Electron Holography

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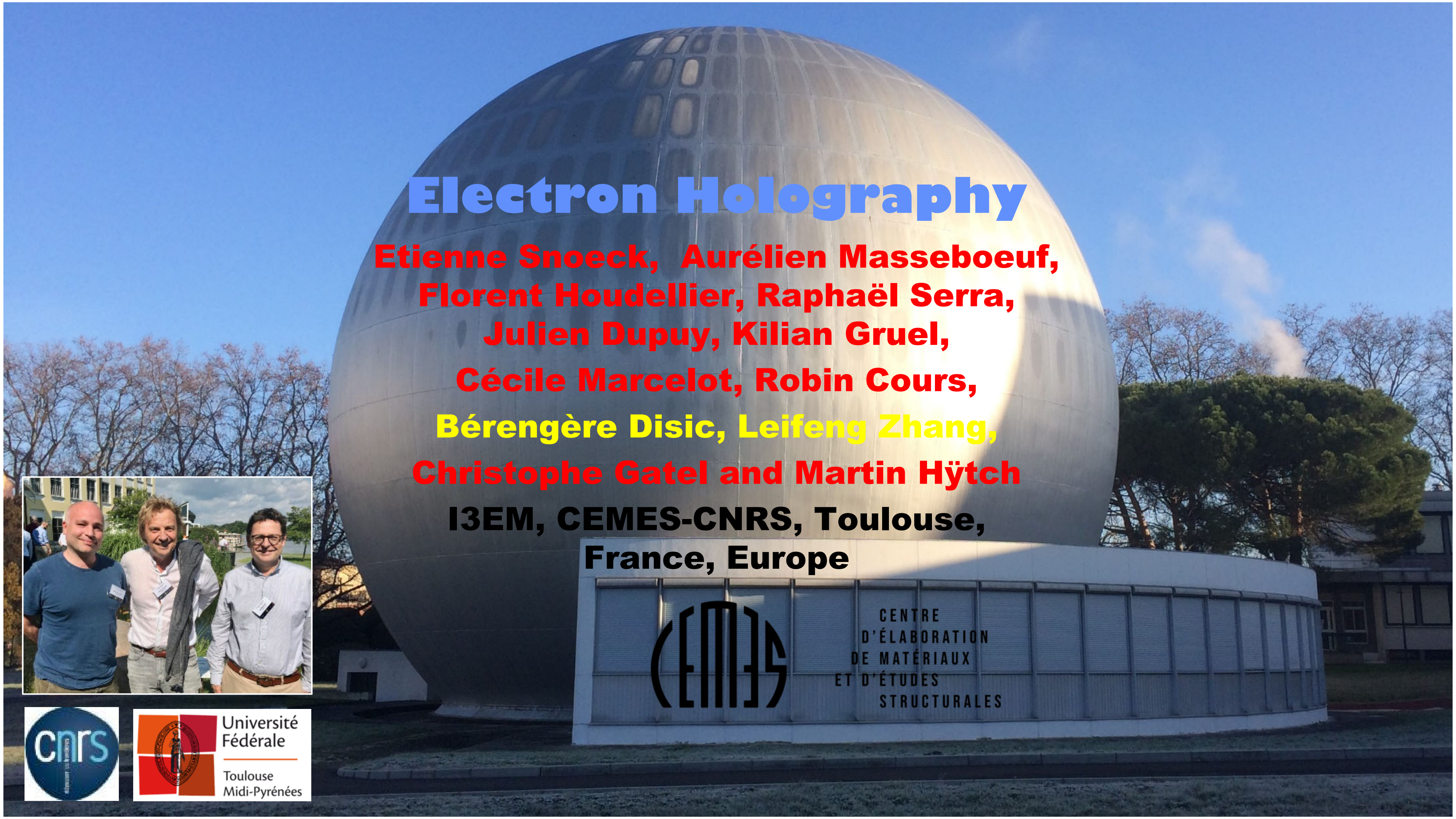
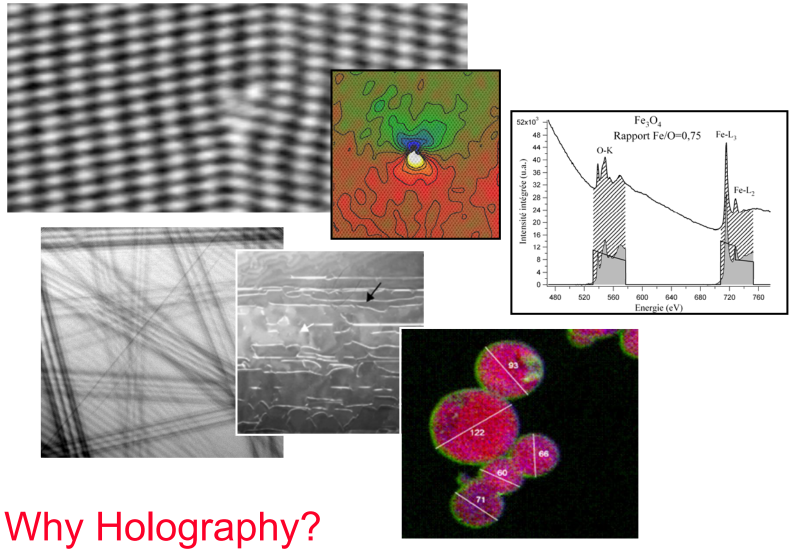


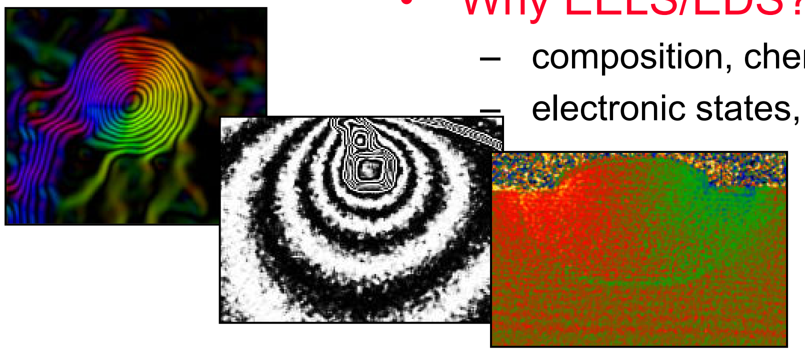
Université Fédérale Toulouse Midi-Pyrénées

Why Holography?

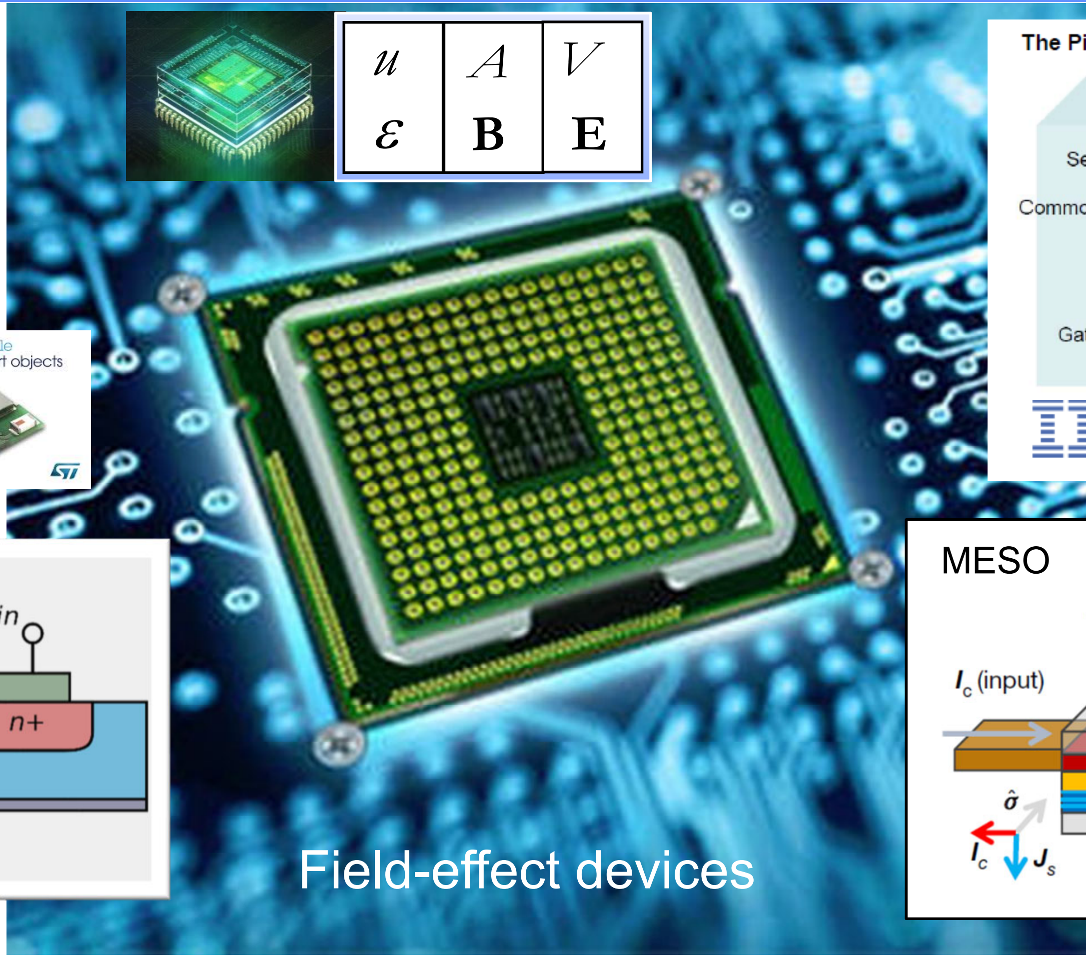


- Why HR(S)TEM?
  - local structure
  - (strain mapping, composition)
- Why Diffraction?
  - crystallography
  - (lattice parameters, bonding)
- Why CTEM?
  - defects
  - (morphology)
- Why EELS/EDS?
  - composition, chemistry
  - electronic states, (magnetism)

- Why Holography?
  - magnetic fields
  - electric fields
  - strain fields
  - (super resolution)

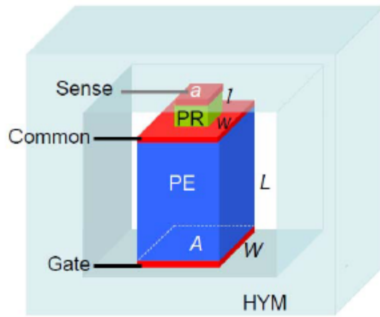


Why fields?

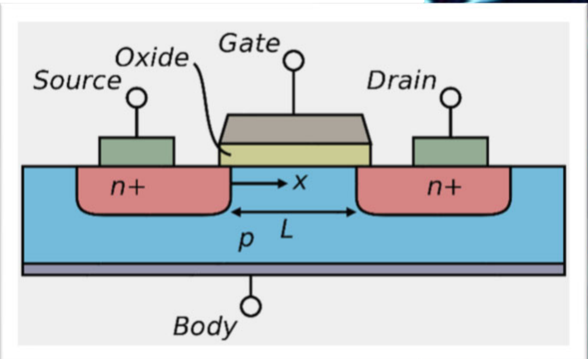


u	A	V
ε	$\mathbf{B}$	$\mathbf{E}$

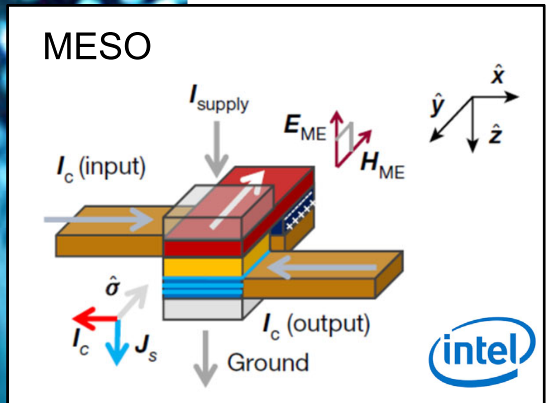
The PiezoElectronic Transistor (PET)



Field-effect transistor



MESO



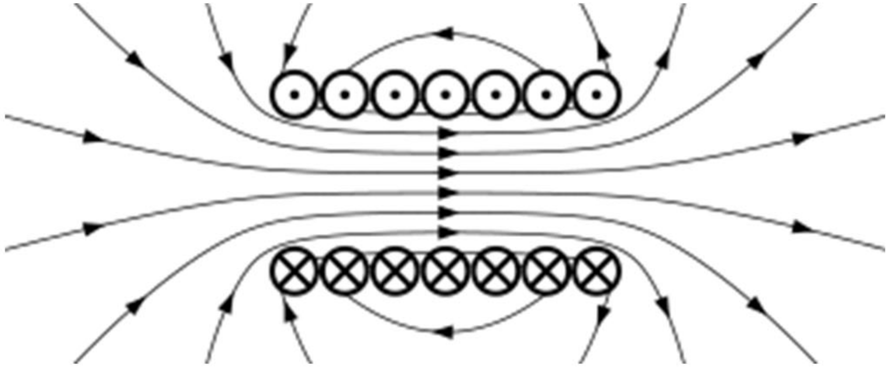
Maxwell's Equations

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\varepsilon_0}$$

$$\nabla \cdot \mathbf{B} = 0$$

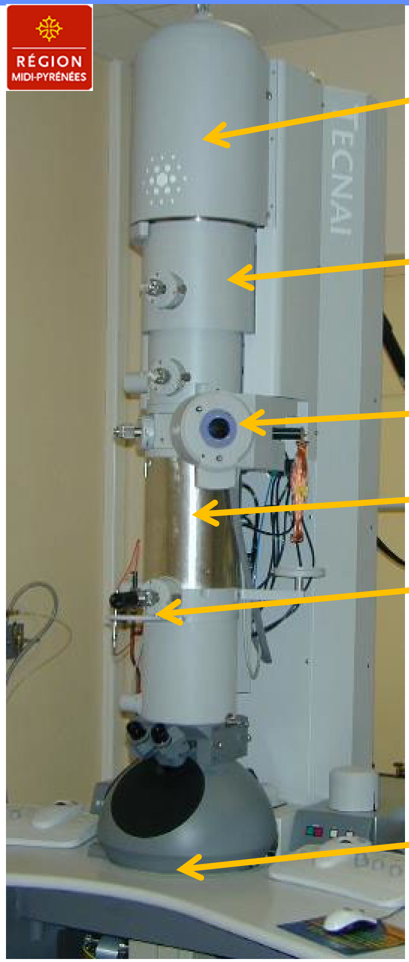
$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$

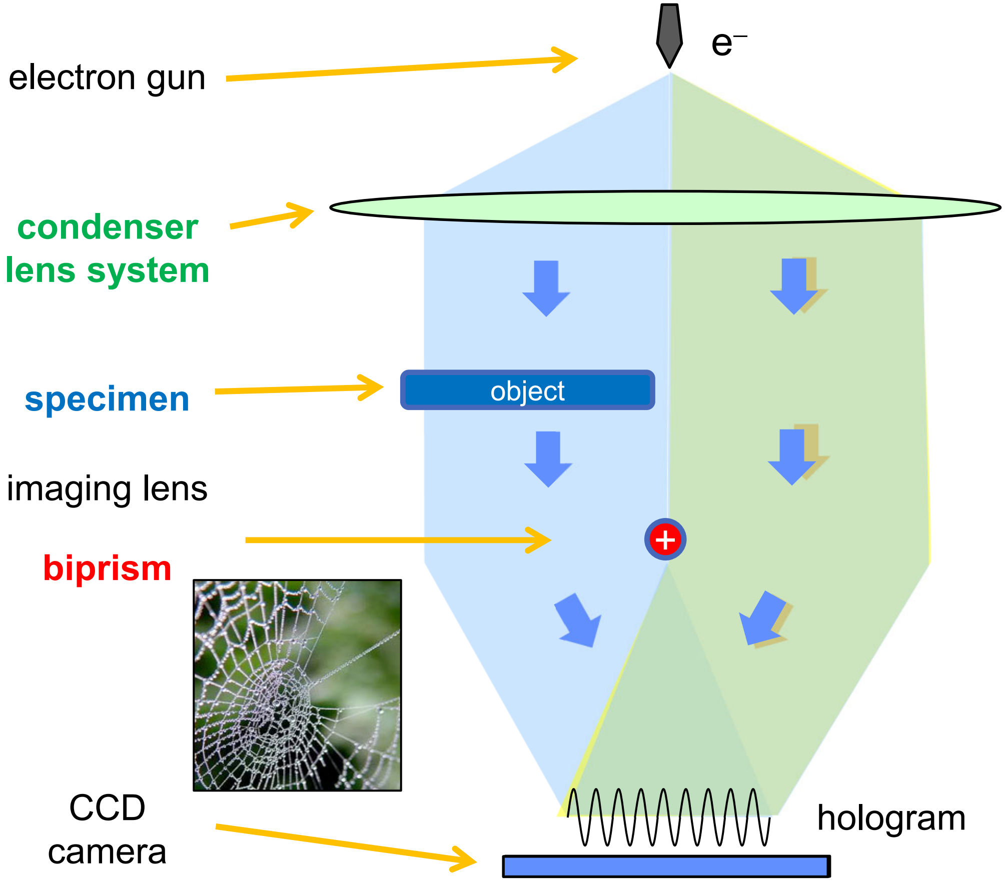
$$\nabla \times \mathbf{B} = \mu_0 \mathbf{j} + \frac{1}{c^2}\frac{\partial \mathbf{E}}{\partial t}$$



James Clerk Maxwell, Phil. Mag (1861).
On the Physical Lines of Force.

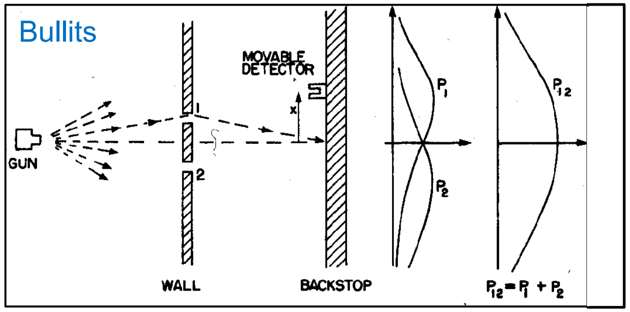
Electron Holography



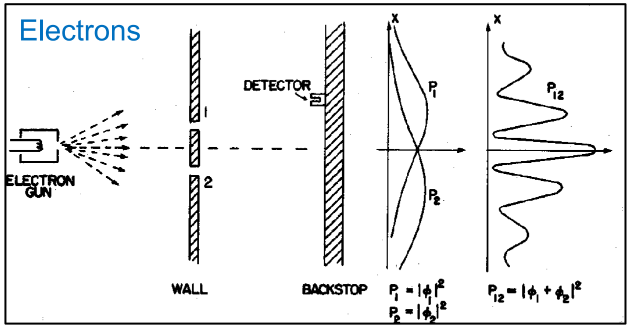


Wave-Particle Duality

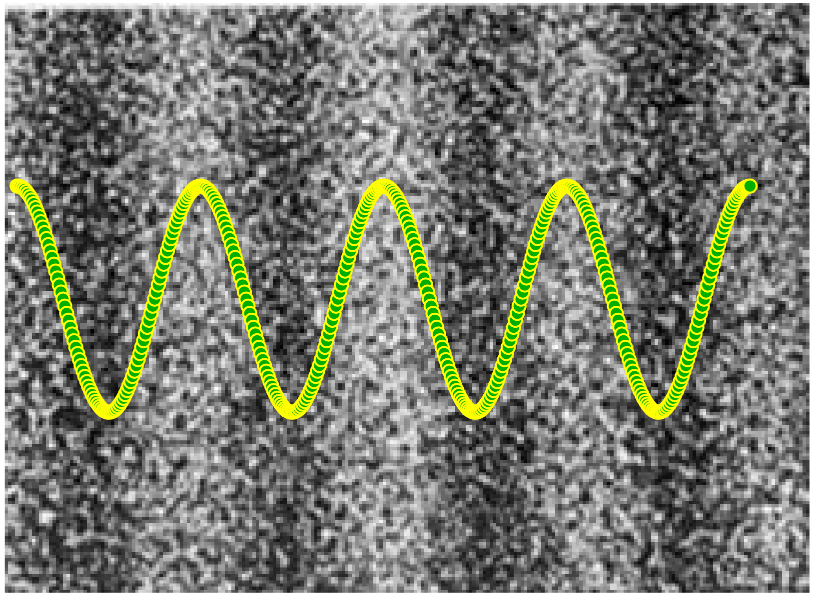
Double-slit thought experiment



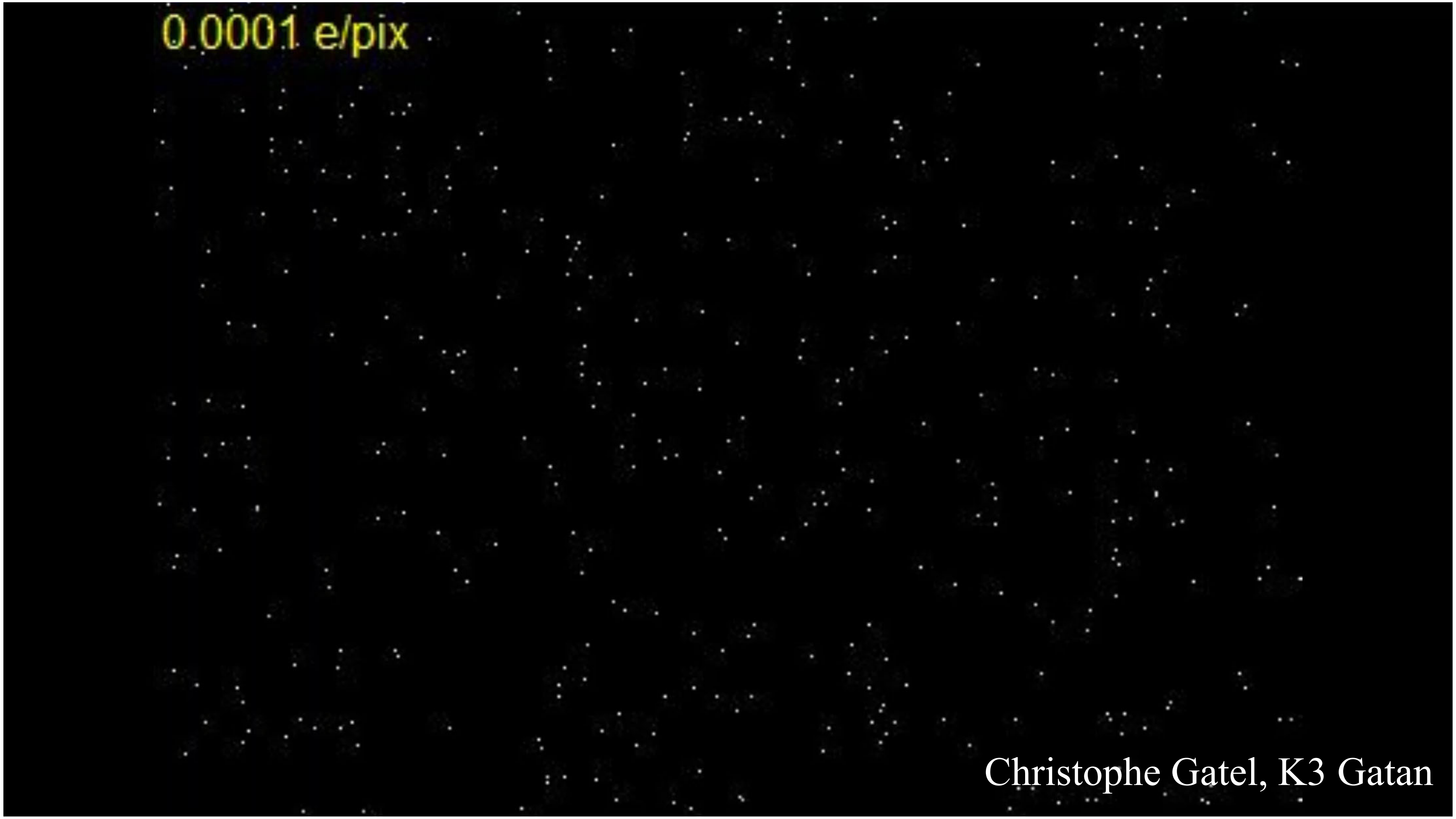
R. Feynman, Lectures on Physics, Volume III (1964)



Experiment: electron holography, one electron at a time



P. G. Merli, G. F. Missiroli, and G. Pozzi, *American Journal of Physics* **44** (1976), 306–307


0.0001 e/pix
Christophe Gatel, K3 Gatan

Phase approximation

Wavefunction: $$\Psi(\boldsymbol{r}, z) = \psi(\boldsymbol{r}) e^{2\pi i k_0 z}$$

$$k_0 = 1/\lambda$$

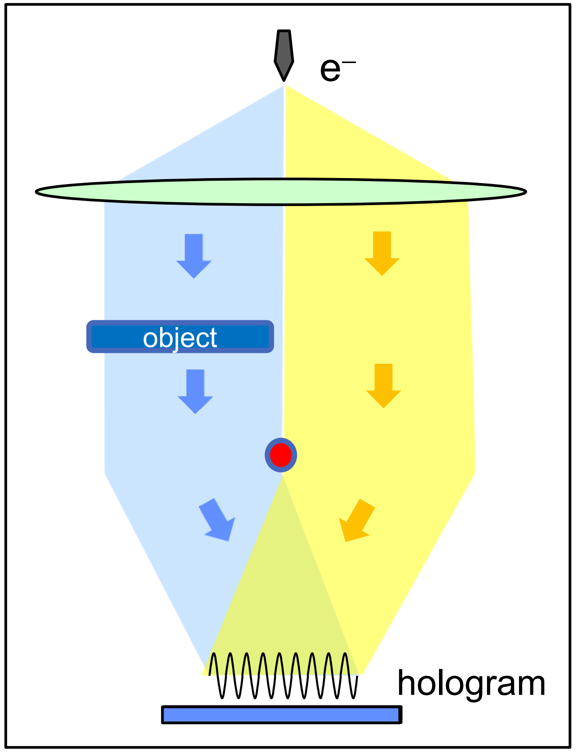
Incident wavefunction:

$$\psi_0(\boldsymbol{r}) = 1$$

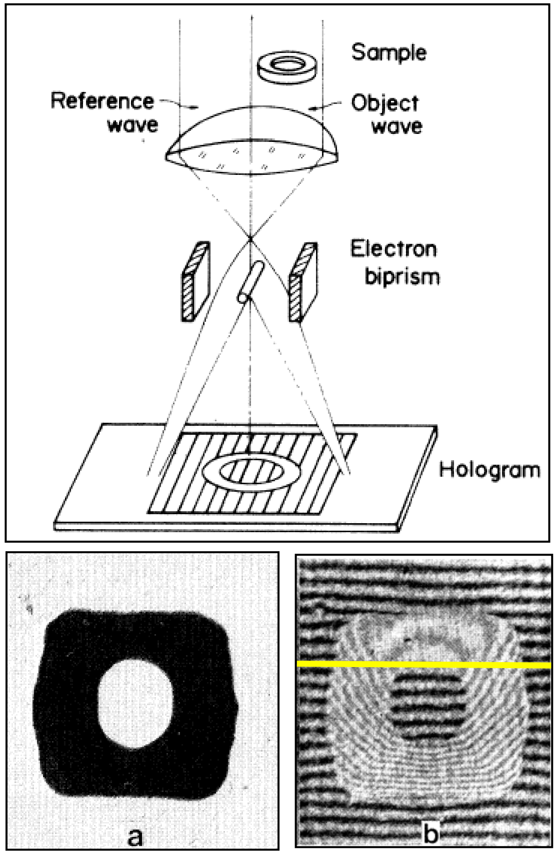
Object wavefunction:

$$\boxed{\psi_{\text{obj}}(\boldsymbol{r}) = e^{2\pi i \phi(\boldsymbol{r})}}$$

- Pure phase object
- Medium-resolution electron holography



Aharonov-Bohm Phase



$$\boxed{\phi = c_E \int V^E(\mathbf{r})dz - \frac{e}{\hbar}\int A_z(\mathbf{r})dz}$$

$$c_E = \frac{me\lambda}{2\pi\hbar^2}$$

$$\phi^E = c_E \int V^E(\mathbf{r})dz$$

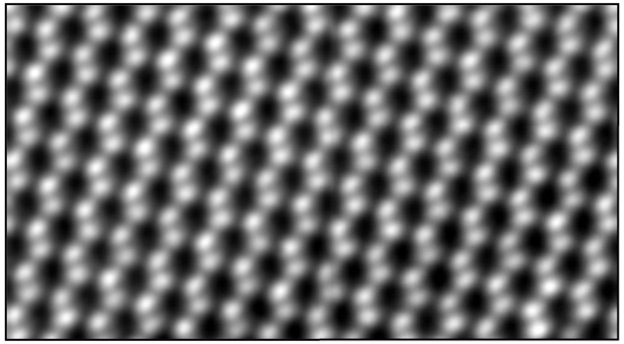
$$\phi^M = -\frac{e}{\hbar}\int A_z(\mathbf{r})dz$$

- **electrostatic phase**
- **magnetostatic phase**

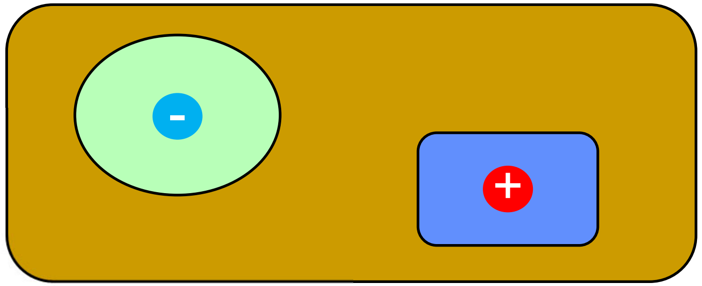
A. Tonomura et al, Phys. Rev. Lett. 56, 1215 (1986)

Continuum Theory

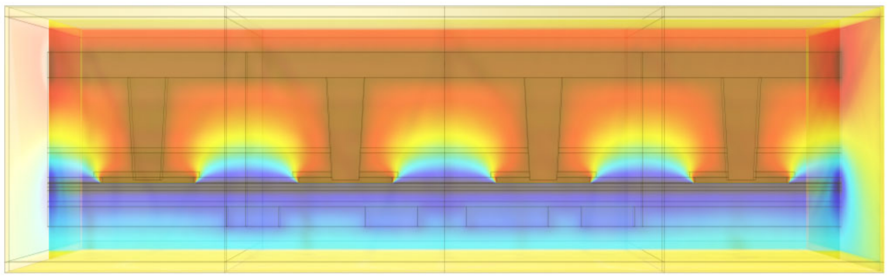
Atoms and Zone-axes



Amorphous or Off-axis

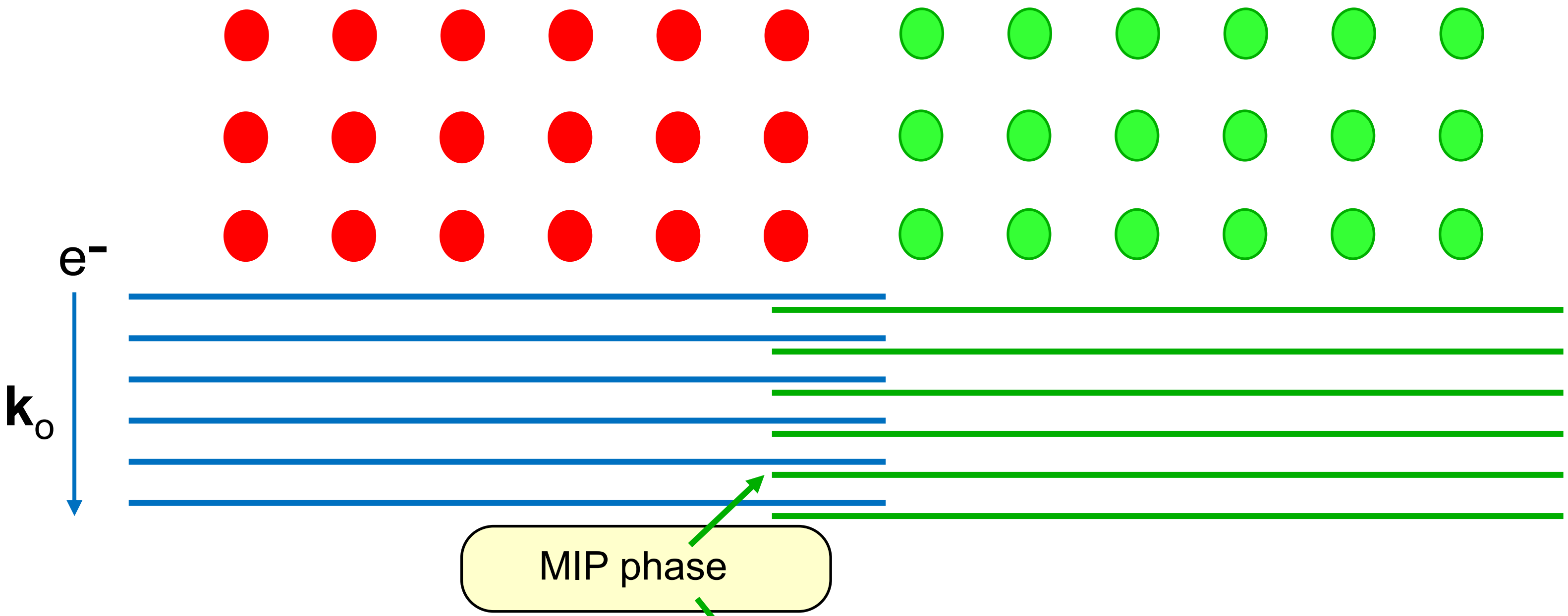


Internal potentials, charges, strain



Finite Element Method (FEM)

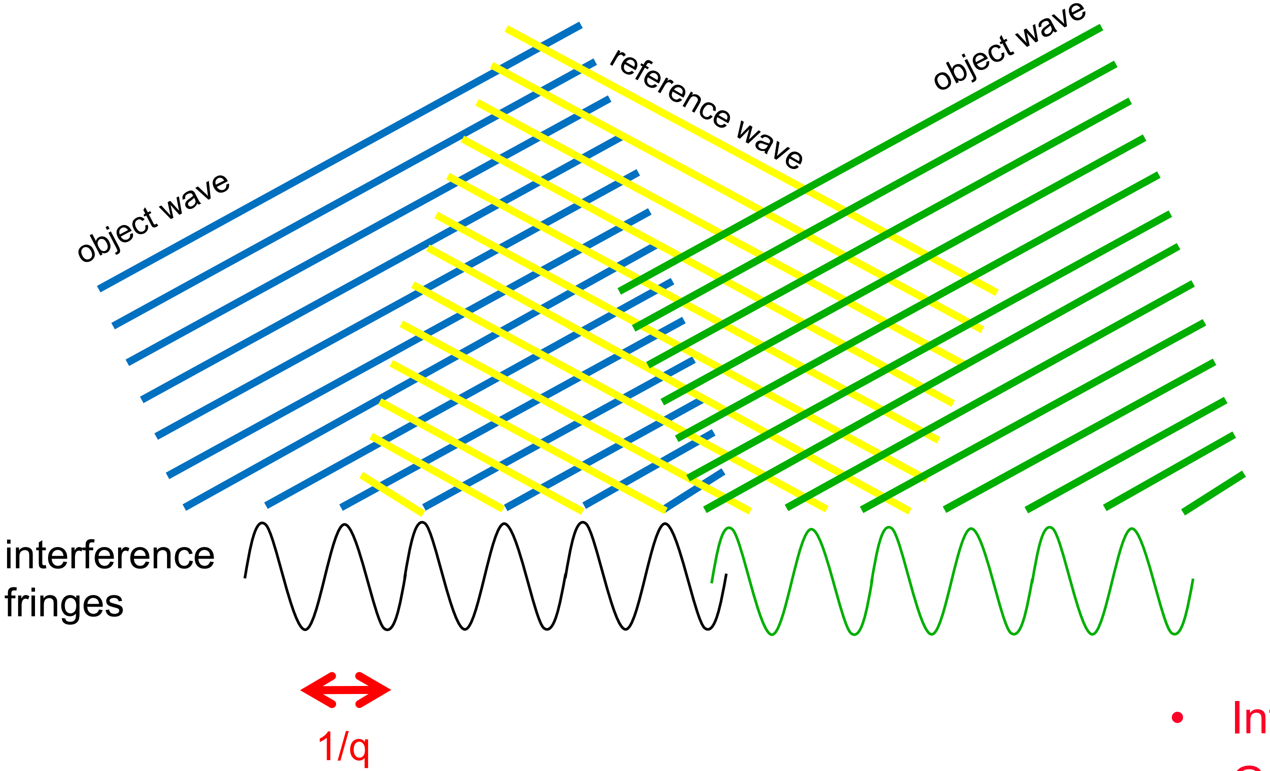
Mean inner potential

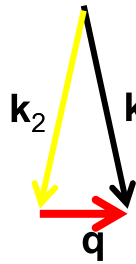


$$\phi_g^E = C_E \Delta V_0 t$$

- But how to measure?

Phase measurements

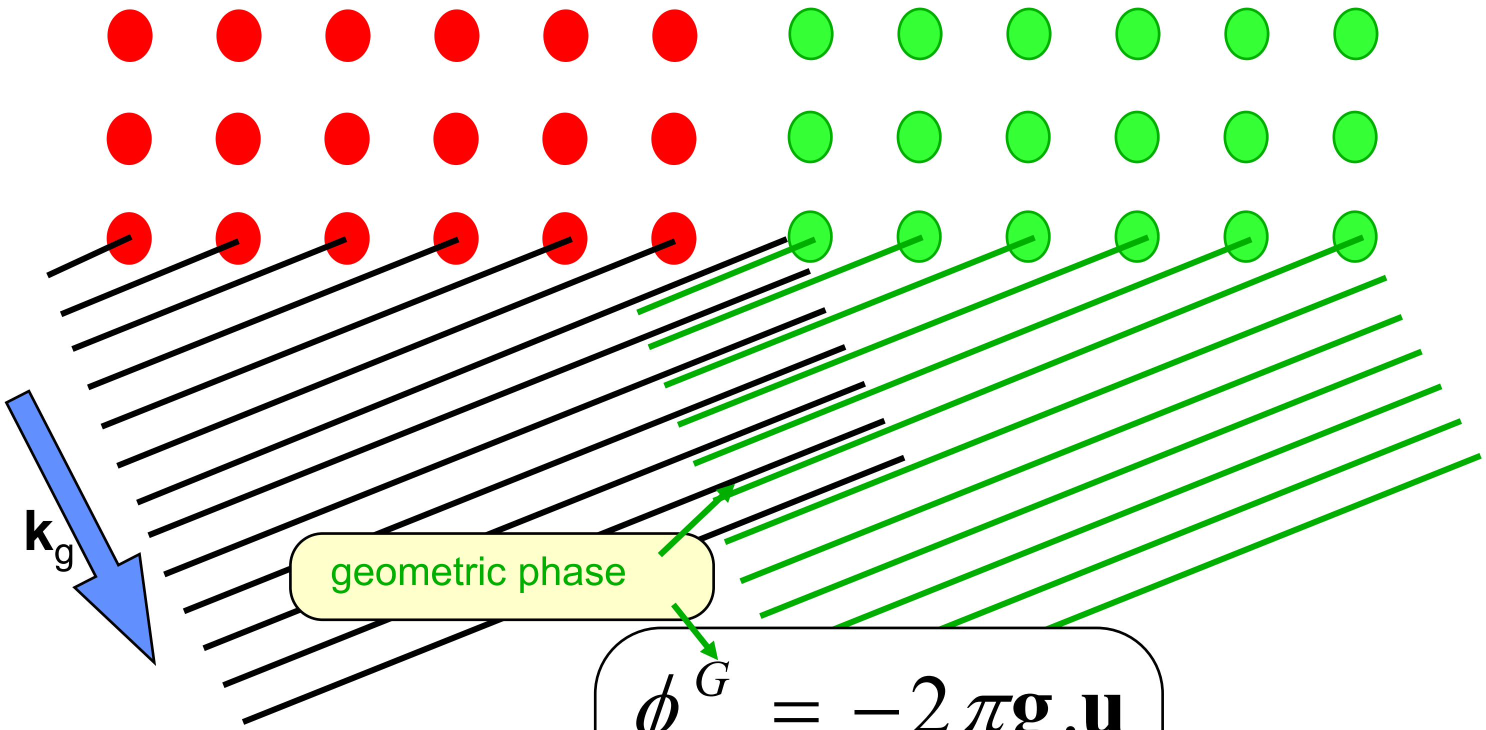




carrier frequency
$\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$

- Interferometry
- Geometry

Geometric phase



$$\phi_g^G = -2\pi \mathbf{g}.\mathbf{u}$$

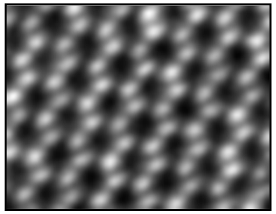
displacement

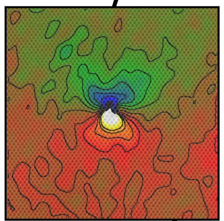
Phases in electron microscopy

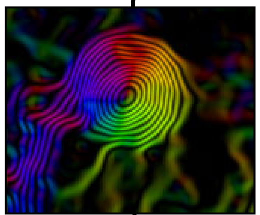
Crystalline Geometric Magnetic Electric

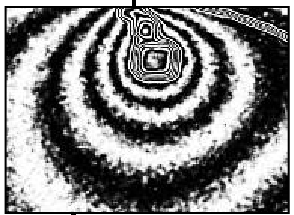
Phase shift

$$\phi = \phi^{C} + \phi^{G} + \phi^{M} + \phi^{E}$$



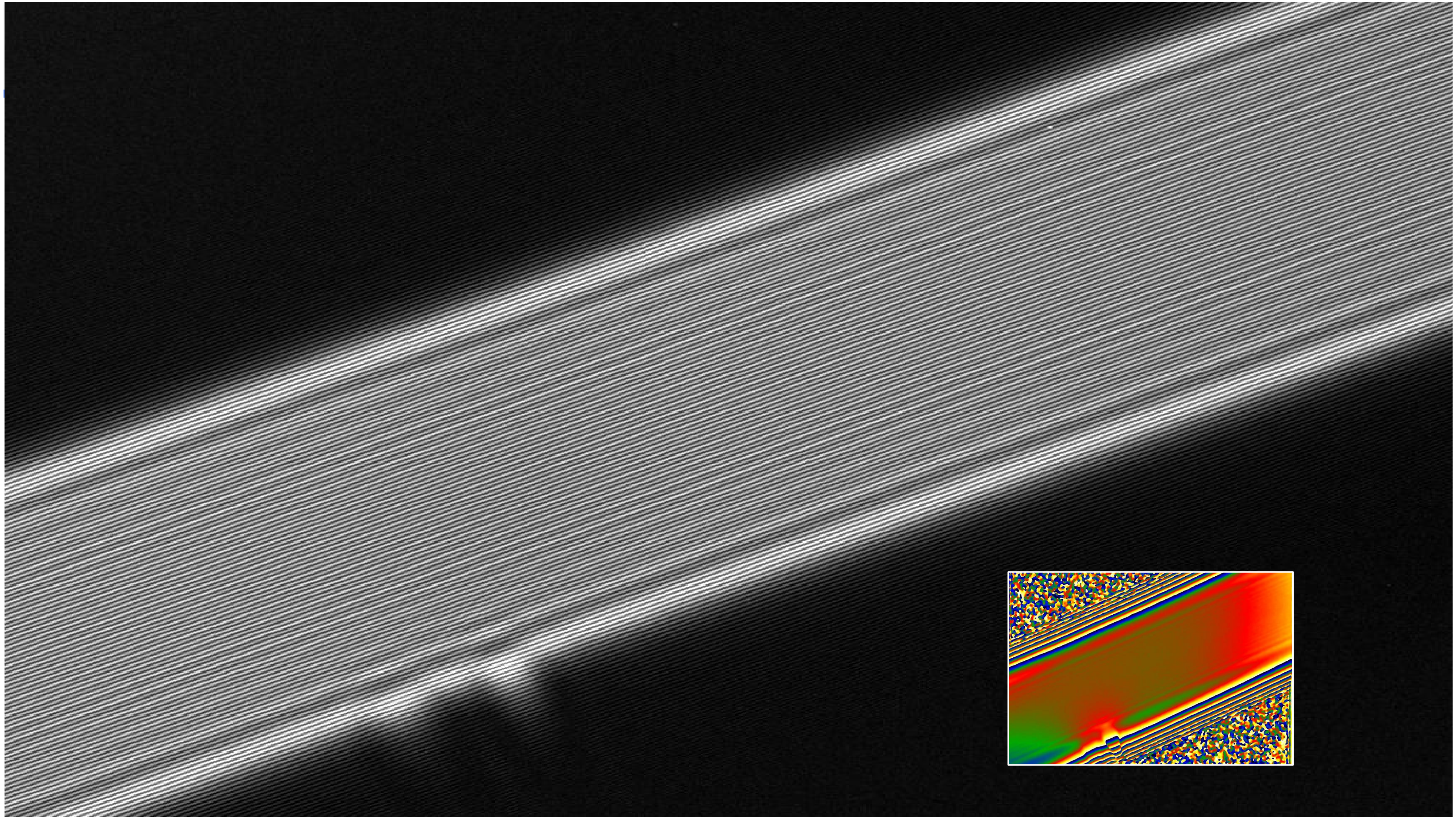






Phase gradient		Fields		
	ϕ	u	A	V
	$\nabla\phi$	ε	$\mathbf{B}$	$\mathbf{E}$

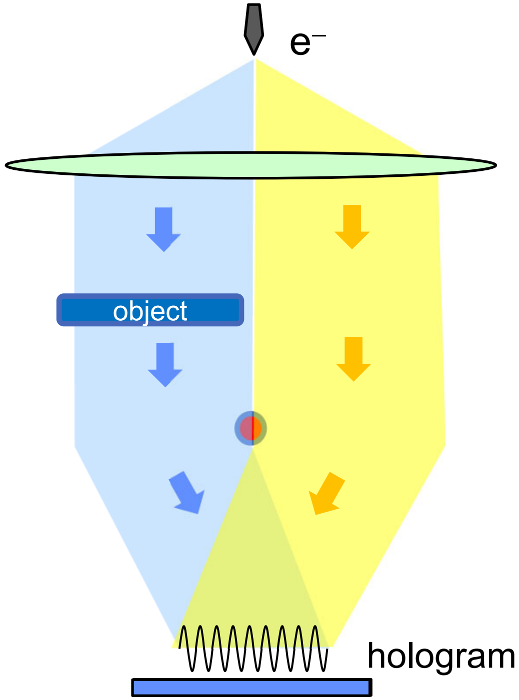
M.J. Hÿtch, F. Houdellier, F. Hüe, and E. Snoeck, Ultramicroscopy 111 (2011) 1328–1337

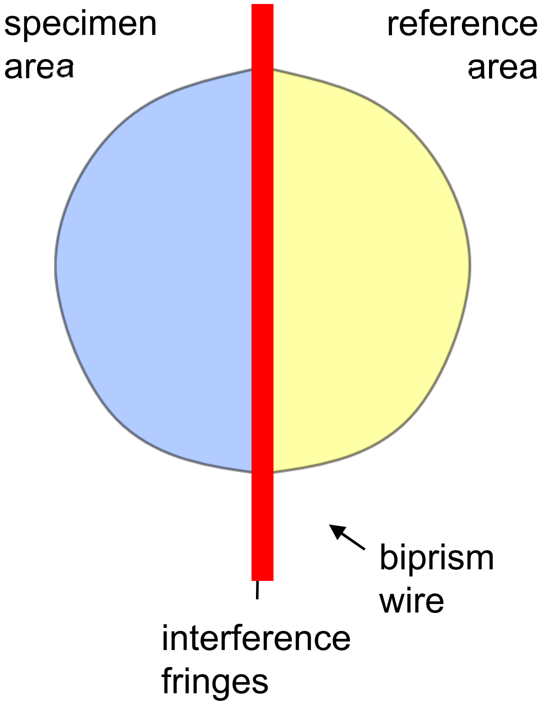


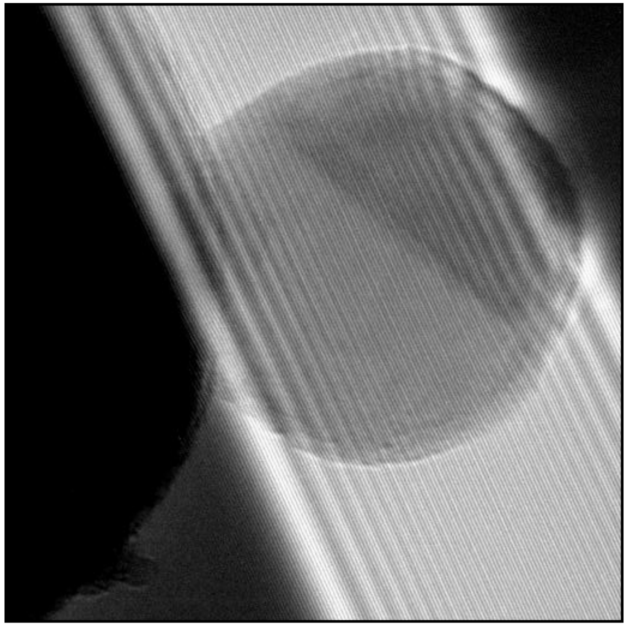
How to make a hologram

and calculate the phase

Electron holograms



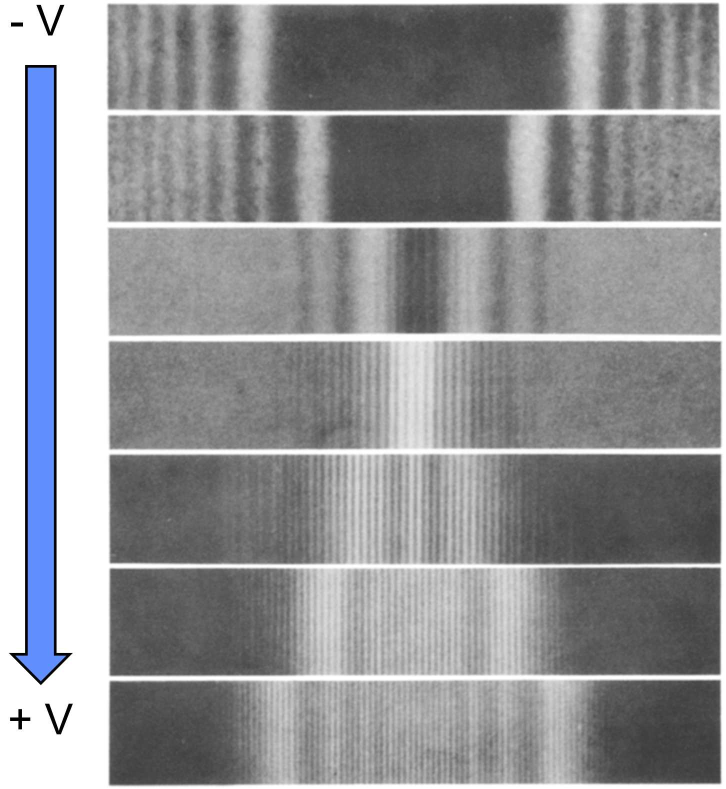




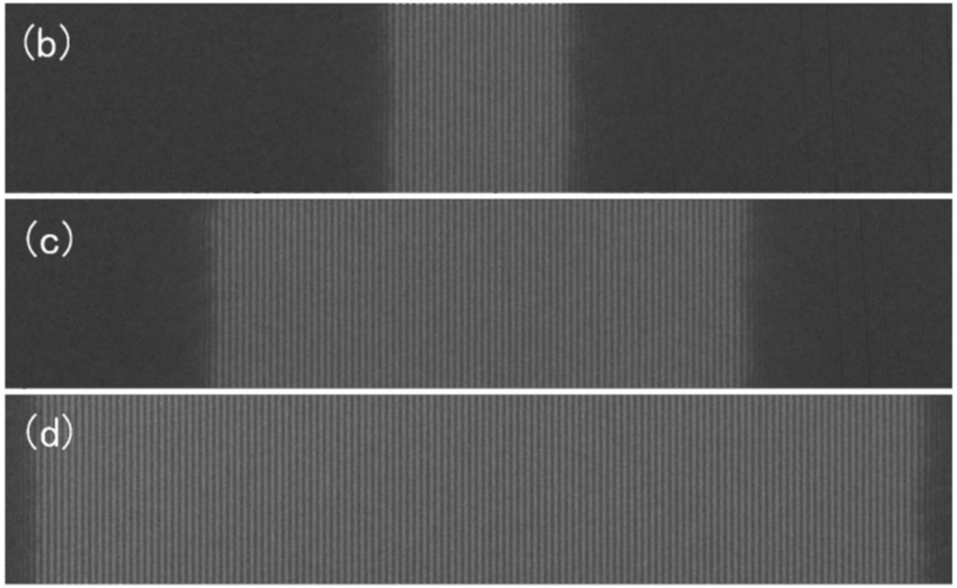
- eliptical illumination

Bi-prisms

Single Bi-prism



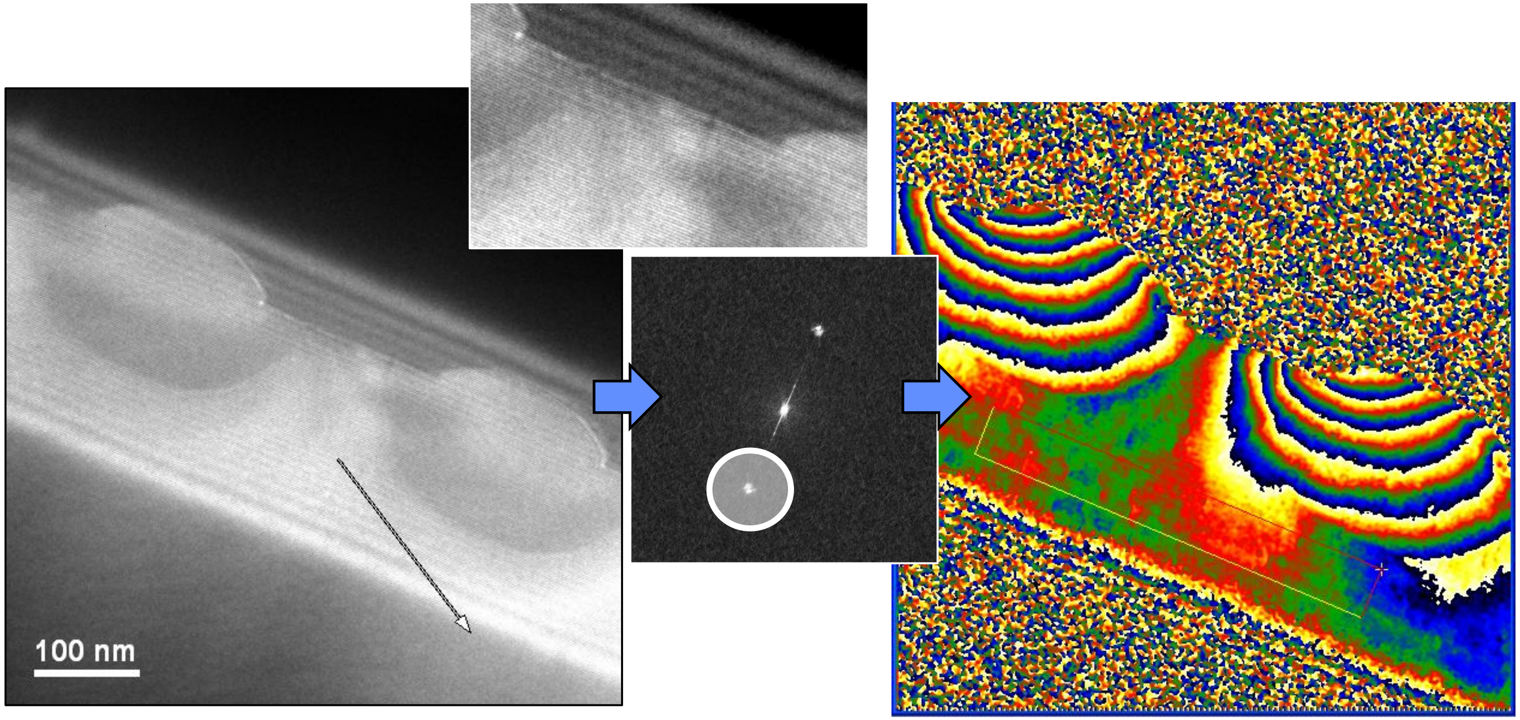
Double Bi-prisms



K. Harada et al., APL (2004)

G. Möllenstedt & H. Düker, Zeitschrift für Physik (1956)

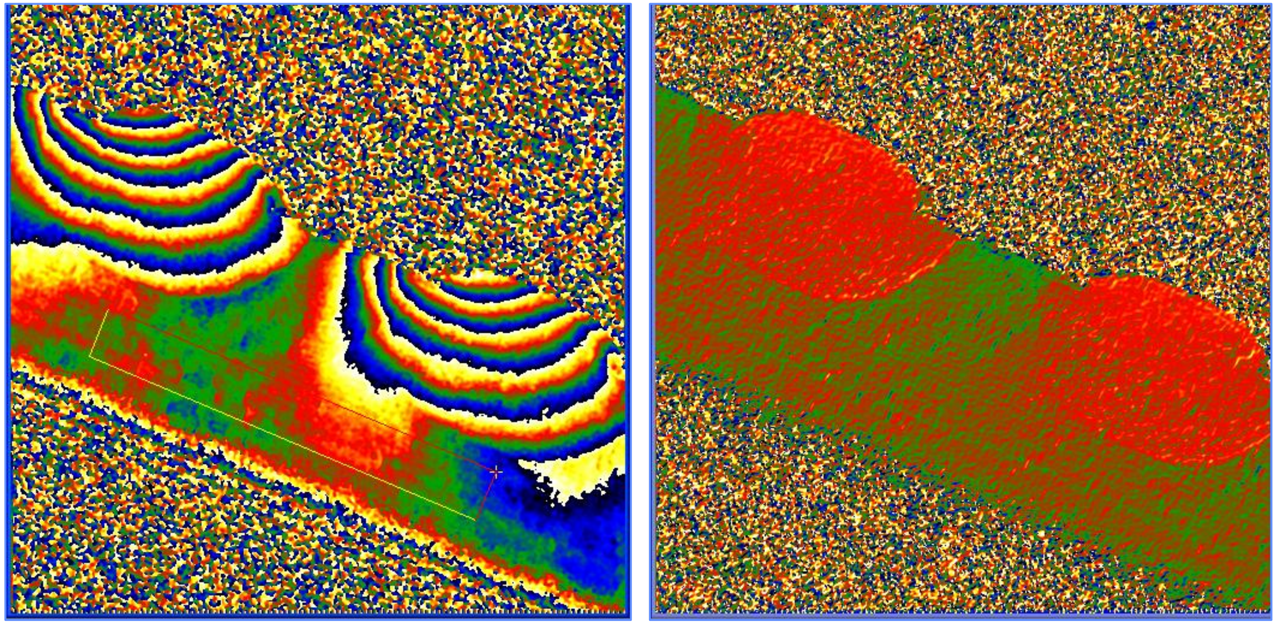
Phase calculation



- choose appropriate mask size

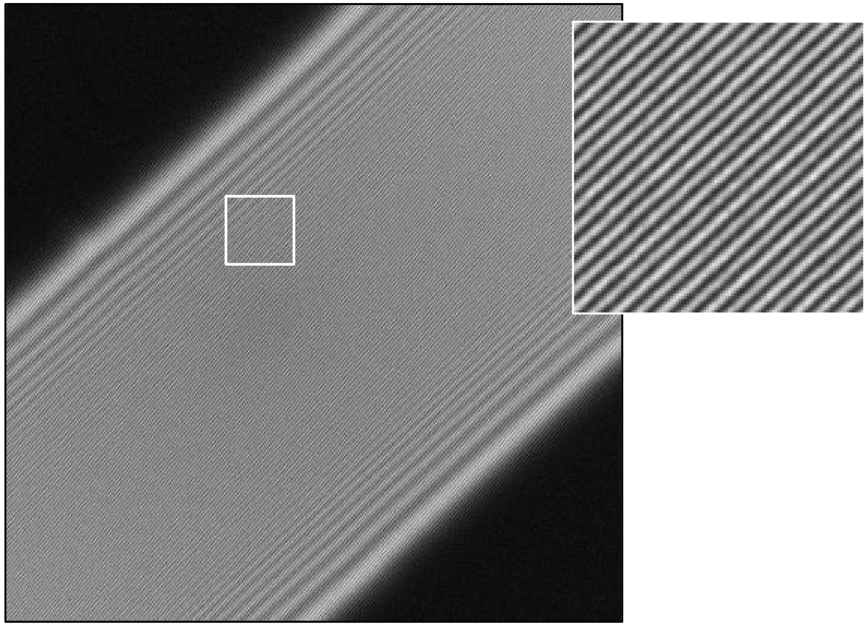
Raw phase image

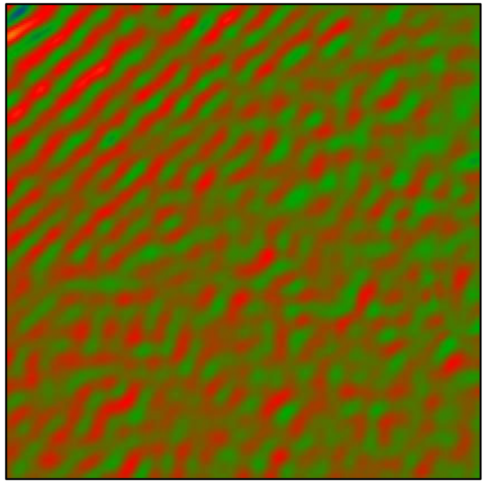
ϕ $\nabla\phi$



- derivative amplifies any problems

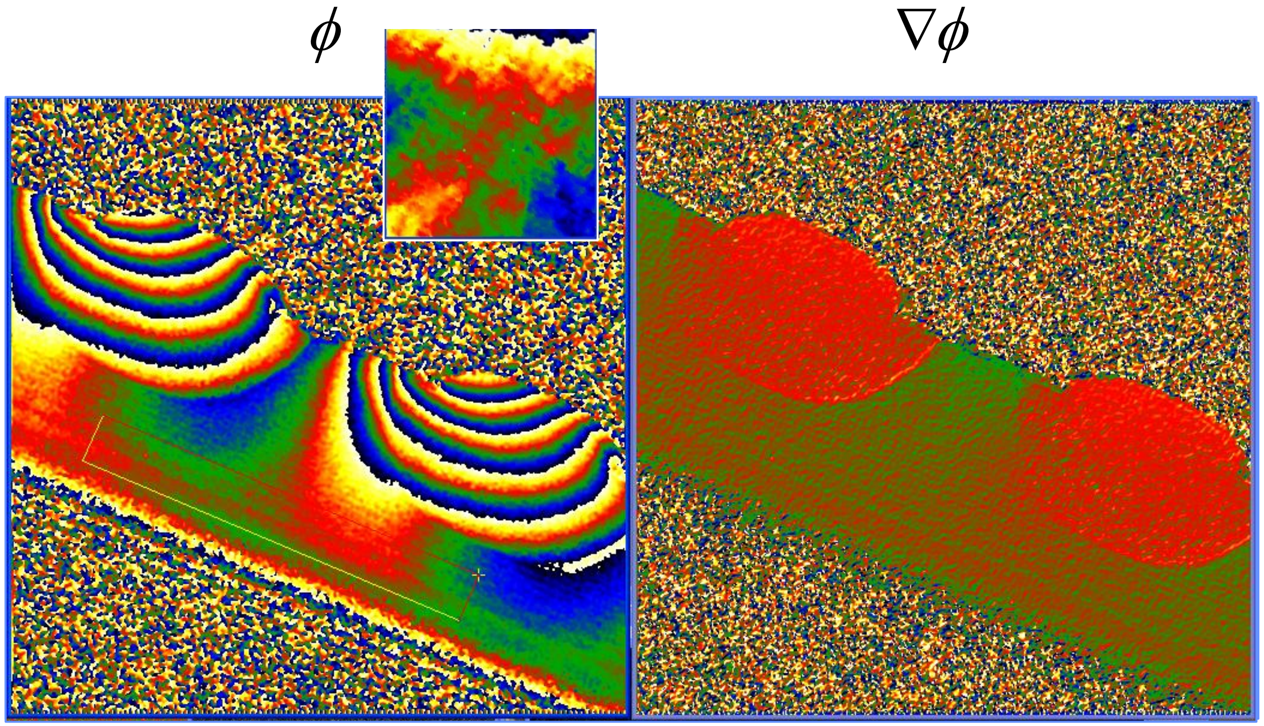
Reference Hologram





- counting statistics
- spatial coherence
- Fresnel fringes

Corrected phase

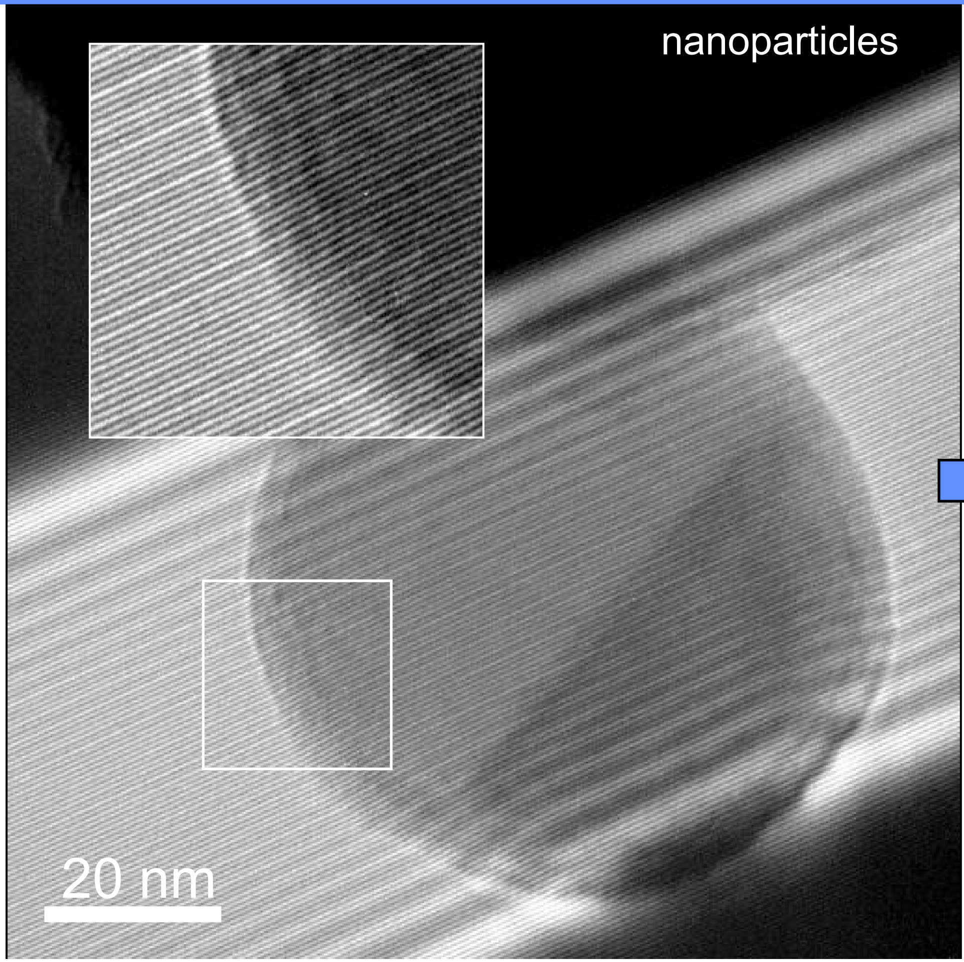


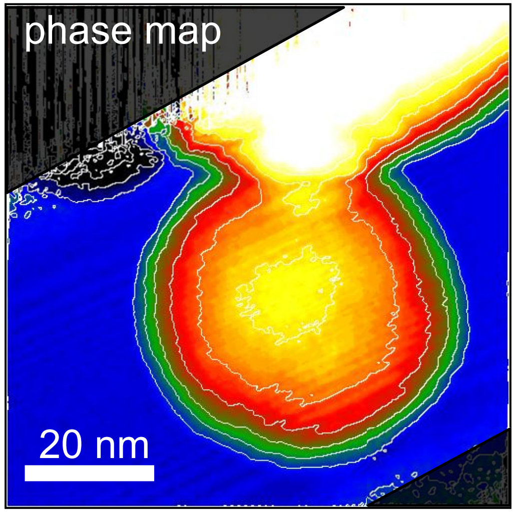
- better

Electric Fields

ϕ	u	A	V
$\nabla\phi$	ε	$\mathbf{B}$	$\mathbf{E}$

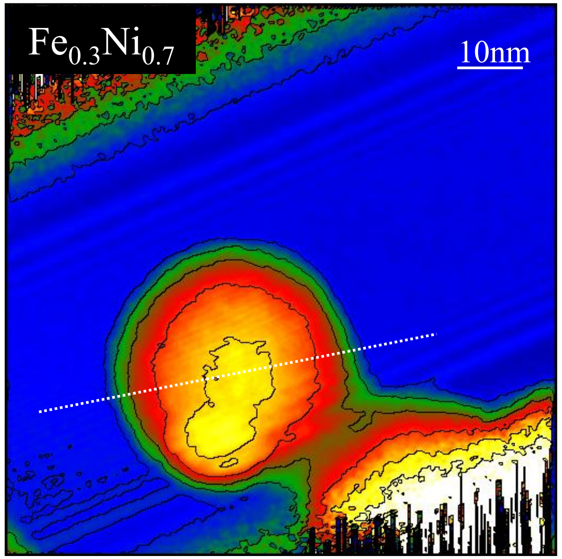
Electron holograms

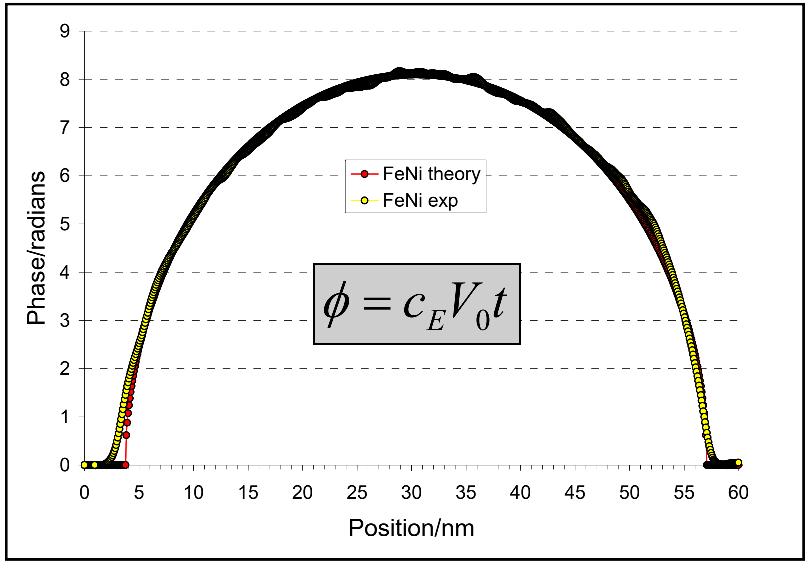




- electrostatic potential

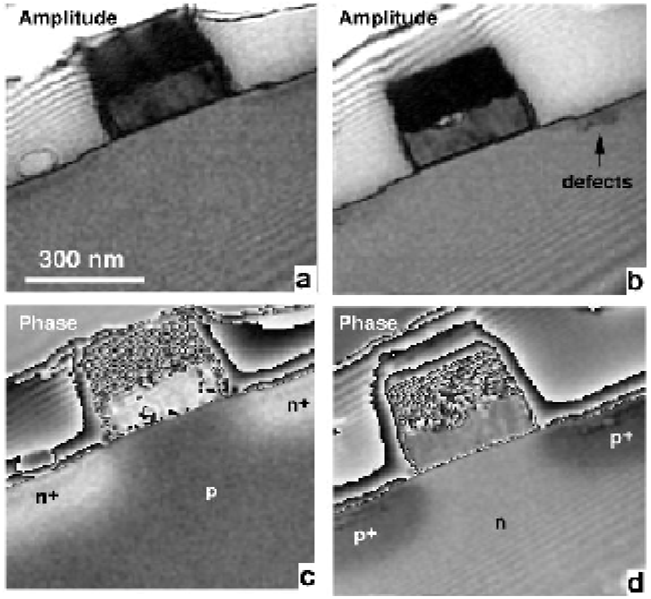
Mean inner potential

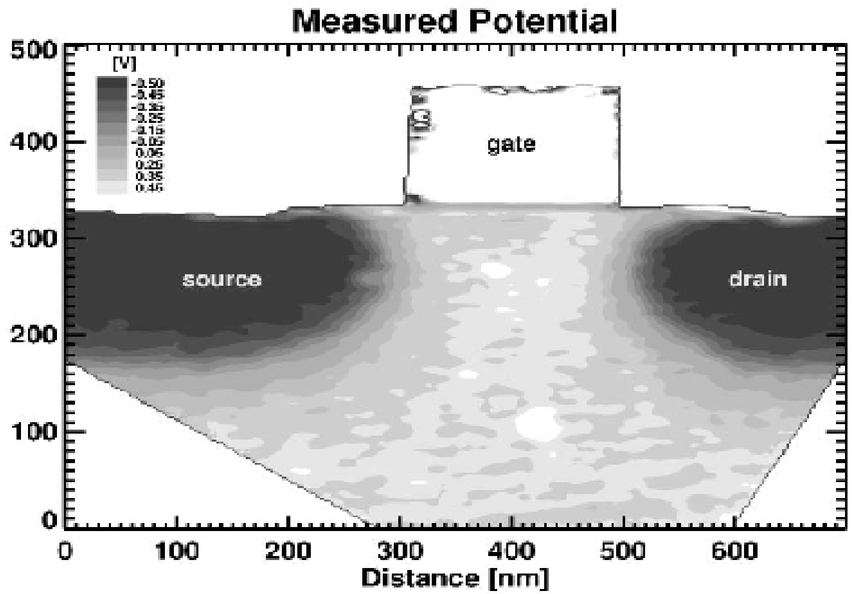




- mean inner potential measurements
- observation of small particles

Dopant profiling





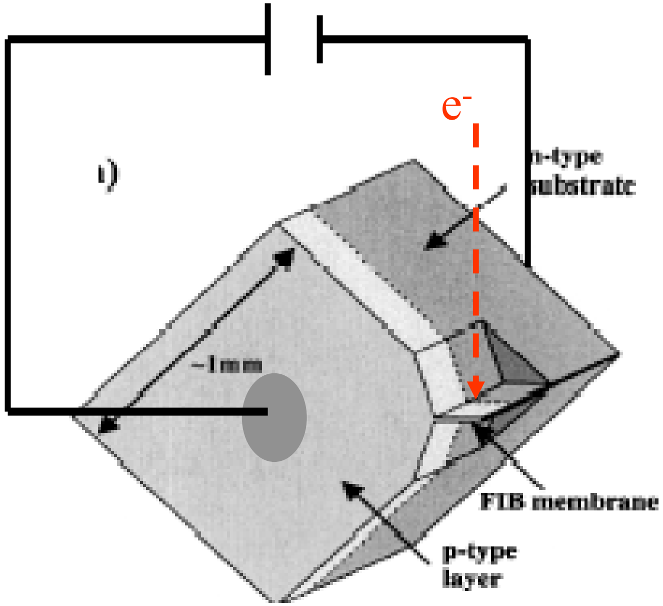
n-MOS and p-MOS transistors

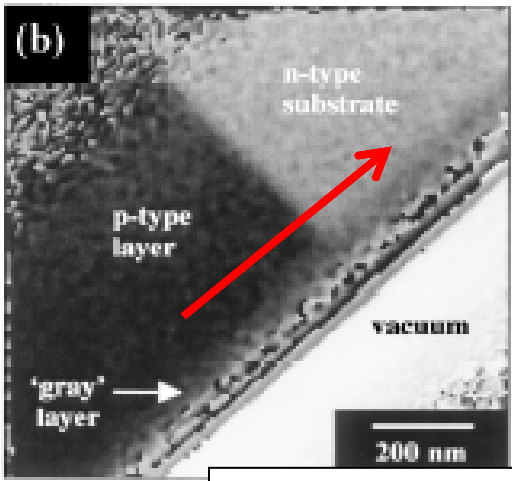
Variation of the electrostatic potentiel across the junction = 0.9 ± 0.12 V

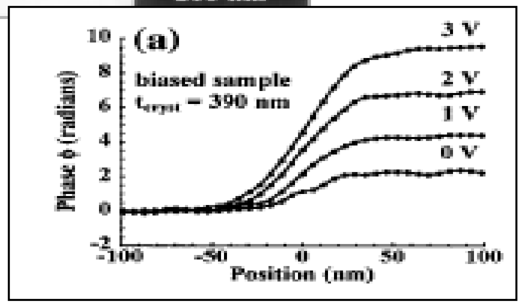
- specimen preparation the key

W.D. Rau, P. Schwander, F.H. Baumann, W. Höppner and A. Ourmazd PRL (1999) ***82****, 2614*

In-situ biasing



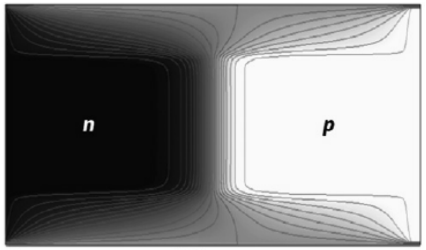




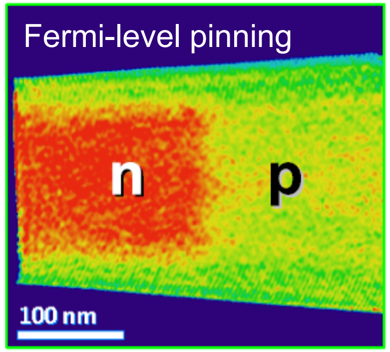
- dead layers

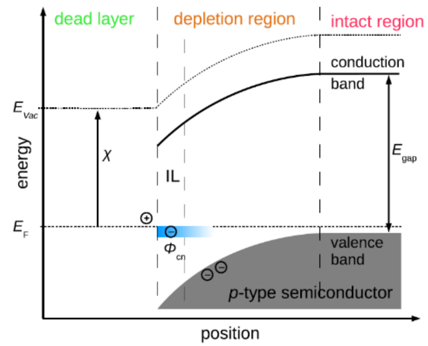
A.C. Twitchett, R.E. Dunin-Borkowski, P.A. Midgley, Phys. Rev. Lett. 88 (2002) 238302

Many Years later



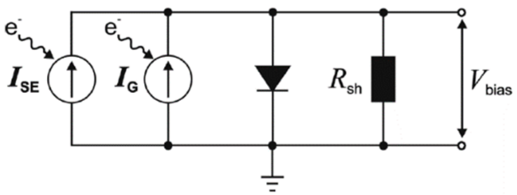
D. Somodi et al. Ultramic 134, 160 (2013)

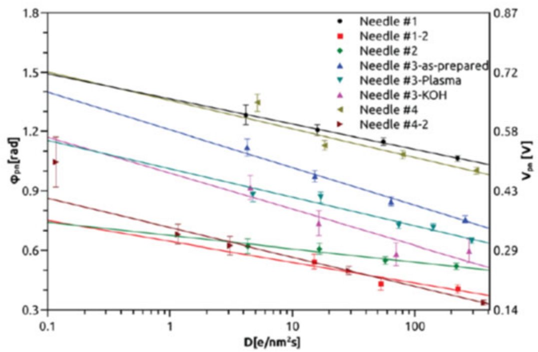




D. Wolf et al. APL 103, 264104 (2013)

p-n junction under e- irradiation





J.B. Park et al. APL 105, 094102 (2014)

- electrostatics
- electrodynamics

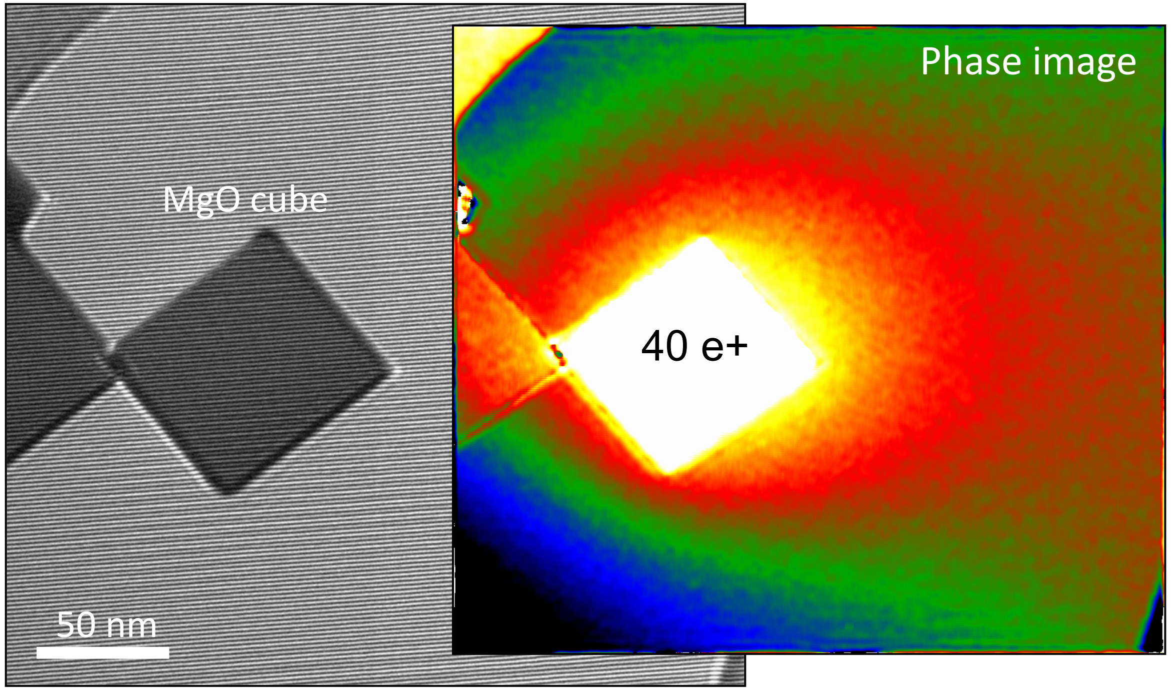
Question to Ask

What technique is best for my material or problem?

What material or problem is best for my technique !

Charge counting

ϕ	u	A	V
$\nabla\phi$	ε	$\mathbf{B}$	$\mathbf{E}$

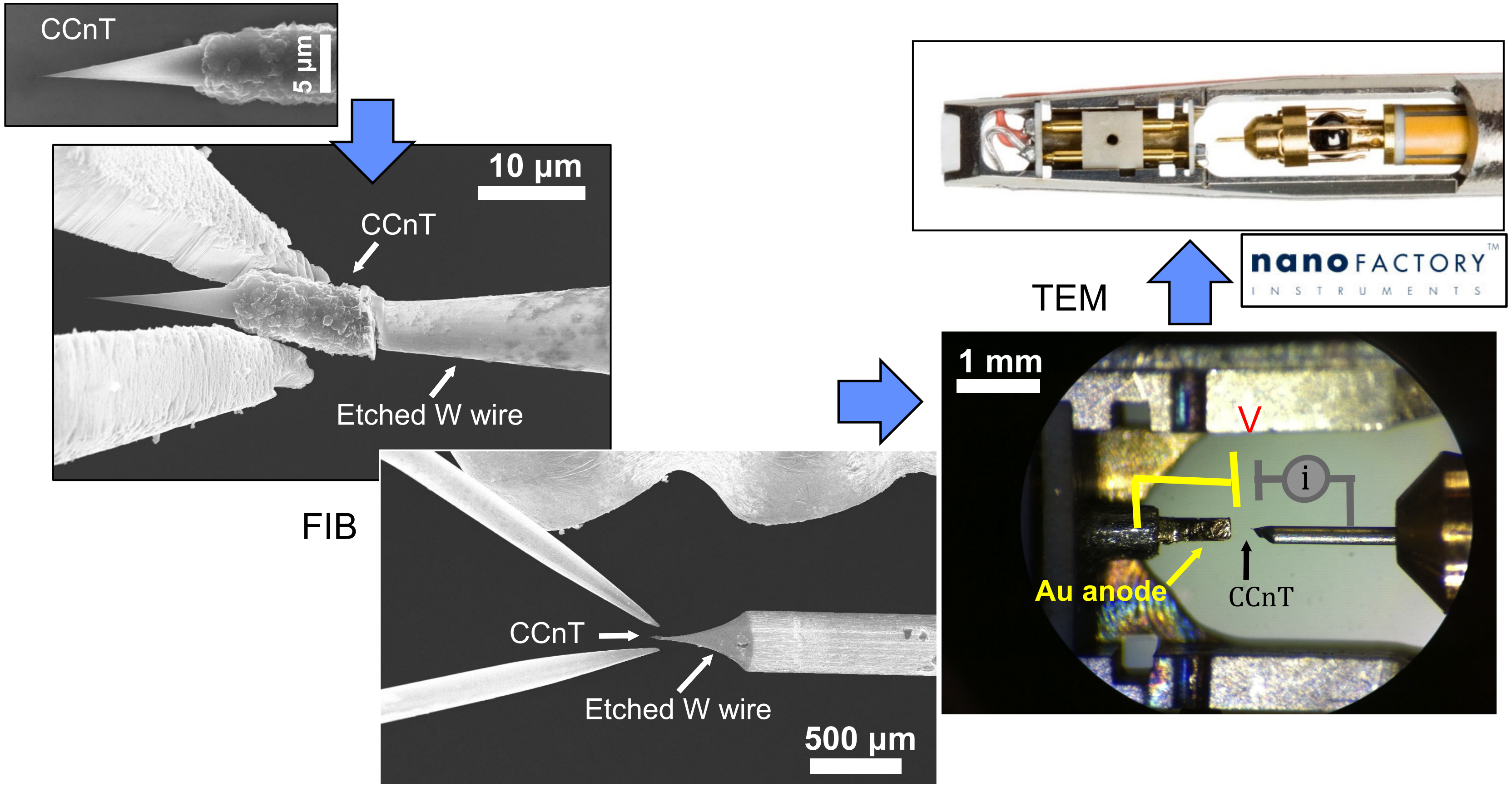


$$Q = \varepsilon_o \oiint_S \boldsymbol{E}.\boldsymbol{dS}$$

- Electric field in the vacuum
  - cube is charged!
- Maxwell's Equations
  - Gauss's Law

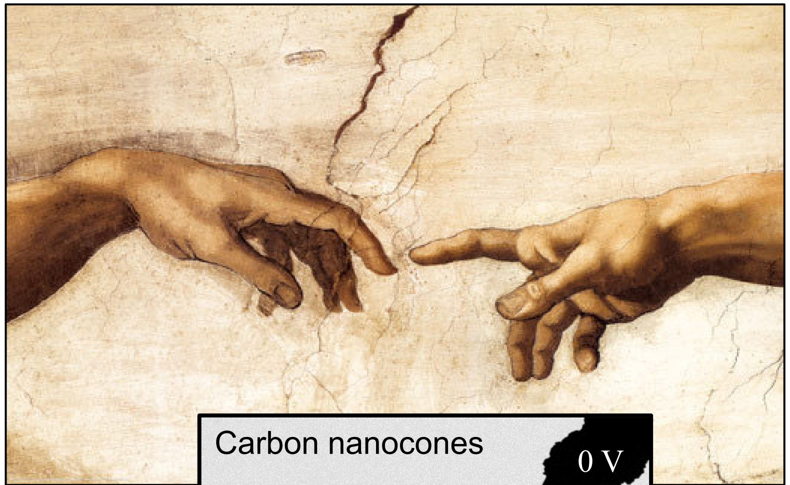
C. Gatel, A. Lubk, G. Pozzi, E. Snoeck, and M.J. Hÿtch, Phys. Rev. Lett. 111, 025501 (2013)

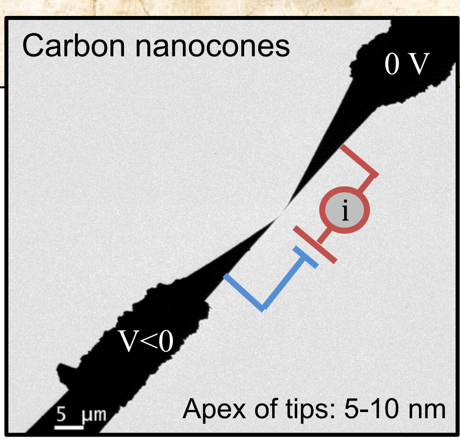
Nanoprobe in-situ biasing

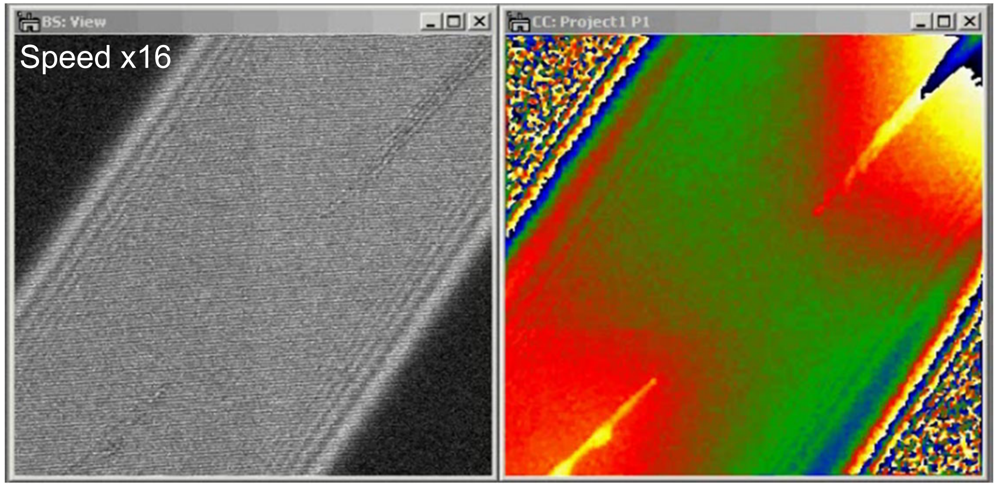


F. Houdellier, A. Masseboeuf, M. Monthioux, M.J. Hÿtch, Carbon 50 (2012) 2037

Michelangelo Experiment







- In situ experiments
  - Biasing the tips between 0 and 85V

L. de Knoop, C. Gatel, F. Houdellier, M. Monthioux, A. Masseboeuf, E. Snoeck, and M.J. Hÿtch, APL 106, 263101 (2015)

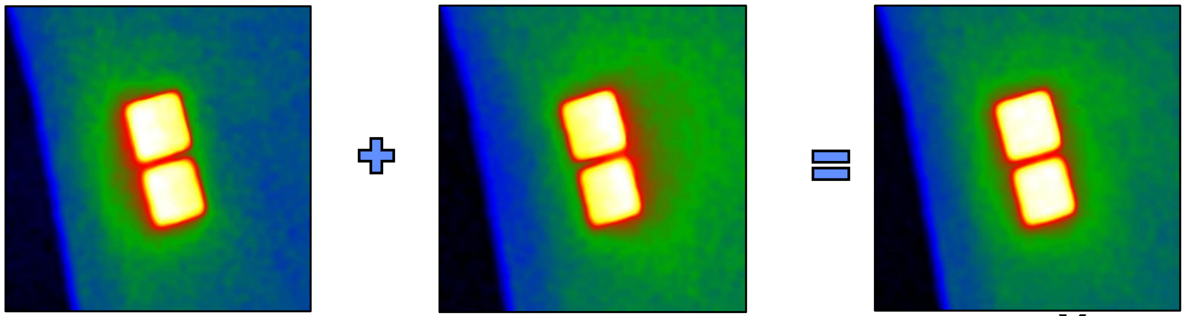
Magnetic Fields

ϕ	u	A	V
$\nabla\phi$	ε	$\mathbf{B}$	$\mathbf{E}$

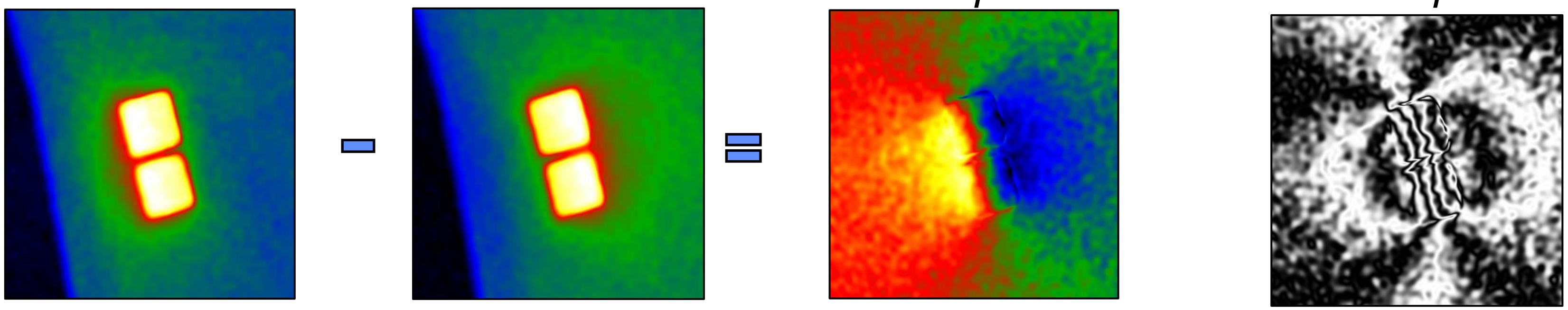
Electric and Magnetic Phase

$$\boxed{\phi = \phi^E + \phi^M}$$

$\phi_a = \phi^E + \phi^M$ $\qquad$ $\phi_b = \phi^E - \phi^M$ $\qquad$ $2\phi^E$



$2\phi^M$ $\qquad$ $\cos 2\phi^M$

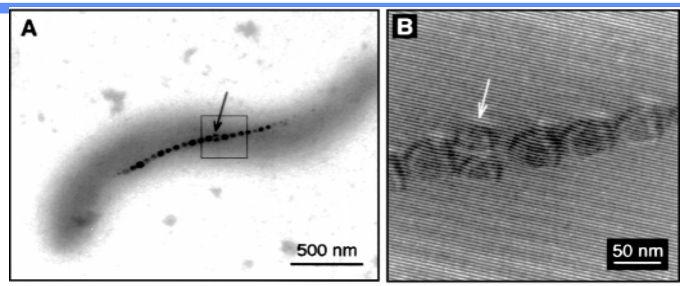


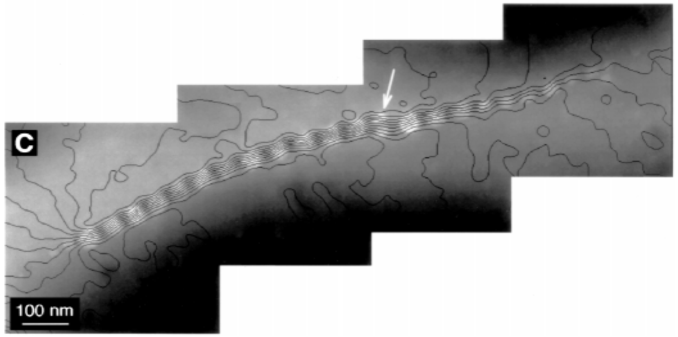
Christophe Gatel, Martin Hÿtch and Kazuo Ishizuka qHolo software (HREM Research)

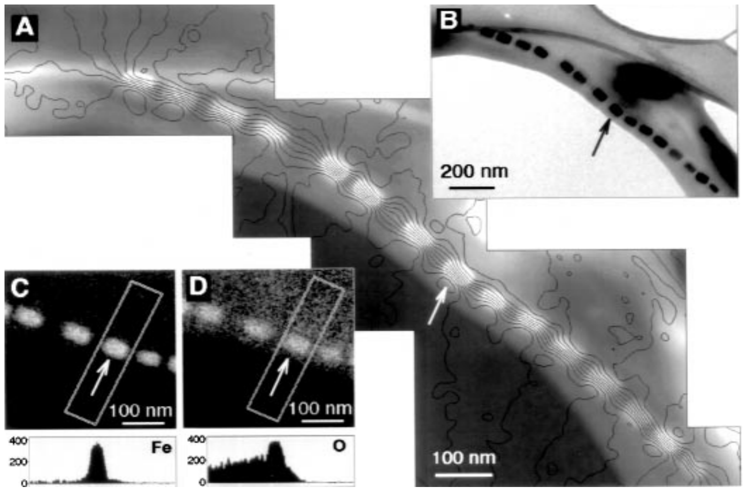
Magnetotactic bacteria

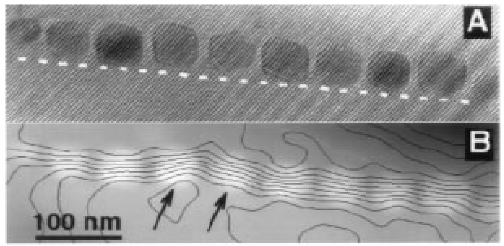
Magnetic Microstructure of Magnetotactic Bacteria by Electron Holography

Rafal E. Dunin-Borkowski, Martha R. McCartney, Richard B. Frankel, Dennis A. Bazylinski, Mihály Pósfai,* Peter R. Buseck



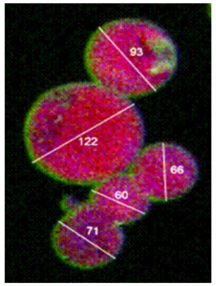


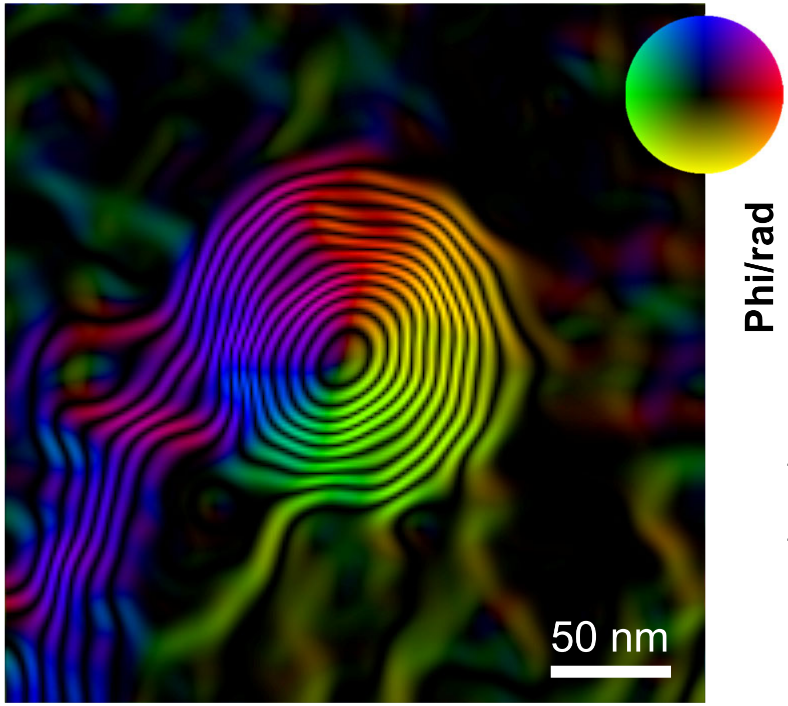




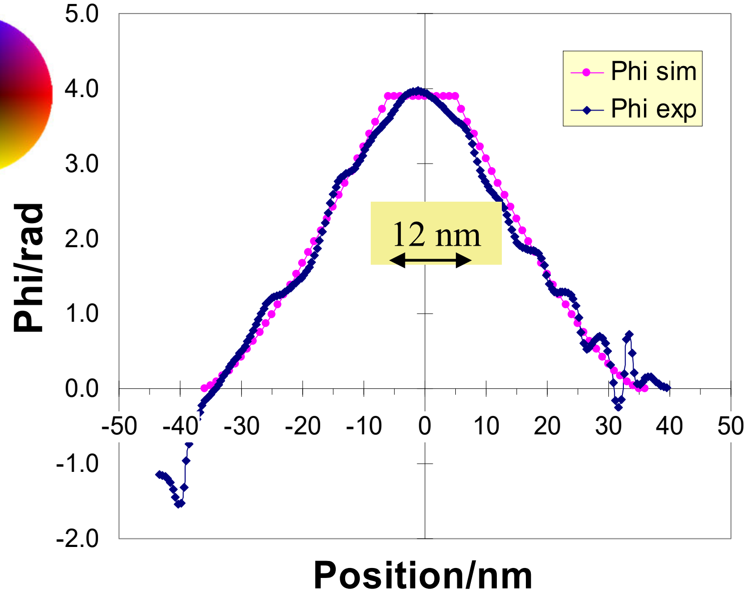
R. E. Dunin-Borkowski et al, Science 282, 1868 (1998)

Vortex cores





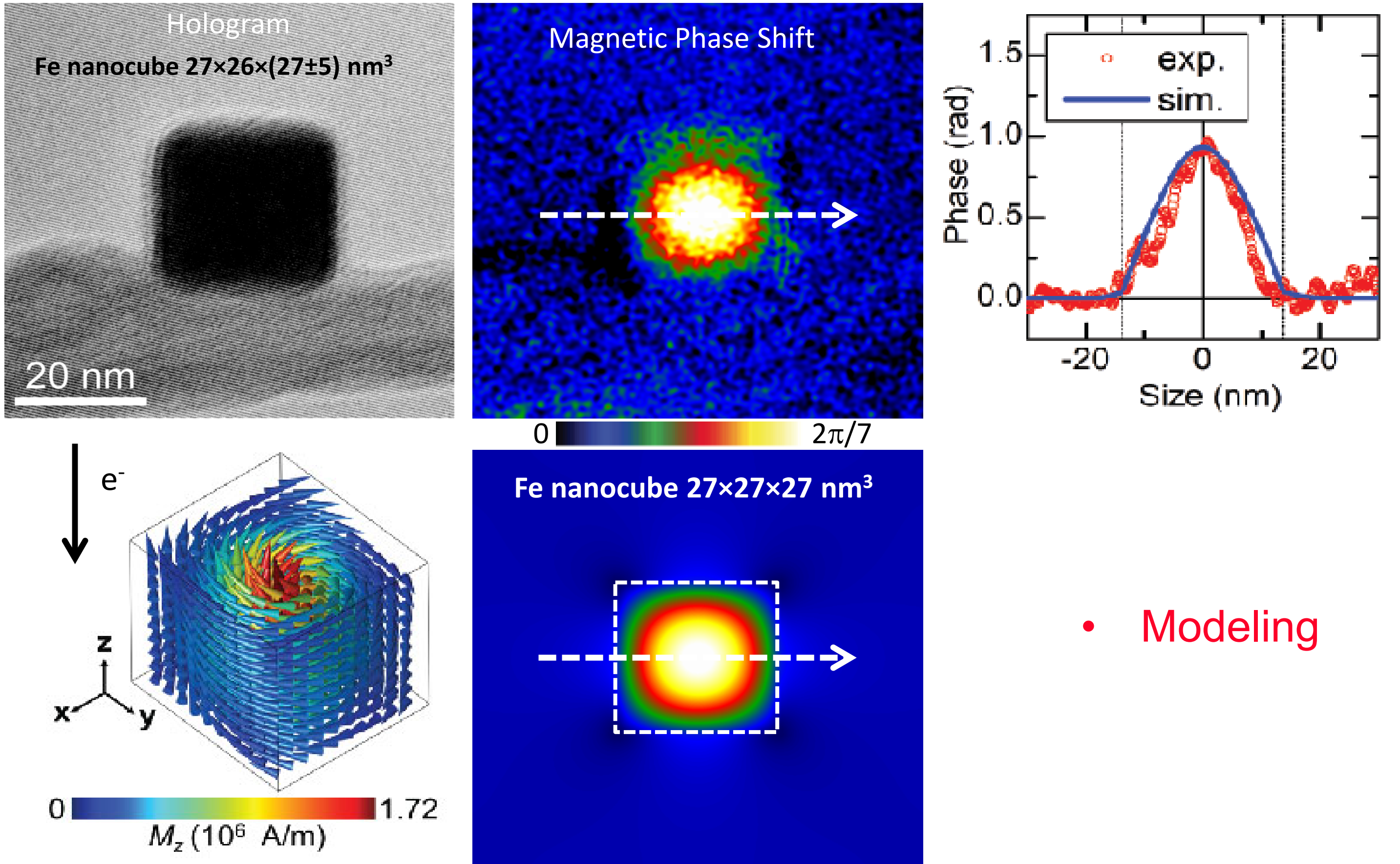
FeNi nanoparticles



- measure the vortex core size
- compare with theory

M. J. Hÿtch et al., Phys. Rev. Lett 91, 257207 (2003)

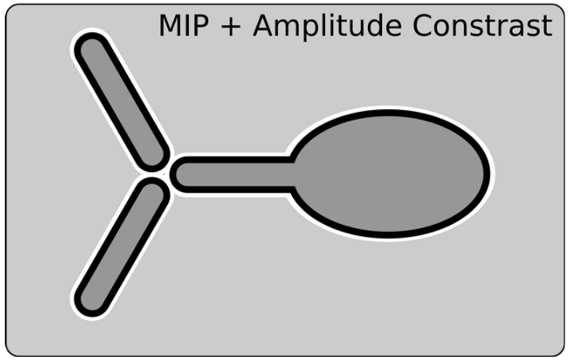
Fe nanoparticles



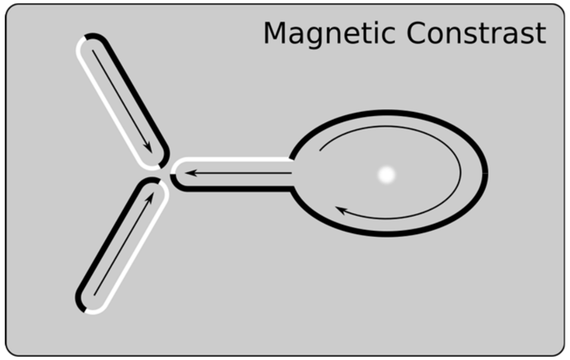
- Modeling

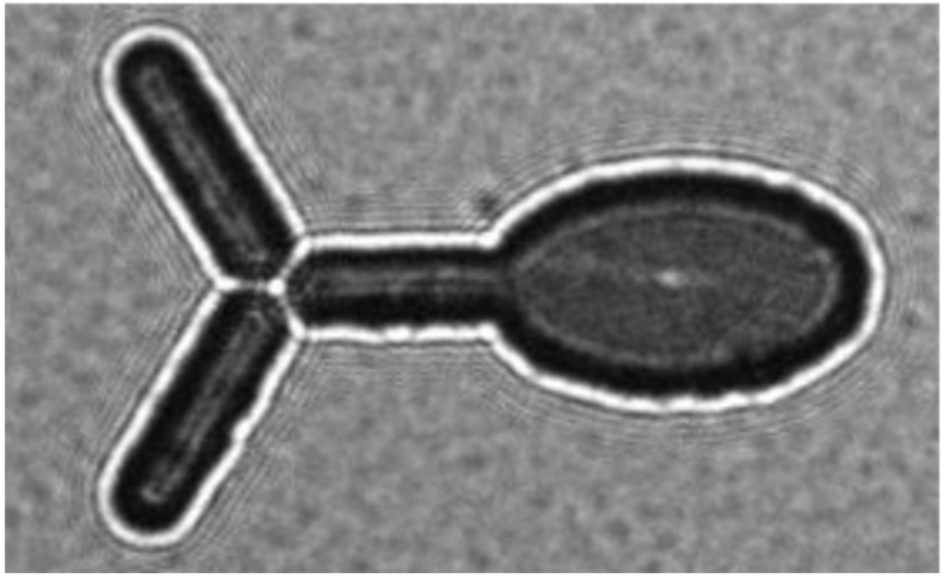
C. Gatel et al., Nano Letters 15, 6952 (2015)

Beam me up, Régis



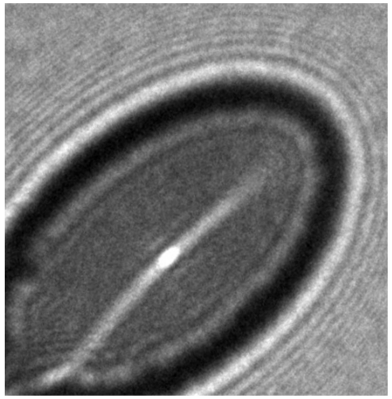
+



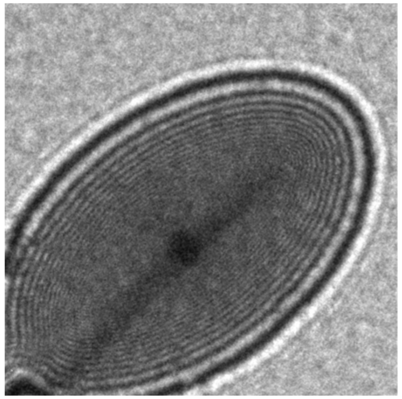


Courtesy of Aurélien Masseboeuf

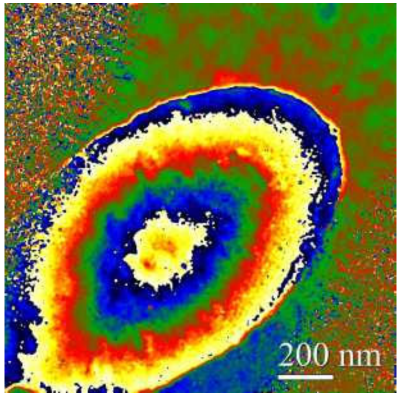
Magnetic holography



Fresnel - Under-Focused
(Greyscale)

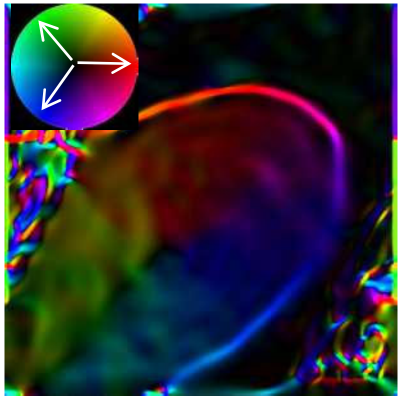


Fresnel - Over-Focused
(Greyscale)

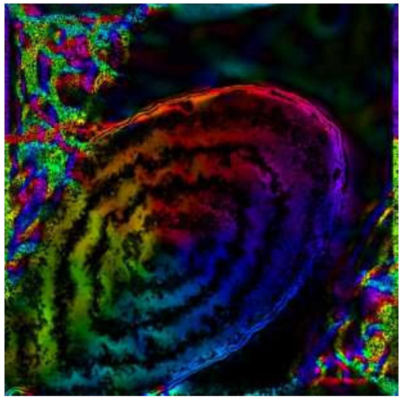


Holography - Phase (raw)
(temperature scale)

Phase Cosine
(iso-phase <=> induction flux)

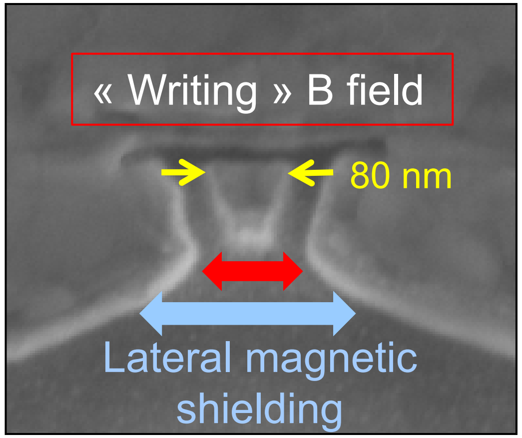


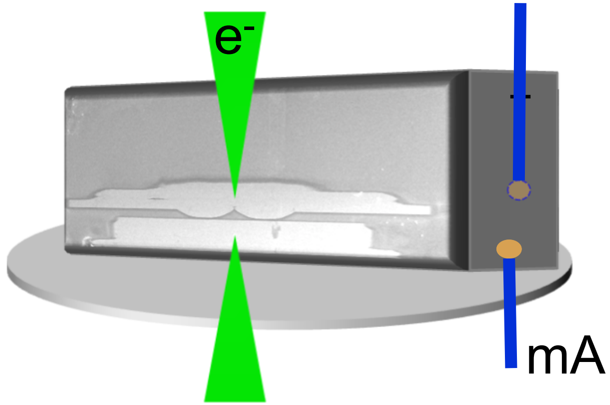
Induction Map
(Phase gradients - Colour wheel)

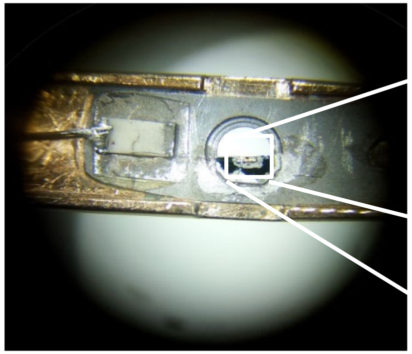


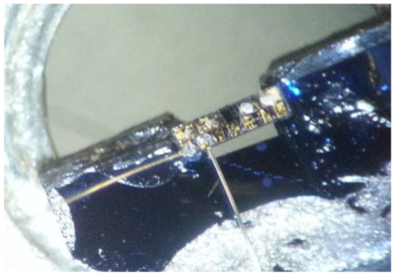
Composite Map
(Phase gradients x Contour)

Operando experiments





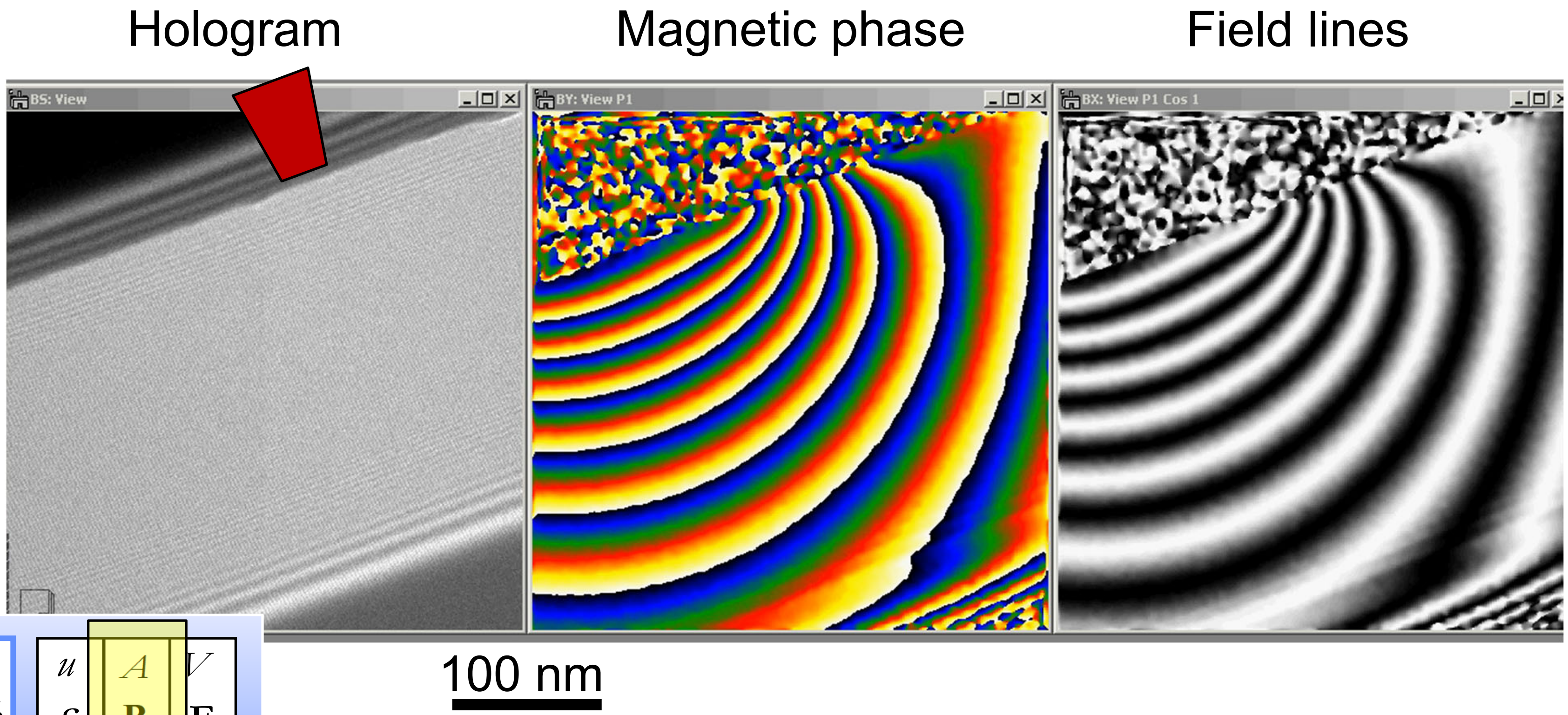


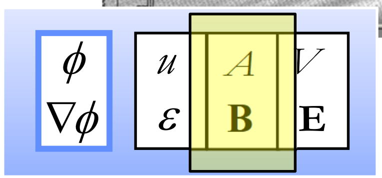


- Heroic experiment

J. F. Einsle, C. Gatel, A. Masseboeuf, R. Cours, M. A. Bashir, M. Gubbins, R. M. Bowman and E. Snoeck, Nano Research 8, 1241 (2015).

Movie

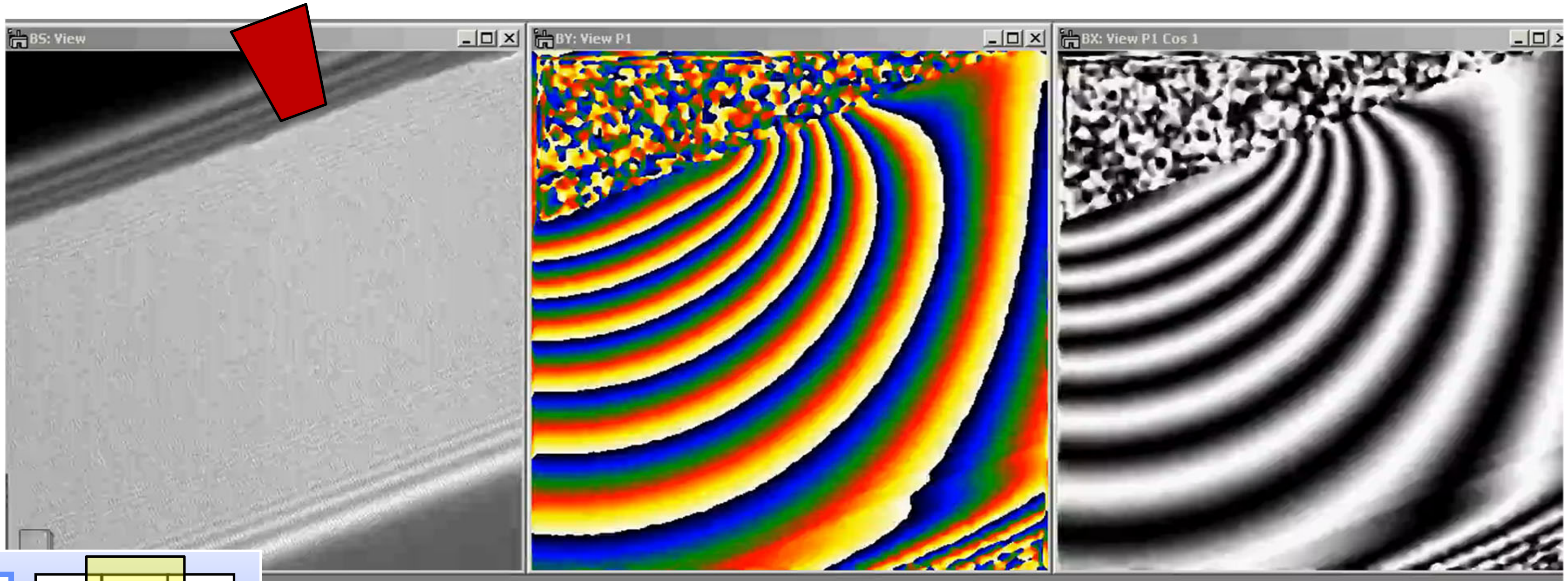


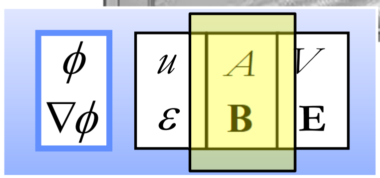


Christophe Gatel & Martin Hÿtch, Holo Live! (HREM Research Inc.)

Movie x3

Hologram Magnetic phase Field lines





100 nm

Christophe Gatel & Martin Hÿtch, Holo Live! (HREM Research Inc.)

Strain Fields

ϕ	u	A	V
$\nabla\phi$	ε	$\mathbf{B}$	$\mathbf{E}$

Geometric phase

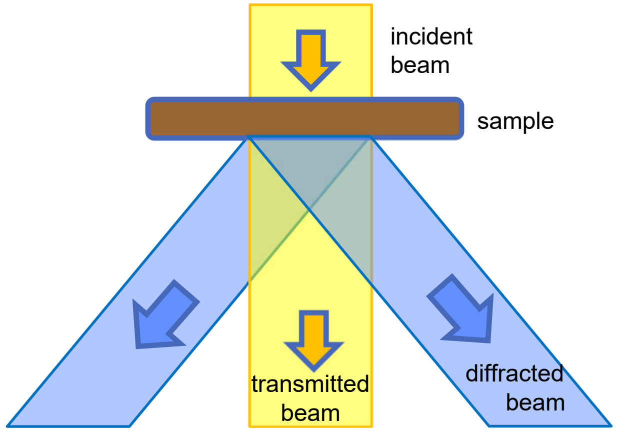
wave function

$$\psi(\mathbf{r}) = \sum_g \tilde{\psi}_g(\mathbf{r}) exp\{2\pi i \mathbf{g.r}\}$$

diffracted beams

$\mathbf{r} \rightarrow \mathbf{r} - \mathbf{u}$ displacement

$$\Rightarrow \tilde{\psi}_g \rightarrow \tilde{\psi}_g e^{-2\pi i \mathbf{g.u}}$$



geometric phase

$$\boxed{\phi_g^G = -2\pi \mathbf{g.u}}$$

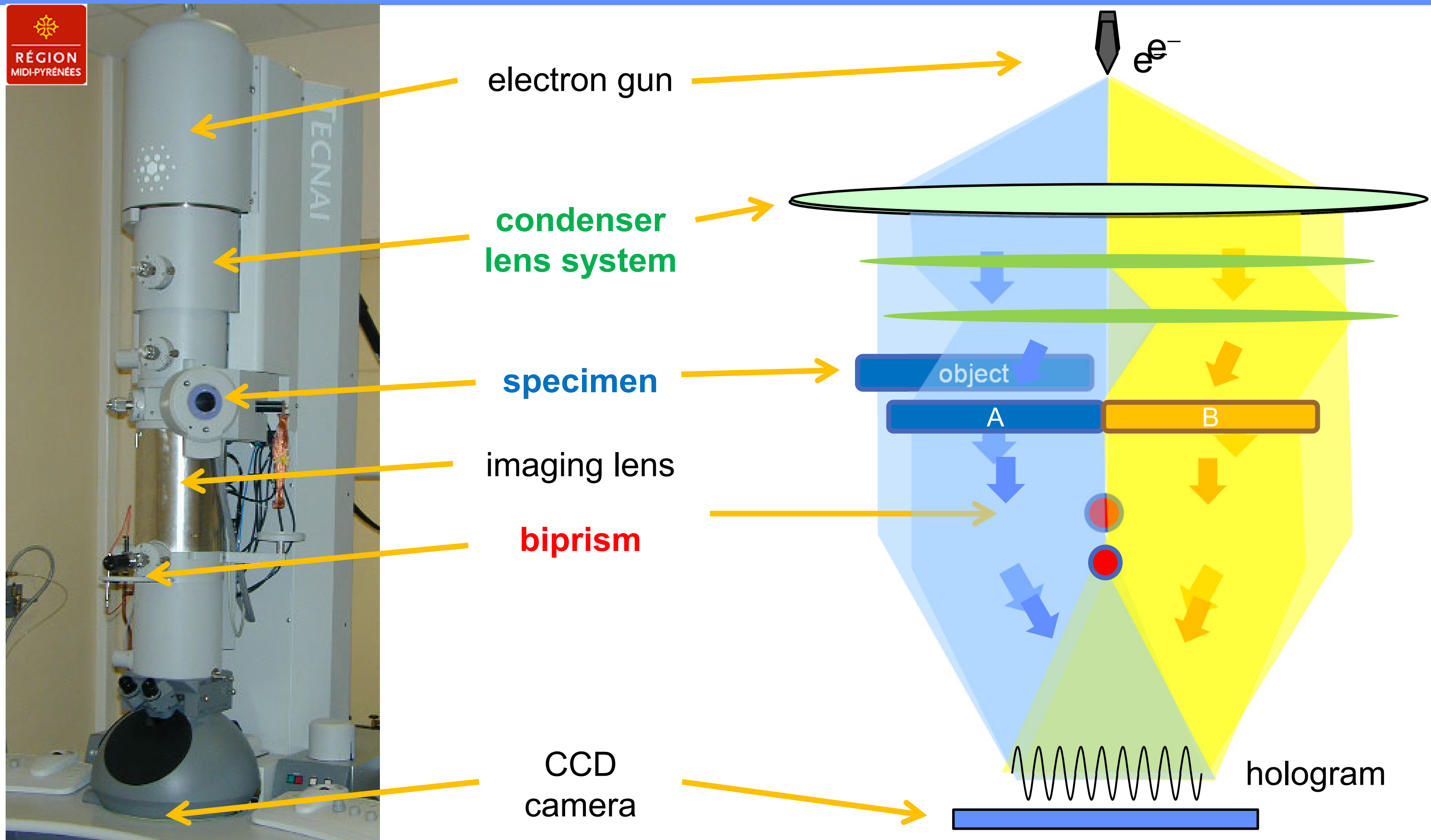
displacement

$\Rightarrow$ ϕ^G NO ϕ^G ϕ^G

$$\Rightarrow \boxed{\varepsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)}$$

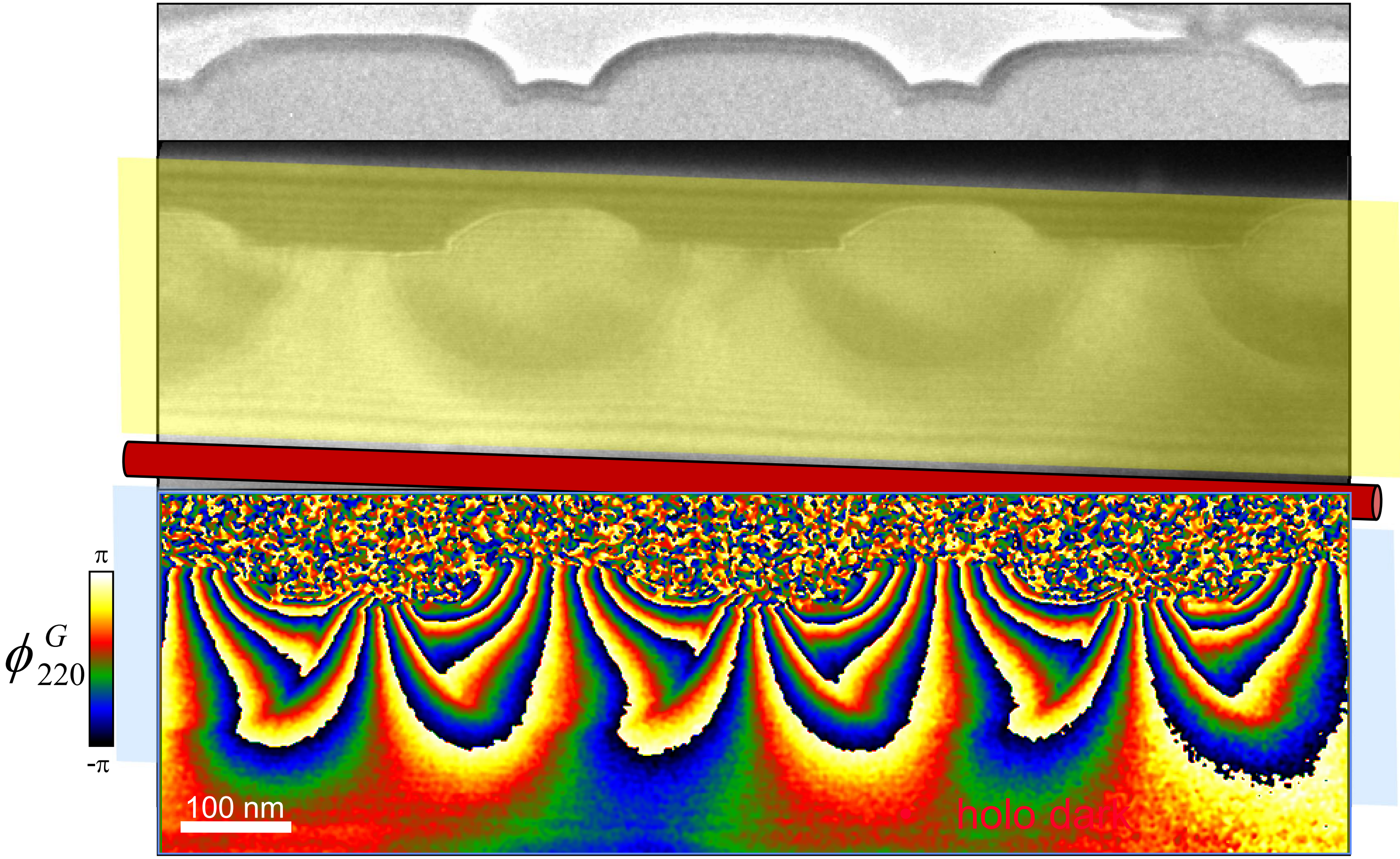
strain

Dark-Field Electron Holography

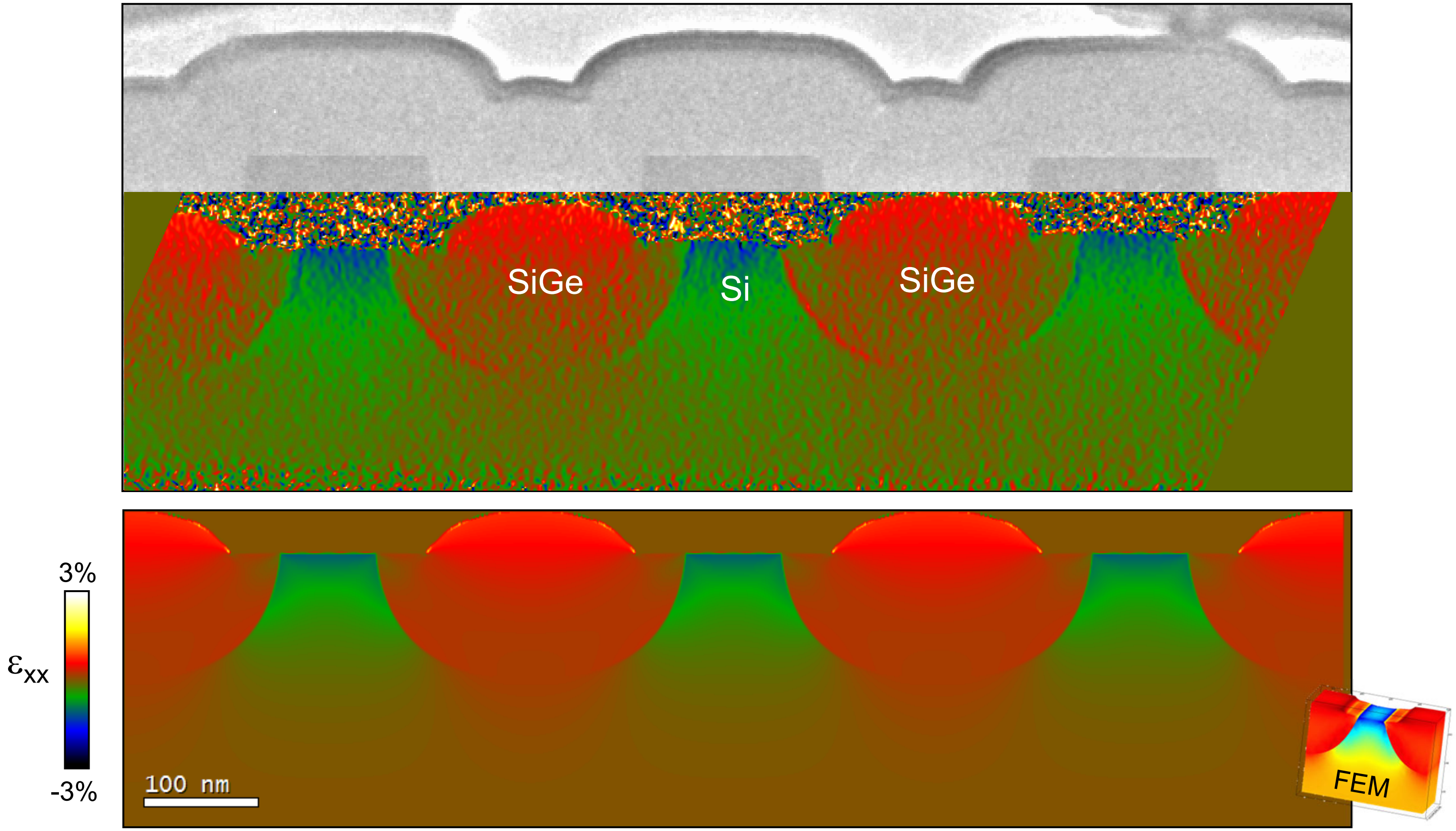


K.J. Hanszen J. Phys. D 19 (1986) 373

Experiment

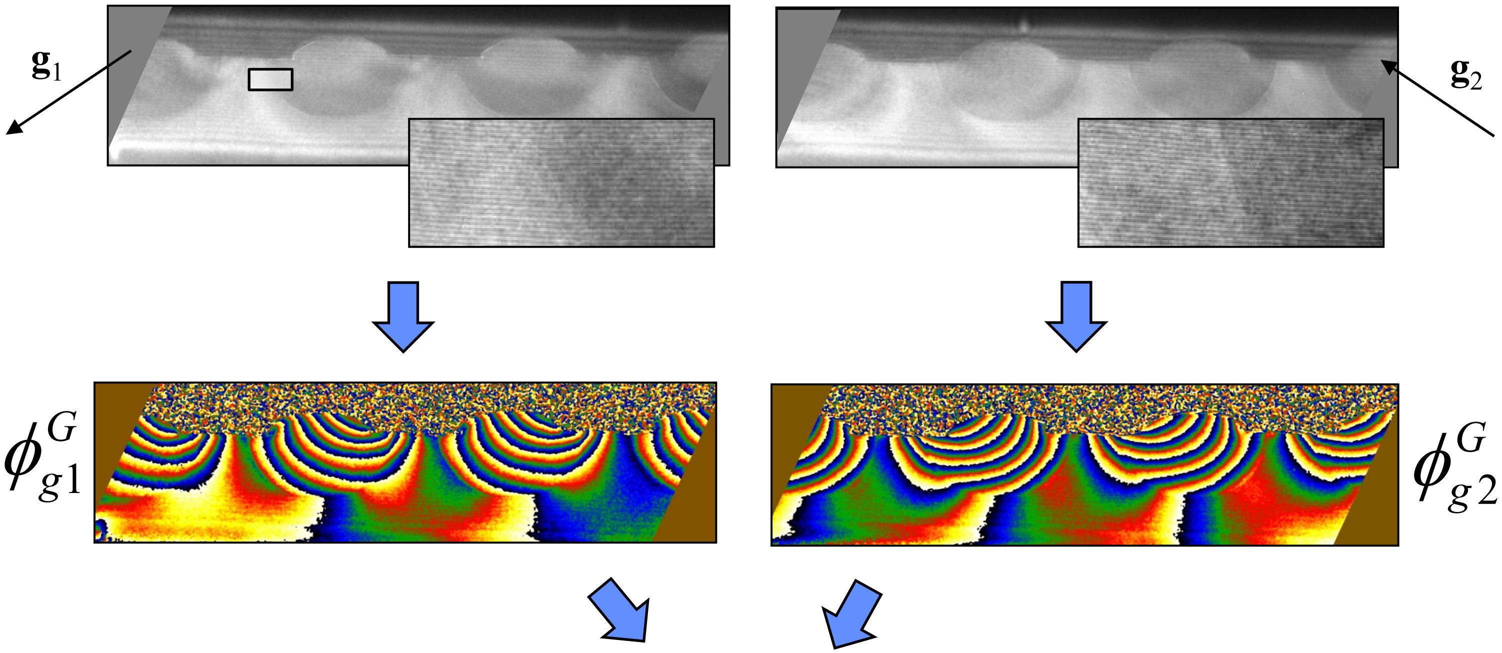


Strained-Si p-MOSFET



M.J. Hÿtch, F. Houdellier, F. Hüe & E. Snoeck, Nature 453 (19th June 2008) 1086

2D Deformation

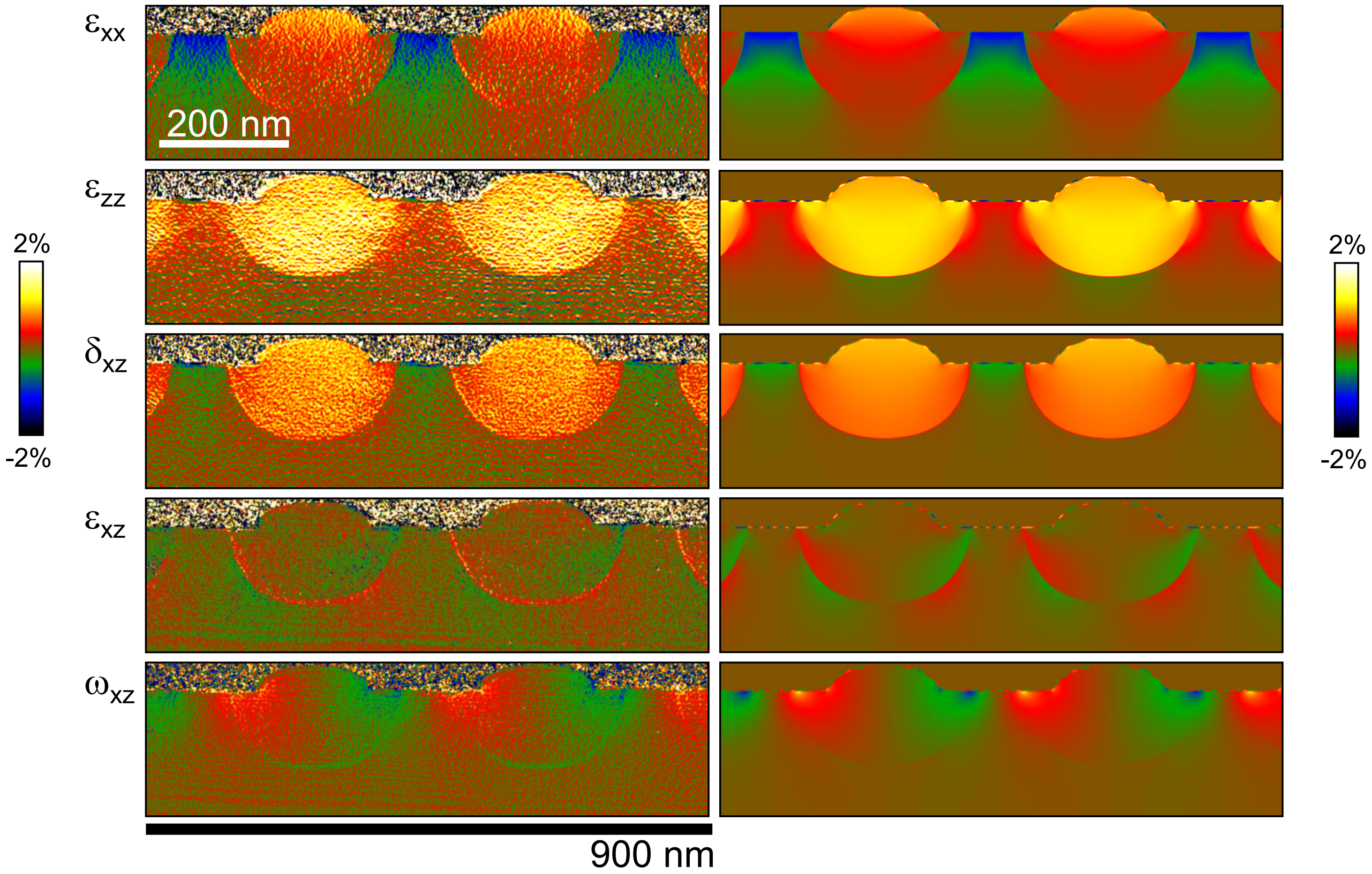


$$\mathbf{u}(\mathbf{r}) = -\frac{1}{2\pi}\left[\phi_{g1}^G(\mathbf{r})\mathbf{a}_1 + \phi_{g2}^G(\mathbf{r})\mathbf{a}_2\right]$$

- as with GPA

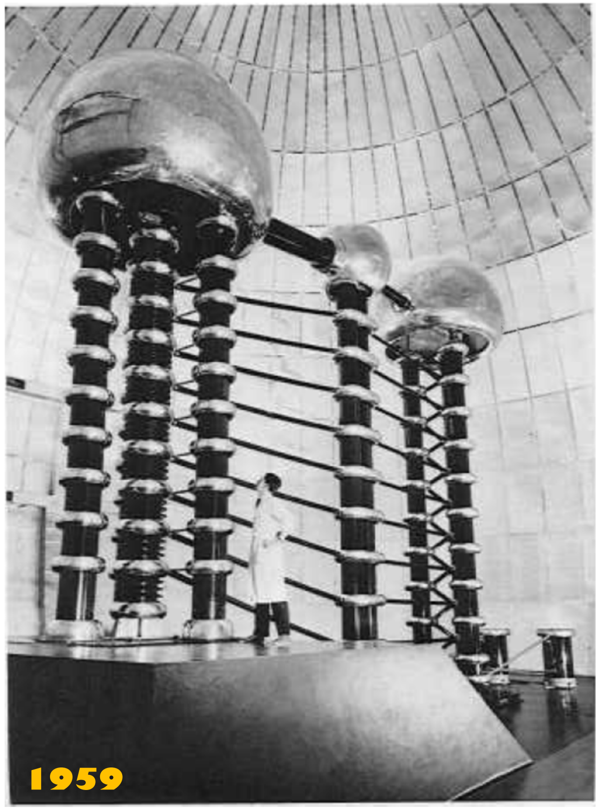
Martin Hÿtch, Christophe Gatel and Kazuo Ishizuka HoloDark software (HREM Research)

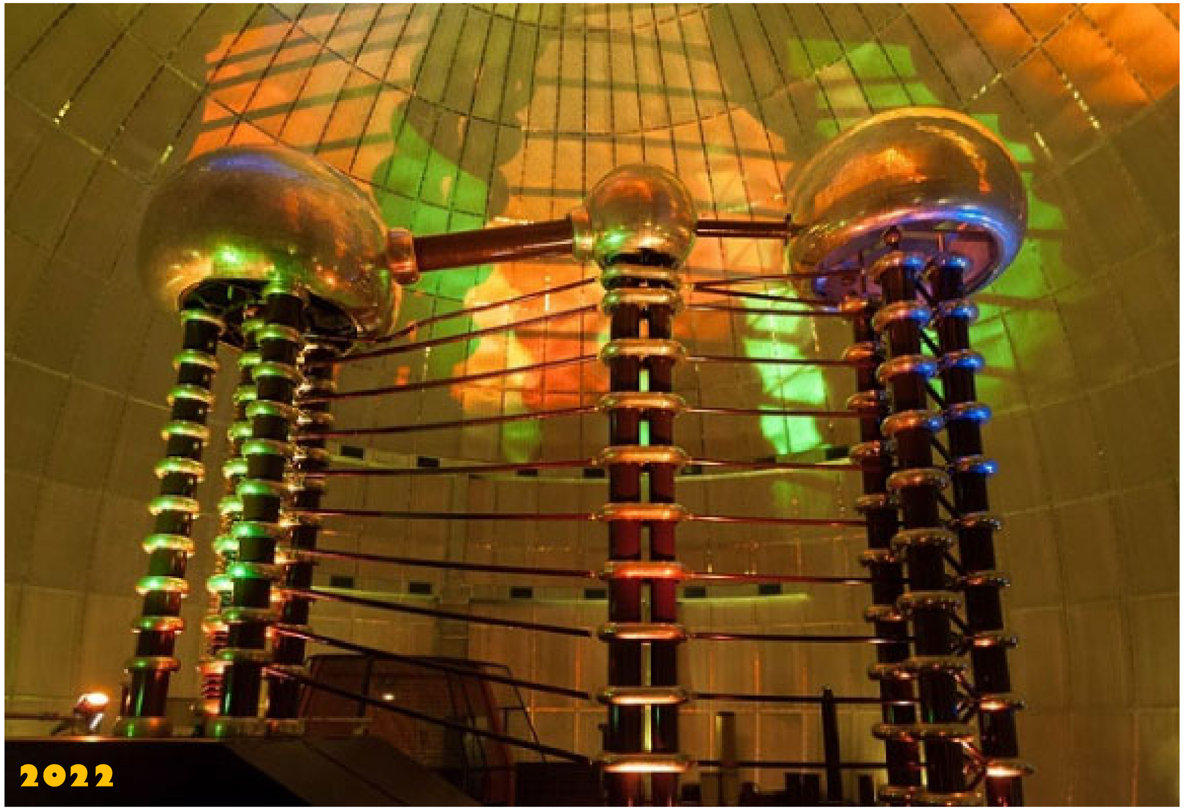
Strain components




Back to La Boule

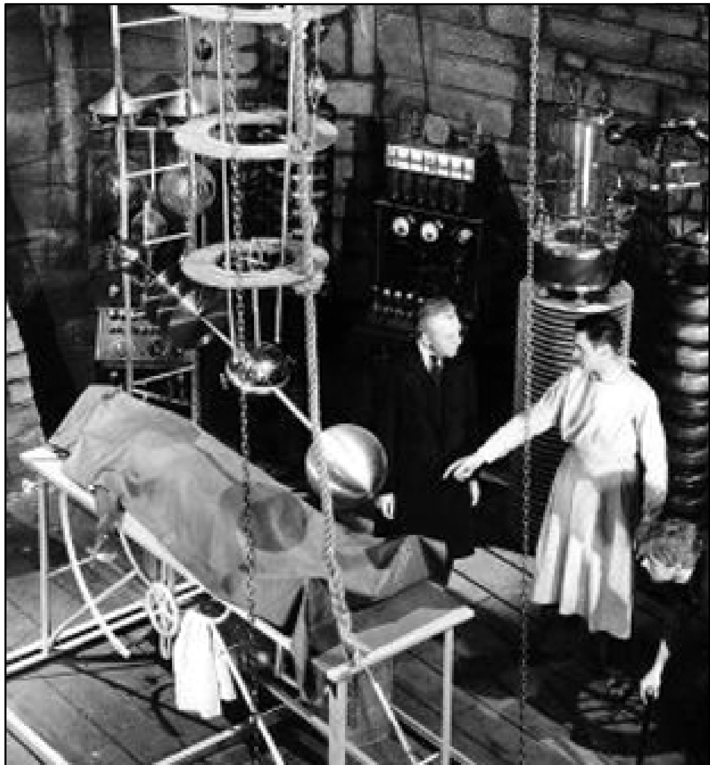
Inside « La Boule »



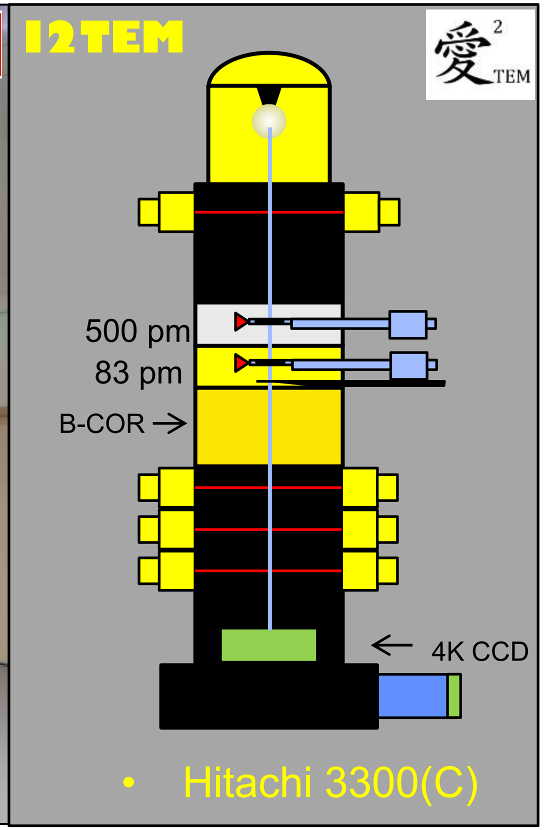


- 1 MeV electron accelerator, in free air

Downstairs « La Boule »



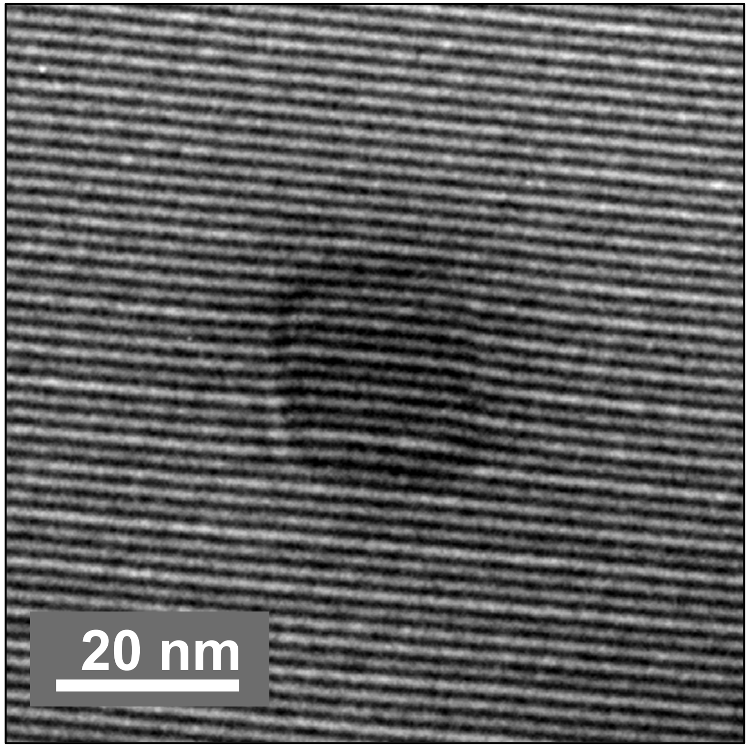
J. Whale & M. Shelley (1931)



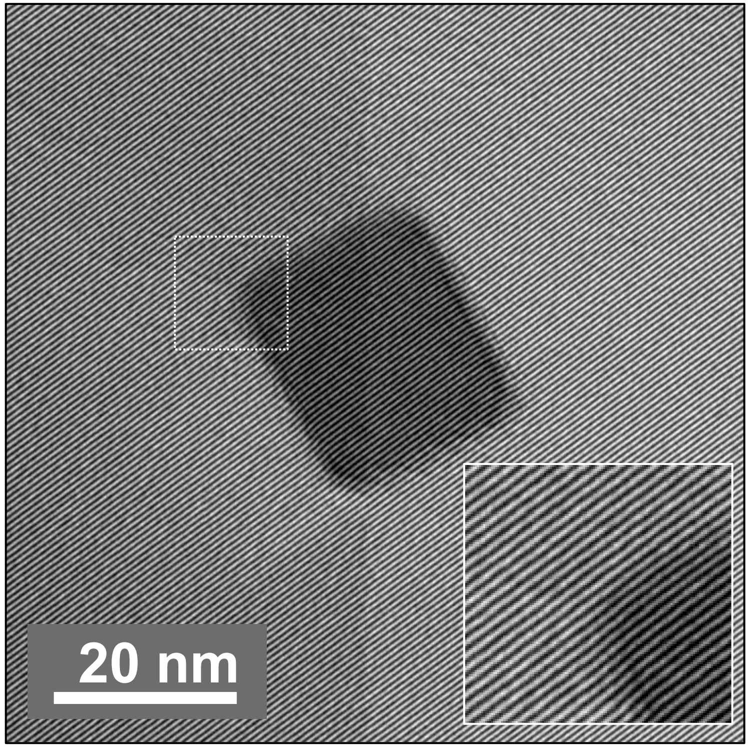
- Powerful and unique electron microscopes

New Instrumentation

SACTEM

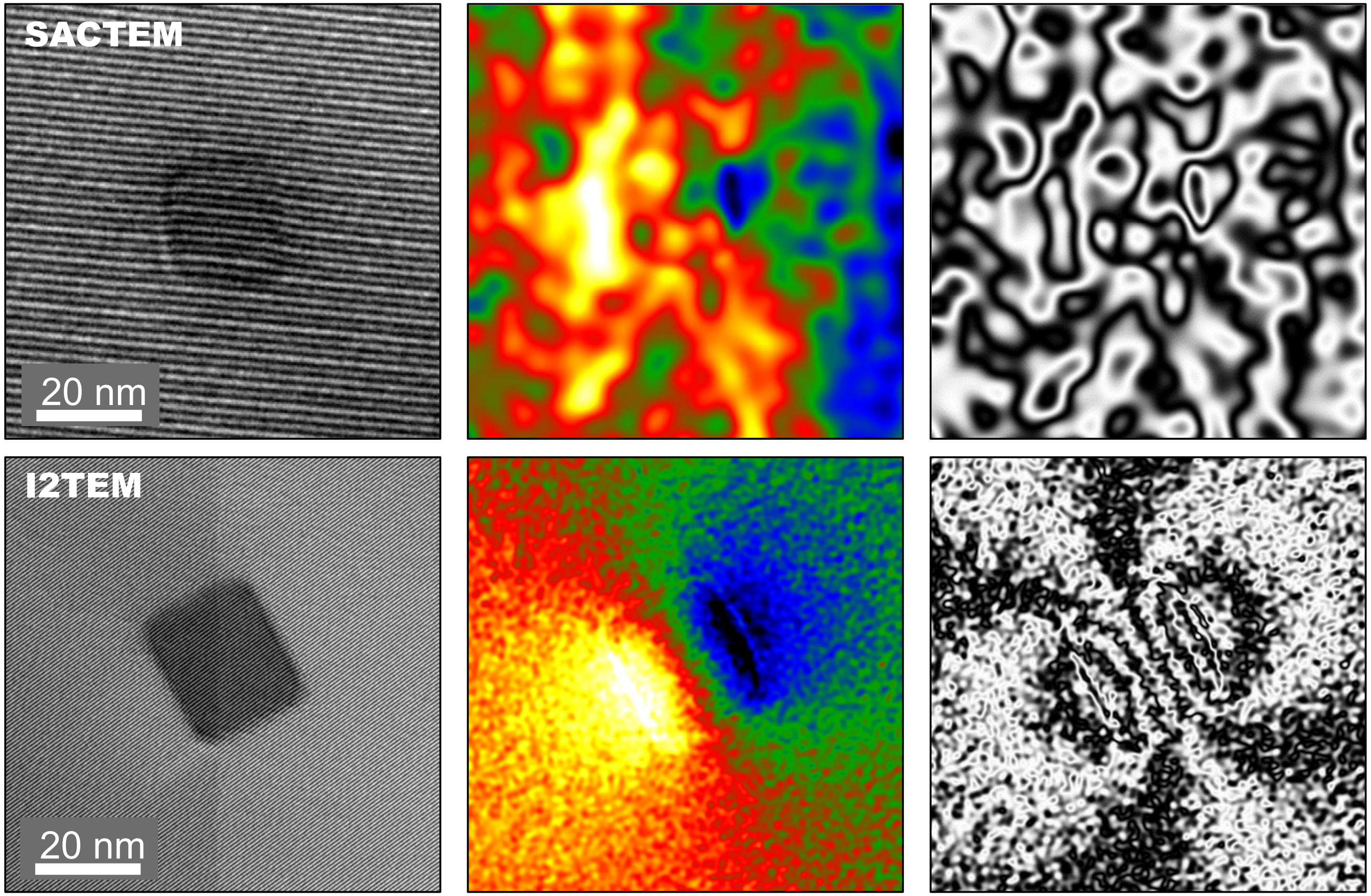


I2TEM

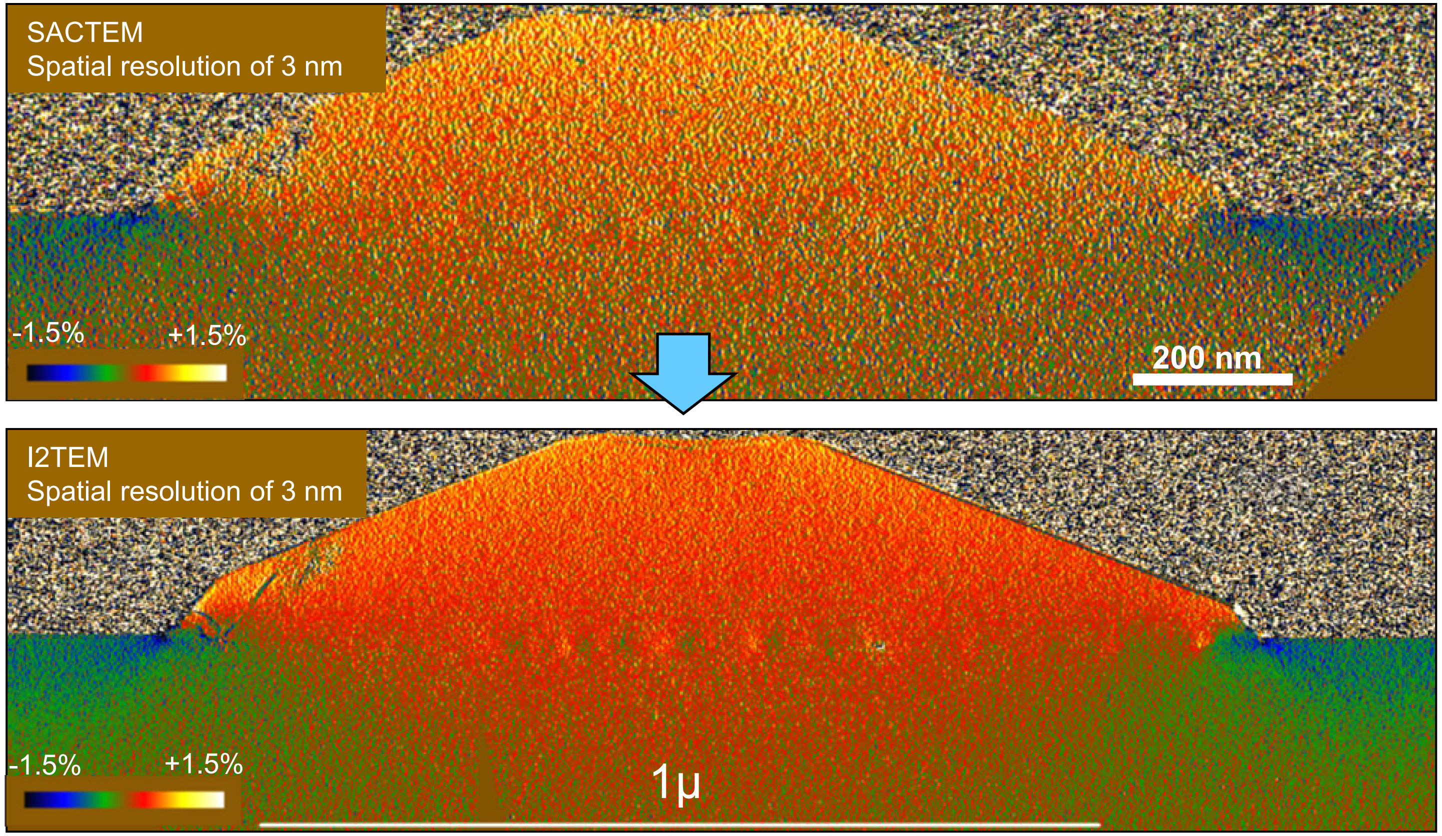


L.-M. Lacroix et al., Nano Letters 12, 3245−3250 (2012)

Magnetic phase: nc-Fe



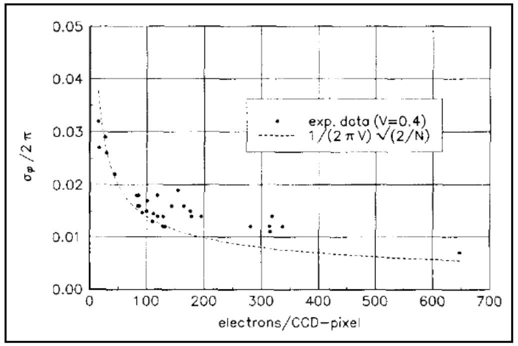
Stain mapping with DFEH

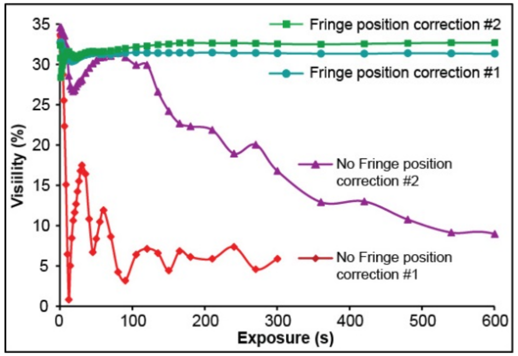


Courtesy of Nikolay Cherkashin

Longer exposure times

More electrons
better phase sensitivity





$$\sigma_\phi \propto \frac{1}{V}\sqrt{\frac{1}{N_e}}$$

Visibility Dose

Instabilities
reduce visibility

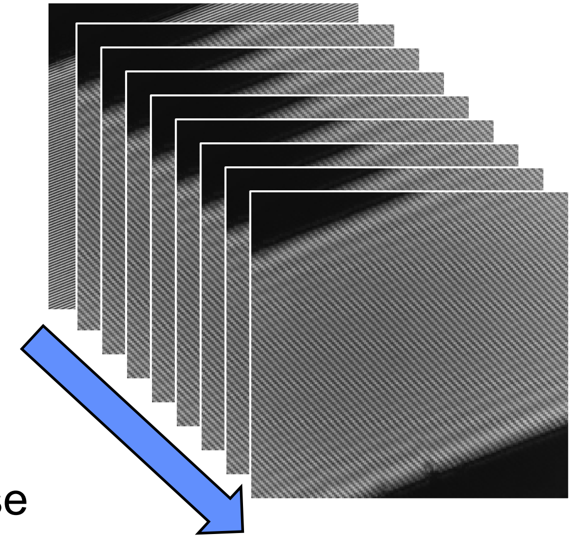


Image stacks
- short exposure times
- many images

A. Harscher & H. Lichte
Ultramicroscopy 64, 57-66 (1996)

E. Voelkl & D. Tang
Ultramicroscopy 110, 447–459 (2010)

Automation: Stabilisation

CEMES - Toulouse

C. Gatel, J. Dupuy, F. Houdellier, M.J. Hÿtch, Applied Physics Letters 113, 133102 (2018)

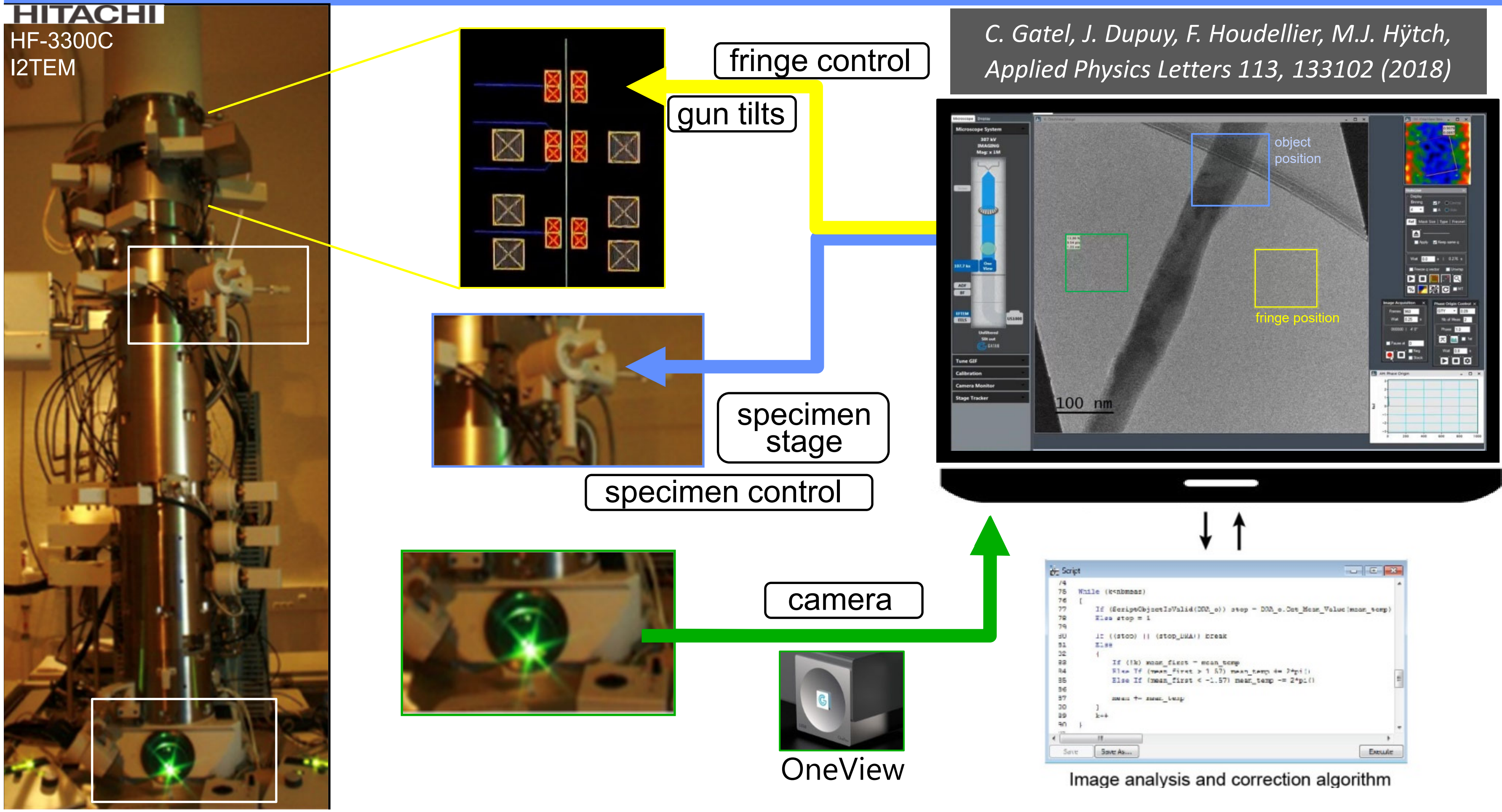
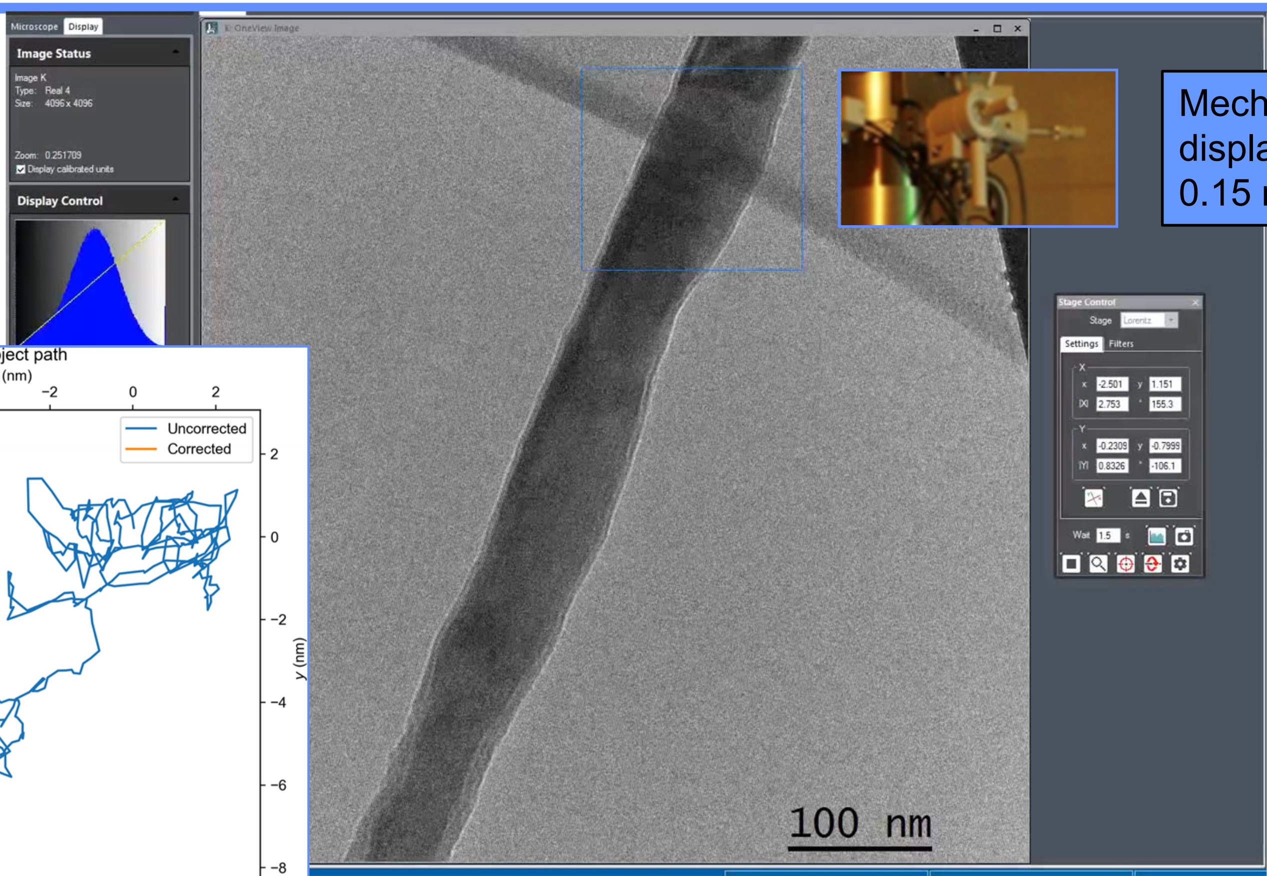
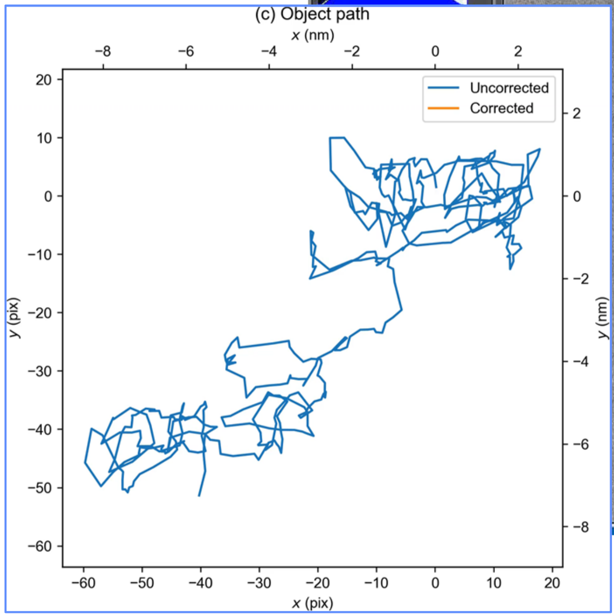


Image analysis and correction algorithm

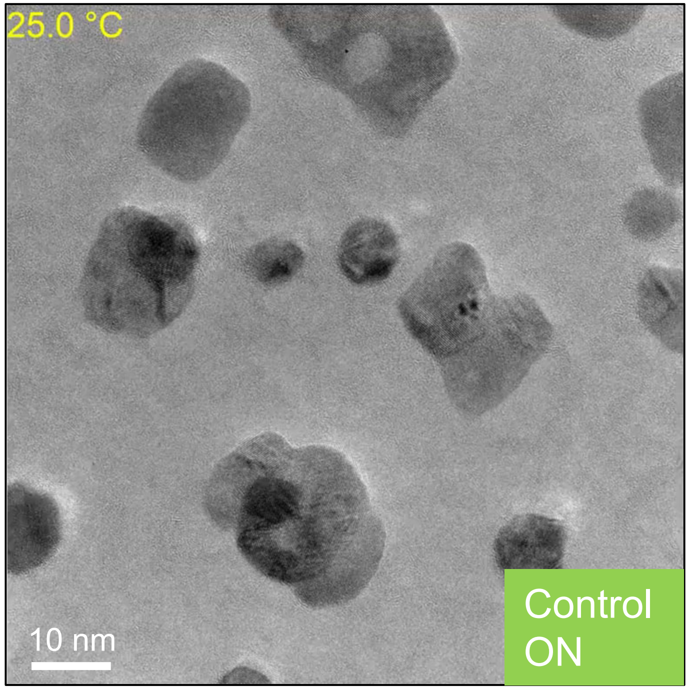
Stage control

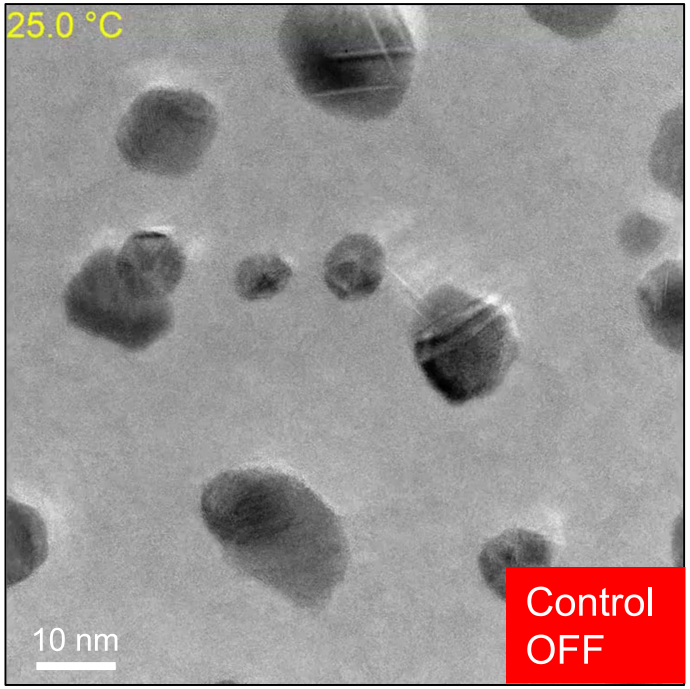


Mechanical displacements of 0.15 nm



Specimen drift control

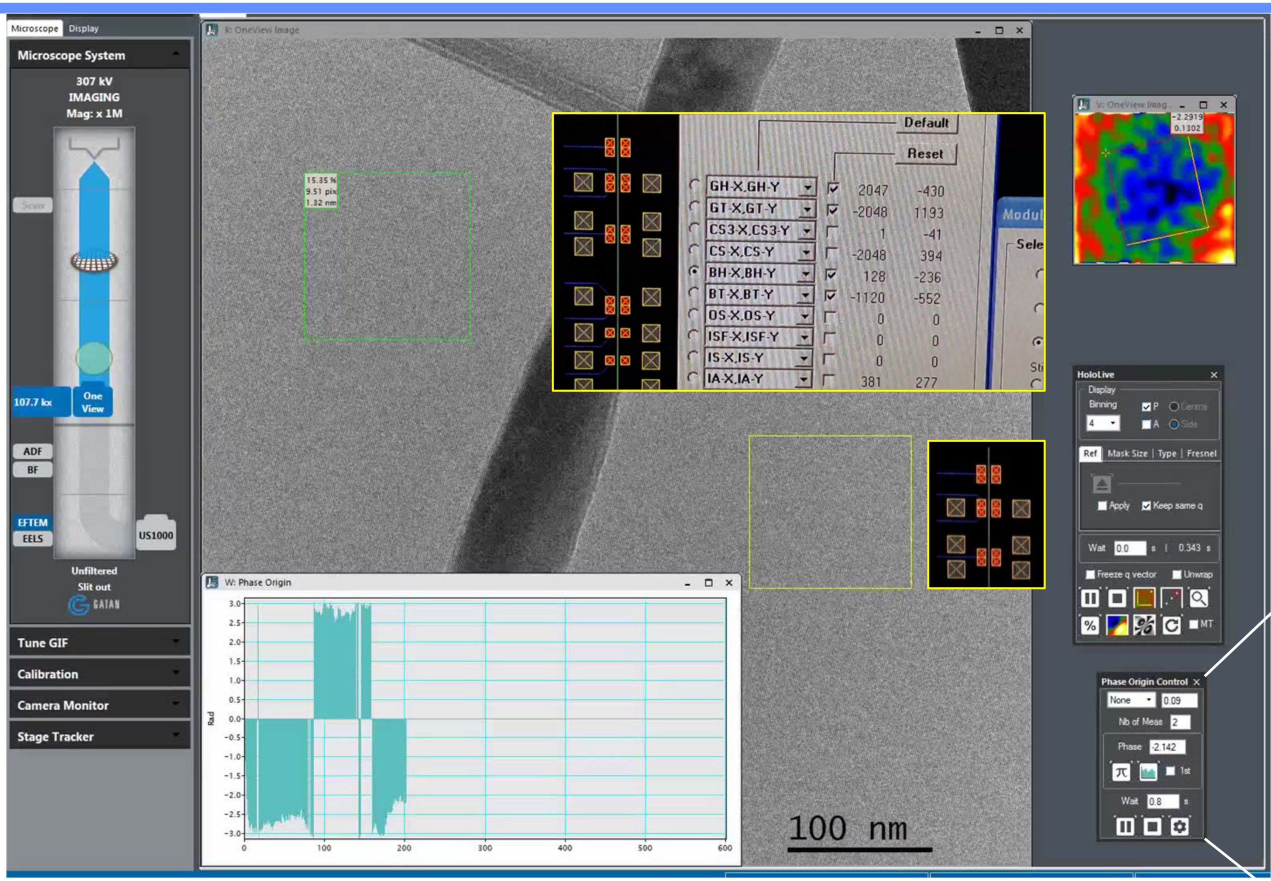




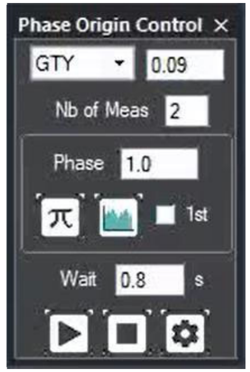
JEOL ARM 200

Collaboration C. Genevois and C. Bouillet, Platform MACLE Centre Val de Loire, Orléans

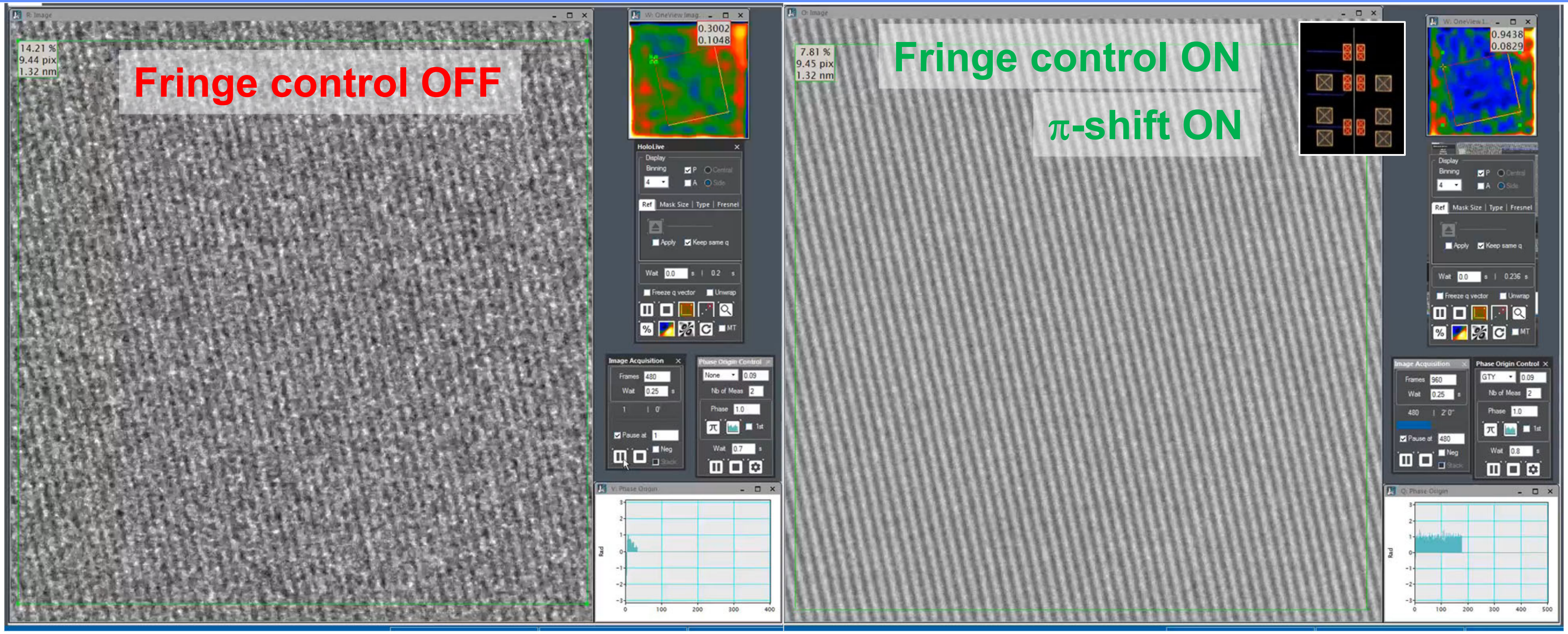
Fringe Control

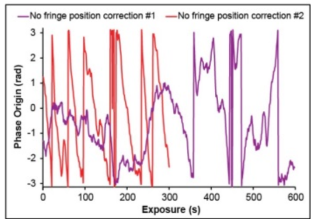


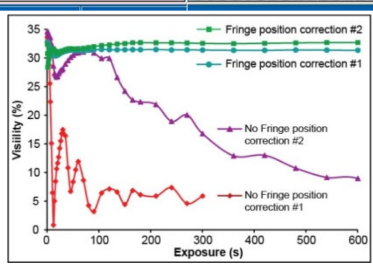
beam tilts of 0.13 µrads

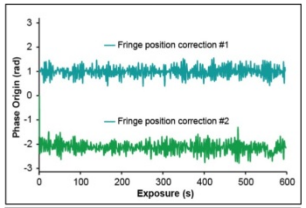


Fringe Control



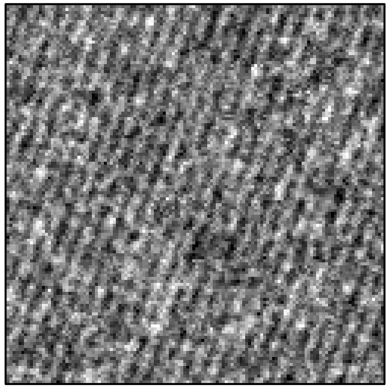




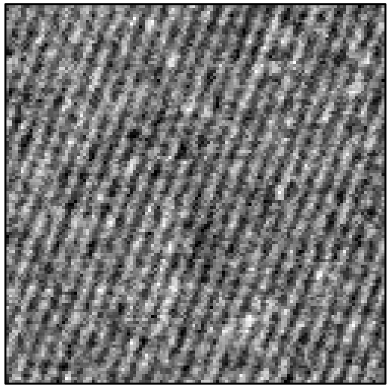


Time series

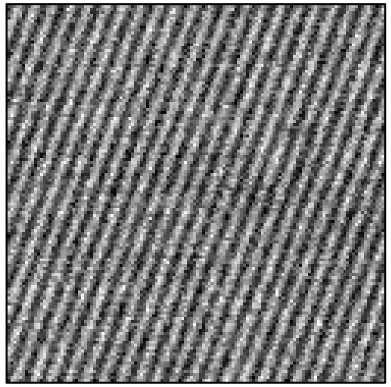
2 s

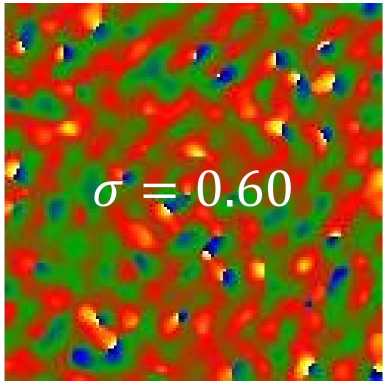


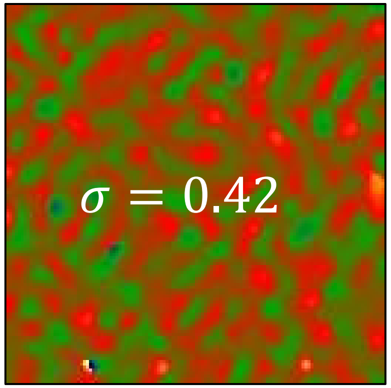
4 s



900 s

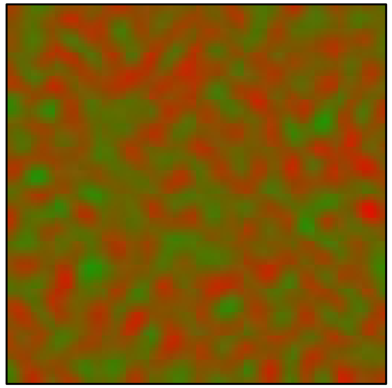




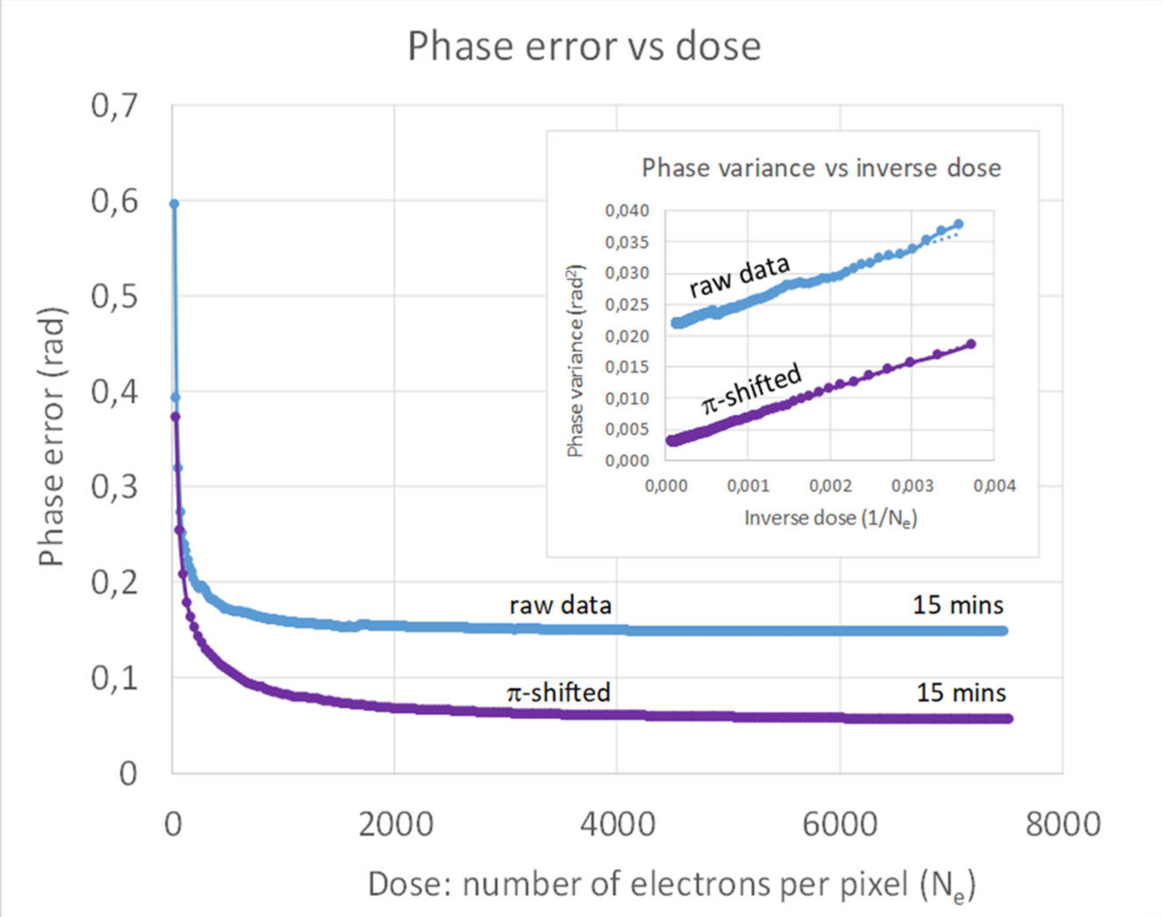


Every 2s

Measure phase standard deviation



Results



$$\sigma_\phi = \frac{1}{V_0}\sqrt{\frac{2}{N_e}}\sqrt{\frac{a}{\mathrm{DQE}(q)}}$$

Visibility

Dose

Phase error from electron dose

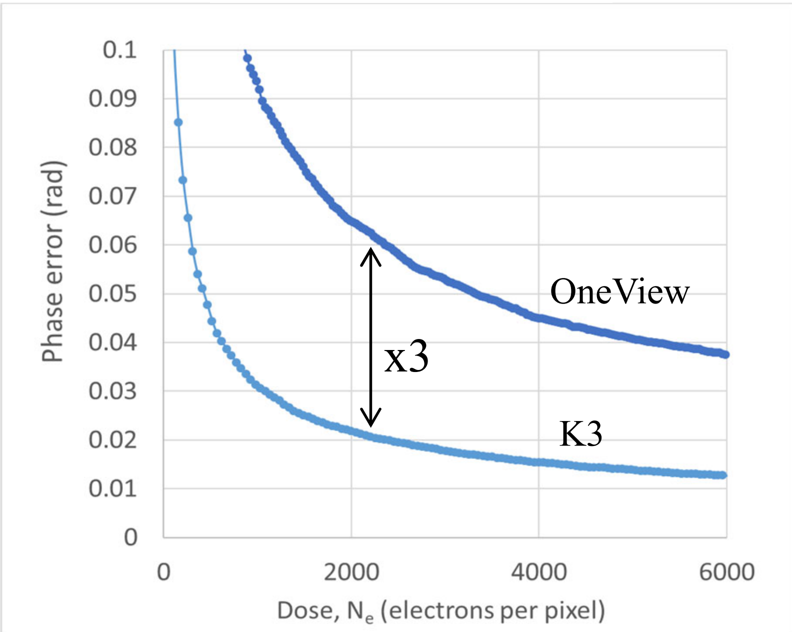
Unknown constant phase error

$$\sigma^2(\phi) = \frac{\sigma_e^2}{N_e} + \sigma_\infty^2$$

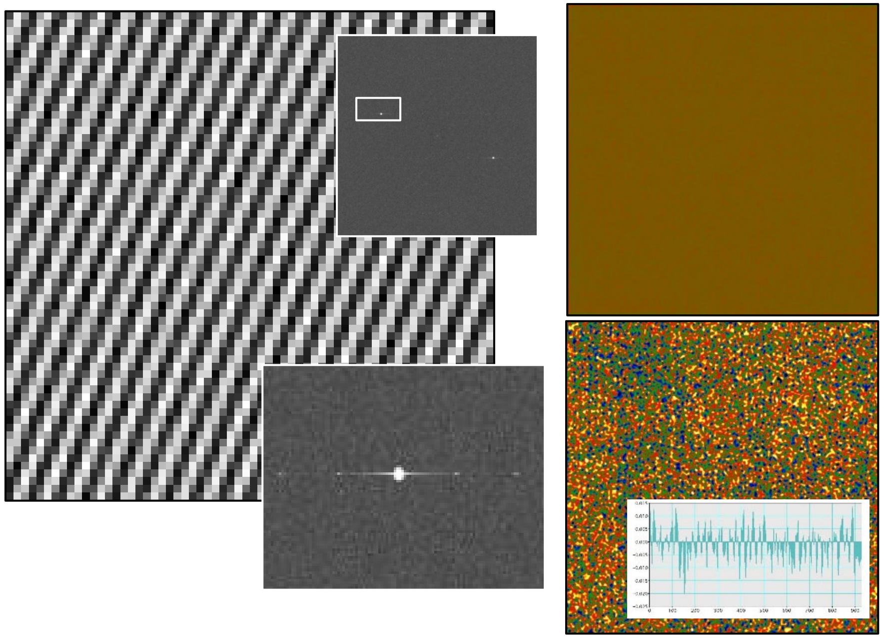
A Harscher and H Lichte, *Ultramicroscopy* 64, 57-66 (1996)

ONEVIEW vs K3

Corrected holograms



Same pixel resolution



0.004 pixel displacement
20 nm on chip !

In-situ Holography

and quantification

In-situ Holography

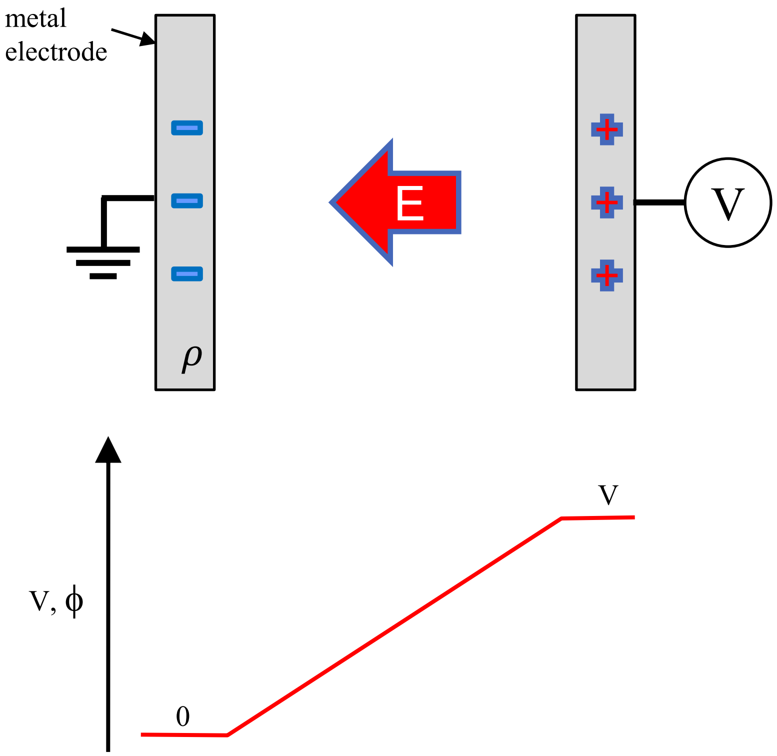
Holograms contain artefacts
Holograms contain unwanted terms

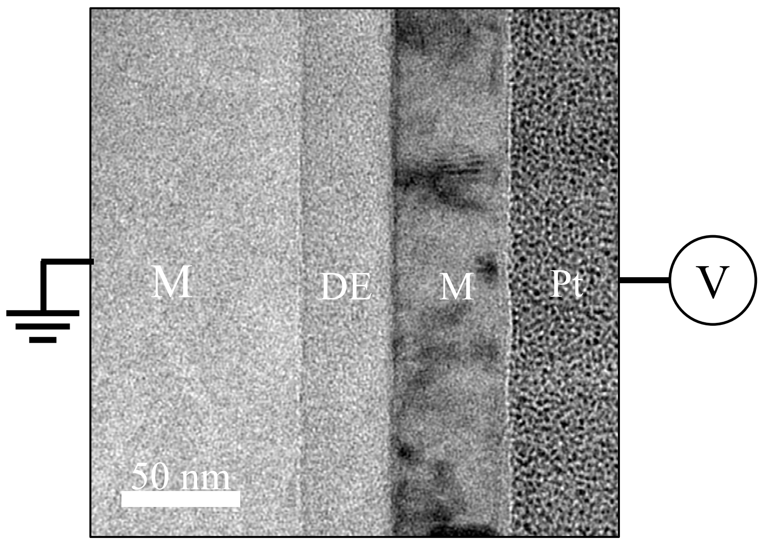
$$\phi = \phi^{C} + \phi^{G} + \phi^{M} + \phi^{E}$$

Vary the parameters

In-situ or operando

Model System: the capacitor

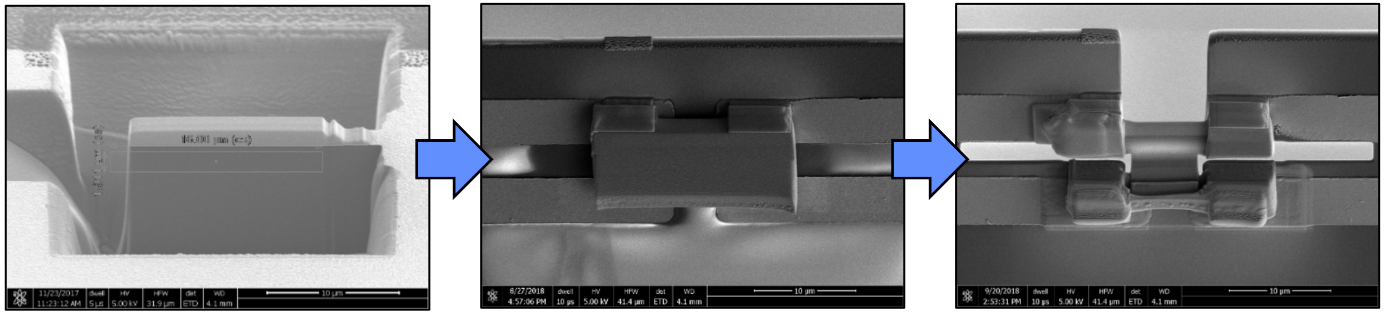


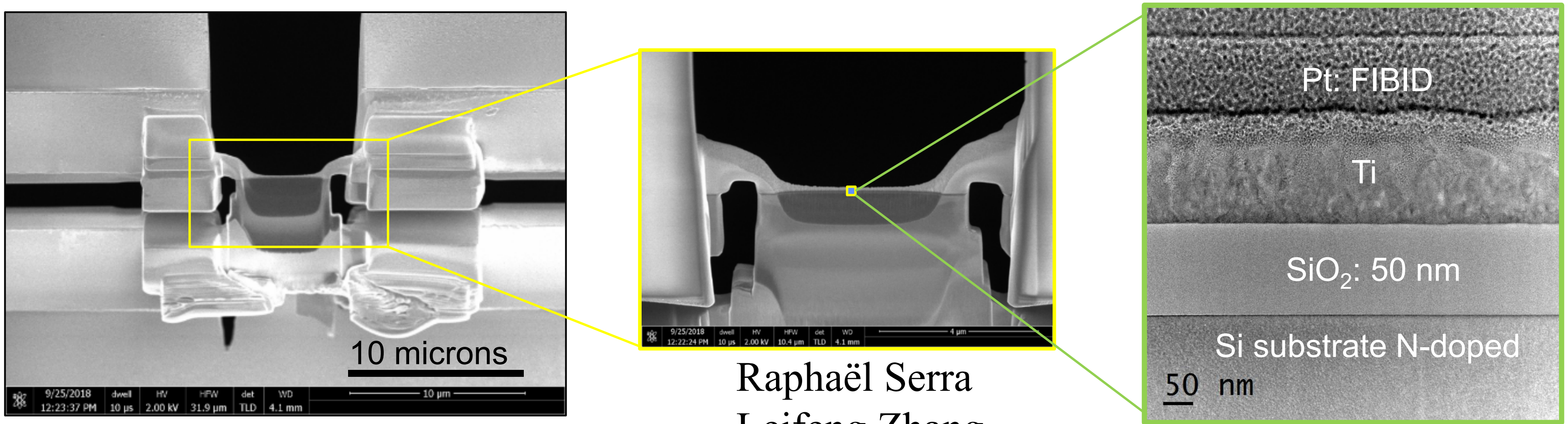


FIB preparation

Nanocapacitor

FIB liftout | Placement on chip | Thinning

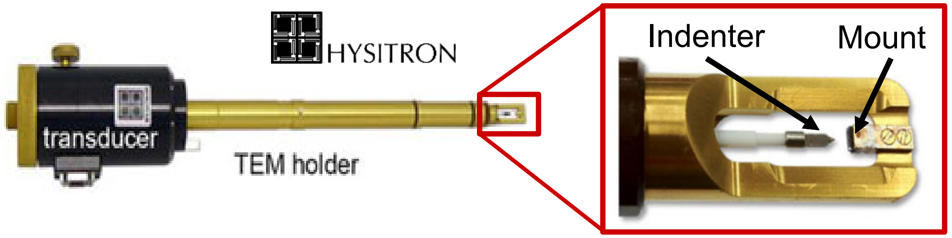


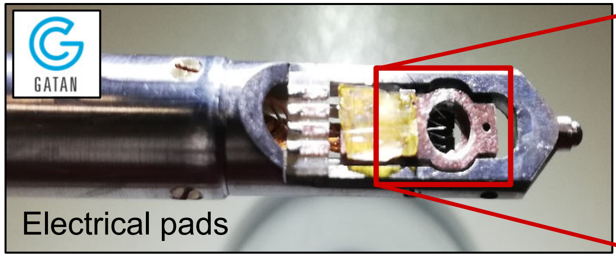


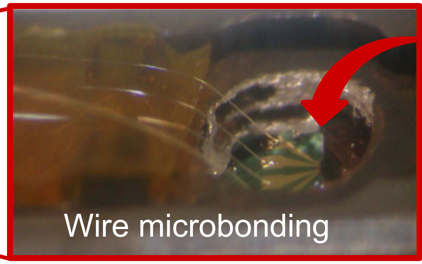
Raphaël Serra
Leifeng Zhang

Dedicated holders

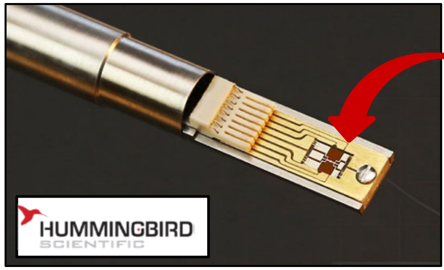
Nanoprobe

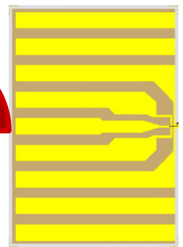






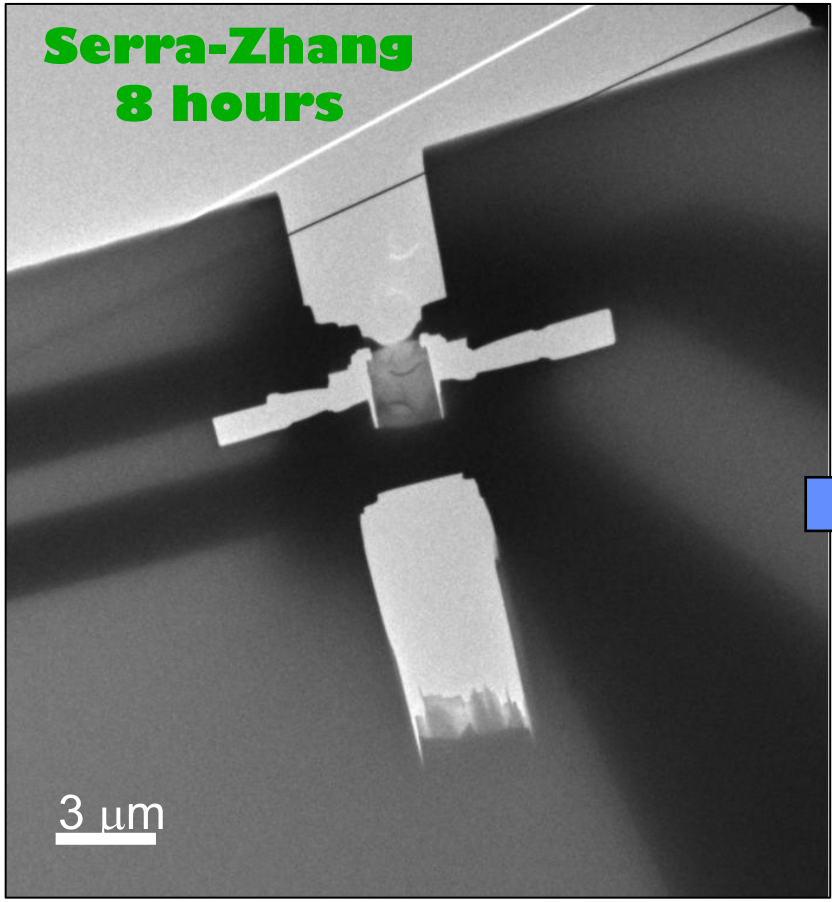
Multiple contacts

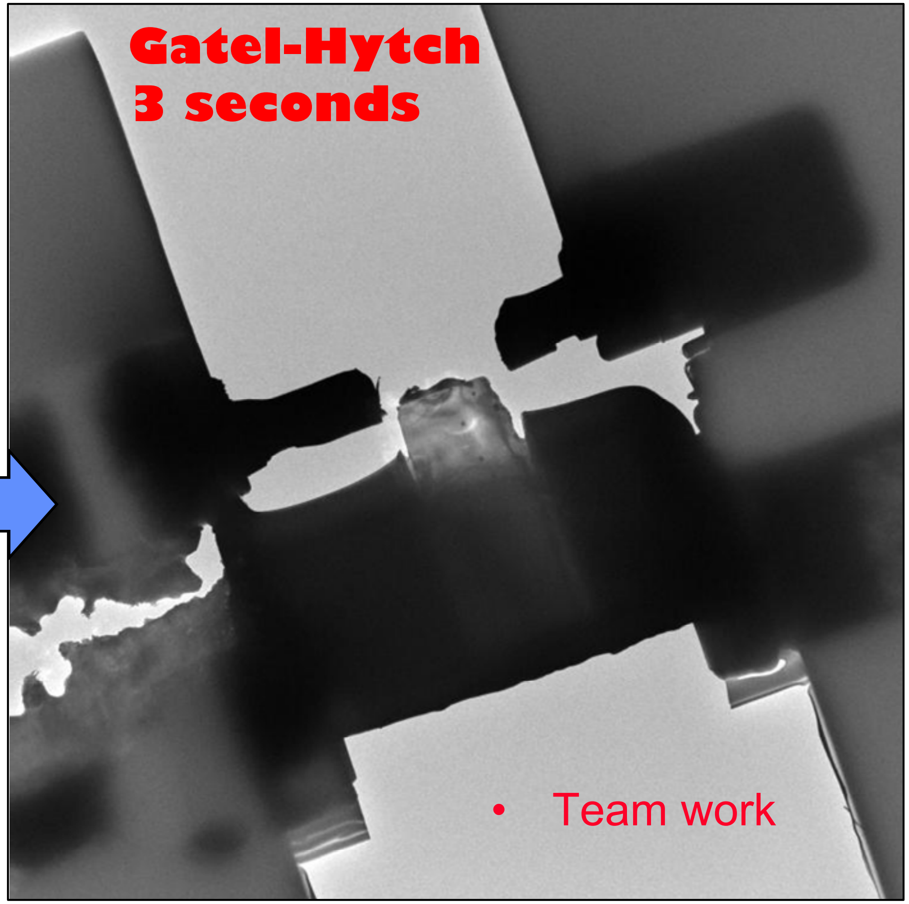




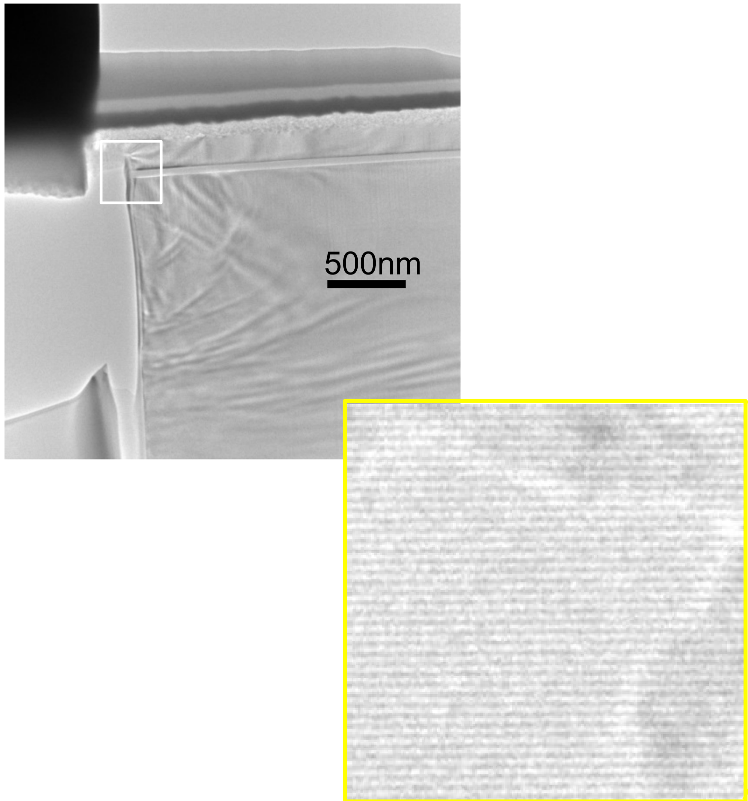
Direct chip/MEMS insertion

Exciting phenomena

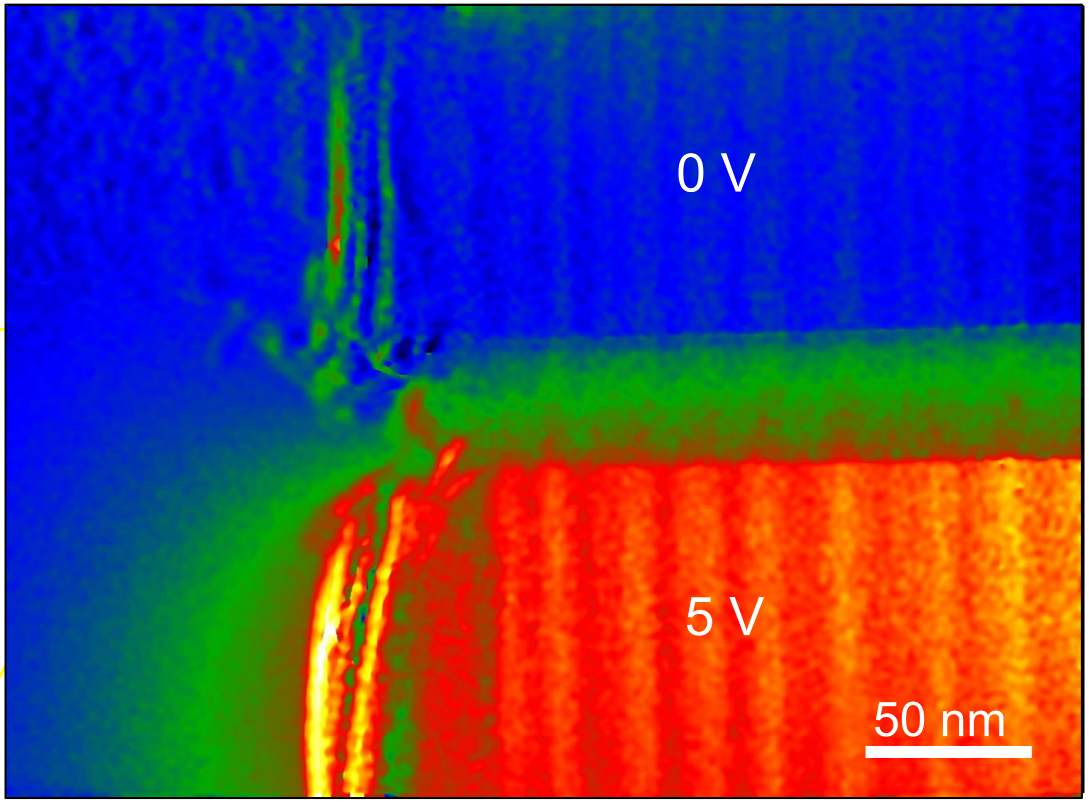




In-situ EH: test structure

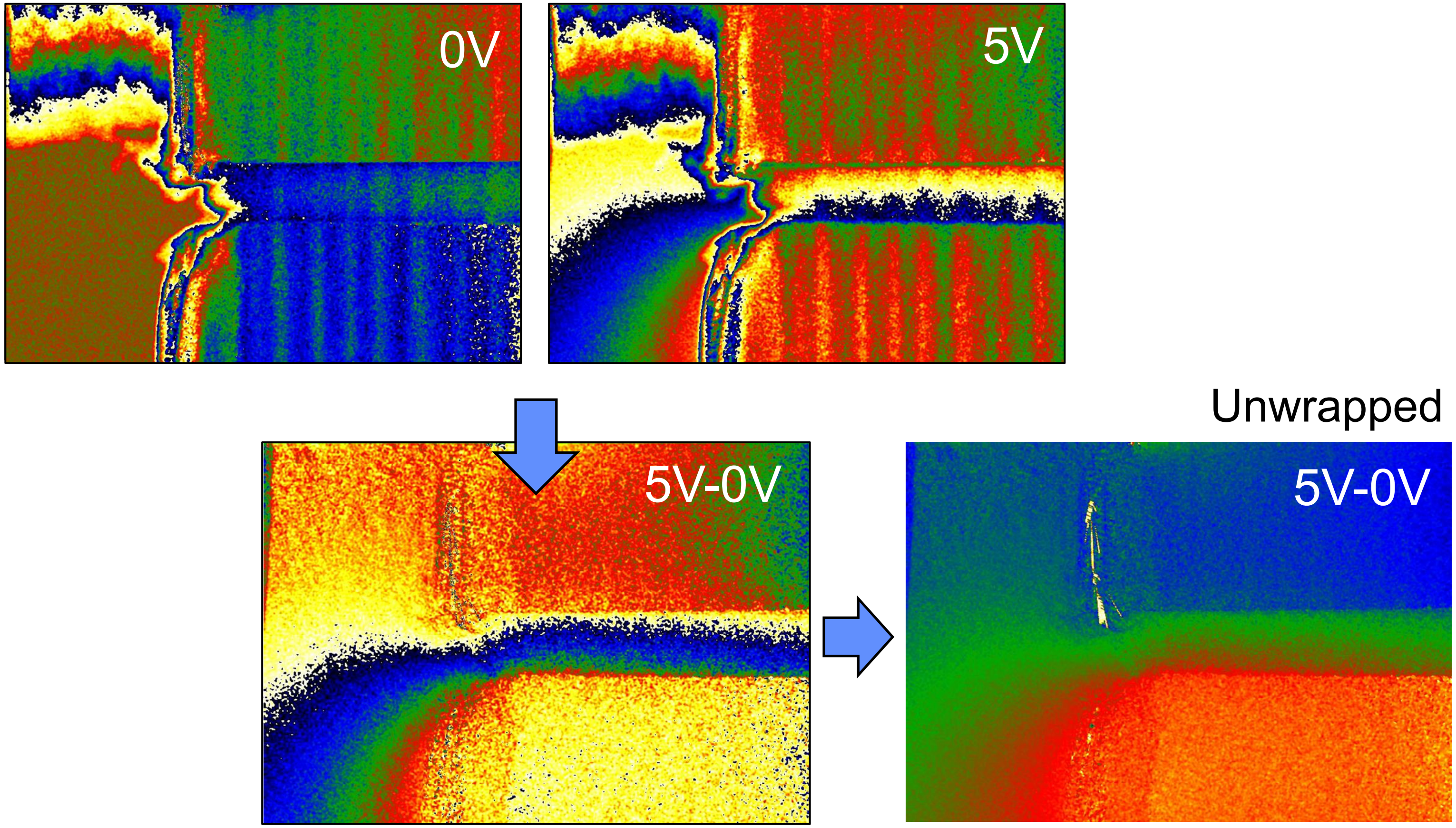


Electron Holography

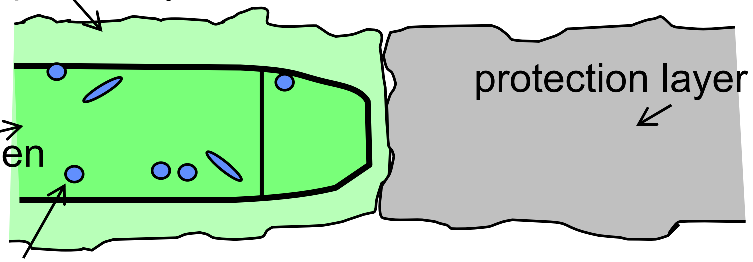


$$\phi^E = c_E \int V(\mathbf{r})dz$$

Image processing

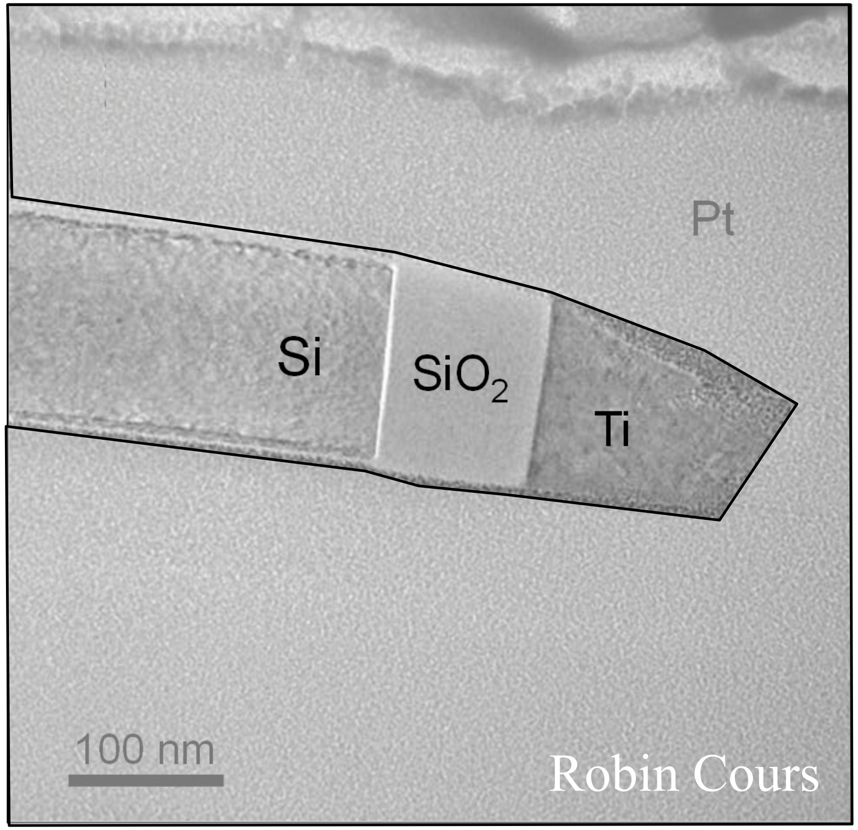


FIB sample preparation

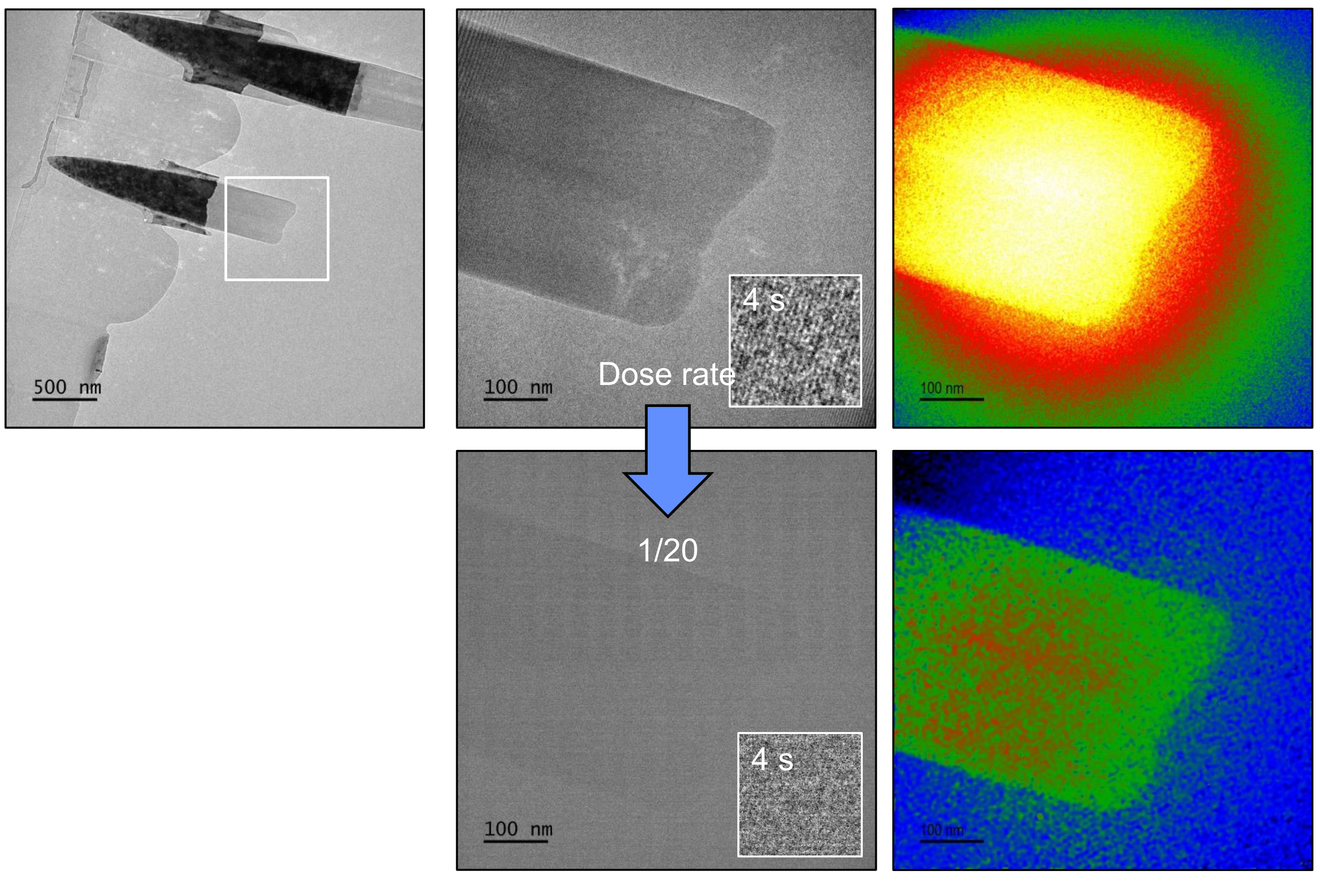


- surface protection layer
- amorphous layer
- damage and impurities
- specimen curvature and bending

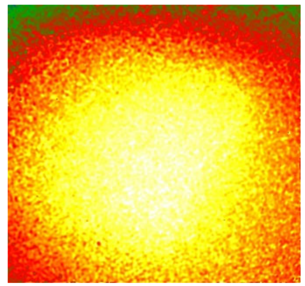
Cross of Cross-section



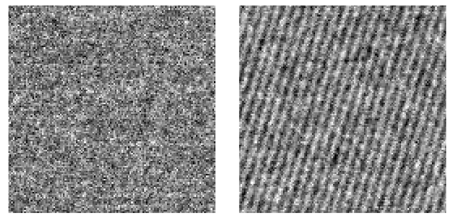
Beam Charging



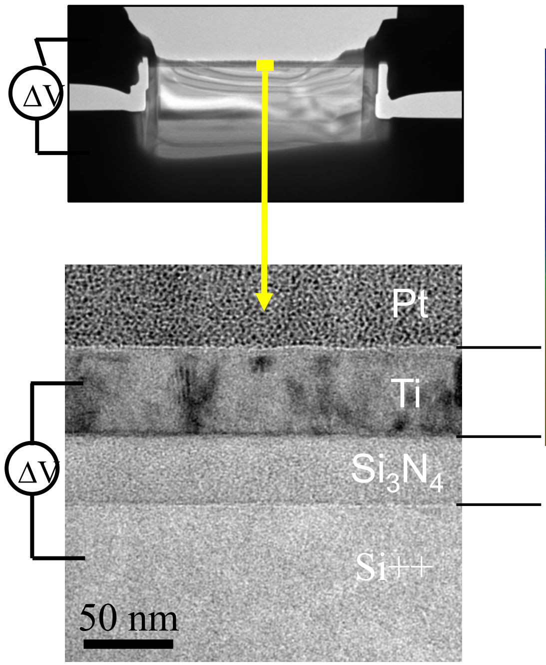
Phase difference



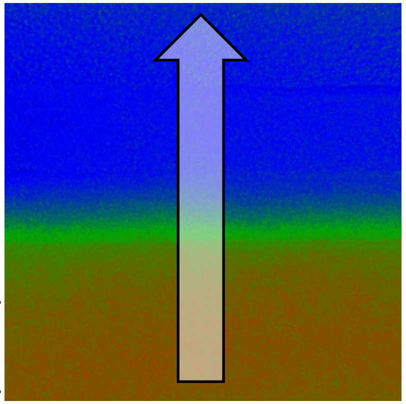
- Less charging
- Need long exposure times



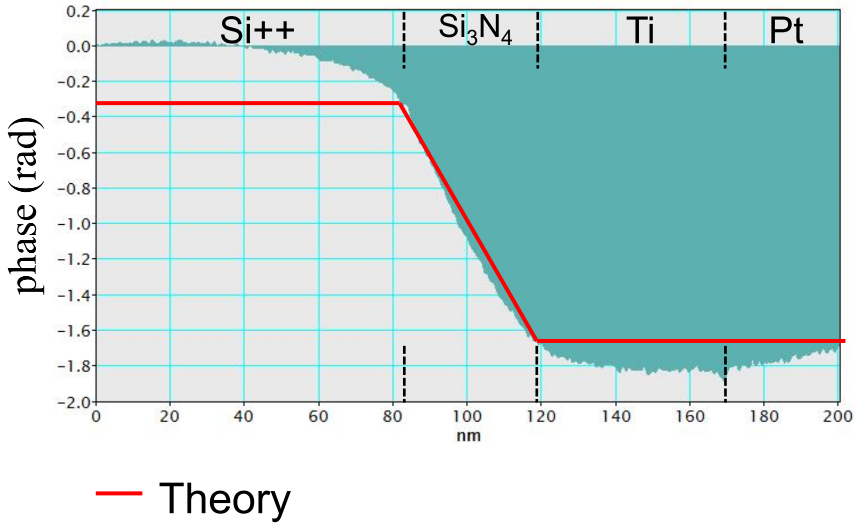
Interpretation and Quantification



Biasing Phase

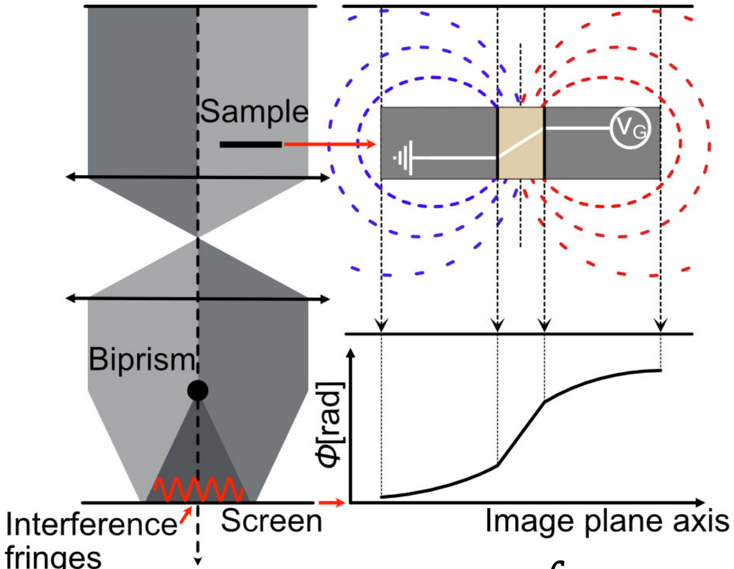


Biasing Phase profile

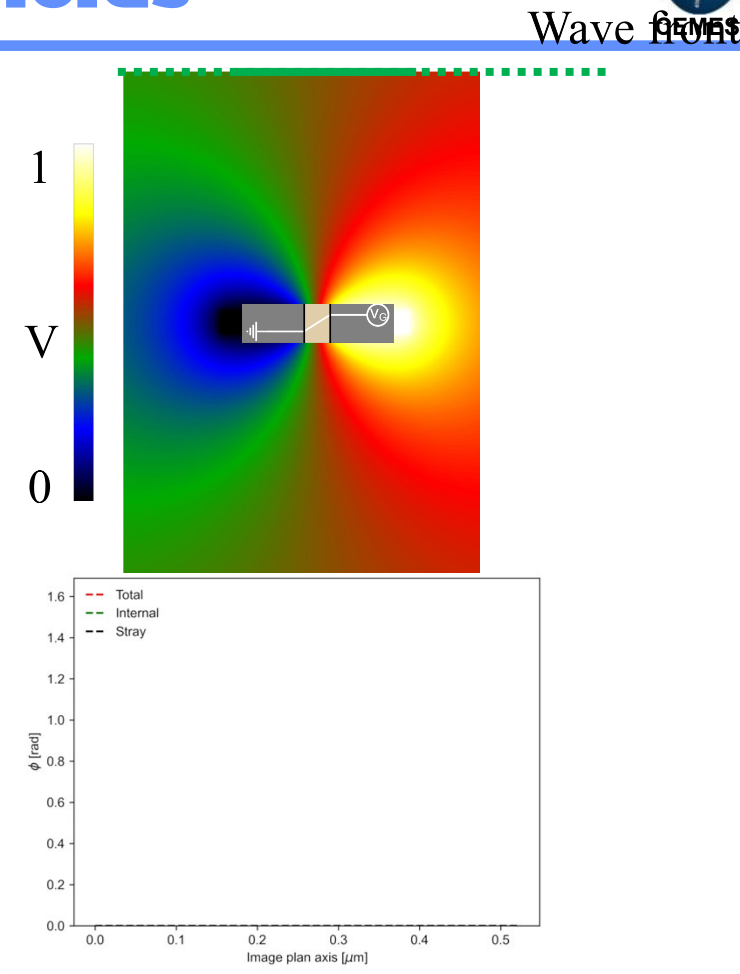


1 rad ≈ 1 Volt

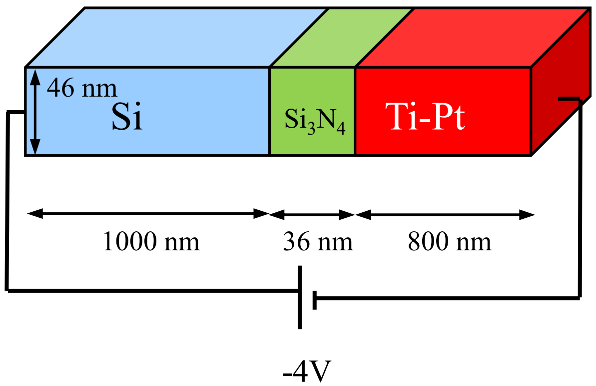
Stray Fields



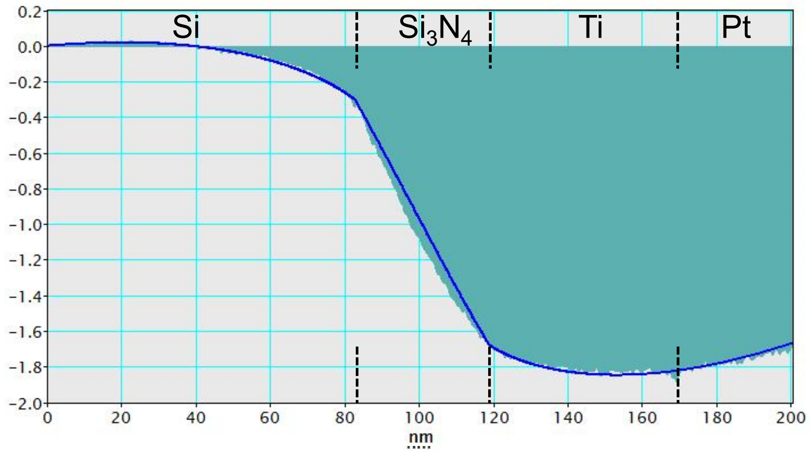
$$\phi^E = c_E \int V(\mathbf{r})dz$$



Fitting Solution

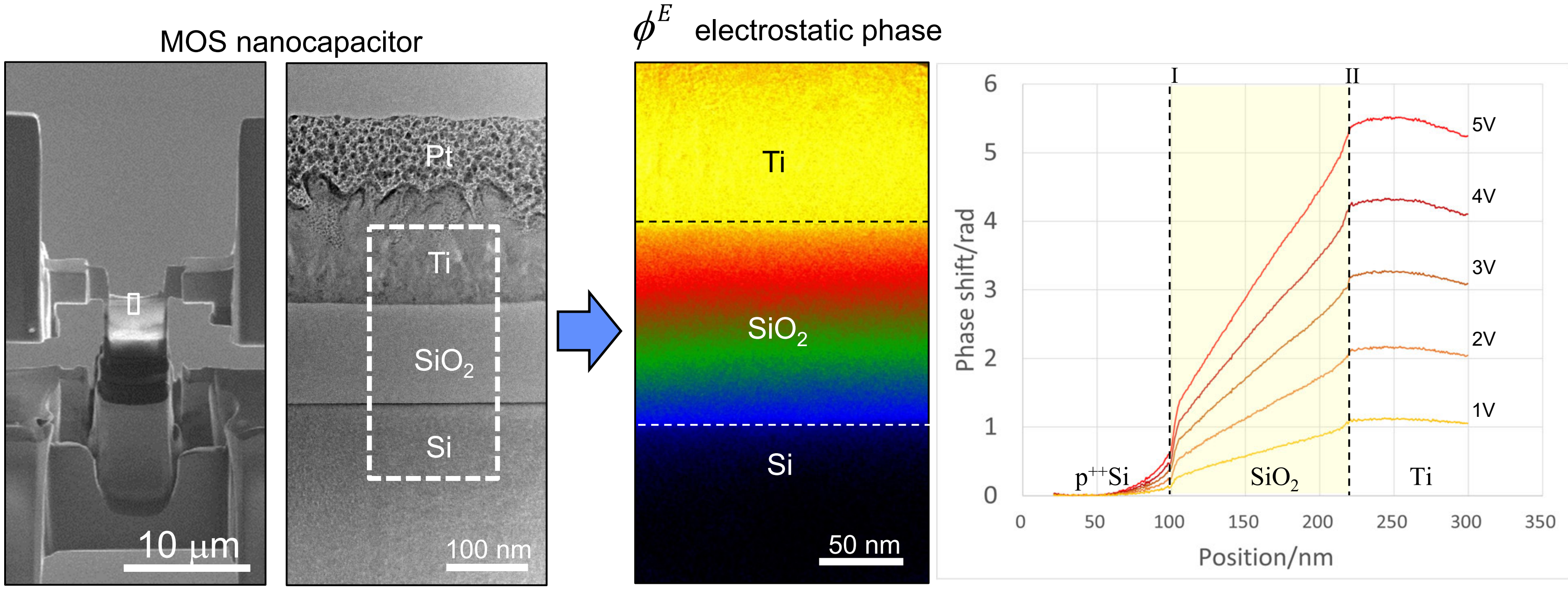


Phase profile



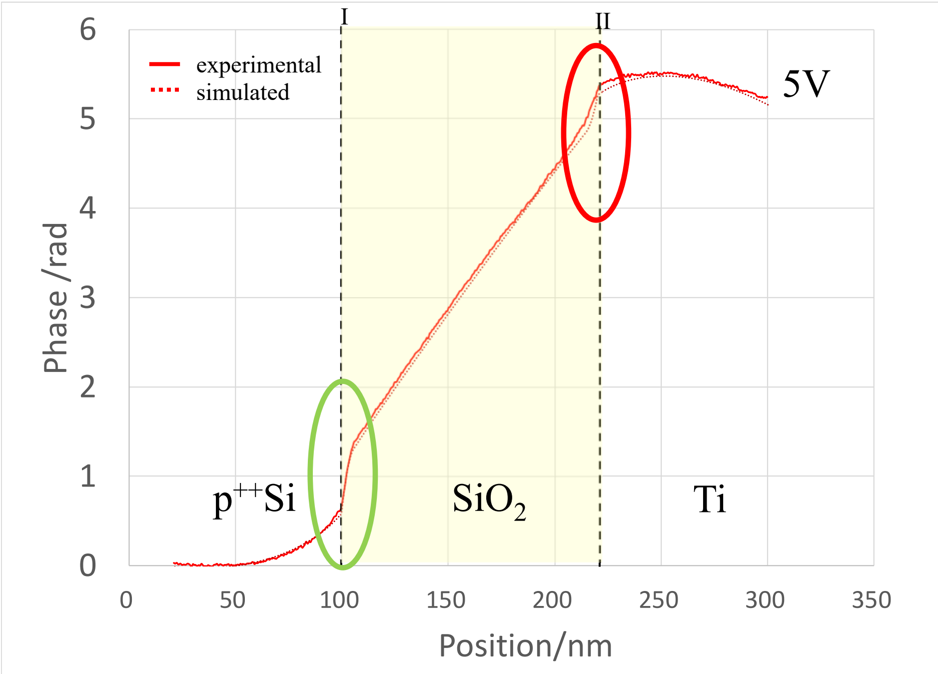
Perfect agreement with
volume charge density in Si_3N_4 of -2.10^5 C.m^{-3}

MOS: Si-SiO2-Ti: MIM

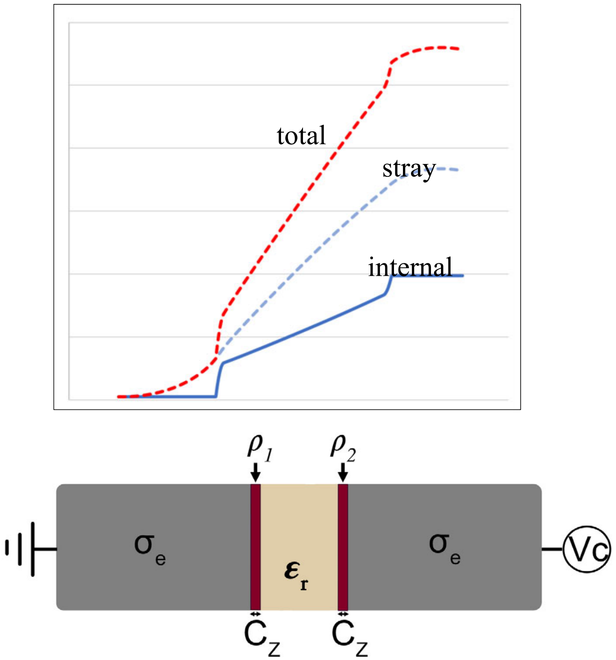


Stray fields and Charge Layers

Phase profile

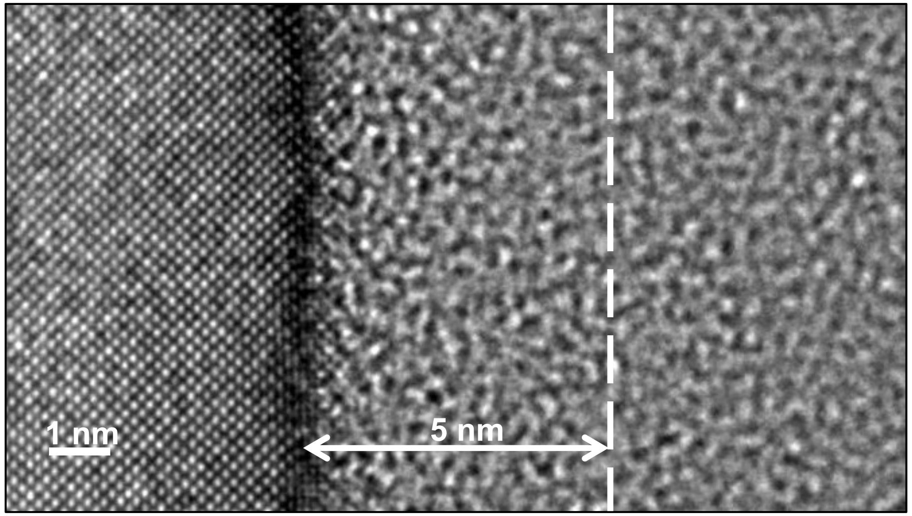


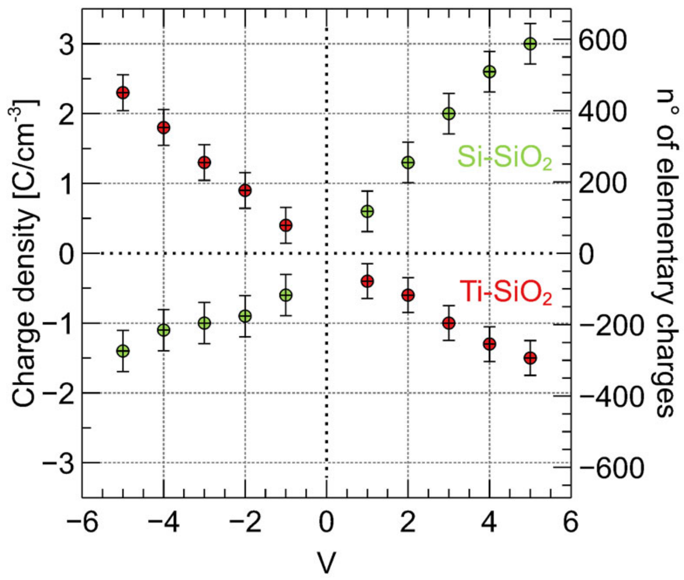
Modeling



Charge Layer Quantification

Interface Si-SiO_2

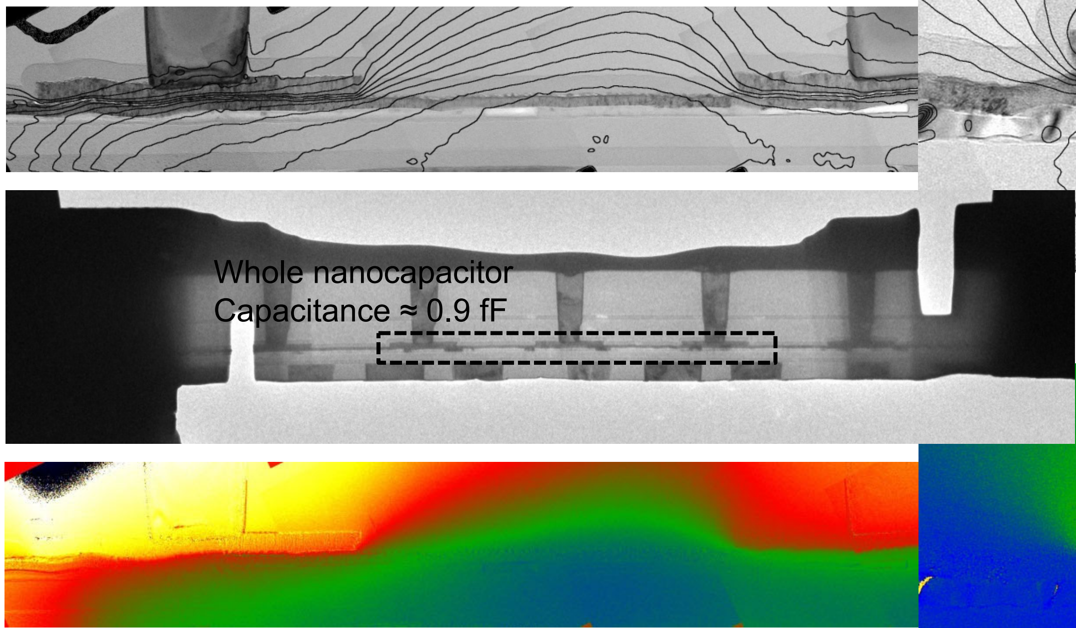


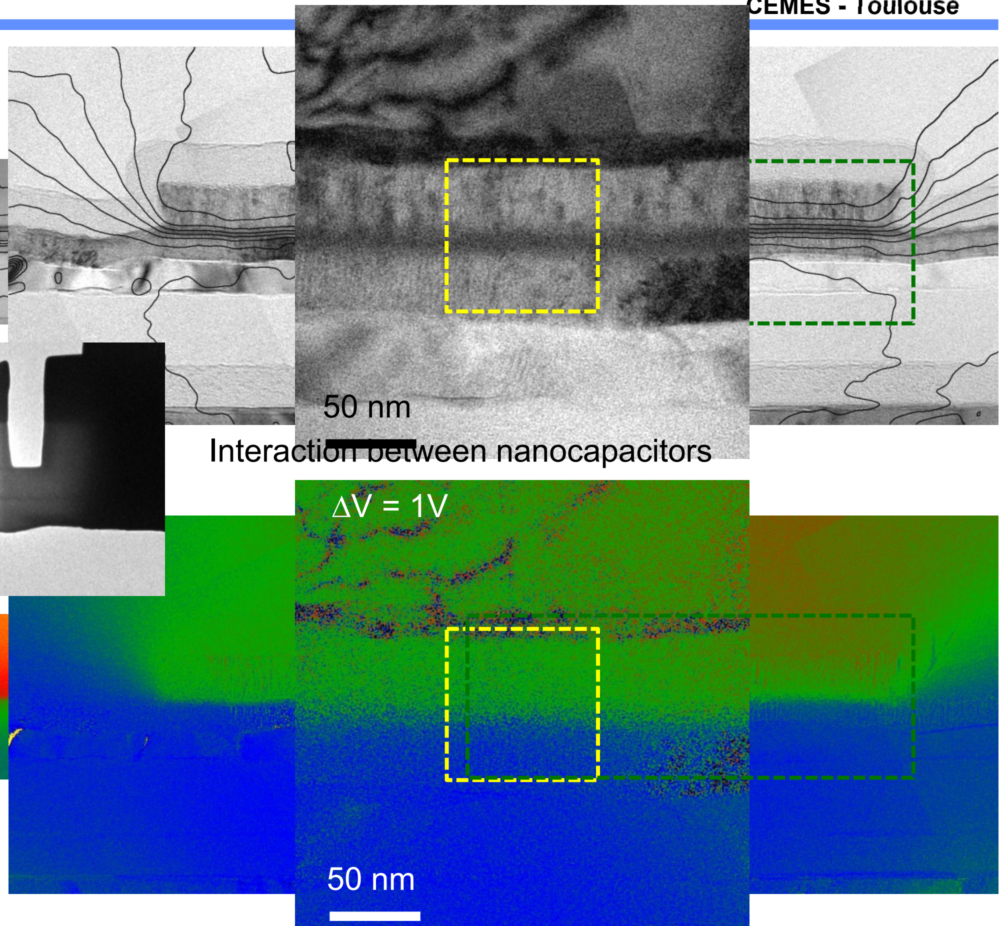


C. Gatel, R. Serra, K. Gruel, A. Masseboeuf, L. Chapuis, R. Cours, L. Zhang, B. Warot-Fonrose, and M. J. Hÿtch, Phys. Rev. Lett. 129, 137701 (2022)

Active components

Real device in production





Field of view = 6.05 µm x 500 nm

Spatial Res. = 0.8 nm

M. Brodovoi, K. Gruel, L. Chapuis, A. Masseboeuf, C. Marcelot, M. J. Hÿtch, F. Lorut, and C. Gatel, APL (2022)