



Evolution of magnetism on a curved nano-surface

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To design custom magnetic nanostructures, it is indispensable to acquire precise knowledge about the systems in the nanoscale range where the magnetism forms. In this paper we present the effect of a curved surface on the evolution of magnetism in ultrathin iron films. Nominally 70 Å thick iron film was deposited in 9 steps on 3 different types of templates: a) A monolayer of silica spheres with 25 nm diameter b) a monolayer of silica spheres with 400 nm diameter and for comparison a flat silicon substrate. The in-situ iron evaporation took place in an ultrahigh vacuum chamber using molecular beam epitaxy technique. After the evaporation steps, time differential nuclear forward scattering spectra, grazing incidence small angle X-ray scattering images and X-ray reflectivity curves were recorded. In order to reconstruct and visualize the magnetic moment configuration in the iron cap formed on top of the silica spheres, micromagnetic simulations were performed for all iron thicknesses. We found a great influence of the template topography on the onset of the magnetism and on the developed magnetic nanostructure. We observed individual magnetic behaviour for the 400 nm spheres which was modelled by vortex formation and collective magnetic structure for the 25 nm spheres where magnetic domains spread over several particles. Depth selective nuclear forward scattering measurements showed that the formation of magnetism begins at the top region of the 400 nm spheres contrary to the 25 nm particles where the magnetism first appears in the region where the spheres are in contact with each other.

1 Introduction

The immense spread of internet and cloud computing requires a constant development of data storage devices. Nowadays, discoveries such as giant magnetoresistance (GMR)^{1,2}, tunneling magnetoresistance (TMR)^{3,4} and innovations like dynamic flying height (DFH)⁵ or perpendicular magnetic recording (PMR)⁶⁻⁸ made possible the continuous increase of stored information on a unit surface area. However, exceeding 1 Tbit/inch² storage density is not possible with the latter techniques⁹ hence further developments are essential. The next generation of high density recording technologies such as thermally assisted recording (TAR)^{10,11} combined with bit patterned media (BPM)^{12,13} extends the theoretical storage limit as high as 20 Tbit/inch²¹⁴. In order to develop efficient BPM devices, there is strong demand for nanosystems with ordered structure. Several methods, like electrodeposition¹⁵, etching¹⁶, self assembly¹⁷⁻²¹, electron-beam lithography^{22,23} and others²⁴⁻²⁶ are known for producing highly ordered nanostructures. Self-assembled spherical nanoparticles

prepared by Langmuir-Blodgett technique^{27,28} are great candidates for future BPM devices due to their cost effective and large scale preparation. Because of the curved surface, magnetic thin films on such a spherical template exhibit unusual properties. Several theoretical and experimental studies have been reported discussing this phenomenon. It has been shown that a Co/Pd multilayer on top of self-assembled spherical particles can exhibit radial symmetric anisotropy orientation²⁹. On the same system, serpentine-like magnetic domain patterns were observed reflecting the topographic structure³⁰. Chirality frustrated vortex states were found in soft magnetic permalloy caps induced by the reduced coupling between particles³¹. Anisotropic magnetostatic interaction was evidenced in case of rolled nanomembranes induced by thickness gradient³² and also several magnetic simulations were done to study magnetism in curved surface³³.

To have a better understanding of such systems it is indispensable to investigate magnetic thin films in the thickness range where magnetism starts to evolve. To avoid external effects in-situ experiments are needed, which have been great challenges due to the dedicated sample environment. Such experiment has been performed by K. Schlage and co-workers where the formation of magnetism of iron on highly ordered vertical cylinder template was studied³⁴.

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In this work, a comprehensive characterization using the combination of in-situ and ex-situ tools, i.e., time resolved nuclear forward scattering (NFS), grazing incidence small angle X-ray scattering (GISAXS), X-ray reflectometry (XRR), atomic and magnetic force microscopy (AFM, MFM), field emission scanning electron microscopy (FESEM), and various calculation methods – vertical susceptibility profiles, micromagnetic simulations – has been applied to understand the onset and evolution of magnetism appearing on top of spherical templates. Our study can help to explore the potential and the limits of the formation of magnetic nanostructures in the applied size range of the nanoparticles, interparticle distances and magnetic layer thicknesses.

2 Experimental and methods

Single monolayer of silica spheres with nominal diameter of 25 nm and 400 nm were prepared on top of silicon wafers using Langmuir-Blodgett technique with controlled hydrolysis of tetraethyl-orthosilicate (Stöber's method³⁵). The nanosphere-film deposition was carried out in the Centre for Energy Research, Institute of Technical Physics and Materials Science in Budapest by KSV2000 film balance using vertical deposition method. The (100)Si substrates were cleaned with 2% HF solution, rinsed with de-ionized (Milli-Q) water and dried at room temperature. The alcohols were diluted by chloroform at a volume ratio of 1:2. The sol samples were homogenized for 10 min in an ultrasonic bath and spread onto the water surface in the film balance. After evaporation of the spreading liquid the layer was compressed at a rate of 40 cm²/min. The surface-pressure of the layer was monitored with the Wilhelmy-plate method. At ca. 80% of the collapse pressure of the sample, the surface-pressure of the sample was kept constant for 15 min. Afterwards the monolayer was deposited on the substrate with a withdrawal speed of 5 mm/min. The films were dried at ambient temperature. The Langmuir-Blodgett monolayers were analyzed by Field Emission Scanning Electron Microscopy (FESEM) using a LEO 1540 XB cross-beam system. The size distribution analysis of

the nanospheres resulted in 26 nm and 398 nm median diameters with full width at half maximum of 11 nm and 54 nm, respectively (Figure 1). In case of the 400 nm particles, the mean interparticle distance was found to be 63 nm with FWHM 50 nm. The local hexagonal symmetry of the spheres vanishes on long scale and the hexagonal cells start to arrange in twisted pattern^{36,37}. This feature is the consequence of the size distribution of the silica spheres and the domain-like structure of the deposited monolayer. The 25 nm particle template consists of closely packed domains and a smaller amount of particle free regions. Since the closely packed domains give ~90% of the full coverage, we consider an interparticle distance of 0 nm indicating that the particles are in direct contact.

Nominally 70 Å thick ⁵⁷Fe layer was evaporated in 9 steps, on top of the nanospheres as well as on a flat silicon substrate. The deposition was carried out using molecular beam epitaxy (MBE) technique in the UHV³⁸ system at the nuclear resonant beamline ID18³⁹ at European Synchrotron Radiation Facility (ESRF). Prior to deposition, the wafers were annealed at 500 °C for 30 minutes in order to remove any surface contamination. ⁵⁷Fe was evaporated from a rod using an Oxford Applied Research 4-pocket electron-beam evaporator which was mounted on the nuclear resonant scattering (NRS) chamber³⁶ of the UHV system (Figure 2). The base pressure was 8×10⁻¹¹ mbar which increased to 1×10⁻¹⁰ mbar during evaporation. After each deposition step, in-situ time differential nuclear forward scattering (NFS) spectra and X-ray reflectivity (XRR) curves were recorded using a 32 element avalanche photo diode detector system with sub-nanosecond time resolution and grazing incidence small angle X-ray scattering (GISAXS) images were acquired utilizing a standard ESRF MAXIPIX⁴⁰ image plate. The experiment was performed at 14.4 keV, the excitation energy of the ⁵⁷Fe nucleus, using a high resolution monochromator with 2 meV energy resolution.

After the synchrotron radiation experiment the sample was covered by 5 nm niobium to avoid surface oxidation. The magnetic structure, together with surface topology were studied using atomic force microscopy (AFM) and magnetic force microscopy (MFM) at Katholieke Universiteit Leuven. The magnetization measurements were carried out with a Dimension 3100 scanning probe microscope (Bruker Inc.) operated in the tapping/lift mode using commercial MFM probes with tips magnetized along the tip axis. The MFM

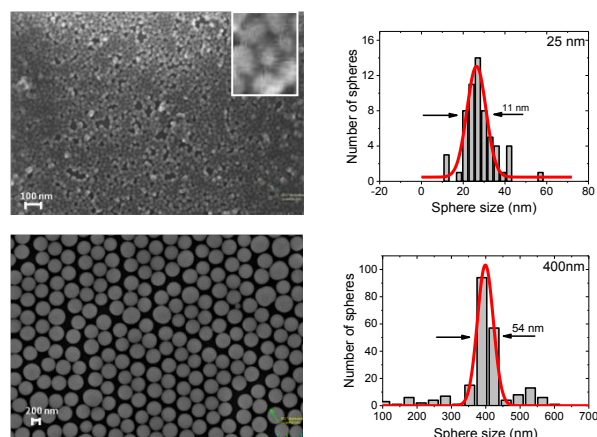


Figure 1 FESEM image of 25 nm (top) and 400 nm (bottom) diameter silica spheres deposited on silicon substrate (left) with the results of size distribution analysis (right).

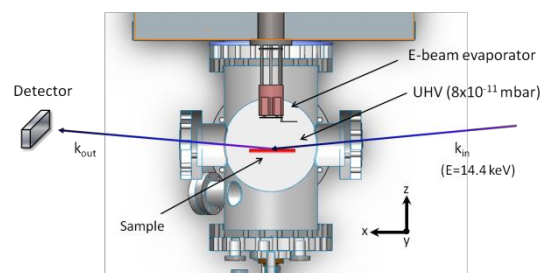


Figure 2 The arrangement of the experiment. The sample situated in a UHV chamber where the electron-beam evaporator was mounted and the synchrotron radiation enters and exits through beryllium windows.

measurements presented in this article are performed at a lift mode of operation, i.e. using the dual pass technique across each scan line and the frequency of the cantilever was set at its resonant frequency (~ 60 kHz). During the first pass AFM data is taken. Then the tip is raised to the lift scan height (typically 20–50 nm) and a second scan is performed while maintaining a constant separation between the tip and the local surface topography. This allows obtaining information about the overall domain structure from an MFM image. The comparison of an MFM image with the corresponding AFM image is able to establish correlations between the domain structure and the topographic data.

Both the AFM, MFM and also FESEM images were evaluated using Gwyddion⁴¹ and WSxM⁴² software.

The magnetic moment configuration was studied by micromagnetic simulations. We used NMAG⁴³, which is an open source, python based software, in order to perform the finite element calculations based on the Landau-Lifshitz and Gilbert equation⁴⁴. The obtained simulations were visualized using ParaView⁴⁵, an open-source, multi-platform data analysis and visualization application.

The evaluation of the XRR data was done by using the program FitSuite⁴⁶, which calculates the reflectivity based on differential propagation matrices⁴⁷.

The GISAXS simulations were done by using BornAgain, an open source, multiplatform software package⁴⁸ which utilizes Distorted Wave Born Approximation (DWBA) in the physical description of the scattering process.

Results and discussion

In order to study the evolution of magnetism in the deposited iron layer, first the growth mechanism should be investigated. XRR curves were recorded after each deposition step on the 25 nm and 400 nm diameter silica nanospheres and on the flat silicon substrate (Figure 3c–e). The red line corresponds to the fitted data (black circles) and the numbers indicate the determined iron thicknesses after each evaporation step.

In our model the monolayer of nanospheres were divided into ten sublayers and an average susceptibility χ was assigned to each of the sublayers (Figure 3a). The susceptibility can be expressed as $\chi = (4\pi N/k^2)f$ and related to the refraction index as $n = 1 + \chi/2$ ⁴⁹. Here N is the density of the scattering centers, k is the vacuum wave number, I is the unit matrix and f is the scattering amplitude.

The three main features in the reflectivity curve: the critical angle, the frequency of oscillations and the slope of the decay carry information about the refraction index (or susceptibility) of the material and the thickness as well as the surface roughness of the layer (Figure 3b). The frequency of the Kiessig oscillation is a function of layer thickness d and refractive index δ . This oscillation is superposed to the curve above the critical angle which decreases with $\sim 1/\theta^4$, where θ is the incident angle. The decrease of the reflected intensity can be further changed by the surface roughness, according to $e^{-k_z^i k_z^r \sigma^2}$ expression, where σ is the uncorrelated surface roughness and k_z^i, k_z^r are the vertical component of the incident and refracted wave number of the beam. In case of flat sample the roughness of the surface was 9 Å while the roughness of iron layer started with 5 Å and increased to 10 Å for the thickest layer. For the spherical templates it is more difficult to define roughness. From one hand side there is a substrate roughness which is 10 Å for both sizes but also a roughness between the sublayers were defined in order to smooth out the susceptibility profile. Finally the upmost layer has a separate roughness which was between 2 Å and 6 Å for the 400 nm diameter and between 1 Å and 7 Å for the 25 nm diameter samples. The critical angle of a X-ray reflectivity curve is at $\theta_c = \sqrt{2\delta}$, where δ is proportional to the density of the material. In case of the pristine templates, a well defined critical angle at about 0.13 degrees can be seen. With increasing iron thickness, the most intense change in the total reflection region appears in case of the flat sample (Figure 3c). Since the evaporated iron forms a homogenous layer on the substrate the shift of the critical angle becomes more and more pronounced. However, if the amplitude of the Kiessig

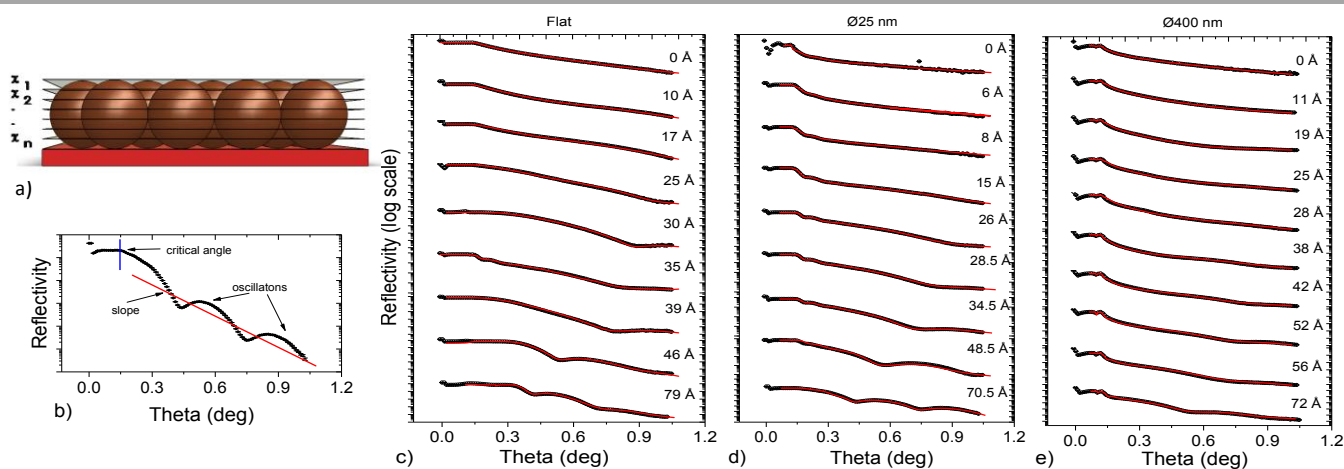


Figure 3 a) Division of the spherical particle template into sublayers. b) A selected reflectivity curve and the definition of the key features such as critical angle, oscillations and the slope of the decay marked. c–e) X-ray reflectivity curves of increasing iron layer thickness on flat silicon substrate and on 25 nm and 400 nm diameter silica spheres (black points). The red line corresponds to the fitted data.

oscillation has an extreme at the vicinity of the critical angle, it alters the continuity of this shift which is evidenced at higher iron thicknesses by the enhanced critical angle. In case of 400 nm diameter spheres (Figure 3e), hardly any change of the critical angle can be seen, only the Kiessig oscillations appear evidencing iron evaporation. The reason for this is that the iron layer cannot form a laterally homogenous layer due to the topography and hence the average density of a sublayer never overcomes the density of the silicon substrate. The change of the x-ray reflectivity curve of the 25 nm diameter sphere sample (Figure 3d) is between the two previously discussed templates. At the very low iron thicknesses the reflectivity curve behaves similarly to the 400 nm samples. After 8 Å thick evaporated iron, a small plateau corresponding to iron critical angle appears and with further iron evaporation becomes more and more dominant. In order to reconstruct the nanosphere structure as well as the vertical distribution (Z direction) of the iron layer on top of the nanospheres the sublayer susceptibilities were determined by analyzing the reflectivity curves. In figure 4 the variation of the susceptibility as a function of the position along Z axis is presented for the 25 nm diameter (a) and 400 nm diameter (b) silica spheres. All susceptibility curves have constant value in the “negative” region along Z axis representing the silicon substrate. At zero position (surface of the substrate) where no iron was deposited, the susceptibility profile drops and then follows the curvature of the spheres as their cross section in the sublayer varies with the thickness along the z-axis.

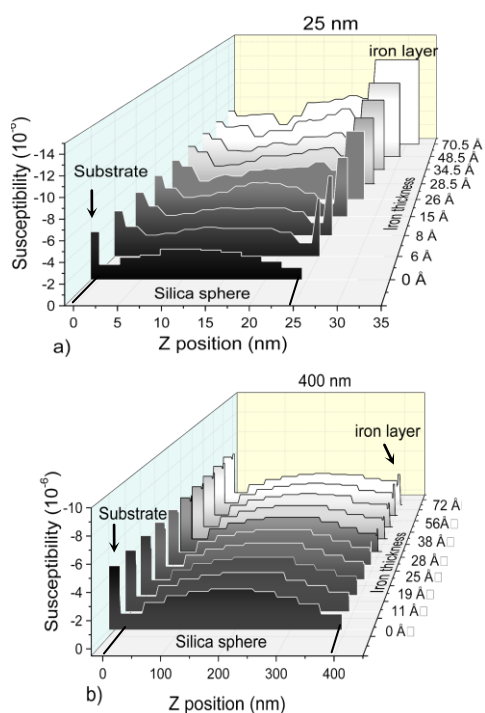


Figure 4 The variation of the susceptibility as a function of the position along the z axis of the substrate for the 25 nm diameter (a) and 400 nm diameter (b) silica spheres. The zero distance represents the substrate/sphere interface

In As expected, the shape of the profiles are similar for the two sphere sizes. However, when the iron layer is evaporated, the shape of the profiles change differently. In case of the 25 nm diameter spheres the evaporated iron thickness is comparable to the sphere size resulting in a significant change in the susceptibility profile. The obtained profiles are also perturbed due to the relatively large inhomogeneities in the template. The initial maximum of the profile at about 12 nm become less and less pronounced and after 29 Å thick iron layer evaporated, the profile doesn't reflect the shape of the nanospheres anymore. In certain regions on the substrate where there were no spheres, the iron was deposited directly on the substrate which is reflected in the increase of the profile in the 0 nm to 7 nm region. Some portion of the iron was deposited on the side of the spheres increasing the susceptibility in the 12 nm to 25 nm region. The definite maximum above 25 nm corresponds to the iron on top of the spheres. In case of 400 nm diameter spheres, the evaporated iron thicknesses are almost negligible compared to the size of the spheres and therefore the susceptibility profiles remained mainly unchanged except for a small region at the top and the bottom of the spheres.

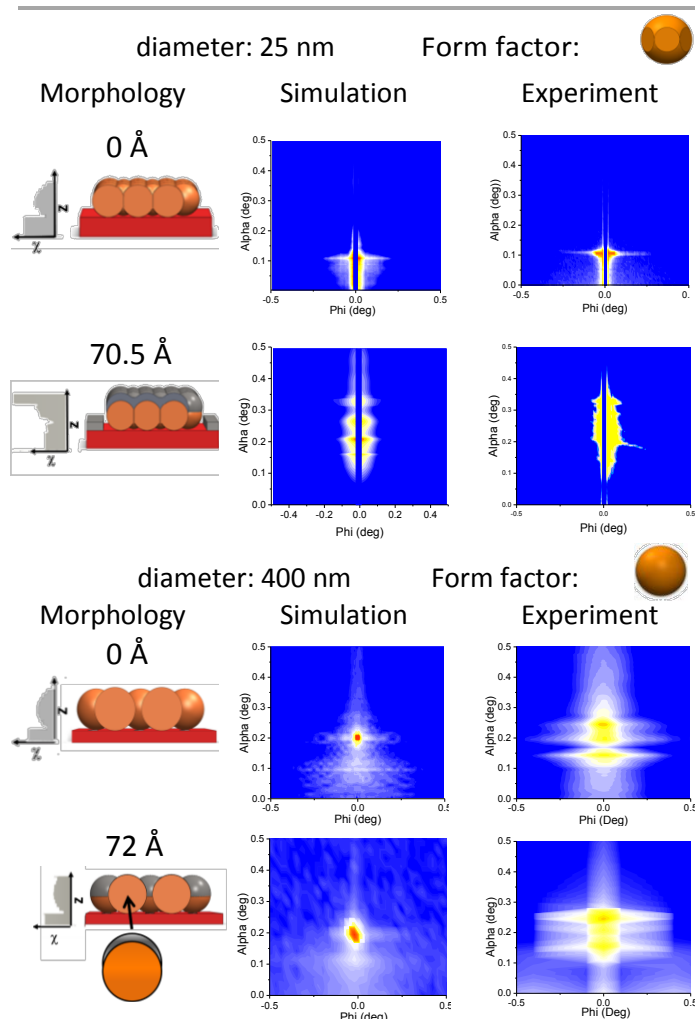


Figure 5 GISAXS images recorded on 25 nm and 400 nm diameter spheres before and after iron deposition and the corresponding simulations. The simulated particle morphology with the corresponding susceptibility profile is given in the first column.

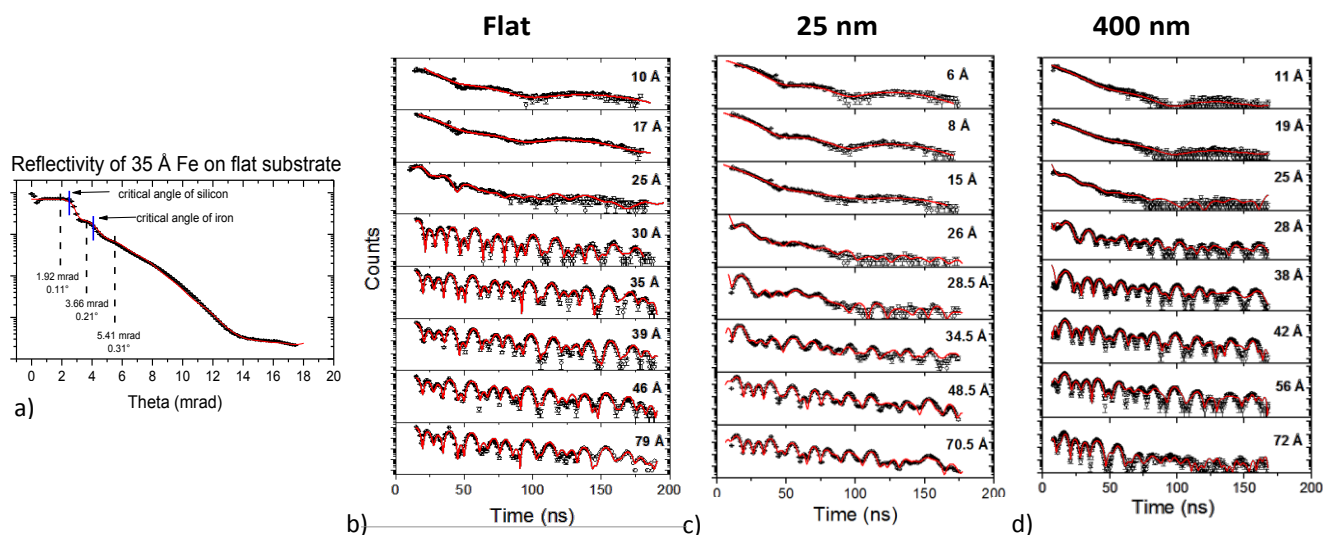


Figure 6 Time resolved NFS spectra (black circles) measured at incident angle of 0.11° after each iron deposition phase (b) on flat Si substrate (c) on 25 nm and (d) on 400 nm diameter spheres. (a) X-ray reflectivity curve measured on 35 Å iron on flat substrate. Dashed lines represent the angles where timespectra were recorded and blue lines indicate the critical angle of silicon and iron. The red line corresponds to the simulated spectra

Based on the extracted layer profiles, the in-situ recorded GISAXS images for both sphere sizes were calculated. Figure 5 shows the recorded images for the 25 nm (top) and 400 nm diameter spheres (bottom) before and after ~ 70 Å iron deposition and the corresponding simulations. In front of the GISAXS images, a schematical picture of the simulated spheres with the assumed iron morphology is shown. In our model the coherent scattering is assumed and treated via a 2D paracrystal interference function based on a hexagonal lattice structure with a Cauchy distribution function. The system consisting of 400 nm diameter spheres could be well described by using full sphere form factor and with particle morphology of randomly oriented domains with an average size of $5 \mu\text{m}$. In case of 25 nm diameter particles, the characterization of the system was different because, contrary to the 400 nm spheres, the particles could not be properly modeled by a perfect full sphere form factor. The small plateau in the susceptibility profile together with the FESEM image indicate that the shape of the small particles may deviate from the perfect sphere, therefore, instead of using a full sphere form factor, it lead to a better result if we used a modified form factor, as presented in figure 5. In the simulation we assumed randomly oriented particle domains of the size of an average 200 nm. Relying on the layer profiles extracted from XRR and on the simulated GISAXS images, it was found that in case of the 25 nm diameter spheres, the evaporated iron forms a continuous layer on the top of the spheres (Figure 5 top), hence the iron cannot be treated as a sum of individual caps formed on the spheres. Due to the larger interparticle distance and the lower ratio of the evaporated iron compared to the sphere size, the 400 nm diameter particles were modeled as independent particles with a small iron cap on top (Figure 5 bottom). The cap has a thickness gradient, starting from the evaporated iron

thickness at the top which decreases continuously as it goes down on the side of the spheres.

In order follow the onset of magnetism as well as to extract the depth dependence of magnetic moment configuration in iron grown on nanospheres, in-situ time resolved NFS measurements were performed (Figure 6b - d). The depth dependence was obtained by measuring nuclear forward scattering at different, carefully selected incident angles of the incoming beam: at 0.11° below the critical angle of silicon (0.12°), at 0.21° below the critical angle of iron (0.22°) and at 0.31° above all critical angles. The latter incident angles together with the corresponding critical angles are indicated in figure 6a.

The evaluation of the NFS data was based on the susceptibility profile obtained from X-ray reflectivity measurements. Utilizing the same sublayer structure, the hyperfine parameters were assigned to these sublayers. However the complexity of the evaluation due to the great amount of independent hyperfine parameters made it necessary to make constraints between the parameters. The hyperfine parameters in the adjacent sublayers were correlated such way that the very top of the sphere was described with one set of parameters and the middle of the sphere with another set of parameters as it is indicated in the right side of figure 7b and c. The NFS data obtained at different incident angles, corresponding to the same iron thickness, were fitted simultaneously.

At the lowest iron thicknesses (below ~ 20 Å) the NFS spectra do not depend on the substrate morphology. The iron layer was found to be not magnetic and the timespectra could be fitted by a quadrupole splitting of $0.63(2) \text{ mm s}^{-1}$ representing the symmetry breaking at the surface.

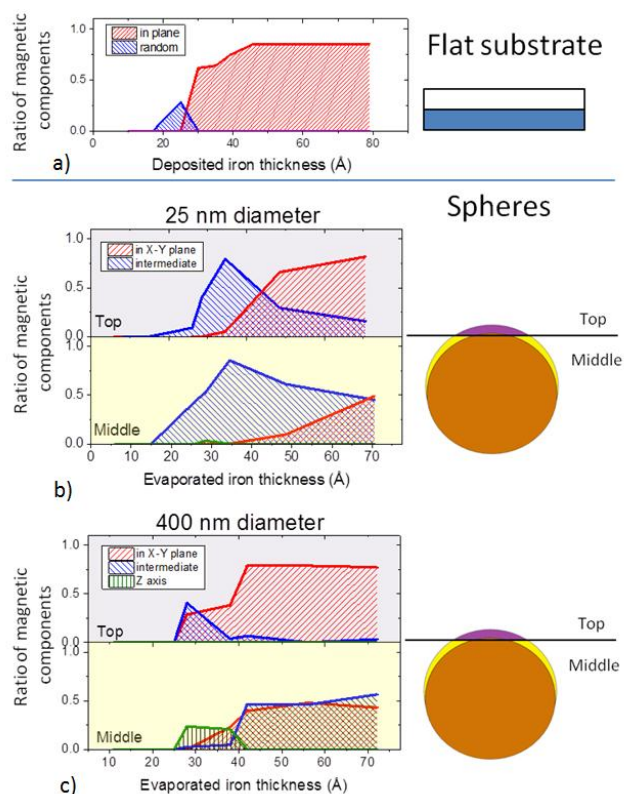


Figure 7 Variation of the differently oriented magnetic components with increasing thickness of deposited iron in case of a) Flat substrate, b) 25 nm and c) 400 nm diameter silica spheres. The green line corresponds to the magnetic orientation parallel to the Z direction, the red line corresponds to the magnetization lying in the X-Y plane and the blue line corresponds to the intermediate magnetization direction. For the 25 nm and 400 nm particles the frame is divided into two parts representing the ratio of the magnetic components on the top of the sphere and on the middle of the sphere as it is indicated in the picture next to the frame

In case of the flat Si sample, the first magnetic component appears at an iron thickness of 25 Å and gives 28% contribution to the spectrum. Here the magnetic moments are oriented 42° to the surface (Figure 7a). With further iron deposition – as expected – the magnetization turned in plane due to the shape anisotropy.

When the iron deposition occurred on nanosphere template the magnetization orientation varied in a more complex way.

In figure 7, the contribution of the different magnetic components to the NFS spectrum is shown as a function of the deposited iron thickness. The components are grouped according to their orientation: a) lays in the X-Y plane, b) intermediate, meaning that it has an out of (X-Y) plane angle distribution and c) where it points along the Z direction. In figure 7b and c, the frame is divided into two parts representing the top and the middle region of the spheres.

In case of 25 nm diameter spheres, the first magnetic component in the spectrum appears as fast oscillations at the iron thickness of 26 Å. The analysis showed that the contribution of the magnetic component in the middle of the sphere is 44% while in the upper part is only 9% and both have random angular distribution. A possible explanation is that,

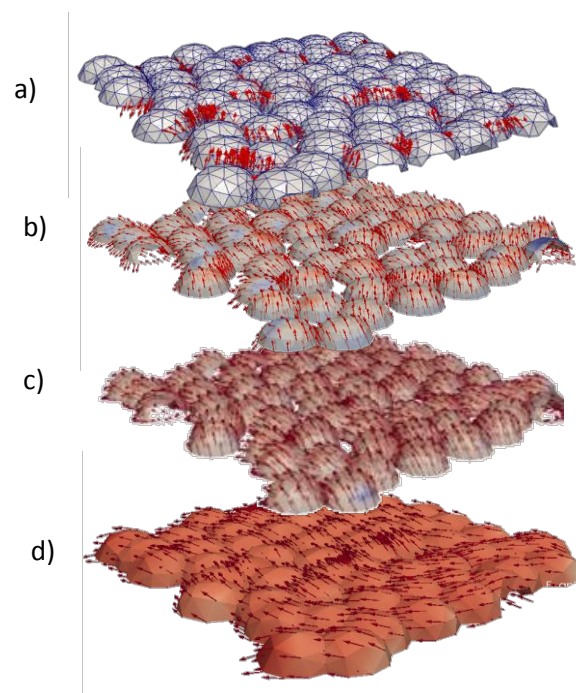


Figure 8 Magnetic moment configuration calculated by micromagnetic simulation for a) 26 Å, b) 34.5 Å, c) 48.5 Å and d) 70.5 Å evaporated iron thickness on 25 nm diameter spheres. The bluish color represents the magnitude of the Z component while the red stands for the X-Y in-plane component.

first, the regions where the spheres are in contact with each other become magnetic due to the locally increased iron concentration. These magnetic volumes are expected to be magnetostatically coupled. Micromagnetic simulations were performed in order to reconstruct the possible magnetic moment configuration. The 3D structure of the nanosphere template was constructed based on a representative FESEM image by taking the position and size of a set of 60 particles and in the simulation this 3D structure was periodically repeated. The saturation magnetization (M_s) was set to $1.7 \times 10^6 \text{ A m}^{-1}$, the exchange stiffness constant to $1 \times 10^{11} \text{ A m}^{-1}$ and no anisotropy was applied. According to the simulations shown in Figure 8a the magnetic domains are isolated from each other and distribute uniformly which makes this system interesting for magnetic applications. By reaching an iron thickness of $\sim 35 \text{ Å}$, the iron cap becomes essentially magnetic with intermediate magnetic distribution, but on the top a small fraction (5%) of the magnetic moments starts to turn in plane (Figure 8b). It can be seen that the structure of magnetization of the iron caps are not limited to only one particle but it involves several particle through exchange coupling. At an iron thickness of $\sim 48 \text{ Å}$ the magnetization on the top part mainly lays in plane. However, in the middle region, the vertical orientation is still distributed, but already, a rotation towards the X-Y plane can be observed (Figure 8c). By the last evaporation step the iron forms a continuous layer on the top of the spheres which was already seen from the reflectivity measurements (Figure 8d). This explains why it is favorable for the system

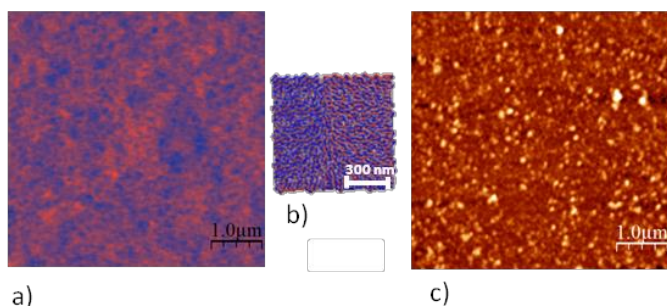


Figure 9 a) MFM image taken on 25 nm diameter nanospheres covered with ~ 70 Å iron and 50 Å Nb capping layer. The magnetic tip was magnetized along the tip axis; hence the measurement was sensitive to the perpendicular component of the magnetization. b) The simulated magnetic orientation distribution in the Fe layer and c) the acquired topography image are shown.

magnetization to lay in the X-Y plane with an off plain distribution along the lower edge of the cap. Figure 9a shows the MFM image recorded on the 70.5 Å thick iron layer (covered by 50 Å Nb capping layer) and the simultaneously acquired topography image (Figure 9a, c). For comparison the result of micromagnetic simulation is plotted in the middle (Figure 9b). The measurement shows the perpendicular component of magnetization in iron. The colors represent the phase shift of the oscillation of the magnetic tip. The MFM measurement indicates that the sizes of magnetic domains are in the order of one micron, much larger than the size of the spheres and no correlation between the magnetic domains and the topology of the surface could be observed.

The evolution of the magnetism in the iron film deposited on the 400 nm diameter spheres template is shown in figure 7c.

At 25 Å thick iron layer, the evaluation of the time spectra revealed, that in the lower part of the cap, the magnetic moments points along the Z direction and has $\sim 25\%$ magnetic fraction while on the top, the orientation is intermediate or lies in the X-Y plane with about 50% contribution to the whole spectrum.

It is known that ultrathin iron film can exhibit complex magnetic structures up to several monolayers⁵⁰⁻⁵². Internal stress or asymmetric crystal lattice can also be the source of magnetic anisotropy³⁰. In our case the orientation of magnetic moments along the z axis stems from the nonmagnetic/magnetic transition region with an ultrathin iron film with gradient thickness on the side of the spheres. Also this gradient thickness is reflected in reduction of ferromagnetism in the lower region which explains the different amount of magnetic contribution in the top and lower part of the iron cap.

The micromagnetic model as well as the boundary conditions were tailored so, that the resulting magnetic moment configuration would show agreement with the experimental data obtained from our nuclear resonant scattering experiments. A strict condition was that at the onset of magnetism, the magnetic moments should be aligned parallel to the z axis at the side of the sphere.

The micromagnetic model is shown in figure 10a. The simulation parameters were identical to what we applied for the 25 nm diameter spheres but in addition a small uniaxial

anisotropy (K_u) along the Z direction was assigned to the transition region in order to reproduce the magnetic configuration extracted from the NFS spectra. We have varied K_u between 0 and $5 \times 10^5 \text{ J m}^{-3}$ and the best fit to the experimental data was found at $K_u = 8 \times 10^4 \text{ J m}^{-3}$. According to the simulation the orientation of the magnetic moments turns from the Z direction to the X-Y plane and on the top it forms a vortex. With further iron deposition (at a thickness of 38 Å) the transition zone moves down on the side of the sphere reducing the effect of anisotropy. As a consequence the vortex starts to spread down on the iron cap resulting in an almost fully in plane magnetization at the top region. In addition, at the lower region, a mixture of intermediate, plane parallel and plane perpendicular magnetic orientation could be observed (Figure 10b). By reaching the 42 Å iron thickness the contribution of magnetic components become dominant in the spectrum. The vortex formation continues and the magnetic moments parallel to the Z axis disappear even at the lower part of the cap. The simulated arrangement of the magnetic moments is shown in figure 10c. We found the best agreement with the extracted data applying a reduced uniaxial anisotropy of $K_u = 1.6 \times 10^4 \text{ J m}^{-3}$. The further increase of the amount of deposited iron to 72 Å only results of a slight change in the magnetic structure (Figure 10d). The reason of this saturated behavior is different for the upper part and for the lower part of the iron cap.

On the top of the spheres the vortex has formed already at the thickness of 42 Å and it doesn't change (significantly) with further iron evaporation explaining the saturated magnetic behavior in the upper part. On the other hand, this argument is not valid at the side of the sphere where one would expect the enhancement of the in-plane magnetization at the expense of the intermediate magnetic component. The magnetic saturation effect is caused by the presence of the nonmagnetic/magnetic interface together with an ultrathin iron film with gradient thickness on the side of the spheres. In our model the transition region with gradient iron thickness favors a magnetization orientation parallel to the Z axis. With

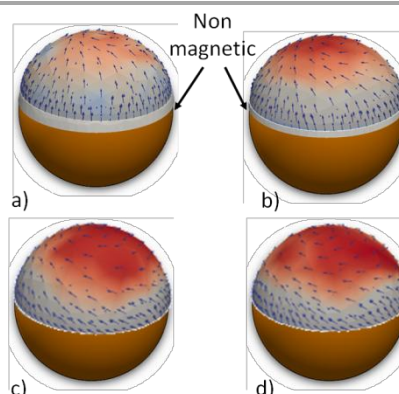


Figure 10 Magnetic moment configuration calculated by micromagnetic simulation for a) 28 Å, b) 38 Å, c) 42 Å and d) 72 Å evaporated iron thickness on 400 nm diameter spheres. The bluish color represents the magnitude of the Z component while the red stands for the X-Y in-plane component. With white, the non magnetic part of the iron layer is shown.

increasing thickness of evaporated iron, the transition zone moves down on the side of the sphere meanwhile its relative contribution decreases. This movement is faster at the beginning when the nonmagnetic/magnetic interface is positioned higher on the side and slows down as the transition zone approaches the lower part of the side of the sphere and disappears when the iron thickness reaches the critical thickness everywhere. In the questioned thickness range (42 Å – 72 Å) the transition zone already reached the bottom of the iron cap and cannot go further down. This is a steady state of the system until the thickness of the iron will exceed a critical iron thickness eliminating this transition zone.

In figure 11 the MFM and AFM image of the 72 Å thick iron film with 50 Å Nb capping layer on top is shown. The MFM measurement features the perpendicular component of magnetization. The hexagonal structure built from the simulated iron caps is shown in Figure 11b in order to compare the simulation with the measurement. As it was assumed in our simulations, each iron cap, contrary to the 25 nm spheres, has an individual magnetic structure. The micromagnetic simulation can be well correlated with the measurement, assigning the red regions to the magnetic vortices on the top with in-plane magnetization and the blue regions to the intermediate component at the side of the sphere.

The two main differences between the 25 nm and 400 nm diameter spheres are: (i) the ratio of the evaporated material thickness compared to the particle diameter, (ii) the amount of contact area between the particles which is manifested in different interparticle interactions. In case of the 25 nm spheres the spheres are in direct contact with each other and also deformed (according to GISAXS) implying a strong interparticle interaction. On the other hand, at low iron thicknesses the magnetic regions appear in the junction of the spheres creating an isolated series of magnetic volumes. Since the magnetic stray fields are long ranged, these regions are expected to exhibit magnetostatic coupling. With further iron evaporation, the independent magnetic regions get in contact, introducing magnetic exchange interaction between the particles. When a continuous iron layer forms on the template, the exchange coupling becomes dominant and it is more reasonable to consider the system as a magnetic layer with

local inhomogeneities than as a strongly coupled assembly of individual magnetic particles. In case of 400 nm diameter spheres are not in direct contact. It has been shown for a short period planar nanodisks array that the interdot interaction has a strong destabilizing effect on the vortex spin state if the interdot distance is less than the radius of the nanodisk.⁵³ However, the destabilizing energy barrier is significantly reduced if a thickness gradient is introduced at the edge of the disks⁵⁴. In our case the thickness gradient is given by the spherical template and for this reason a reduced or no magnetostatic coupling is expected.

In case of 400 nm diameter spheres are not in direct contact. It has been shown for a short period planar nanodisks array that the interdot interaction has a strong destabilizing effect on the vortex spin state if the interdot distance is less than the radius of the nanodisk.⁵⁵ However, the destabilizing energy barrier is significantly reduced if a thickness gradient is introduced at the edge of the disks⁵⁶. In our case the thickness gradient is given by the spherical template and for this reason a reduced or no magnetostatic coupling is expected.

Conclusions

In this paper we have investigated in detail how the size of silica nanospheres composing the template layer affects the evolution of magnetism in an iron ultrathin film, evaporated on top of the particles. We have focused on the thickness range of the nonmagnetic/magnetic transition. In order to gather extensive knowledge about the delicate interaction of the various parameters, we have carefully characterized the systems using different in-situ (NFS, GISAXS, XRR) and ex-situ (AFM, MFM, FESEM) techniques and with micromagnetic simulations we reconstructed the development of magnetic moment configurations.

We have found that the template morphology is cardinal and precise control of magnetization, necessary for industrial application, is not possible without careful selection of the size and distribution of the spherical particles. The ratio of the evaporated iron layer to the sphere size as well as the interparticle distances determine how the magnetic structure form in the capping layer through the exchange coupling between particles and the shape of the particle.

When iron was evaporated on 25 nm diameter particles the iron started to become magnetic in regions where the particles touch each other, creating a magnetic structure, consisting of isolated magnetic domains distributed uniformly which are essential for magnetic storage media. With further iron deposition a magnetic structure involving several particles could be observed due to the enhanced exchange interaction between particles.

On the other hand, the 400 nm diameter spheres, the system could be considered as a sum of individual magnetic particles. The formed magnetic structure reflected the topology of the spheres. By evaporating iron, vortex formation on the top of the cap and magnetic anisotropy along the z axis at the nonmagnetic/magnetic interface was assumed and modeled. With increasing iron thickness, the vortex spreads down on the

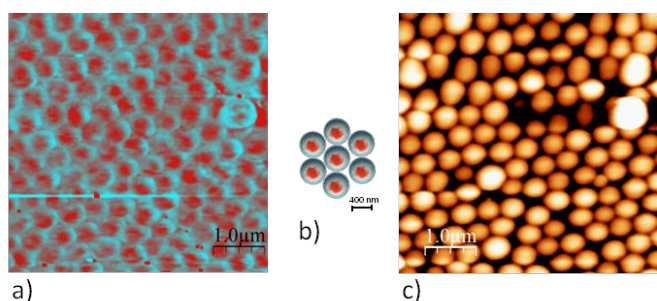


Figure 11 a) MFM image taken on ~70 Å thick iron layer evaporated on 400 nm spheres. The magnetic tip was magnetized along the tip axis; hence the measurement was sensitive to the perpendicular component of the magnetization. b) The simulated hexagonal structure and c) acquired topography image is shown

sphere thus eliminating the Z component of magnetization at the lower edge of the caps.

As a conclusion we can say that the magnetic structure can be fine-tuned through the size of the spherical template together with the thickness of magnetic layer on top and this is a fundamental requirement for the future 20 Tbit/inch² applications.

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