In situ transmission electron microscopy and differential phase contrast imaging of the magnetostructural transition in nanoscale FeRh-based thin films

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Equi-atomic iron-rhodium ($Fe_{48}Rh_{52}$ to $Fe_{56}Rh_{44}$) has attracted considerable attention due to its first-order transition from its antiferromagnetic (AF) to ferromagnetic (FM) phase [1] and can hence present AF / FM phase co-existence. The co-existing phases are separated by a phase-boundary (PB) and effective control over the creation and motion of these PBs is considered desirable for potential application in a new generation of novel nanomagnetic or spintronic devices [2]. Previous studies have shown that domain walls (DWs) can be created and driven in FeRh films by combining heating with gradients of chemical doping [3-5]. However, our knowledge of the dynamic behaviour of DWs or PBs in FeRh is often limited to bulk magnetic measurements or low magnification imaging (resolution in the order of 10 s of nm), *i.e*. magnetic force microscopy, x-ray magnetic circular dichroism. Hence, in order to fully understand the magnetostructural transition and dynamic motion of both DWs and PBs in FeRh thin films, it is necessary to investigate their thermomagnetic behaviour on a localised scale through their entire thickness, whilst applying external stimuli, *i.e. in situ* heating.

The scanning transmission electron microscopy (STEM) technique of differential phase contrast (DPC) imaging permits nanometre-scale imaging of magnetisation within nanostructured thin films as a function of applied electric and magnetic fields, as well as temperature. This is the only technique that is presently able to provide both high-resolution images of DW and PB motion in thin magnetic films *in situ* within the TEM and quantitative measurements of saturation induction from the DWs, directly.

In this study, we perform several (S)TEM techniques to examine the localised chemical, structural and magnetic properties of FeRh films grown epitaxially grown on MgO substrates or NiAl buffers on GaAs substrates [6]. Fig. 1a presents a cross-section of the FeRh thin film with a thickness of ~ 50 nm, whilst the energy dispersive X-ray spectroscopy (EDS) chemical maps (Fig. 1b) acquired from the boxed region (red) in Fig. 1a display its elemental distribution. The DPC image of Fig. 1c shows well-resolved DWs between FM regions (arrowed) at 120°C and measurements of electron-beam deflection from the DW (boxed region, red) as a function of temperature allowed for calculation of integrated magnetic induction, and subsequent charting of the magnetostructural transition (Fig. 1d). The growth and co-existence of phases in a HF-etched planar FeRh film are observed directly during *in situ* heating using Fresnel imaging, small-angle electron scattering (SAES) and pixelated DPC imaging, as presented in Fig. 2. At 75°C, small magnetic domains nucleate in the form of vortices (Fig. 2a&c, arrowed) and provide minimal deflection in the SAES pattern (Fig. 2b). As the FeRh is heated further, the vortices coalesce into larger domains at 90°C (Fig. 2d&f) and induce formation of the large ring in the SAES pattern (Fig. 2e), representing a state of the AF / FM phase co-existence separated by PBs, until the FeRh is fully FM at 100°C (Fig. 2g-i). Hence, this localised analysis provides fundamental insight into the mechanistic details of the AF / FM magnetostructural phase transition.

References

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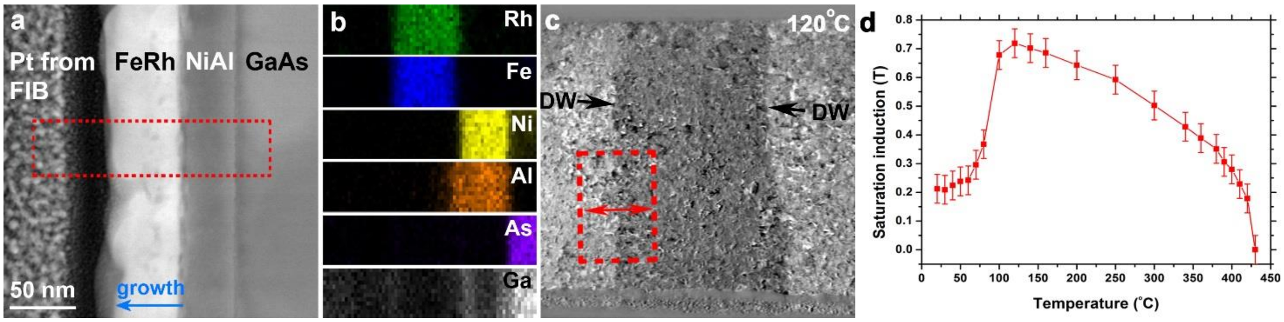

Fig. 1 (a) STEM image of an FeRh thin film epitaxially grown on a NiAl buffer layer and GaAs substrate. (b) EDS spectrum image acquired from the boxed region in (a), and chemical maps showing their elemental content. (c) DPC image of magnetic domains/DWs in a planar FeRh thin film acquired at 120°C. (d) Graph of saturation induction as a function of temperature, calculated using direct measurements of electron-beam deflection from the DW in the boxed region (red) defined in (c).

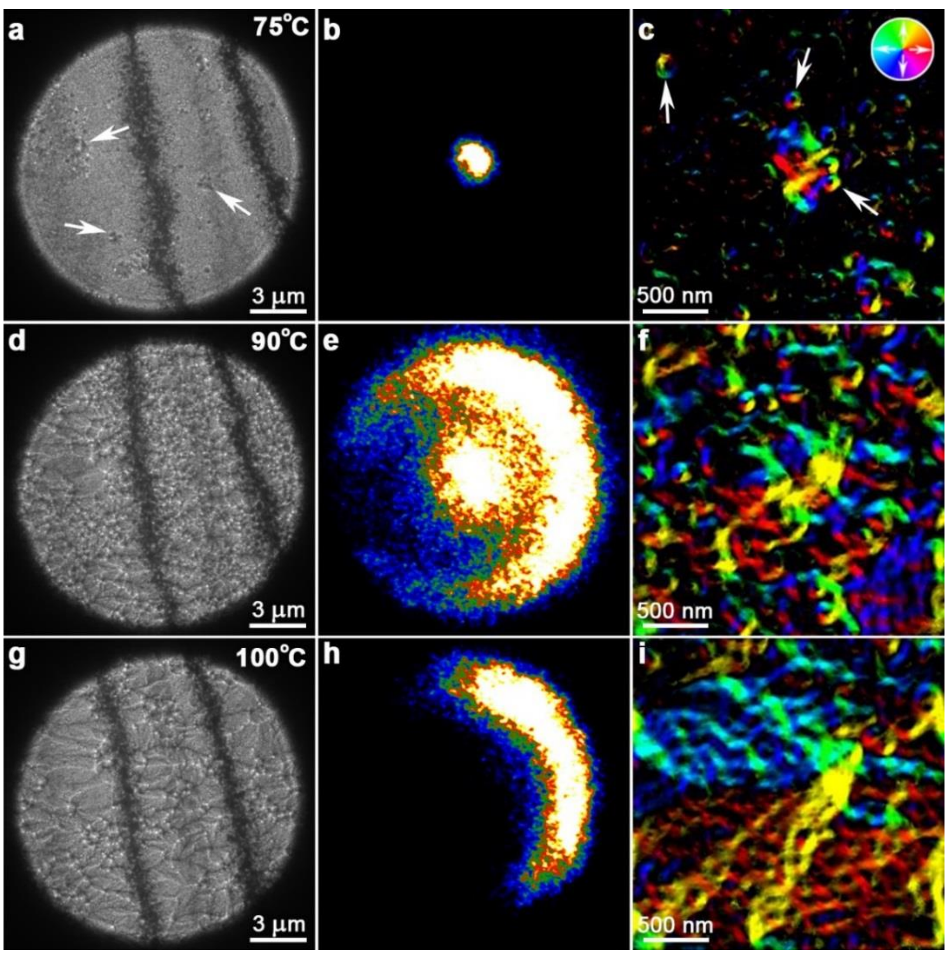

Fig. 2 (a,d,g) Fresnel; (b,e,h) SAES; and (c,f,i) pixelated DPC images of an HF-etched FeRh thin film acquired during *in situ* heating to (a-c) 75°C; (d-f) 90°C; and (g-i) 100°C. The images demonstrate the different stages of the magnetostructural transition, including (a-c) nucleation of magnetic vortices (arrowed); (d-f) AF / FM phase co-existence; and (g-i) a fully FM state. The direction of magnetization in the DPC images is depicted in the colour wheel (Fig. 2c, inset).