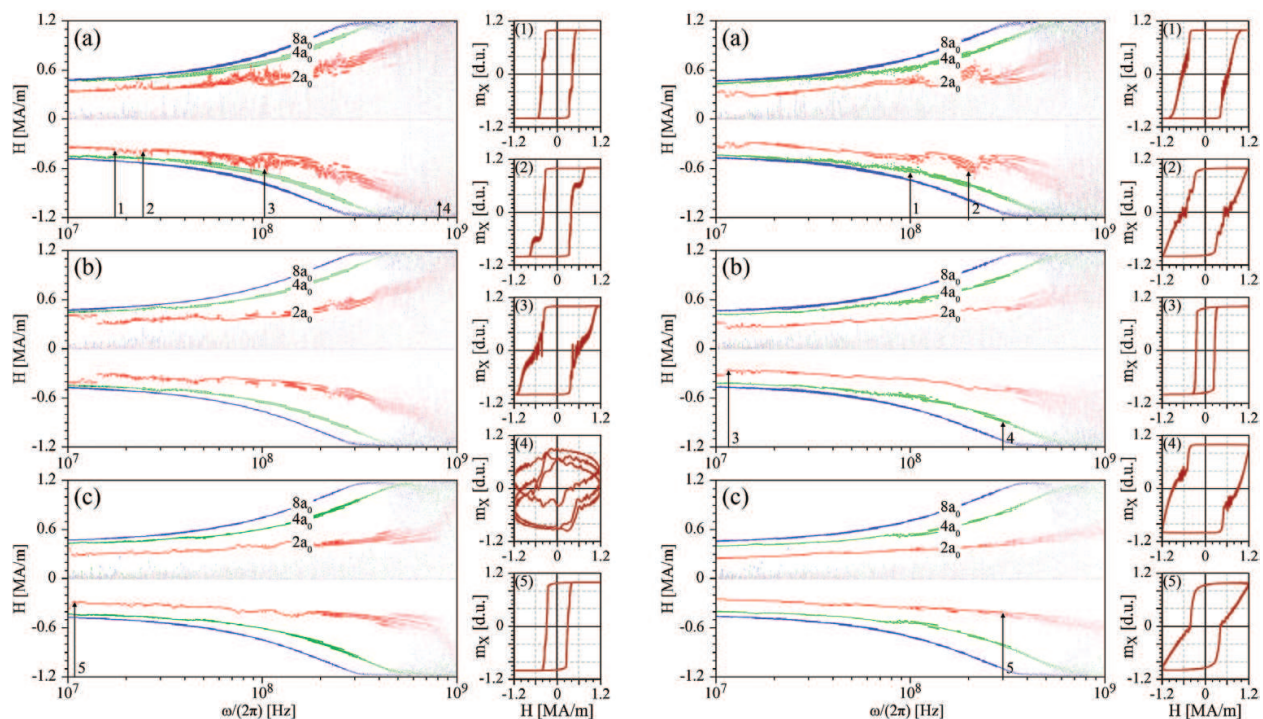


Figure 2. Coercivity histograms for macrospins arranged in a square (left) and hexagonal (right) lattice with different numbers of particles: (a) 3×3 , (b) 5×5 , and (c) 10×10 . The distance between the particles is marked as $d = 2a_0, 4a_0$, and $8a_0$ (see [28]).

an interesting finding that could be exploited by trying to switch the magnetization only for of small subgroups of MQDs from a bigger array without affecting the rest of the magnetic system.

In all cases, by increasing the proximity of the nanoparticles, i.e., with a tighter packing of the assembly, and the intensity of the dipole interactions becomes more relevant, this in turn stabilizes the reversal at higher frequencies; so, in a macrospin system, the minimum of the switching time for a given frequency of an oscillating external field is limited by the packing of the nanoparticles.

As an additional note, it is noteworthy that experimentally, it is certainly possible to synthesize magnetic nanoparticles of these sizes by chemical methods. Nevertheless, the fabrication of colloidal nanoparticles and its self-assembly via solvent evaporation cannot yet attain the level of control that would be required to fabricate small uniform clusters of the sizes studied. On the other hand, lithographic techniques that allow a more refined control over the position, sizes, and number of particles in an array are still far from reaching the length scales of the system proposed.

3.2. Micromagnetics

To contrast the time evolution of a single-domain small-sized array, let us now consider the work done by Semenova et al. in 2013. In [29], the quasistatic hysteresis of close-packed arrays of NiFe nanodisks is studied. The motivation of this work is quite different from the one presented above and relates to the desire of understanding the phenomena in the field of *magnonics*, where there is a great interest in better grasping the propagation and confinement of spin waves in (among other materials) ordered assemblies of MQDs, which would have potential applications in the fabrication of reprogrammable crystals, magnetoelectronic devices, and metamaterials to name a few.

Although part of the work is focused on obtaining the spin-wave excitation spectra on the arrays of MQDs, we will only discuss the results on the role of the array geometry and packing over the hysteresis loops measured.

The specific set of MQD assemblies studied consisted in two two-dimensional hexagonal arrays of $\text{Ni}_{80}\text{Fe}_{20}$ nanodisks prepared by etched nanosphere lithography. The disks were of near 350 nm in diameter with an edge-to-edge separation of 65 and 12 nm in average for the two arrays, respectively. Accordingly, there is one tightly packed array, and another one is almost closed-packed array.

The system analyzed requires resolving over much larger distances than it is customary, making the problem of numerically predicting the magnetization dynamics a challenging exercise—mainly because of the complexity required to adequately consider periodic boundary conditions for the long-range interactions of the QDs.

The experimental results and magnetic simulations in [29] indicated that the overall hysteretic behavior in the system is dictated by the nucleation and escape of vortices within each nanodisk. Indeed, the observed sudden drop (see **Figure 3**) in the magnetization near an applied null external field is attributed to the formation of vortices near the center of the QDs; eventually, as the applied field pushes the system toward saturation, it manages to move around the center of every vertex to the edge of its disk. Similar phenomenology has been observed in square arrays of MQDs with some degree of anisotropy on the vortex nucleation [30, 31].

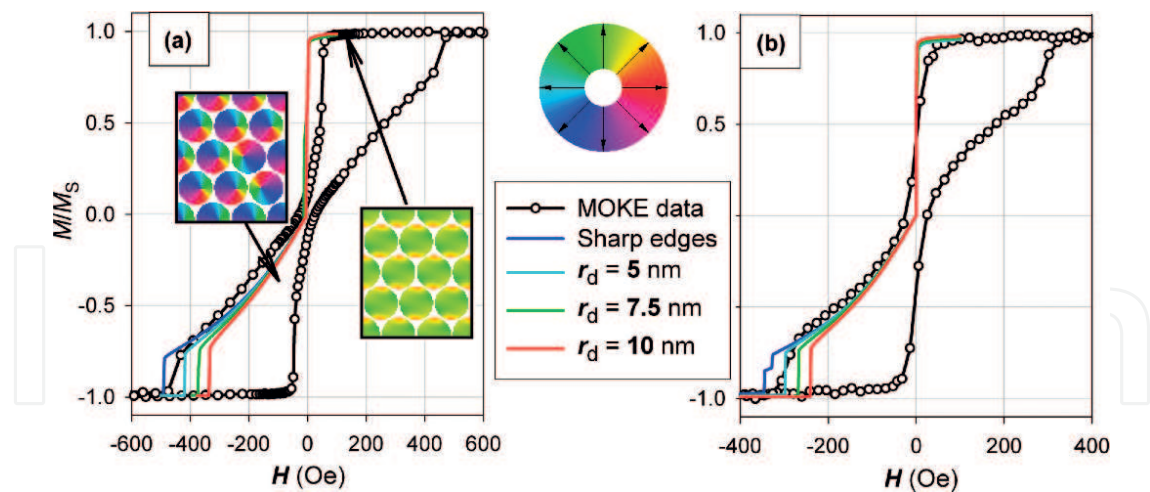


Figure 3. Simulated and measured hysteresis loops for the (a) the tightly packed array and (b) the almost closed-packed array from [29].

In the assemblies described above, the dipolar interactions play a less important role than the internal micromagnetics of each particle near almost null applied fields, and to some extent, the inter-dot dipolar interaction can be thought in an effective way contributing only to the displacement of the vertices out of the disks.

The lesson learned so far is that the net effect of the geometry of arrays of MQDs is of greater importance for monodomain systems where one of the dominant interactions is the dipolar one—for sufficiently packed systems—and that it enters in a much less important fashion for large systems where intra-dot micromagnetics dominate the magnetization reversal properties.

3.3. In between domains

To conclude the section, let us look one final assembly of MQD where the particle's size is of the order of other multi-domain nanostructures but for which the array geometry and the macrospin description might play an important part.

In 2010, Redondo et al. [32] reported experimental observations of the magnetization reversal mechanism for arrays of equally irregularly shaped MQDs aligned and distributed over an ordered square grid. The research team found that for different directions of an applied external field, the magnetization reversal would behave in some cases as if the switching mechanisms were dominated by nucleation and displacement of vortices, but in other directions, for the same system, the behavior would be that of a macrospin array.

For such systems one of the demagnetizing fields or the exchange interactions will benefit from the presence of the external field to balance out the contention.

Conflict of interest

The authors declare no conflicts of interest.

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