Tailoring the magnetodynamic properties of nanomagnets using magnetocrystalline and shape anisotropies

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Dynamical properties of nanomagnets are affected by the demagnetizing fields created by the same nanoelements. In addition, magnetocrystalline anisotropy produces an effective field that also contributes to the spin dynamics. In this report we show how the dimensions of magnetic elements can be used to balance crystalline and shape anisotropies, and that this can be used to tailor the magnetodynamic properties. We study ferromagnetic ellipses patterned from a 10 nm thick epitaxial Fe film with dimensions ranging from 50 × 150 nm to 150 × 450 nm. Our study combines ferromagnetic resonance spectroscopy (FMR) with analytical calculations, and proves that the dynamical properties can be effectively controlled by changing the size of the nanomagnets.

I. INTRODUCTION

The magnetodynamic properties of nanostructures have received extensive attention, from both fundamental and applications viewpoints. While technological applications are important, there is also significant interest in understanding the fundamental behaviour of magnetic materials when they are confined to nanoscale dimensions. In confined magnetic elements, there is a complex competition between exchange, dipolar and anisotropic magnetic energies. Understanding the interplay between the various energy terms is thus of importance when investigating the magnetodynamics of such systems.

The magnetization dynamics in patterned magnetic structures has been extensively studied previously. However, the dynamic properties of structures utilizing both crystalline and shape anisotropies of the magnetic elements, remains relatively unexplored. The goal of this study is thus to investigate a system where all these energy terms contribute to determine the dynamics of the system.

We have investigated a system utilizing epitaxial Fe as the ferromagnetic (FM) material, patterned to an array of elliptical nano-magnets. This results in a system combining the cubic crystalline anisotropy of Fe with the shape anisotropy due to the elliptical shape of the confined magnetic elements.

The dynamic properties were investigated by ferromagnetic resonance (FMR) experiments for ellipses with a thickness of 10 nm and lateral dimensions of 50 × 150 nm, 100 × 300 nm and 150 × 450 nm. The experimental results are compared with a macrospin model considering the total free energy density of a ferromagnetic structure containing both crystalline and shape anisotropies. From the free energy density we calculate the ferromagnetic resonance conditions, showing a good agreement with the experimental results.

Using the macrospin model, we also explore the properties of ellipses with lateral dimensions ranging from 50 × 150 nm to 500 × 1500 nm, showing how the ellipse size governs the balance between crystalline and shape anisotropy. In addition, we briefly investigate how the orientation of the easy/hard crystalographic anisotropy axis with respect to the long/short axis of the ellipse affects the dynamics.

II. EXPERIMENTAL SETUP

The samples are based on a single crystalline Fe film epitaxially grown on MgO(001) substrates. The ferromagnetic ellipses were patterned by e-beam lithography and ion beam milling from a 10 nm thick Fe layer, and had lateral dimensions of 50 × 150 nm, 100 × 300 nm and 150 × 450 nm. Further details concerning sample growth and processing are similar to that described earlier.

The FMR experiments were performed using two complementary setups. The cavity FMR measurements were carried out in a commercial X-band electron paramagnetic resonance (EPR) setup with a fixed microwave frequency of 9.4 GHz (Bruker Bio-spin ELEXSYS 500, with a cylindrical TE-011 microwave cavity). The magnitude of the external field is then swept to locate the resonance field, $H_R$. The sample is attached to a quartz rod connected to a goniometer, allowing to rotate the sample 360 degrees in order to accurately resolve the angular dependence. The FMR measurements were performed with a low amplitude ac modulation of the static field, which allows lock-in detection to be used in order to increase the signal to noise ratio.

For the broadband FMR measurements, we used a vector network analyzer (VNA) FMR setup with a coplanar waveguide (CPW) excitation structure. The static external field was applied in the sample plane, and perpendicular to the microwave field from the CPW. This was used to obtain the standard microwave S parameters as a function of frequency for various fixed values of the static field. This allows for a complete field versus frequency
map of the resonance absorption, not being limited to a fixed frequency as for the cavity measurements. Data was then collected in a field range of ± 500 mT, and a frequency range of 1-25 GHz. Typical absorption maps had a step size of Δf = 0.1 GHz and ΔH₀ = 5 mT.

III. FREE ENERGY DENSITY AND THEORETICAL FMR SPECTRUM

Due to the size and shape of the ellipses, we consider the individual magnetic elements to be in a single domain state. This was also confirmed by MFM imaging of similar samples. Having a single domain state allows us to use an analytical macrospin model to investigate the ferromagnetic resonance properties of the system.

The array of ellipses have an inter-particle spacing of two times the corresponding ellipse dimension in each direction, as illustrated in Fig. 1a. This spacing is sufficient to significantly reduce the dipolar coupling between the individual elements. A more detailed study of the dipolar coupling will be presented elsewhere and as a first approximation we consider the ellipses as uncoupled magnetic elements.

We start by defining the geometry of the system, and consider the free energy density of the individual magnetic elements.

From the sample geometry illustrated in Fig. 1b (magnetic element in the x-y plane), one gets that:

\[
\begin{align*}
M_x &= M_s \sin \theta \cos \phi \\
M_y &= M_s \sin \theta \sin \phi \\
M_z &= M_s \cos \theta,
\end{align*}
\]

where \(M_s\) is the saturation magnetization. Assuming the external applied field, \(H_0\), is oriented in the sample plane, \(\theta_H = \pi/2\), gives

\[
\begin{align*}
H_x &= H_0 \cos \phi_H \\
H_y &= H_0 \sin \phi_H
\end{align*}
\]

After defining the geometry, one can calculate the free energy density of the system by adding up the various energy terms. Using a macrospin model, we do not consider the exchange energy. The total free energy density of the system is then given by, \(E_{\text{tot}} = E_{\text{Zeeman}} + E_{\text{Demagnetization}} + E_{\text{Anisotropy}}\).

\[
E_Z = -\vec{M} \cdot \vec{H} = -M_s H_0 [\sin \theta \cos \phi \cos \phi_H + \sin \theta \sin \phi \sin \phi_H] = -M_s H_0 \sin \theta \cos(\phi - \phi_H),
\]

where \(\mu_0\) is the vacuum permeability, \(N_i\) are the demagnetization factors and \(N_x + N_y + N_z = 1\). The units for the saturation magnetization and magnetic field are \([M_s] = \text{A/m}\) and \([H] = \text{T}\) respectively. We assume cubic crystalline anisotropy for the epitaxial Fe film, with the easy axis oriented parallel to the long/short axis of the ellipse. The lowest order term in the crystalline anisotropy energy is then the fourth order term:

\[
E_{\text{Anis}} = K_1 [\alpha_x^2 \alpha_y^2 + \alpha_y^2 \alpha_z^2 + \alpha_z^2 \alpha_z^2]
\]

\[
= K_1 \left[ \sin^4 \theta \sin^2 \phi \cos^2 \phi + \sin^2 \theta \sin^2 \phi \cos^2 \theta \right],
\]

where \(K_1\) is the magnetocrystalline anisotropy constant and \(\alpha_i = M_i/M_s\). After adding the terms, one can write the total free energy density as:

\[
\begin{align*}
E_{\text{tot}} &= -M_s H_0 \sin \theta \cos(\phi - \phi_H) \\
&+ \frac{\mu_0 M_s^2}{2} \left[ \sin^2 \theta \cos^2 \phi \left( N_x + \frac{2K_1}{\mu_0 M_s^2} \sin^2 \theta \sin^2 \phi \right) \\
&+ \sin^2 \theta \sin^2 \phi \left( N_y + \frac{2K_1}{\mu_0 M_s^2} \cos^2 \theta \right) \\
&+ \cos^2 \theta \left( N_z + \frac{2K_1}{\mu_0 M_s^2} \sin^2 \theta \cos^2 \phi \right) \right].
\end{align*}
\]
important to note that the orientation of the magnetization, given by $\phi$, might not be parallel to the applied field, $\phi_H$. Thus, to investigate the resonance conditions of the system one must first find the equilibrium orientation of the magnetization. The equilibrium orientation was found by minimizing the free energy density of the system given by Eq. (6) for each value of $H$ and $\phi_H$, and was performed numerically. After obtaining the equilibrium orientation of the magnetization, one can calculate the resonance frequency $\omega$, given by [15]

$$
\omega = \frac{\gamma}{\mu_0 M_s \sin \theta} \sqrt{\left( \frac{\partial^2 E_{\text{tot}}}{\partial \theta^2} \right)^2 + \left( \frac{\partial^2 E_{\text{tot}}}{\partial \phi^2} \right)^2 + \left( \frac{\partial^2 E_{\text{tot}}}{\partial \theta \partial \phi} \right)^2}. \tag{7}
$$

By solving Eq. (7), one can obtain the resonance frequency as a function of magnitude and direction of the applied field, $\omega(H, \phi_H)$. Calculating the various terms in Eq. (7), one obtains:

$$
\frac{\partial^2 E_{\text{tot}}}{\partial \theta^2} = M_s H \sin \theta \cos(\phi - \phi_H)
\quad + \frac{K_1}{4} \left[ \cos 2\theta - 4 \mu_0 M_s^2 N_x / K_1 \right]
\quad + \frac{2 \mu_0 M_s^2}{K_1} \left( N_x + N_y + (N_x - N_y) \cos 2\phi \right)
\quad + \left( \cos 4\phi + 7 \right) \cos 4\theta \tag{8}
$$

$$
\frac{\partial^2 E_{\text{tot}}}{\partial \phi^2} = M_s H \sin \theta \cos(\phi - \phi_H)
\quad + 2 K_1 \sin^2 \theta \left[ \cos 4\phi \sin^2 \theta \right]
\quad + \frac{\mu_0 M_s^2}{2 K_1} \left( N_y - N_x \right) \cos 2\phi \tag{9}
$$

$$
\frac{\partial^2 E_{\text{tot}}}{\partial \theta \partial \phi} = M_s H \cos \theta \sin(\phi - \phi_H)
\quad + 8 K_1 \sin \phi \cos \phi \sin \theta \cos \theta \left[ \cos 2\phi \sin^2 \theta \right]
\quad + \frac{\mu_0 M_s^2 (N_y - N_x)}{4 K_1} \tag{10}
$$

For thin films, one can simplify these expressions by assuming that the magnetization is oriented in the film plane, $\theta = \pi/2$. After introducing the anisotropy field, $H_k = 2K_1/M_s$, one obtains the resonance frequency given by Eq. (7):

$$
\left( \frac{\omega}{\gamma} \right)^2 = \left[ H \cos(\phi - \phi_H) + \mu_0 M_s \left( N_z - \frac{N_x + N_y + (N_x - N_y) \cos 2\phi}{2} \right) \right] + \frac{H_k}{4} \left( 3 + \cos 4\phi \right)
\quad \times \left[ H \cos(\phi - \phi_H) + H_k \cos 4\phi \right]
\quad + \mu_0 M_s (N_y - N_x) \cos 2\phi \right]. \tag{11}
$$

Equation (11) gives the resonance frequency for the general case, with the assumption that the magnetization is oriented in the sample plane. Depending on the shape and size of the magnetic elements, one can then adjust the demagnetization factors $N_i$ to obtain the resonance conditions for various samples.

In addition to the four fold symmetry from the cubic anisotropy, one notices that in this case there are additional terms of two fold symmetry due to the shape anisotropy along the long/short axis of the ellipse. The resonance conditions of the system are thus more complicated, and is determined by the interplay of shape and crystalline anisotropies. This brings us to the main topic of the study, to investigate how tuning the various energy terms changes the magnetodynamic properties of the system.

### IV. RESULTS AND DISCUSSION

#### A. Cavity FMR measurements

The experiments to investigate the angular dependence were performed in the X-band cavity FMR setup described previously. This gives an angular FMR spectrum for both the continuous film and an array of ellipses of dimension $150 \times 450$ nm, as shown in Fig. 2.

Going from a continuous film to a patterned array of ellipses, there is a significant difference. For the continuous film, the four fold symmetry due to the cubic crystalline anisotropy in Fe is dominating. For the ellipses the situation is more complicated, as there are competing energies also from the shape anisotropy.

To investigate this, we compare the experimental and theoretical results. By solving Eq. (11) after first minimizing the free energy density for each value of $H$ and $\phi_H$, one gets the dispersion relations shown in the lower part of Fig. 3. From Eq. (11), the relevant parameters determining the dispersion are the demagnetization factors $N_i$, the anisotropy field $H_k$, and the saturation magnetization $M_s$. In nanometer-dimension magnetic structures, estimates of the demagnetization factors using an ellipsoidal formulae are considered to represent the anisotropy fields well. The factors $N_i$ were found from [13].
for an ellipse of dimension $10 \times 150 \times 450$ nm they are: $N_x \approx 0.005$, $N_y \approx 0.05$ and $N_z = 1 - N_x - N_y$. The anisotropy field $H_k$ was determined from the experimental FMR spectrum in Fig. 2, and was found to be approx. 50 mT. In the calculations, $M_s$ was adjusted to obtain the best fit between the experimental and theoretical spectrum, and the best fit was found for $M_s = 1.5 \times 10^6$ A/m (a reduction of approx. 10% compared to textbook values of $M_s$ for Fe).

To compare the angular dependence of the theoretical spectrum with experimental results from the cavity measurements shown in Fig. 2, one can invert the solution. This rather gives the resonance field $H_R$, as a function of rotation angle for a fixed excitation frequency of 9.4 GHz, and the inverted solution is shown in the upper part of Fig. 3. To distinguish the effect of crystalline anisotropy and shape anisotropy, the same calculations were also performed assuming polycrystalline Fe, setting $H_k = 0$.

Comparing theory and experiment in Fig. 2 and 3, one notice that for the continuous film, both show the expected 4-fold cubic symmetry. For the ellipses, the theory replicates the “heart shape” of the resonance well. In the experimental data in Fig. 2a), there are also some additional weak resonance lines. It is known that regions of non-uniform magnetization along the sample edges could lead to a spectrum of additional edge modes. The assumption that the magnetization in the individual ellipses is uniform is a good approximation at the center of the ellipse, but along the edges the magnetization will be less uniform due to the shape anisotropy. In addition, there could be other spin wave excitations with nonzero wave vectors, and correspondingly varying frequencies. Experimentally, we observe that the amplitude of the main mode is dominating compared to the additional weak resonance. In the following, we thus focus on the main mode.

FIG. 2. Experimental FMR spectrum for a) continuous film and b) ellipses of dimension $150 \times 450$ nm from the X-band cavity FMR setup.

FIG. 3. Upper figures: Theoretical data for resonance field versus rotation angle for a) continuous film and b) ellipse of dimension $10 \times 150 \times 450$ nm, with (red) and without (blue) crystalline anisotropy. Lower figures: Dispersion for c) continuous film and d) ellipse of dimension $10 \times 150 \times 450$ nm, with (solid lines) and without (dotted lines) crystalline anisotropy.

### B. Broadband FMR measurements

To obtain a complete field versus frequency map of the FMR absorption, we performed experiments using a broadband setup. The experimental FMR absorption peaks were extracted, and are shown in Fig. 4. Red dots represent the main FMR mode, and the blue squares the additional weak resonance. For clarity only a few selected datapoints are included, where the uncertainty in determining the absorption peak position is of the order of the dot size. The experimental results are then compared with the theoretical FMR spectrum, shown as dotted black lines.

The agreement between theory and experiment is good for an applied field oriented along the long axis of the ellipse, as indicated in Fig. 4. Sweeping the field from negative to positive, one also notices the switching of the magnetization. As the field is swept from negative to zero, the FMR frequency drops as expected. This continues also for positive fields until the external field is strong enough to overcome the anisotropy favouring the magnetization along the long axis of the ellipse. The switching is then observed as an abrupt jump in the FMR spectrum. Further details concerning the switching behaviour of the ellipses is presented elsewhere.
When applying the field along the short axis there are now two parallel dispersing lines, as shown in Fig. 4a. A high frequency resonance and an additional weak resonance at lower frequency, which corresponds well to the additional resonance also seen in Fig. 2b.

Comparing the measurements along the short axis with the theoretical dispersion, one does not observe the low field resonance in Fig. 4 (the black dotted line below 100 mT). This can be understood, as in this field range the magnetization will be oriented along the long axis of the ellipse, parallel to the microwave (MW) pumping field in this experimental setup (parallel pumping). However, in the cavity measurement this resonance can be observed as the pumping field in this case is oriented out of plane, always perpendicular to the magnetization. This can be seen in Fig. 2b, at a rotation of 90 degrees (field along short axis). The first resonance is observed at a field of approx. 50 mT, and a second one at approx. 100 mT. This corresponds quite well with the expected resonance fields from the theoretical curve in Fig. 4b at a frequency of 9.4 GHz. At higher fields the magnetization in the ellipse will reorient itself along the external field such that it is perpendicular to the MW pumping field. In this field region, the theoretical spectrum corresponds well with the high frequency branch of the experimental data.

Using our macrospin model, we can only account for the main FMR mode. However, from our experiments we observe that the amplitude of the main mode is dominating compared to the weak additional modes. Our model should thus be a good approximation to describe the main FMR modes of the system.

C. Size of the Ellipses

To investigate the interplay of shape anisotropy and crystalline anisotropy, we studied ellipses of various lateral dimensions but with the same aspect ratio of 1:3. There are two limiting cases worth noticing: In the limit of a very large ellipse, one should expect a behaviour close to that of a continuous film, where crystalline anisotropy is dominating. By gradually reducing the size of the ellipse, shape anisotropy becomes increasingly important. This means one can use the size of the magnetic elements to tune the ratio between crystalline and shape anisotropies, and thus change the magnetodynamic properties of the system.

Changing the lateral dimensions of the ellipse affects the free energy density of the system, given by Eq. (6). The transition from a continuous film to a small ellipse can be observed by looking at the energy landscape of the system as a function of the ellipse dimension, as shown in Fig. 5.

Figure 5 indicates how the free energy density changes when one gradually reduce the size of the ellipse from the upper limit of a continuous film, to an ellipse of dimension 50 × 150 nm. As expected, one sees that in all cases the magnetization favours an orientation in the sample plane (θ = 90, from sample geometry as defined in Fig. 1). For the continuous film and the largest ellipse in Fig. 5a and b, one can clearly see the dominating crystalline anisotropy, with a four fold symmetry between the energy minima along the ϕ axis.

In the intermediate case for an ellipse of dimension 150 × 450 nm, one has two dominating energy minima at ϕ = 0 and ϕ = 180 (magnetization along the long axis of the ellipse). In addition, there is a quite flat saddle point at ϕ = 90 (which corresponds to a magnetization along the short axis of the ellipse). This not a stable energy minima, but the flatness of the saddle point means that applying a small magnetic field along this axis will create a local energy minima along this direction.
For the smallest ellipse, the energy landscape is dominated by the two fold shape anisotropy along the long axis of the ellipse. To align the magnetization along the short axis of the ellipse ($\phi = 90$) will thus require a quite large external field.

As shown previously, the FMR frequency given by Eq. [11] is determined by the free energy density of the system. Adjusting the lateral dimension of the ellipse is thus an important parameter controlling the FMR frequency. From Eq. [11], one notices that the resonance frequency is determined by contributions of both two fold and four fold symmetry. From this expression, the relevant ratio to determine which term will dominate is given by $H_K/\mu_0M_s(N_x - N_y)$. Changing the ellipse dimension, and thus the demagnetization factors $N_x$, affects the resonance frequency significantly, as shown in the upper panel of Fig. 6.

For an ellipse of dimension $500 \times 1500$ nm the dispersion starts to look similar along the long/short axis of the ellipse, as indicated in Fig. 7. If the only contribution was from the crystalline anisotropy, the dispersion should be identical along the long/short axis due to the four fold symmetry. Comparing the FMR spectrum for the largest ellipse in Fig. 7 to that of a continuous film in Fig. 3, they look very similar. This indicates that as the sample dimensions approach the $\mu m$ scale, shape anisotropies play a minor role compared to the crystalline anisotropy.

To summarize the size dependence, we have shown that for sample dimensions above approx. 1 $\mu m$, crystalline anisotropy will dominate. In the opposite size limit, shape anisotropy will dominate for sample dimensions below approx. $50 \times 150$ nm. In this intermediate regime, one can thus effectively use the sample size as a parameter to tune the balance between crystalline and shape anisotropies.

D. Direction the crystallographic axis

To this point, we have investigated the case where the crystallographic axis of the Fe film is oriented such that the easy axis is parallel to the long/short axis of the ellipse. For completeness, we consider here the case of rotating the crystallographic axis by 45 degrees, such that rather the hard axis is oriented parallel to the long/short axis of the ellipse. The rotation of the crystallographic axis was done by making the substitution $\phi \rightarrow \phi + \pi/4$ in Eq. [5], and results in a new expression for the free energy density used to calculate the FMR spectrum.

Comparing the dispersion for the two different orientations of the crystallographic axis in Fig. 8a, one now has a stronger angular dependence of the resonance frequency when the external field is oriented along the short axis of the ellipse. Solid lines represents the new dispersion af-

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FIG. 7. a) Theoretical dispersion for ellipses of dimension $500 \times 1500$ nm (Black), $250 \times 750$ nm (Blue) and $150 \times 450$ nm (Red). b) Angular dependence of same data.
ter rotating the crystallographic axis by 45 degrees, and dotted lines are included for comparison with the other direction. The strong angular dependence is illustrated in more detail in Fig. 8b, where rotating the external field by less then 5 degrees away from the short axis of the ellipse drastically changes the resonance frequency.

![Graph showing angular dependence of resonance frequency](image)

**E. Thickness of the Ferromagnetic film**

Another important parameter is the thickness of the ferromagnetic film. So far we have only considered the case of a 10 nm film, but increasing the Fe thickness has several important effects. The demagnetization factors \( N_i \), are determined by the ratio between the length, width and thickness of the ellipse. Adjusting the thickness changes \( N_i \) and thus the FMR frequency. However, increasing the thickness can also introduce a transition where the ellipses change from being in a single domain state to multi domain states.

The switching properties and domain formation in samples with a Fe thickness of 10, 30 and 50 nm is presented elsewhere, and for the 10 nm ellipses they were always found to be in a single domain state. Increasing the thickness makes it energetically favourable to form flux closure domains, and already at a thickness of 30 nm some of the particles were found to be in such multi domain states.

We also investigated the FMR spectrum of samples with a thickness of 30 nm and 50 nm, which had a rich spectrum of modes compared to the 10 nm case. However, the formation and dynamics of multi domain states is not the focus of this paper and will not be discussed in detail. The main point is that to make sure the magnetic elements are in single domain states, one has to keep the thickness well below 30 nm for ellipses of the dimensions we have investigated.

**V. CONCLUSIONS**

In this study, we have investigated how the combined interplay between shape anisotropy and crystalline anisotropy affects the magnetodynamic properties of confined magnetic elements.

We have shown that a simple macrospin model for the FMR frequency gives good agreement with the experimental results for the main FMR mode. Using the macrospin model for sample dimensions in the range of 50-1500 nm and a film thickness of 10 nm, we show how changing the sample size and orientation of the crystallographic easy/hard axis affects the magnetodynamic properties.

For the smallest ellipses, shape anisotropy is dominating, whereas for the largest ellipses crystalline anisotropy is the dominating energy term. From Eq. (11), the relative contributions to the resonance frequency from crystalline and shape anisotropy is given by: \( H_k/\mu_0 M_s(N_x - N_y) \), determined by the anisotropy field \( H_k \), the saturation magnetization \( M_s \) and the demagnetization factors \( N_i \).

For the case of a 10 nm thick epitaxial Fe film, one has an intermediate regime between approximately 50 nm to 1 \( \mu \)m where one can use the sample size as an additional tuning parameter for the dynamic properties. For other materials with a different \( H_k \) and \( M_s \), this regime can be shifted to smaller/larger sample sizes. The tunability of the relative contributions means that by changing the material parameters and sample size one can tailor the magnetodynamic properties of the magnetic elements, which could be of importance for magnonics applications.

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