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Competing anisotropies in exchange biased nano-structured thin films

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The magnetic anisotropies of a patterned, exchange biased Fe\(_{50}\)Mn\(_{50}\)/Ni\(_{80}\)Fe\(_{20}\) system are studied using ferromagnetic resonance, supplemented by Brillouin light scattering experiments and Kerr magnetometry. The exchange biased bi-layer is partially etched into an antidot geometry so that the system approximates a Ni\(_{80}\)Fe\(_{20}\) layer in contact with antidot structured Fe\(_{50}\)Mn\(_{50}\). Brillouin light scattering measurements of the spin wave frequency dependence on the wave vector reveal a magnonic band gap as expected for a periodic modulation of the magnetic properties. Analysis of the ferromagnetic resonance spectra reveals 8-fold and 4-fold contributions to the magnetic anisotropy. Additionally, the antidot patterning decreases the magnitude of the exchange bias and modifies strongly its angular dependence. Softening of all resonance modes is most pronounced for the applied magnetic field aligned within 10\(^\circ\) of the antidot axis, in the direction of the bias. Given the degree to which one can tailor the ground state, the resulting asymmetry at low frequencies could make this an interesting candidate for applications such as selective/directional microwave filtering and multi-state magnetic logic.

I. INTRODUCTION

Periodically patterned holes in a magnetic film can be used to affect microwave frequency response, and are used in magnonic devices, spin wave logic \[3\], interferometry and filtering devices \[3\]. An interesting aspect of patterned films is that they induce a non-uniform magnetisation reversal process, which can be advantageous for the development of multi-state memory devices \[3\]. A stepped magnetisation reversal, necessary for such applications, can be achieved by patterning a bi-component structure, typically comprised of antidot structuring with the holes filled with a different magnetic material \[7\] or by adding positive or negative exchange bias on a single ferromagnetic layer system \[4,11\]. Exchange bias can also be seen as a way to modify the dynamic response of the magnetic system, as a result of the competing unidirectional and anisotropic field distribution \[12\]. In particular, an asymmetric microwave response with respect to the applied field has been demonstrated to originate in the unbalanced pole distribution at the edges of the holes in the antidot lattice (ADL) \[13\].

We report the magneto-dynamic properties of an antiferromagnet - ferromagnet system with antidot structuring on the antiferromagnet, but incomplete holes in the ferromagnetic layer, studied using ferromagnetic resonance (FMR) and Brillouin light scattering (BLS). This type of structuring results in a periodic modulation of the ferromagnetic layer thickness and exchange bias between the holes. The layer configuration is illustrated in Fig. 1(a). Having a continuous film adjacent to the ADL allows the spin wave modes to have a complex dependence on the applied field angle, as spin waves can potentially be allowed to propagate in all directions \[4\]. The concept of partially etched ADL has been reported in Ref. \[14\], where the two resonance modes observed both via all-electric spin wave spectroscopy and micro-focused BLS on an ADL with 1 µm pitch and 435 nm hole diameter, have been attributed to the hole and inter-hole channels, which are perpendicular to the applied magnetic field direction. The sample reported here is different in terms of the lattice parameters, etch depth and presence of the exchange bias field. Applied magnetic field angle dependent FMR results show a combined 8-fold symmetry for the lowest frequency resonance modes and a dominant 4-fold symmetry for the highest frequency resonances. Also, the variation of the exchange bias field with applied field direction exhibits an interesting dependence. It is widely known that exchange bias in continuous films possesses a unidirectional symmetry with regards to an external field \[15,16\]. However, in the case of the ADL here studied, the effect of exchange bias on the FMR properties appears to vary greatly with the angle between the external magnetic field and the symmetry of the ADL structuring. Previous reports focused mainly on the effect of ADL on the net exchange bias field \[4,11,17,18\] which in turn was observed to affect the static magnetisation, without providing detail on the angular dependence of the FMR
modes we address in this manuscript. Using micromag-137
etic modelling we obtain simulated resonance spectra138
which is in good agreement with the experimental FMR139
and BLS data. The resonance spectra as a function of the140
applied field angle and the spatial distribution of the pre-
cessional modes were investigated in order to determine141
the origin of the 8-fold anisotropy observed in the exper-
iments. Modes are considered to be localised when the142
precession amplitude is confined to the edges of the holes143
and extended when the maximum precession amplitude144
extends across the ADL. This nomenclature has been in-
troduced in Refs. [19, 20] and since then used in several145
Refs. [21–23]. Here, it is found that an 8-fold anisotropy146
emerges due to the partial patterning in the FM layer,
causing an overlap in resonance frequency between the
localised (or edge) mode and the first extended mode.
Importantly, despite being partially patterned across the
whole thickness, the structure behaves as magnonic crys-
tal exhibiting characteristic spin wave band gaps induced
by the artificial periodicity of the ADL.

The manuscript is structured as follows: the details of
the film growth, patterning, structural analysis and mag-
etometry data are presented in Sec. II. The magneto-
dynamic properties of the patterning process studied
by FMR and BLS, and the effect of patterning on the mag-
netic anisotropies are discussed in Sec. III. In Sec. IV we
present and discuss the micromagnetic simulations car-
ted out to interpret the origin of the anisotropy in this
particular ADL system.

II. FILM GROWTH AND PATTERNING

A Si substrate was sputtered with an 8 nm thick layer of
tantalum (Ta), acting as a buffer layer, and followed151
by the sequential deposition of Fe$_{20}$Ni$_{30}$ and Fe$_{50}$Mn$_{50}$ in
a sputtering system (Shamrock SFI) at a base pressure154
of 10$^{-8}$ mbar in the presence of an in-plane magnetic155
field of 9 mT. The bi-layer was capped with an 8 nm Ta$_{317}$
layer to prevent oxidation. Following deposition, the158
sample was annealed at 498 K in a 0.2 T magnetic field159
(3°C/minute ramp, 120 minutes at maximum tempera-
ture) in order to set the exchange bias direction. To
initiate the patterning process, a 220 nm thick layer of160
silicon nitride (SiN) was deposited on top of the Ta layer162
with the purpose of using it as a hard mask. The sample163
was subsequently patterned using electron beam lithog-
rphy combined with reactive ion etching. The whole165
process was carried out in three main steps: (1) a 200 nm
thick layer of ZEP520 resist was deposited on top of the166
SiN, subsequently exposed to a Vistec VB6 UHR168
EWF e-beam writer and developed in O-Xylene; (2) the169
developed pattern was transferred to the hard mask via170
a reactive ion etching process (RIE) using CHF$_3$/O$_2$ in171
a 80+RIE etching tool; and (3) the NiFe/FeMn/Ta was172
etched on a ET340 RIE tool using CH$_4$/H$_2$ [23]. The es-
timated etching rate for the NiFe/FeMn/Ta layers was 3 nm/min. The ADL covered an area of 1.5 $\times$ 1.5 mm$^2$.

The unit cell size is 420 nm, with 280 nm diameter holes. A scanning electron microscopy (SEM) image of the structure is shown in Fig. 1(b).

An area of the substrate (5 $\times$ 5 mm$^2$) which was subject to the fabrication process but remained unpattered was used as a reference in the study of the magnetic properties of the continuous films. In Appendix Sec. A we compared the resonance properties of this continuous film with those of the continuous film as-deposited. The results suggest that the fabrication process did not affect the exchange bias properties of the continuous film.

Conventional transmission electron microscopy and electron energy loss spectroscopy (EELS) studies on a cross section of the sample were performed in order to determine the thickness and the elemental composition of each layer. EELS measurements were performed in a probe corrected JEOL ARM200F scanning transmission electron microscope operated at 200 kV and equipped with a cold field emission electron gun and a GIF Gatan Quantum ER spectrometer. The EELS data were collected at a dispersion of 1.2 meV/channel (5 eV / 4096 channels) and pixel size of 1.54 nm. Figures 2(a)-(c) show a cross-section of the ADL which was prepared by focused ion beam (FIB) with a cut along the lattice diagonal. We note that the unetched sections of the ADL are protected by a trapezoid shaped section of SiN.

FIG. 1. (a) Schematic of the cross-section view of the partially etched ADL. The ADL consists of a fully etched FeMn layer adjacent to a partially etched NiFe layer. The thickness of the remaining NiFe is labelled as $t_h$. A non-zero exchange bias field, $H_b$, is expected right underneath the FeMn layer while in the regions from which the FeMn has been removed the expected value for $H_b$ is zero. (b) SEM image of the NiFe(20 nm)/FeMn(10 nm) ADL fabricated. The red dashed square illustrates the ADL unitary cell whose side is 420 nm long. The hole diameter is 280 nm. The yellow dashed rectangle illustrates the orientation of the cross-section used in the morphology and elemental analysis presented in Fig. 2.
This shape is a result of the etch rates for the different elements during the RIE process. In Fig. 2(b), the regions labelled as I and II refer to the Ta/NiFe and the Ta/NiFe/FeMn/Ta, respectively. The elemental maps of each region are shown in Fig. 2(d). From analysing region I, the presence of Si, Ta, Ni, Fe, C, O, Pt and Ti were detected, from the substrate to the top of the film. Although it has not been possible to identify the origin of the Ti, it is believed that its origin is related to contamination during the etching process since this element only appears in regions exposed to the reactive ion etching process. The presence of C and O are the result of etching and the fact that this surface, rich in Ni and Fe, was not capped in any way, promoting the formation of oxides at the interface as well as a carbon layer. In region II, one is able to identify the elements Si, Ta, Ni, Fe, Mn, Ta and N. Based on the elemental composition shown in Fig. 2(c) relative to the several elements across the sample, the thickness is estimated for the case of the Ta, NiFe and FeMn layers. This is done by measuring the width at half height of the elemental distribution.

Magnetooptic Kerr effect (MOKE) magnetometry was employed to study the hysteretic behaviour of the continuous (a) and the patterned ADL (b) films. MOKE hysteresis loops are shown in Fig. 3 for both the (a) continuous NiFe/FeMn films and (b) ADL. The setup was operated in the longitudinal configuration, with a laser spot size of 500 µm in diameter. For the continuous film, the hysteresis was obtained while the external magnetic field was applied parallel (θ_H = 0°) and perpendicular (θ_H = 90°) to the exchange bias direction. The results show a clear easy axis direction with an exchange bias field magnitude H_b ~ 5 mT. In the case of the ADL, the hysteresis was measured with the external field applied along the lattice edges. For both θ_H = 0° and θ_H = 90° the exchange bias field magnitude |H_b| = 1.5 mT. In a patterned film, the low field magnetisation processes are dominated by the ADL anisotropy so the effect of exchange bias is not as trivial as a lateral shift in the hysteresis loop commonly observed in continuous exchange bias films. The fact that both directions exhibit a net exchange bias field suggests that a transformation of the exchange bias symmetry has occurred due to the ADL patterning. This aspect will be discussed when looking at the ferromagnetic resonance results.

### III. MODE STRUCTURE

Broadband ferromagnetic resonance spectroscopy was performed using a vector network analyser (Rohde & Schwarz ZVA40), VNA-FMR. The sample was placed on top of a coplanar waveguide with the system operating in a 2-port configuration. Each measurement was initiated well above the saturation field, at μ_0H = 150 mT, where a reference spectrum was acquired for background correction. Then, starting at a maximum applied field μ_0H = 65 mT, the VNA frequency was swept and the averaged (5 times) forward scattering parameter, S_{21} recorded. The static external magnetic field was linearly reduced once the frequency sweep was completed. This procedure was repeated in the applied field range of |μ_0H| ≤ 65 mT in field steps of 1.2 mT. As a final outcome, we obtained the relative variation of the magnitude of the parameter S_{21} as a function of the microwave frequency and external magnetic field.
The principle consists of the interaction of photons with a certain energy and momentum \((\hbar \omega_1, \hbar \vec{k}_1)\) with magnons \((\hbar \omega, \hbar \vec{k})\). The terms \(\omega\) and \(\vec{k}\) correspond to the frequency and wavevector of the incident photons and magnons. The annihilation or creation of optically excited magnons can be retrieved by measuring the energy and momentum transfer of the scattered photons \(\hbar \omega_{S}(\vec{k}_S) = \hbar (\omega_1(\vec{k}_1) \pm \omega(\vec{k}))\). BLS experiments were performed in the backscattering configuration using a Sandercock \((3+3)\)-type tandem Fabry-Perot interferometer. Spectra were acquired in the Damon-Eshbach (DE) scattering configuration \((\vec{k} \perp \vec{H})\). The wavelength of the incident laser light was \(\lambda = 532\) nm. Due to a photon-magnon conservation of momentum in the scattering process, the in-plane component of the excitation wavevector \((k)\) varies with the incidence angle of light \((\theta)\) according to \(k = \frac{(4\pi/\lambda)}{\sin \theta}\), where \(\lambda\) is the light wavelength. A static external field of \(\mu_0 H = 50\) mT was applied in the direction parallel to the lattice edge and collinear with the exchange bias direction, consistent with \(\theta_H = 0^{\circ}\) of Fig. 1(b).

**A. Field dependent magneto-dynamics using VNA-FMR and BLS**

The ferromagnetic resonance spectra shown as colour plot in Fig. 3 were obtained from the ADL sample,296 where the magnetic field is applied parallel to the lattice edge \((\theta_H = 0^{\circ})\) and the exchange bias direction. The full spectra contains four resonances and all are centered at around \(\mu_0 H \approx 0\) mT. The modes are labelled as I,294 CF, II and III, from lower to higher frequencies. The mode CF is labelled differently since its origin is related to the continuous film underneath the ADL (recall Fig. 1(a) and Fig. 2(a)). The modes I, II and III are intrinsically related to the patterned layer. Features worth noting are the field regions at which softening of modes I and II occur. These are indicated with arrows numbered as 1 and 2, respectively. For the case where the applied field is aligned with the edges of the lattice, the netisation reversal undergoes a hard-axis like behaviour,312 i.e. at a certain stage of the reversal, the external field cancels the effective anisotropy, allowing for local reorientation of the magnetic domains. Consequently the resonance frequency drops to a minimum value, as a result of the vanishing torque along the applied field direction.317 When the applied field is lower than the anisotropy field, the torque is restored and the resonance frequency increases. The regions in the spectra highlighted with dashed circles and numbered as 3 (\(\mu_0 H \approx 15\) mT) and 4 (\(\mu_0 H \approx 40\) mT) indicate the overlap between the resonance mode CF and modes I and II, respectively.319

One should also note the effect of exchange bias in the FMR response. This appears as a lateral displacement of the spectra and a noticeable asymmetry in resonance frequency between positive and negative applied fields. The asymmetric behaviour results from the unidirectional nature of the exchange bias field, which at positive applied fields shifts the resonance frequency downwards, whereas for negative fields the resonances are shifted upwards, giving rise to an asymmetry noted in the spectra by \(\Delta f_R\). For the case of mode I, an asymmetry \(\Delta f_R = 1\) GHz is obtained when evaluating the difference in resonance frequency at the applied field of \(\mu_0 H = 20\) mT and \(\mu_0 H = -20\) mT. A similar behaviour is observed for resonance mode II, where the asymmetry \(\Delta f_R = 0.4\) GHz for \(\mu_0 H = 20\) mT. The asymmetry is expected to be larger when \(\mu_0 H \approx H_k\) since that, due to mode softening, the exchange bias field becomes the only ordering parameter for the magnetisation. Field dependent BLS data (triangular symbols) is also shown in Fig. 4. Note the good agreement with the FMR data. The small deviations between the FMR and BLS data can originate from a possible misalignment of the sample relative to \(\theta_H = 0^{\circ}\). The wavevector dispersion studied using BLS is discussed in the following section.

**B. Spin wave dispersion using BLS**

Figure 5 shows the spin wave frequency dispersion (frequency vs wavevector) of the ADL (open circles) and the continuous exchange biased film (red squares). The
The largest wave number measured was $k_{\text{max}} = 2.6 \times \pi/a$, with $a$ being the hole spacing (420 nm), which, in the reciprocal space, corresponds to a wave vector just above the second Brillouin zone. When the light is focused at normal incidence upon the sample surface ($k = 0$), the modes agree well with the resonance peaks obtained from the VNA-FMR experiments. Similarly to the FMR results, at $k = 0$ the resonance mode CF overlaps with mode II from the ADL.

FIG. 5. BLS measurements at $\theta_H = 0^\circ$, while in the presence of an external field $\mu_0 H = 50$ mT, of the ADL (open circles) and the continuous film (red squares). For $k = 0$ the resonances match the FMR data. The periodicity of the lattice causes a change in the slope of the dispersion modes at $k = \pi/a$ and $2\pi/a$ as highlighted by the red dashed arrows. The magnonic band gap is highlighted by the grey shaded area at $(\pi/a \text{ rad/cm}^{-1}, \sim 10 \text{ GHz})$. The spin wave wave vector is perpendicular to the applied field direction (DE configuration).

In materials with modulated magnetic properties, the spin wave dispersion relation exhibits prohibited and allowed frequency bands, similarly to the case of electron scattering due to an atomic lattice or the diffraction of photons in the case of a photonic crystal. In Fig. 5, note the existence of a forbidden bandgap, 0.6 GHz wide, at $k = 0.77 \times 10^3 \text{rad cm}^{-1} (\pi/a)$, located at the boundary of the first Brillouin zone (grey shaded rectangle). The emergence of a band gap is evidence of repulsive interactions due to Bragg scattering of spin wave modes. Experimental evidence for Bragg scattering of spin waves was first demonstrated in Ref. 27 for the case of simple two-dimensional nano-structures. In Ref. 28 the spin wave dispersion for a type of bi-component ADL demonstrated the emergence of a BG. Reference 29 demonstrates that having a non-magnetic Ag ADL on top of a ferromagnetic CuZn continuous film can induce selectivity in the spin wave spectra. The results presented in the present manuscript show that the formation of band gap is also possible in the current geometry while in the presence of spatially modulated exchange bias field. The periodicity of the lattice is reflected in the BLS data, as can be seen by following the dispersion branches which have inflection points at the boundaries of the Brillouin zone. The frequency variation of these modes is highlighted by the red dashed arrows.

C. Magnetic anisotropies

In-plane angular dependent FMR measurements were performed in order to evaluate the anisotropic behaviour of the resonance modes. In the angular range of $\theta_H = [0^\circ - 190^\circ]$, FMR spectra were acquired every $10^\circ$, while around $[0^\circ, 45^\circ, 90^\circ]$ the step was reduced to $5^\circ$. Representative spectra are shown in Fig. 6. On all spectra the dashed lines represent the fit to each resonance mode using a Kittel-like resonance equation shown in Eq. 2.

The data corresponding to $\theta_H = 10^\circ$ shown in Fig. 6 is used to demonstrate the existence of two softening regions (black arrows) in the field range presented. In particular, at an applied field of $\mu_0 H \sim -15$ mT, the decrease in resonance frequency of mode I is associated with the balance between the external field and the effective anisotropy which includes the ADL anisotropy, $H_k$, due to patterning and the exchange bias field, $H_b$. The difference in resonance behaviour at positive and negative applied fields is related to the exchange bias field which, for positive applied fields, counts as a positive contribution to the anisotropy, whilst for negative applied fields, counts as a negative contribution.

In the low field regions, near $\mu_0 H \sim 0$ mT, the magnetisation undergoes the reversal process, as we have also observed in the MOKE data shown in Fig. 3. The reversal process is widely understood as mode softening followed by complete reversal of the magnetisation via domain formation and rotation. Similar resonance behaviour is observed when the applied field is set along, for example, $\theta_H = 20^\circ$ and $30^\circ$. In these spectra, the resonance frequency of mode I is higher when compared to that of $\theta_H = 0^\circ$, and therefore the resonance in the direction between $20^\circ$ and $30^\circ$ behaves like an easy-axis, as opposed to the directions $\theta_H = 0^\circ$, $45^\circ$, $90^\circ$ where a hard-axis behaviour is observed. It is important to note that in principle, the $\theta_H = 45^\circ$ hard-axis anisotropy is lower than the anisotropy along the directions $\theta_H = 0^\circ$, $90^\circ$, since the drop in the resonance frequency is less pronounced. This can be easily seen in Fig. 7 where we show a detailed analysis of the anisotropy field as a function of the applied field angle, $\theta_H$. A detailed description of the anisotropies of the different resonance modes is now presented and discussed. The anisotropy parameter for each mode was obtained by simultaneously fitting all the resonance modes obtained at each spectra. The multi-peak fit done in the field range within $60 \text{ mT} < |\mu_0 H| > 35 \text{ mT}$, thus considering only the saturated states. In this field range we assume that the domain textures which are inherent to low field region are suppressed due to the presence of a relatively large applied field magnitude. The fittings were performed on
the derivative of the experimental data so a derivative of the Lorentzian function was used to fit each resonance line, with an average $R^2$ of 0.88±0.04. An example of the fitting process is given in Appendix Sec. A. At each angle, the data was fitted with four resonance lines, as illustrated in Fig. 6 by the four dashed lines, allowing the assessment of the anisotropy field, the exchange bias field, and the effective magnetisation, following a generalised Kittel formula [34]:

$$f^2 = \gamma^2 (|H + H_b| + H_k)(|H + H_b| + H_k + \mu_0 M)$$  \hspace{1cm} (1)

where $\gamma$ is the electron gyromagnetic ratio. Assuming $\mu_0 M \gg |H + H_b| + H_k$, in Eq (1) leads to the approximation

$$f^2(\theta) \approx \gamma^2 (|H + H_b| + H_k) \mu_0 M_{eff},$$  \hspace{1cm} (2)

where $M_{eff}$ is the effective magnetisation. The anisotropy field as a function of angle, $H_a(\theta_H)$, obtained from fitting to the experimental data with Eq. (2) is plotted in Fig. 7. The results suggest the presence of an overlapping 8-fold an 4-fold anisotropy terms. The solid lines represent a fit to Eq (3) which accounts for an offset constant, $H_k^0$, and 4- and 8-fold anisotropy constants, which are labelled as $H_k^4$ and $H_k^8$, respectively. In particular for mode I, the 8-fold and 4-fold dependence can be clearly seen by following the continuous blue line. An uniaxial anisotropy constant was not used here given that both the CF mode and the continuous film (Appendix Sec. A) data exhibited a negligible anisotropy field.

$$H_k(\theta_H) = H_k^0 + H_k^4 \cos^2(2(\theta_H + \theta_0)) + H_k^8 \cos^2(4(\theta_H + \theta_0))$$  \hspace{1cm} (3)

$$H_b(\theta_H) = H_b^0 + H_b^1 \cos(\theta_H + \theta_0)$$  \hspace{1cm} (4)

Figure 7 shows the angular variation of the exchange bias field, $b$, for all modes. The continuous lines are fits of Eq. (1) to the experimental data. It is important to note that the angular variation of the exchange bias field is in agreement with the angular variation of a continuous exchange biased film [35], as also demonstrated using micromagnetic simulations (see Fig. A4(d) of Appendix Sec. C).

The exchange bias field magnitude obtained in mode I appears to change from positive to negative amplitude when $\theta_H$ changes by $22.5^\circ$. This angular dependence illustrates an interesting property of this magnetic system whereby the combined anisotropies give rise to an apparent anisotropic dependence of the exchange bias field with regards to the applied field direction, which may be associated with modifications in the coercivity due to the ADL structuring. Alternatively, one could consider the occurrence of deformations in the spin texture at the interface between the NiFe and the FeMn films, allowing...
for the exchange bias to behave as a rotatable anisotropy.\textsuperscript{466}
Having obtained a residual anisotropy for the CF mode\textsuperscript{467} and on the other obtained a good agreement between the\textsuperscript{468} Eq. 4 and the exchange bias field dependence across all\textsuperscript{469} field directions, we considered that effects such as rotatation\textsuperscript{470} able anisotropy and training effects are negligible in this\textsuperscript{471} exchange bias system. These effects would in any case be\textsuperscript{472} highly suppressed due to the strong local modification\textsuperscript{473} to the anisotropy imposed by the structuring. Moreover, in Sec.IV we will demonstrate that micromagnetic simulations support our decision to exclude such artefacts.\textsuperscript{506}
Although the mechanism for this is not fully understood,\textsuperscript{507} this feature could possibly enable the use of such systems\textsuperscript{480} as tunable microwave filtering devices.

\begin{table}[h]
\centering
\begin{tabular}{c|cccccc}
\hline
mode & $H_{b0}$ (mT) & $H_{b}^\theta (\deg)$ & $H_{b}^{\phi}$ & $H_{b}^{\phi} (\deg)$ & $M_{eff}$ \\
\hline
CF & -1.6 & 1.8 & -8.9 & -4.5 & 8.6 & 1.1 \\
I & 0.05 & 3.5 & -1.7 & -1.0 & 1.3 \\
II & -1.7 & 2.4 & 1.4 & -1.2 & -7.6 & 1.2 \\
III & -0.1 & 1.1 & 13.6 & 22.1 & 5.6 & 1.2 \\
\hline
\end{tabular}
\caption{Fitting parameters for the anisotropy, bias and effective magnetisation of the ADL. Units of the fitting parameters expressed in mT, with exception of $M_{eff}$, which is expressed in T. The fitting parameter $\theta_0$ is $\sim 11^\circ$.}
\end{table}

The angular variation of modes II and III is rather complicated to follow, given the strong dependence on the anisotropy and the overlapping of the modes. However, it is noted that the angular dependence of these two modes appears to be shifted by $45^\circ$ with regards to one another. Also, in the angular range of $\theta_H = [22.5^\circ - 67.5^\circ]$, it appears that the anisotropy field of mode III increases substantially, which suggests that in the VNA-FMR data this mode could be shifting to higher frequencies than covered in this measurement. The limited sensitivity of the VNA-FMR apparatus constrained observations at higher frequency. Thus, it is only possible to make a qualitative description of these modes. In Fig. 7 at angles corresponding to the range $\theta_H = [30^\circ - 50^\circ]$, two sets of symbols were added (square and circular shapes) to highlight two frequency modes which appear within the frequency and field range marked with the dashed grey rectangles. To aid the discussion, the region of the spectra where these two modes appear is shown in more detail in Fig. 9. The modes in the frequency region between 7-10 GHz are labelled as IV and V and highlighted with blue and green dashed lines, respectively, so that they are more easily followed. Note that when $\theta_H$ changes from $30^\circ$ to $40^\circ$, mode IV moves upwards in frequency, while mode V moves downwards. At $\theta_H = 40^\circ$ both modes intercept at $(\mu_0 H, f) \sim (14$ mT, $8.2$ GHz). In the spectra corresponding to $\theta_H = 45^\circ$ and $50^\circ$, modes IV and V continue their ascending and descending movements, respectively. The ascending movement of mode IV may be understood as the continuation of the mode III, as suggested by the trend of the anisotropy field discussed in Fig. 6. An alternative description is that mode III remains in the same frequency range as mode II. However, our interpretation is that mode III undergoes an increase in resonance frequency, whose movement is given by mode IV, which is consistent with the combined 8-fold and 4-fold anisotropy obtained from modes I and II and III. Mode V moves downwards in frequency with increasing $\theta_H$ and may be related to the resonance mode observed at $12.01$ GHz in the BLS spectra (Fig. 5). The fact that these two modes change rapidly with $\theta_H$ suggests great sensitivity to the anisotropy of the patterned structure.

Following the cross-sectional studies of Fig. 2 the NiFe remaining in the holes is approximately 14.5 nm in thickness and therefore it is expected to play a significant role in defining the FMR properties. With this in mind, a comparative micromagnetic study was performed in order to understand the effects of the partial patterning on the anisotropy dependence of the ADL system.

\section{Partial Patterning and Mode Analysis}

The FMR spectra as function of the applied field angle ($\theta_H$) were obtained from micromagnetic simulations using Mumax\textsuperscript{36}. The angular dependence was obtained at a fixed external field magnitude of 50 mT, thus vary-
FIG. 9. Resonance modes IV and V with low FMR amplitude appear the high frequency region. These spectra correspond to sections of the data shown in Fig. 6 with improved contrast. The arrows indicate the direction of motion with increasing $\theta_H$.

These spectra were obtained by performing a Fourier analysis of the time dependent out-of-plane component of the magnetisation, $m_z(t)$, after applying a spatially uniform field pulse in the form of $A_0 \text{sinc}(t-t_0)$, where $A_0=1$ mT and $t_0=3$ ns.

A broad study was performed, covering the angular variation of the FMR data for $t_h=0$, 5, 10, 15 and 20 nm. In this study, the magnetic properties, the hole size, and periodicity were kept constant, whilst varying the thickness, $t_h$, of the magnetic material in the holes (see Fig. 1(a)). The angular dependence of the resonance modes, in the case of $t_h=0, 5, 10$ and 20 nm are shown in Appendix Sec. C, in Fig. A4. In summary, we observed the emergence of an edge mode in the low frequency range at $t_h=0$ nm, whose resonance frequency tends to increase with an increasing $t_h$. In addition, we observed that for $t_h=0$ nm the first fundamental mode exhibits a 4-fold dependence, which is in agreement the literature. However, when considering $t_h=15$ nm, which is shown in Fig. 11, the resonance of the edge mode overlaps with that of the first fundamental mode and as a result, the angular dependence of the resonance frequency exhibits a combined 4-fold and 8-fold components. Under these conditions, we were able to qualitatively reproduce the angular dependence of mode I discussed in Fig. 7.

Note that the 8-fold symmetry emerges in the lowest frequency mode, M1. Additionally, it is also noted that the higher frequency modes, M4 and M5 exhibit 4-fold dependence shifted by $90^\circ$ with respect to one another. This is important to note as a similar trend was observed in the experimental data, when discussing the modes shown in Fig. 6. One can also note that along the main directions of the ADL ($\theta_H \sim 0^\circ$ and $90^\circ$), mode M3 may correspond to mode V of Fig. 9. Magnetic properties used in Mumax: $M_3=1$ T, $A_2=1.3 \times 10^{-12}$ J/m, Landau-Lifshitz damping constant $\alpha=0.02$ and $\theta_h=11.25^\circ$. The discretisation unit was set to 3.28 nm in the film plane and 5 nm along the direction perpendicular to the film.

FIG. 10. Angular variation of the FMR spectra for $t_h=15$ nm. The dark regions correspond to the resonance position. Symbols illustrate the position of the resonance modes M1-5 at $\theta_H=2.8^\circ$ and these can be associated with modes I, CF, II and III obtained experimentally, while M5 may correspond to mode V of Fig. 9. Magnetic properties used in Mumax: $M_3=1$ T, $A_2=1.3 \times 10^{-12}$ J/m, Landau-Lifshitz damping constant $\alpha=0.02$ and $\theta_h=11.25^\circ$. The discretisation unit was set to 3.28 nm in the film plane and 5 nm along the direction perpendicular to the film.

Figure 11 shows the spatial distribution of the resonance modes obtained at $\theta_H=2.8^\circ$ with an applied field magnitude of 50 mT, while considering $t_h=15$ nm. The normalised precession amplitude is colour coded in the images and each panel appears labelled with its corresponding precession frequency. It can be noted that in the lowest frequency mode, the mode amplitude at the edges of the holes (edge mode) overlaps with the amplitude profile corresponding to the first order mode which extends throughout the ADL. The coupling between these two modes is maintained along all directions since the existence of the continuous layer allows the first order mode to rotate uniformly with the applied field angle. The modes at higher frequencies correspond to
in the form of the magnetic anisotropies with 8-fold and 4-fold components, which we observed from the angular dependence of four resonance modes. This feature has not been explored so far in the literature as one usually expects a 4-fold component in ADL systems. The addition of the exchange bias field induces an asymmetric response of the resonance frequencies with regards to the applied field direction. These asymmetries are tunable, reaching up to 2 GHz for the lowest resonance frequency mode, and can be advantageous for microwave filtering applications. In addition, we observed that as a result of the local modifications due to structuring, the exchange bias field becomes highly dependent on the direction of the external applied field with regards to the ADL lattice, suggesting that a stepped reversal may be achieved by carefully controlling the lattice parameters and etch depth.

The TEM cross-sectional analysis allowed a detailed characterisation of the multi-layered system, especially in terms of the layer configuration and the morphology of the holes. In particular, it allowed to measure the thickness of the NiFe remaining in the holes (14.5 nm). This information helped producing an accurate micromagnetic picture which in turn allowed to understand the origin of the 8-fold anisotropy component. We have concluded that the 8-fold symmetry emerges due to an overlap between the edge mode and the first order extended mode. This may be understood from the perspective of the dipolar fields which emerge with structuring. Due to partial patterning, the dipolar interactions among the nearest holes and next-nearest holes become comparable and thus raising such anisotropic behaviour. The etching of only 5 nm has ensured that the nanostructured system maintains the properties of a magnonic crystal as the spin wave dispersion (frequency vs wave vector) measured by BLS reflects the periodicity of the ADL, and the emergence of a frequency band gap of 0.6 GHz, at the edge of the first Brillouin zone.

VI. ACKNOWLEDGEMENTS

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Appendix A: FMR data of the continuous films

To ensure that the magnetic properties of the films have not been affected by the temperature changes throughout the fabrication process, a comparison was made between the ferromagnetic resonance data relative to the continuous film as-deposited (AD) and after the patterning process (AP). The results demonstrated that the magnetic properties have not been altered throughout the patterning process by, for example the resist baking step. This can be seen from the comparison of the VNA-FMR data shown in Fig. A1. A summary of the fitting parameters is shown in Table I. As can be noted, the magnetic properties are identical for both AD and AP samples. The anisotropy field amplitude, \( H_b \), obtained from the fitting is small on both AD and AP films. The anisotropy field amplitude, \( H_b \), is set to a negligible value.

### Table I. Fitting parameters for the continuous films at the as-deposited (AD) and post-processing (AP) stages

<table>
<thead>
<tr>
<th>Sample</th>
<th>( H_b(T) )</th>
<th>( H_b(T) )</th>
<th>( M_{eff}(T) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD</td>
<td>0.005</td>
<td>0.001</td>
<td>1.16</td>
</tr>
<tr>
<td>AP</td>
<td>0.0052</td>
<td>0.0011</td>
<td>1.21</td>
</tr>
</tbody>
</table>

Figure A2 corresponds to an example the multi-peak fitting procedure. The derivative of the experimental data shown in Fig. A2(a) is fitted with four Lorentzian functions. All functions are fit simultaneously and across the whole field range. As we consider only the field region where the specimen is saturated, the fitted functions are also set to satisfy Kittel’s resonance equation. The initial conditions necessary for the fitting are the estimates of the parameters for the Kittel equation (\( M_{eff}, H_a \), and \( H_b \)) for each of the resonances and from there we obtain an estimate for the resonance peaks and line-widths. This set of initial conditions is then fit to the experimental data. An example of the fit is shown in Fig. A2(b). From the residual plot shown in Fig. A2(c) we note a good agreement between the experimental data and the fitted functions.

### Appendix C: Case study

Figure A3 demonstrates the agreement between the BLS data (blue markers) and the simulated FMR spectra, suggesting that the ground state for the micromagnetic simulations has been successfully determined. In particular, note the agreement in the three lowest resonance modes and the softening of the lowest mode, at around \( \mu_0 H = 20 \) mT. The disagreement in the low field region may be due to the existence of imperfections at the hole edges in the ADL which were not recreated in micromagnetic simulations. These imperfections will affect the reversal of the magnetisation via local pinning of domains.

Figure A4 shows the calculated resonance frequency as a function of the angle, \( \theta_H \), between the applied field and the edge of the ADL. From Fig. A4(a) to (d) the thickness of the material in the region of the holes, \( t_h \), is set to 0, 5, 10 and 20 nm, respectively. For \( t_h = 0 \) nm we recreate a fully etched ADL and the limiting case of \( t_h = 20 \) nm all resonances merge in a single mode, resembling the FMR response of the mode CF discussed in the Fig. 8. In Fig. A4(a), the high frequency resonance...
FIG. A3. (a) Simulated FMR data (colour plot) compared with BLS data (blue markers). Magnetic properties used in the micromagnetic simulations: $M_S = 1$ T, $A_{ex} = 1.3 \times 10^{-12} \text{ J/m}$ and $\alpha = 0.02$ and $\theta_b = 11^\circ$.

FIG. A4. (a)-(d) Angular variation of the resonance modes of the ADL for $t_h = 0$, 5, 10 and 20 nm. The exchange bias field direction was kept fixed at $\theta_b = 11.25^\circ$. Using Mumax, we simulated a $2 \times 2$ array and applied boundary conditions to ensure the finite dimensions of the system do not affect the results.

band appears labelled as HF (11 - 14 GHz), the intermediate frequencies as IF (5.0 - 11 GHz) and the lowest frequency band as LF (1.5 - 4.8 GHz). When comparing the results of the Fig. A4 (a)-(d), we observed the following: 1) the modes in the HF band become weaker in amplitude with the increase in $t_h$; 2) the frequency range of the IF band becomes narrower, the amplitude of the modes becomes dominant and the band itself appears to shift downwards in frequency, with increasing $t_h$; 3) for $t_h = 0$ nm the resonance mode in the LF band appears in the frequency range of 2-4 GHz, when $t_h = 5$ nm the IF band appears between 3-5 GHz and in the case of $t_h = 10$ nm the LF band shifts to a range of 5-6 GHz. Importantly, we note that the angular dependence of the LF band (dashed lines) evolves to an 8-fold symmetry, as $t_h$ increases.

Appendix D: Asymmetry

In section III C of the manuscript the anisotropic and biased magneto-dynamic response of the ADL was discussed. It was noted that only the $H_b(\theta_H)$ of mode CF follows the $\cos(\theta_H)$ behaviour of Eq. [4] which is normally obtained in continuous exchange biased films. The modes I, II, and II exhibit an oscillatory behaviour different to $\cos(\theta_H)$. In mode I the oscillation of $H_b(\theta_H)$ is more pronounced, with the largest exchange bias field amplitude obtained at $\theta_H = -10^\circ$, 10$^\circ$, 80$^\circ$ and 100$^\circ$, where $\Delta H_b \approx \pm 2 \text{ mT}$ (Fig. 8). At $\theta_H = 0^\circ$, the effect of the exchange bias field is not observable mainly because the spin configuration is dominated by the anisotropy of the ADL. By combining the anisotropy of the ADL and the exchange bias one adds a degree of tuning to the resonance frequencies, whereby positive and negative frequency shifts are obtained by small variation in the applied field angle. Although the mechanism is not fully understood, the change in the polarity of the bias field may be associated with the magnetic domain texture which forms during magnetisation reversal [33].

Figure A5 shows the difference (asymmetry), $\Delta f_R$, in resonance frequency between $\sim \mu_0 H = 20 \text{ mT}$ and $\mu_0 H = -20 \text{ mT}$, as a function of applied field angle. The applied field $|\mu_0 H| = 20 \text{ mT}$ represents a good approximation to the magnitude at which the softening of mode I occurs. The magnetic configuration at this applied field will be highly non-linear at certain angles due to the proximity to the hard-axis of the ADL, for example
at $\theta_H = \pm 10^\circ$. Given the 8-fold oscillation in the asymmetry of $\Delta f_{AS}$, we can confirm that the asymmetry is caused by the competition between ADL anisotropy and the exchange bias field. The asymmetry appears to have different behavior in the angle range $\theta_H \approx \pm (112.5^\circ - 135^\circ)$, possibly due to non-uniform reversal behavior of the magnetisation, caused by pinning near the edges of the holes which may give rise to complex domain structures.

The micromagnetic simulation results shown in Fig. A4 do not exhibit this change in polarity, as can be understood by comparing the spectra at $\theta_H = 10^\circ$ and $\theta_H = 350^\circ$, suggesting that by simply defining the exchange bias as a pinning field may be an incomplete approach to the problem of exchange bias in patterned structures. Local deformations of the magnetic domain structure imposed by the patterning of the holes or roughness at the interface between the Ni$_{50}$Fe$_{20}$ and Fe$_{50}$Mn$_{50}$ films may also occur, allowing for non-uniform magnetisation reversal.

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