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Differential Phase Contrast Imaging of the Magnetostructural Transition and Phase Boundary Motion in Uniform and Gradient-doped FeRh-based Thin Films

Trevor P. Almeida[1]*, Laura Clark[1], Rowan Temple[2], Jamie Massey[2], Thomas Moore[2], Damien McGrouther[1], Christopher H. Marrows[2] and Stephen McVitie[1]

[1.] SUPA, School of Physics and Astronomy, University of Glasgow, UK.
[2.] School of Physics and Astronomy, University of Leeds, UK.
* Corresponding author: trevor.almeida@glasgow.ac.uk

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) domain walls (DWs) 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 the PBs can be created and driven in FeRh films by combining heating with differential gradients of chemical doping, where dopants of iridium and palladium have been shown to decrease and increase the transition temperature, respectively[3]. However, our knowledge of the dynamic behavior of PBs in FeRh is often limited to to bulk magnetic measurements or low magnification imaging (resolution in the order of 10 s of nm), *i.e.* magnetic force microscopy, Kerr microscopy, x-ray magnetic circular dichroism. In order to fully understand the magnetostructural transition and dynamic motion of PBs, it is necessary to examine the effect of temperature directly. The scanning transmission electron microscopy (STEM) technique of differential phase contrast (DPC) imaging permits nanometer-scale imaging of magnetization within nanostructured thin films as a function of temperature. Here, the first use of DPC to examine the magnetic domain evolution and heat-induced PB motion in uniform and gradient-doped cross-sectional FeRh thin films is presented.

Uniform and Ir / Pd gradient-doped thin films of ordered α'-FeRh alloy were grown epitaxially on clean (001) MgO substrates by conventional DC magnetron sputter co-deposition, as described previously[4]. Cross-sectional TEM samples were prepared from bulk substrates and transferred onto *in situ* heating (DENSsolution WildfireTM) TEM electronic (e-) chips by focused ion beam methods[5]. Conventional and high-resolution STEM imaging of the FeRh films and substrates was performed using a JEOL ARM cFEG instrument at 200kV, whilst energy dispersive X-ray (EDX) spectroscopy provided chemical analysis. In order to recover the ordered α' FeRh structure after FIB preparation, the cross-sectional samples were annealed *in situ* in the TEM at 650°C for 1 hour using the DENSsolution e-chips. The magnetic structure of the films is visualized using DPC imaging under low-magnetic field conditions, and *in situ* heating up to 200°C provides direct access to the dynamics of the FM domain nucleation / evolution within the uniform FeRh films. In addition, PB motion in the gradient-doped FeRh cross-sections is systematically induced as a function of temperature.

Figure 1 presents a cross-section of a uniform FeRh thin film, providing information on its thickness, localized structure and interface with the MgO substrate[6], as well as its magnetization. The dark-field (DF) STEM image of Fig. 1a reveals the FeRh film to be grown with a uniform thickness of ~ 52 nm, whilst the high-resolution STEM image of Fig. 1b presents the interface between the single crystalline FeRh and MgO substrate, revealing their well-matched orientation and confirming the epitaxial growth of the deposited FeRh. The DPC image of Fig. 1c reveals the growth of a green magnetic domain (~ 200 nm wide, right-hand side) at the FeRh / MgO interface at 93°C. As the temperature is increased to 200°C, a large magnetic domain (red) with opposite direction of magnetism is observed to form on the left hand-side of the FeRh thin film (Fig. 1d), where a small domain (blue, arrowed) considered to be a transverse DW is seen to separate the larger domains (green and red). Hence, this localized analysis provides fundamental insight into domain evolution during the AF to FM transition.

Figure 2 presents a cross-section of the Ir / Pd gradient-doped FeRh thin film, providing information on its chemical distribution and magnetism as a function of temperature. The EDX chemical maps (Fig. 2a) display the elemental distribution of palladium and iridium dopants within the FeRh thin film. The palladium dopant content is uniformly distributed along the cross-section, whilst the iridium dopant exhibits a gradient from high (top) to low (bottom) content. The DPC images of Fig. 2b presents the AF / FM PB motion as a function of increasing temperature. At 50°C, no magnetic signal is evident in the top Ir-rich side of FeRh thin film and hence considered AF, distinct from the FM bottom side (green), suggesting they are separated by an AF/FM PB. As temperature is increased, the green FM domain migrates upwards, inducing upward PB motion, until the FeRh film is fully FM at 200°C. Fig. 2c displays the AF / FM PB motion as a function of decreasing temperature from 150°C, where it is in a fully-FM state. As the FeRh thin film is cooled to 20°C, the very top layer of the FeRh becomes AF, and the PB progressively migrates downwards towards the center. Hence, this study shows that PB motion can be controlled systematically using temperature and Ir-gradient doping.

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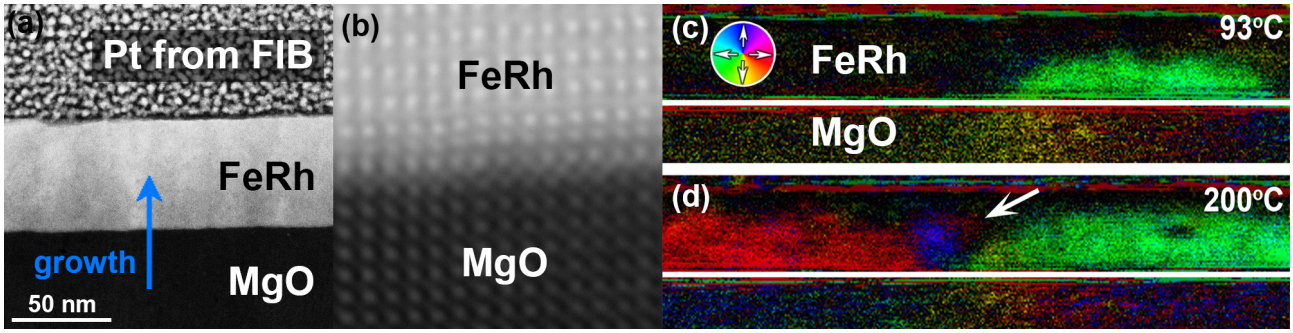

Figure 1. (a) DF STEM image of a FeRh thin film grown epitaxially on the MgO substrate. (b) High resolution STEM image showing the epitaxially grown FeRh and its interface with the MgO substrate. (c,d) DPC imaging of a uniform FeRh film cross section showing: (c) growth of a FM domain (green) at the FeRh / MgO substrate at 93ºC; and (d) green and red FM domains at 200 °C separated by a transverse DW (blue, arrowed). The direction of magnetization is depicted in the color wheel (inset).

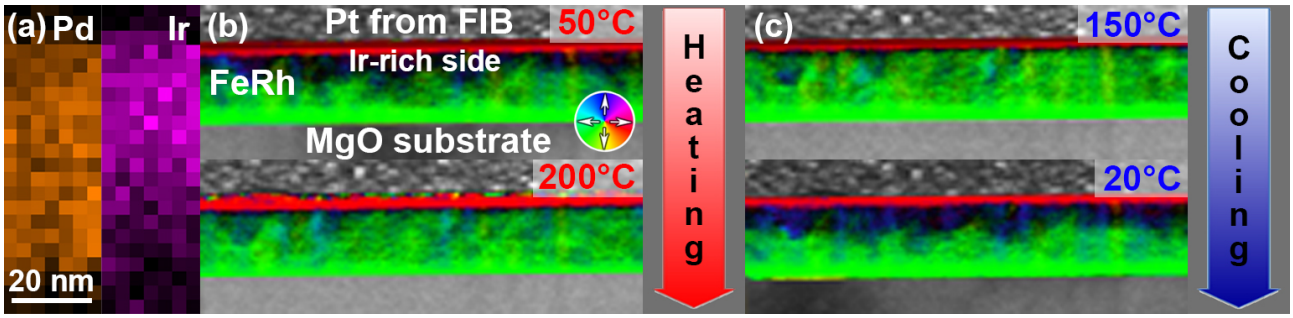

Figure 2. (a) EDX chemical maps displaying the elemental distribution of palladium (orange) and iridium (pink) content, revealing the high (top) to low (bottom) gradient in iridium. (b) DPC imaging of the Ir/Pd gradient-doped FeRh film as a function of increasing temperature, displaying the upward PB motion to the Ir-rich side. The direction of magnetization is depicted in the colour wheel (inset). (c) DPC imaging as a function of decreasing temperature, displaying the downward PB motion.

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