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Lubk, C Gatel

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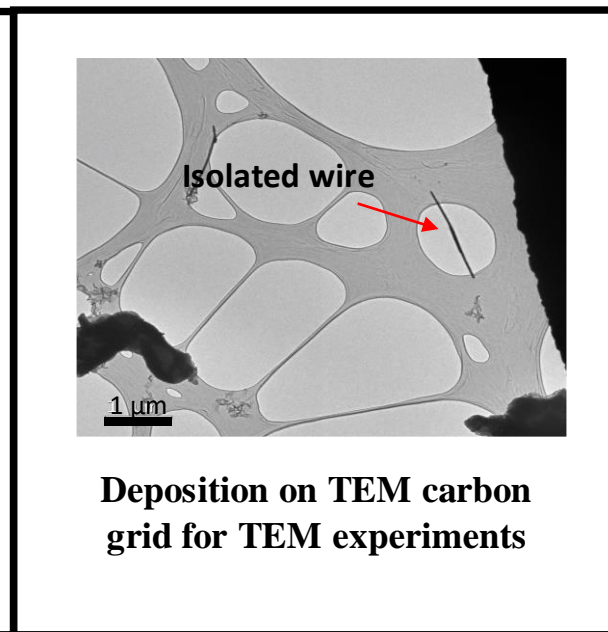
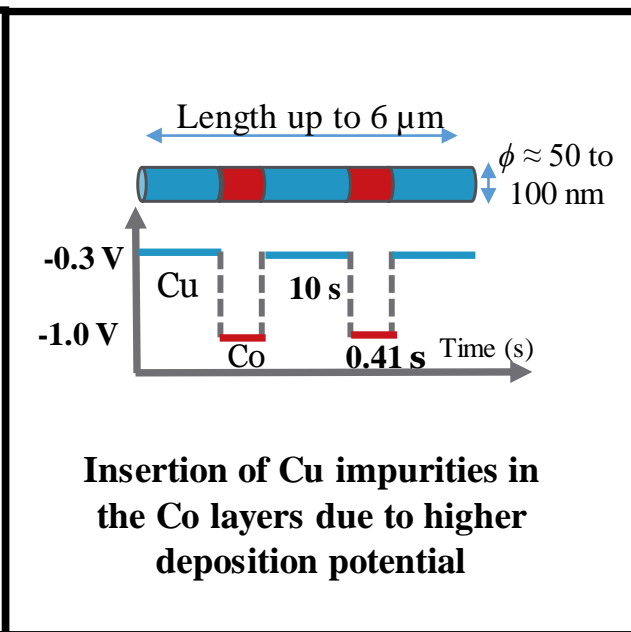
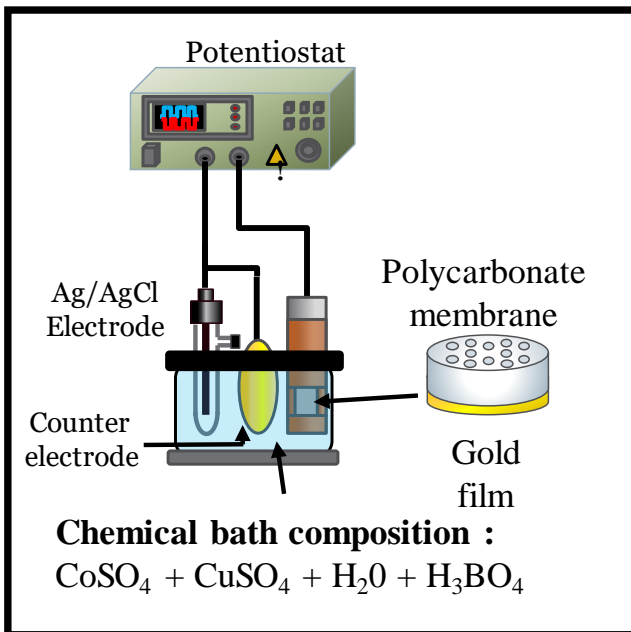
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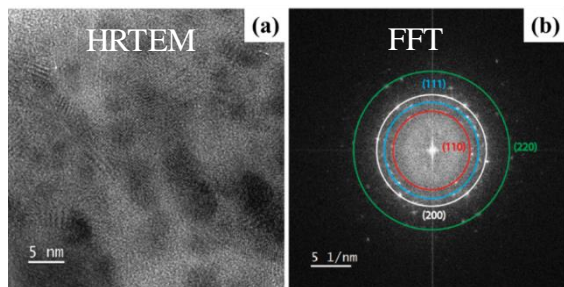
# Micromagnetism of Co/Cu multi-layered nanocylinders observed by electron holography

**N. Biziere**<sup>1</sup>, D. Reyes<sup>1</sup>, T.L. Wade<sup>3</sup>, D. Wolf<sup>3</sup>, B. Warot-Fonrose<sup>1</sup>, S. Sturm<sup>3</sup>,  
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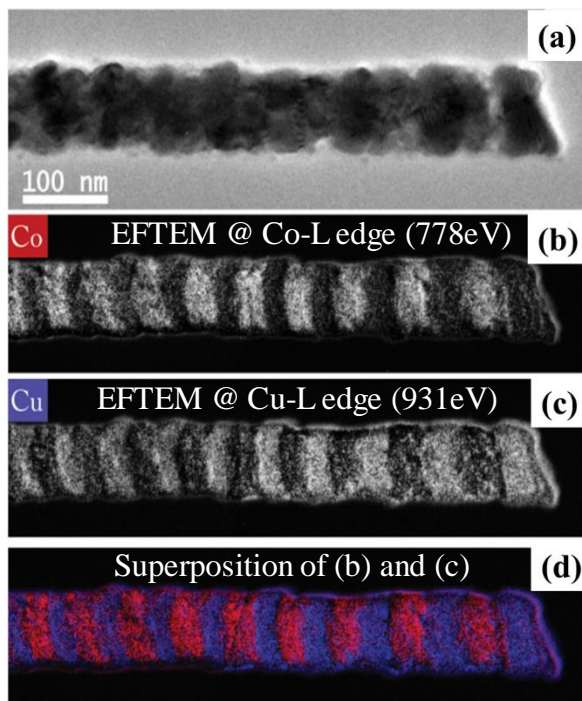
- Interest of template synthesis for magnetic multilayers :
  - ⇒ easiness to modify the thickness of the layers
  - ⇒ nice playground for micromagnetism studies
  - ⇒ potential for spin torque nano-oscillators connected in series?





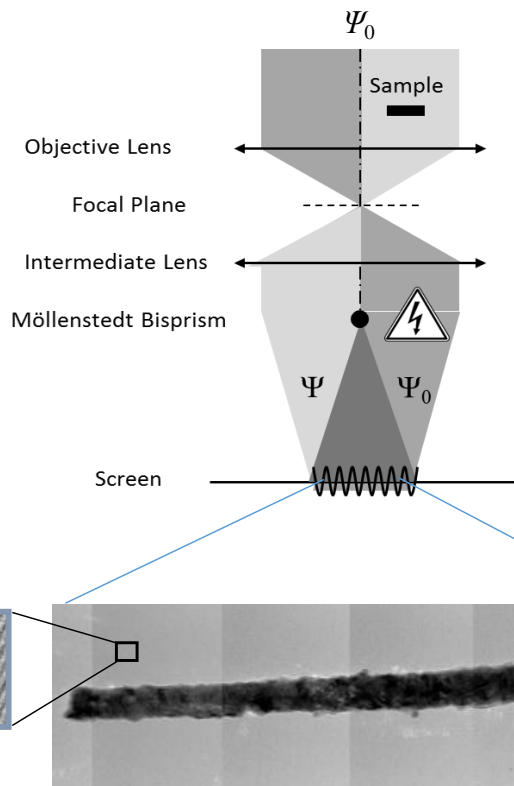
## High Resolution TEM (HREM)

- random distribution of Co and Cu polycrystals.
- grain mean size of  $7 \pm 2$  nm
- fcc phases for Co and Cu (as expected)



## Chemical analysis by Energy Filtered Transmission Electron Microscopy (EFTEM)

- Classical TEM imaging does not allow distinguishing between Co and Cu layers
- EFTEM images at the Co and Cu energy edges reveal the position of the different layers.
- Variations of diameter and thickness along the wire
- Averaged tilt angle  $\approx 20^\circ$  from wire axis and up to  $45^\circ$



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

$$\Psi(\mathbf{r}) = A_S(\mathbf{r}) \exp i((\mathbf{k}\cdot\mathbf{r}) + \phi_S(\mathbf{r}))$$

Incident beam

After the sample

The information on magnetization is in  $\phi_S(\mathbf{r})$

$$\phi_S(\mathbf{x}) = \phi_{elec}(\mathbf{x}) + \phi_{Mag}(\mathbf{x})$$

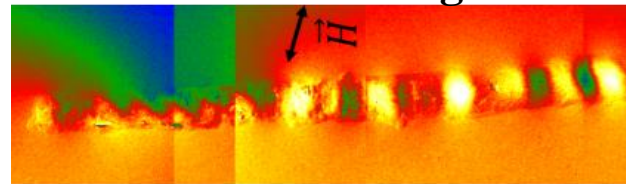
Electrostatic part

$$\phi_{elec}(\mathbf{x}) = C_E \int V(\mathbf{x}, \mathbf{y}, \mathbf{z}) d\mathbf{z}$$

Magnetic part

$$\phi_{Mag} = -\frac{e}{\hbar} \int B_{\perp}(\mathbf{x}, \mathbf{z}) d\mathbf{x} d\mathbf{z}$$

Phase shift image



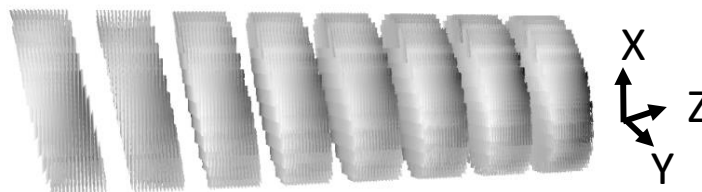
- Integration of  $\vec{B}$  along the electron path
- ⇒ Sensitive to in plane components of  $\vec{B}$
- ⇒ **2D projection of the 3D magnetic state**

⇒ *Need for micromagnetic simulations and phase shift reconstruction to recover the full 3D magnetic state*

- OOMMF simulations of magnetic remanent states after saturation // or  $\perp$  to the wire axis
- Calculation of magnetic induction  $\vec{B}$  and reconstruction of electron phase shift  $\phi_{Mag}$  image :

$$\vec{B} = \mu_0(\vec{M} + \vec{H}_D) \Rightarrow \phi_{Mag} = -\frac{e}{\hbar} \iint \mathbf{B}_{\perp}(x, z) \cdot dx \cdot dz$$

Cell size  $2 \times 2 \times 2 \text{ nm}^3$   
From 1 to 20 layers of Co

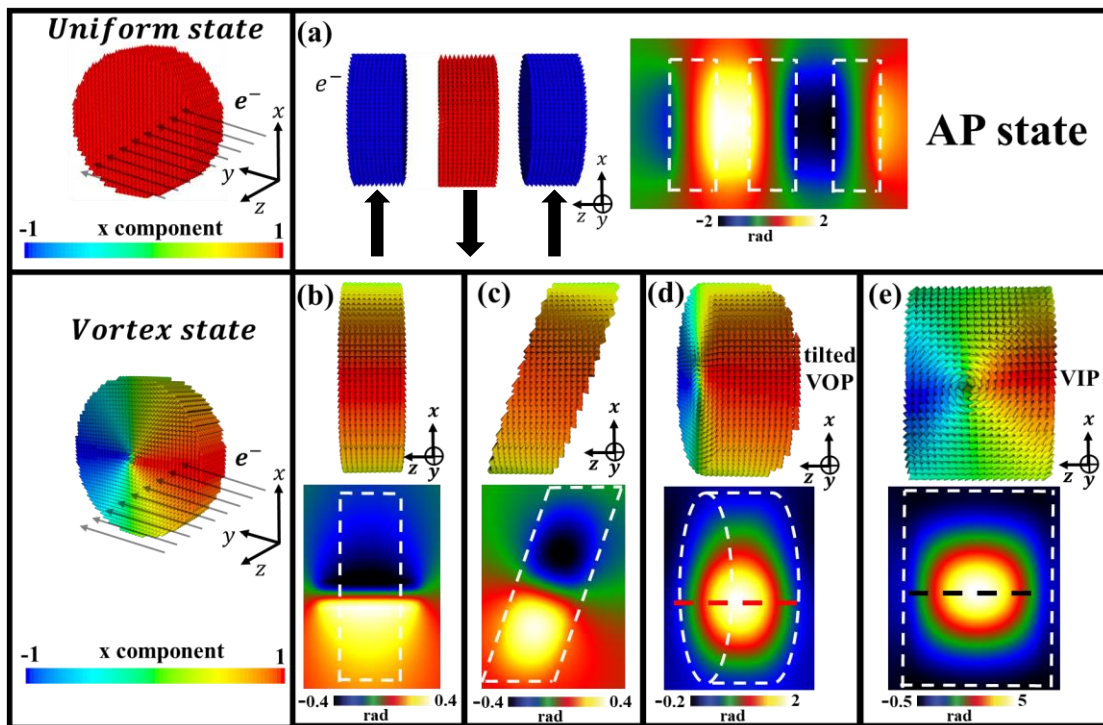


*Use of CALMIP parallel computing center of Toulouse, France*

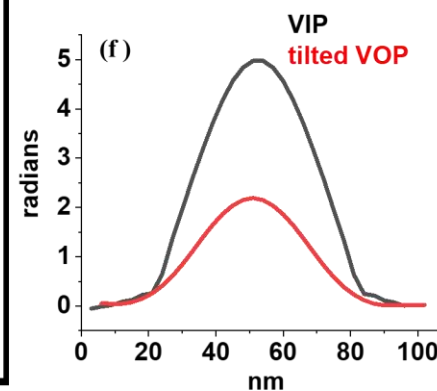
## Difficulty : high number of geometrical and magnetic inputs !

- Geometry extrapolated from EFTEM : layers tilted around Y and X axis (**more complex geometry than a simple disk!**)
- Adjustable magnetic parameters :
  - $\mu_0 M_s = 1.5 \text{ T}$  (obtained from other measurements)
  - exchange value  $A \approx 22 \cdot 10^{-12} \text{ J/m}$  (bulk =  $30 \cdot 10^{-12} \text{ J/m}$ )
  - **uniaxial crystal anisotropy field  $H_k$  because of polycrystals** (value  $K_u$ , direction  $\theta_{HK}$ ) with  $K_u$  between  $100$  and  $200 \cdot 10^3 \text{ J/m}^3$  (bulk =  $400 \cdot 10^3 \text{ J/m}^3$ )

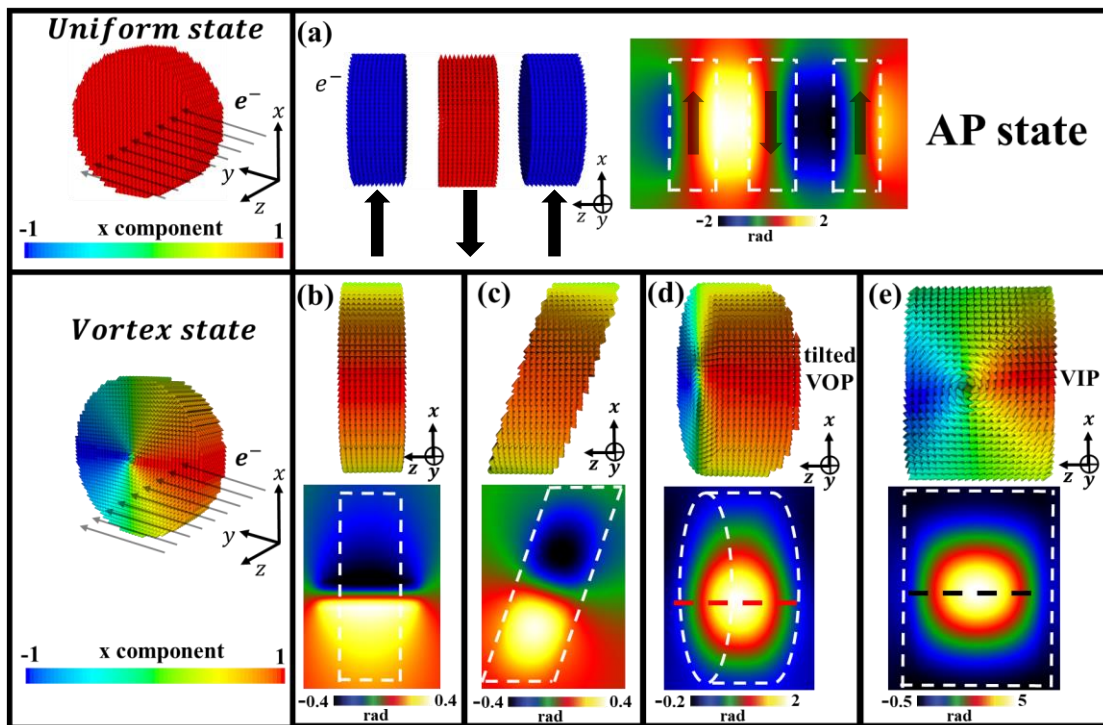
$\Rightarrow$  **Values lower than bulk because of Cu impurities in Co**



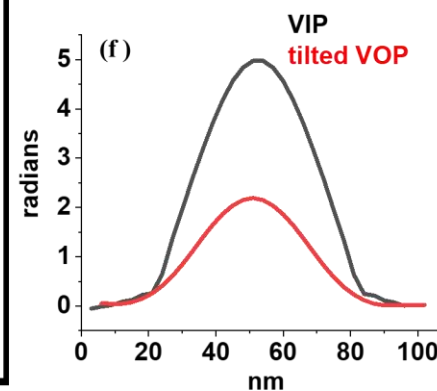
Direction of  $e^-$  beam =  $y$



- Phase shift images depend on magnetic states and geometry of the layers.
- **Problem** : different magnetic states “look” similar because of the layer geometry  
 $\Rightarrow$  *need for qualitative and quantitative analysis of the images*

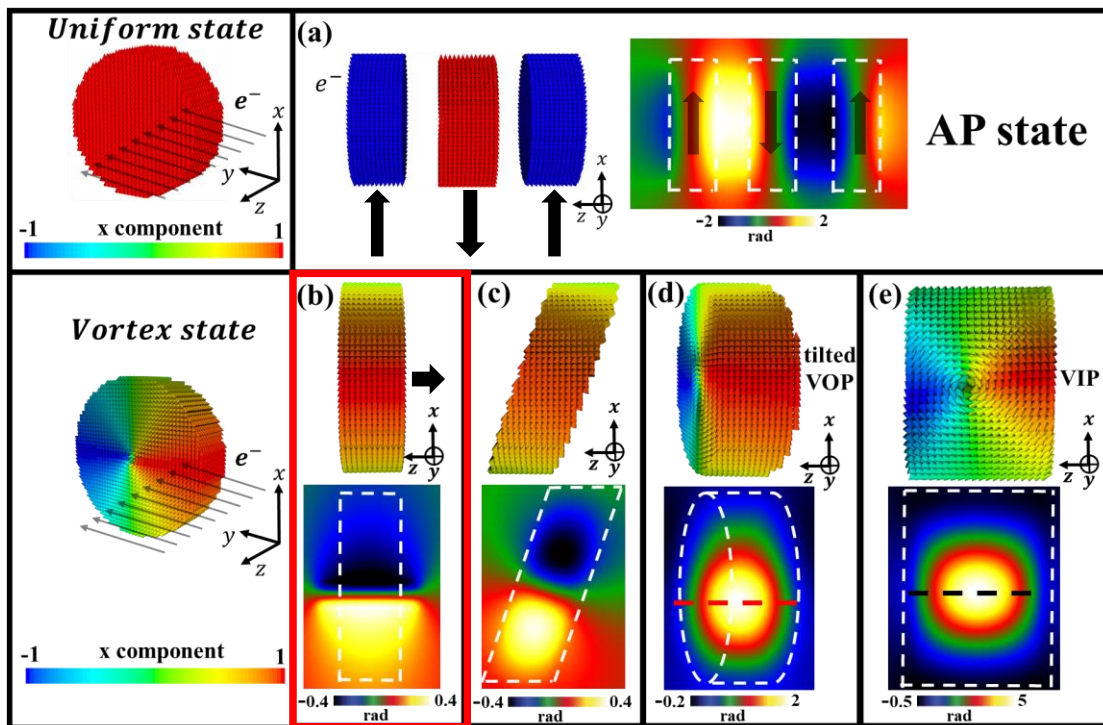


Direction of  $e^-$  beam =  $y$

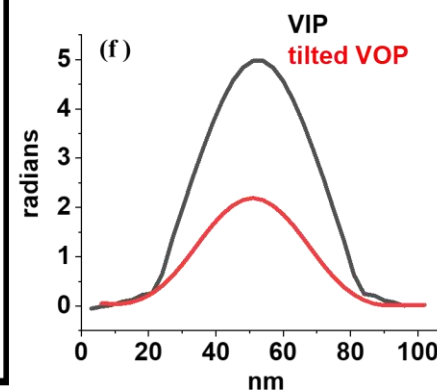


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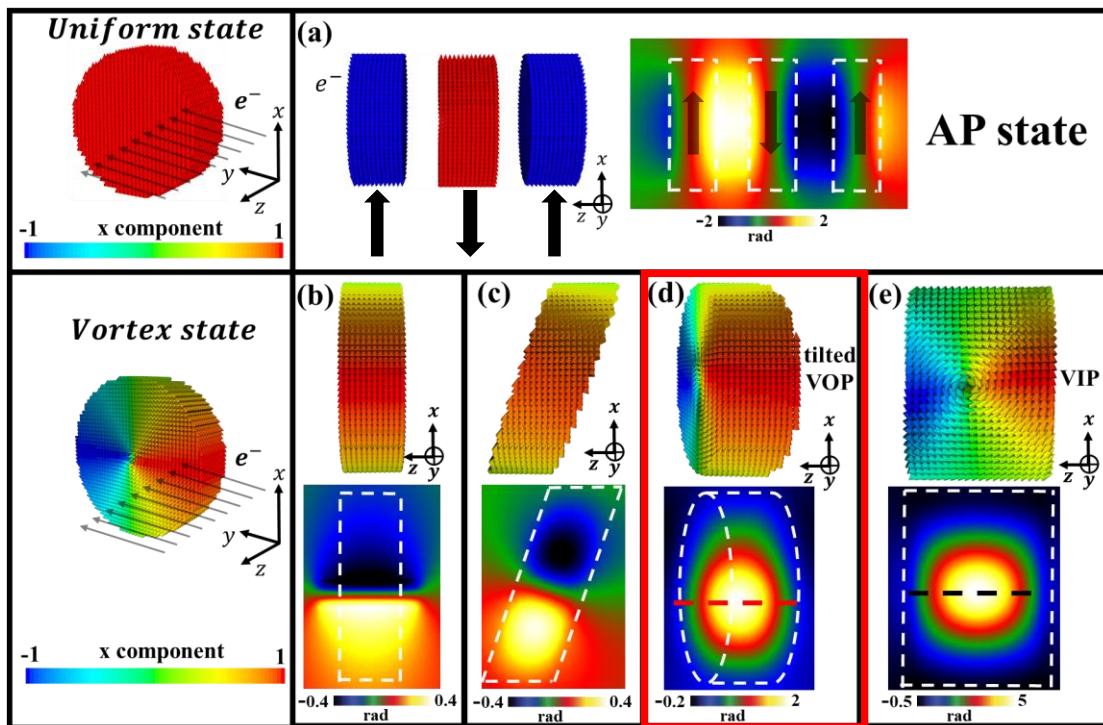


Direction of  $e^-$  beam =  $y$

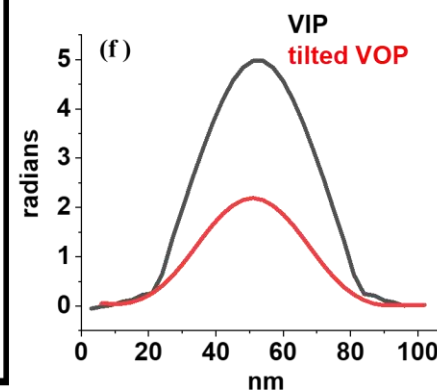


- Phase shift images depend on magnetic states and geometry of the layers.
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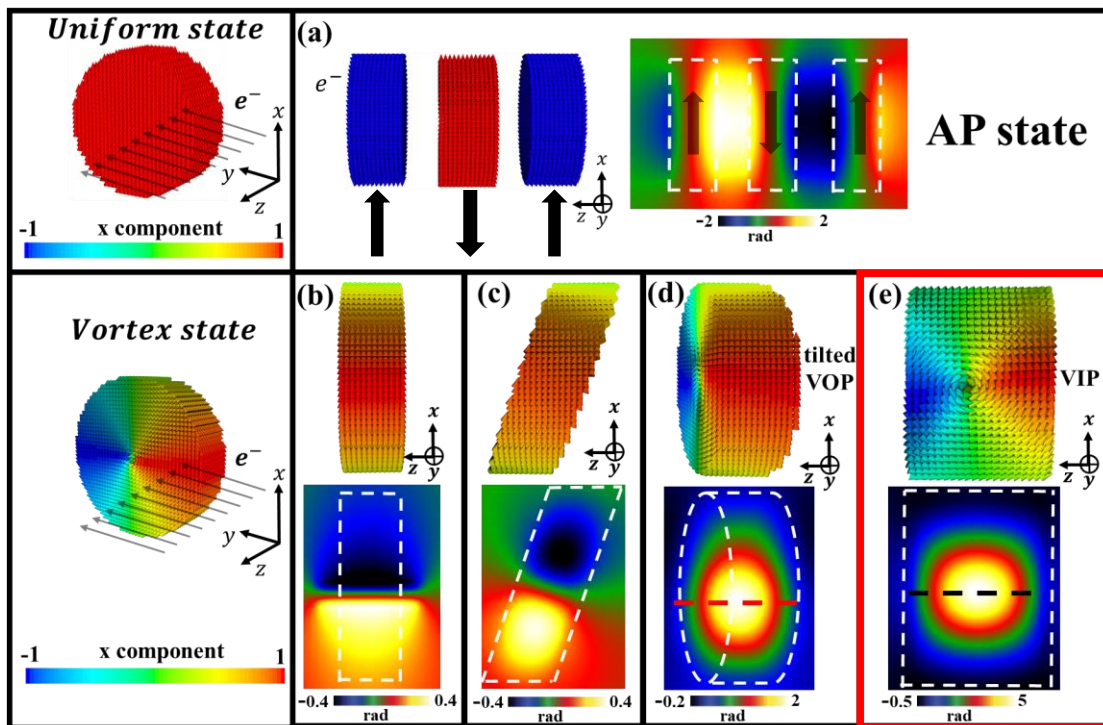


Direction of  $e^-$  beam =  $y$

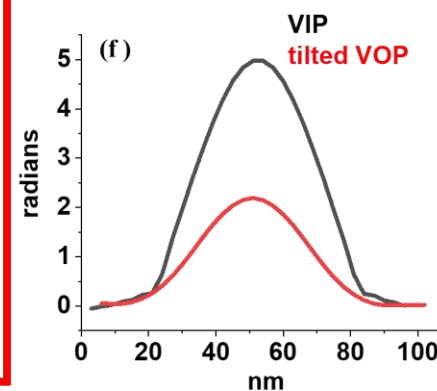


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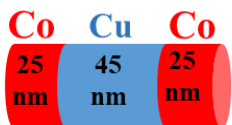
*⇒ need for qualitative and quantitative analysis of the images*



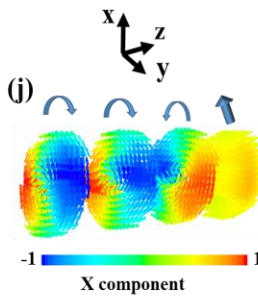
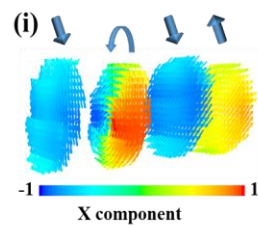
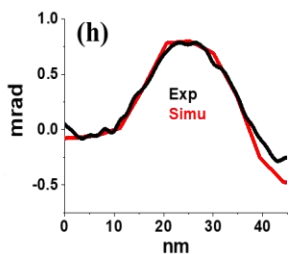
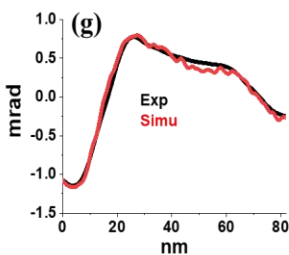
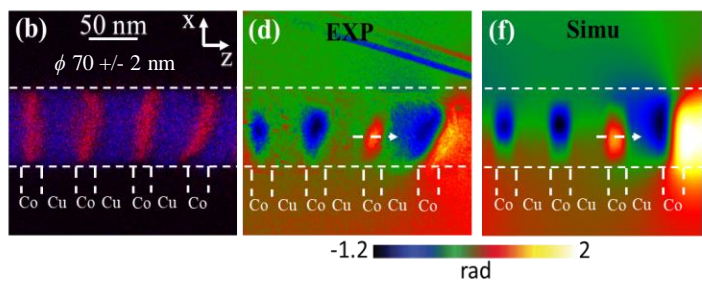
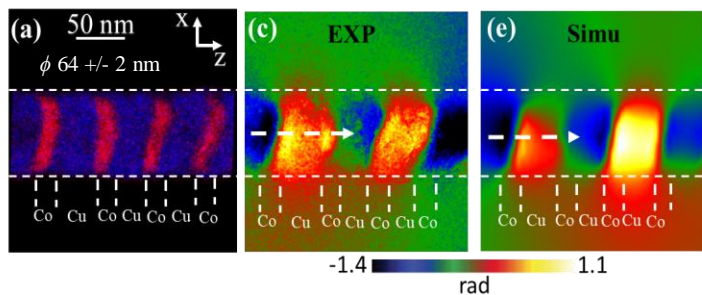
Direction of  $e^-$  beam =  $y$



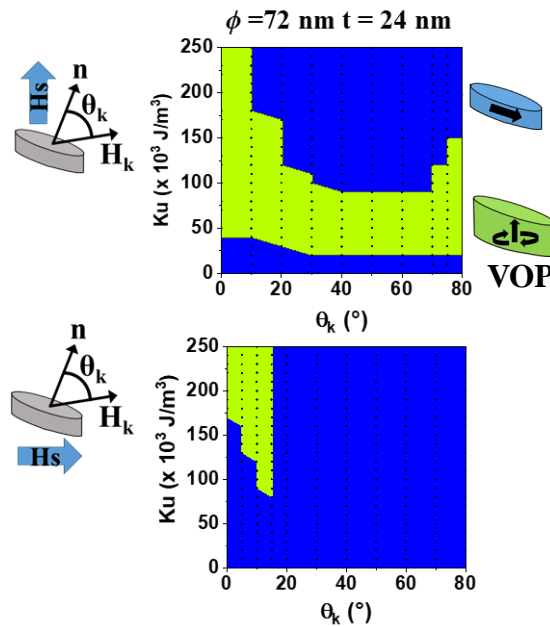
- Phase shift images depend on magnetic states and geometry of the layers.
- **Problem** : different magnetic states “look” similar because of the layer geometry  
 $\Rightarrow$  need for qualitative and quantitative analysis of the images



## Multilayer

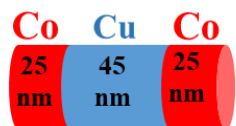


## Phase diagram of single layer

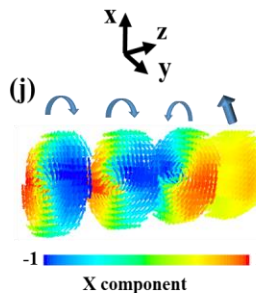
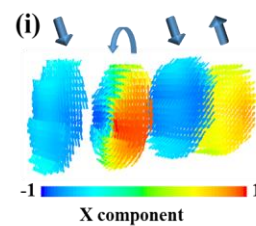
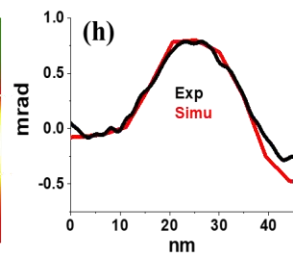
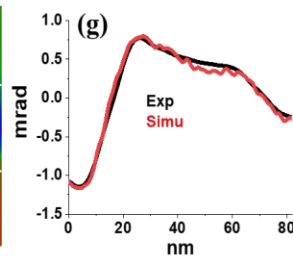
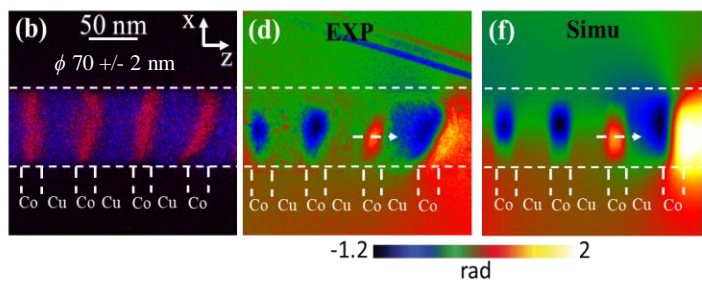
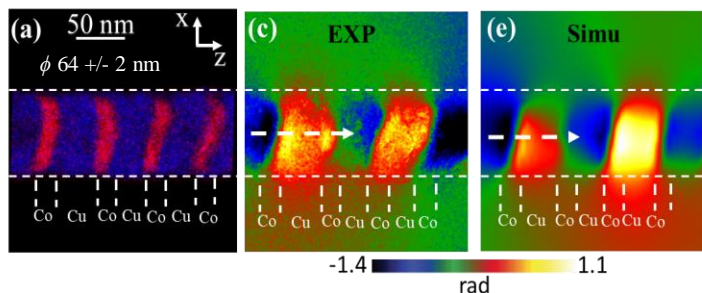


- No effect of saturation field  $H_s$  on the remanent states of multilayers
- $\approx 60\%$  of layers in antiparallel (AP) states and  $\approx 40\%$  in vortex states (VOP).

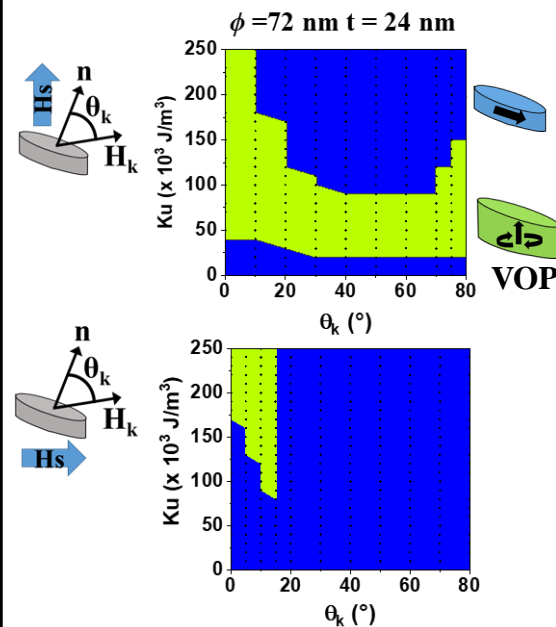
*N. Biziere et al., J. Appl. Phys. 126, 163906 (2019)*



## Multilayer



## Phase diagram of single layer

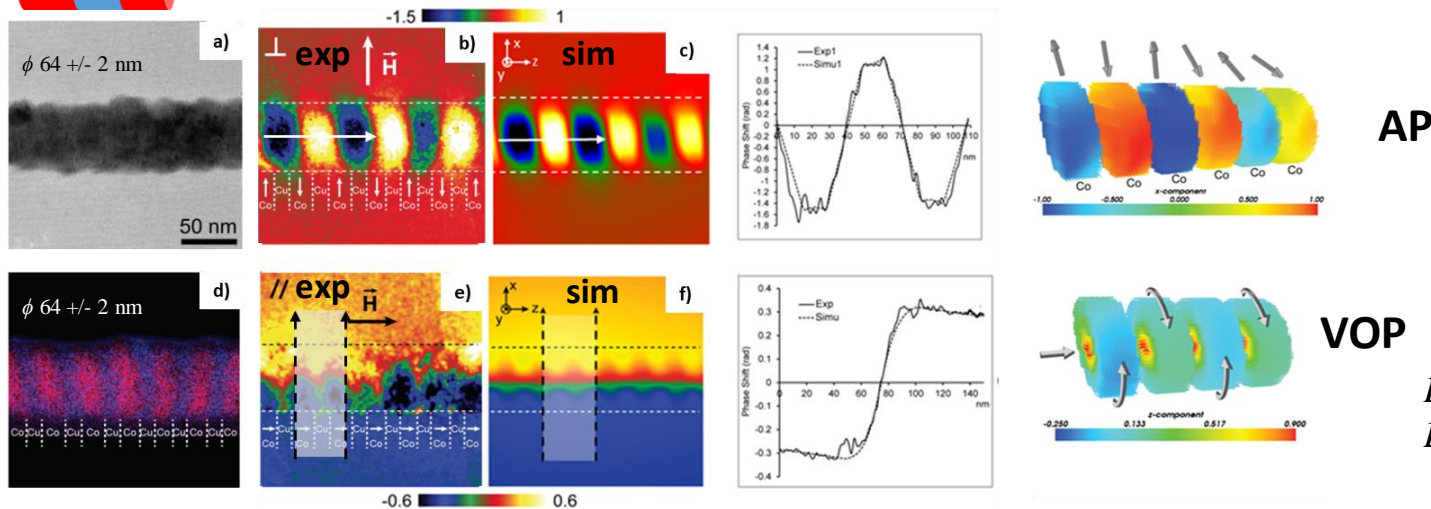


- No effect of saturation field  $H_S$  on the remanent states of multilayers
- $\approx 60\%$  of layers in antiparallel (AP) states and  $\approx 40\%$  in vortex states (VOP).
- Simulations on 20 layers show that :
  - Anti parallel states are stable whatever  $H_S$  if  $\theta_k \geq 20^\circ$
  - vortex states are stable whatever  $H_S$  only if  $\theta_k \leq 20^\circ$

*N. Biziere et al., J. Appl. Phys. 126, 163906 (2019)*

$\Rightarrow$  significant of a preferential orientation of  $H_k$  toward the wire axis ?

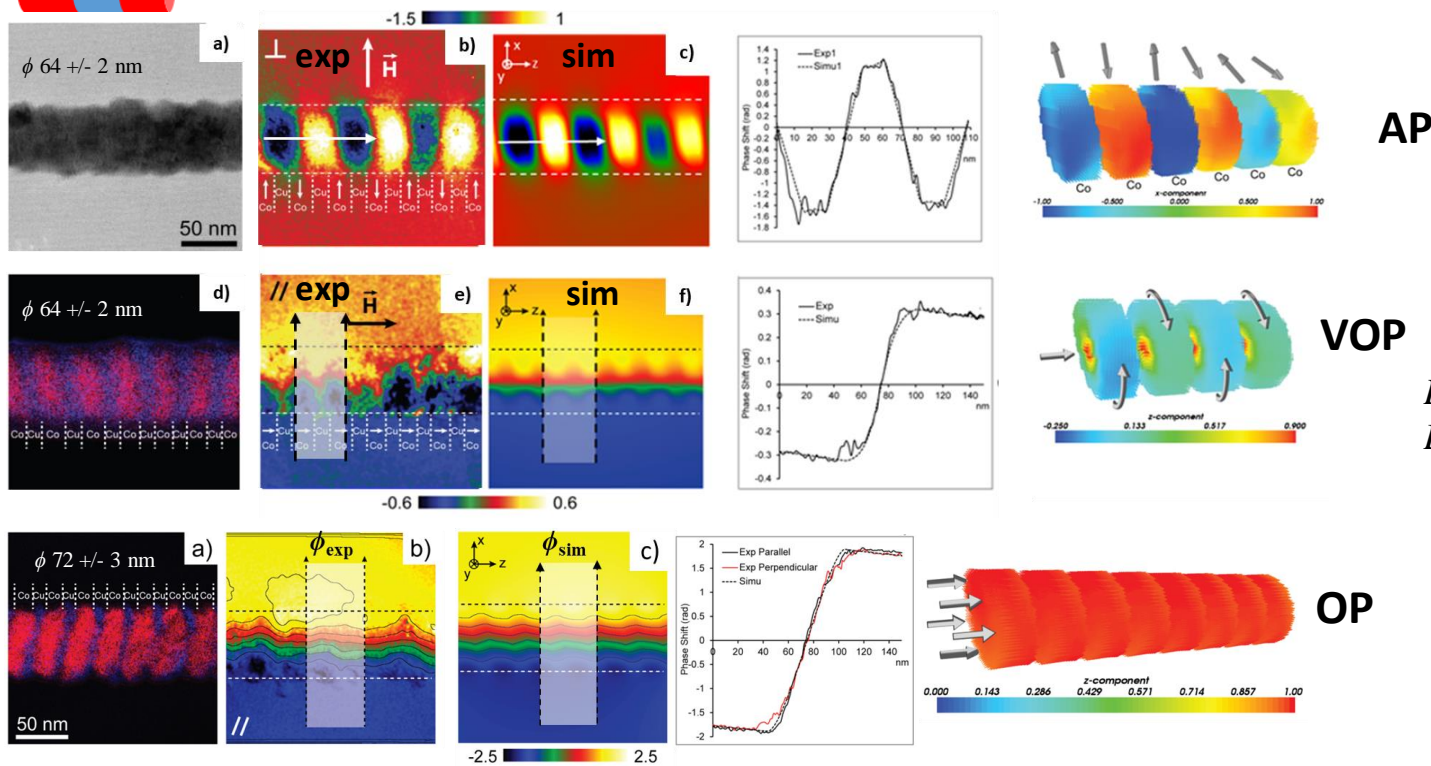
Co Cu Co  
25 nm 15 nm 25 nm



*D. Reyes et al., Nano Lett. 16, 1230, 2016.*

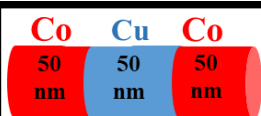
- 70% of the layers show AP configuration whatever  $H_s$  : favored by stronger dipolar coupling
- 20% of layers switch between AP and VOP states when rotating  $H_s$  (only possible if  $H_k$  close from the normal of the layers).

Co Cu Co  
25 nm 15 nm 25 nm

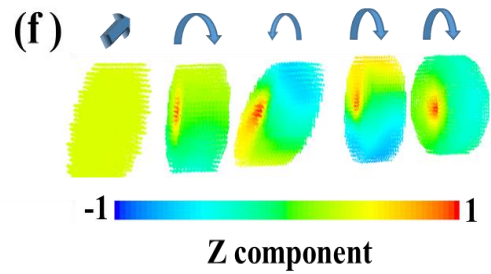
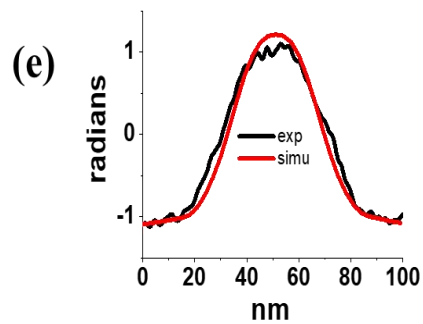
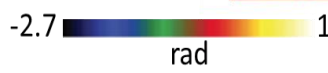
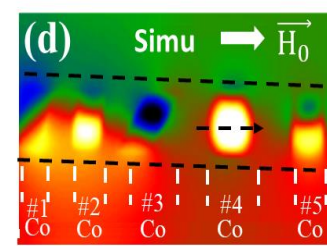
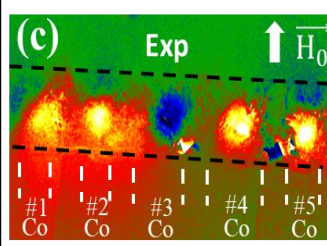
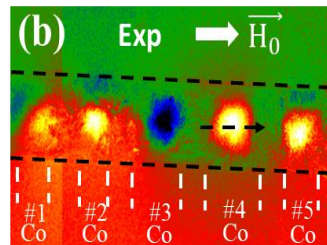
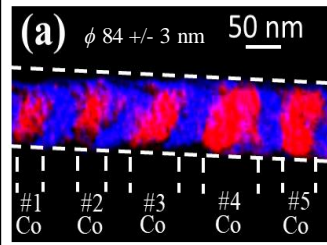


*D. Reyes et al., Nano Lett. 16, 1230, 2016.*

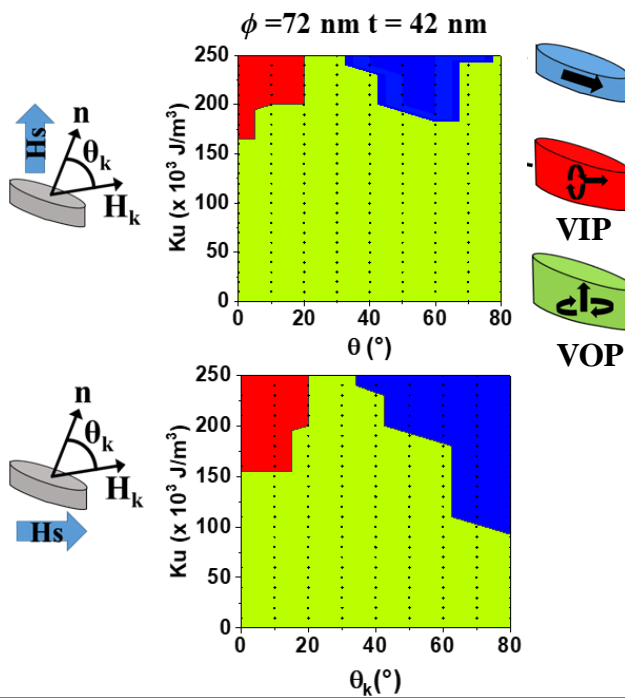
- 70% of the layers show AP configuration whatever  $H_s$  : favored by stronger dipolar coupling
- 20% of layers switch between IP and VOP states when rotating  $H_s$  (only possible if  $H_k$  close from the normal of the layers).
- 10% of out of plane state (OP) whatever  $H_s$  : impossible without dipolar coupling



## Multilayer



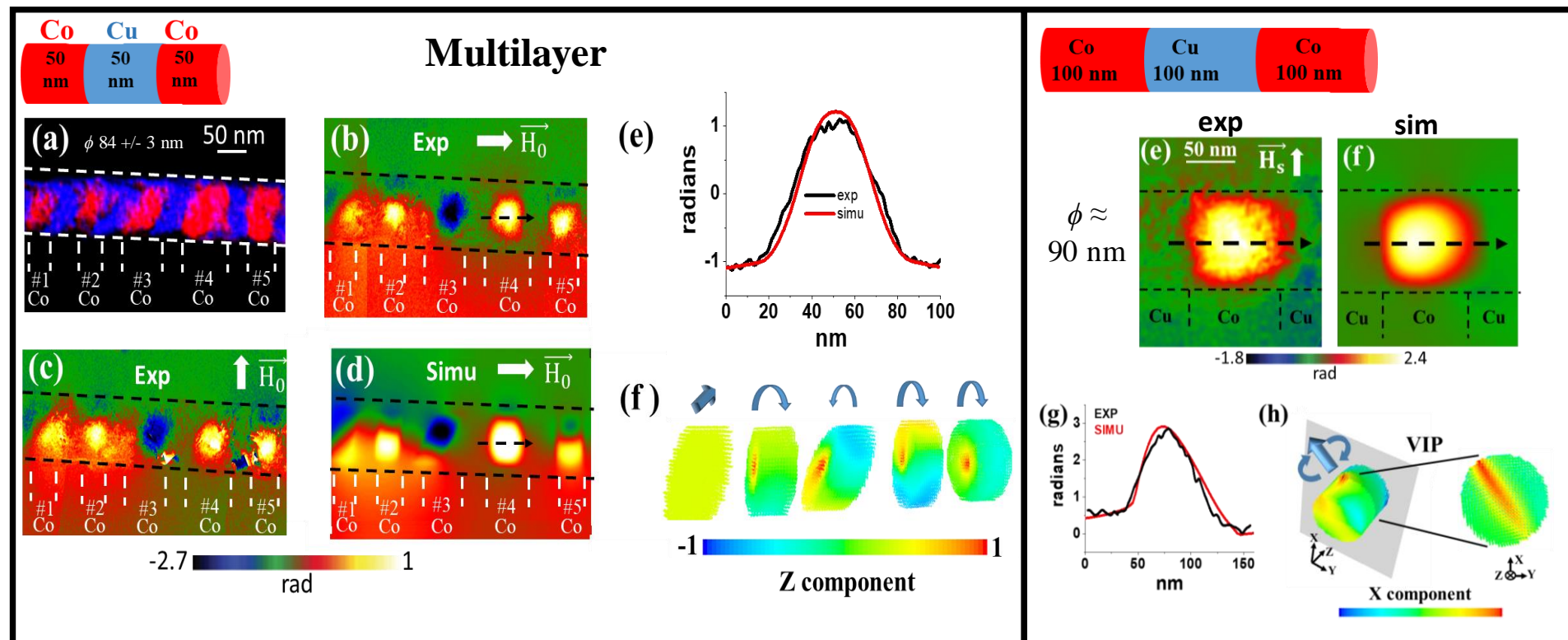
## Phase diagram of single layer



*N. Biziere et al., J. Appl. Phys. 126, 163906 (2019)*

- No effect of H<sub>s</sub> and 95% of vortex states (VOP state, with access to chirality for tilted layers)
- Simulations on multilayers show that VIP states disappear for Co<sub>50 nm</sub>/Cu<sub>50 nm</sub> because of dipolar coupling

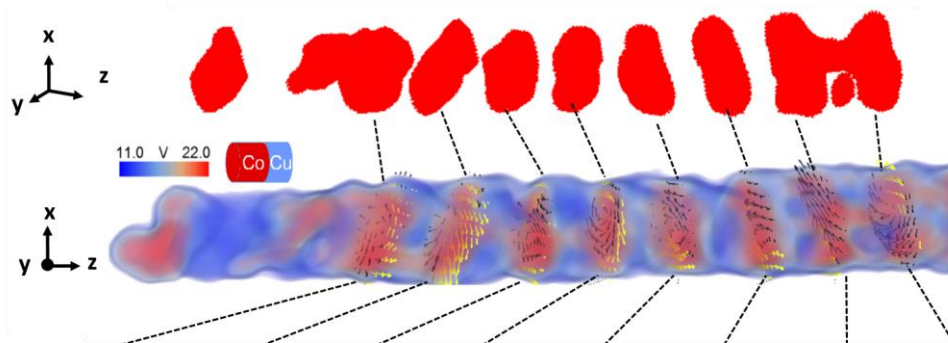




N. Biziere et al., *J. Appl. Phys.* 126, 163906 (2019)

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- Simulations on multilayers show that VIP states disappear for Co<sub>50 nm</sub>/Cu<sub>50 nm</sub> because of dipolar coupling
- VIP states observable for very thick Co layers with diameter  $\geq 80$  nm

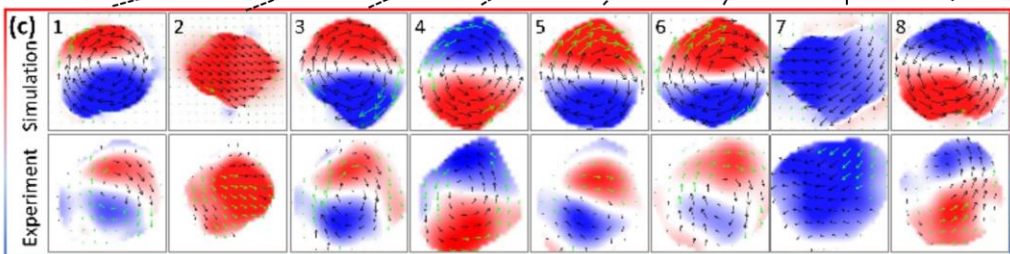
- **Principle of 3D holographic vector field electron tomography** : numerical reconstruction of 3D geometry and B field components of a nano-object via multiple tilted holograms



Implementation in OOMMF of the layer shape



Electrostatic potential of  $\text{Co}_{25\text{ nm}}/\text{Cu}_{15\text{ nm}}$  nanowire (spatial resolution 7 nm)



← Simulated B field with OOMMF

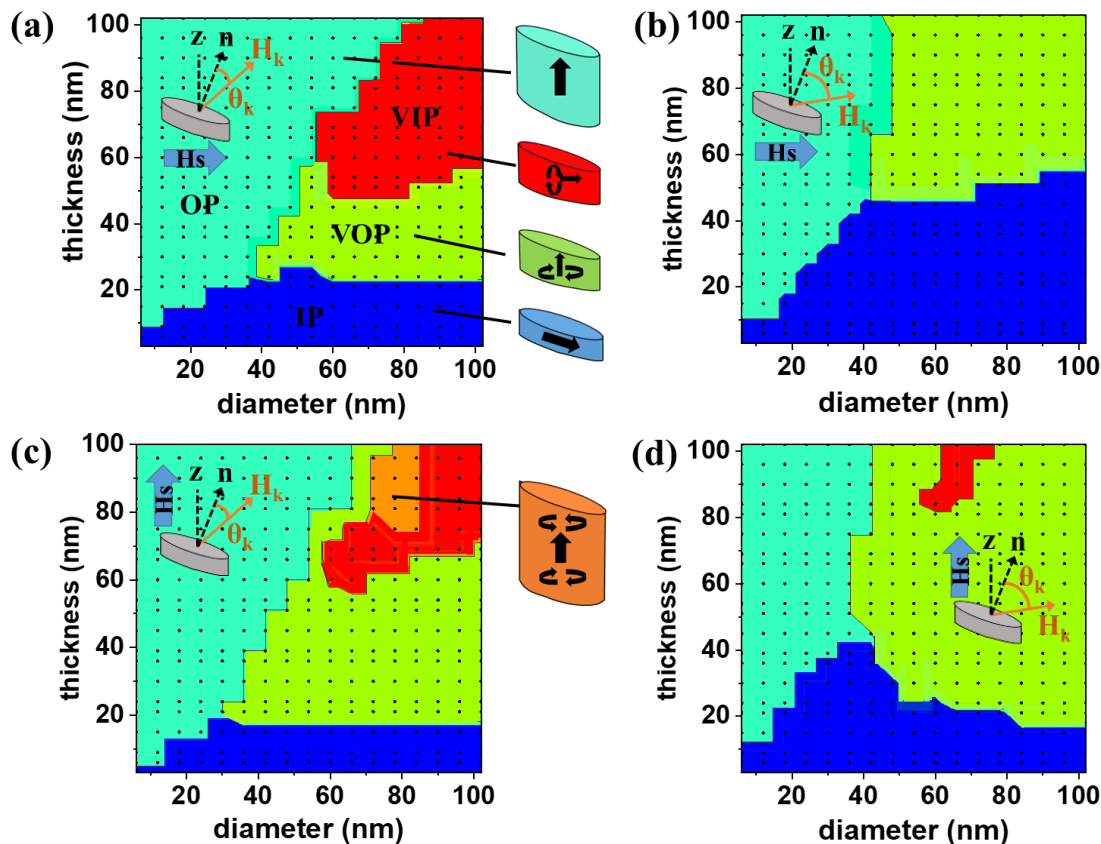
← Experimental B field recovered from 3D tomography

*D. Wolf et al., Communications Physics 2, 87, 2019.*

- Preliminary results : quantitative agreement but calculated induction overestimated (10-20%)
- Interest : removing geometrical uncertainties allow focusing on magnetic parameters of individual layers

- Due to reduced magnetic constants as compared to bulk, electrodeposited Co layers show multiple remanent states as a function of aspect ratios.
- Inter layer dipolar coupling favors antiparallel coupling of the layers but also allows transition between uniform and vortex states.  
⇒ For spin torque oscillators : possibility to obtain two different working frequencies at remanence by switching the direction of the saturation field.
- Drawback : the control of the electrodeposition conditions must be improved in order to use it in a reproducible way!

***Thank you !***

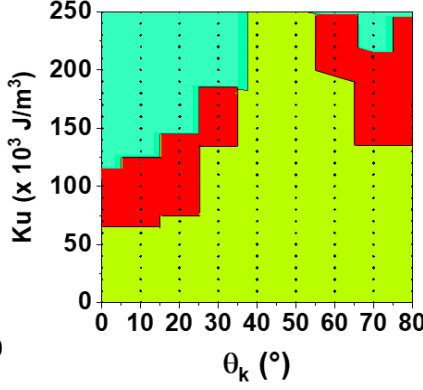
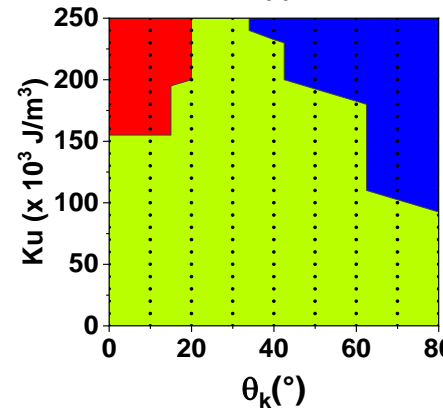
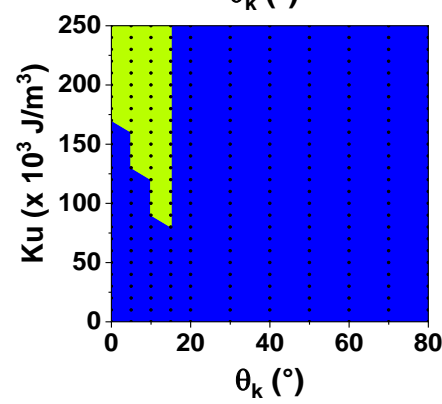
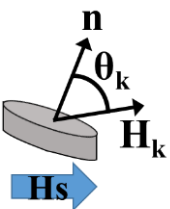
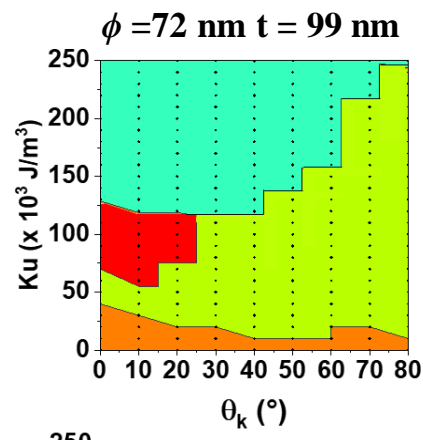
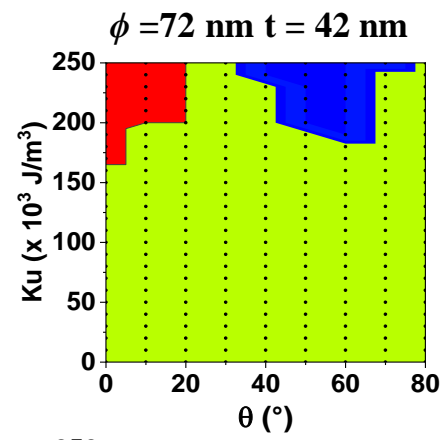
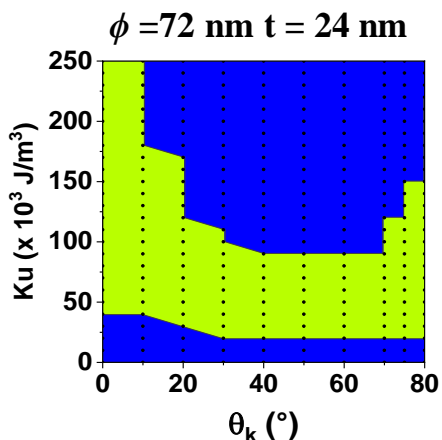
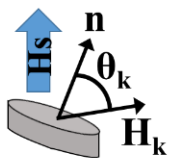


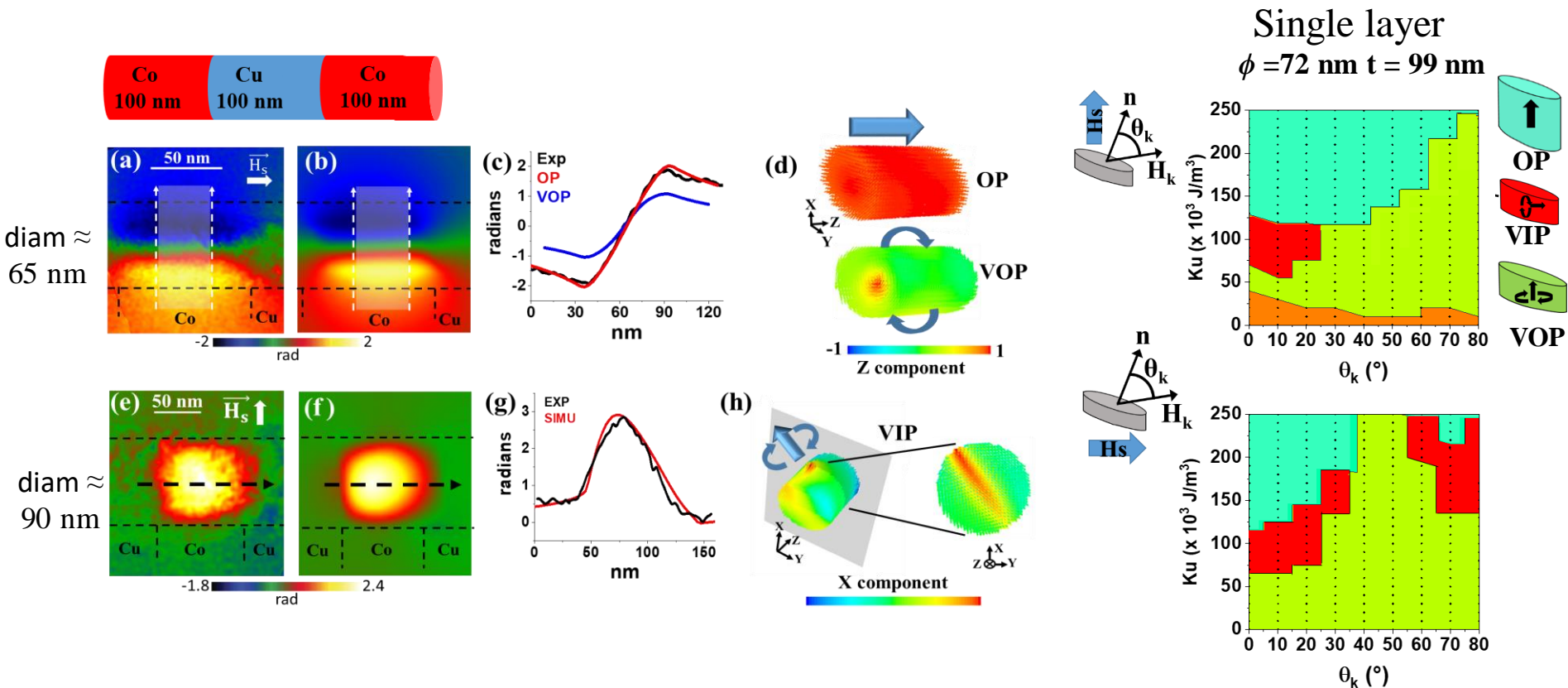
Magnetic parameters :

- $\mu_0 M_s = 1.5 \text{ T}$  (bulk = 1.7 T)
- Exchange value  $A = 22 \cdot 10^{-12} \text{ J/m}$  (bulk =  $30 \cdot 10^{-12} \text{ J/m}$ )
- $K_u = 130 \cdot 10^3 \text{ J/m}^3$

$\Rightarrow$  Values lower than bulk because of Cu impurities in Co

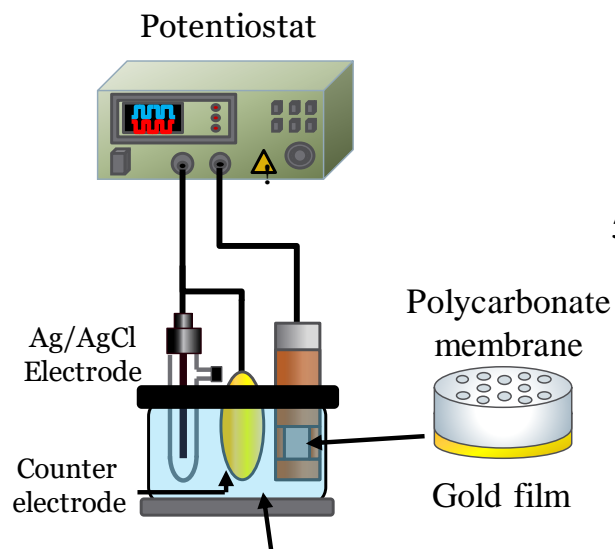
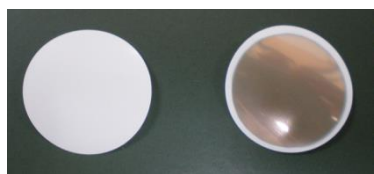
- 4 different states can be expected depending on aspect ratio
- Strong impact of  $H_k$  and  $H_s \Rightarrow$  possibility to get information on  $H_k$  by changing  $H_s$



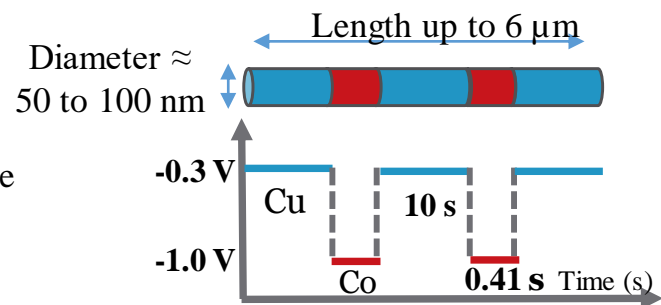


- No statistical analysis but seem that  $H_s$  has no effect.
- For diameters  $\leq 70$  nm, the dipolar coupling avoid vortex states and uniform sates (OP) aligned with the wire axis become the most stable ones for both direction of  $H_s$ .
- For large diameter ( $\approx 90$  nm) vortex states with the core perpendicular to the wire axis (VIP) become the most stable for both direction of  $H_s$

Polycarbonate Membrane



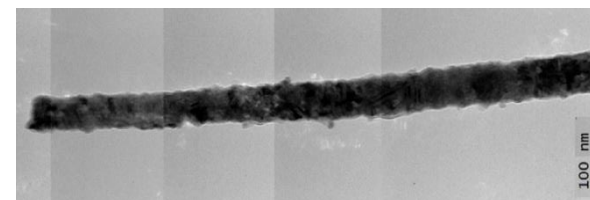
**Chemical bath composition :**  
 $\text{CoSO}_4 + \text{CuSO}_4 + \text{H}_2\text{O} + \text{H}_3\text{BO}_4$



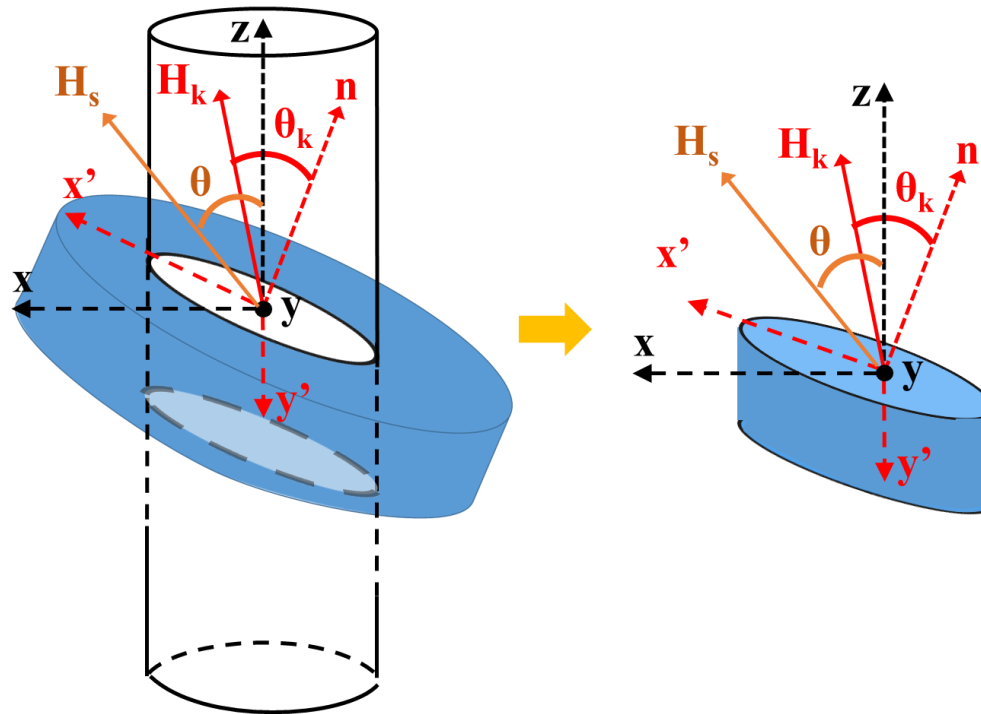
**Insertion of Cu impurities in the Co layers due to higher deposition potential**

Expected thickness Co/Cu (nm/nm)	Measured thickness Co/Cu (nm/nm)	Measured diameter (nm)
25/15	24 +/- 4 / 14 +/- 8	65 +/- 10
25/45	22 +/- 5 / 46 +/- 14	74 +/- 7
50/50	42 +/- 13 / 46 +/- 17	68 +/- 8
100/100	90 +/- 24 / 110 +/- 44	69 +/- 10

TEM analysis







**Layers like nanocylinders with tilted surfaces.**