

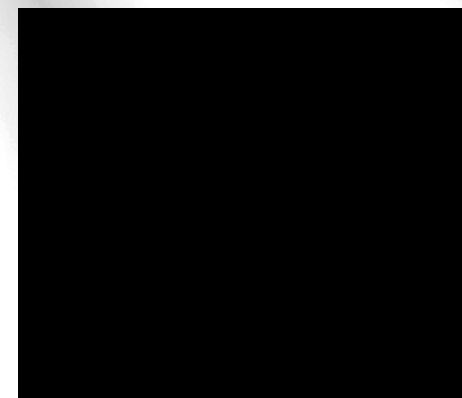
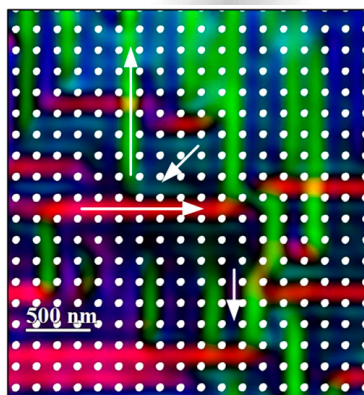
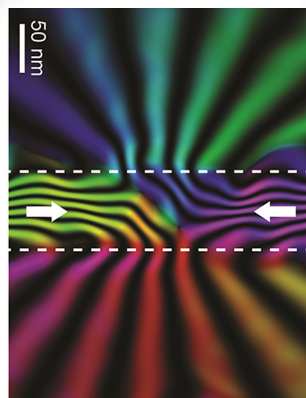
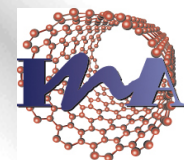
Electron Holography and Lorentz microscopy for the quantitative measurements of local magnetic properties and *in-situ* experiments

Etienne SNOECK, C. GATEL, C. MAGEN*,
A. MASSEBOEUF, L.A. RODRIGUEZ,

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Universidad de Zaragoza - Spain



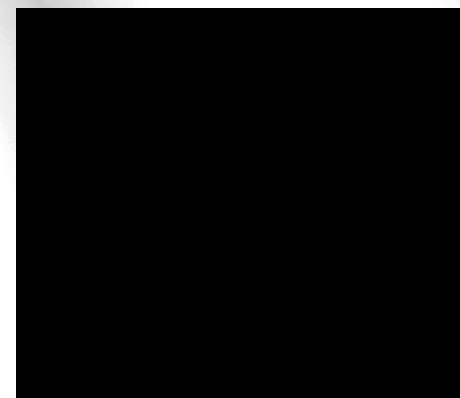
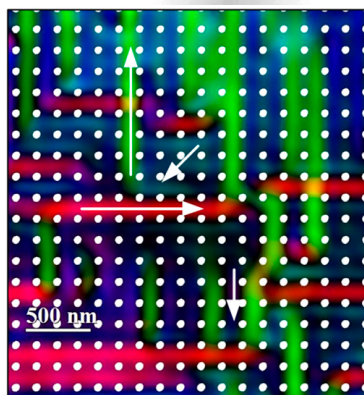
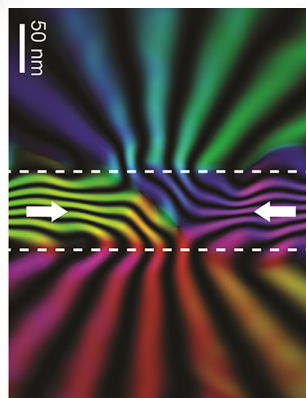
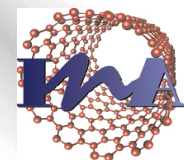
Electron Holography and Lorentz microscopy for the quantitative measurements of local magnetic properties and *in-situ* experiments

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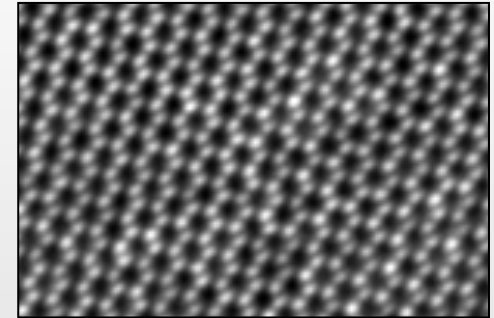
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Transmission Electron Microscopy

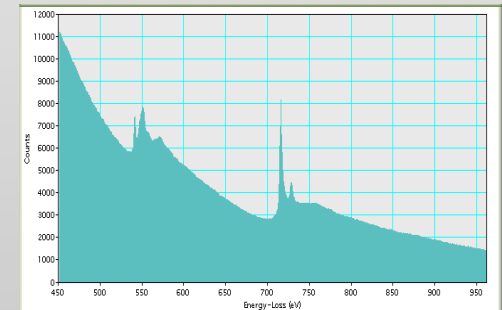
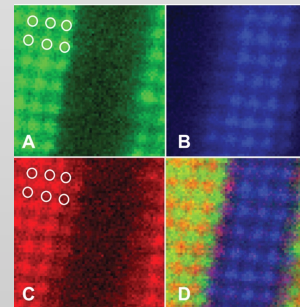
- **Structure imaging**

- Conventional BF-DF imaging
- High resolution TEM (HRTEM)
- HRSTEM - HAADF



- **Local chemical measurements and chemical mapping**

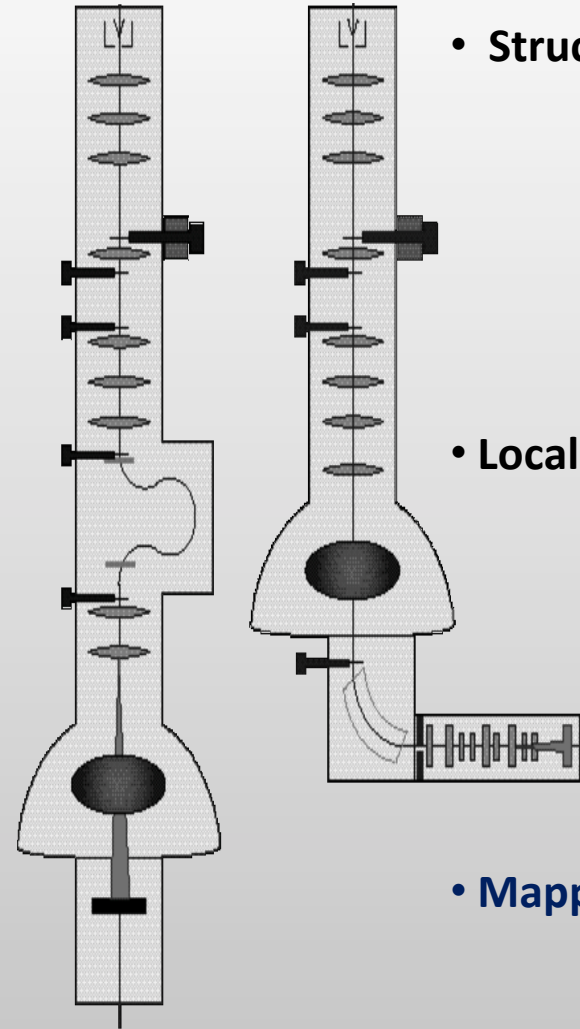
- Electron Energy Loss Spectroscopy (EELS)
- EFTEM



- **Mapping local fields**

- **Magnetic fields**
- Electrostatic fields
- Strain fields

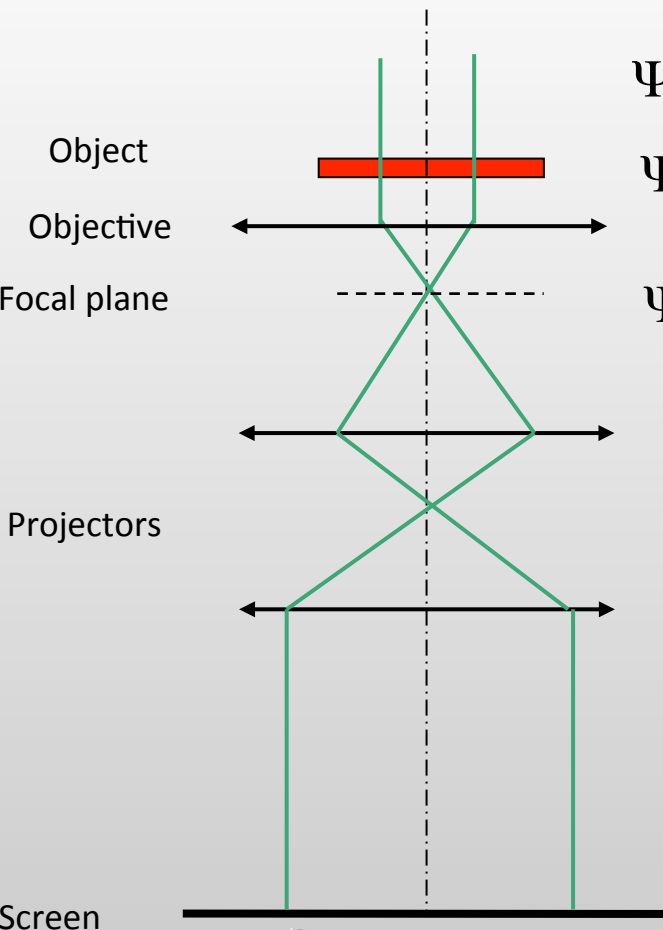
➔ **Electron Holography**



Outlook

- ◆ Lorentz TEM and Electron Holography
- ◆ Exple1 : Study of the phase diagram of Fe nanocubes by EH
- ◆ Exple 2 : *In-situ* Lorentz studies of the magnetic reversal of Co antidot arrays
- ◆ Exple 3 : *In-situ* EH studies of the magnetic induction generated by a HDD writer-head *in operando*

Electron wave in a TEM



$$\Psi_0 = \exp(i\vec{k} \cdot \vec{r})$$

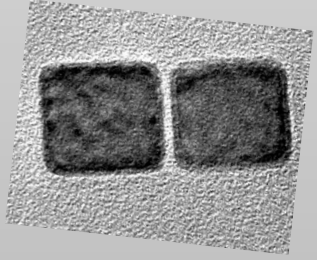
$$\Psi_{obj} = a_0(\vec{r}) \exp(i\vec{k} \cdot \vec{r} + \phi_0(\vec{r}))$$

$$\Psi_s = a_s(\vec{r}) \exp(i\vec{k} \cdot \vec{r} + \phi_s(\vec{r}))$$

Phase shift after interaction with the object

Phase shift below the objective

$$I(x, y) \propto |\Psi_s|^2 \propto a_s^2(\vec{r}) \rightarrow \phi_s(\vec{r}) \text{ is lost!}$$



What information is carried by $\phi_s(\vec{r})$?

$$\Psi_0 = \exp(i\vec{k} \cdot \vec{r})$$



Electromagnetic field

- Electrostatic potential : V
- Magnetic Potential vector : \vec{A}



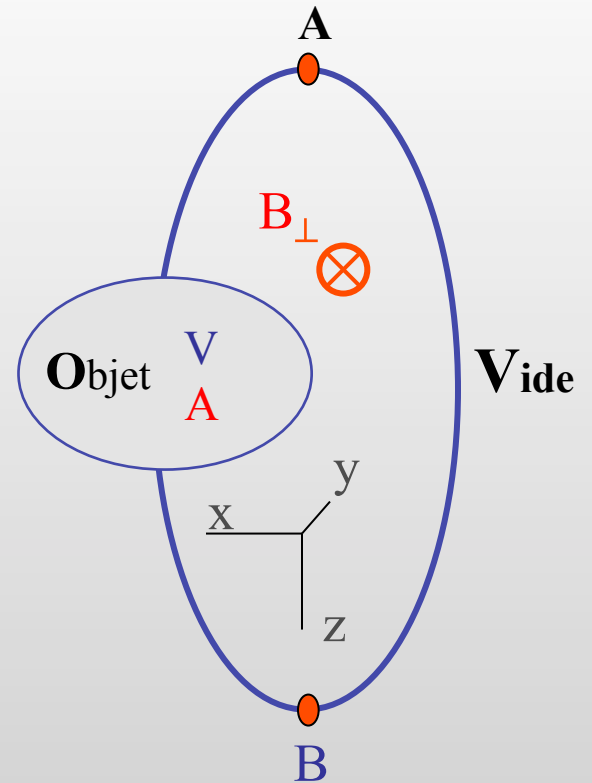
Aharonov-Bohm

$$\Psi_s = a_s(\vec{r}) \exp(i(\vec{k} \cdot \vec{r} + \phi_s(\vec{r})))$$

$$\begin{aligned} \phi_s(x, y) &= C_E \int V(x, y, z).dz - \frac{e}{\hbar} \int A_z(x, z).dz \\ &= C_E \int V(x, y, z).dz - \frac{e}{\hbar} \iint B_{\perp}(x, z).dx.dz \end{aligned}$$

Electrostatic potential contribution to the phase shift

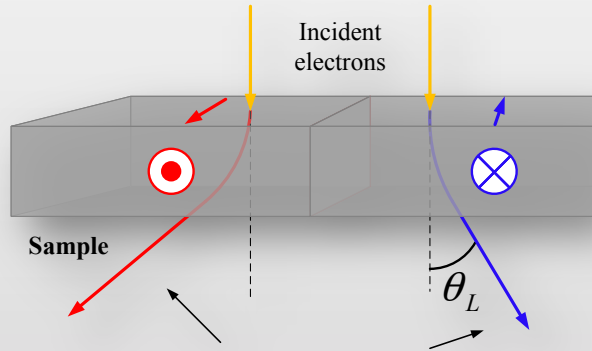
Magnetic contribution to the phase shift



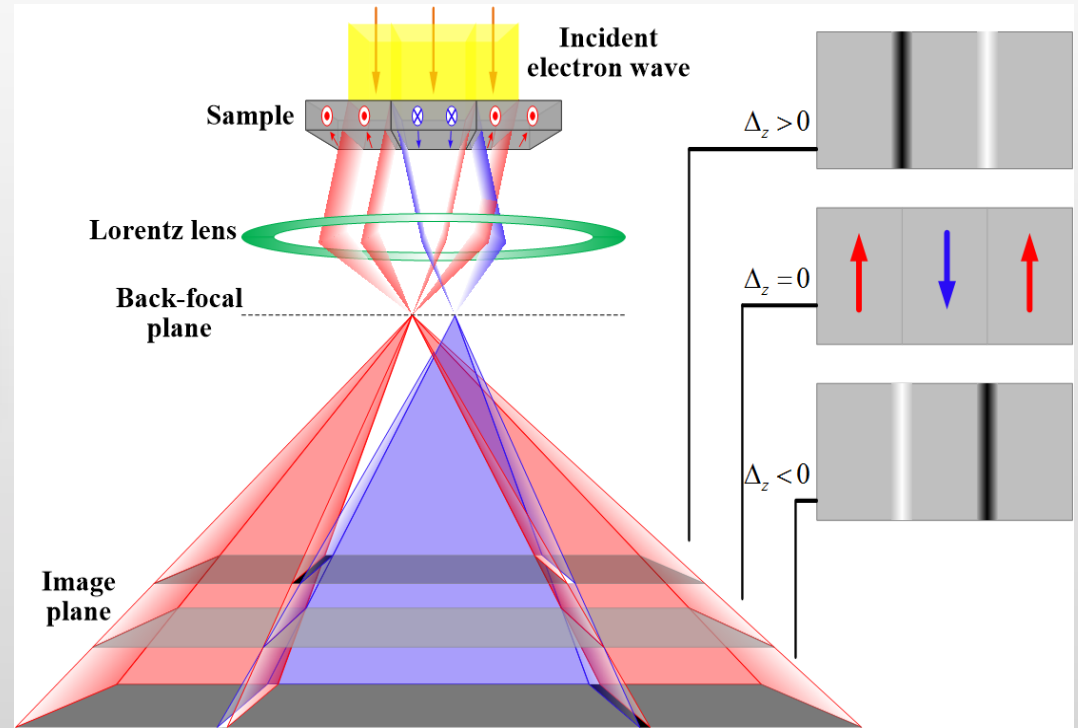
$$C_E = \left(\frac{2\pi}{\lambda} \right) \left(\frac{E + E_0}{E(E + 2E_0)} \right)$$

1. LORENTZ Microscopy (Fresnel Mode)

Lorentz Microscopy is based on the **Lorentz Force** that experiment the electrons



→ We can observe directly **DOMAIN WALLS**.



$$\Psi_{obj} = a_0(\vec{r}) \exp(i(\vec{k} \cdot \vec{r} + \phi_0(\vec{r})))$$

TEM
→

$$\Delta z_0 \rightarrow \Psi_{s_0} = a_{s_0}(\vec{r}) \exp(i(\vec{k} \cdot \vec{r} + \phi_{s_0}(\vec{r})))$$

$$\Delta z_1 \rightarrow \Psi_{s_1} = a_{s_1}(\vec{r}) \exp(i(\vec{k} \cdot \vec{r} + \phi_{s_1}(\vec{r})))$$

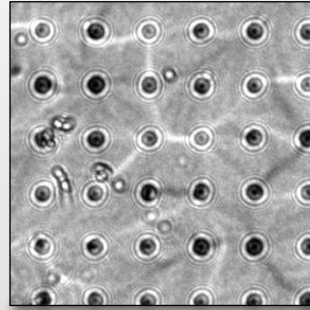
$$\Delta z_2 \rightarrow \Psi_{s_2} = a_{s_2}(\vec{r}) \exp(i(\vec{k} \cdot \vec{r} + \phi_{s_2}(\vec{r})))$$

Phase retrieval Process

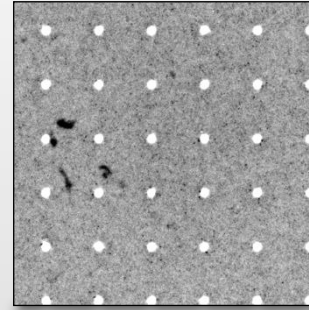
1- Take a series of images in three focal planes:

- **underfocus**
- **overfocus**
- **in-focus**

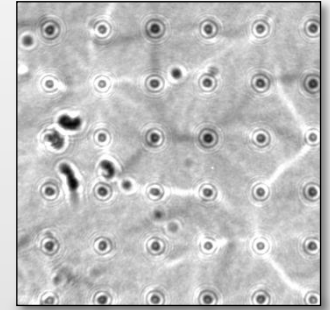
$I(r, \Delta_z)$



$I(r, 0)$



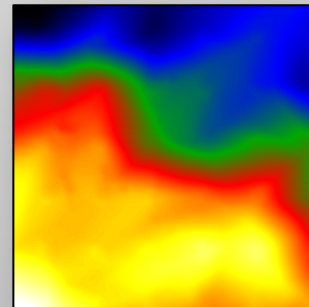
$I(r, -\Delta_z)$



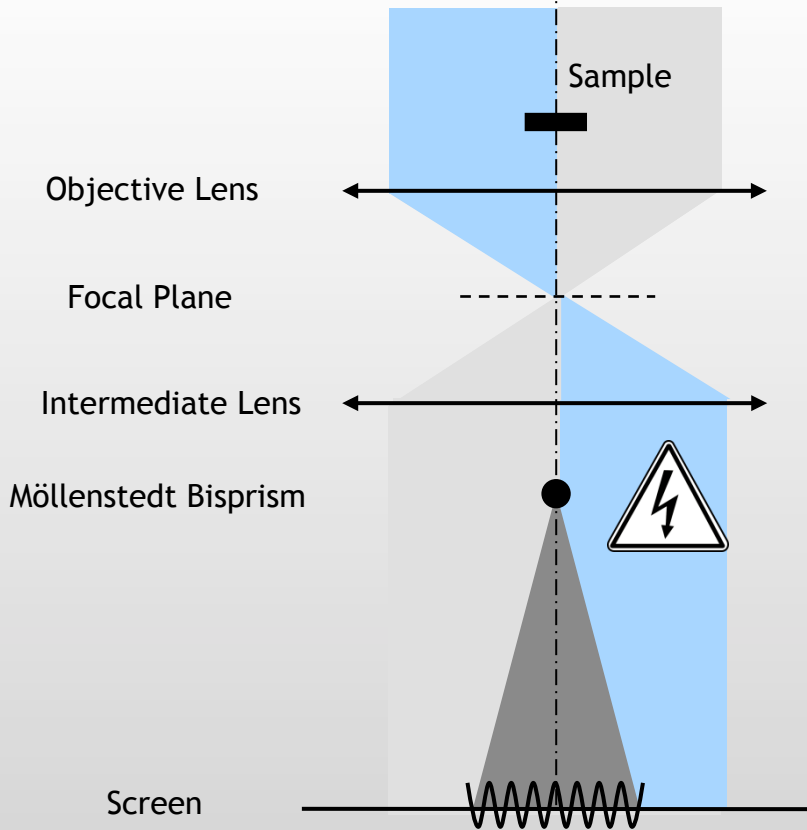
2- Solve TIE

$$\varphi(r) = -\frac{\pi}{\lambda \Delta_z} \nabla^{-2} \nabla \cdot \left[\frac{1}{I(r, 0)} \nabla \left\{ \nabla^{-2} \left[\underline{I(r, \Delta_z)} - \underline{I(r, -\Delta_z)} \right] \right\} \right]$$

In-plane
phase shift
image



2. Off-axis Electron Holography



$$\psi_0 = \exp(i\mathbf{k}\cdot\mathbf{r})$$

$$\psi(\mathbf{r}) = A(\mathbf{r}) \exp i((\mathbf{k}\cdot\mathbf{r}) + \varphi(r))$$

$$\Psi(\mathbf{r}) = A_s(\mathbf{r}) \exp i((\mathbf{k}\cdot\mathbf{r}) + \varphi_s(r))$$

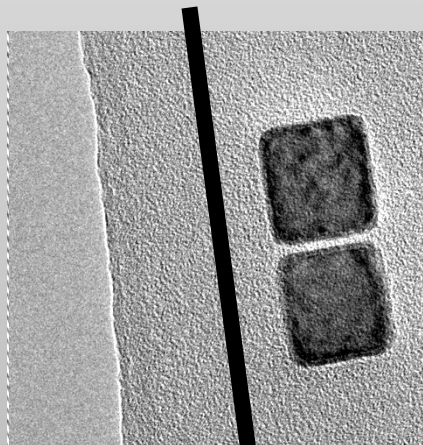
Incident beam



After the sample



After the objective lens



$$\begin{aligned} I_{Holo} &= |\Psi_0 + \Psi^*|^2 + \text{background} \\ &= 1 + A_s^2(x, y) \\ &\quad + 2A_s(x, y) \cdot \cos(2\pi R_0 \cdot x + \varphi_s(x, y)) \\ &\quad + I_{Inelast}(x, y) \end{aligned}$$

Phase retrieval

$$\Psi(\mathbf{r}) = A_s(\mathbf{r}) \exp i((\mathbf{k} \cdot \mathbf{r}) + \phi_s(r))$$

Intensity image (hologram)

$$I_{Holo} = |\Psi_0 + \Psi^*|^2 = 1 + A_s^2(x, y) + 2A_s(x, y) \cdot \cos(2\pi R_0 \cdot x + \phi_s(x, y))$$

FFT of $I_{hol}(\mathbf{r})$

$$FT|I_{hol}(\mathbf{r})| = \delta(u) + FT[A^2(\mathbf{r})] + FT[A_s(\mathbf{r}) \exp(i\phi_s(\mathbf{r}))] \otimes \delta\left(\mathbf{u} + \frac{\alpha_h}{\lambda} \hat{\mathbf{x}}\right) + FT[A_s(\mathbf{r}) \exp(-i\phi_s(\mathbf{r}))] \otimes \delta\left(\mathbf{u} - \frac{\alpha_h}{\lambda} \hat{\mathbf{x}}\right)$$

Sideband

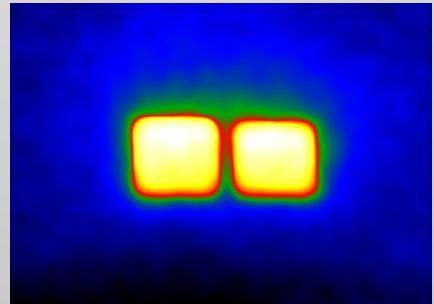
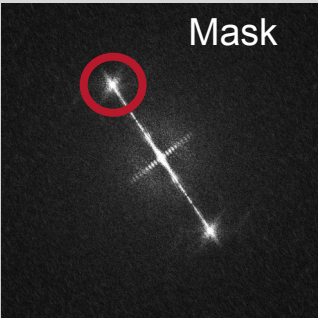
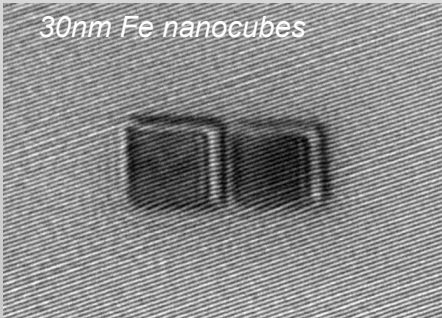
1- Calculate the Fast Fourier Transform (FFT) of the digitally recorded hologram $FT|I_{hol}(\mathbf{r})|$.

FFT

2- Apply a mask to select and center one of the sidebands

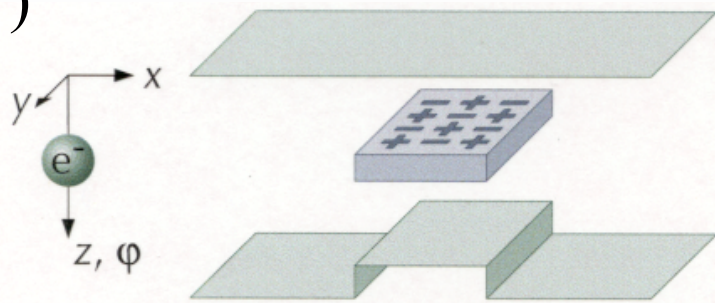
FFT⁻¹

3- Calculate FFT⁻¹ to obtain the amplitude and phase images.



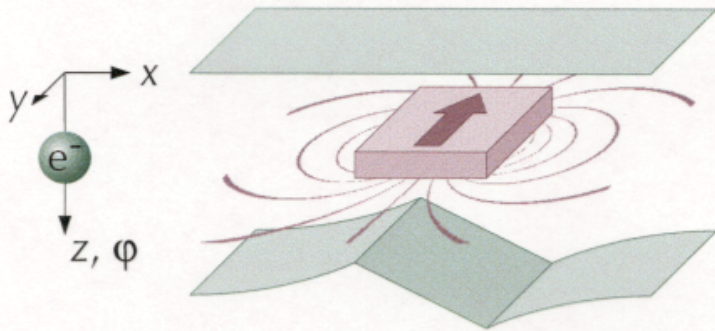
Amplitude: $A_s(\mathbf{r})$

Phase: $\phi_s(\vec{r})$

$\phi_s(\vec{r})$ 

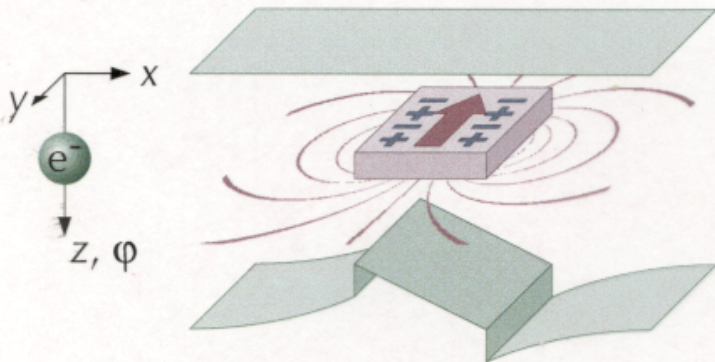
Phase shift induced by the local electrostatic potential:

$$\phi_{Elec}(x) = C_E \int V(x, y, z). dz$$



Phase shift induced by the local magnetic field :

$$\phi_{Mag}(x) = -\frac{e}{\hbar} \iint B_{\perp}(x, z). dx. dz$$



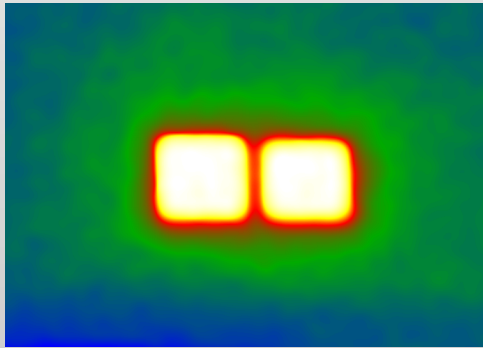
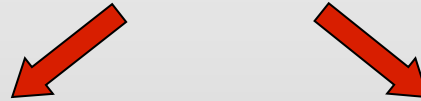
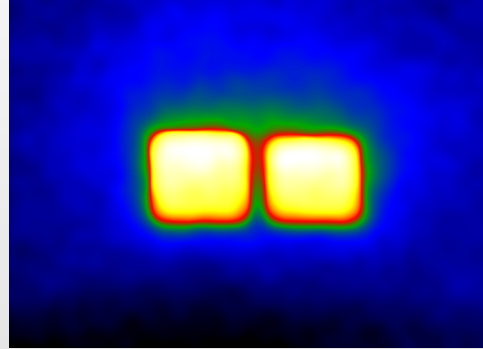
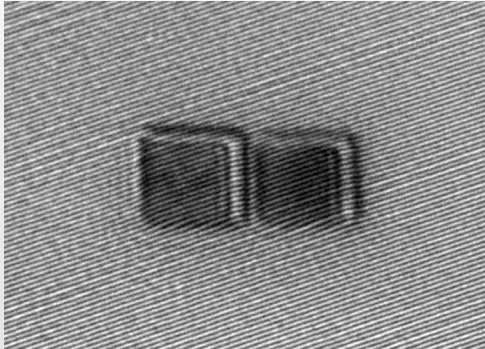
Total phase shift

$$\phi_{Total}(x) = \phi_{Elec}(x) + \phi_{Mag}(x)$$

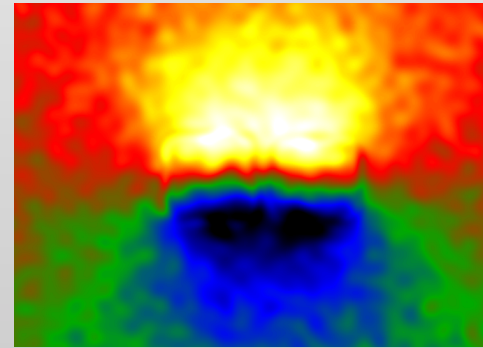
But phase image $\phi(x, y) = \phi^C(x, y) + \phi^M(x, y)$

→ Need to separate different contributions to the phase shift

$$\phi(x, y) = \phi^C(x, y) + \phi^M(x, y)$$



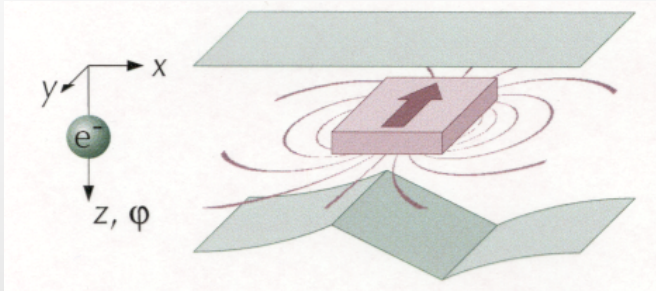
$$\phi_{C, Elec}(x, y) = C_E \int V(x, y, z) dz$$



$$\phi_{Mag}(x, y) = - \frac{e}{\hbar} \iint B_{\perp}(\vec{r}_{\perp}, z) dr dz$$

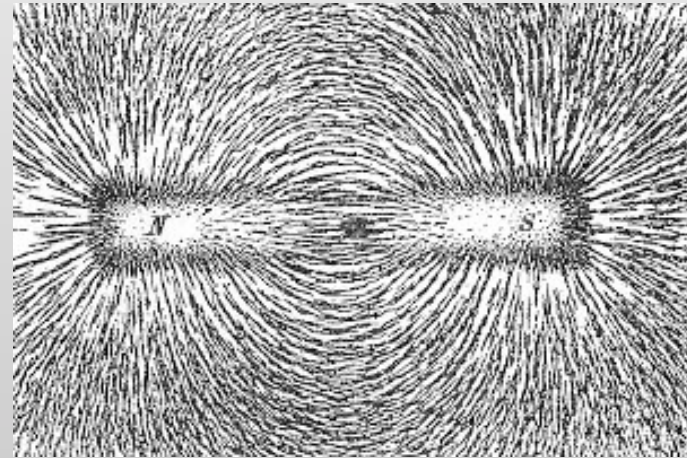
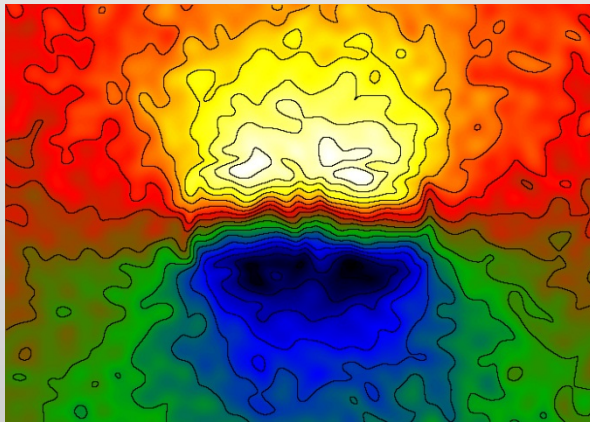
Typical phase sensitivity : few 10 mrad @ 5 nm spatial resolution

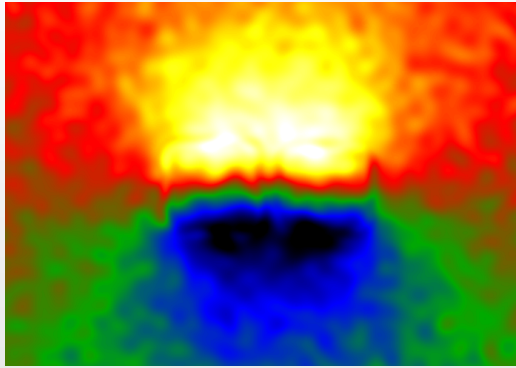
Measurements of the magnetic induction (\mathbf{B}) components from $\phi_{Mag}(\vec{r})$



$$\phi_{Mag}(x) = -\frac{e}{\hbar} \iint B_{\perp}(x,z).dx.dz$$

- The equiphase contours give the B_{\perp} direction \rightarrow contours lines = in-plane magnetic flux





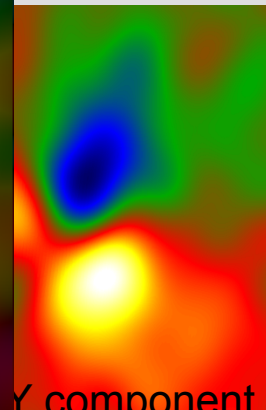
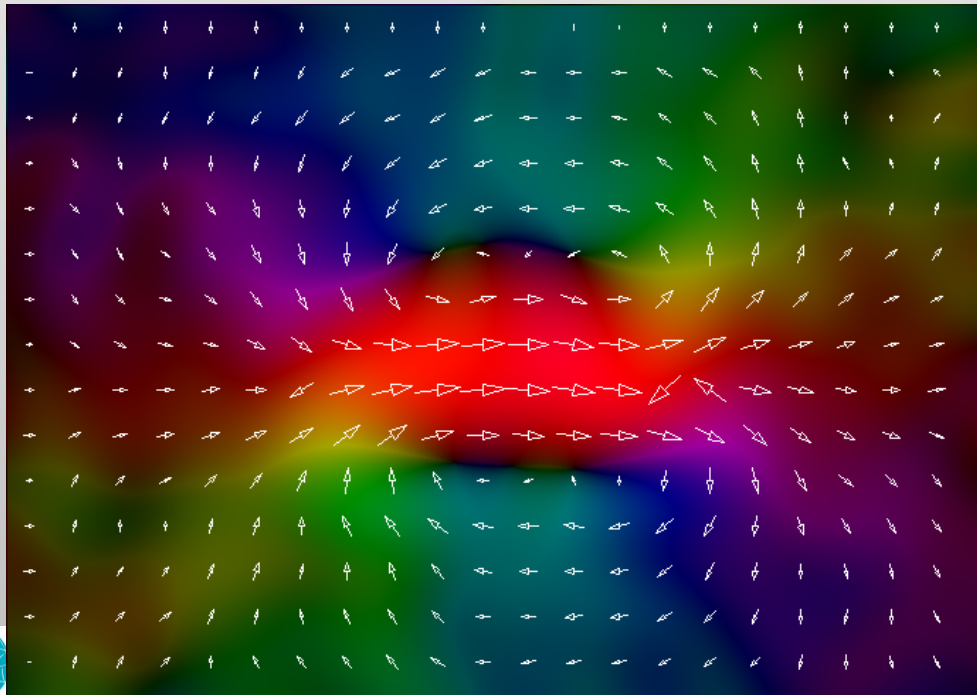
- The phase gradient is proportionnal to the in-plane B_{\perp} components of the induction

→ B_x, B_y components

$$\frac{\partial \phi_{Mag}(y)}{\partial y} = \frac{e}{\hbar} B_x(y).t$$

(t = thickness)

$$\frac{\partial \phi_{Mag}(x)}{\partial x} = \frac{e}{\hbar} B_y(x).t$$



Y component

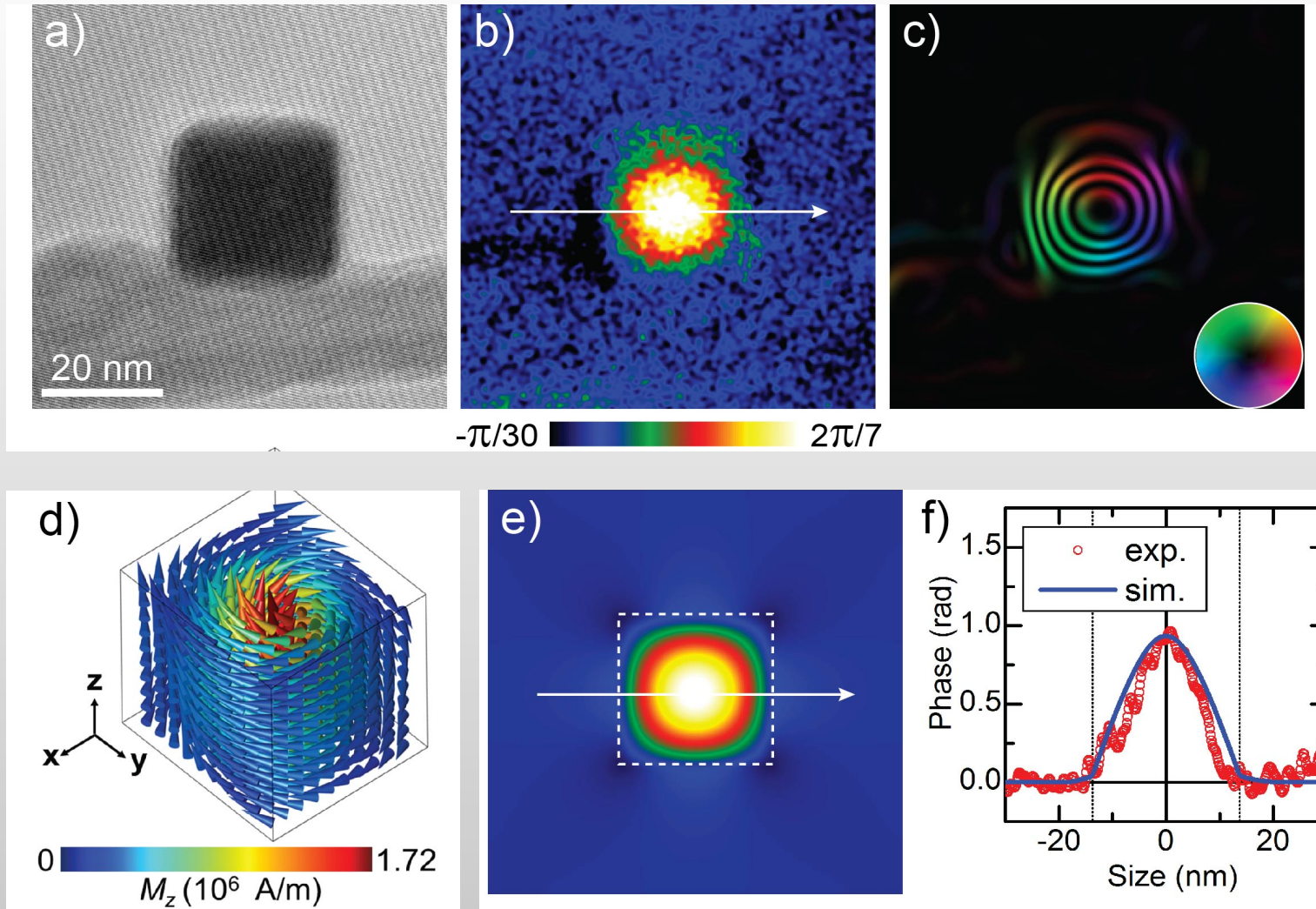


- Only planar components of the induction can be measured

Outlook

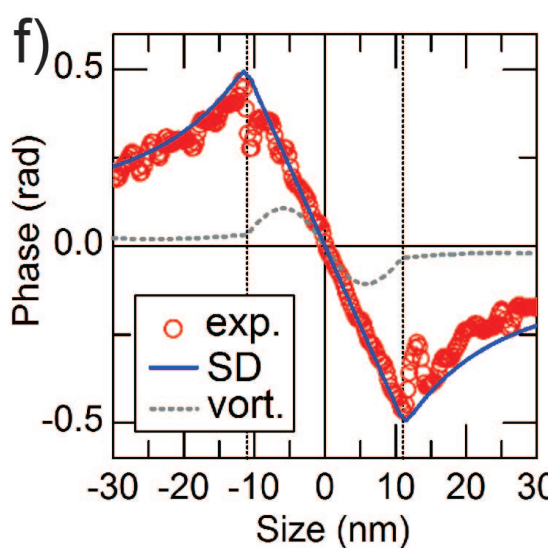
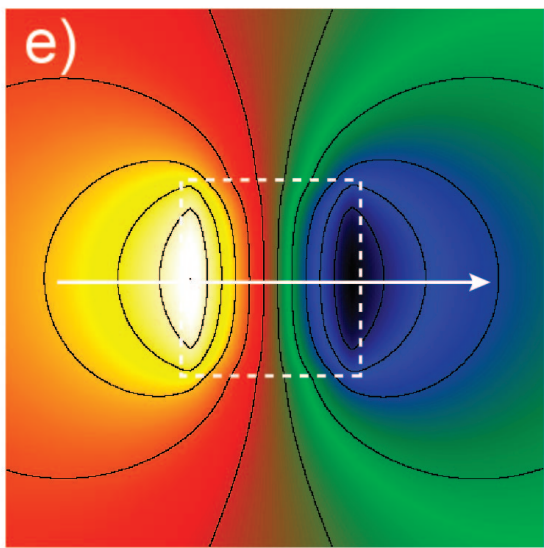
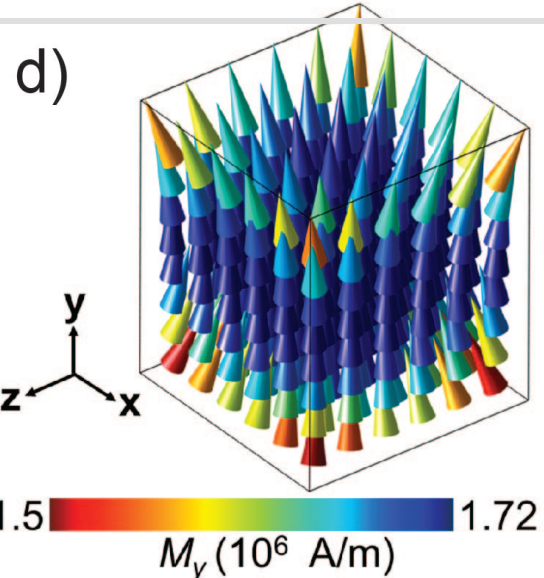
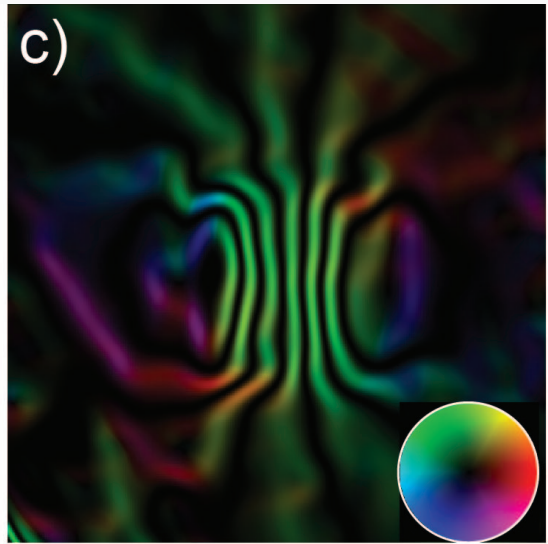
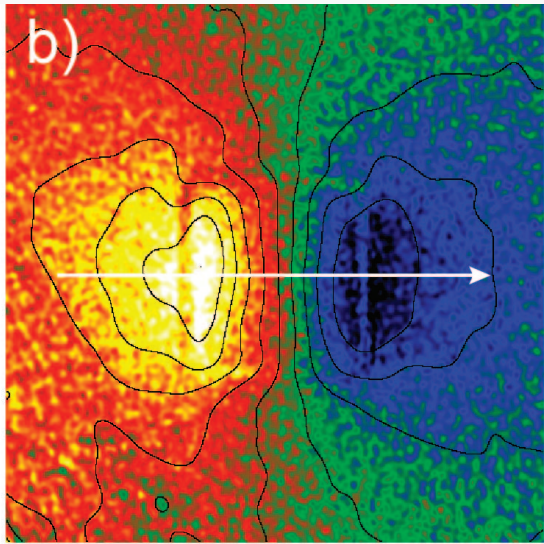
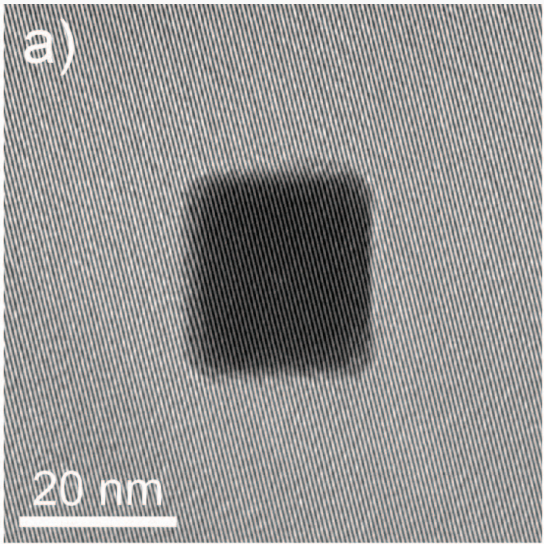
- ◆ Lorentz TEM and Electron Holography
- ◆ Exple1 : Study of the phase diagram of Fe nanocubes by EH
- ◆ Exple 2 : *In-situ* Lorentz studies of the magnetic reversal of Co antidot arrays
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<001> Vortex configuration of a single isolated 29 nm nanocube

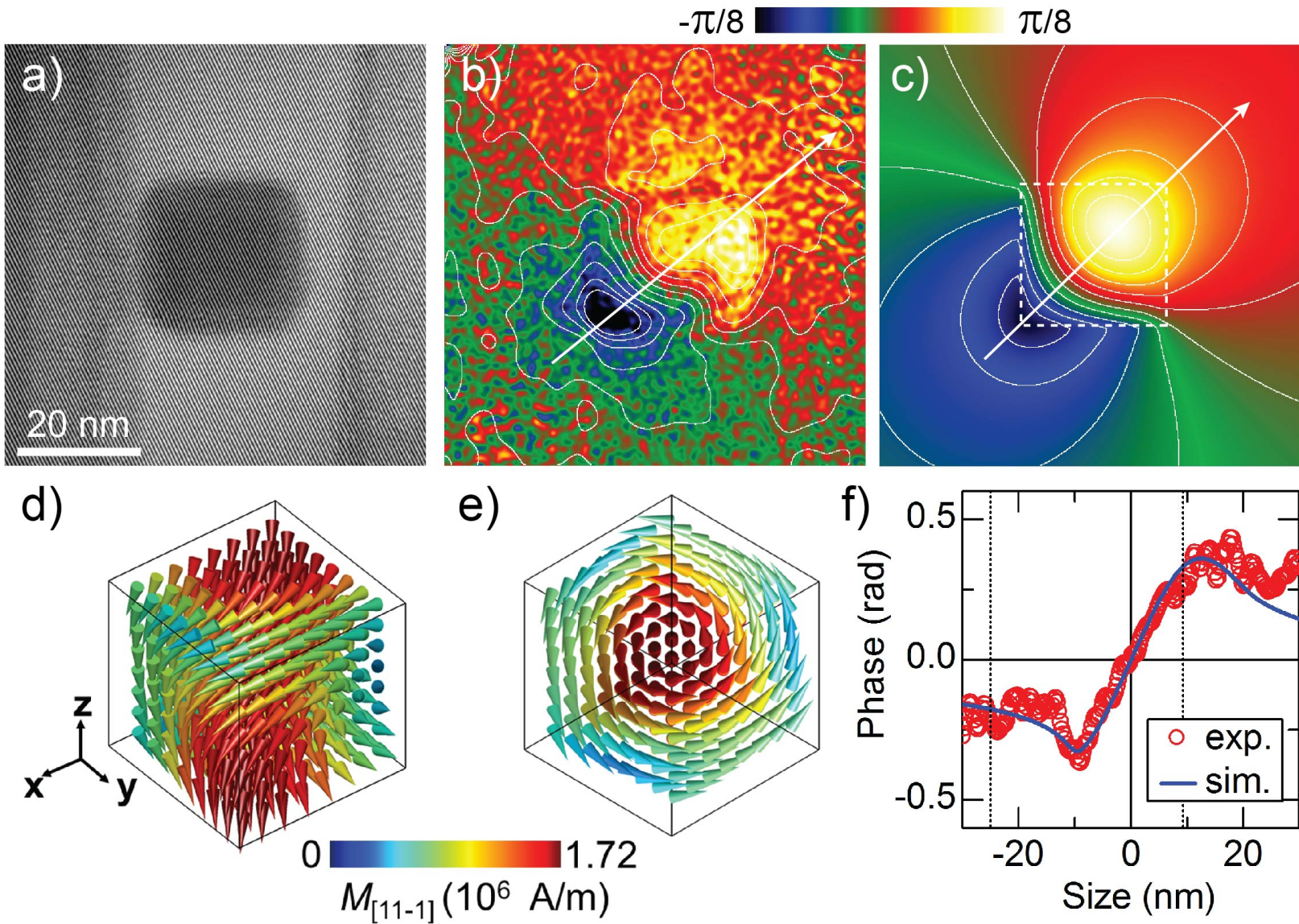


a) Hologram b) Phase image corresponding to the magnetic phase shift, c) Contour map, d) Micromagnetic simulation (with bulk M_S) of the in-plane induction (OOMMF), e) simulated phase image f) phase profiles

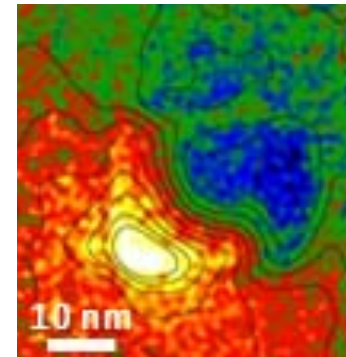
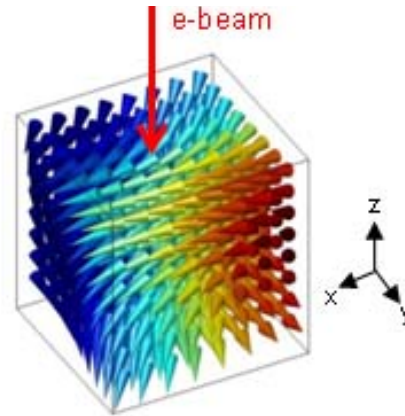
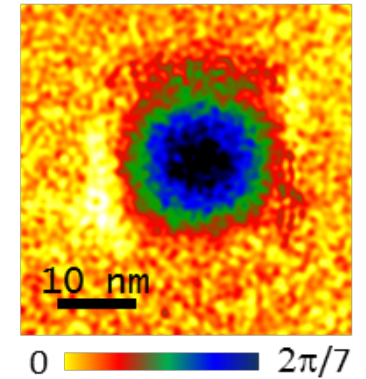
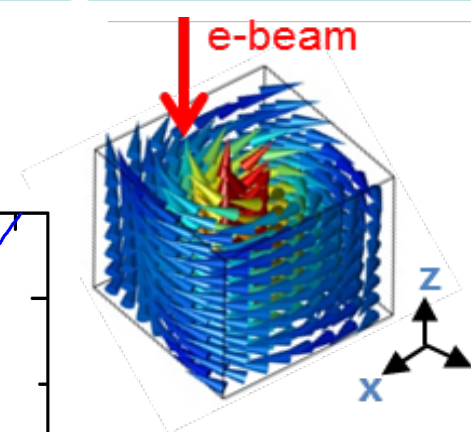
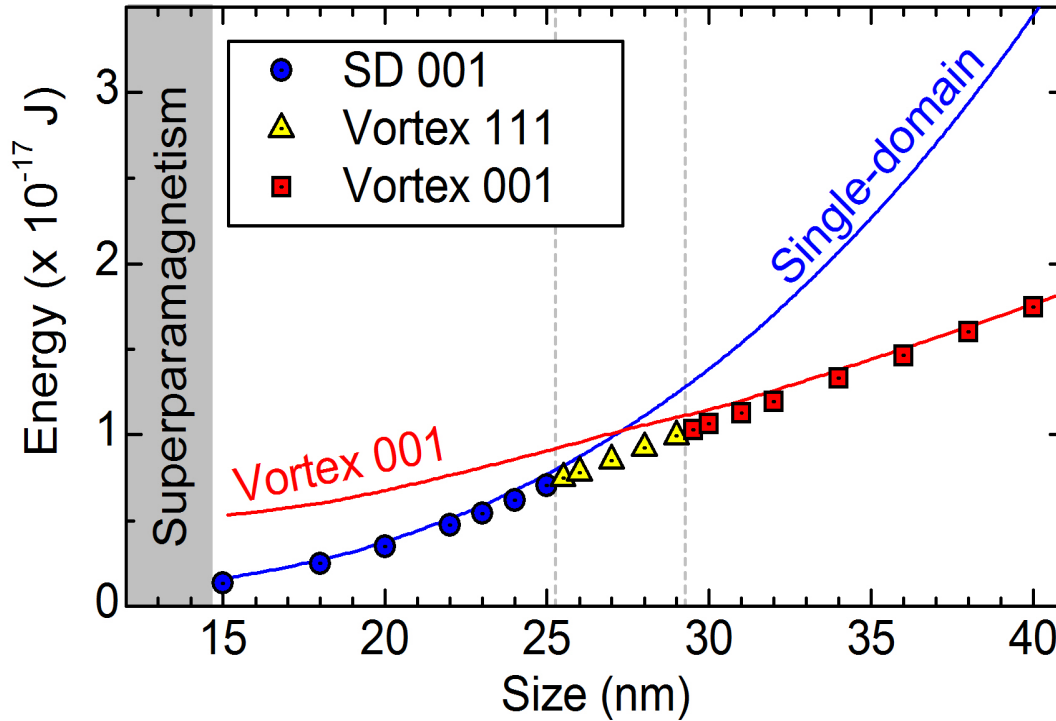
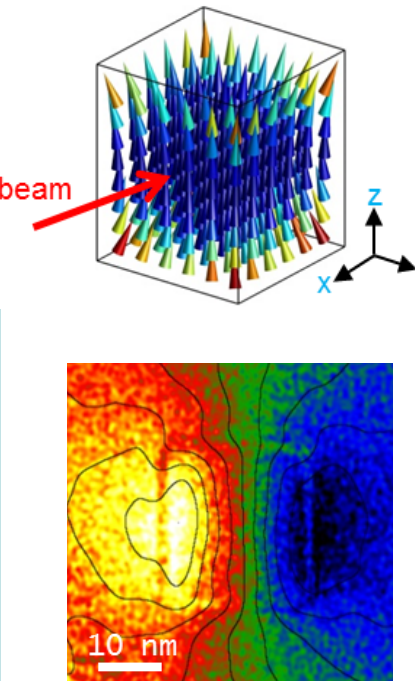
Single domain configuration of a single isolated 25 nm nanocube



<111> Vortex configuration of a single isolated 27 nm nanocube



Phase diagram (considering a 2 nm thick oxide shell layer)



Coll. T. Blon, F. Bonilla, LM Lacroix
INSA-LPCNO (Toulouse)



See talk of
Francisco Bonilla
Fr.E.3-04 @18:00

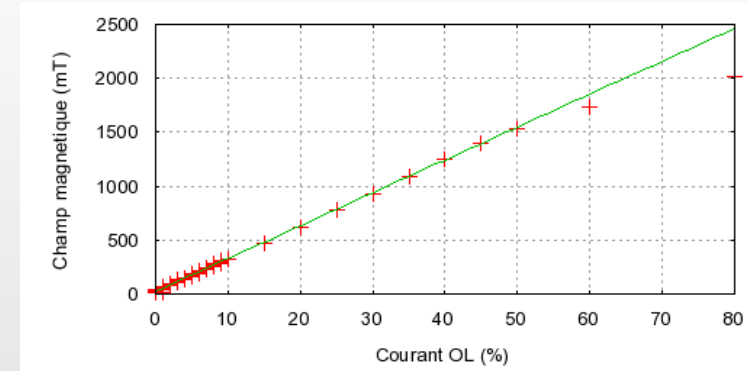
Outlook

- ◆ Lorentz TEM and Electron Holography
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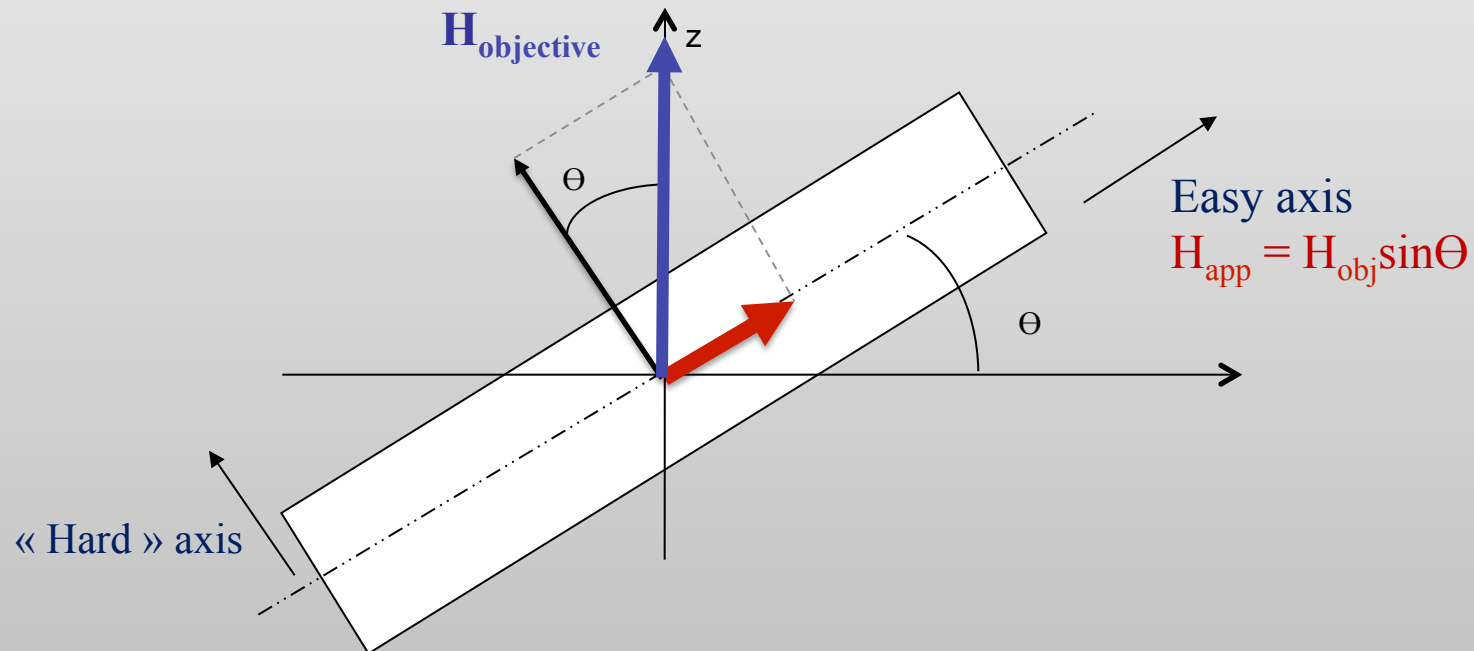
In-situ Lorentz experiments under external applied magnetic field ?

→ Use the magnetic field of the objective lens as external field

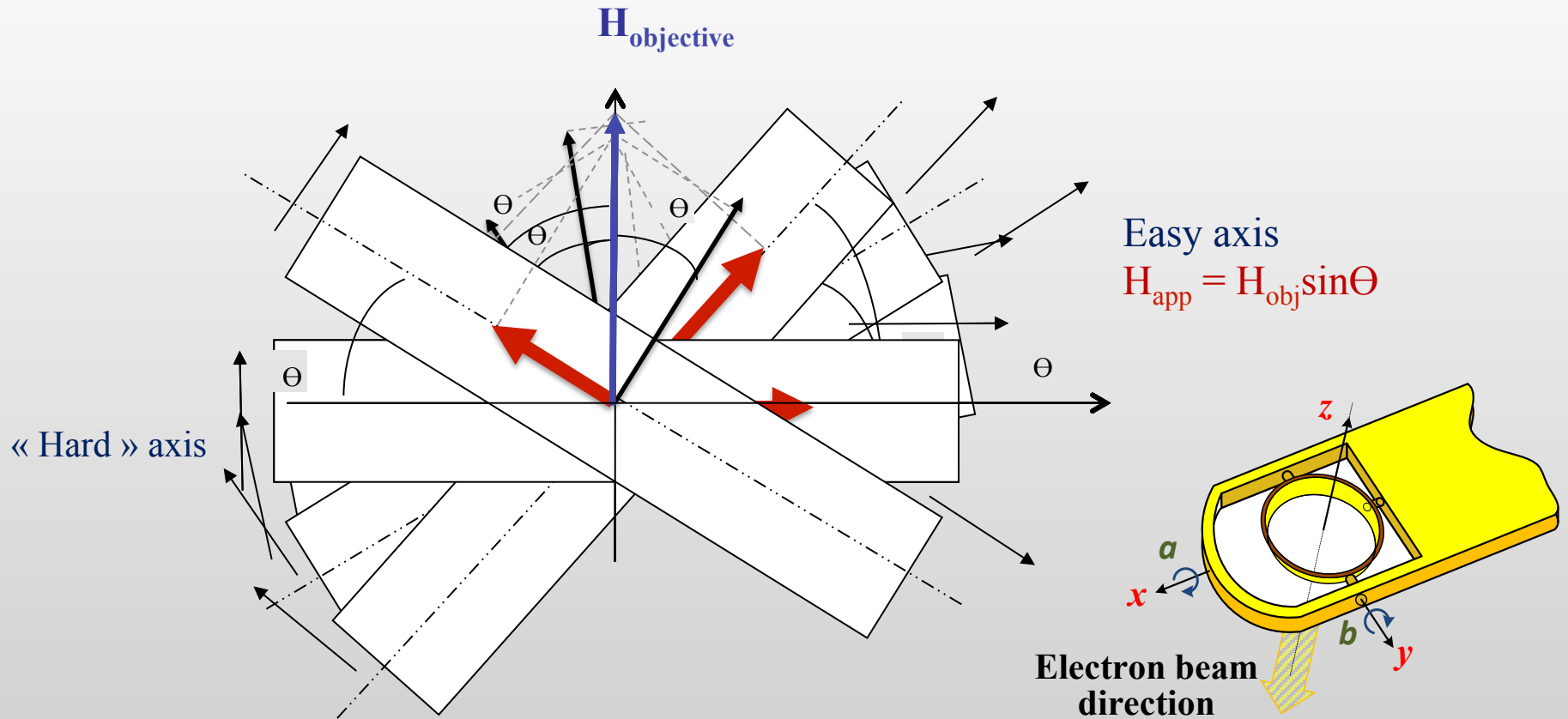
$H_{\max} = 2.1 \text{ Tesla @} 200 \text{ kV}, 3 \text{ Tesla @} 300\text{kV}$



→ Tilting the sample between to apply a magnetic in the sample plane



In-situ Lorentz experiments under external applied magnetic field



Using a rotation lineal transformation, we can control the in-plane magnetic field both in intensity and direction

L. A. Rodriguez et. al. *Ultramicroscopy* 134 (2013) 144

$$H_{x'} = -H \cos(\alpha) \sin(\beta)$$

$$H_{y'} = H \sin(\alpha)$$

$$H_{z'} = -H \cos(\alpha) \cos(\beta)$$

In-situ experiments under external applied magnetic field

MAGNETIC ANTIDOT ARRAYS

Regular antidot arrays in continuous magnetic thin films leads to modifications of :

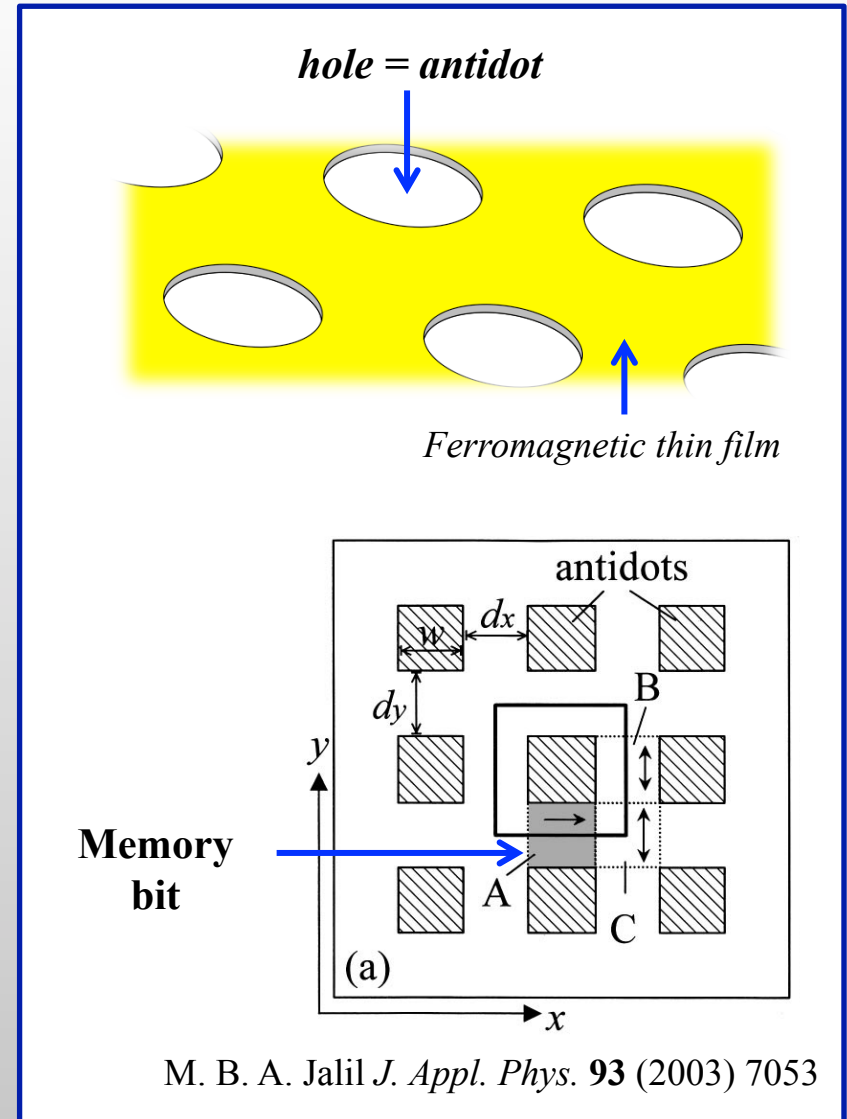
- *Coercivity*
- *Magnetoresistance*
- *Remanence*

→ potential to form individual magnetic entities that could be used as **bits of information up to 10 Gbits/cm²**

Motivation: Exploring the magnetization confinements effect on magnetic domain structures and switching processes at the nanometer scale

→ Arrays of Co antidots have been fabricated on Si₃N₄ membranes (sputtering + FIB etching):

- Thickness = 10 nm
- Hole diameter = 55 nm



* Z.L. Xiao et. al., *Nanotech* **3** (2003) 357

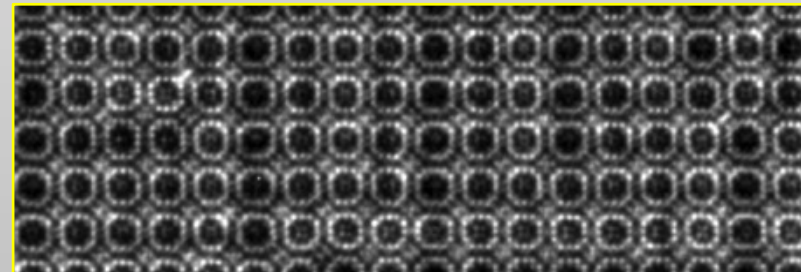
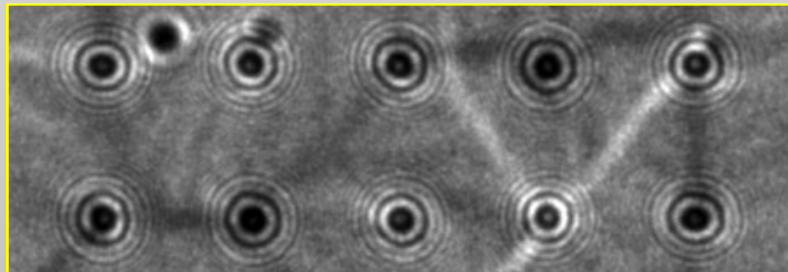
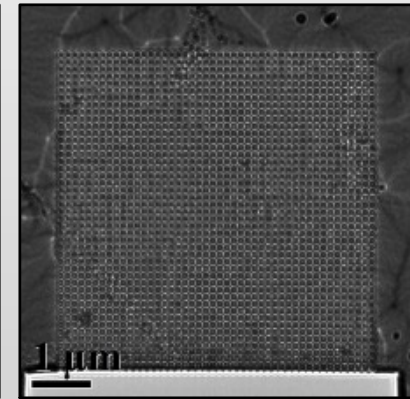
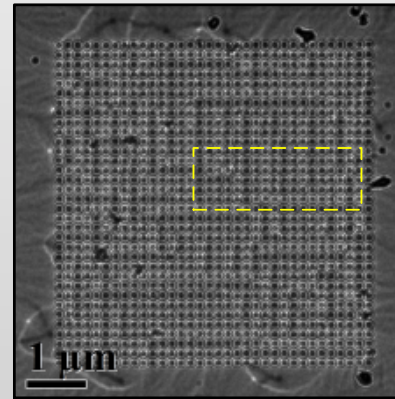
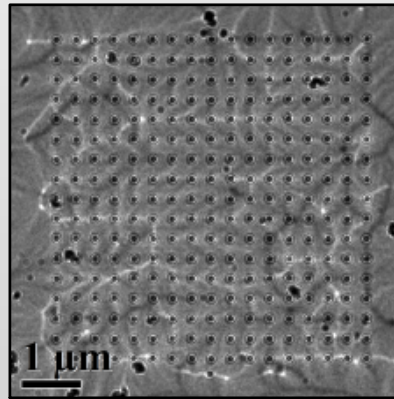
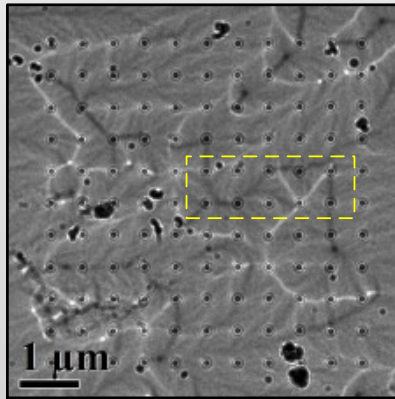
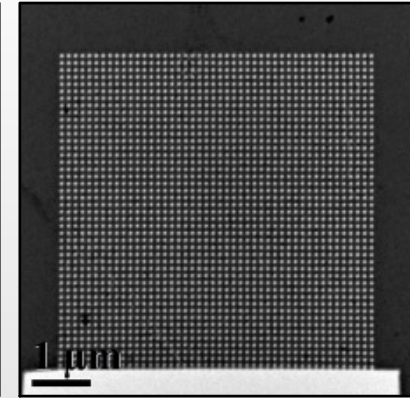
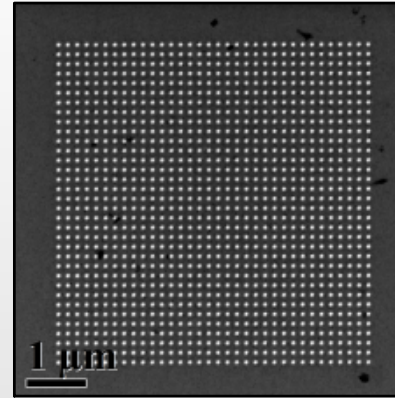
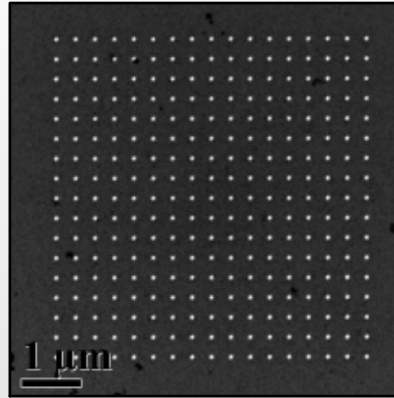
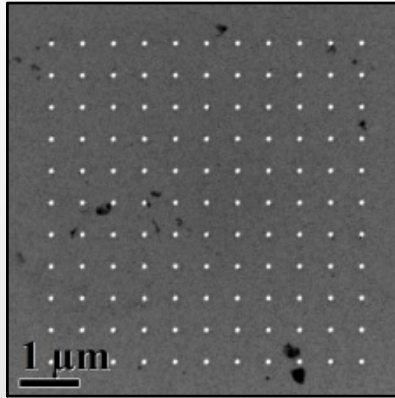
At remanence, Lorentz images show lines of bright and dark contrast → **DOMAIN WALLS**

$p = 524 \text{ nm}$

327 nm

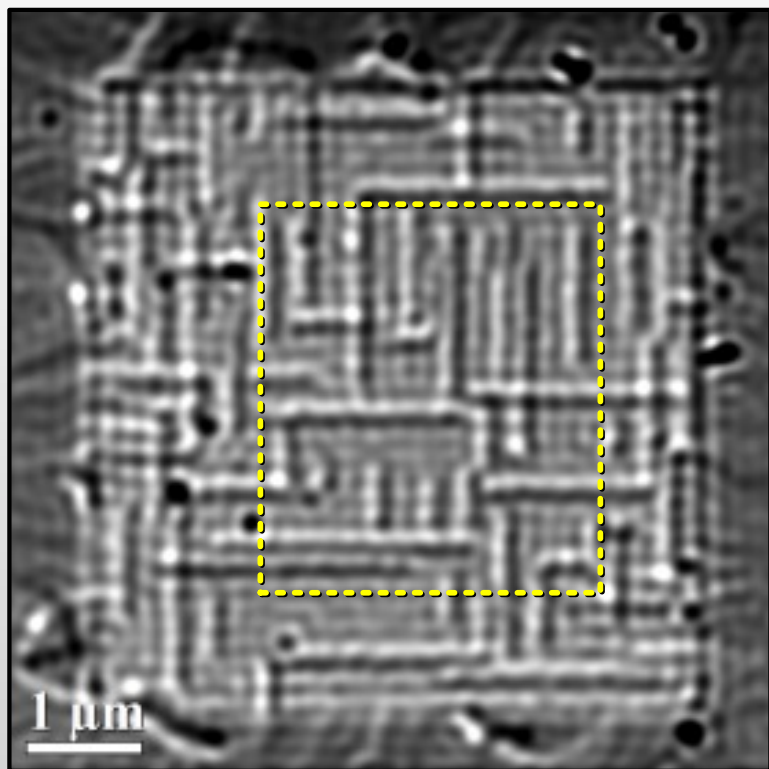
160 nm

116 nm

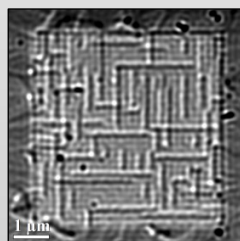
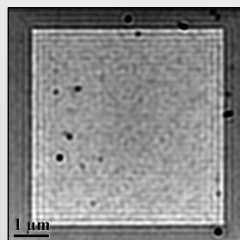
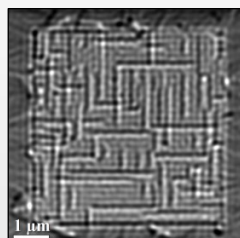


Fresnel Fringes in the defocused images

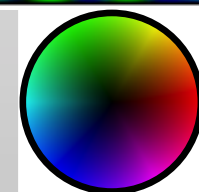
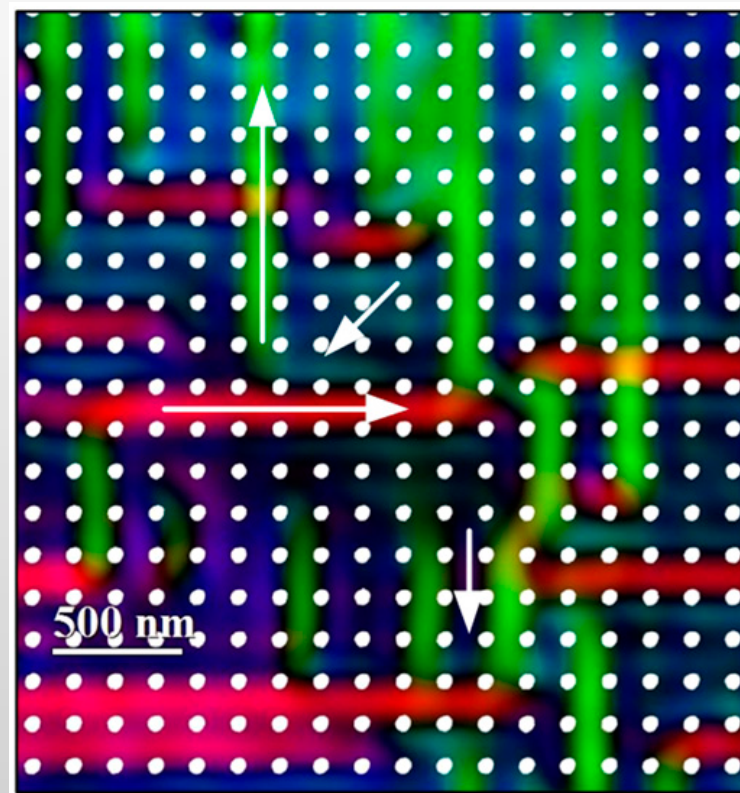
Filtered LM image



Magnetic domain stripes

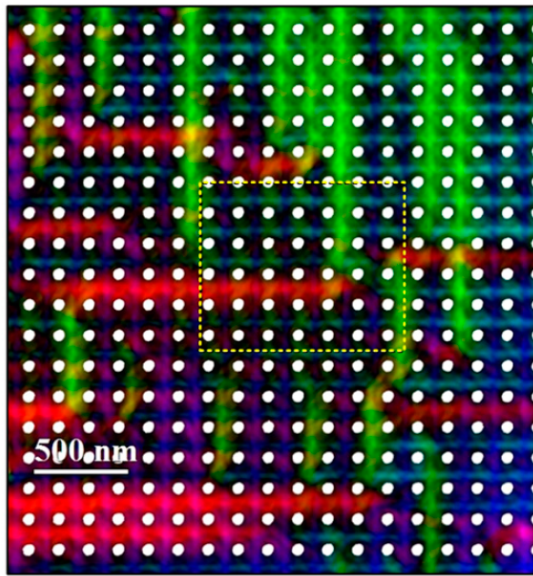


Magnetization map

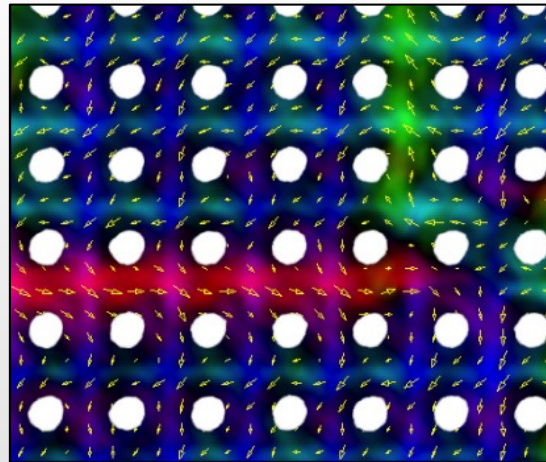


Magnetization
Color scale

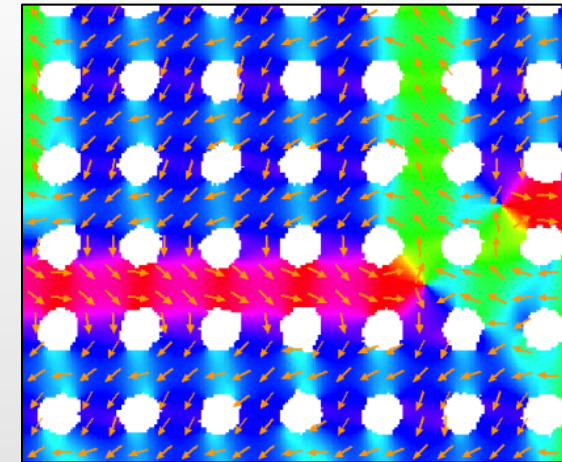
Vector map of the magnetization



Experimental (TIE-Reconstruction)

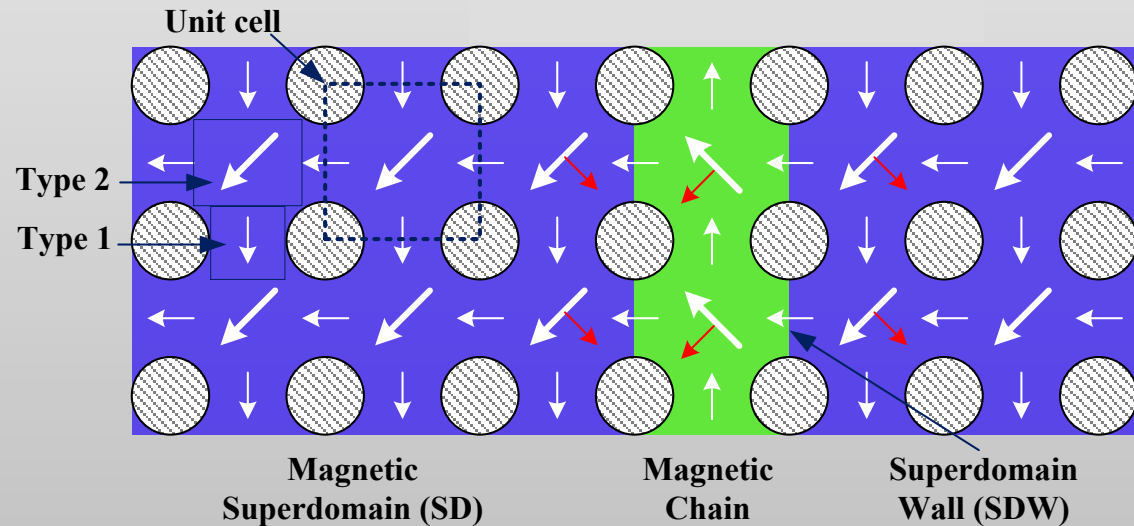


Simulation



Shape anisotropy tends to align the magnetization:

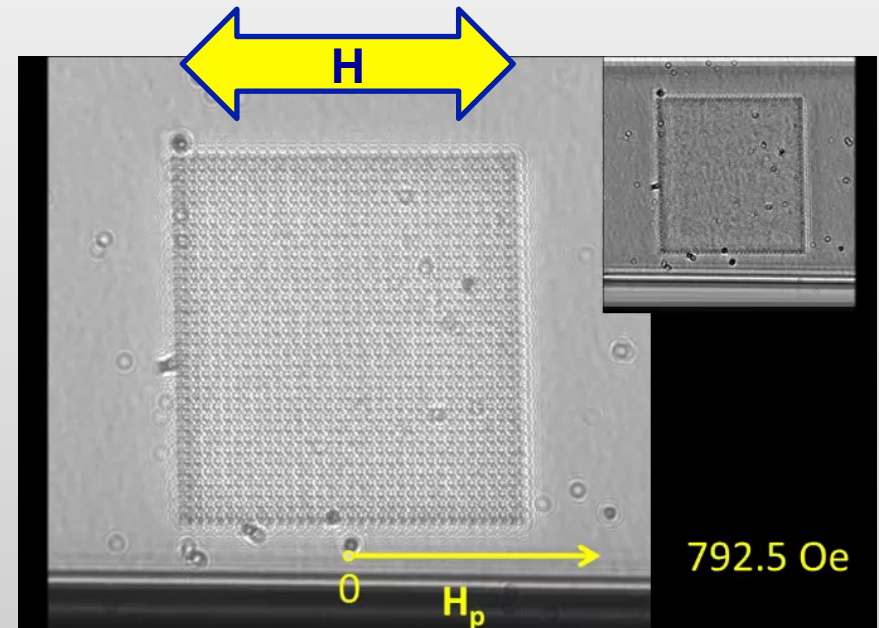
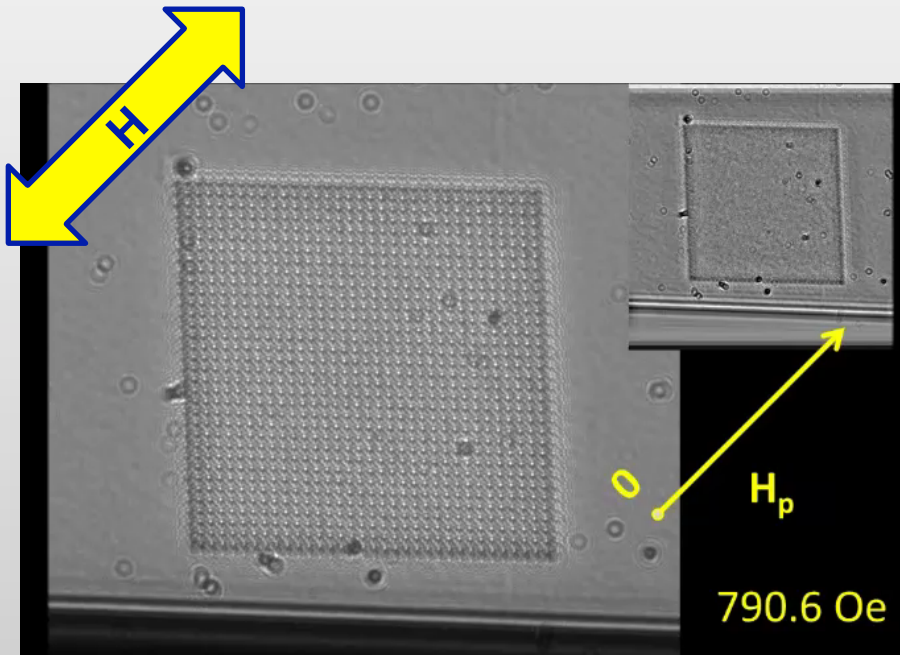
- 1) Parallel to the edge between two holes.
- 2) Transverse to the lattice between four holes.



In-situ Lorentz experiments under external applied magnetic field

Magnetization reversal processes

By *Nucleation (N)* and *Propagation (P)* of Superdomain Walls (SDWs)



Simultaneous N and P of
Vertical and Horizontal SDWs

- 2 steps
(1) N and P of Horizontal SDWs
(2) N and P of Vertical SDWs

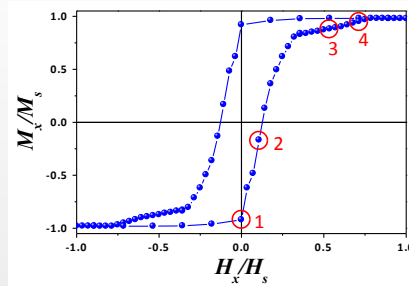
Magnetization reversal processes

Parallel Magnetic field

Magnetization reversal process occurs by **nucleation**

and **propagation** of superdomains:

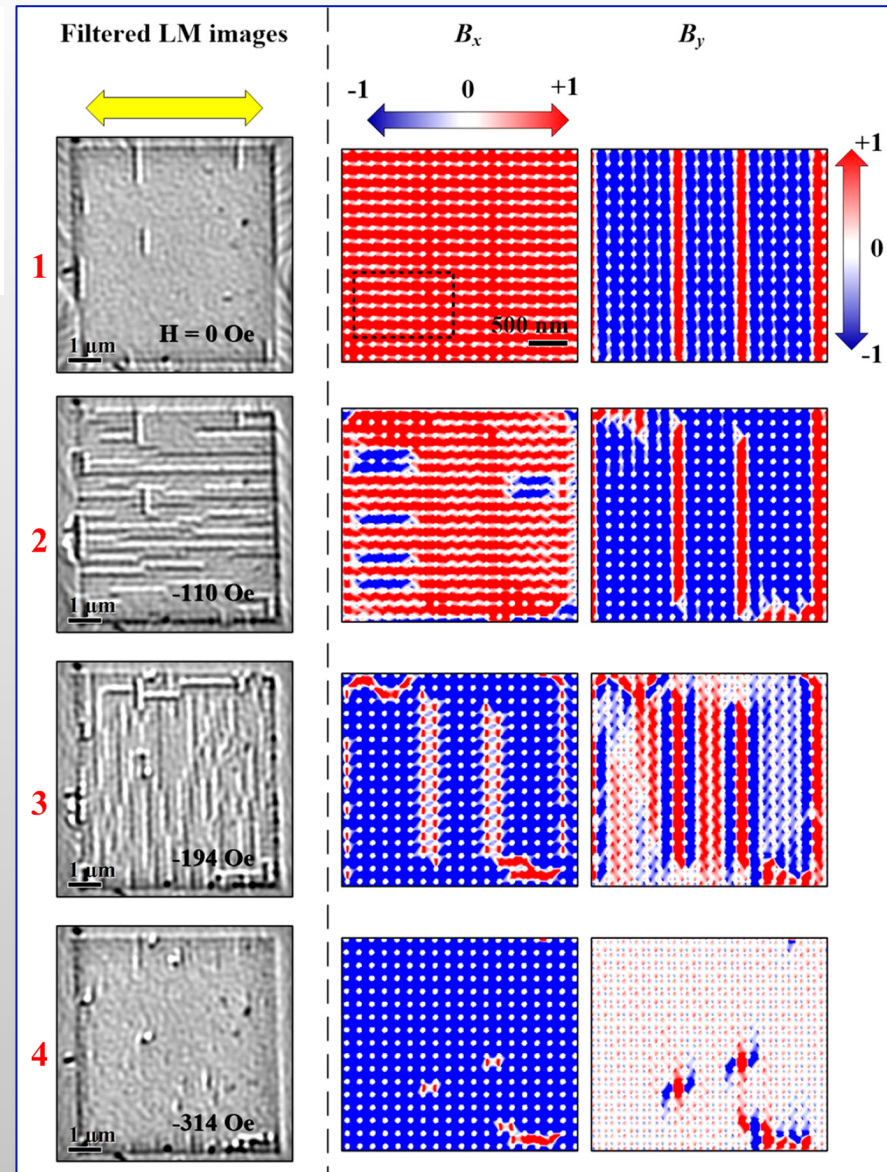
1. For $H = 0$, some vertical superdomains are nucleated
2. Magnetization switching by horizontal superdomains
3. Magnetization switching by vertical superdomains
4. Small magnetic domains just before the saturation



Transverse Magnetic field:

Simultaneous **nucleation** and **propagation** of horizontal and vertical SDWs.

L. A. Rodriguez et. al. *Nanotechnology* **23** (2014) 385703



Outlook

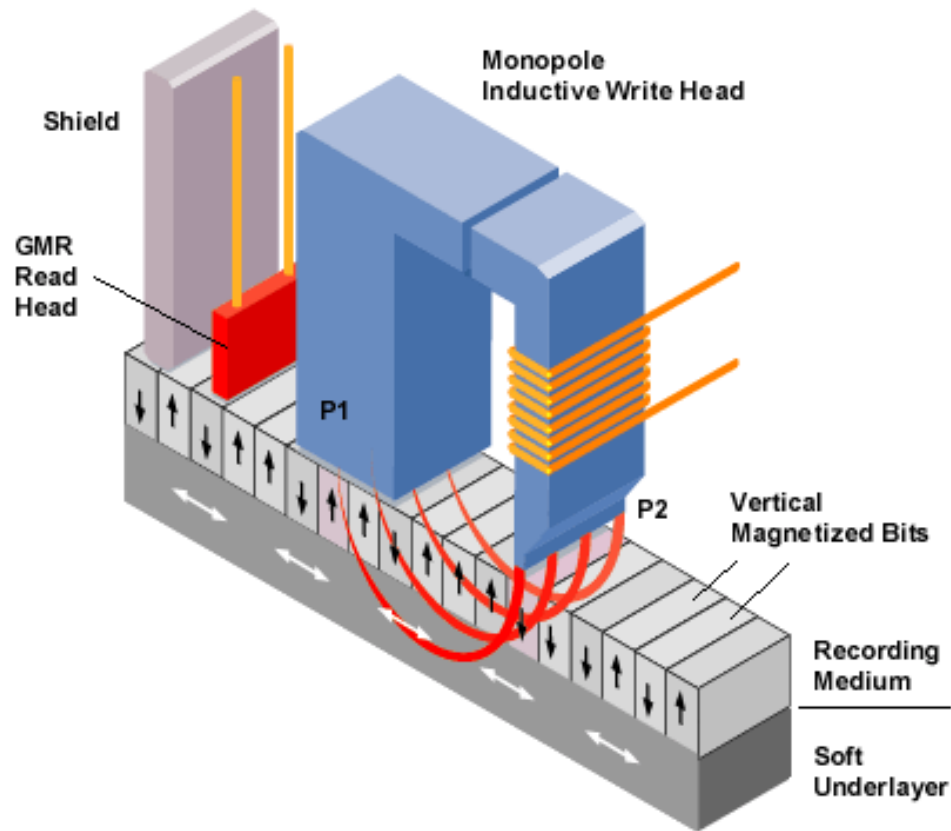
- ◆ Lorentz TEM and Electron Holography
- ◆ Exple1 : Study of the phase diagram of Fe nanocubes by EH
- ◆ Exple 2 : *In-situ* Lorentz studies of the magnetic reversal of Co antidot arrays
- ◆ Exple 3 : *In-situ* EH studies of the magnetic induction generated by a HDD writer-head *in operando*

In-situ EH experiments of a writer head in operando

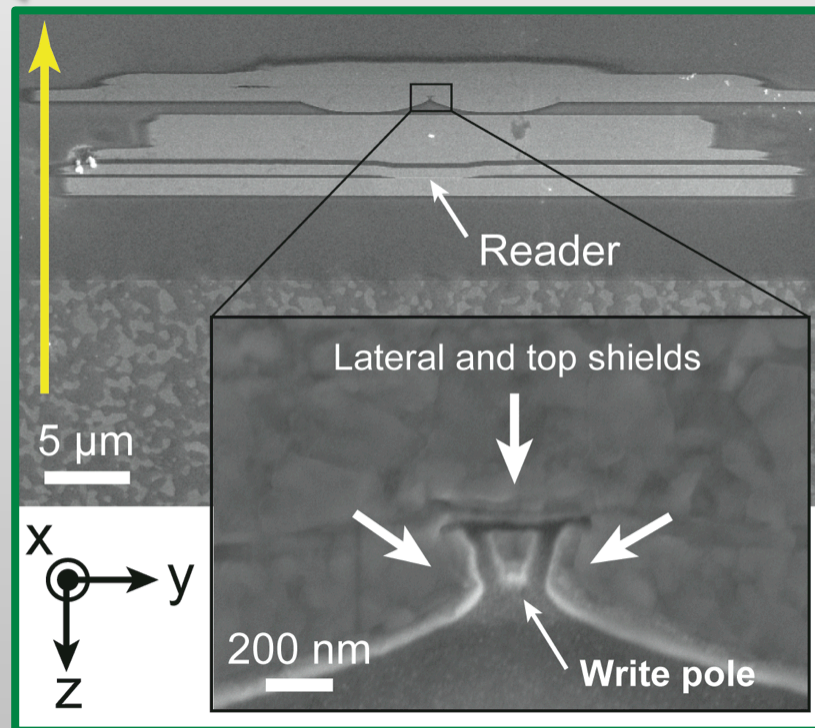
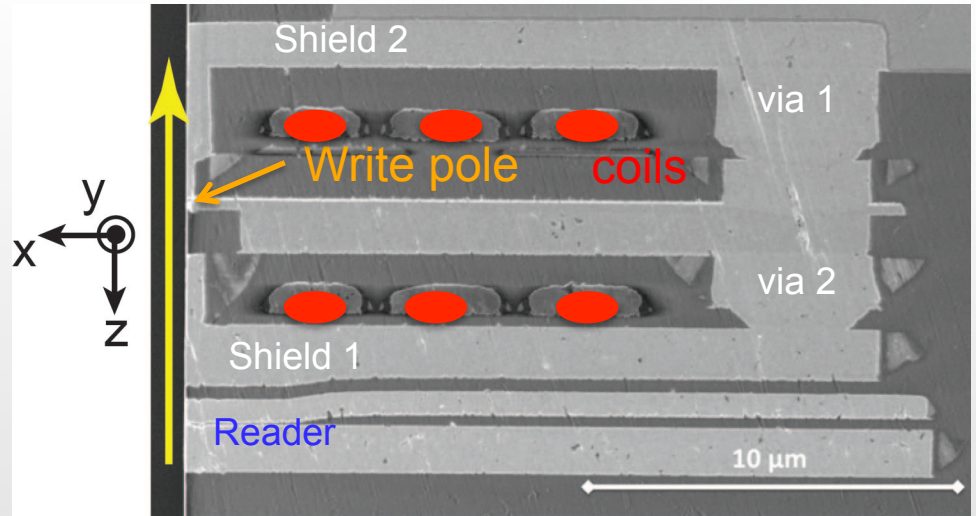
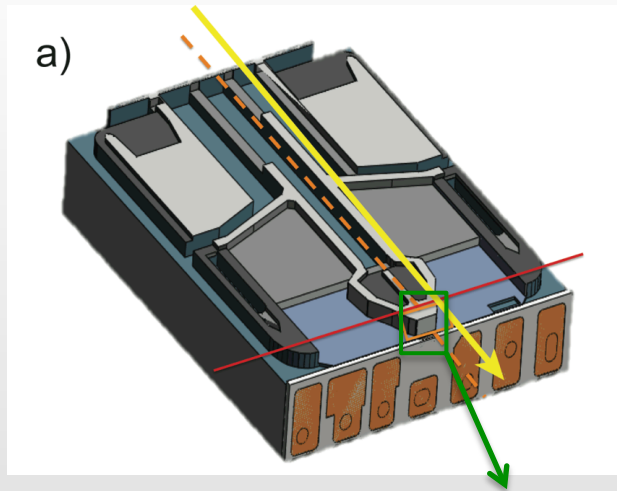


HDD Write Pole for Perpendicular Recording

From Computer Desktop Encyclopedia
© 2006 The Computer Language Company Inc.

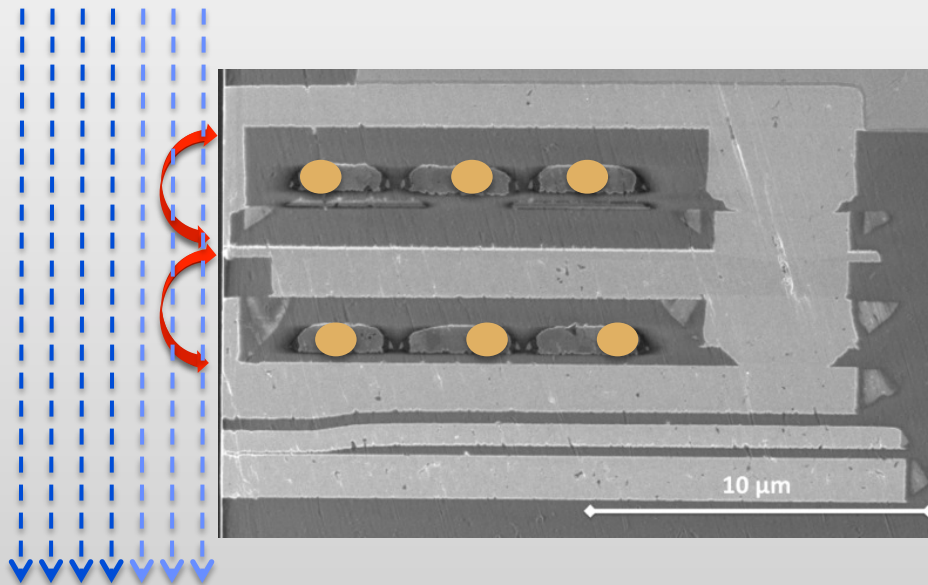


Recording head technology

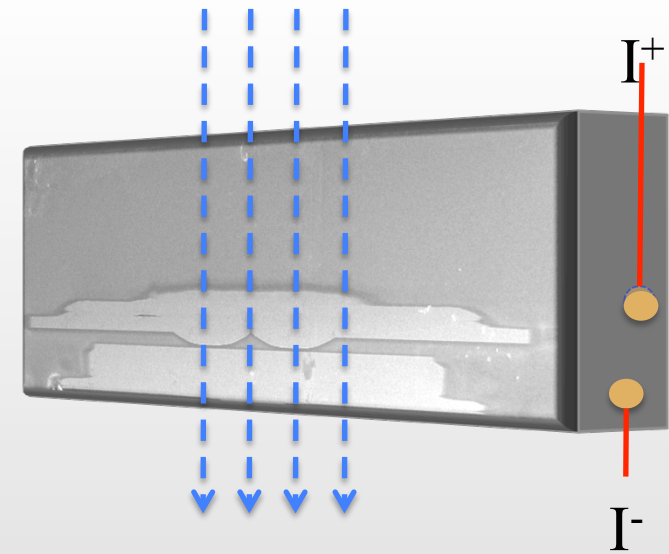


Holography experimental set-up

Electron beam

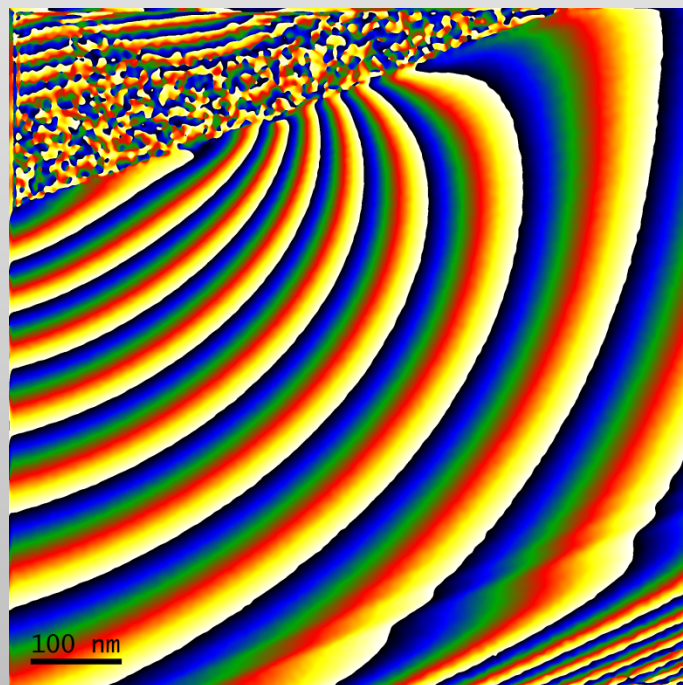
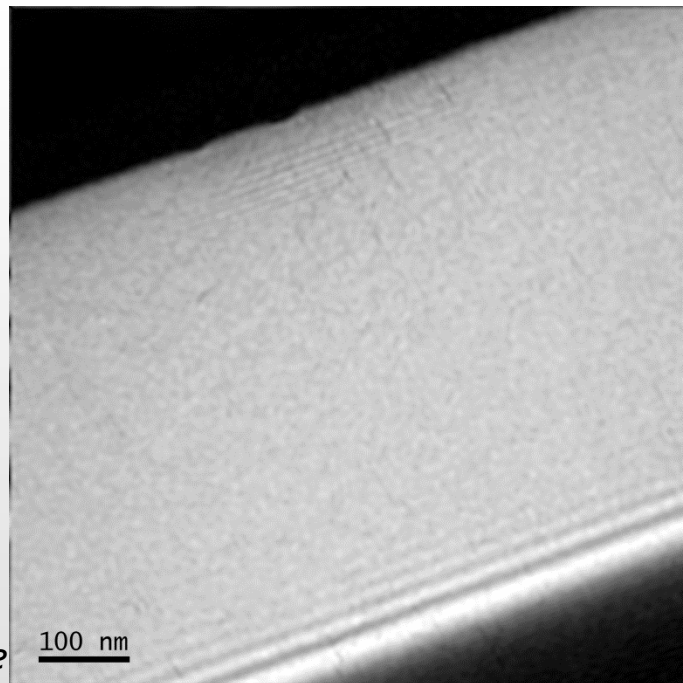
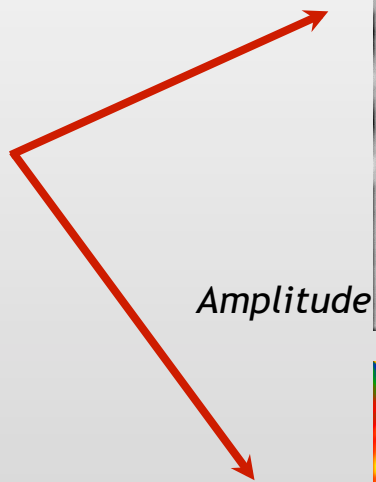
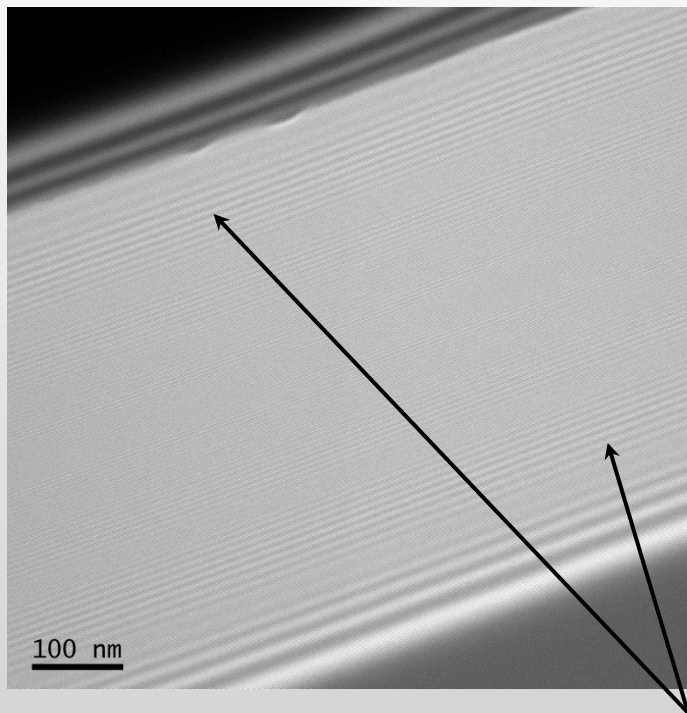


Electron beam



→ Electron beam phase shift is measured in front of the write pole while applying a current through the device.

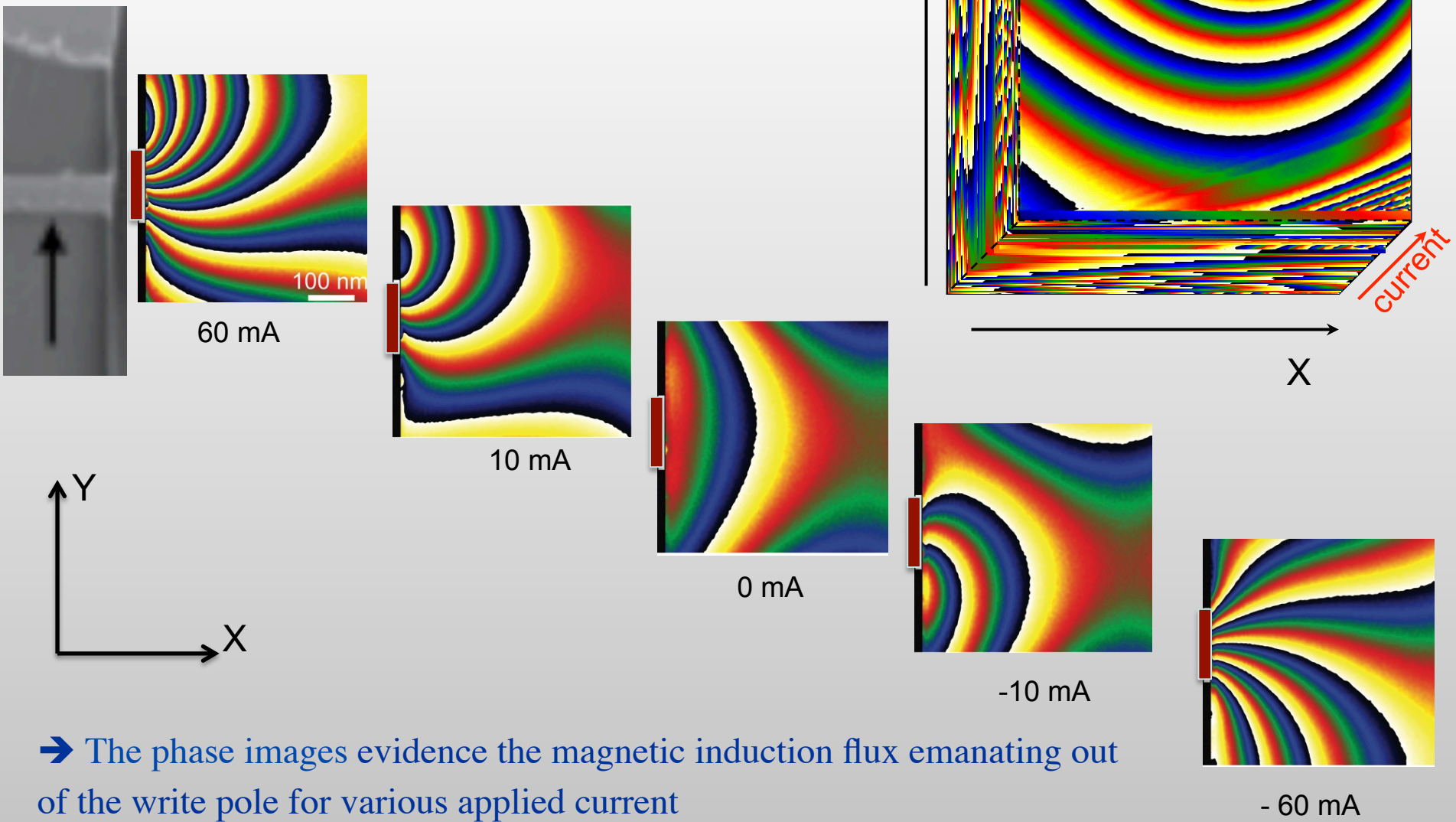
Hologram (dataset, -60 / +60 mA)



- For all holograms (88) remove Fresnel fringes
- Correct for camera distortion
- Extraction of amplitude and phase images (8 nm resolution)

Phase

$$\phi_{Mag}(\mathbf{r}) = -\frac{e}{\hbar} \iint B_{\perp}(\mathbf{r}_{\perp}, z) dr dz$$



→ The phase images evidence the magnetic induction flux emanating out of the write pole for various applied current

$$\phi_{Mag}(\mathbf{r}) = -\frac{e}{\hbar} \iint B_{\perp}(\mathbf{r}_{\perp}, z) dr dz$$

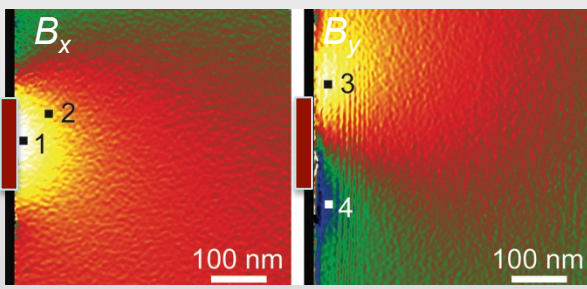
B_x component

$$\int_{-\infty}^{\infty} B_x(r_{\perp}, z) dz = -\frac{\hbar}{e} \frac{\partial \phi_{Mag}(\mathbf{r})}{\partial y}$$

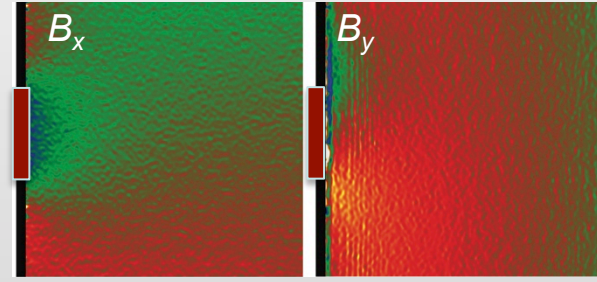
B_y component

$$\int_{-\infty}^{\infty} B_y(r_{\perp}, z) dz = \frac{\hbar}{e} \frac{\partial \phi_{Mag}(\mathbf{r})}{\partial x}$$

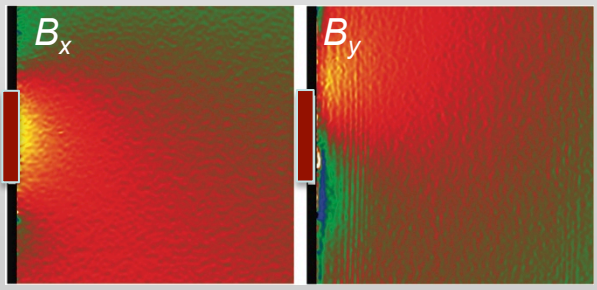
60 mA



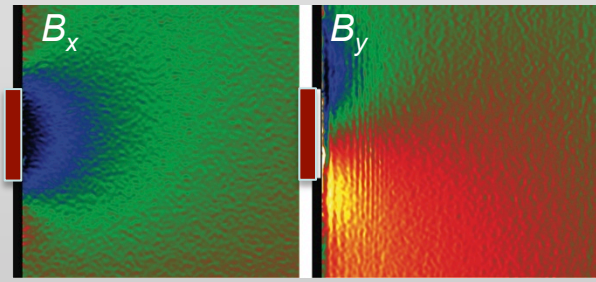
- 60 mA



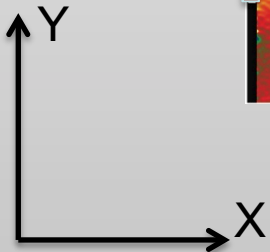
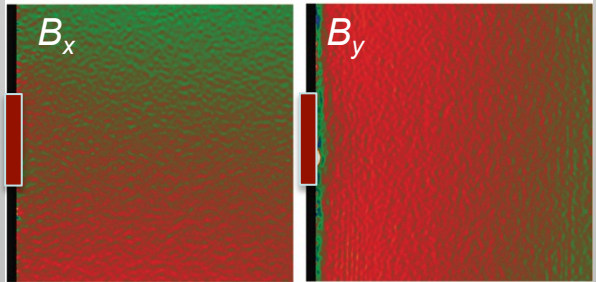
10 mA



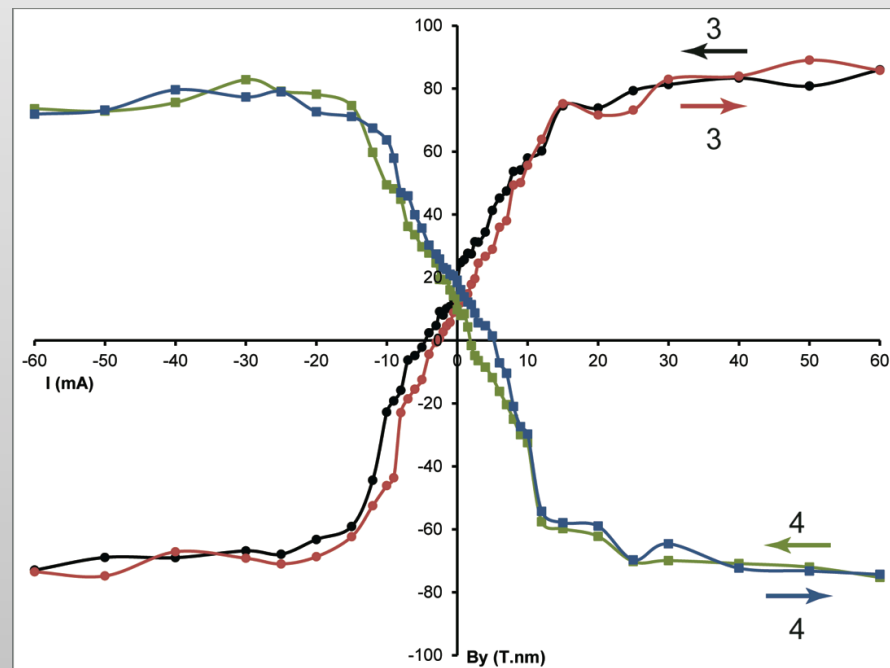
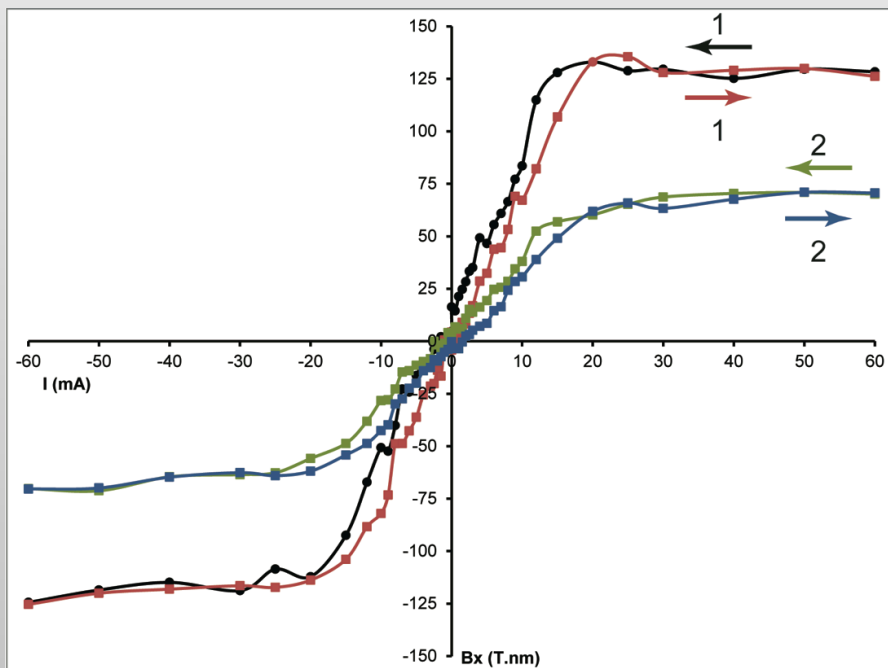
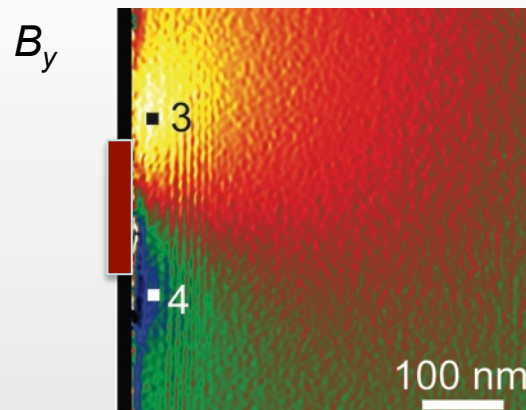
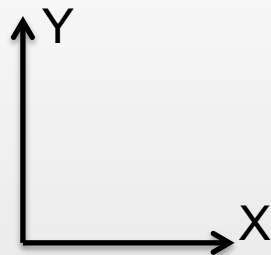
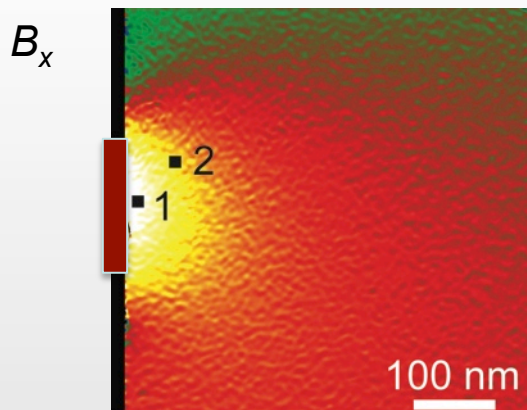
-10 mA



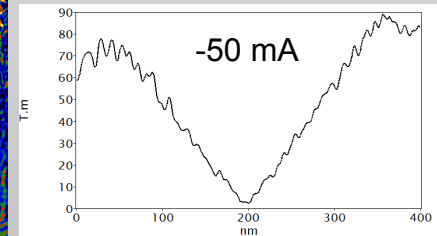
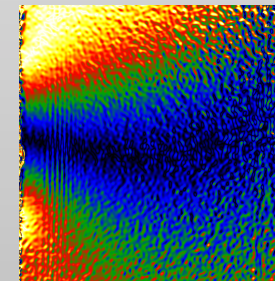
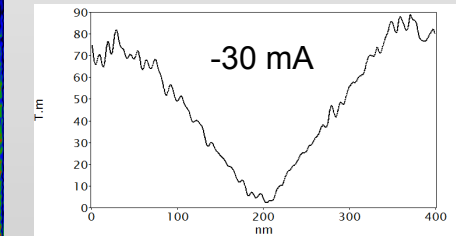
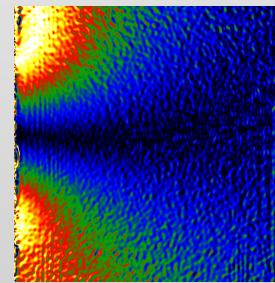
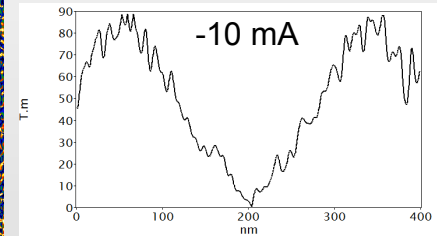
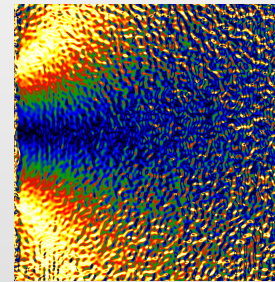
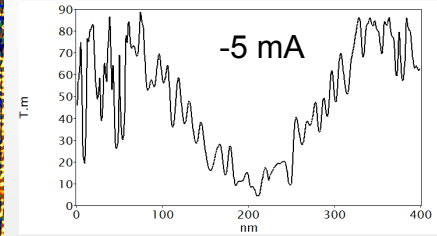
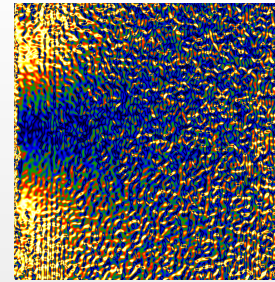
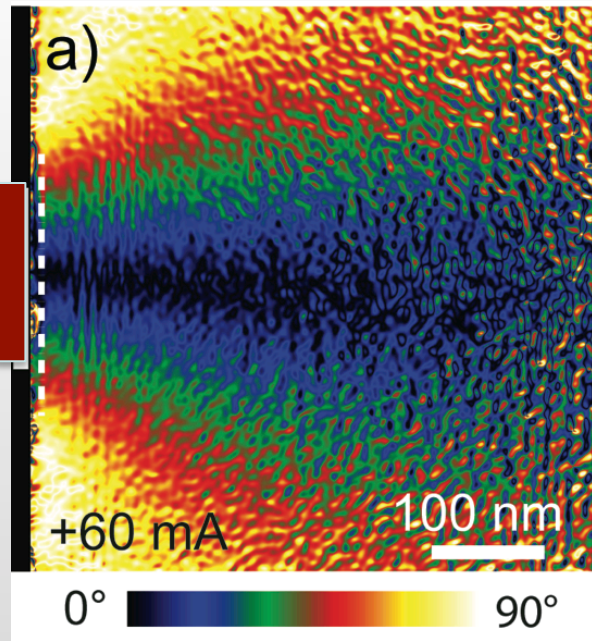
0 mA



... to local hysteresis loops



$$\theta = \arctan(B_Y/B_X)$$

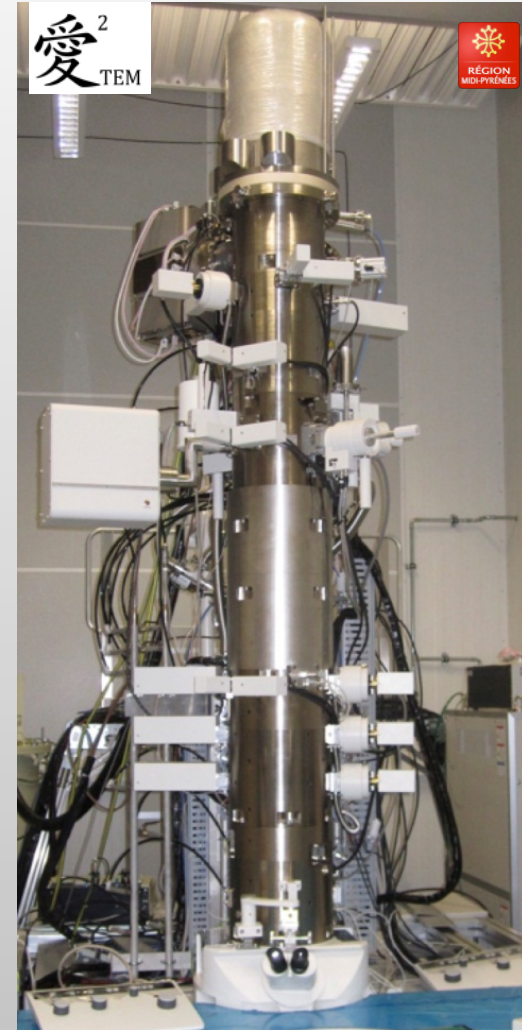


J.F. Einsle, C. Gatel et. al. *Nano Research*, 1-9, Nov. 2014

Back home message

- **Electron Holography** and **Lorentz Microscopy** are TEM methods allowing retrieving the **phase** of the electron beam with resolution of few nanometers.
- EH and LM necessitate **dedicated** instruments (C-FEG, biprisms, ...) and cautious experimental and image analyses processes
- Through the Aharonov-Bohm effect **EH** and **LM** allow quantitative measurements of local **magnetic** fields (*and electrostatic*) therefore allowing determining **local magnetic configurations** and **local magnetic moments** measurements
- ***In-situ*** EH and LM experiments permits to study **phase diagram** of magnetic nanomaterials and magnetic devices **in operando**

EH and *in-situ* dedicated **I2TEM** (Hitachi) TEM with C-FEG & Cs corrected Lorentz lens (B-Corr) & multibiprism set-up
→ Resolution in Lorentz mode : 0.5 nm





ESTEEM2 is an
**INTEGRATED
INFRASTRUCTURE NETWORK**
of electron microscopy facilities
providing access for the academic
and industrial research community
in materials science to the most
powerful TEM techniques
available at the
nanoscale.

14 European schools and workshops

to provide training in innovative methods
in electron microscopy and a forum for
discussing emerging techniques.

(including QEM2013: www.qem2013.com)

ESTEEM2 research program

focuses on the further
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- electron diffraction
- imaging
- spectroscopy
- 3D methods
- time-resolved microscopy



Free transnational access

to the most advanced TEM equipment and skilled operators for HR(S)TEM, EELS, EDX, Tomography, Holography and various in-situ state-of-the-art experiments.

ESTEEM2 available equipment includes:

- 19 FEI-Titan 60-300 and Tecnai G2 aberration corrected microscopes
- 6 JEOL aberration-corrected TEM
- A NION probe corrected Ultrastem 200
- The ZEISS SESAM
- The HITACHI I2TEM Cs corrected C-FEG
- Extensive FIB and sample preparation facilities.



The TEM group @ CEMES - Toulouse



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Warot

Florent
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Masseboeuf

Maïc
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Christophe
Gatel

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Laura Caustardoy

Rong-rong Wang

Cesar Magen

Jaime Gasquet

Luis-Alfredo Rodriguez

Rodrigo Fernandez-Pacheco

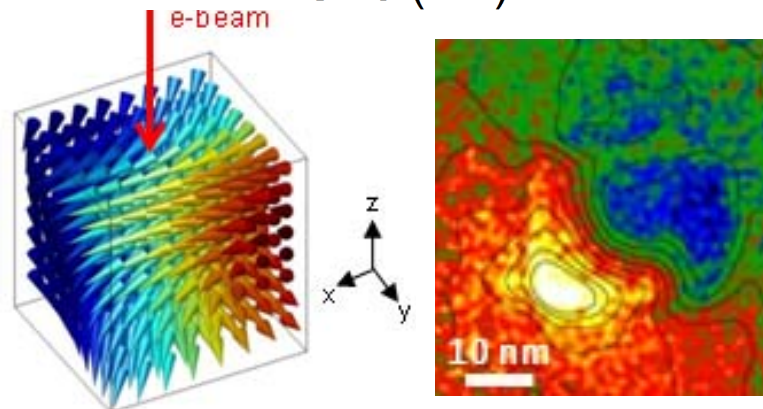
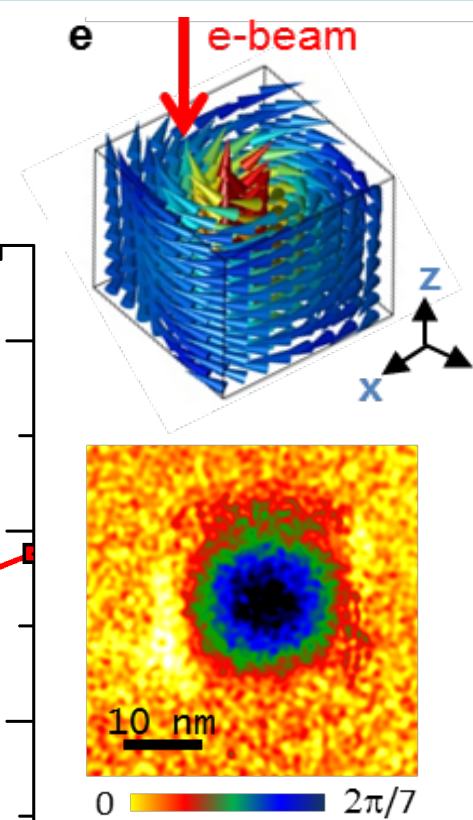
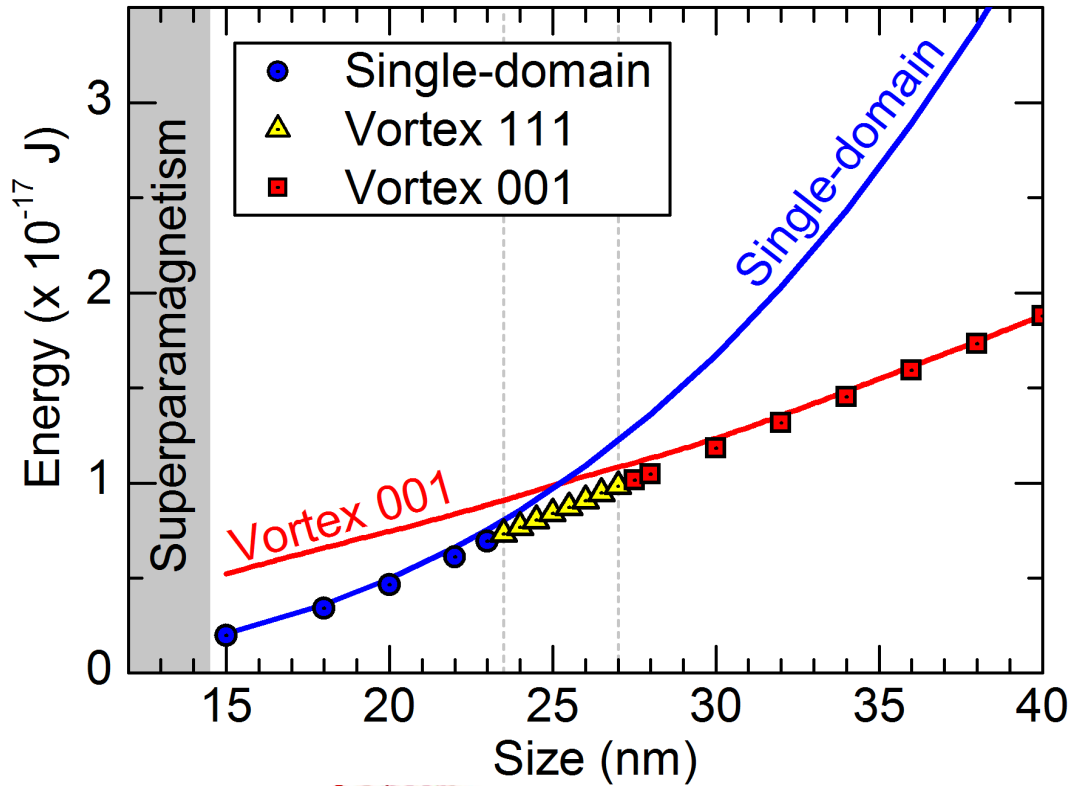
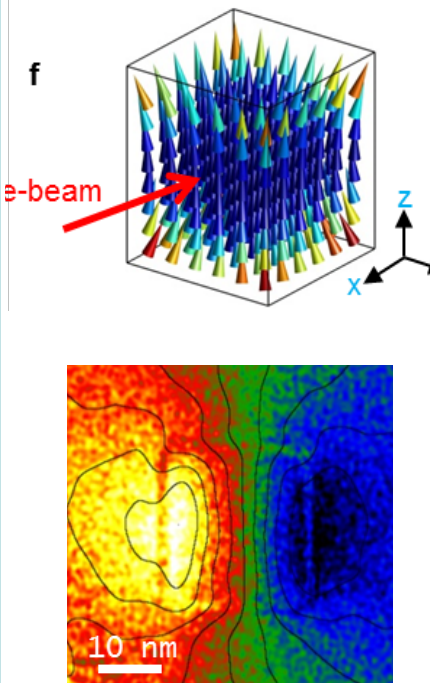
Etienne SNOECK

Raul Arenal

Thank you for listening



Phase diagram (without Oxyde layer)

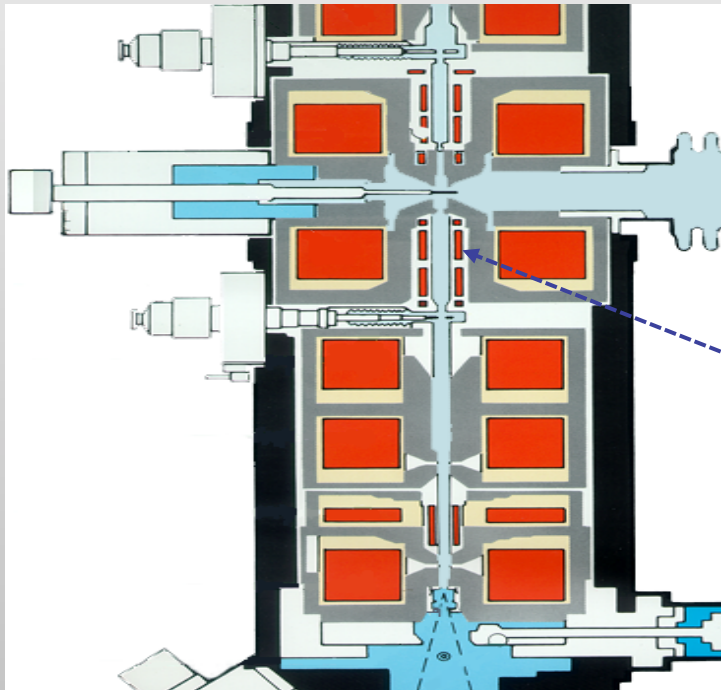


Coll. T. Blon, L.M. Lacroix
 INSA-LPCNO (Toulouse)

$$\varphi(\mathbf{x}) = C_E \int V(x, z) dz - \frac{e}{\hbar} \iint B_{\perp}(x, z) dx dz$$

→ e⁻ beam phase is only sensitive to B_⊥

Pb. 1: **The objective must be switched off** => use of Lorentz lens to get sufficient resolution



- Object in a magnetic field free area
- Field of view up to 1 μm

f = 30 mm

Cs = 7430 mm (!!)

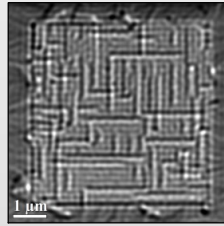
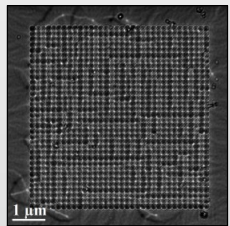
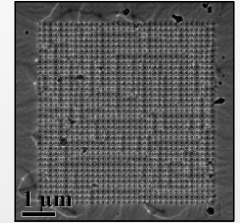
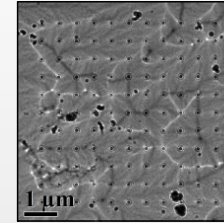
Cc = 39 mm

Ponctual résolution # 5 nm

IN-SITU EXPERIMENTS UNDER EXTERNAL APPLIED MAGNETIC FIELD

SUMMARY/RESULTS

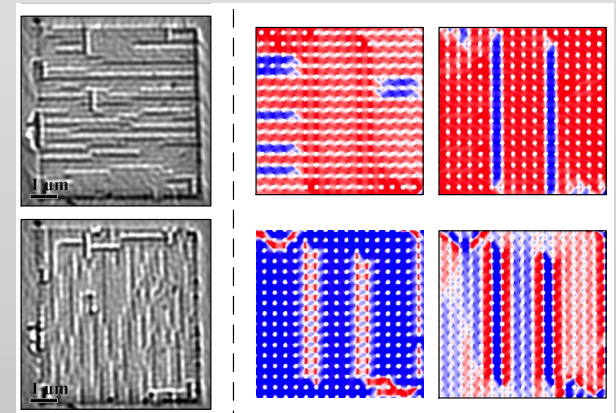
At remanence, Lorentz images allowed detecting a transition in the magnetic structure due to the reduction of the hole periodicity.



A Fourier filter method permitted enhancing the visualization of magnetic contrasts in Co antidot arrays with periodicities down to 116 nm

The magnetization reversal process is given by NUCLEATION (N) and PROPAGATION (P) of SDWs.

- **Transverse Magnetic field:** Simultaneous N and P of horizontal and vertical SDWs.
- **Parallel Magnetic field:** At two steps: 1) N and P of horizontal SDWs. 2) N and P of vertical SDWs.



L. A. Rodriguez et. al. *Nanotechnology* **23** (2014) 385703

Surface plots Hysteresis loops

