

## Research articles

## Dipolar magnetic interaction effects in 2D hexagonal array of cobalt hollow-spheres

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## ABSTRACT

Planar arrangements of cobalt hollow-spheres were studied by means of micromagnetic simulation. The calculated coercivity values are in correspondence with the reported experimental data. Dipole energy effects are determinant and more significant if thickness decreases. We observed the formation of some vortex and onion configurations, solutions for individual hollow-sphere, even so there is predominance of non-homogeneous reversal. This confirms that solutions for individual spheres are not efficient in the analysis of arrays.

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## 1. Introduction

Several magnetic studies on 2D and 3D arrangements of hollow-spheres (H-S) have been made motivated by their technological applications [1–13]. H-S of cobalt has been obtained using various chemical methods [14–19]. Chains of submicron Co hollow-spheres were obtained using a simple soft-assembly strategy [17–19]. Duan et al. [20] and Hu et al. [21] prepared a 2D array of inorganic H-S through deposition onto conductive substrates, combined with thermal treatment and subsequent solution-dipping and electrochemical deposition. In many experimental studies the magnetic properties of H-S arrays are explained on the basis of single sphere behavior. Reversal of magnetic moments in arrangements, is affected by the strong dipolar and exchange interactions [13,22]. Our previous papers present the micromagnetic simulation as an efficient tool for studies in arrangements for Ni hollow-spheres, due to the high complexity of these systems [22]. In the present work we study the magnetic properties of a 2D hexagonal array of connected hollow-spheres of Co, motivated by the competition of different magnetic anisotropies (shape and magneto-crystalline). It was using the micromagnetic simulation with Object Oriented Micromagnetic Framework (OOMMF), based on the finite difference method (FDM) [23].

## 1.1. Micromagnetic model

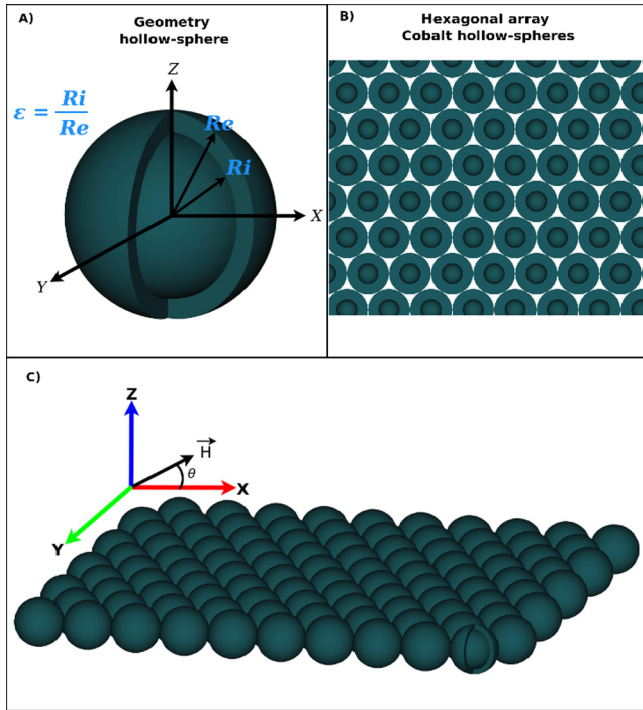
In the micromagnetic model, the magnetization vector field has absolute value at each point equal to spontaneous magnetization of the material,  $M_s$  ( $M = M_s m$ ,  $m \cdot m = 1$ ). The effective field,  $H_{eff}$  is the functional derivative of magnetic energy density,  $E$  [24,25];  $H_{eff} = \frac{1}{\mu_0 M_s} \frac{\delta E}{\delta m}$ . In the present work were included the energetic contributions from exchange interactions, magneto-crystalline anisotropy, external applied field or Zeeman, and demagnetizing energy densities;  $E = E_{exchange} + E_{demag} + E_{crystall} + E_{Zeeman}$ . Magnetization dynamic is described by the Landau–Lifshitz–Gilbert (LLG) equation [24];  $\frac{dm}{dt} = -\gamma_0 (m \times H_{eff}) + \alpha [m \times \frac{dm}{dt}]$ . Here,  $\gamma_0 = 2.211 \times 10^5 \text{ mA}^{-1} \text{ s}^{-1}$ , is the Gilbert gyromagnetic ratio and  $\alpha$  is the Gilbert damping constant.

The easy-axis direction of magneto-crystalline anisotropy was oriented perpendicular to the array plane. For better understanding, the geometry of a hollow-sphere (H-S) with external radius  $R_e = 20 \text{ nm}$  is displayed in Fig. 1A). The thickness ( $R_e - R_i$ ) was systematically changed according to the ratio  $\varepsilon = R_i/R_e$  ( $0 \leq \varepsilon \leq 0.8$ ), with  $R_i$  as the inner radius. For our calculations a 2D hexagonal array constituted by 100 H-Ss of Co connected by the exchange interactions, was used, Fig. 1B). Hysteresis curves were calculated by the minimization of the total energy density by a conjugate gradient algorithm, using the Fletcher–Reeves method implemented in the minimization evolver of OOMMF [23].

The external magnetic field was applied in different directions in relation to the plane of the arrangement, as shown in Fig. 1C). The dynamic of magnetization was simulated by solving the LLG

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**Fig. 1.** A) Geometry for a cobalt hollow-sphere (H-S), showing the inner and outer radii as the principal geometric parameters used in the study here. B) Cross-section of the hexagonal array of cobalt H-S in 2D configuration. C) The external magnetic field applied in different directions, relative to the plane of the array.

equation, through the time evolver implemented in OOMMF, integrating the equations by the Runge-Kutta-Fehlberg method [26,27]. The typical magnetic parameters of cobalt were used [11]: saturation magnetization  $M_s = 1400 \times 10^3$  A/m, exchange stiffness constant  $A = 30 \times 10^{-12}$  J/m, magneto-crystalline anisotropy constants  $K_1 = 400 \times 10^3$  J/m<sup>3</sup> and damping constant  $\alpha = 0.013$ . According to our computational capacity, the cell size was  $4 \text{ nm} \times 3.35 \text{ nm} \times 4 \text{ nm}$ . This applies for all cell sizes and is smaller than the exchange length for cobalt,  $L_{\text{ex}} \sim 5 \text{ nm}$ . The array dimensions were  $420 \text{ nm} \times 351 \text{ nm} \times 40 \text{ nm}$  and the mesh was with a total of 110250 computational cells.

## 2. Results and discussion

Our initial calculations were oriented to determinate the magnetization curves applying the external magnetic field in different directions, relative to the plane of the array ( $0 \leq \theta \leq 90^\circ$ ) as shown in Fig. 1C). It is motivated because magnetization curves are a fundamental and primary measurement indicating the global properties of any magnetic system, this measure is presented in all experimental works. It is important to remember that our work is motivated by researching the reversal of magnetic moments in arrays in order to understand the role of the known modes in a single cobalt hollow-sphere (H-S) [11–13,28]. For all calculations the external field was varied slowly from 13 kOe until  $-13 \text{ kOe}$  and then back to 13 kOe, to close the thermodynamic cycle. The calculations were using an external field shift step of 80 Oe. Hysteresis curves were performed by plotting the projection of magnetization along the direction of the external field versus the value of the applied field.

Fig. 2 presents the calculation for the applied field perpendicular ( $\theta = 90^\circ$ ) and parallel ( $\theta = 0^\circ$ ) to the array. These calculations were made for different thicknesses in order to verify the effects of this geometric modification on the magnetic properties. There is the manifestation of the competition between two anisotropies.

The magnetization in direction perpendicular to the plane of the arrangement ( $\theta = 90^\circ$ ) is always favored by the easy-axis of magneto-crystalline energy with a contribution depending on the material and the orientation of magnetic moments in all points of ferromagnetic volume. On the other hand, there is always the auto-demagnetizing energy of the overall arrangement. This contribution is from the inter-spheres dipolar interaction and depends on the magnetization vector field configuration. In conjunction with this last contribution the self-demagnetizing energy with dipolar origin is present, and is originated from the interaction between decomposed dipoles on the inner and outer surfaces of each hollow-sphere (H-S). It should be noted that this contribution is the one that most changes during the study due to the different thickness of the spheres used. It should also be pointed out that uncompensated dipoles on the surface depend on the way the moments are oriented in each field situation applied. It is also known that in the hollow-sphere, the demagnetizing field energy depends on the square of the thickness [29]. A decrease in thickness decreases the demagnetizing energy which favors the onion configuration. For connected spheres, there will be the contribution that modifies the already known solutions for a single and isolated sphere. In the case of the H-S arrangement, the configuration of the moments during the reversion will no longer be the same as for an isolated sphere, due to the strong dipole interactions between the H-S. In this way the 2D array will tend to behave like a film but otherwise there will be the already mentioned inter-surface interaction for each sphere together with magneto-crystalline energy. The orientation of the moments tends to be parallel to the plane of the arrangement and the magneto-crystalline energy tries to force moments into the perpendicular direction. In addition thinner H-S favors the orientation of the moments in the direction parallel to the surface of each sphere. Because the distances presented here are a few nanometers, the dipole energy is very strong compared to magneto-crystalline and the competition is evident. On the other hand, the moments are no in coherent configuration (uniform mode) and the result is that in a global view all factors favor the direction perpendicular to the plane of the arrangement. This causes a widening of the hysteresis cycle as the thickness of the spheres decreases.

The  $\varepsilon$  value is related to the hollow-sphere stiffness and a great value for this parameter corresponds to a thick hollow-sphere. From Fig. 2 in curves with the field applied parallel to the arrangement there is a rapid initial increase and then a second stage of increase in magnetization. In the first stage we see in general the effects of the 2D arrangement with global behavior characteristic of the film. In this way the momentum grows rapidly along the direction parallel to the plane of the arrangement. The profile of the magnetization curves for very low fields is modified as the hollow-spheres become increasingly thinner i.e. when the value of  $\varepsilon$  increases. In the second stage of the curve the competition between the dipole energy coming from the interaction between the inner and outer surfaces of each sphere and the rest of the energies is manifested. For  $\varepsilon = 0.1$  a rapid growth of the magnetization value and then a change in derivative until 2500 Oe occurs. It is due to the difficulty of the alignment of the magnetic moments due to the effect of the magneto-crystalline easy-axis direction, perpendicular to the array. For  $\varepsilon = 0.3$  the energetic competition is more evident because the inner and outer surface of all individual H-S are closer, then magnetostatic energy increases and becomes significant in the magnetization process. The reversion of magnetic moments for  $\varepsilon = 0.6$  is a new point in the discussion because the magnetic moments are now confined in the thick hollow-sphere and the reversion is directly affected. For  $\varepsilon = 0.8$  we have other regime because the diminution of the stiffness has a direct influence over the dynamic of magnetic moments in the material.

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