Visualizing Dynamic Magnetism in Nanostructures using Electron Microscopy

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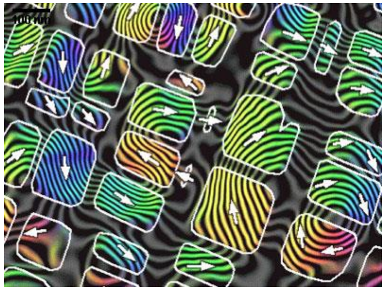
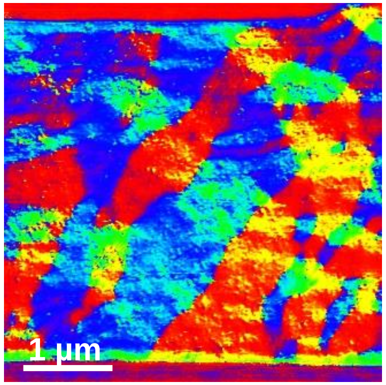
Outline

- **Motivation**
- **Transmission Electron Microscopy**
- **Magnetic Minerals**
  - Electron holography
- **Phase transition in FeRh thin films**
  - Differential phase contrast imaging (DPC)





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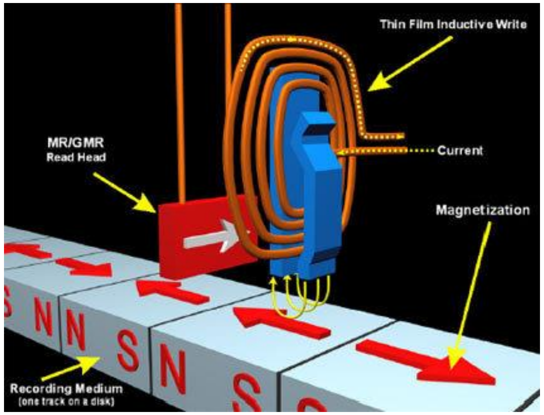
Motivation

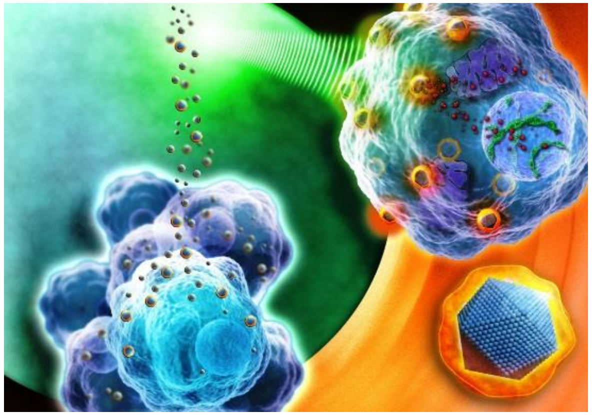
The demand for improved functionality and reduction in size of a range modern devices has led to the rapid development of new magnetic materials

→ driving the need to visualise localised magnetism on the nanoscale

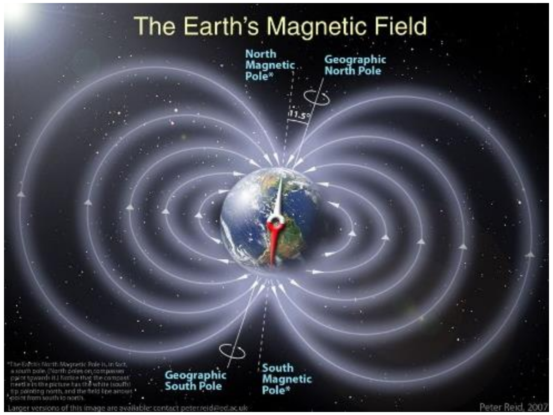
Magnetic data storage



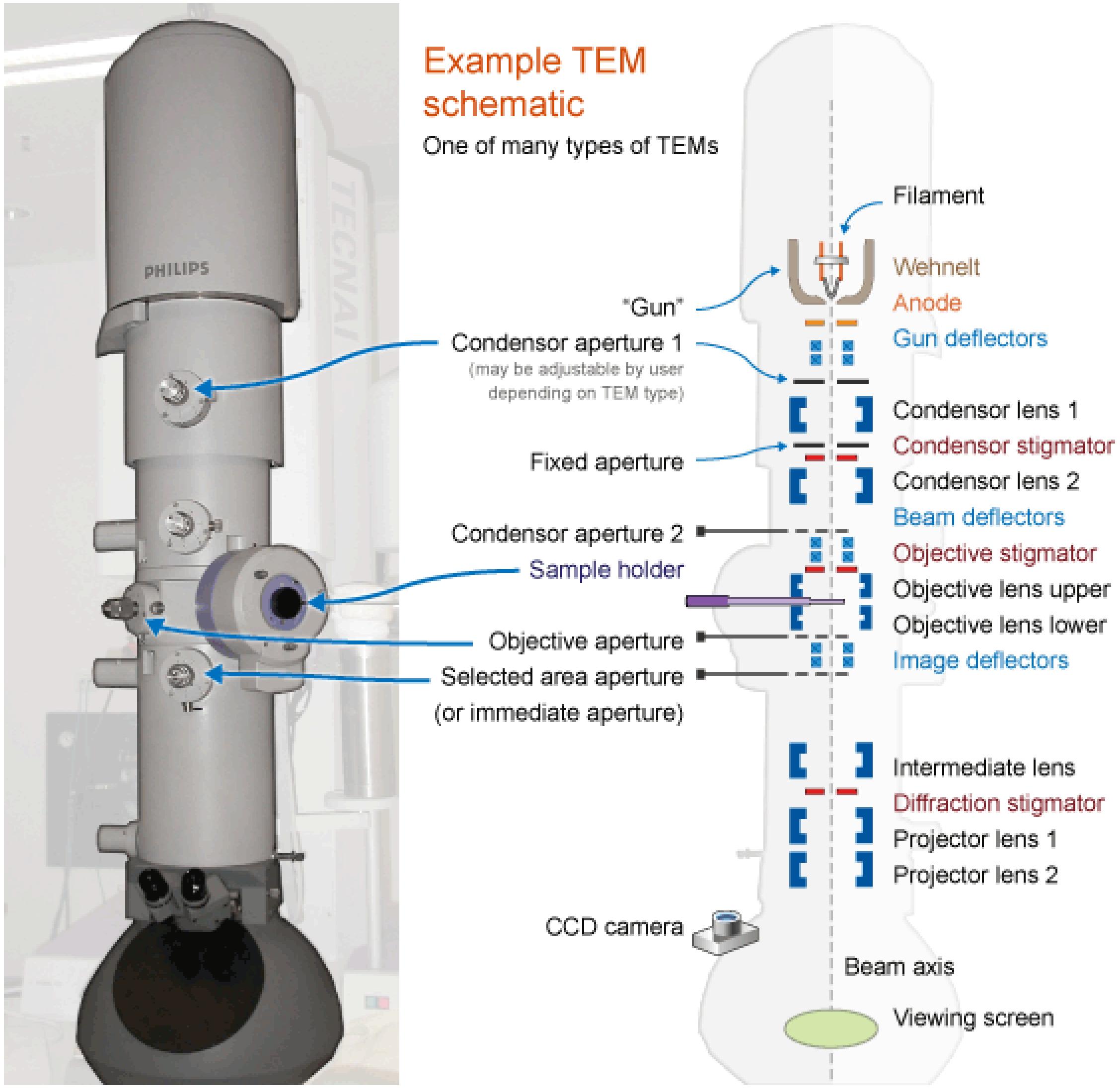
Nanomedicine



Earth science



Transmission electron microscopy



Sample thickness
< 200 nm

Resolution
~ 80 pm

Information:

- **Morphology**
- **Chemistry**
- **Structure**
- **Magnetism**
- **Electronic**
- **Etc.**

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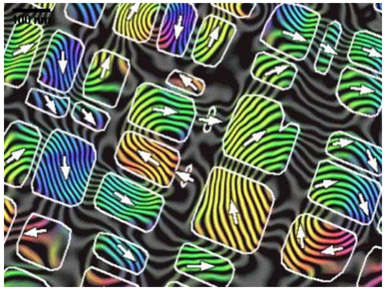
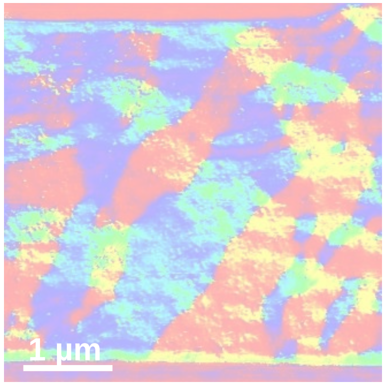
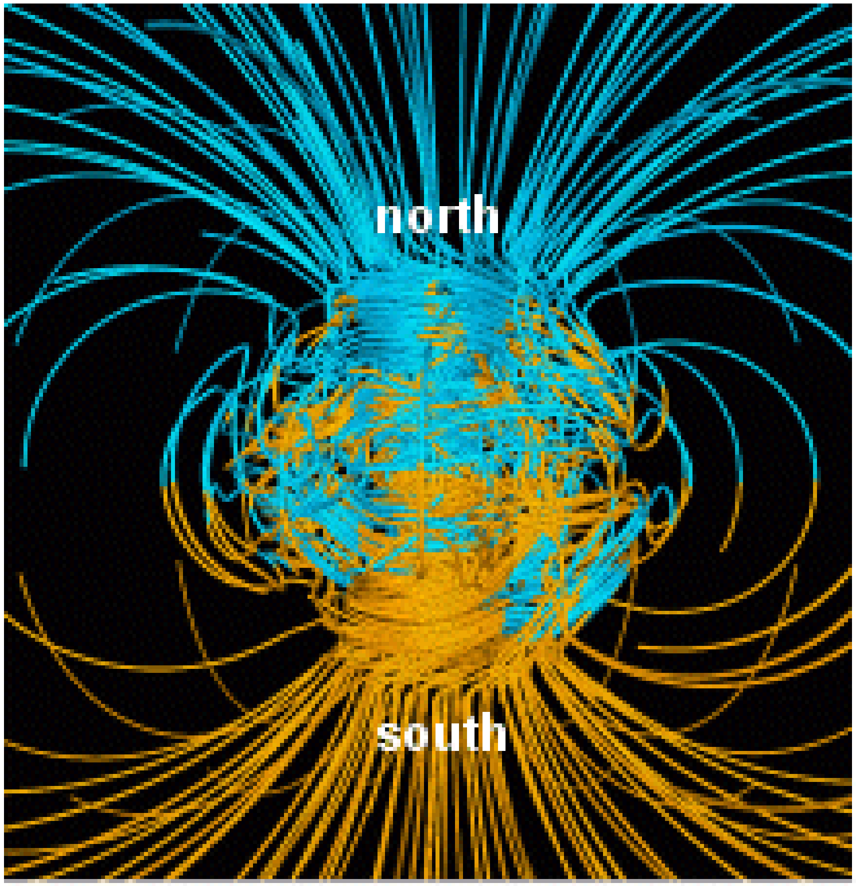
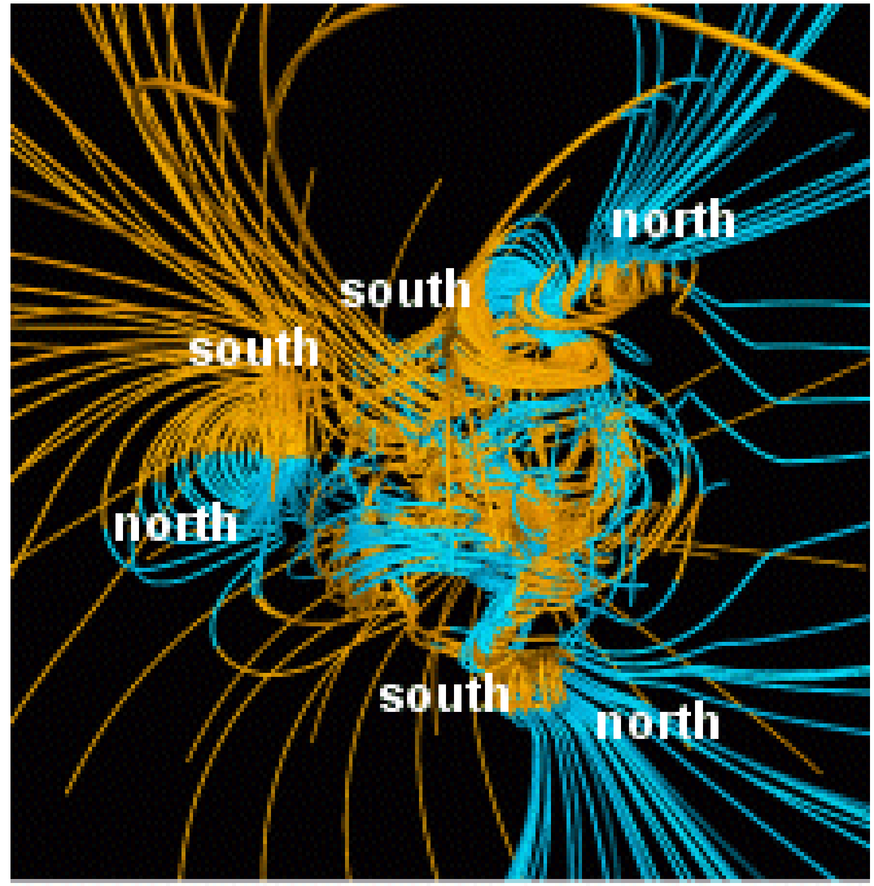




Magnetism in Earth science



between reversals



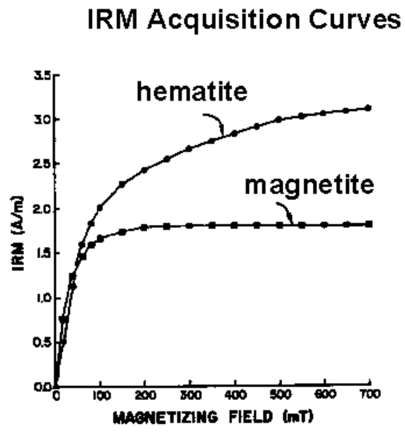
during a reversal

Magnetism in Earth science

To interpret palaeomagnetic data we need to understand mechanisms that:

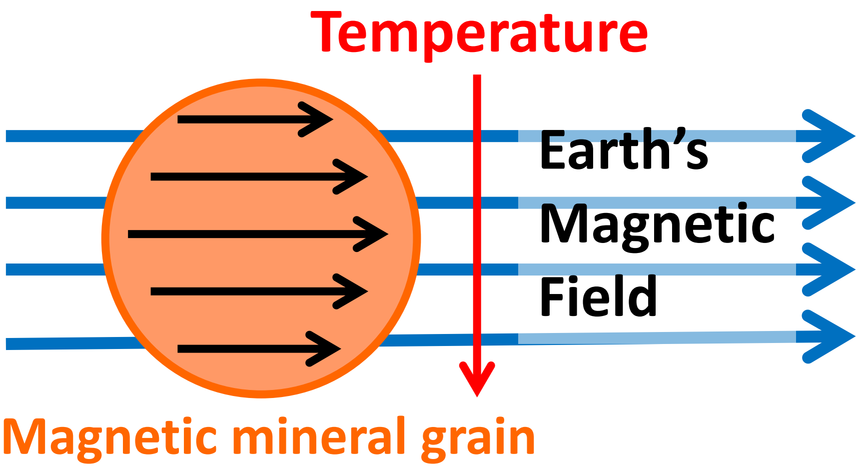
1. **induce magnetic remanence**
2. **alter magnetic remanence**

1. Chemical alteration – change in oxidation state



after Bulter (1982)

2. Thermomagnetic behaviour



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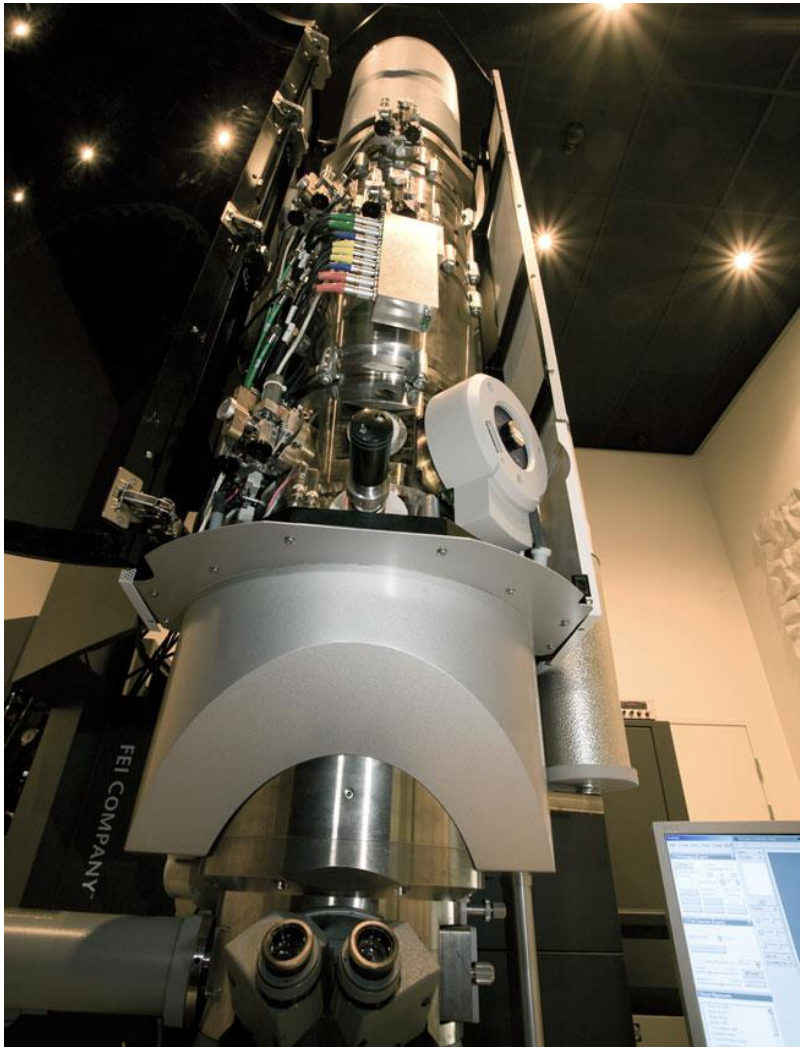
Electron holography

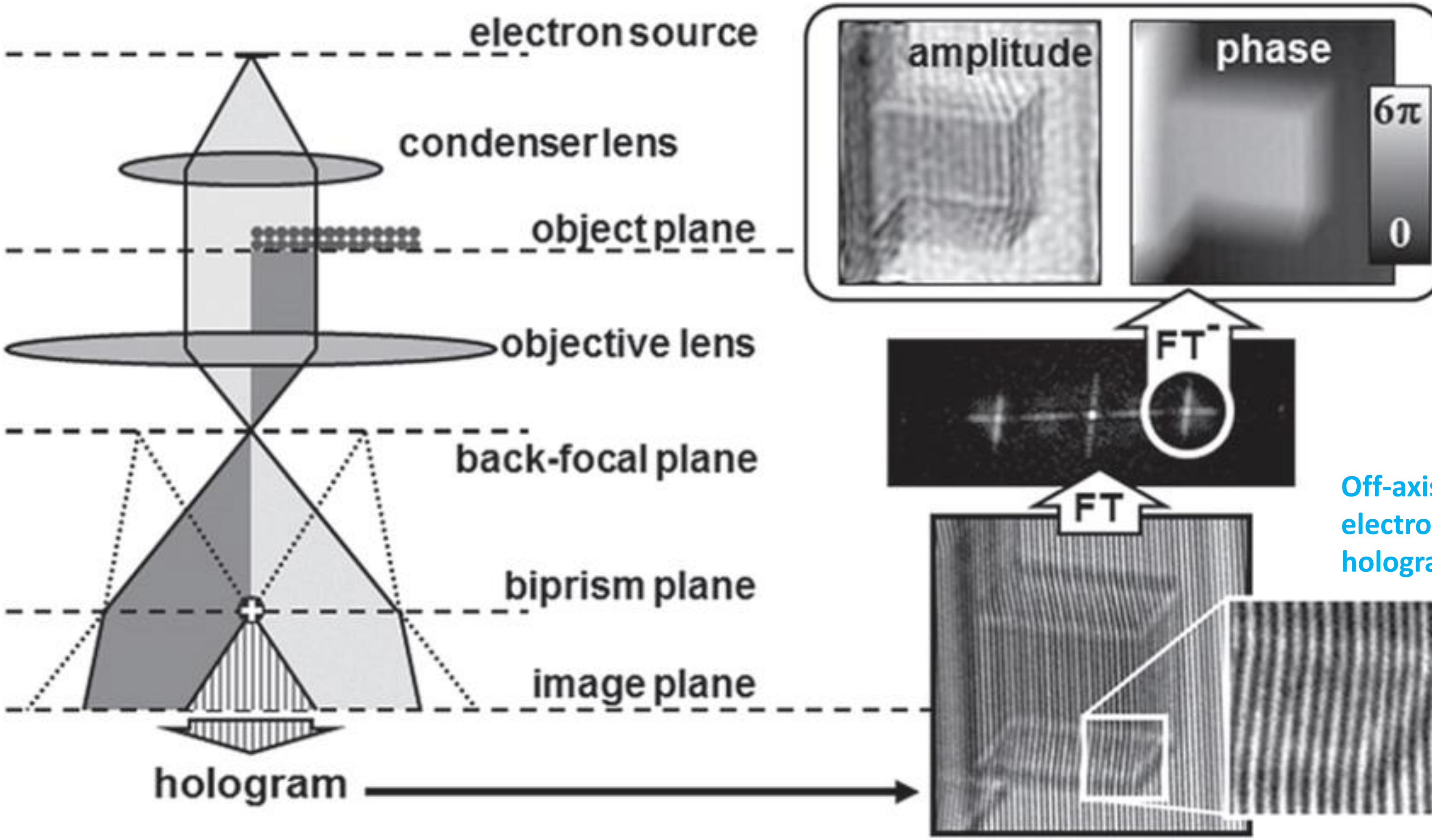


- **FEI Titan Analytical**
- **C_S correction on condenser lens, *i.e.* probe corrected**
- **Operated at 300kV**
- **HR-STEM and chemical mapping at atomic scale**
- **Biprism and Lorentz lens for electron holography of magnetic fields**

DTU

DTU Cen
Center for Electron Nanoscopy

Electron holography



Off-axis electron hologram

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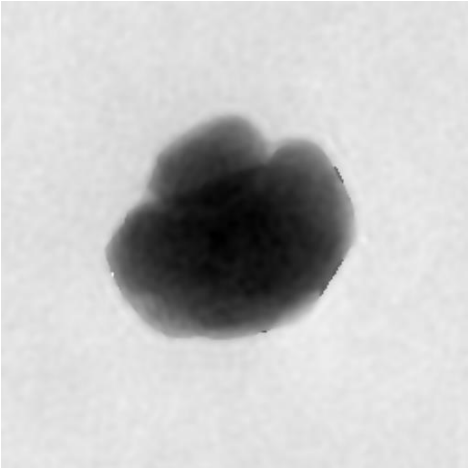
Electron holography

Phase shift: $$\varphi(X) = C_E \int V(x,z)\,dz - \left(\frac{e}{h}\right) \iint B_{\perp}(x,z)\,dx\,dz$$

Mean inner potential — **Magnetic induction**

Total phase shift



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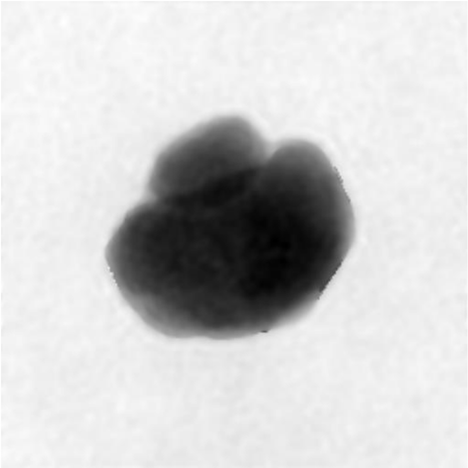
Mean inner potential



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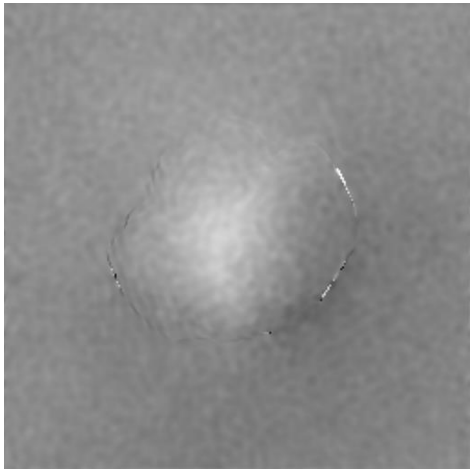
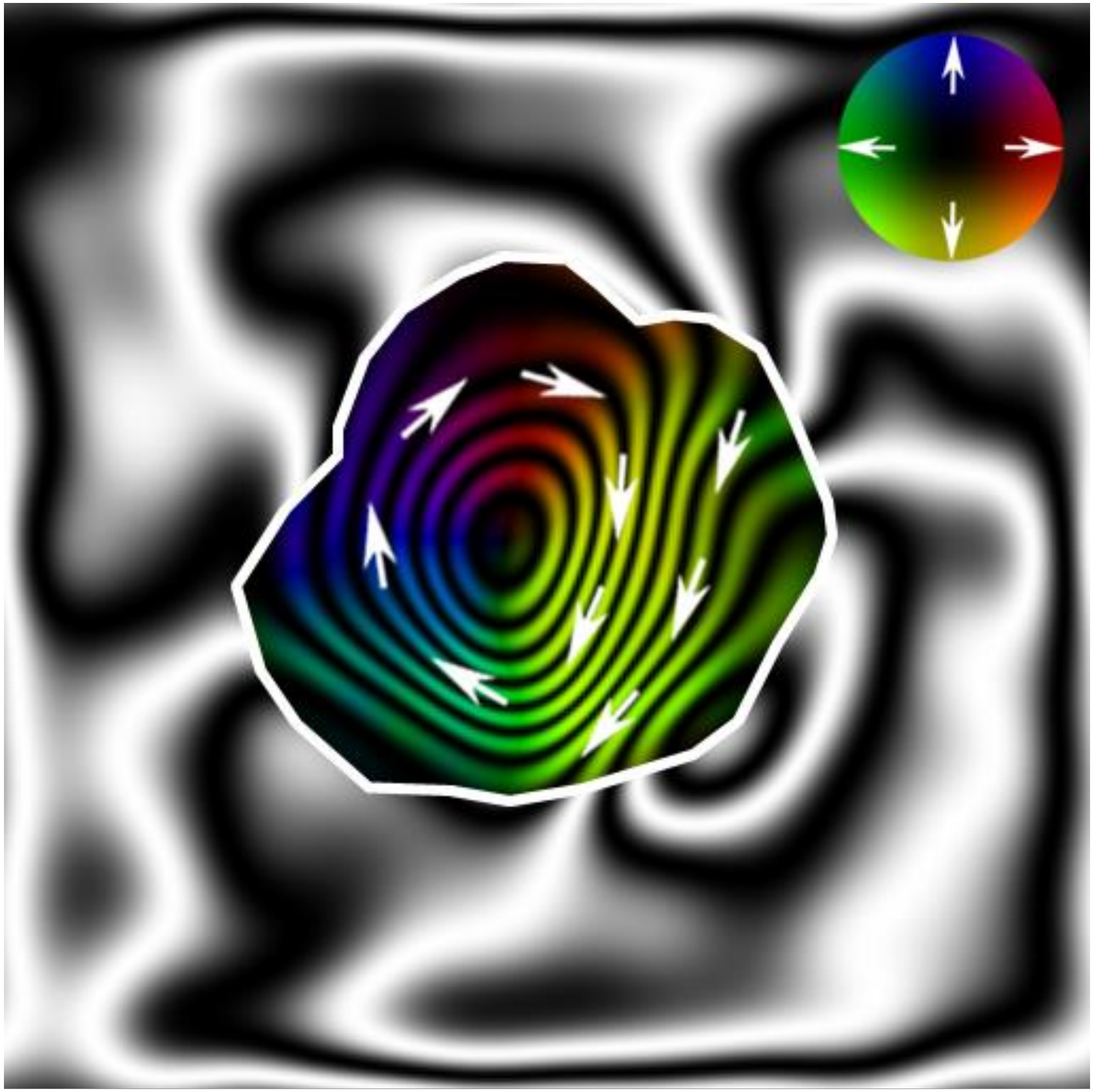
Magnetic contribution



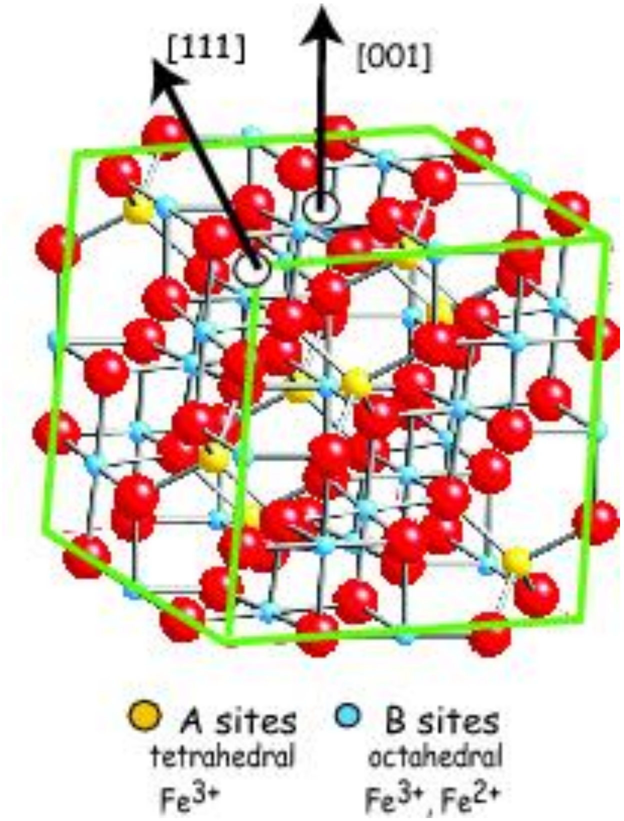
Electron holography

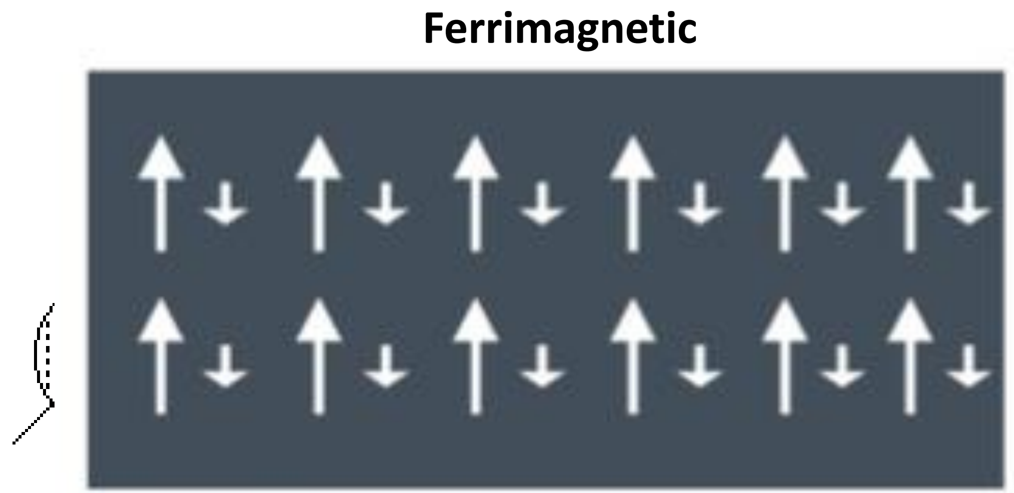
Vortex state and stray magnetic field – PSD particle



Oxidation of Fe_3O_4:

$$2Fe_3O_4 + \frac{1}{2}O_2$$





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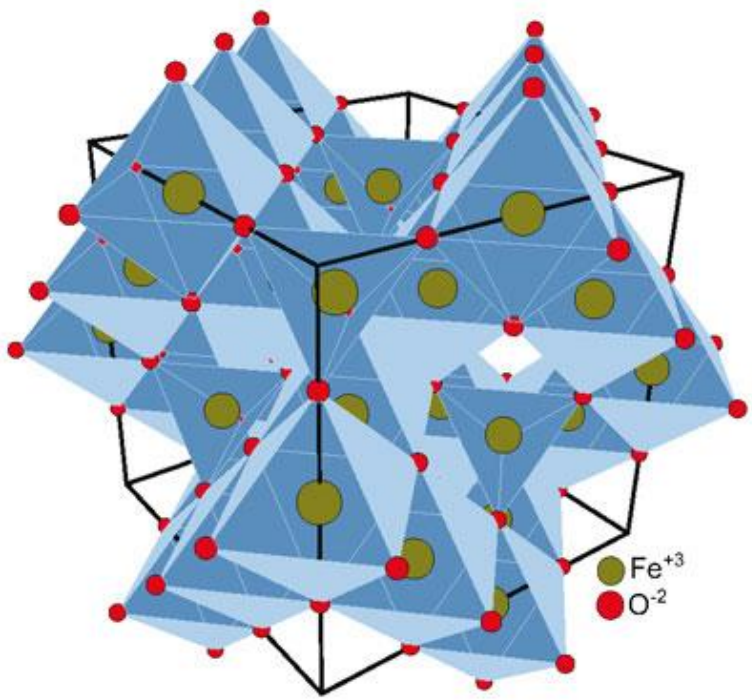
Oxidation of magnetic minerals

Oxidation of Fe_3O_4:

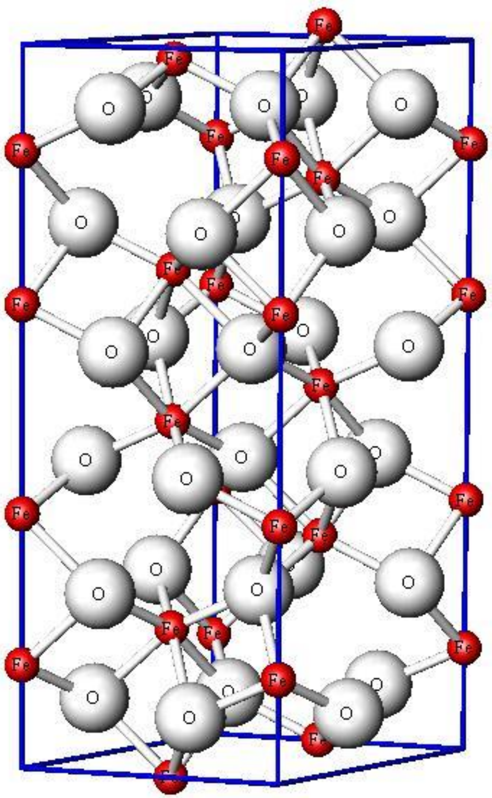
$$2Fe_3O_4 + \frac{1}{2}O_2 \rightarrow \text{ϒ-}3Fe_2O_3$$



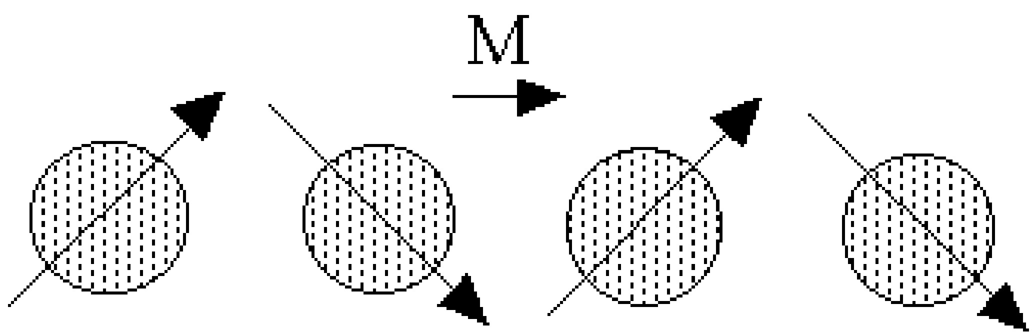
Slightly weaker ferrimagnetic

Oxidation of Fe_3O_4:

$$2Fe_3O_4 + \frac{1}{2}O_2 \rightarrow \Upsilon\text{-}3Fe_2O_3 \rightarrow \alpha\text{-}3Fe_2O_3$$



Canted antiferromagnetic



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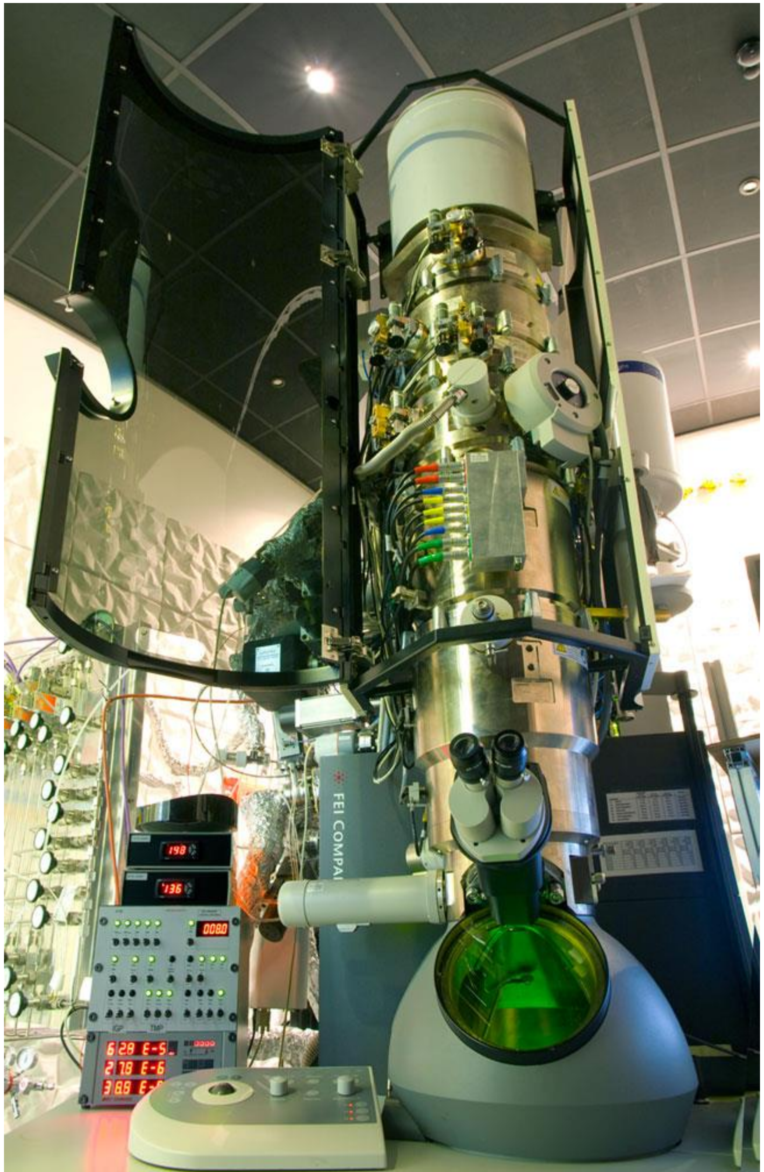
Environmental TEM



- **FEI Titan E-Cell**
- **C_S correction on objective lens, *i.e.* image corrected**
- **Operated at 300kV**
- **Various gases, *i.e.* H_2, He and $H_2$0 up to 1000 Pa**
- **Heating specimen holder**

  < 1000°C

DTU

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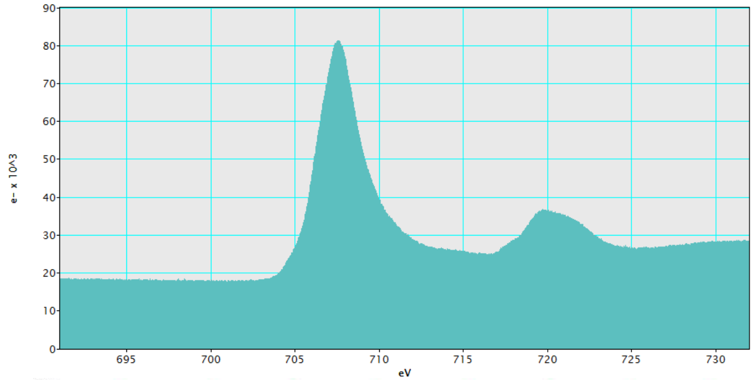
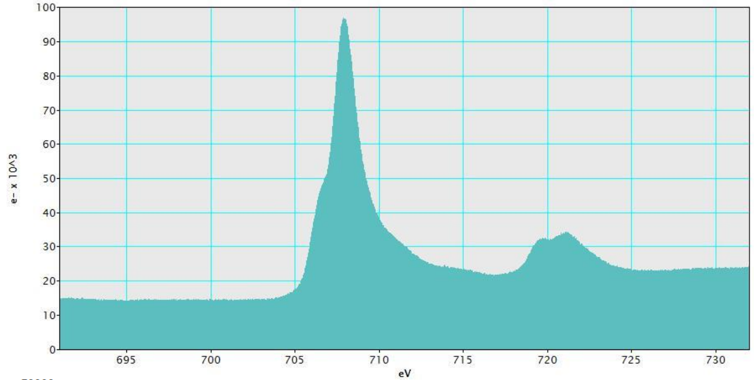
Center for Electron Nanoscopy

Environmental TEM

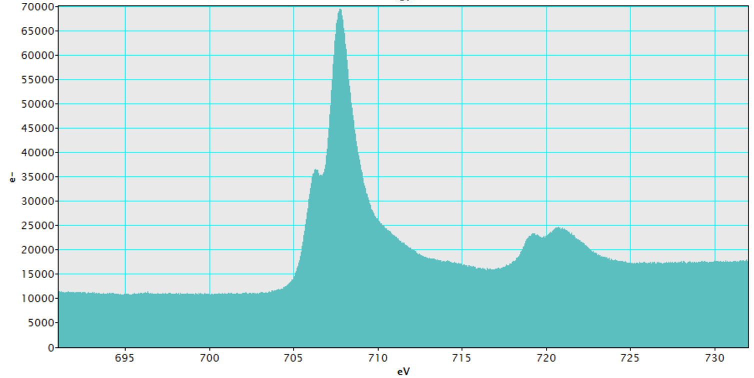
Energy dispersion of 0.02eV & resolution of 0.5eV



EEL spectrum from Fe_3O_4 particles



EEL spectrum from ϒ-Fe_2O_3 sample



EEL spectrum from reference α-Fe_2O_3 sample

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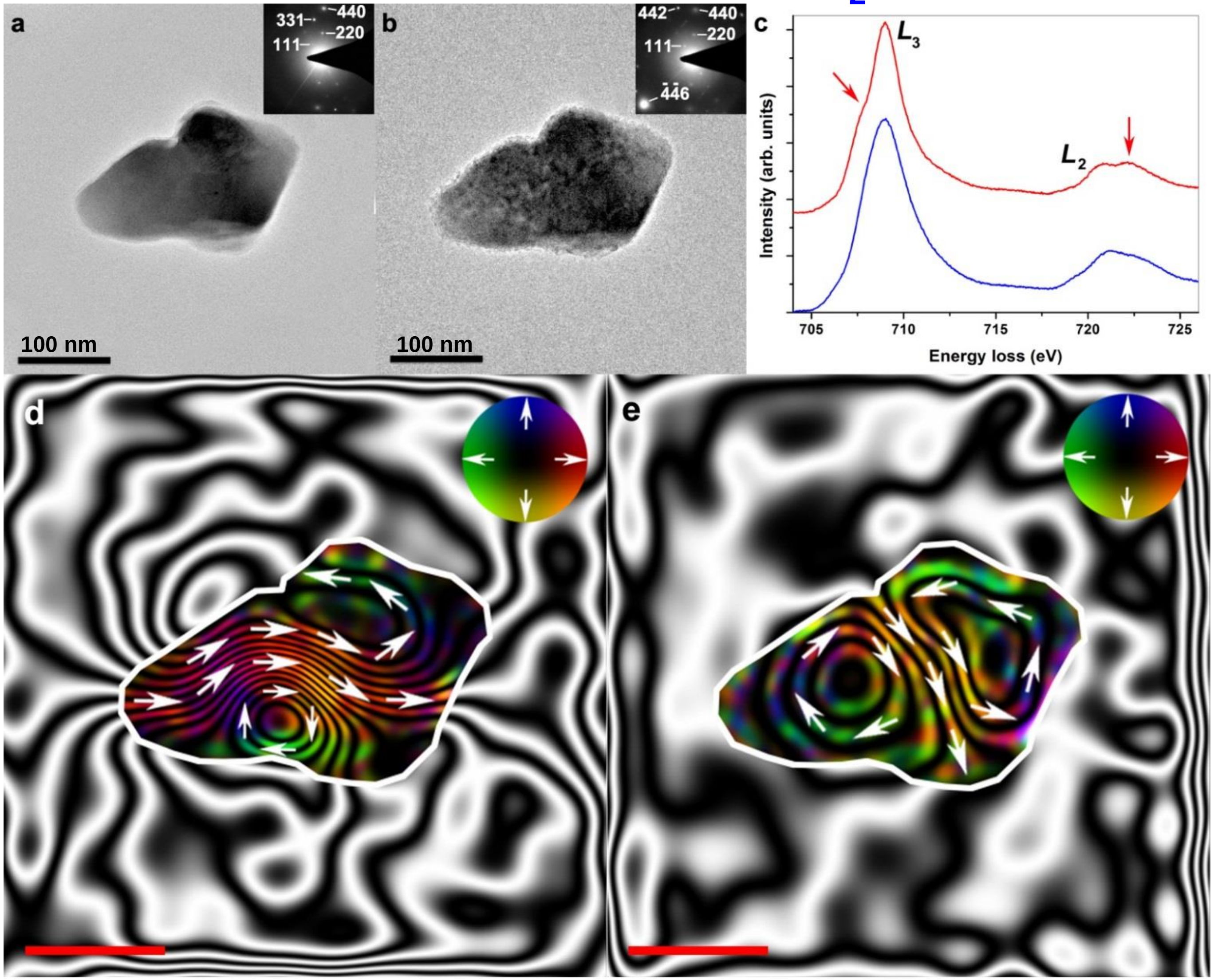
Environmental TEM

Heated at 700°C, under 9mbar O_2 for 8 hours



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Almeida, T. P *et al.*, *Nature Communications*, 2014

Environmental TEM

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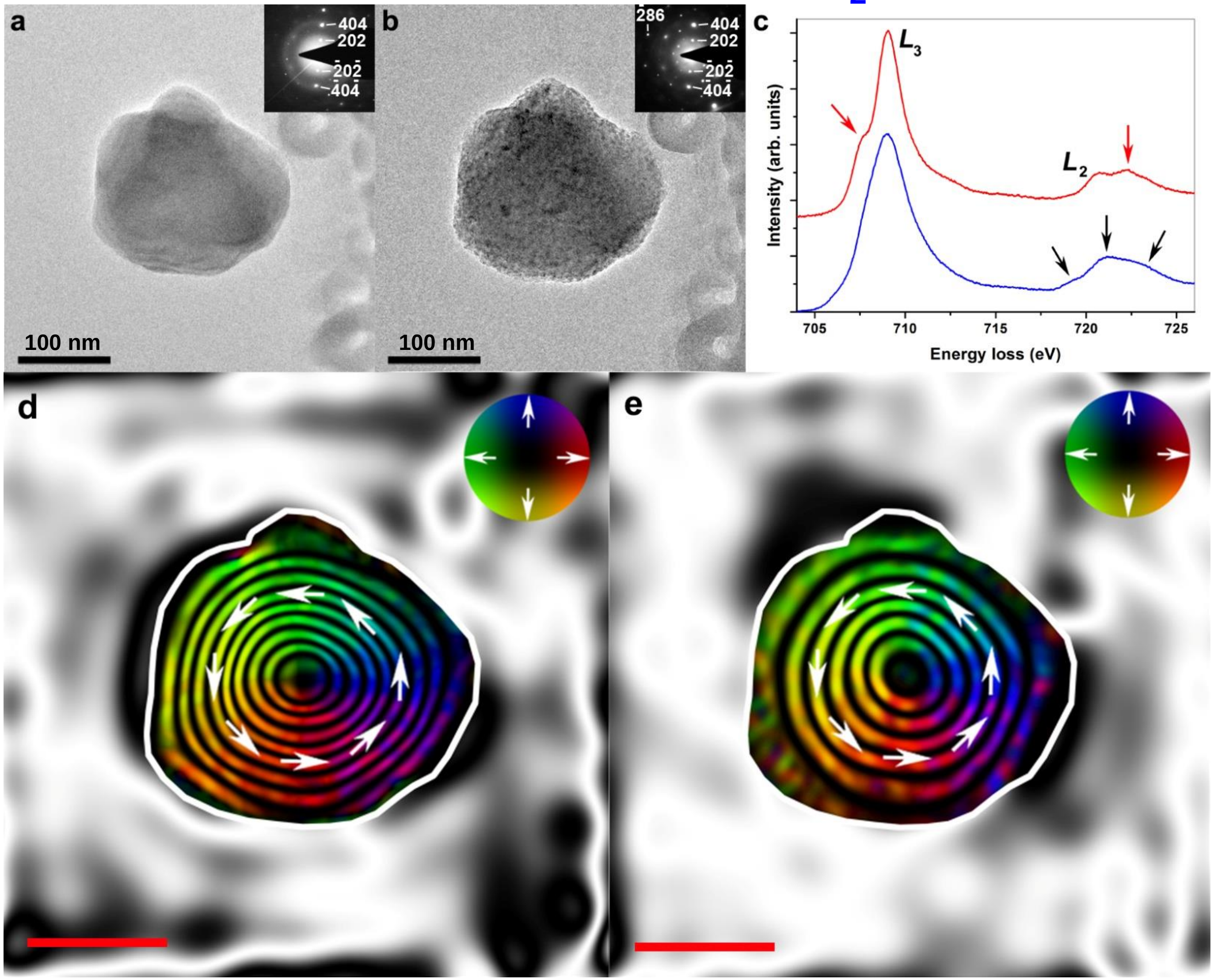
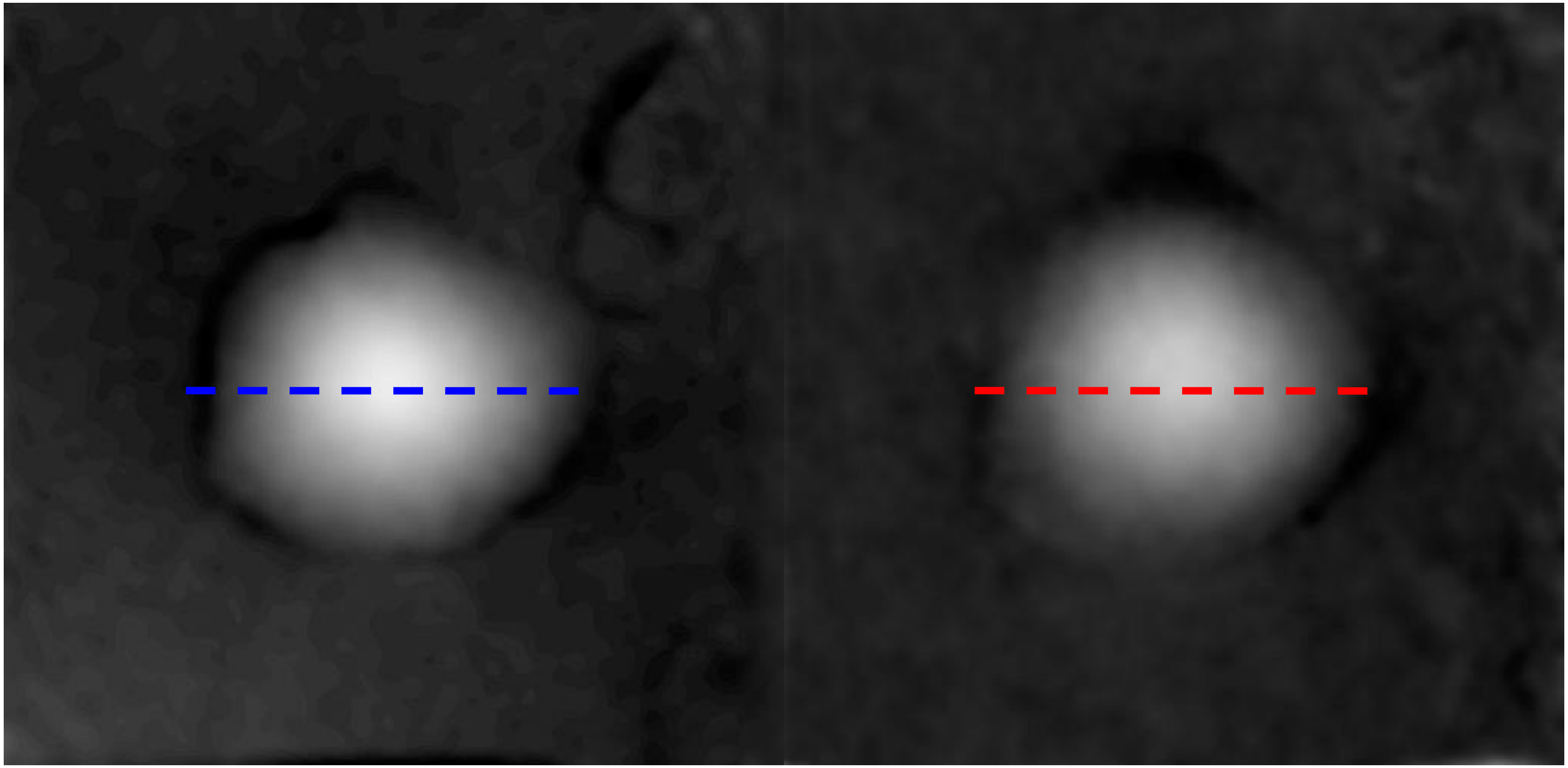


Almeida, T. P *et al.*, *Nature Communications*, 2014

Environmental TEM

Before oxidation **Oxidised**



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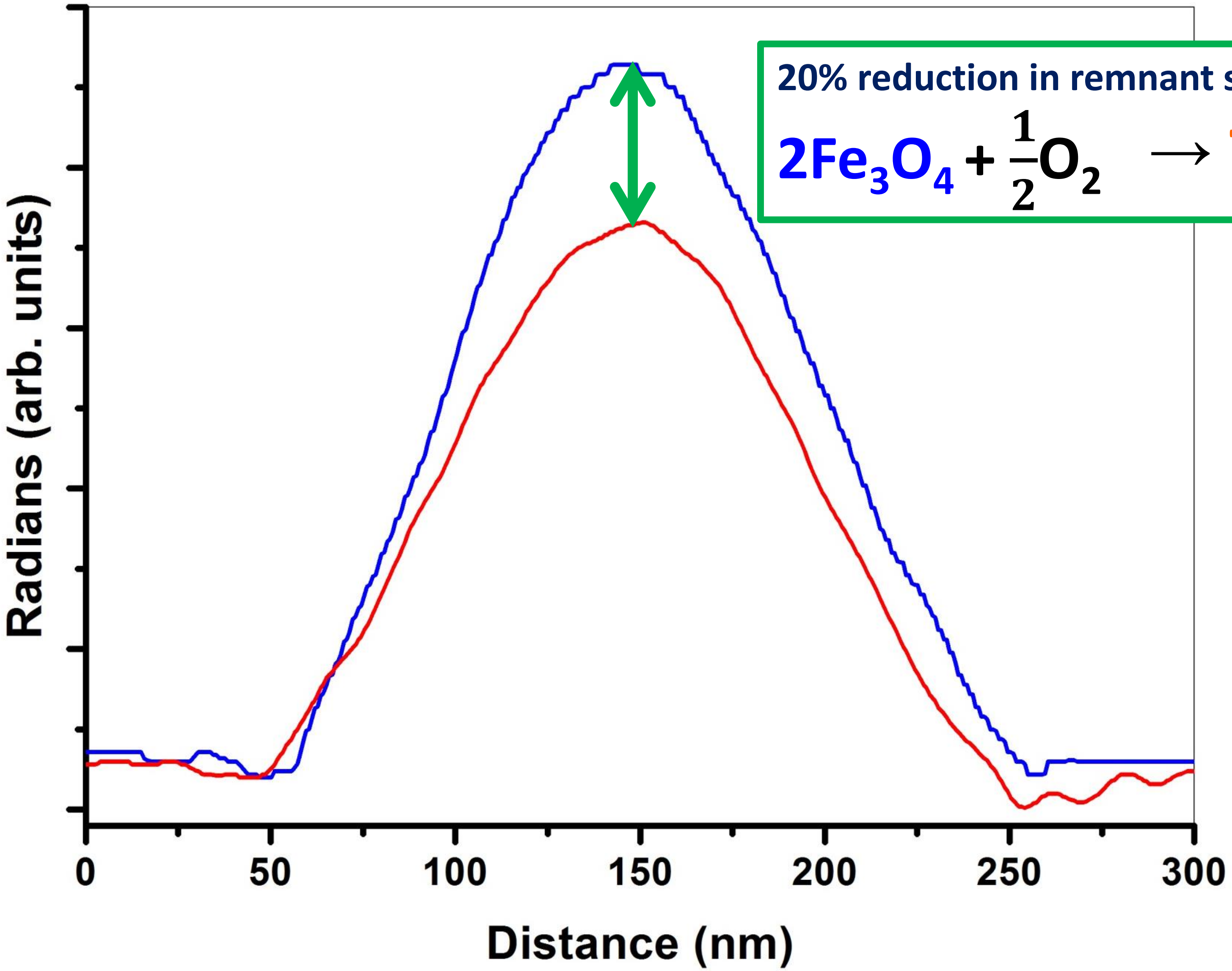
Environmental TEM



20% reduction in remnant saturation

$$2Fe_3O_4 + \frac{1}{2}O_2 \rightarrow \gamma\text{-}3Fe_2O_3$$

Almeida, T. P *et al.*, *Nature Communications*, 2014

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Thermomagnetic behaviour

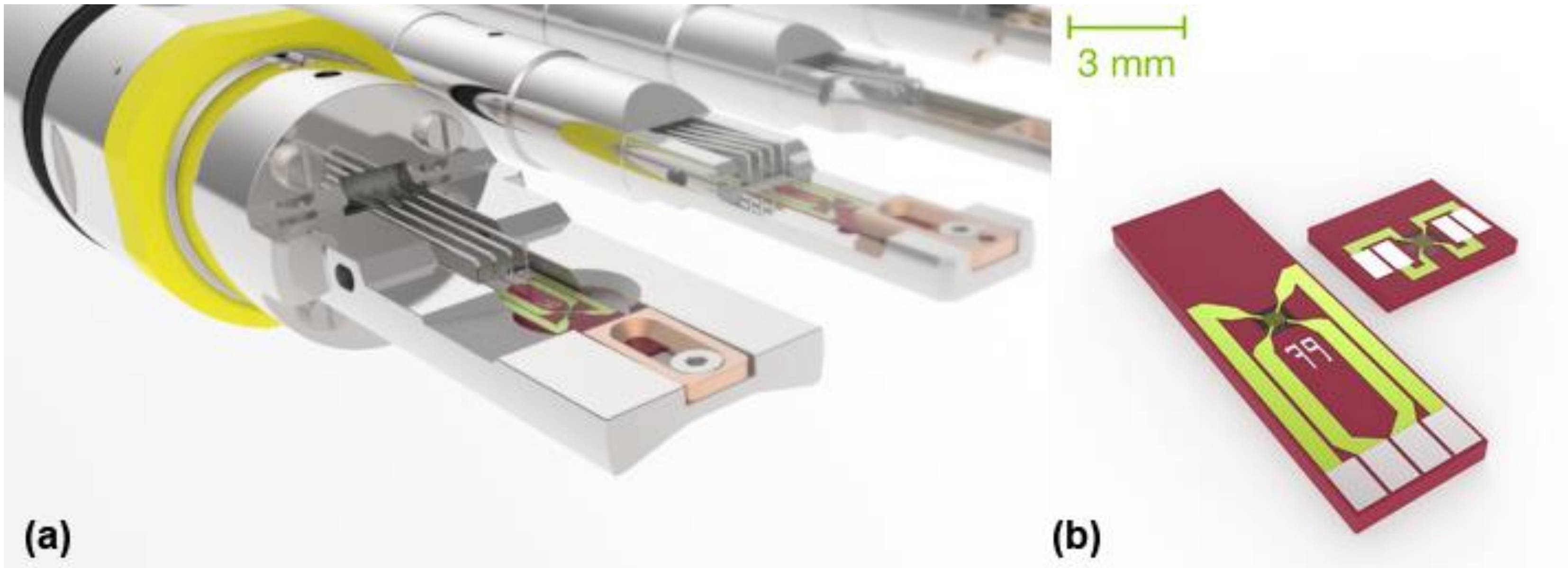
- **FEI Titan HOLO**
- **C_S correction on objective lens, *i.e.* image corrected**
- **Operated at 60 - 300kV**
- **3 biprisms and Lorentz lens for electron holography of magnetic fields**
- **11 mm pole piece gap to allow tilting to ± 75°**
- **Specifically designed to allow for *in situ* experiments**

Ernst Ruska-Centrum für Mikroskopie und Spektroskopie mit Elektronen ER-C

Thermomagnetic behaviour

DENS solutions

Wildfire heating holder and EMheaterchips^{TM}.

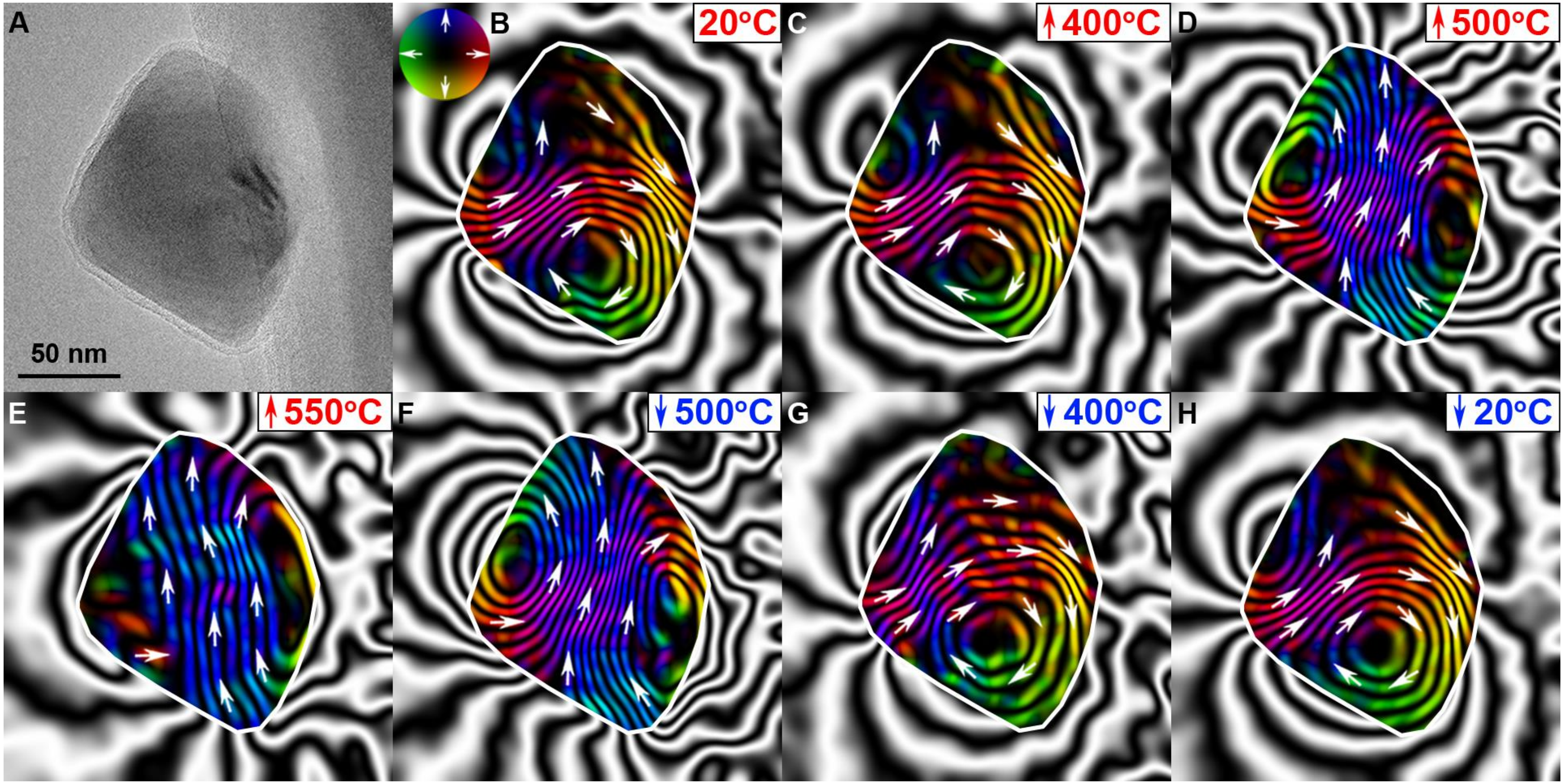


Ultra stable holder during heating up to < 1000°C

TRM = total phase shift – MIP (measured at each temperature in separate experiment)

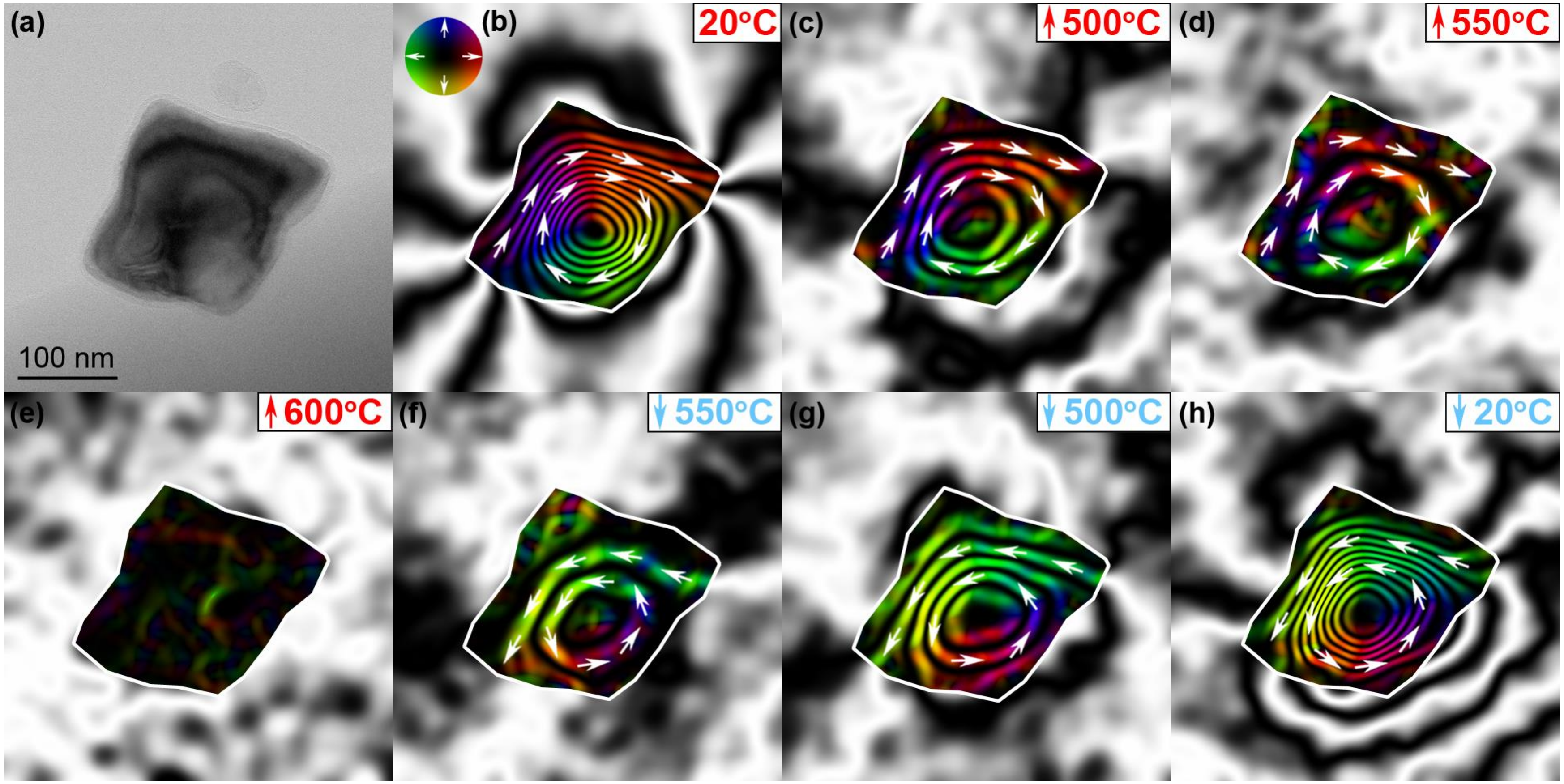
Thermomagnetic behaviour

Fe_3O_4 grain at remanence upon heating to 550°C and cooling to room temperature



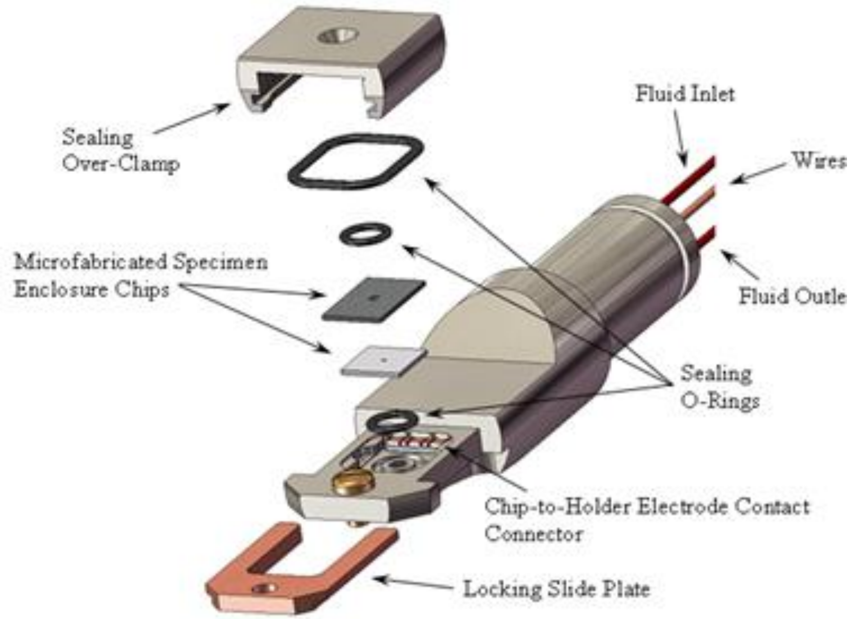
Thermomagnetic behaviour

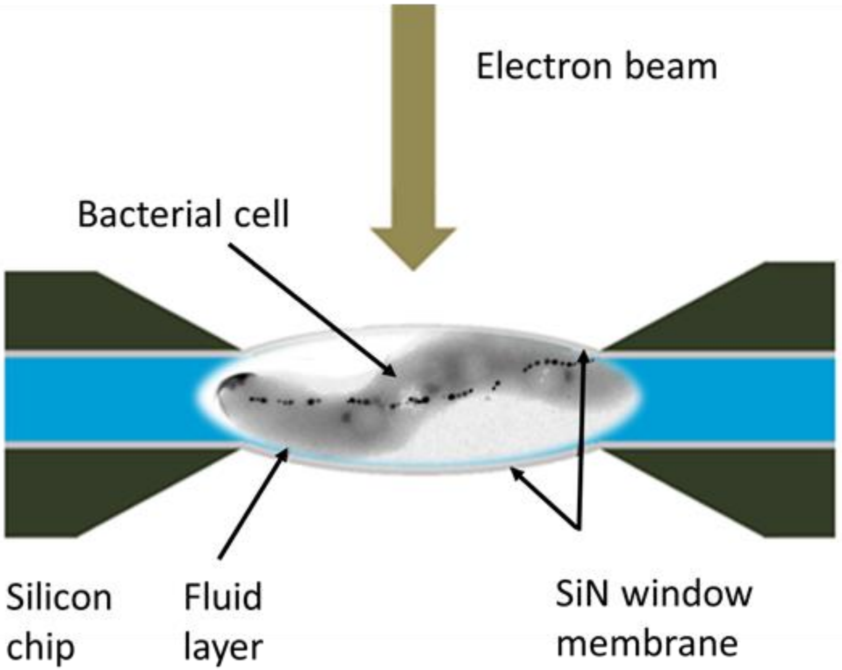
Fe_3O_4 grain at remanence upon heating to 600°C and cooling to room temperature



Liquid cell electron holography

Combining electron holography with liquid-cell TEM holder

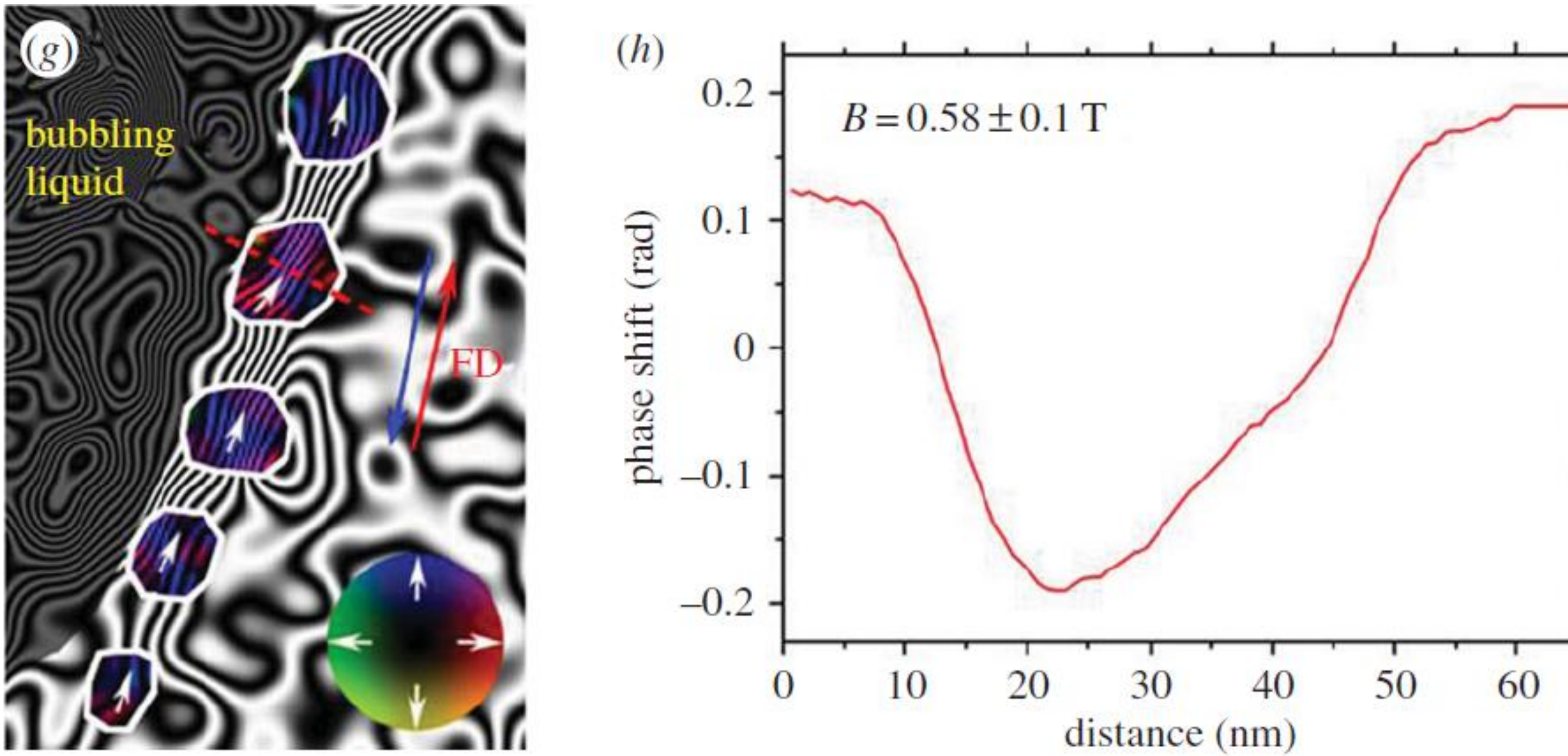




Prozorov T. *et al. Journal of Royal Society: Interface*, 2017

Liquid cell electron holography

Combining electron holography with liquid-cell TEM holder



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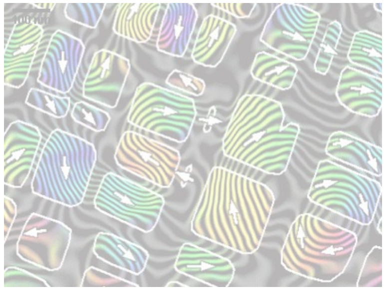
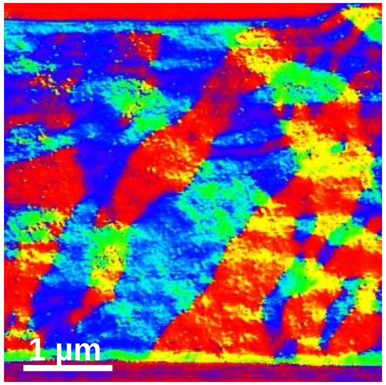
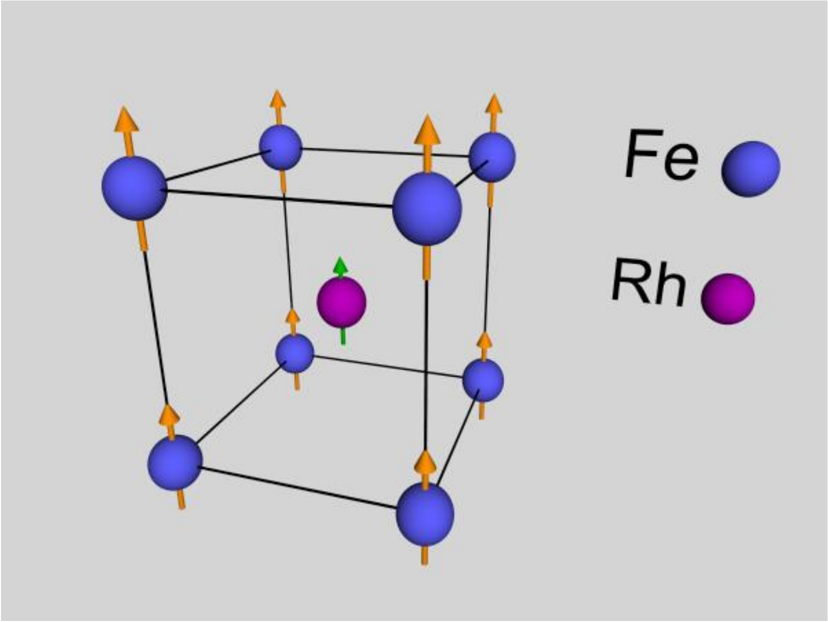
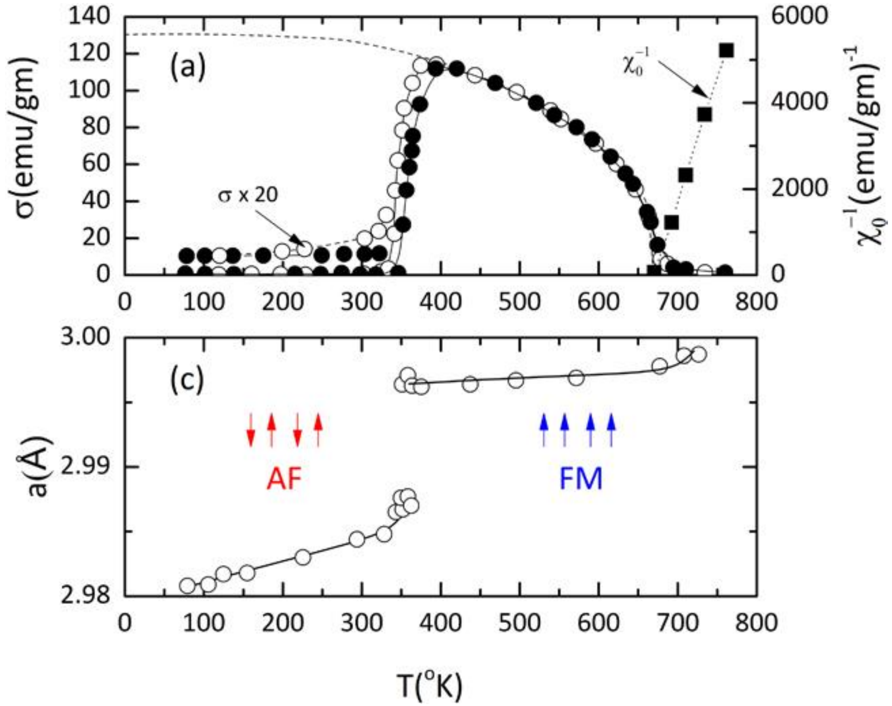
- **Phase transition in FeRh thin films**
  - ➢ Differential phase contrast imaging (DPC)





Equiatomic intermetallic iron-rhodium compound ($Fe_{48}Rh_{52}$ to $Fe_{56}Rh_{44}$)





380 K: AF → FM

Fe: ~3.1 μ_B

Rh: ~1 μ_B

Zakharov A I *et al.*, *J. Exp. Theor. Phys.*, 1964
Kouvel J S *et al., J. Appl. Phys.* 1962

Sample preparation

DC magnetron sputter co-deposition

- FeRh targets
- MgO and GaAs substrates
- Samples:
  1) FeRh on MgO
  2) Planar FeRh TEM samples via HF-etching

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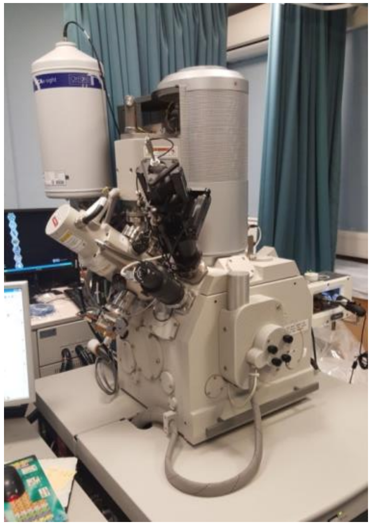
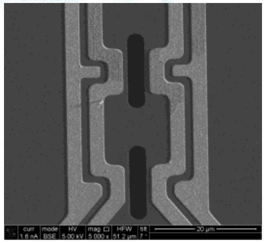
Focused Ion Beam / Scanning Electron Microscope (FIB-SEM)

- Cross-sectional and planar FeRh samples

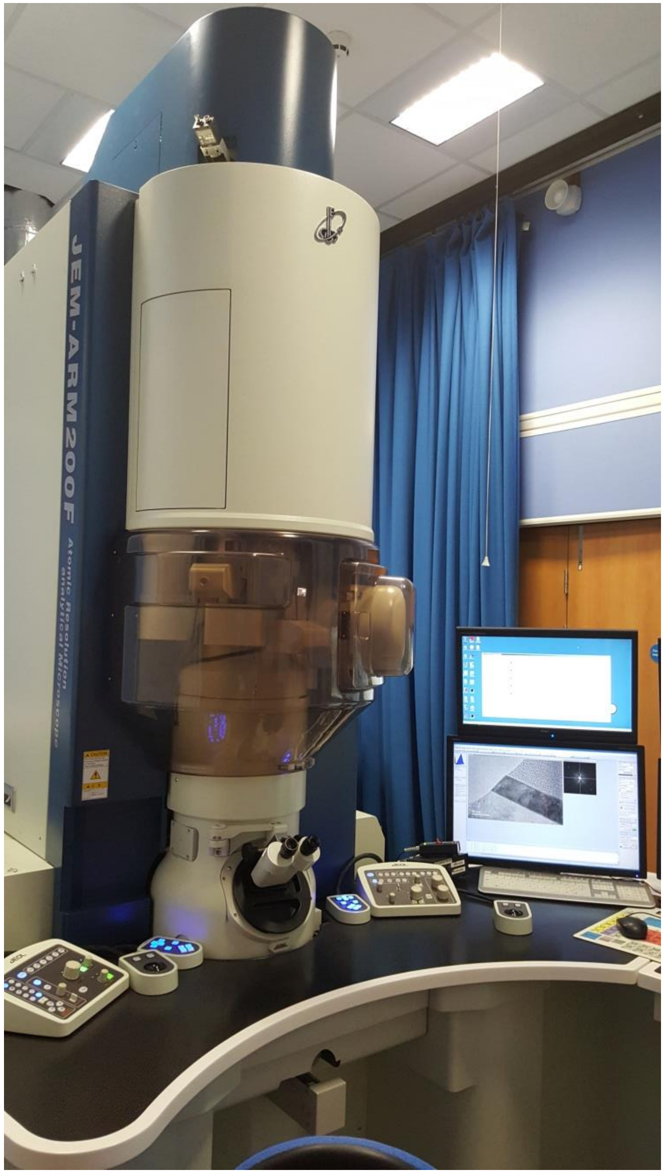


DENS solutions

Lightning holder with heater / biasing MEMS chip



Sample characterisation



- **JEOL ARM200cF - "MagTEM"**
- **C_S correction on condenser lens, *i.e.* probe corrected**
- **Operated at 60kV - 200kV**
- **HR-STEM and chemical mapping at atomic scale, with EDX and EELS**
- **Lorentz lens and segmented / pixelated detectors for imaging of magnetisation**

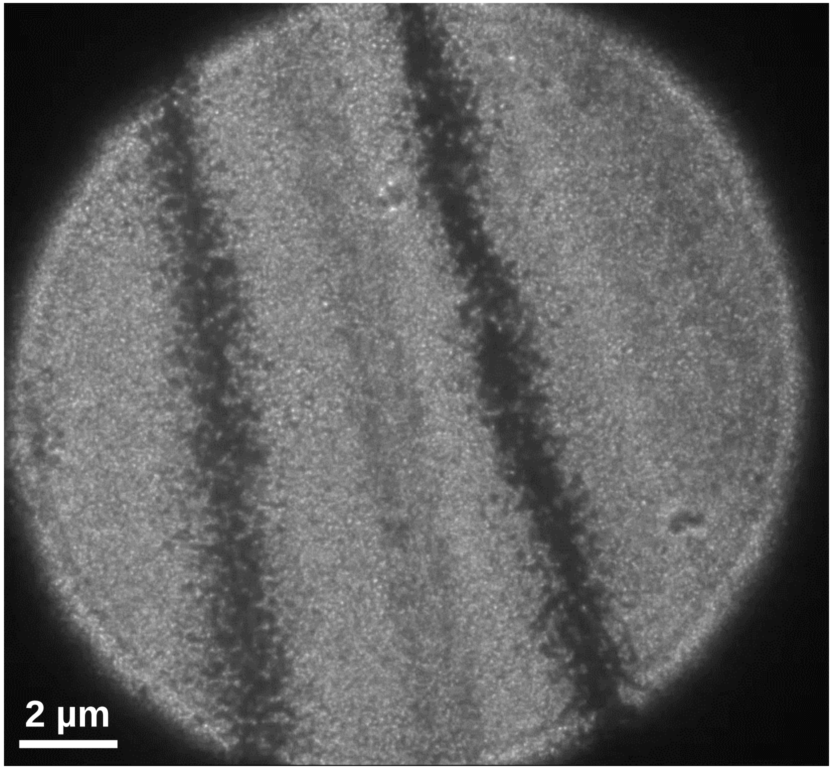
Kelvin Nanocharacterisation Centre

Phase transition in FeRh

HF-etched FeRh planar sample as a function of temperature

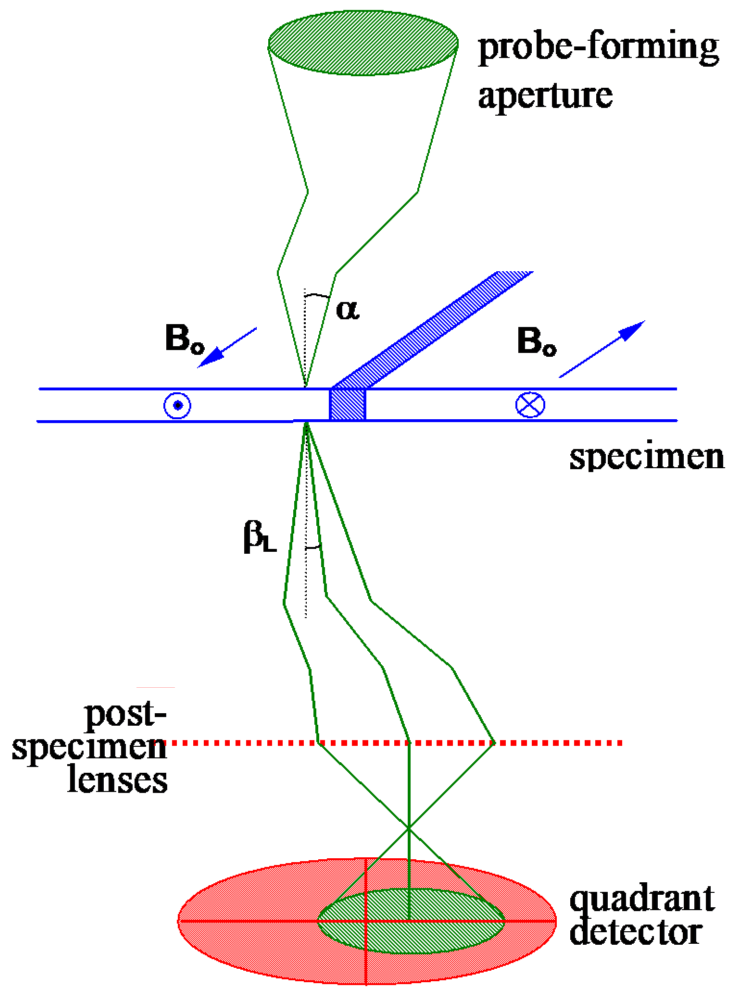
20°C → 140°C → 20°C

Bright field TEM imaging

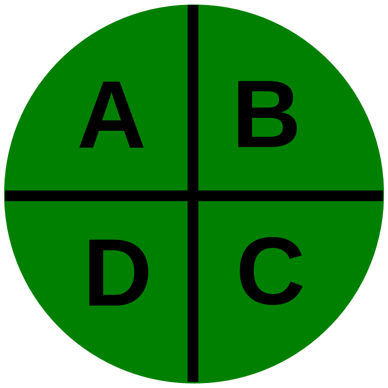


Low-angle electron diffraction

Differential phase contrast imaging

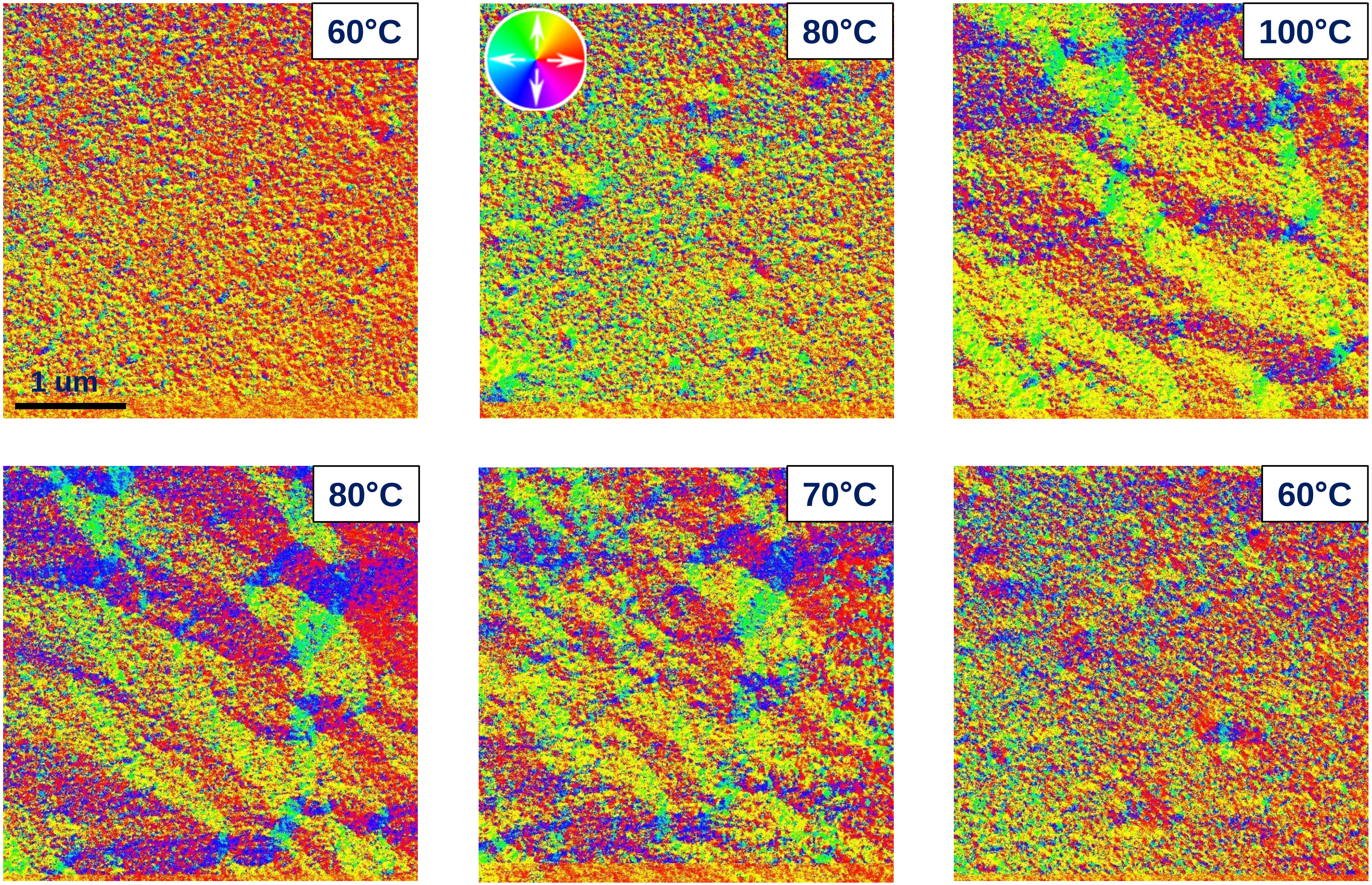


- STEM mode focused probe on sample, probe semi angle α.
- Beam deflected by Lorentz force.
- Segmented detector can then be used to map deflection by taking difference signals from opposite segments (quadrants or halves)



In situ imaging of FeRh transition

HF-etched FeRh planar sample as a function of temperature



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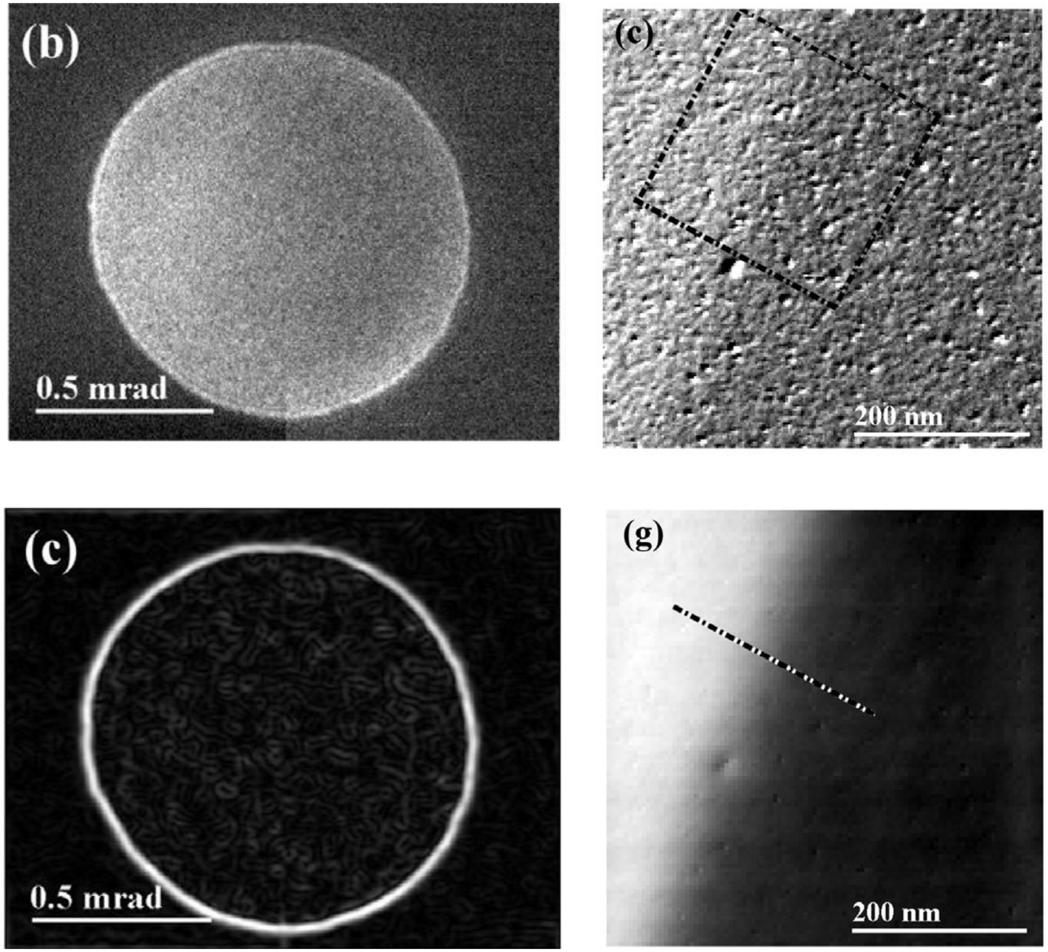
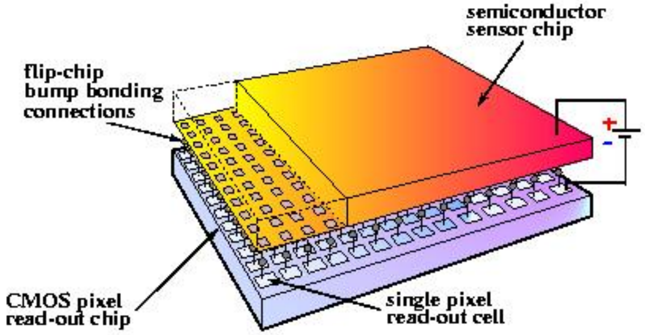
In situ imaging of FeRh transition

Medipix pixelated detector can remove effects of diffraction contrast



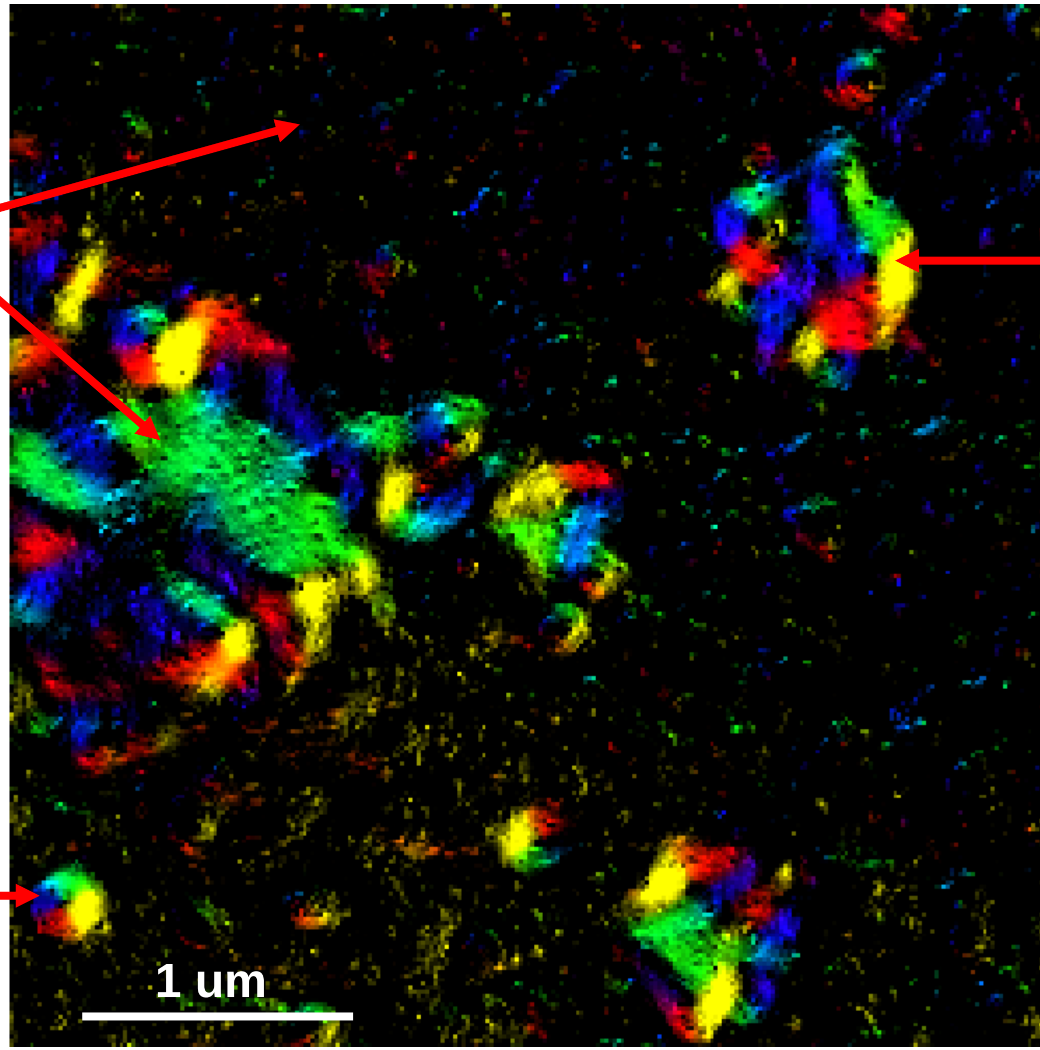


Krajnak, M. *et al. Ultramicroscopy*, 2016

In situ imaging of FeRh transition

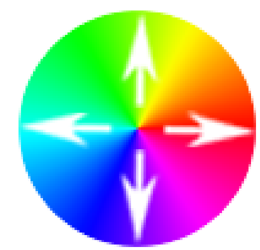
HF-etched FeRh examined using the Medipix detector at 80°C

AFM / FM co-existence



Complex domain structures

Small vortex ~ 200 nm



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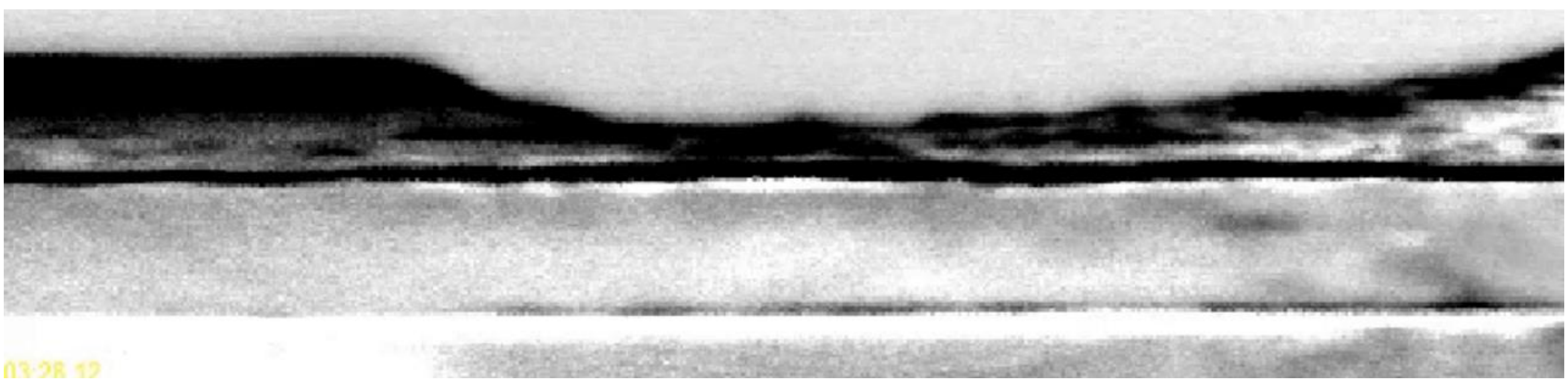
University of Glasgow

In situ imaging of FeRh transition

Cross-section of 55 nm FeRh on MgO substrate

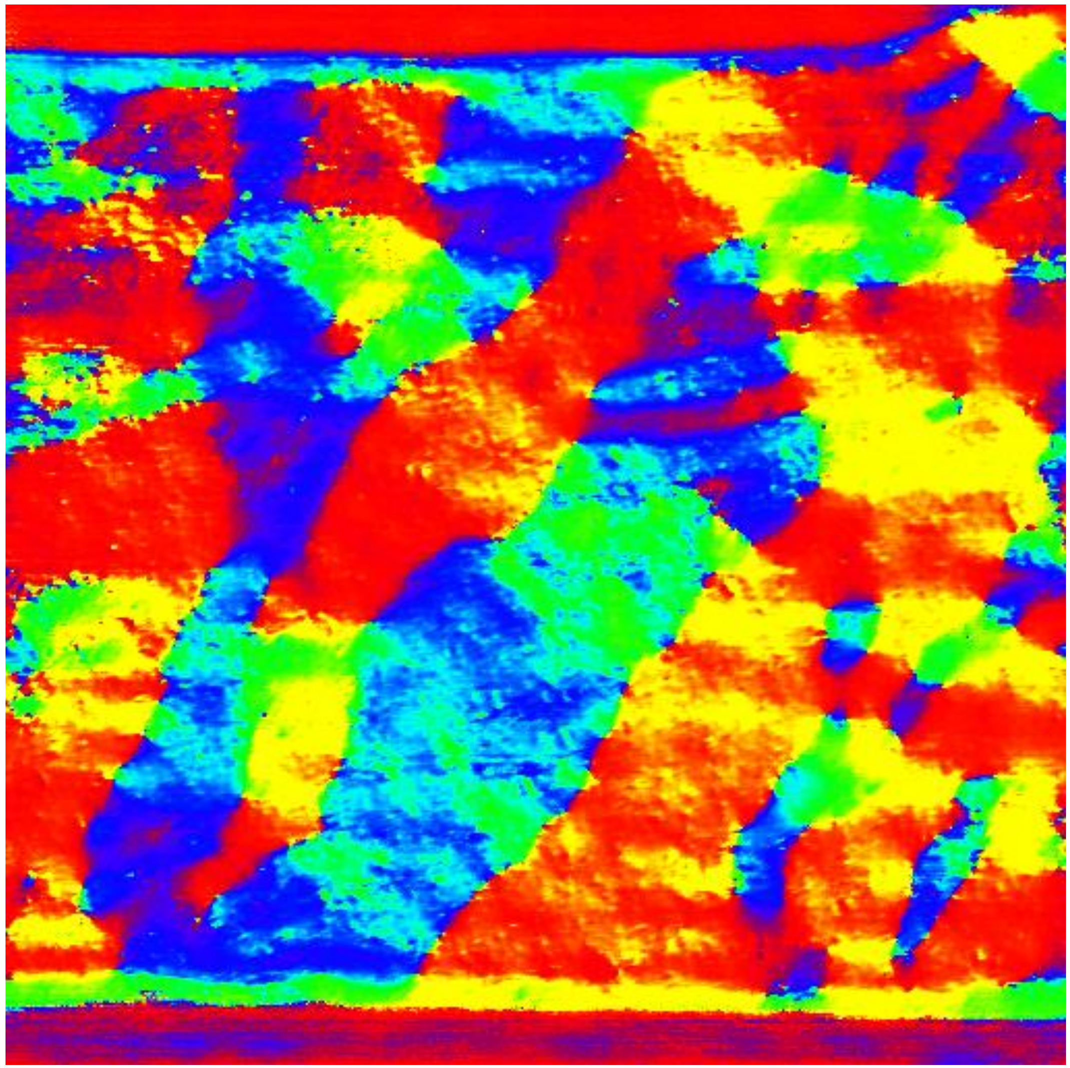
20°C → 200°C

200°C → 20°C

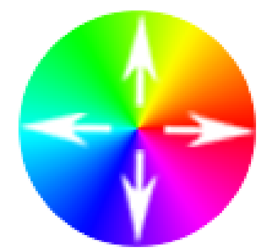


Current-driven domain wall motion

Planar TEM lamella of FeRh on NiAl on GaAs substrate at 150°C

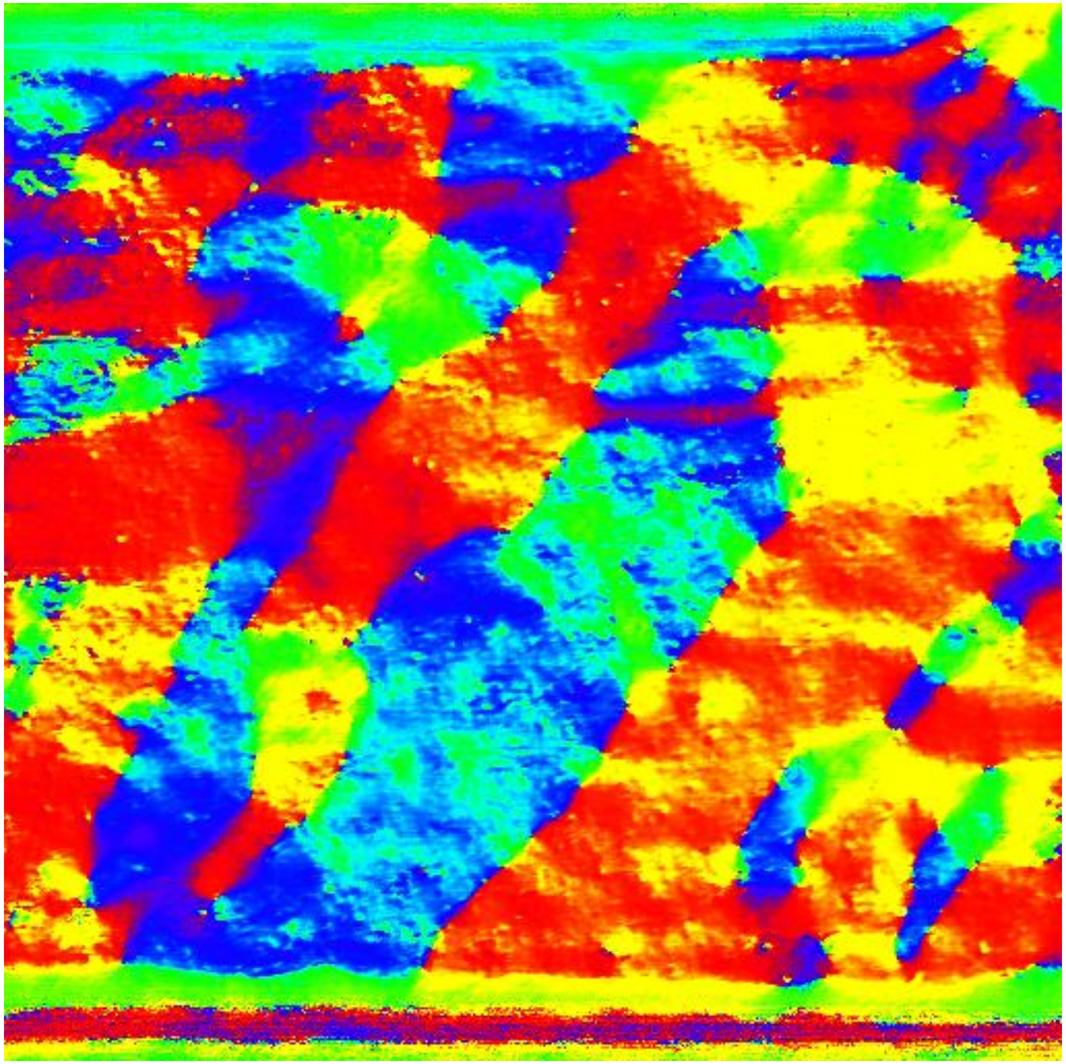


1 µm



Current-driven domain wall motion

Biasing - 500μs pulses increasing from 100mV to 1.1V

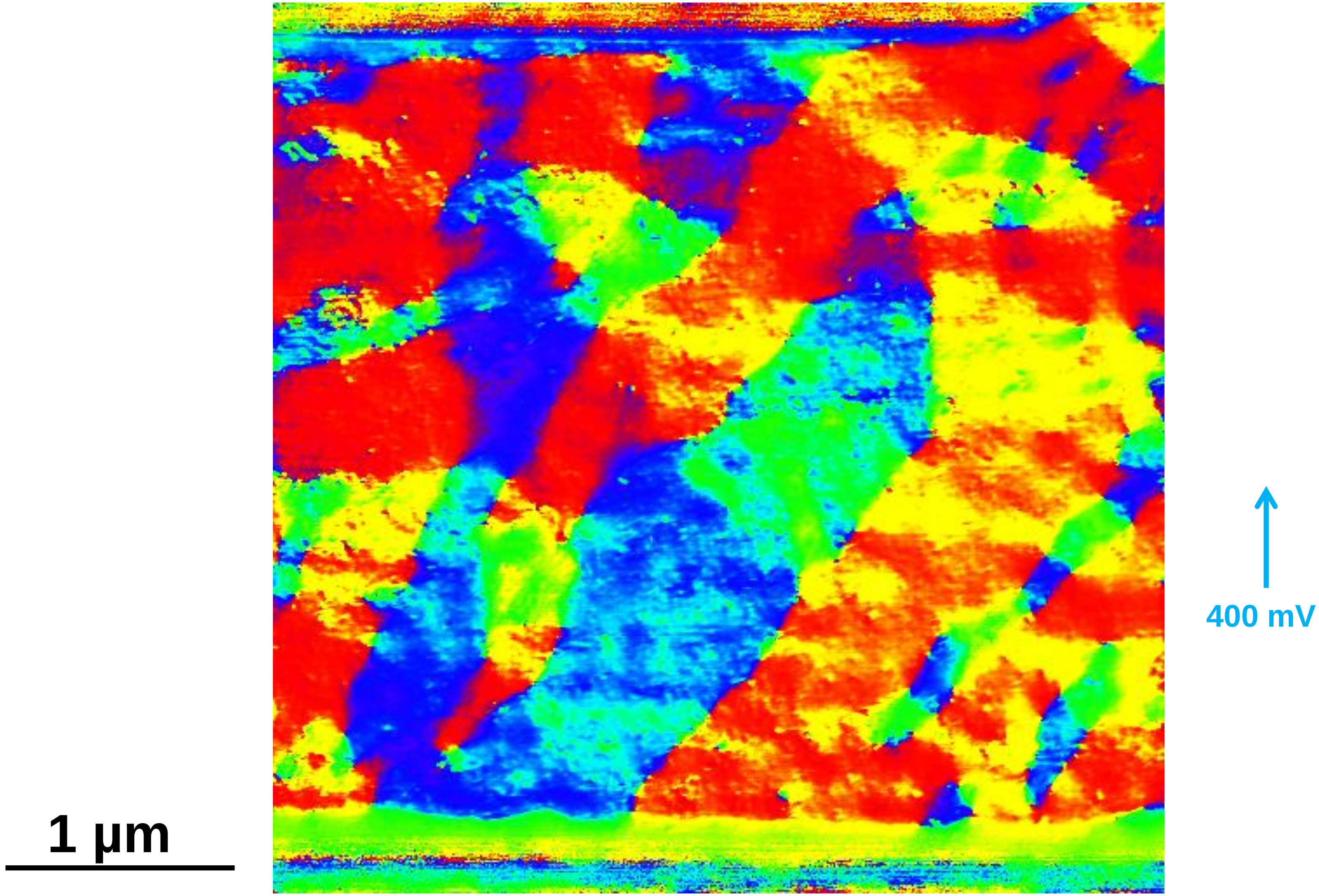


1 μm

100 mV

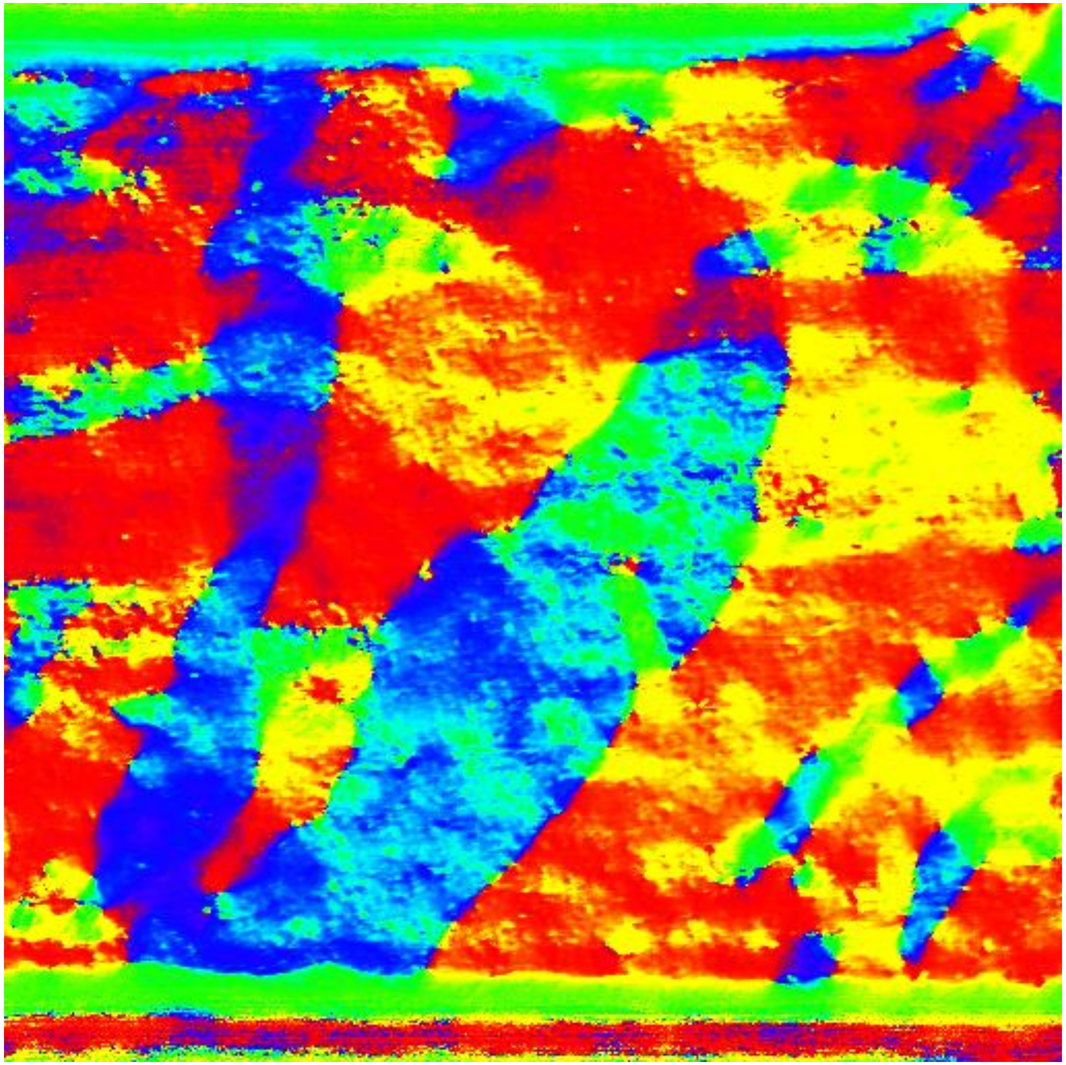
Current-driven domain wall motion

Biasing - 500µs pulses increasing from 100mV to 1.1V



Current-driven domain wall motion

Biasing - 500µs pulses increasing from 100mV to 1.1V

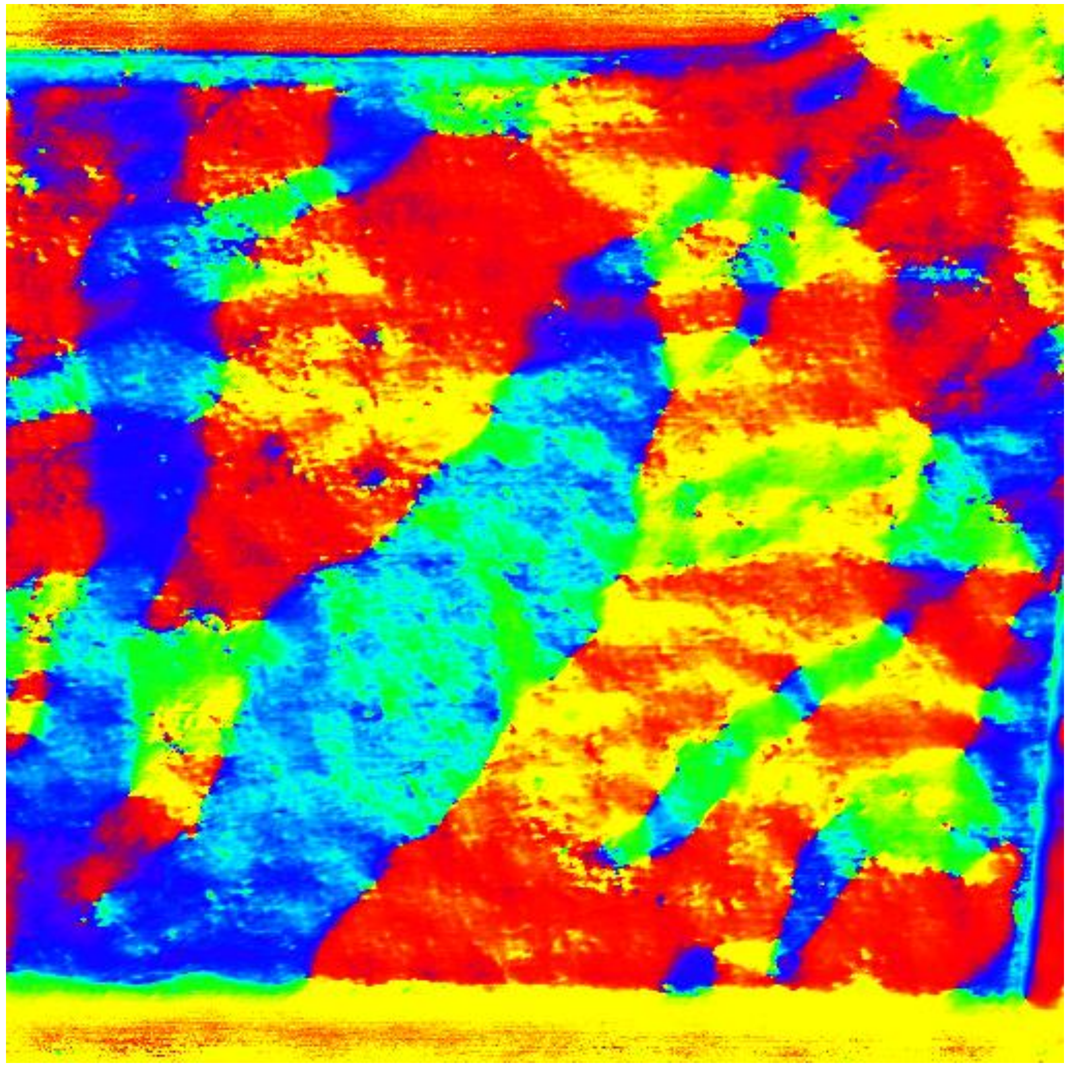


700 mV

1 µm

Current-driven domain wall motion

Biasing - 500μs pulses increasing from 100mV to 1.1V

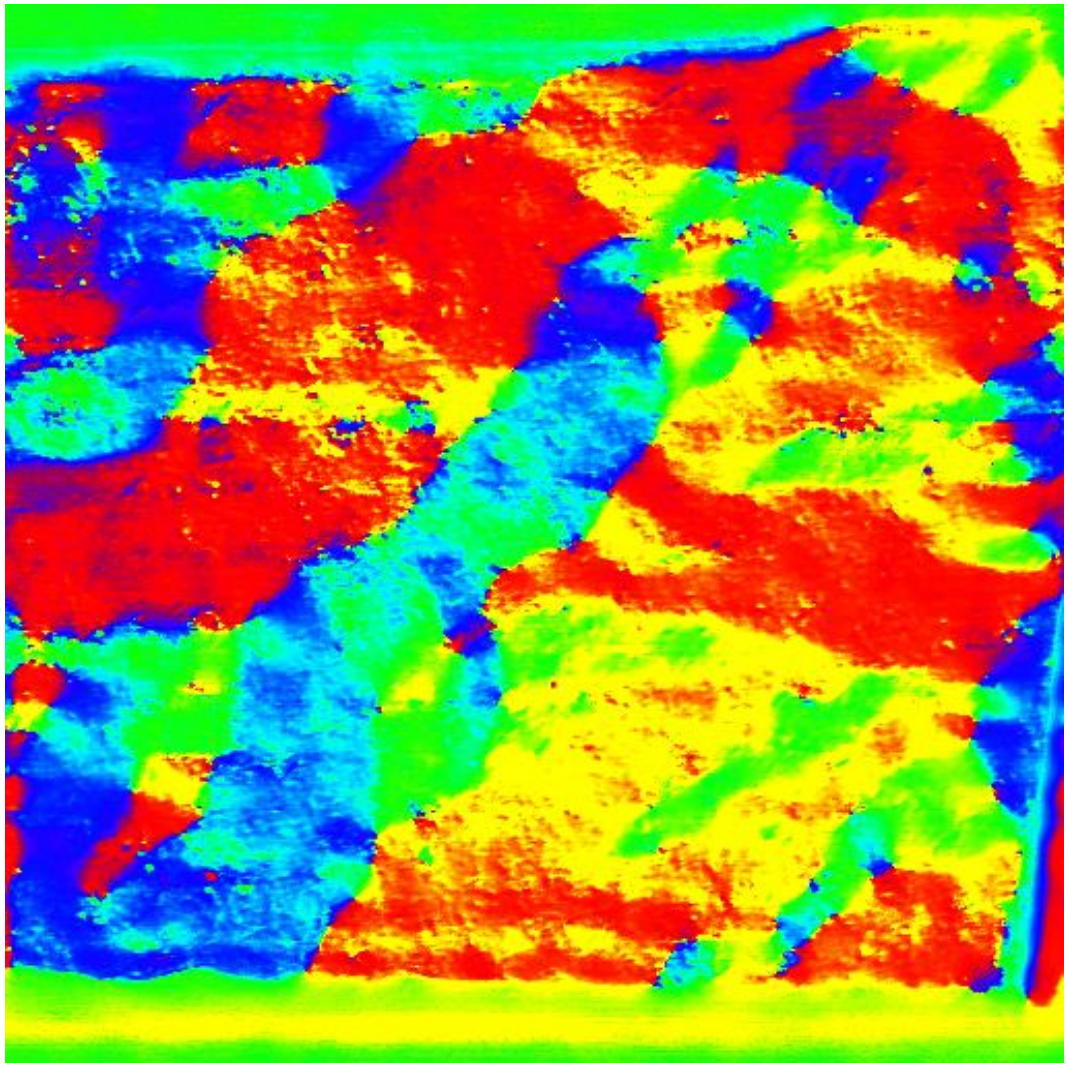


1 μm

800 mV

Current-driven domain wall motion

Biasing - 500µs pulses increasing from 100mV to 1.1V

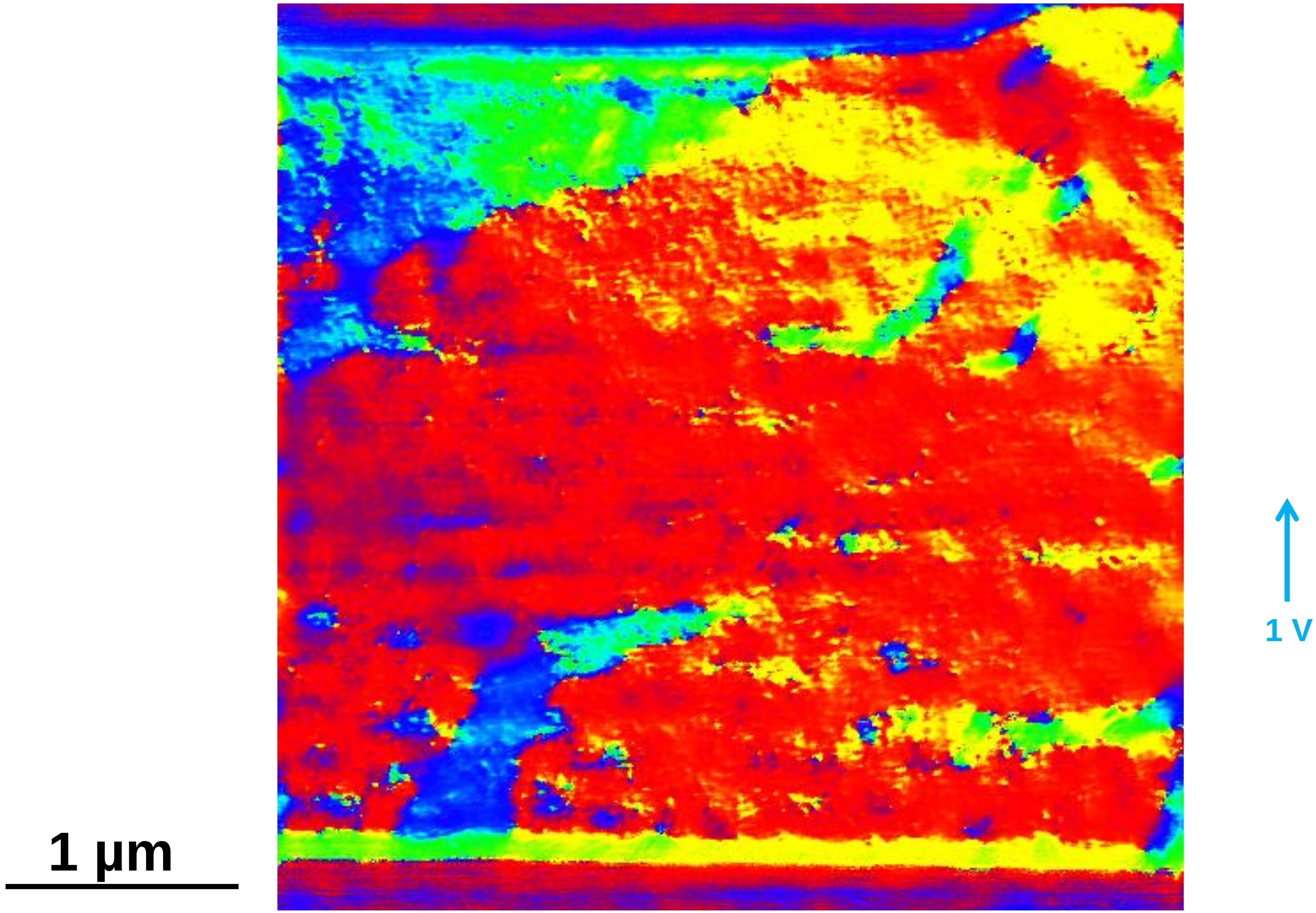


1 µm

900 mV

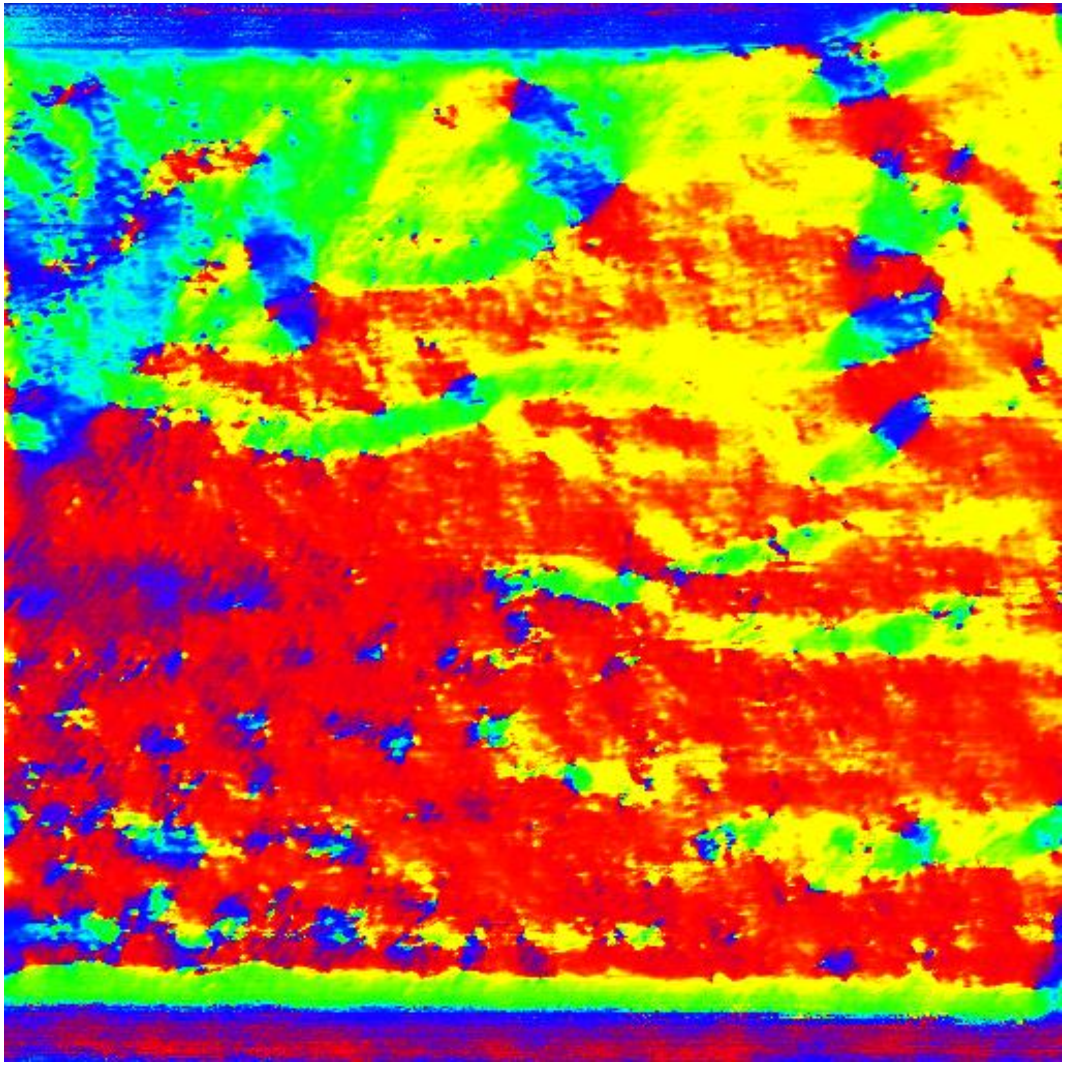
Current-driven domain wall motion

Biasing - 500µs pulses increasing from 100mV to 1.1V



Current-driven domain wall motion

Biasing - 500µs pulses increasing from 100mV to 1.1V



↑ 1.1 V

1 µm

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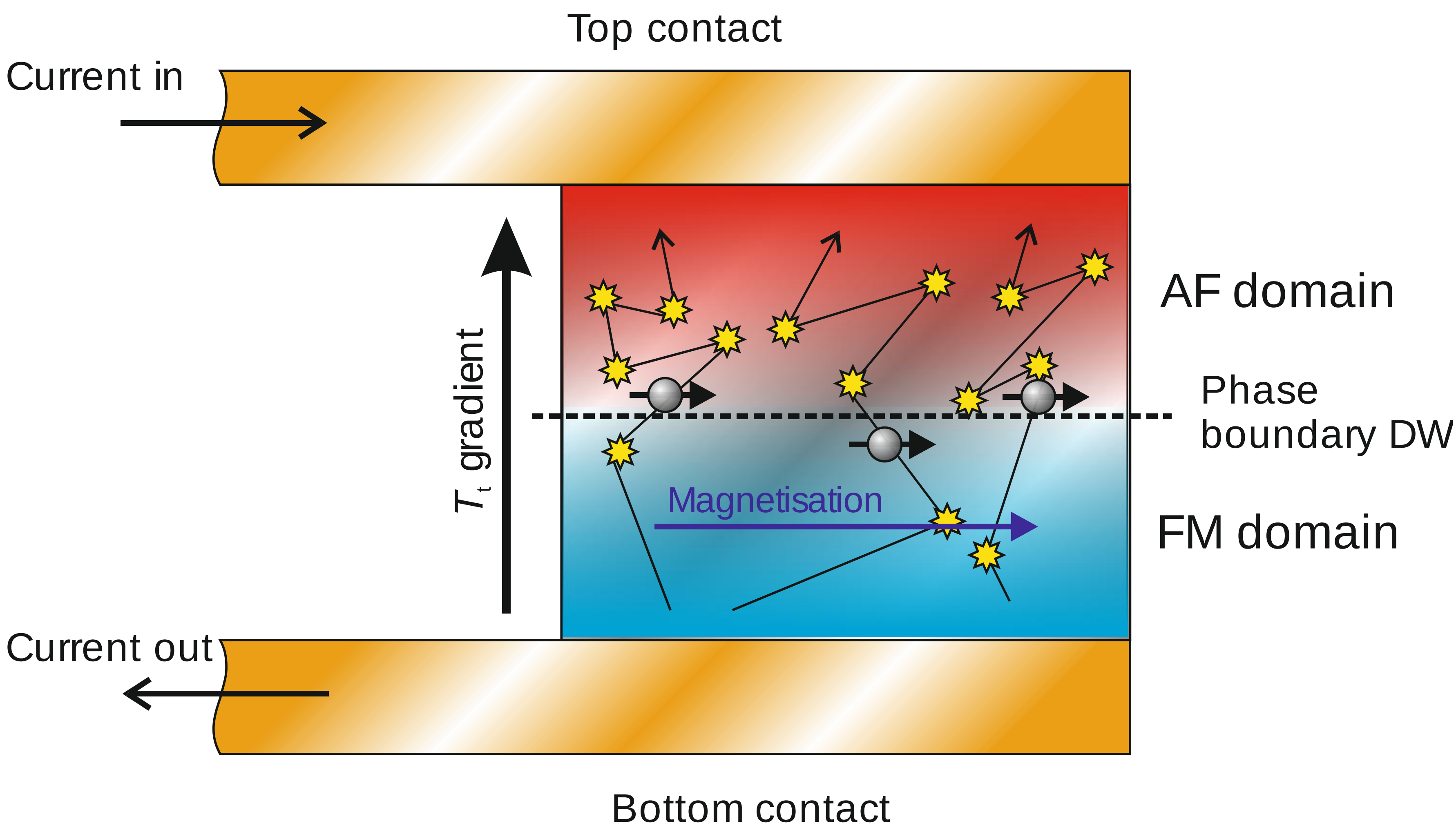
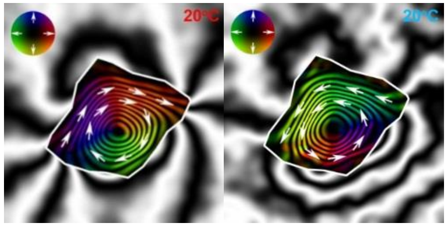
Current-driven domain wall motion



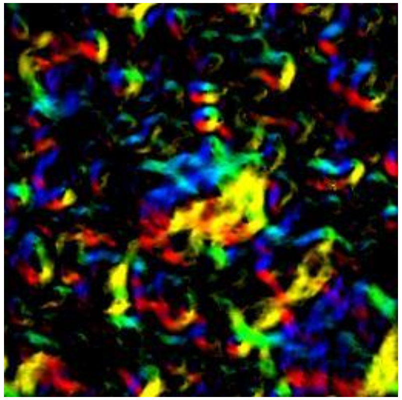
Summary

- **Specialised TEM techniques can be used to visualise nanoscale magnetism and provide fundamental insight for a range of applications**

- **Combining electron holography with *in situ* TEM and ETEM provided direct access to:**
  - **The effect of oxidation on vortex-state Fe_3O_4 grains**
  - **Thermomagnetic behaviour of vortex states**



- **Lorentz techniques like Fresnel, SAES, conventional and pixelated DPC imaging allow for:**
  - **Visualising the dynamic nucleation and agglomeration ferromagnetic domains during transition in FeRh**
  - **DW and phase boundary motion can be controlled by current pulsing and Ir / Pd doping of the FeRh films**



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Thank you