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1 **Direct visualization of the magnetostructural phase transition in nano-scale**
2 **FeRh thin films using differential phase contrast imaging**

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4 Trevor P. Almeida^{1*}, Damien McGrouther¹, Rowan Temple², Jamie Massey², Yue Li¹, Thomas
5 Moore², Christopher H. Marrows², Stephen McVitie¹

6
7 ¹ SUPA, School of Physics and Astronomy, University of Glasgow, Glasgow, G12 8QQ, UK.

8 ² School of Physics and Astronomy, University of Leeds, Leeds LS2 9JT, UK.

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11 **ABSTRACT**

12 To advance the use of thermally-activated magnetic materials in device applications it is necessary to
13 examine their behaviour on the localised scale *operando* conditions. Equi-atomic FeRh undergoes a
14 magnetostructural transition from an antiferromagnetic (AF) to a ferromagnetic (FM) phase above
15 room temperature (~ 350 to 380 K) and hence is considered a very desirable material for the next
16 generation of novel nanomagnetic or spintronic devices. For this to be realised, we must fully
17 understand the intricate details of AF to FM transition and associated FM domain growth on the scale
18 of their operation. Here we combine *in-situ* heating with a comprehensive suite of advanced
19 transmission electron microscopy techniques to investigate directly the magnetostructural transition in
20 nano-scale FeRh thin films. Differential phase contrast imaging visualizes the stages of FM domain
21 growth in both cross-sectional and planar FeRh thin films as a function of temperature. Small surface
22 FM signals are also detected due to interfacial strain with the MgO substrate and Fe deficiency after
23 HF etching of the substrate, providing a directional bias for FM domain growth. Our work provides
24 high resolution imaging and quantitative measurements throughout the transition, which were
25 previously inaccessible, and offers new fundamental insight into their potential use in magnetic
26 devices.

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35 * Correspondence to: trevor.almeida@glasgow.ac.uk

36 I. INTRODUCTION

37 The ordered α'' alloy of iron-rhodium ($\text{Fe}_{48}\text{Rh}_{52}$ to $\text{Fe}_{56}\text{Rh}_{44}$) has gained significant interest due to its
38 magnetostructural transition from its antiferromagnetic (AF) to ferromagnetic (FM) phase¹. This equi-
39 atomic, CsCl-structured α'' alloy undergoes a first-order transition from its room-temperature AF state
40 to FM state between ~ 350 to 380 K, which is accompanied by a 1% lattice volume expansion².
41 Hence, nano-scale thin films of FeRh can present AF / FM phase co-existence and hysteresis, where
42 the transition temperature and associated thermal hysteresis depend on the film thickness³, lattice
43 strain behaviour and additional chemical doping⁴. At intermediate stages during the transition, the co-
44 existing AF / FM phase regions dynamically evolve or disintegrate with temperature and are
45 separated by phase boundary (PB) domain walls (DWs). Exploitation of the PB motion and its
46 effective control is considered very desirable for the next generation of spintronic devices. For
47 example, the PBs can be systematically driven by heating FeRh films grown with differential
48 gradients of elemental Ir- and Pd-doping, as determined by a corresponding change in resistivity⁵.
49 However, the dynamical FM domain nucleation, growth and coalescence stages on heating, and
50 subsequent separation, disintegration and annihilation stages on cooling, as well as the PB behaviour,
51 are not accessible by bulk magnetic measurements.

52 Imaging techniques sensitive to magnetic structure and with spatial resolution in the 10's of
53 nanometres range, including magnetic force microscopy^{6,7}, X-ray magnetic circular dichroism
54 (XMCD)^{7,8} and scanning electron microscopy with polarization analysis (SEMPA)⁹ have been used to
55 observe the phase coexistence in FeRh thin films, showing the first order transition from the
56 nucleation of domains regime to be distinct from the domain growth regime^{8,10}. Further, X-ray
57 photoemission microscopy in XMCD has revealed the effects of lateral confinement of FeRh islands
58 on the transition temperature, resulting in a 20 K variation in transition temperature in small islands,
59 and showed that their ion-beam-damaged edges act as favourable nucleation sites¹¹. Nevertheless,
60 these techniques are typically limited to a spatial resolution of ~ 20 - 30 nm and penetration depth of a
61 few nm^{12,13}. Hence, in order to elucidate fully the localised and dynamic domain evolution /
62 dissipation throughout the magnetostructural transition with sufficient detail, it is necessary to
63 investigate the thermally-induced domain growth at the highest spatial resolution during *in-situ*
64 heating.

65 Aberration-corrected transmission electron microscopy (TEM) techniques are well known for
66 enabling the imaging of both physical and chemical structure of sufficiently thin, electron transparent,
67 samples with atomic-scale spatial resolution. High spatial resolution magnetic imaging can also be
68 performed utilising a family of techniques collectively known as Lorentz microscopy. These include
69 Fresnel imaging¹⁴⁻¹⁶, off-axis electron holography¹⁷⁻²⁰ and differential phase contrast (DPC)
70 imaging^{16,21,22}. Recent advancements of modern aberration-corrected TEMs have also improved the

71 spatial resolution of magnetic imaging to approach $\sim 1 \text{ nm}^{23}$. Fresnel imaging has revealed the FM
72 domain structure in high quality 2D planar-view FeRh thin films produced for magnetic imaging
73 within the TEM²⁴. Electron holography has also exposed an inhomogeneous spatial distribution of the
74 transition temperature along the growth direction in cross-sectional FeRh thin films, as well as a
75 regular spacing of the nucleated FM domains²⁵. However, preparation of cross-sectional thin film
76 TEM lamellae is inherently destructive and shape anisotropy dominates magnetically compared to its
77 continuous film state. Accordingly, conventional DPC imaging of planar FeRh thin films has provided
78 quantitative measurements from individual DWs as a function of temperature, including a general
79 overview of the nucleation, growth and coalescence stages during the transition²⁶. Yet this type of
80 conventional DPC imaging is susceptible to unwanted signal variation due to strong diffraction
81 contrast from grain boundaries and crystallinity, as it employs segmented detectors to measure
82 differential signals that relate to in-plane magnetism but which can also arise from crystallographic
83 directional scattering. To overcome this, the recent advent of fast direct electron pixelated detectors
84 has revolutionised the ability to acquire large amount of images in relatively short time periods. DPC
85 type imaging is performed by capturing images of the transmitted electron disc for every electron
86 beam scan location with advanced data processing, based on disc edge detection, enabling isolation of
87 small Lorentz deflections from directional artefacts introduced by diffraction contrast²⁷. This
88 technique belongs to a wider, relatively new, family of techniques termed “4D STEM”. Thus, when
89 reporting results from both DPC techniques we refer to these as being from segmented detector DPC
90 or 4D STEM DPC.

91 Herein, this study employs a comprehensive suite of advanced magnetic TEM imaging and
92 scattering techniques to investigate the magnetostructural transition in cross-sectional and planar
93 FeRh thin films, as a function of temperature. Conventional TEM characterisation confirms their
94 chemical and structural properties, whilst segmented detector DPC imaging reveals the origin of a
95 small FM signal at room temperature and FM domain growth of cross-sectional FeRh films during
96 heating. Localised insight into the AF to FM domain evolution within a planar FeRh film, and
97 subsequent reversal, is provided by three complementary Lorentz microscopy techniques to
98 investigate the nature of the appearance/disappearance of the FM phase. Defocused Fresnel imaging
99 in TEM mode was utilised to obtain high contrast, real space imaging of the phase transition. Small
100 angle electron scattering (SAES)²⁸ provided a counterpart quantitative, reciprocal space, analysis of
101 the same regions imaged by Fresnel. Lastly, advanced 4D STEM-based²⁷ DPC imaging was used to
102 obtain high-spatial-resolution images of the domain structures and states occurring during the
103 transition. Taken together, these reveal new fundamental details of the mechanisms associated with
104 the AF to FM phase transition at the highest spatial resolution, which were previously inaccessible.

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109 **II. MATERIALS AND METHODS**

110 **1. Fabrication**

111 Ordered α'' -FeRh alloy thin films were grown epitaxially on a clean (001) MgO substrate or (001)
112 NiAl, GaAs and AlAs multi-layered buffer layers on a GaAs substrate by conventional DC magnetron
113 sputter co-deposition, as described previously²⁹. We studied two TEM specimens. Firstly, a cross-
114 sectional FeRh TEM specimen was prepared from its bulk MgO substrate and transferred onto *in-situ*
115 heating electronic (e-) chip (DENSsolution WildfireTM) by Ga⁺ focused ion beam (FIB) methods²⁴.
116 After FIB preparation, the cross-sectional sample was annealed at 873 K under vacuum in the TEM
117 for 1 hour to recover any damage to the FeRh structure from Ga⁺ implantation. A second planar FeRh
118 TEM specimen was prepared through a process of HF-etching of the AlAs, GaAs and NiAl buffer
119 layers, as well as GaAs substrate³⁰, and subsequently transferred onto a Cu TEM grid for *in-situ*
120 heating using a Gatan heating TEM holder.

121

122 **2. Electron microscopy**

123 All the imaging, diffraction and spectroscopy described in this paper were carried out on a JEOL
124 Atomic Resolution Microscope (JEM-ARM200F) TEM, operating at 200 kV²³. This microscope is
125 equipped with a cold field emission gun and a CEOS (Corrected Electron Optical Systems GmbH)
126 probe corrector for STEM imaging. Conventional and HR STEM imaging were performed on cross-
127 sectional and planar TEM samples of the FeRh films / substrates, whilst SAED acquired in TEM
128 mode provided structural information. Both EDX and EELS provided chemical analysis of the
129 samples. The sample thickness was determined by the spectrum imaging technique in STEM mode³¹,
130 whereby low-loss EELS spectra acquired from each pixel were used to calculate values of t/λ . These
131 calculations were performed using the Digital MicrographTM software package. The mean free path, λ ,
132 was determined from the density of equi-atomic FeRh and the TEM beam conditions, *i.e.*,
133 accelerating voltage, convergence and divergence angles, *etc.* and substituted into t/λ to calculate the
134 relative thickness repetition, with a standard deviation of 6%. The magnetic structure of the FeRh
135 films was visualised using segmented and 4D STEM DPC in Lorentz mode under low-magnetic field
136 conditions. DPC imaging was carried out with the HR objective lens pole piece switched off, with the
137 samples positioned in the low-strength remanent field of the lens (~ 12 kA/m). An 8-segment silicon
138 photodiode array detector (supplied by DebenUK Ltd) was used for the segmented DPC imaging. The
139 signal from the detectors was converted and amplified using the "Superfast" amplifier (Andrew Armit
140 Designs). The 8 detected signals were acquired, mixed and displayed via four Gatan DigiscanII units.
141 For 4D STEM DPC imaging, the central diffraction disk was recorded at each scan position by a
142 Medipix3 pixelated detector, whilst using a 50 μ m condenser aperture and camera length of 800cm.

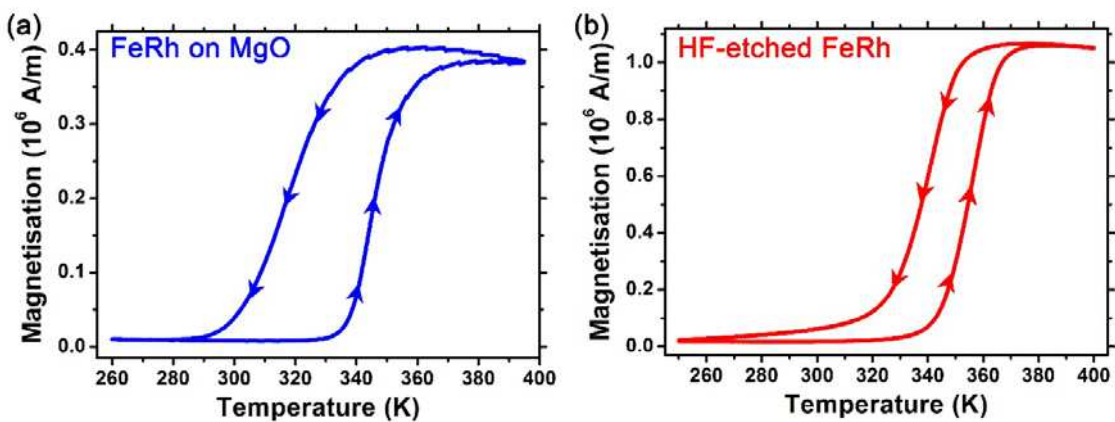
143 The cross-correlation disk deflection registration method was used to reconstruct the 4D STEM DPC
 144 images²⁷. In addition, combining DPC with *in-situ* heating using the DENSSolution e-chips (up to 473
 145 K) or Gatan heating holder (up to 413 K) allowed for direct access to the thermomagnetic behaviour
 146 of the DWs and magnetostructural transition within the FeRh films. SAES patterns were acquired in
 147 TEM mode using a 30-micrometre diameter condenser aperture which yielded parallel illumination of
 148 a circular region of the sample, 15 microns in diameter. A custom post specimen lens excitation series
 149 was used to either obtain Fresnel images or SAES patterns with 460m camera length at each
 150 temperature step. Switching between the imaging / SAES mode was performed by only adjusting the
 151 strength of the first lens immediately after the main imaging lens.

152

153 III. RESULTS

154 To provide an overview of the magnetostructural transition in the two FeRh thin films, Figure 1
 155 presents their SQUID-VSM measurements as a function of temperature. The FeRh on MgO substrate
 156 (Fig. 1a) exhibits a sharp increase in magnetisation from ~ 10 kA/m at ~ 330 K to ~ 380 kA/m at ~ 370
 157 K, with a noticeable asymmetrical hysteresis curve, where the reduction of magnetisation occurs at \sim
 158 335 K on cooling. Similarly, the HF-etched FeRh thin film (Fig. 1b) exhibits an increase in
 159 magnetisation from ~ 40 kA/m at ~ 320 K and stabilises to ~ 1060 kA/m at ~ 380 K, with a ~ 20 K
 160 difference in the symmetric hysteresis curve on cooling. Both samples present hysteresis as expected
 161 with a first-order transition, but it is noteworthy that the HF-etched FeRh sample exhibits a slightly
 162 higher moment in the AF state compared to the FeRh on MgO, as well as a significantly higher
 163 magnetisation in the FM state (~ 1100 kA/m $\gg \sim 380$ kA/m). It is also considered that since the latter
 164 FeRh thin film is clamped to the MgO substrate, the change in lattice parameter of the FeRh is more
 165 reliant on the thermal expansion of the substrate. This produces a larger hysteresis compared to the
 166 HF-etched FeRh thin film, which is less restricted to provide a thermomagnetic response.

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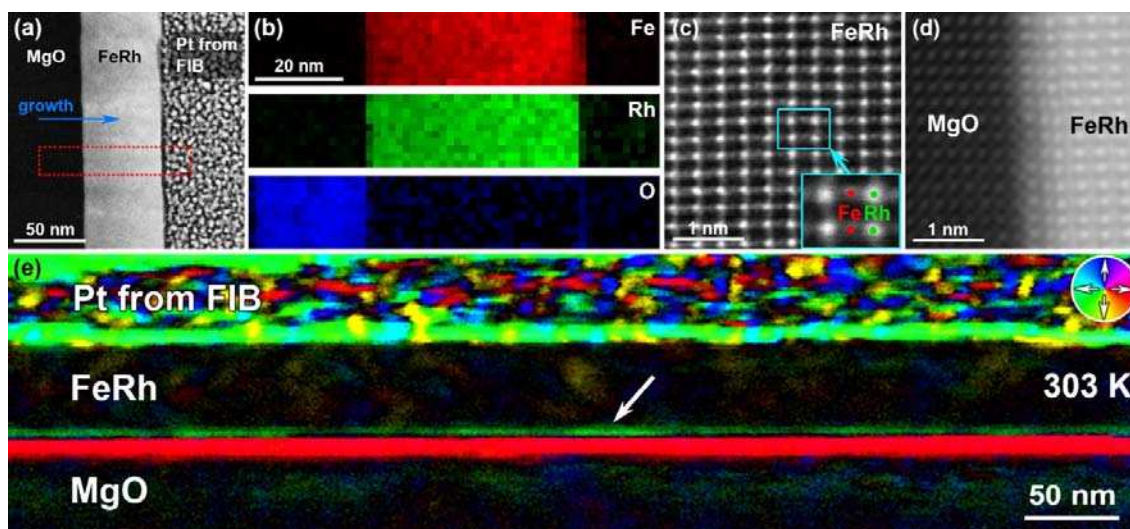
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169 **Figure 1. Magnetization (M) vs. temperature (T) plots.** (a) FeRh thin film on MgO substrate; and (b) free-
 170 standing FeRh thin film after HF-etching of its AlAs, GaAs and NiAl buffer layers, as well as the GaAs
 171 substrate.

172

173 Figure 2 presents a cross-sectional view of the FeRh thin film grown on the MgO substrate,
174 providing information on its thickness, chemical distribution, localised structure and interfacial
175 magnetism with the MgO substrate. The high angle annular dark-field (HAADF) STEM image of Fig.
176 2a reveals the FeRh film to be grown with a uniform thickness of ~ 53 nm, whilst the electron-energy
177 loss spectroscopy (EELS) chemical maps (Fig. 2b) acquired from the boxed region (red) in Fig. 2a
178 display the elemental distribution of iron, rhodium and oxygen. The high resolution (HR) STEM
179 image of Fig. 2c presents the localised CsCl crystal structure of the FeRh thin film along the $\langle 110 \rangle$
180 zone axis, where the alternating columns of Rh atoms appear brighter compared to the Fe atoms due
181 to their higher atomic number (Fig. 2c, inset). Similarly, Fig. 2d displays the localised interface
182 between the single crystalline FeRh and MgO substrate, revealing their well-matched orientation and
183 confirming the epitaxial growth of the deposited FeRh. Segmented detector DPC imaging, presented
184 in Fig. 2e, provides low temperature (303 K) magnetic information on the FeRh thin film. Within the
185 FIB Pt protective layer, multi-coloured contrast is observed which is purely electrostatic in origin and
186 arises due to it containing many grain boundaries. Within the FeRh layer, there is an absence of strong
187 contrast, commensurate with it being in the AF state at this temperature. Strong contrast is observed
188 between the FeRh layer and its interfaces. At the interface with the FIB Pt protective layer, this
189 contrast relates to the sharp electron wave phase gradient which arises due to the two materials having
190 different mean electrostatic potential (analogous to refractive indices in optics). This gives rise to a
191 single green coloured band that indicates the strength and direction of this phase gradient which is
192 purely electrostatic in origin. The situation at the interface between the FeRh/MgO substrate appears
193 to be more complicated. As for the previously discussed interface, a change in the mean electrostatic
194 potential between the two materials would be expected to contribute a single coloured band. However,
195 here, both a strong red coloured band and a weaker green coloured band (indicated by an arrow) are
196 observed. We ascribe the strong red band to being electrostatic in origin but the weaker green band to
197 being associated with the existence of a finite magnetic moment over a region ~ 8 nm wide. This is
198 explained in more detail in the Supplementary Material³² (Fig. S1-S3) and the Discussion section. In
199 order to isolate the magnetic contrast induced by thermal effects during the magnetostructural
200 transition, Fig 2e acts as a reference and is subtracted from DPC images in the temperature series
201 presented in Figure 3. The steps for this subtraction process are also described in the Supplementary
202 Material³² (Fig. S4).

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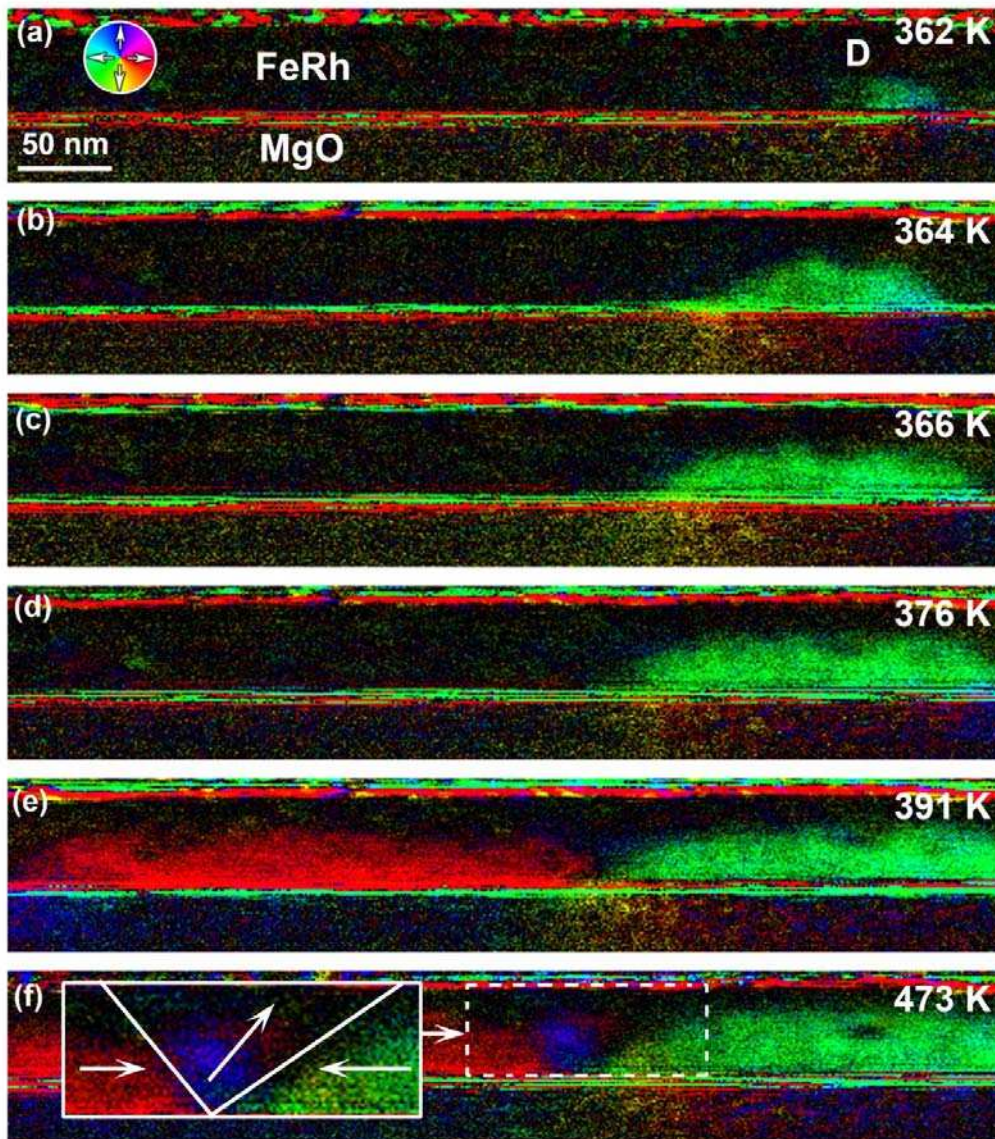
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Figure 2. Overview of the TEM cross-section of the FeRh thin film grown on a MgO substrate. (a) HAADF-STEM image of the cross-sectional TEM lamella showing the FeRh thin film grown on the MgO substrate. (b) EELS chemical maps acquired from the box region in (a) displaying the elemental distribution of iron (red), rhodium (green) and oxygen (blue). (c,d) High-resolution HAADF-STEM images showing the localised structure of the (c) FeRh thin film with labelled atoms (inset); and (d) its interface with the MgO substrate. (e) Segmented detector DPC image of the FeRh thin film, revealing magnetic contrast at its interface with the MgO substrate (~ 8 nm green layer, arrowed). The direction of magnetization is depicted in the colour wheel (inset).

215 The segmented detector DPC images of Figure 3 present the magnetic domain evolution
216 associated with the magnetostructural transition of the FeRh thin film as a function of temperature.
217 Fig. 3a reveals nucleation of a small, green magnetic domain (~ 50 nm long, ~ 20 nm wide, labelled
218 'D'), with magnetisation directed from right to left, on the right side of the FeRh / MgO interface at
219 362 K. As the temperature is increased to 364 K the small domain is observed to act as a nucleation
220 site for expansion to the left (Fig. 3b), and further growth at 366 K (Fig. 3c) and 376 K (Fig. 3d).
221 Weak contrast associated with the domain's resulting dipolar magnetic field (yellow to blue) in Fig.
222 3a-d is observed in the MgO substrate as the magnetostructural transition proceeds. At 391 K, a large
223 magnetic domain (red) is observed to form on the left hand-side of the FeRh thin film (Fig. 3e), with
224 the magnetisation pointing from left to right. As the temperature is increased to 473 K, the two larger
225 domains (red and green) are seen to be separated by a head-to-head transverse DW (blue, inset), like
226 those seen in permalloy nanowires¹⁴. It is considered that the top region of the FeRh thin film has not
227 fully recovered its α'' structure after annealing, resulting in the domains not progressing towards the
228 very top layer.

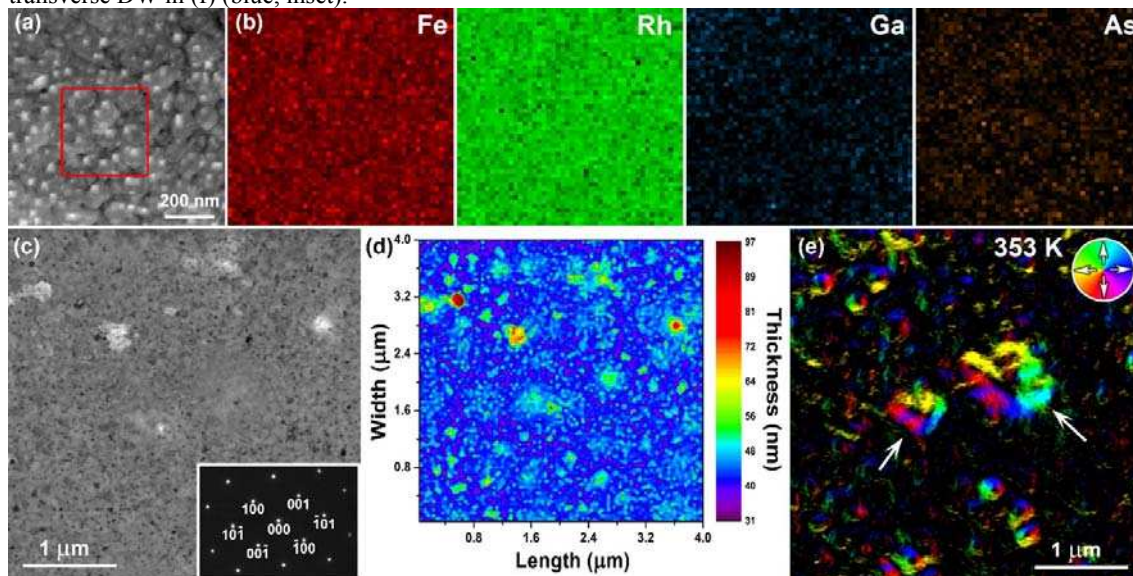
229 Figure 4 presents a planar view of the HF-etched FeRh thin film, providing details of its
230 surface, morphology, chemistry, relative thickness and magnetism. The DF STEM image of Fig. 4a
231 reveals the FeRh thin film to exhibit a non-uniform surface and morphology, with variations in
232 contrast attributed to debris on the surface. The EDX chemical maps (Fig. 4b) acquired from the
233 boxed region (red) in Fig. 4a display the elemental distribution of iron, rhodium, gallium and arsenic,

234 revealing a relatively uniform distribution of rhodium and iron, traces amounts of gallium (Ga-1%at)
235 and small concentrated areas of arsenic (As-3%at). The arsenic-rich areas coincide well with the
236 surface debris in Fig. 4a and are expected to be caused by the HF-etching process. Fig. 4c presents a
237 HAADF image of a large square area ($\sim 4 \mu\text{m} \times \sim 4 \mu\text{m}$) and the SAED pattern (Fig. 4c, inset)
238 confirms that the HF-etched FeRh film is single crystalline. Fig. 4d presents the thickness map
239 acquired from the entire region of Fig. 4c, where the relative thickness ranges from $\sim 85 \text{ nm}$ at the red
240 spots, artefacts attributed to grains lying on a crystallographic zone axis which scatter the electron
241 beam away from HAADF detector, to a more uniform thickness of ~ 40 to $\sim 45 \text{ nm}$. The DPC image
242 of Fig. 4e reveals the presence of several magnetic domain structures (arrowed), including vortices,
243 when heated to 353 K, while most of the sample is in the AF state, represented by the black regions,
244 where no magnetic deflections are detected.
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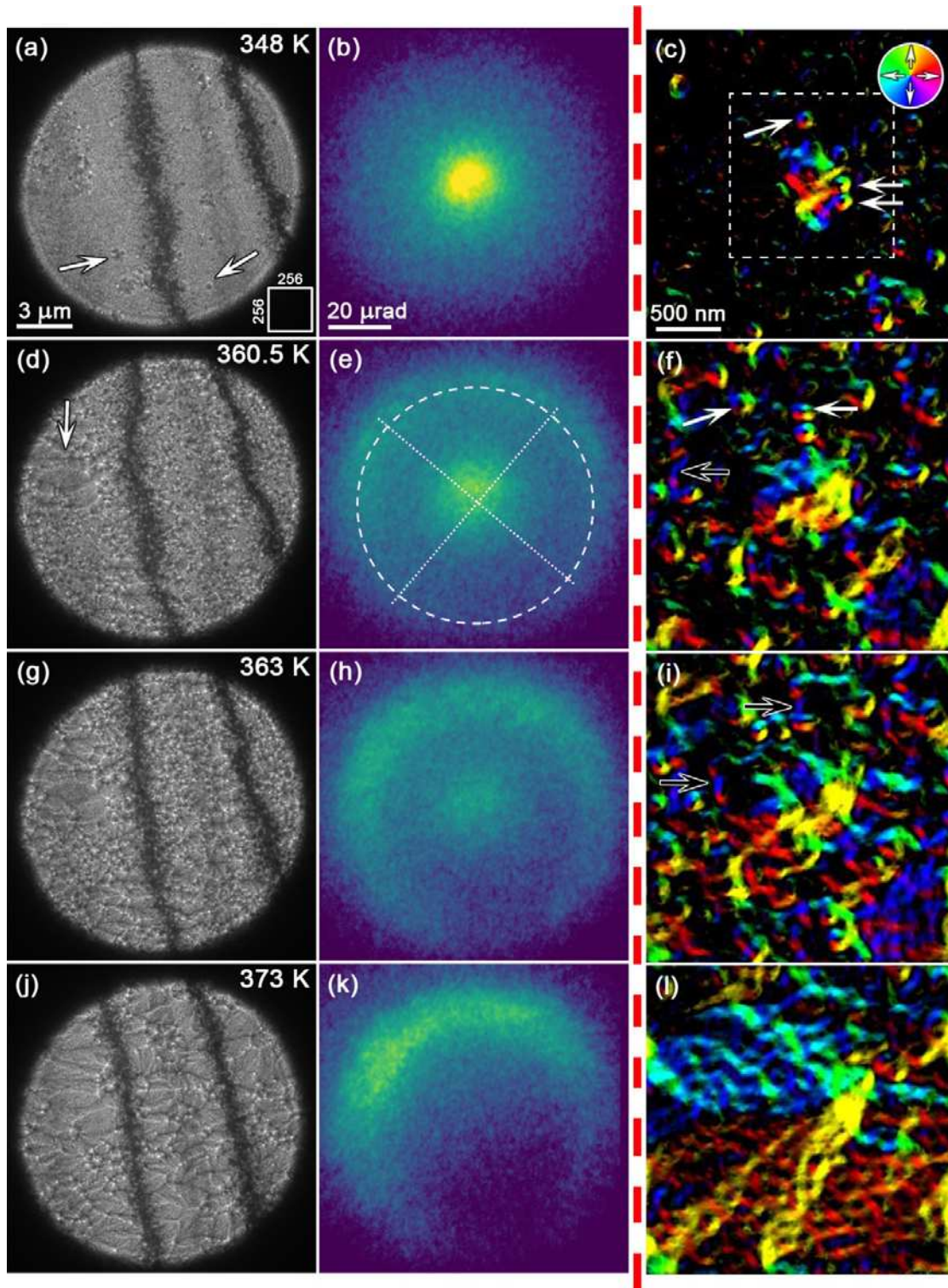
247 **Figure 3. Segmented detector DPC imaging of magnetic domain evolution in the FeRh thin film as a**
 248 **function of temperature.** (a-f) DPC imaging of the cross-sectional TEM sample of the FeRh thin film / MgO
 249 substrate during *in-situ* heating to (a) 362 K; (b) 364 K; (c) 366 K; (d) 376 K; (e) 391 K; and (f) 473 K. The
 250 direction of magnetization is depicted in the colour wheel (a, inset) and is illustrated in the head-to-head
 251 transverse DW in (f) (blue, inset).



252 **Figure 4. Overview of the planar FeRh thin film after HF-etching of its AlAs, GaAs and NiAl buffer**
 253 **layers, as well as the GaAs substrate.** (a) DF STEM image of the HF-etched planar FeRh thin film. (b) EDX
 254 chemical maps acquired from the box region in (a) showing the distribution of iron, rhodium, gallium and
 255 arsenic. (c) HAADF STEM image showing several white spots that are attributed to grains lying on a zone axis
 256 which deflect the electron beam away from HAADF detector, with the SAED (inset, bottom right). (d)
 257 Thickness map calculated from the low-loss EEL spectrum acquired from the entire area of (c), with small
 258 regions of large thickness (red) coinciding with the white spots seen in (c). (e) DPC image of the same region in
 259 (c&d) during *in-situ* heating, showing the presence of a magnetic domain structures at 353 K (arrowed), as well
 260 as small vortices.
 261
 262

263 Figure 5 provides direct visualisation of the thermomagnetic behaviour displayed by the HF-
 264 etched planar FeRh thin film using complementary techniques of Fresnel TEM imaging, small-angle
 265 electron scattering (both acquired sequentially from the same sample region) and 4D STEM DPC
 266 imaging (acquired from the same sample but from a different location). The relative scale of the 4D
 267 STEM scan region (256 x 256 pixels) to the Fresnel images is denoted in the bottom-right corner of
 268 Fig. 5a. The Fresnel TEM image of Fig. 5a, acquired at 348 K, shows a large area of the planar film
 269 (~ 15 μm in diameter) which includes two relatively strong contrast bend contour features (dark, near
 270 vertical bands) and Fresnel contrast that indicate the presence of small vortex structures (arrowed).
 271 The corresponding SAES pattern (Fig. 5b) is acquired from the same area shown in Fig. 5a (obtained
 272 by alteration of the excitation of only the post-specimen lenses of the TEM) and reveals that the
 273 intensity of the electron beam is concentrated in the central spot, recording minimal dispersion from
 274 FM domains. This is supported by the associated 4D STEM DPC image acquired at 348 K (Fig. 5c),
 275 which documents the existence of vortex-like structures (~ 130 nm, white arrows) in close proximity
 276 to a more complex domain (centre of Fig. 5c). Nevertheless, the majority of the DPC image is covered
 277 by dark regions and provides complementary evidence indicating that the FeRh thin film is mostly in

278 the AF state. After increasing the temperature to 360.5 K, the Fresnel TEM image (Fig. 5d) showed a
279 significant increase in vortex state nucleation across the entire illuminated area, as well as formation
280 of larger magnetic domains elongated along the orthogonal x-axis (arrowed) and slight variation in the
281 bend contours.



283 **Figure 5. Magnetic domain evolution in the planar FeRh thin film as a function of temperature during *in-***
284 ***situ* heating to 373 K.** (a,d,g,j) Fresnel; (b,e,h,k) SAES; and (c,f,i,l) 4D STEM DPC images of a HF-etched
285 FeRh thin film acquired at (a-c) 348 K; (d-f) 360.5 K; (g-i) 363 K; and (j-l) 373 K. The images demonstrate the
286 different stages of the magnetostructural transition, including (a-c) nucleation of magnetic vortices (arrowed);
287 (d-f) AF / FM phase co-existence; (g-i) domain coalescence; and (j-l) a fully FM state. The relative scale of the
288 256 x 256 4D STEM DPC images to the Fresnel images is denoted in the bottom-right left corner of Fig. 5a,
289 while the associated direction of magnetization is depicted in the colour wheel (Fig. 5c, inset). The
290 superimposed crosshairs in (e) are used to show the geometric centre of the SAES pattern.
291 Fig. 5e reveals the accompanying development of the SAES pattern through the redistribution of
292 intensity from the central spot to the outer concentric ring. Further insight is provided by the
293 corresponding 4D STEM DPC image (Fig. 5f), revealing nucleation of additional FM vortex
294 structures (arrowed, white) and their interconnection via small, uniformly magnetised ‘string’
295 domains (arrowed, black). Fig. 5g demonstrates that increasing the temperature further, to 363 K,
296 further nucleation, and string domains (arrowed, black) are induced which coalesce into larger
297 magnetic domains. The SAES pattern of Fig. 5h shows that most intensity is now located in the outer
298 ring rather than the central spot, indicating that the film is in a mostly FM state with a full range of
299 magnetic orientations present. This description of the state is supported by the 4D STEM DPC image
300 of Fig. 5i. The Fresnel TEM image of the Fig. 5j shows that heating to 373 K promotes agglomeration
301 into large, fully formed FM domains and reveals the bend contours to be straighter and more parallel
302 compared to Fig. 5a. All the electron intensity in the corresponding SAES pattern (Fig. 5k) is
303 segregated to the outer top-left arced ring pattern, which indicates the sample is in the fully FM state
304 at 373 K, supported by the existence of large, fully-formed FM domains in the 4D STEM DPC image
305 (Fig. 5l).

306 In a similar fashion to Figure 5, Figure 6 charts the magnetostructural transition of the HF-
307 etched planar FeRh thin film during stages of cooling from the FM state. The large FM domains in the
308 Fresnel and 4D STEM DPC images of Fig. 6a and Fig. 6c, respectively, and outer arced ring pattern
309 of the corresponding SAES pattern (Fig. 6b) suggests the FeRh is in the fully-FM state at 353 K,
310 revealing hysteresis in the transition during cooling compared to heating. It is clear that the FM
311 domains start to disintegrate at 343 K (Fig. 6d,f), as evidenced by electron intensity migrating towards
312 the centre of the SAES pattern (Fig. 6e). The FM to AF transition proceeds further when cooled to
313 338 K, with almost all the large FM domains in Figs. 6g and 6i transforming into smaller vortex or
314 ‘string’ states, and increased intensity in the central spot of the SAES pattern (Fig. 6h). The bend
315 contours are observed to increasingly widen and ripple in Figs. 6g and 6j, with very few magnetic
316 domains still present in the latter at 333 K. The majority of electron intensity in the SAES pattern
317 acquired at 333 K (Fig. 6k) is concentrated in the central spot, with the associated 4D STEM DPC
318 image of Fig. 6l displaying only a few vortex or string domain states.

319 Figure 7 presents quantitative analysis of selected SAES patterns acquired during heating
320 through the AF to FM transition. The SAES pattern of Fig. 7a, at 358 K, contains two contributions:
321 the central diffuse spot and the outer ring. For the central spot, the peak intensity was located at a
322 deflection angle, β_i , of 6.4 μ rad from the geometric pattern centre (denoted by crosshairs in Fig. 5e).

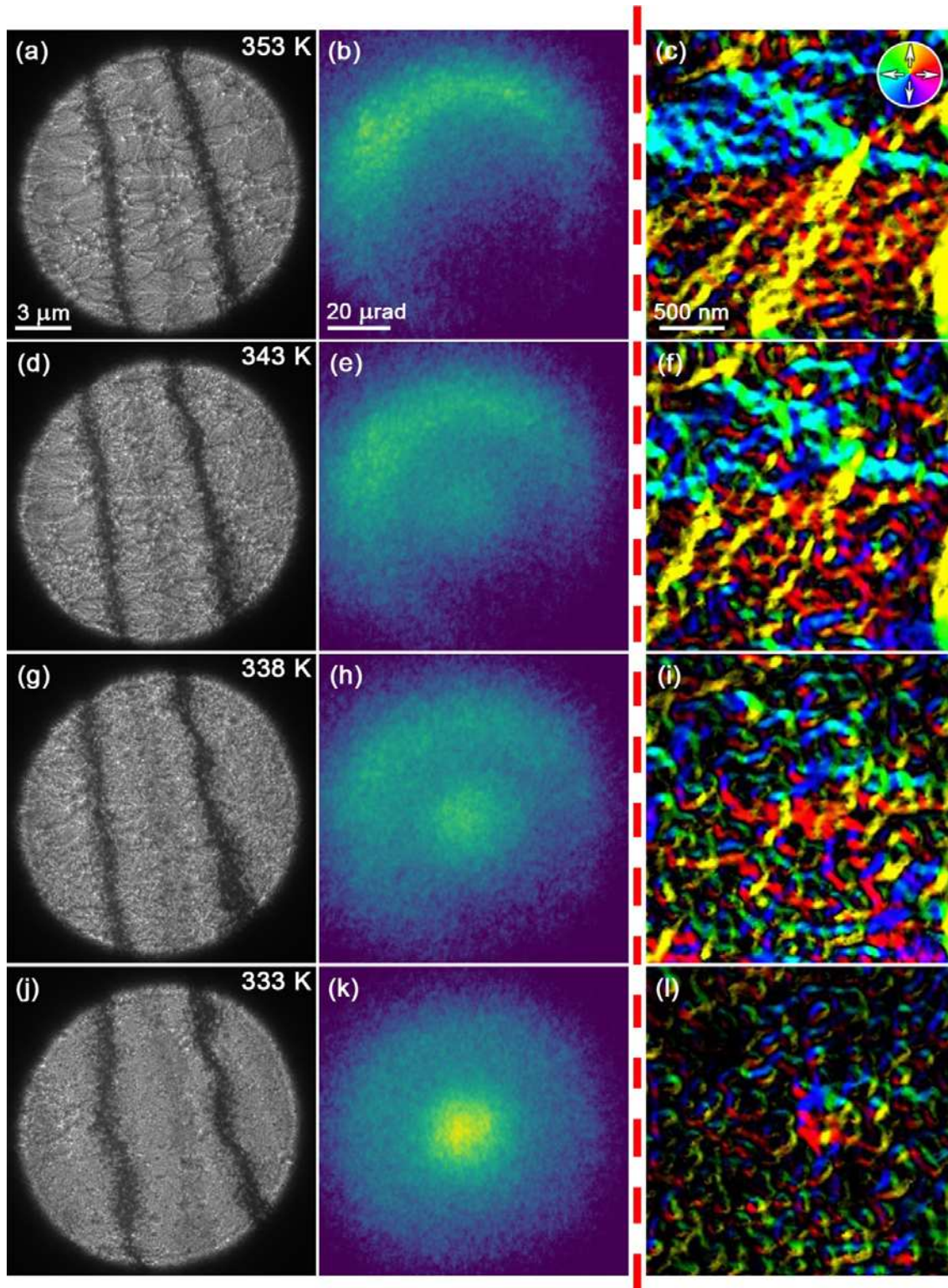
323 The outer ring corresponds to fully FM regions possessing the complete range of in-plane orientation
324 directions and its diameter is due to magnetic deflection of the electrons through a maximum angle β_l
325 of $\sim 35 \mu\text{rad}$. Fig. 7b displays the SAES pattern acquired at 413 K where the intensity has evolved,
326 becoming concentrated in an arc of restricted angular range on the outer ring (top-left side) possessing
327 the same deflection angle amplitude, β_l of $\sim 35 \mu\text{rad}$. This deflection angle, β_l , can be related to the
328 saturated magnetic induction for the FeRh thin film using the equation:

$$329 \quad B_s = \frac{\beta_l h}{e \lambda t} \quad (1)$$

330 where B_s is the saturation induction, t is the thickness of the magnetic thin film (measured from the
331 EELS low-loss thickness map), h is Planck's constant, e is the magnitude of electronic charge and λ is
332 the electron wavelength. Using equation 1, the saturation induction can be calculated from the SAES
333 pattern for the fully-FM state, assuming the film is uniformly magnetised throughout its thickness.
334 The β_l of $35 \mu\text{rad}$ in the SAES pattern of Fig. 7b corresponds to a saturation induction of $1.34 \text{ T} \pm$
335 0.15 T , where the uncertainty is attributed to the full width half maximum spread of intensity in the
336 arced outer ring of the SAES pattern. This value is in very good agreement with the bulk saturation
337 magnetisation measurements from this HF-etched FeRh thin film in its FM state of 1060 kA/m , or
338 1.33 T^{26} . Considering the AF to FM transition is of first order, without intermediate stages of reduced
339 B_s from the FM phase, it is assumed the β_l of $6.4 \mu\text{rad}$ in Fig. 7a corresponds to FM regions of
340 reduced thickness. Again, using equation 1 and the $B_s = 1.33 \text{ T}$, the β_l of $6.4 \mu\text{rad}$ is calculated to
341 correspond to FM regions with a thickness of $7.9 \pm 0.2 \text{ nm}$, which is commensurate with the $\sim 8 \text{ nm}$
342 wide FM region observed in Fig. 2e, and will also be explained in more detail in the Discussion
343 section.
344

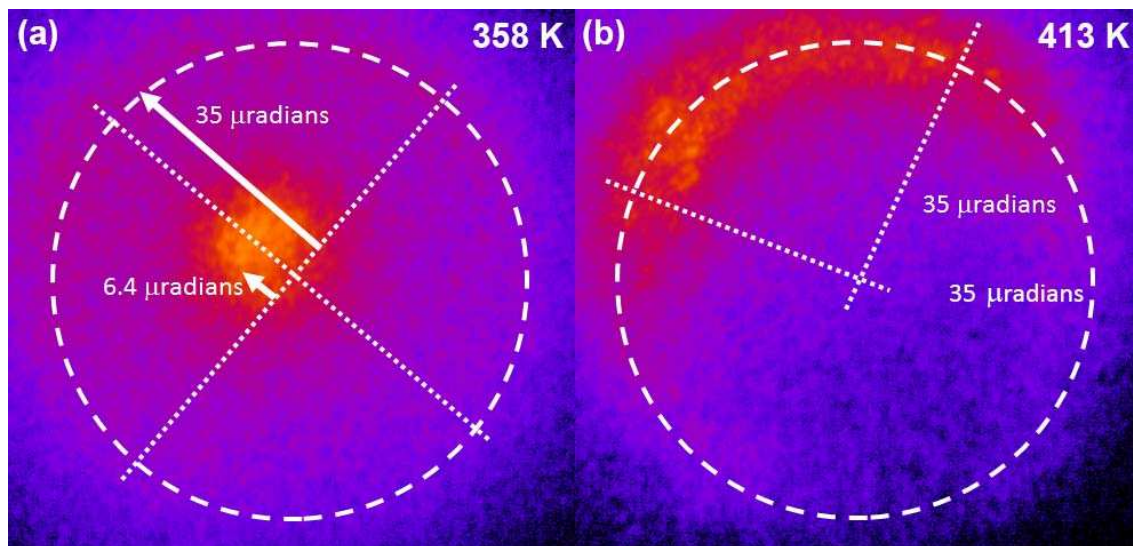
345 The 4D STEM DPC images of Figure 8 provides a localised examination of domain evolution
346 from the white-boxed region in Fig. 5c, as a function of temperature. Fig. 8a shows that the central
347 complex domain at 343 K comprises a vortex (arrowed, white) attached to a blue 'string' domain
348 flowing from right to left and separated from the rest of the domain by an elongated AF region, before
349 wrapping around on the left-hand side. As the temperature is increased to 348 K (Fig. 8b), the
350 elongated AF region disappears and the two vortices (arrowed) on the right-hand side become more
351 prominent. Further heating to 353 K promotes the merging of these two vortices (arrowed) into the
352 larger central domain (Fig. 8c). This domain is extended to the left at 358 K in Fig. 8d through growth
353 of a large vortex-like state (arrowed, white) and 'string' domain (arrowed, black). Increasing the
354 temperature to 360.5 K promotes separation of the oppositely magnetised top and bottom of the left-
355 hand side of this magnetic structure (blue and yellow, respectively) by a black DW (arrowed). At 363
356 K, the dark DW disintegrates into a cross-tie DW (arrowed), and the central domain is observed to
357 integrate with domain structures on all sides.

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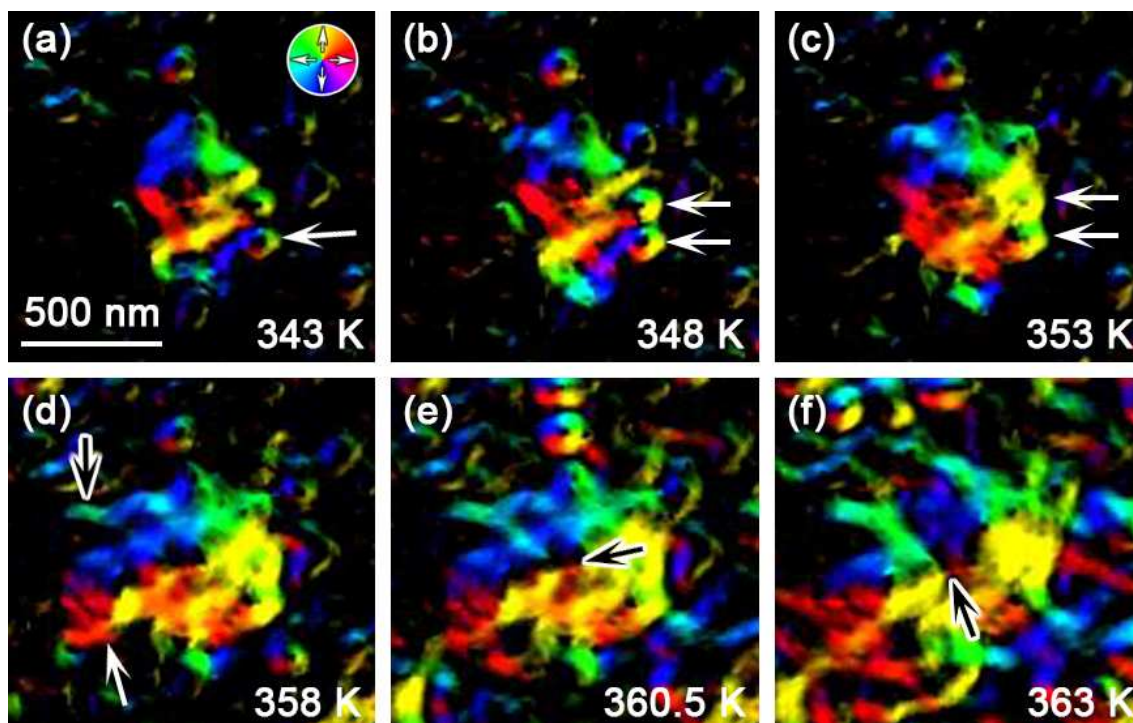
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Figure 6. Magnetic domain evolution in the planar FeRh thin film as a function of temperature during *in-situ* cooling from 353 K. (a,d,g,j) Fresnel; (b,e,h,k) SAES; and (c,f,i,l) 4D STEM DPC images of a HF-etched FeRh thin film acquired at (a-c) 353 K; (d-f) 343 K; (g-i) 338 K; and (j-l) 333 K. The images chart the reversal of the magnetostructural transition from (a-c) large domains of the FM state; (d-f) disintegration of FM domains; (g-i) AF / FM phase co-existence with small FM domains in the form of vortices and ‘strings’; and (j-l) small vortices. The direction of magnetization in the DPC images is depicted in the colour wheel (Fig. 6c, inset).



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Figure 7. Quantitative analysis of selected SAES patterns. (a) SAES pattern acquired at 358 K, showing the highest intensity corresponding to a 6.4 μrad deflection from the central optic axis (denoted by cross-hairs), along with a diffuse ring of intensity at a deflection of $\sim 35 \mu\text{rad}$. (b) SAES pattern acquired at 413 K showing the highest intensity at a deflection of $\sim 35 \mu\text{rad}$ towards the top left of the SAES pattern.



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Figure 8. Localised magnetic domain evolution during *in-situ* heating. (a-f) 4D STEM DPC images of the HF-etched FeRh thin film acquired at (a) 343 K; (b) 348 K; (c) 353 K; (d) 358 K; (e) 360.5 K and (f) 363 K. The images reveal the subtle localised growth dynamics of FM domains during the AF to FM phase transition. The direction of magnetization in the DPC images is depicted in the colour wheel (Fig. 8a, inset).

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383 **IV. DISCUSSION**

384 This multi-mode combined Lorentz microscopy investigation has provided fundamental, localised
385 insight into the magnetostructural transition in FeRh thin films as a function of temperature. The DPC
386 image of Fig. 2e revealed magnetic contrast in the vicinity of the interface with the MgO substrate at
387 303 K. The existence of such an interfacial moment has been previously detected indirectly and is
388 attributed to interfacial strain induced within the surface atomic layers of the FeRh, due to the slight
389 mismatch between the FeRh and MgO unit cells, and its thickness (6 – 8 nm) is consistent with that
390 reported³³. This is one area where the magnetic transition in FeRh thin films can vary from bulk FeRh,
391 as their epitaxial clamping on a substrate surface can mimic pressure effects on the FeRh
392 transformation by creating inhomogeneous strain in the regions nearest to the film-substrate
393 interface³³⁻³⁵. It is also suggested that interfacial oxidation can form a FeO-like layer that enhances the
394 spin polarisation of this small FM surface interfacial barrier³⁶. The DPC image series of Figure 3
395 recorded the AF to FM transition during heating from 362 K to 473 K in the cross-sectional FeRh
396 sample. The first small FM domain (~ 50 nm wide, ~ 20 nm high) nucleated at 362 K on the right-
397 hand side of the FeRh / MgO interface, which is consistent with previous reports of inhomogeneous
398 spatial distribution of the transition along the growth direction, where the transition initiates at the
399 substrate interface²⁵. This domain grows both laterally and upwards into the cross-section with
400 temperature until 391 K, at which point another domain rapidly nucleates and grows on the left-hand
401 side. These domains are separated by an asymmetric transverse DW³⁷, likely due to the shape
402 anisotropy of the cross-sectional FeRh sample, as they are not an energetically favourable
403 configuration for planar FM films. Nevertheless, these domains can be compared to the uniformly-
404 magnetised ‘string’ domains observed in the planar FeRh sample, as the thinner cross-sectional FeRh
405 TEM lamella would not accommodate the small vortex states (~ 130 nm). Hence, this study goes
406 beyond previous work on FeRh thin film cross-sections²⁵ by showing evolution of multiple domains
407 separated by DWs, without the need to magnetically saturate the sample, e.g. for electron holography,
408 which can provide a directional bias or memory, even in the AF state. Further, the domain evolution
409 and formation of DWs in the FeRh cross-section is consistent with that observed in the planar FeRh
410 film (Figs. 4&5).

411 In addition to the small FM signal at room temperature in the cross-sectional TEM sample
412 (Fig. 2e), the SAES patterns also reveal a small FM signal detected from the planar FeRh film. It is
413 considered that preferential HF-etching of the more reactive Fe atoms can result in a Fe-deficient and
414 Rh-terminated surface, where symmetry breaking stabilizes a surface FM layer at room temperature³⁸.
415 This FM layer is believed to be responsible for the dispersed central spot in the SAES pattern acquired
416 at 348 K (Fig. 5b) and provides a directional bias in terms of domain evolution. This is demonstrated
417 by the 6.4 μ rad offset from the optic axis (Fig. 7a) and arced outer ring favoured to the top left of Fig.
418 7b, as well as elongation of the magnetic domains in the Fresnel images. The 6.4 μ rad offset

419 corresponds to a ~ 8 nm thick surface FM layer, which is consistent with the $\sim 6 - 8$ nm interfacial
420 FM region in the cross-sectional TEM sample. Hence, this study provides both visual and quantitative
421 evidence of surface FM layers, as well as insight into initial stages of the domain evolution during the
422 AF to FM transition in the planar FeRh film, the details of which will now be discussed in detail.

423 The 4D STEM DPC images revealed that the thermally-induced nucleation of vortex
424 structures (~ 130 nm in diameter) act as initial or seed FM domain states in an AF matrix, where the
425 reduction of magnetostatic energy at the AF / FM boundary during the formation of the vortex state is
426 consistent with the edges of a FM nanostructure patterned in a non-magnetic film^{23,39}. This is also
427 consistent with the behaviour of a nanoscale magnetic cylinder, which will form a vortex if it has a
428 large enough diameter and is thick enough.⁴⁰ These vortex seeds clearly exhibit continuously flowing
429 magnetization and no DWs, providing an unambiguous interpretation compared to previous XMCD
430 results where a four-domain pattern separated by DWs and a vortex-state would appear identical⁸. The
431 localised expansion of the crystal lattice associated with the FM domain formation is likely to induce
432 strain in adjacent AF regions and make them more susceptible to transition to the FM state. This is
433 due to the film not being constrained to a substrate and hence freer to expand, as demonstrated by the
434 variation of bend contours observed in the Fresnel images. For this reason, isolated single domains are
435 rarely observed at this high spatial resolution due to the ease of their transition to the more
436 energetically favoured vortex state, as compared to substrate constrained FeRh films analysed by
437 surface magnetic imaging techniques like XMCD⁸ and SEMPA⁹. Hence, the initial stages progress
438 through nucleation of vortex states in the close vicinity of FM vortices or ‘string’ domains spreading
439 out from them (Fig. 5f & Fig. 8), where they are then seen to agglomerate and be consumed by the
440 nearby domains with increasing temperature. In the case of the directional growth of string domains,
441 crystal expansion is expected at the string-front prompting further directionally favoured growth,
442 whilst it is considered regions parallel to the string domain may undergo momentary compression
443 which hinders their transition and preserves dark AF regions between adjacent parallel string
444 domains, as observed at the bottom of the central domain in Fig. 8a. At this stage, the inherently
445 multi-directional nature of vortices and ‘string’ domains deflects the transmitted electron beam in all
446 directions and is in good agreement with the concentric ring of intensity in the SAES pattern of Fig.
447 5e. As more FM vortices nucleate, the AF regions decrease in size and FM domains expand through
448 agglomeration (Fig. 5g,i). This stage is considered to comprise small AF regions and all three main
449 types of FM domain: vortices; ‘strings’; and larger elongated domains which exhibit a preferred
450 directionality due to their biased interaction with the FM surface layer. Once the AF regions fully
451 transition to the FM state with increasing temperature, the remaining FM vortices and ‘strings’ lower
452 their energy configurations through coalescence with the larger domains. The fully-FM state will also
453 undergo long-range energy minimisation by merging of large grains through DW annihilation, as well
454 as the associated formation of cross-tie DWs, seen in Fig. 8f.

455 It is clear from both the Fresnel and 4D STEM DPC imaging that the FM domain
456 disintegration during the FM to AF transition behaves differently to the FM domain evolution
457 described above. As FM regions become AF, there is no net localised magnetic moment and hence no
458 need to minimise their energy into vortex structures like the FM states. However, in a similar fashion
459 to the FM domains, the AF regions act as nucleation sites for AF region growth which disrupts the
460 long-range ordering of the large FM domains. The high prevalence of the ‘string’ domains observed
461 in Fig. 6f,i is considered to be due to the retention of directionality during the short-range
462 disintegration of the larger FM domains. This is supported by the associated SAES patterns (Fig. 6e,h)
463 exhibiting fewer concentric outer rings, favouring the top-left side, compared to heating (Fig. 5e,h).
464 As the AF regions grow and ‘strings’ are the dominant FM domains, they are less likely to relax into
465 vortices since their domain width is already < 100 nm. Further, the prominent, complex domain in the
466 centre of Fig. 6l suggests that the first FM domains formed are also likely to be the last to disintegrate
467 during cooling, confirming that local structural defects or thickness variations act as both favourable
468 domain nucleation and final annihilation sites. This may prove beneficial for introducing controlled
469 nucleation sites through FIB techniques³⁹.

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471 V. CONCLUSIONS

472 This comprehensive TEM study has revealed directly the AF / FM phase transition in nanoscale FeRh
473 thin films as a function of temperature. We have provided both visual and quantitative evidence of
474 surface FM layers in the cross-sectional and planar FeRh samples, respectively, which was previously
475 inaccessible. Complementary segmented detector and 4D STEM DPC imaging displayed the intricate
476 details of the evolution and dissipation of the FM domains at an unambiguous level of detail. The
477 imaging was suitably complemented by SAES, providing quantitative measurements of integrated
478 induction matching the bulk magnetic measurements. As a significant step forward in understanding
479 the FeRh magnetic transition, this high-spatial-resolution magnetic imaging provides an explicit route
480 to analysis of the more localised and complex thermally-activated PB motion in gradient-doped FeRh
481 thin films, as well as current-induced spin injection⁴¹.

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602

603 **AUTHOR CONTRIBUTIONS STATEMENT**

604 T.P.A. and D.M designed and carried out the experiments; C.H.M., D.M., S.M. and T. M. conceived
605 the experiment; J.M. and R.T. synthesised and provided the samples; Y.L assisted with the
606 experimental work; T.P.A, D.M., and S. M. analysed the results. T.P.A. led the writing of the paper
607 and all authors reviewed the manuscript.

608

609 **DATA AND MATERIALS AVAILABILITY**

610 All data needed to evaluate the conclusions of this study can be found the at the following

611 <https://dx.doi.org/10.5525/gla.researchdata.987>.