

Laboratory for Neutronen Scattering  
ETH Zürich & Paul Scherrer Institute



Jochen Stahn

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# **Antiphase magnetic proximity effect in perovskite superconductor / ferromagnet multilayers**

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## essence

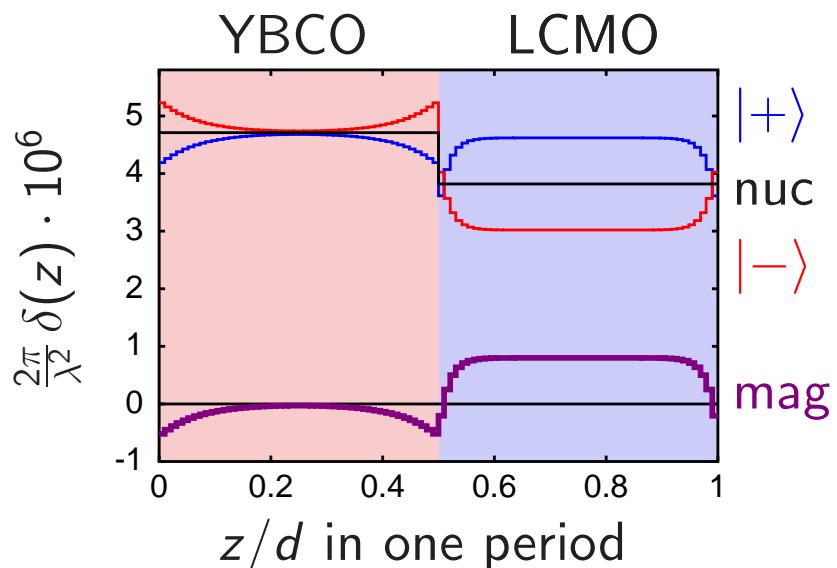
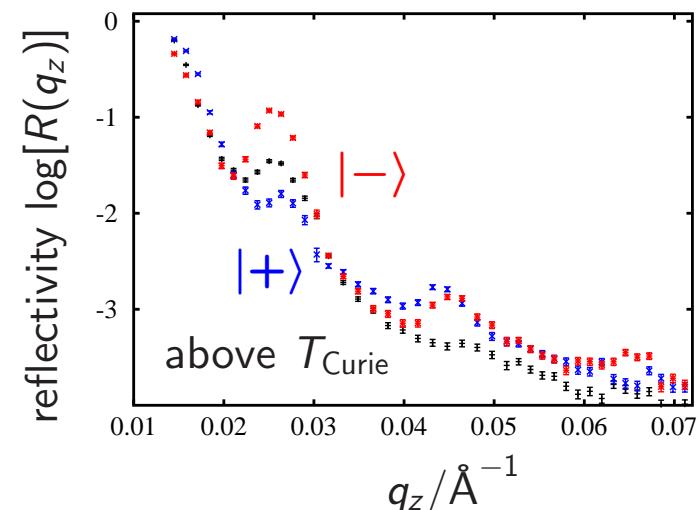
question: What is the magnetic induction (profile) in HTSC / FM multilayers?

method: polarised neutron reflectometry allows for the determination of  $\rho(z)$  and  $B_{\parallel}(z)$

answers: FM layers magnetised parallel

net magnetic moment in SC at the interfaces, antiparallel to FM magnetisation

SC creates and aligns domain walls in FM



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## cooperators:

samples  
G. Cristiani<sup>2</sup>  
HU. Habermeier<sup>2</sup>

analysis	measurements	interpretation
J. Hoppler <sup>1</sup>	C. Niedermayer <sup>1</sup>	
S. Pekarek <sup>1</sup>	E. Kenzinger <sup>5</sup>	
	J. Chakhalian <sup>2</sup>	
T. Gutberlet <sup>1</sup>		C. Bernhard <sup>3</sup>
U. Rücker <sup>5</sup>		B. Keimer <sup>2</sup>
M. Wolff <sup>4</sup>		

- <sup>1</sup> LNS, ETHZ & PSI  
<sup>2</sup> MPI-FKF, Stuttgart  
<sup>3</sup> Universität Fribourg  
<sup>4</sup> RUB / ILL  
<sup>5</sup> FZ Jülich

## interfaces and layered systems      “new physics” and “spintronics”?

general idea: the close contact of materials with different (alternative) properties might lead to **new phenomena**

e.g. – interface of SrTiO<sub>3</sub>/LaTiO<sub>3</sub> (insulators) is metallic

a multilayer **reduces the dimension and forces the interaction**  
coupling phenomena might show up

e.g. – RKKY-interaction  
– colossal magnetoresistance  
– changed characteristic temperatures

present case: multilayers of a FM with a HTSC (both metals) seem to show an metal/insulator transition in ellipsometry transition for small periods — but stay superconducting / magnetic

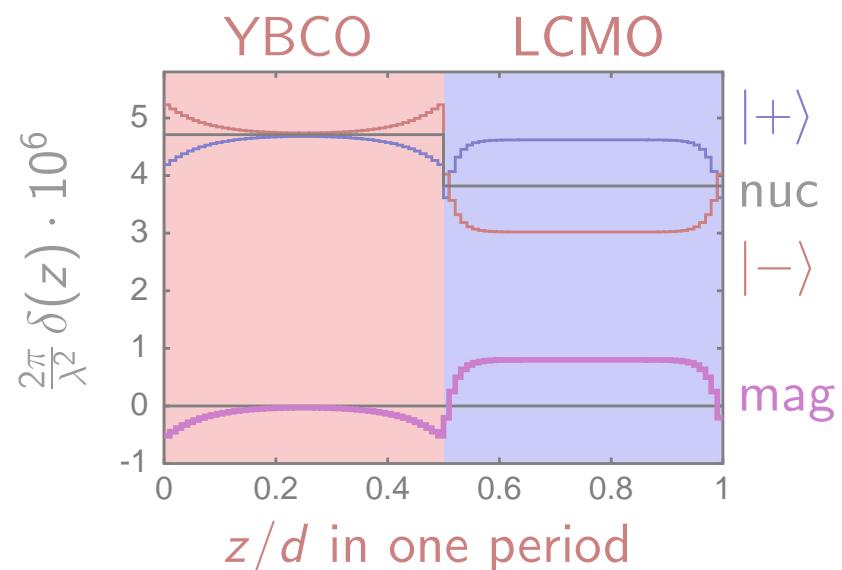
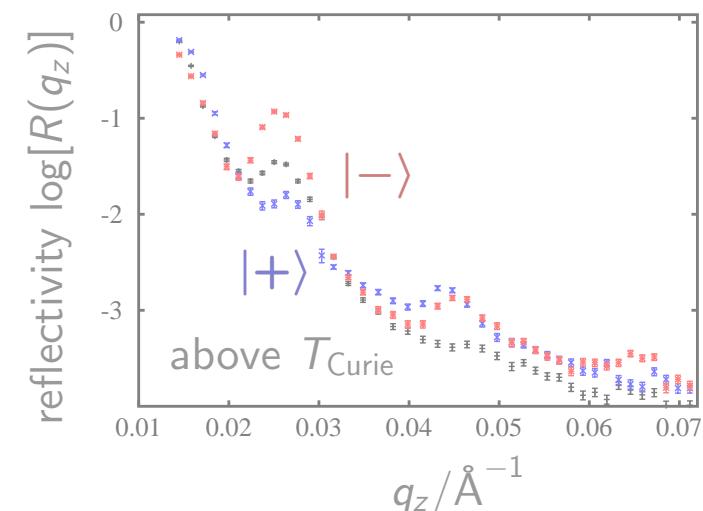
so: **what happens with the magnetisation and the superconduction order parameter?**

## overview

question: What is the magnetic induction (profile) in HTSC / FM multilayers?

method: polarised neutron reflectometry allows for the determination of  $\rho(z)$  and  $\mathbf{B}_{\parallel}(z)$

answers: FM layers magnetised parallel net magnetic moment in SC at the interfaces, antiparallel to FM magnetisation  
SC creates and aligns domain walls in FM

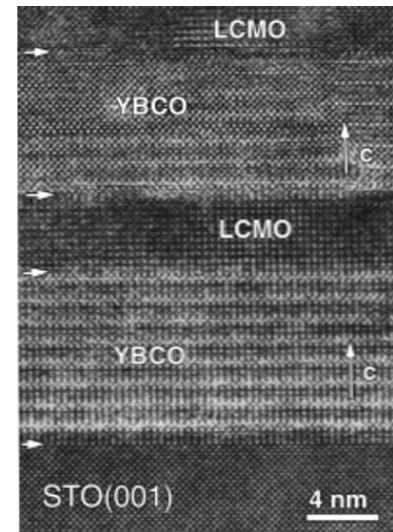
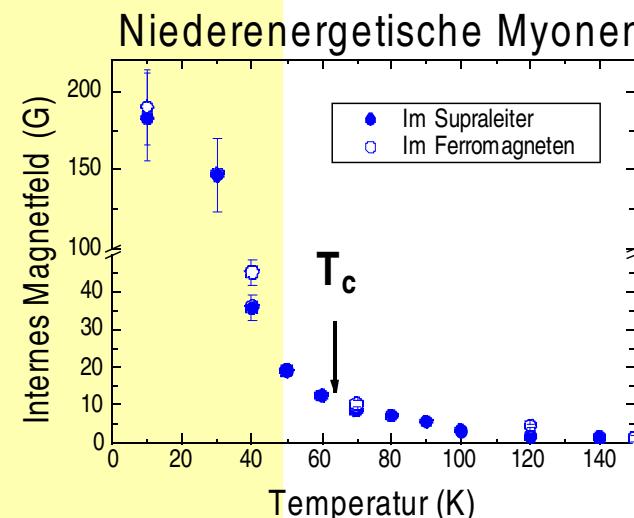
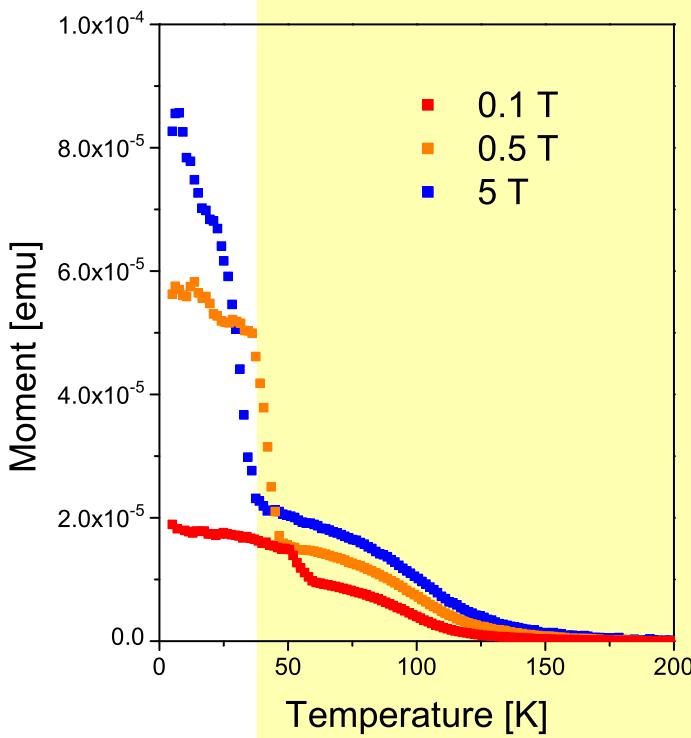


## motivation / history:

spring 2003:

C. Niedermayer presents nice  $\mu$ SR and magnetisation measurements at PSI

enhanced magnetism below  $T_c$



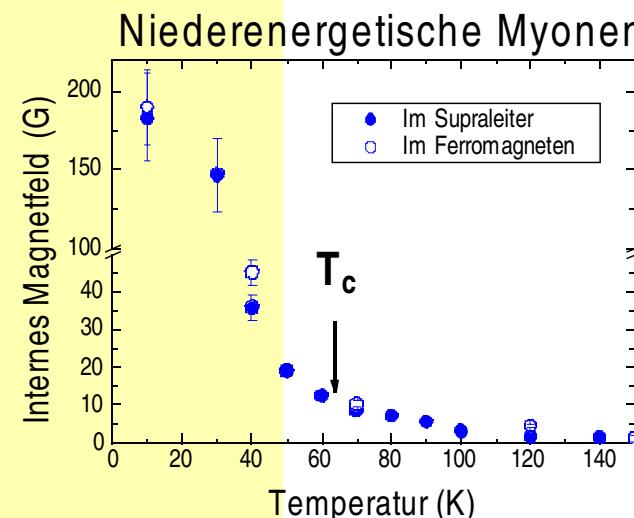
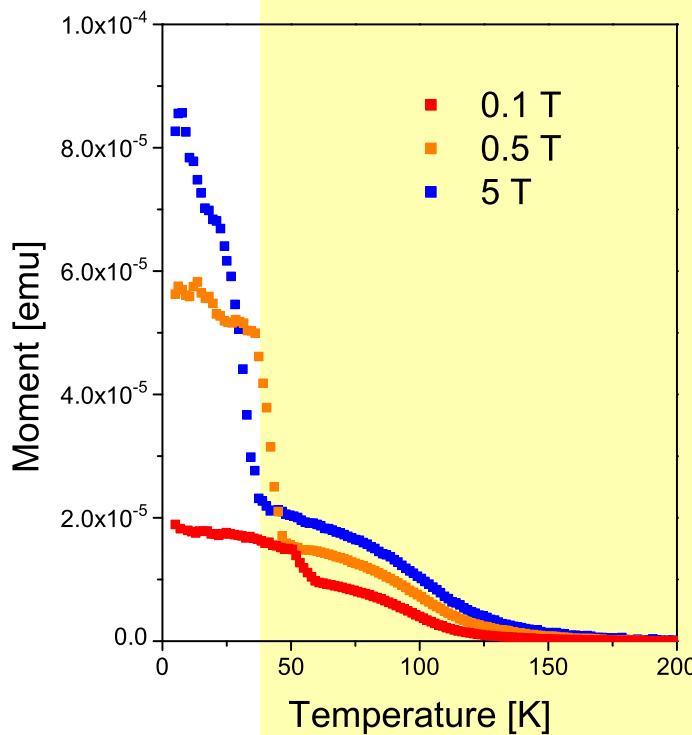
coexistence of FM and SC in RuSrCuGdO  
 → competitive order parameters  
 artificial multilayers to investigate  
 – interaction of FM and SC at the interfaces and  
 – coupling through the layer

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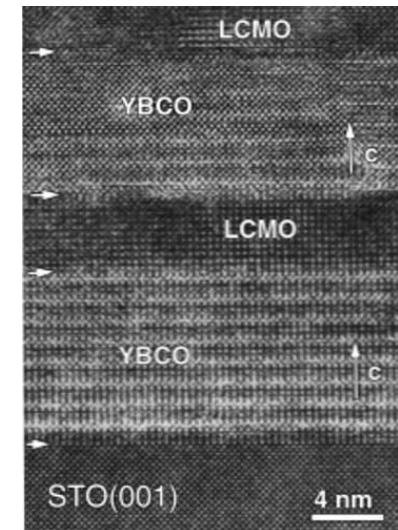
enhanced magnetism below  $T_c$



method of choice  
(for a neutron scatterer):

neutrons!

in particular *polarised n-reflectometry*



## overview

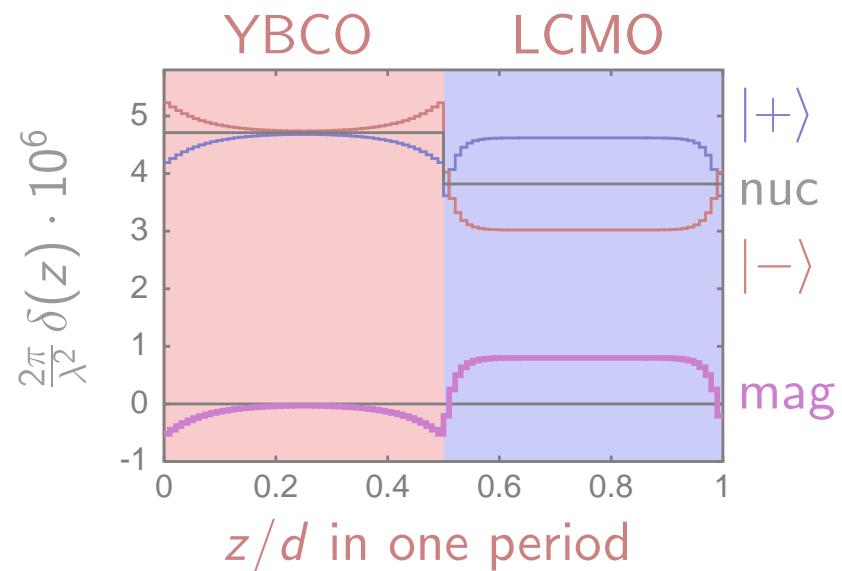
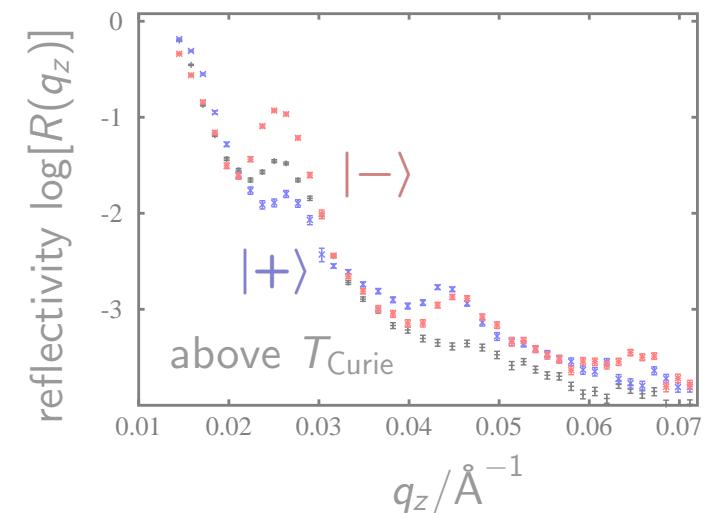
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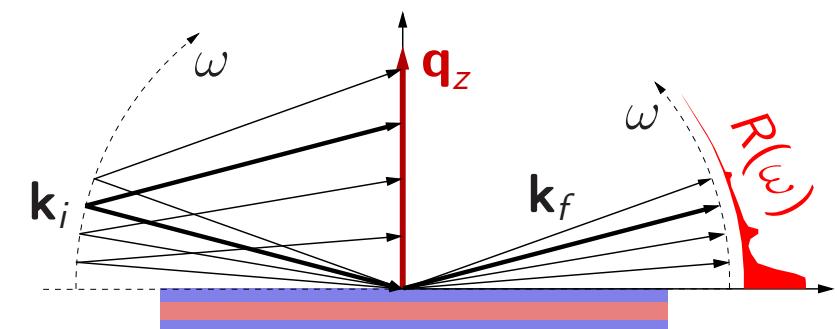
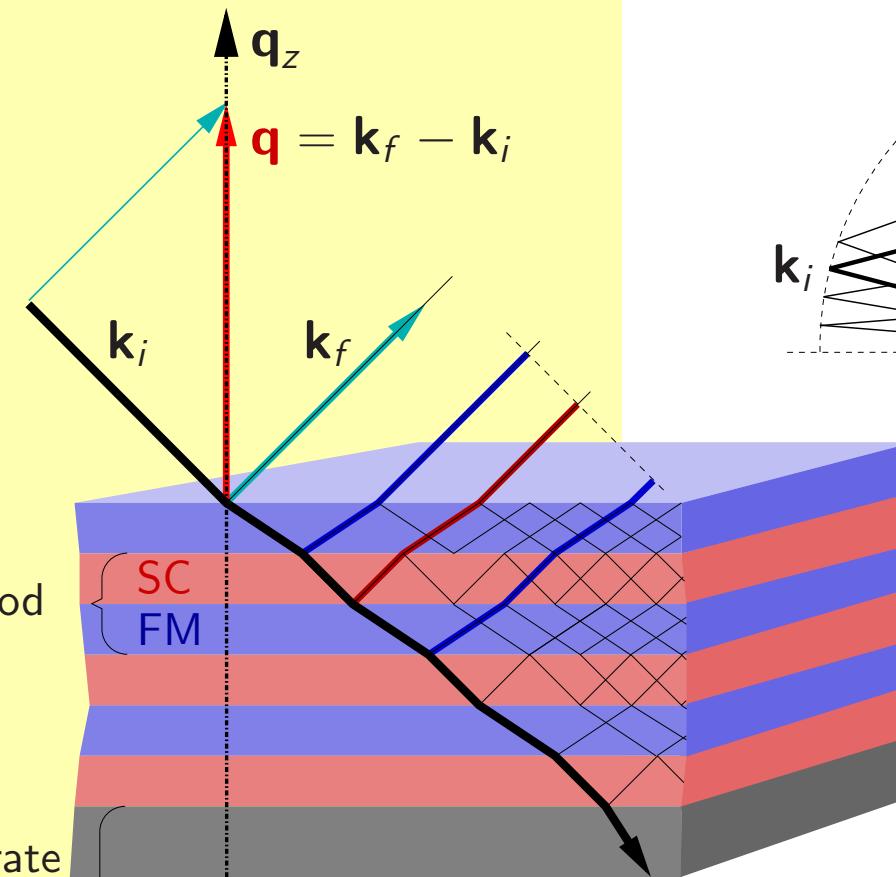
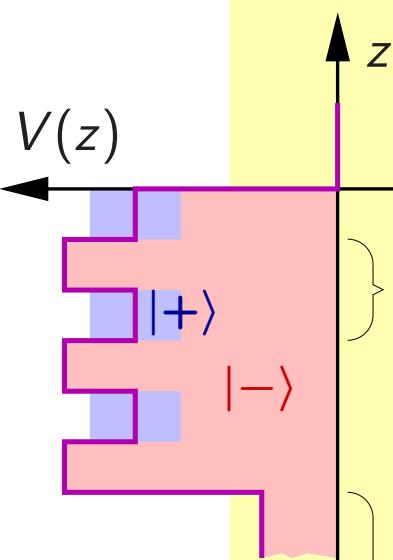
# reflectometry

interference of beams reflected from parallel interfaces

periodic structure  $\Rightarrow$  Bragg-condition for constructive interference

scattering potential:

$$\begin{aligned} V &= V_{\text{nuc}} \pm V_{\text{mag}} \\ &= V_{\text{nuc}} + \mu \mathbf{B}_{\parallel} \end{aligned}$$



layer thickness ratio 1:1  
 $\Rightarrow$  extinction of even Bragg-peaks

## reflectometry      tailored samples

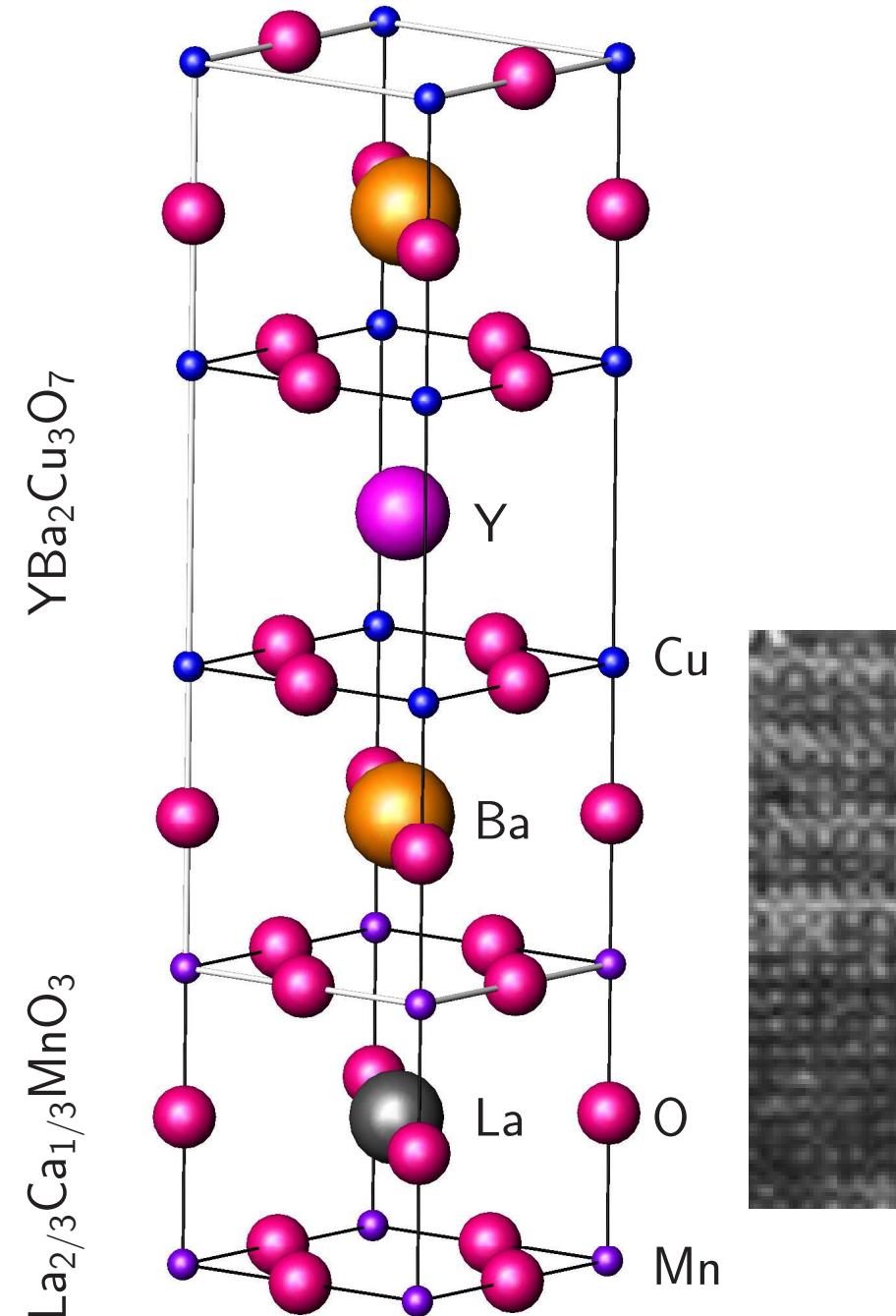
materials: HTSC YBCO  $\text{YBa}_2\text{Cu}_3\text{O}_7$   
               FM LCMO  $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$   
               substr. STO  $\text{SrTiO}_3$

size:  $10 \times 10 \text{ mm}^2$   
       (instead of  $5 \times 5 \text{ mm}^2$ )

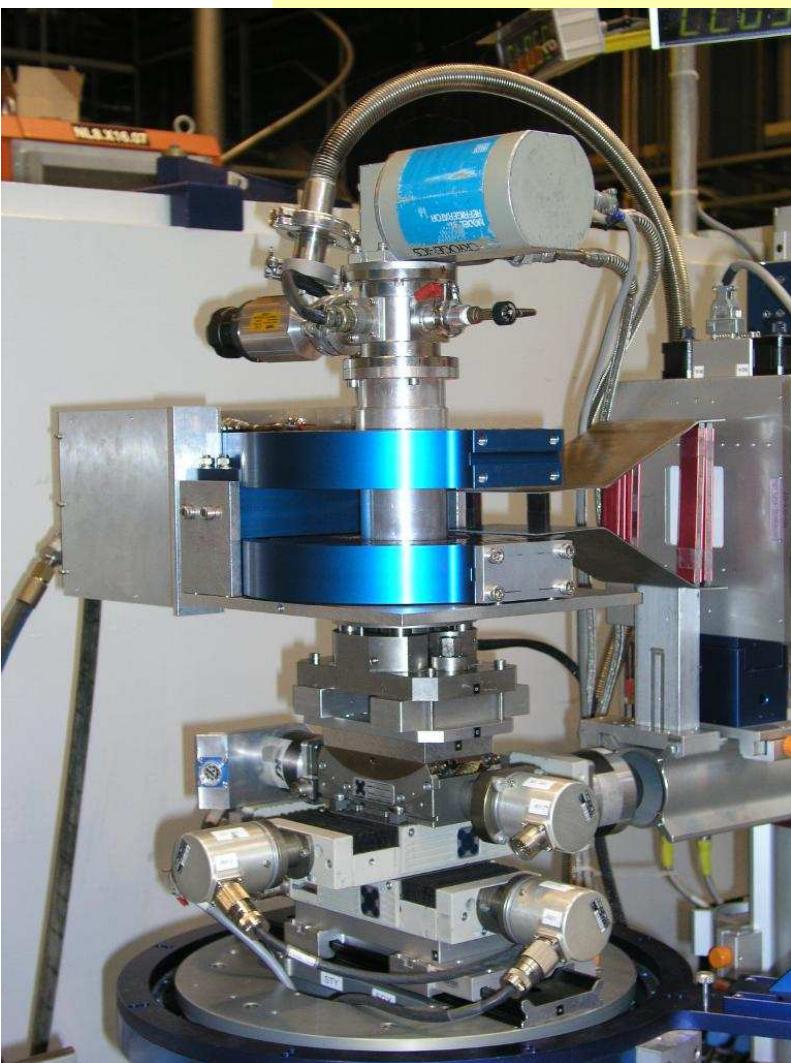
produced: by *Pulsed Laser Deposition*

period:  $200 \text{ \AA}$  to  $500 \text{ \AA}$   
       5 to 16 periods

ratios: 1 : 1 and 1 : 2  
       to cause extinction  
       non-rough interfaces  
       (otherwise used to tune  $T_c$ )



## reflectometry sample environment (at SINQ):



closed cycle refrigerator  
 $8\text{ K} < T < 300\text{ K}$

Helmholtz coils  
 $H \leq 1000\text{ Oe}$   
vol:  $40 \times 40 \times 40\text{ mm}^3$

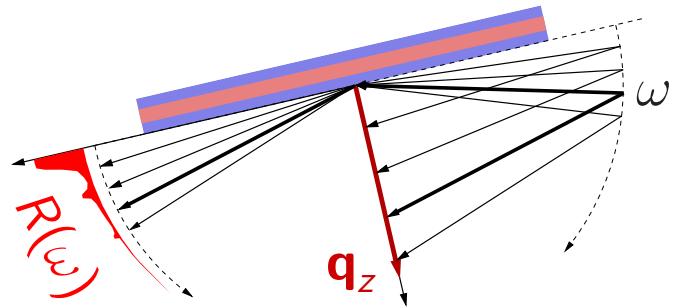
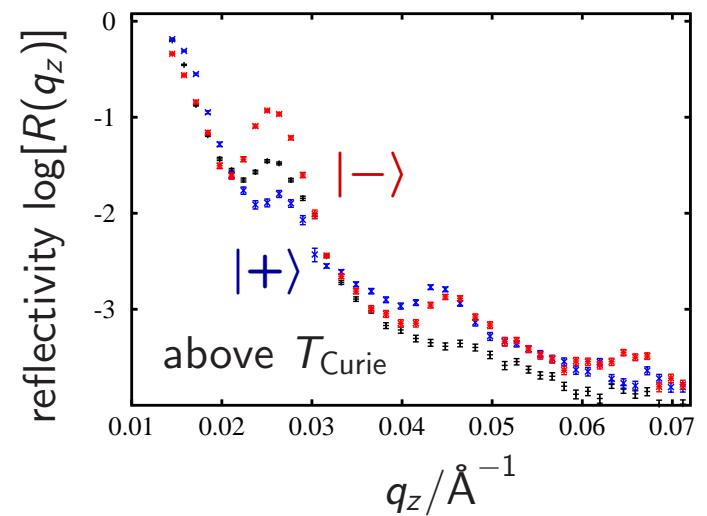
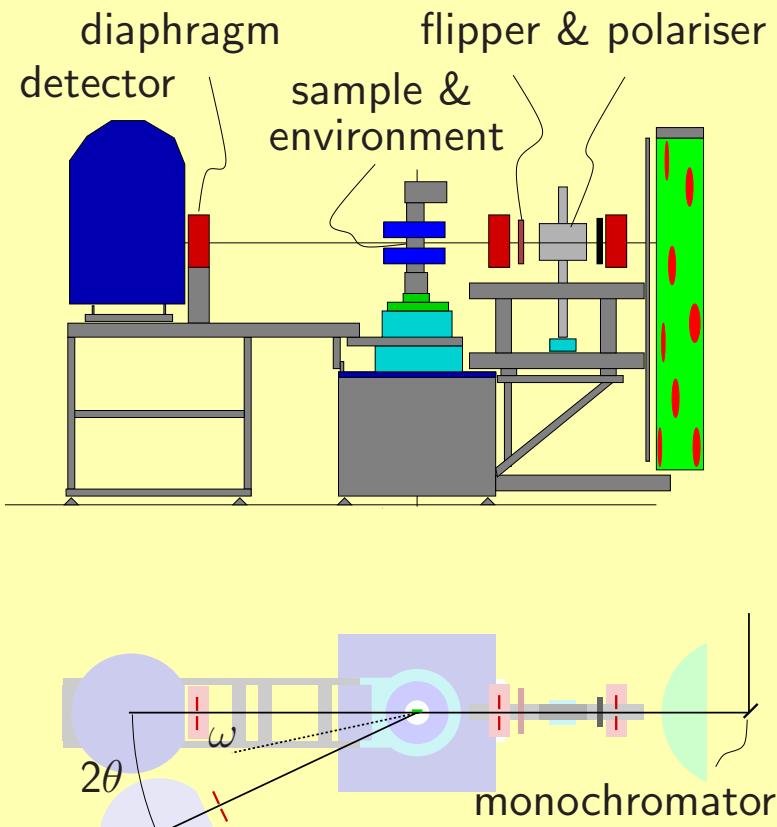
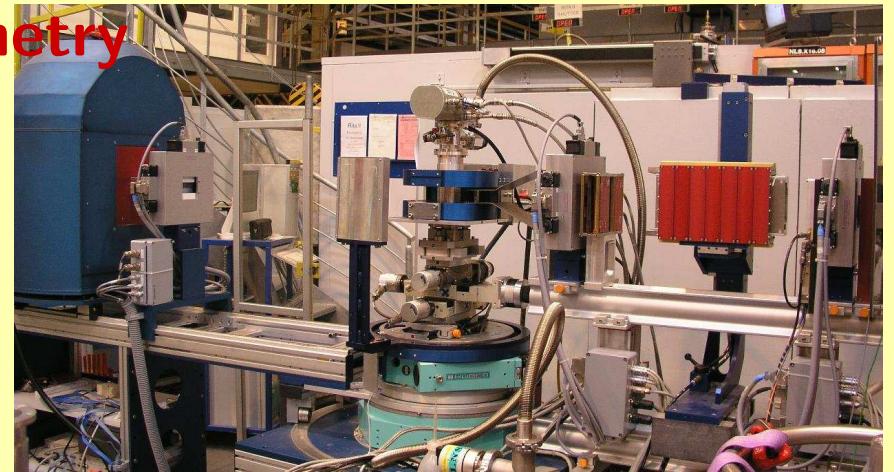
sample holder  
with absorber



translation stages for alignment  
 $\omega$ -rotation stage

# reflectometry

example:  
Morpheus @ SINQ



## overview

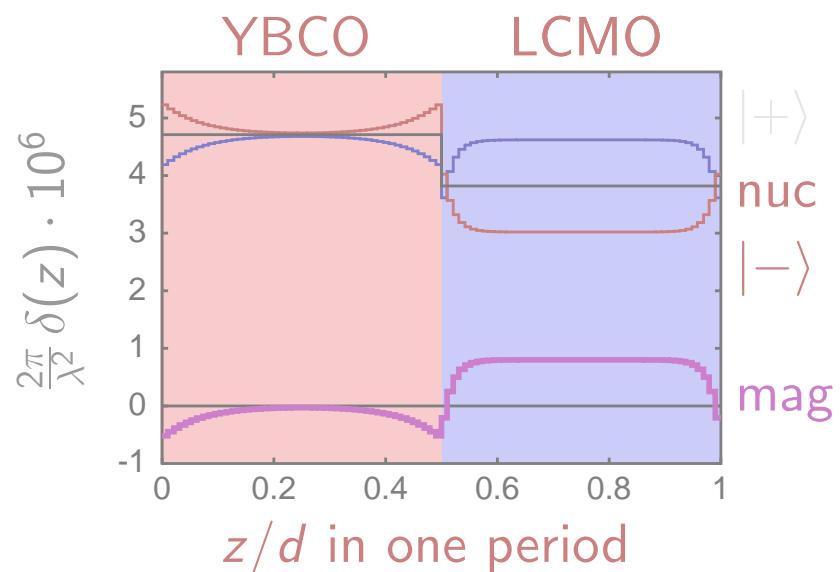
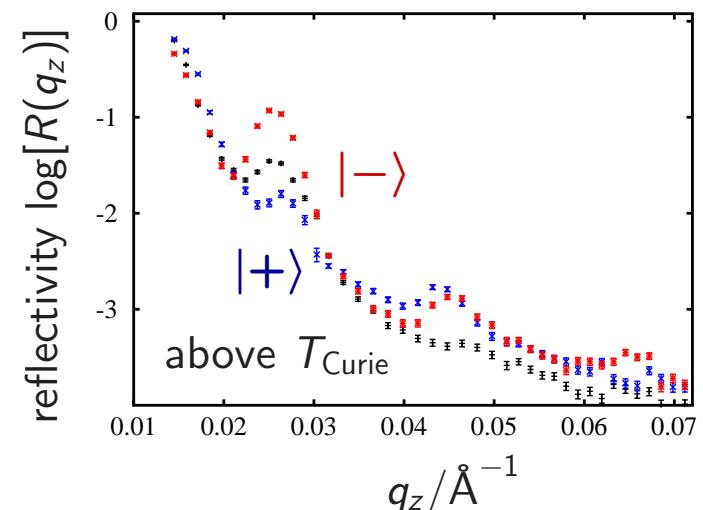
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## reflectometry

## direct interpretation

$H = 100$  Oe

field cooled

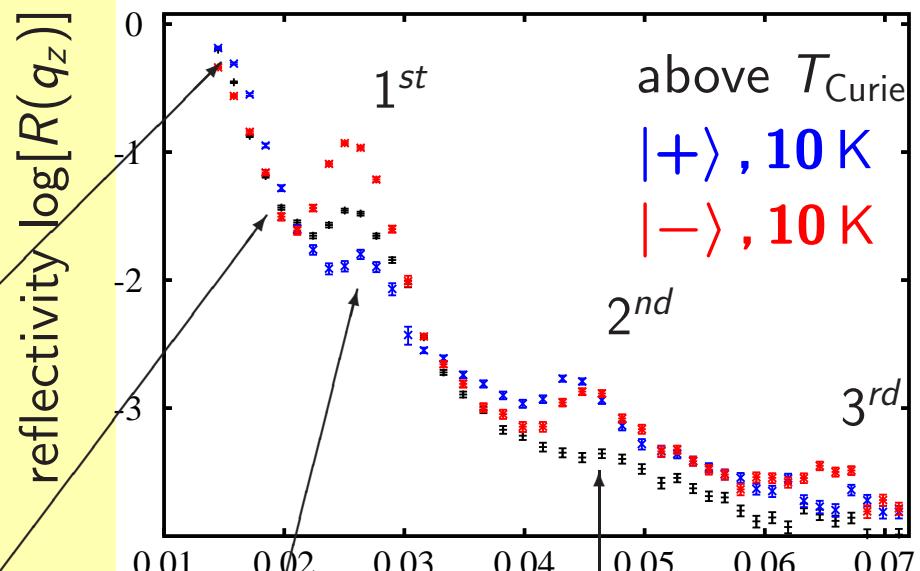
$T = 10, 300$  K

splitting of the edge of total reflection  
 $\Rightarrow$  changed potential of the surface

no half-order Bragg-peak  
 $\Rightarrow$  parallel alignment of  $\mathbf{B}$  in the FM layers

intensity variation of the  $1^{st}$  Bragg-peak  
 $\Rightarrow$  changed potential in the FM layers  
 $B_{\parallel}$  can be determined

appearance of a  $2^{nd}$  order Bragg-peak  
 $\Rightarrow B_{\parallel}(z)$  and  $V_{\text{nuc}}(z)$  have different symmetry



## reflectometry simulations

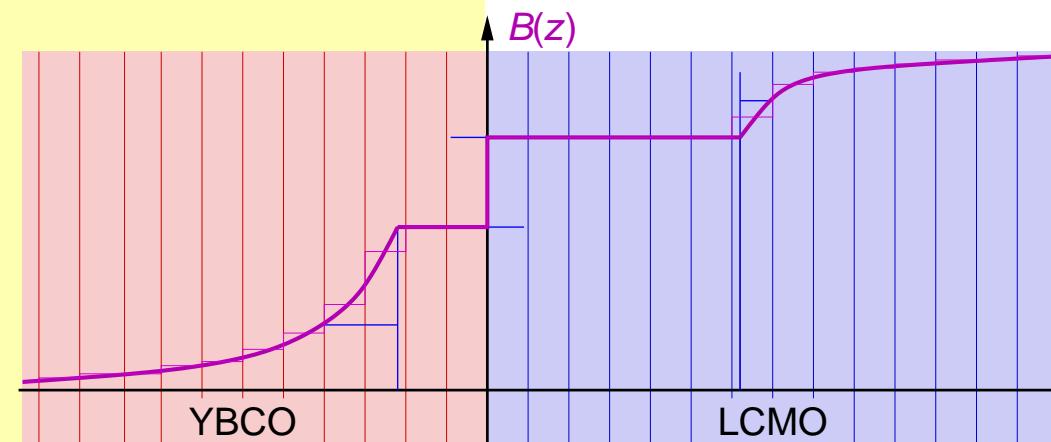
**simulations** performed with EDXR by Petr Mikulík (no fitting)

bilayer structure has been broken down to some 100 sublayers to pay respect to  $\mathbf{B}(z)$ .

analytic expressions for  $B(z)$ :

cosh-functions

off-sets with constant  $B$



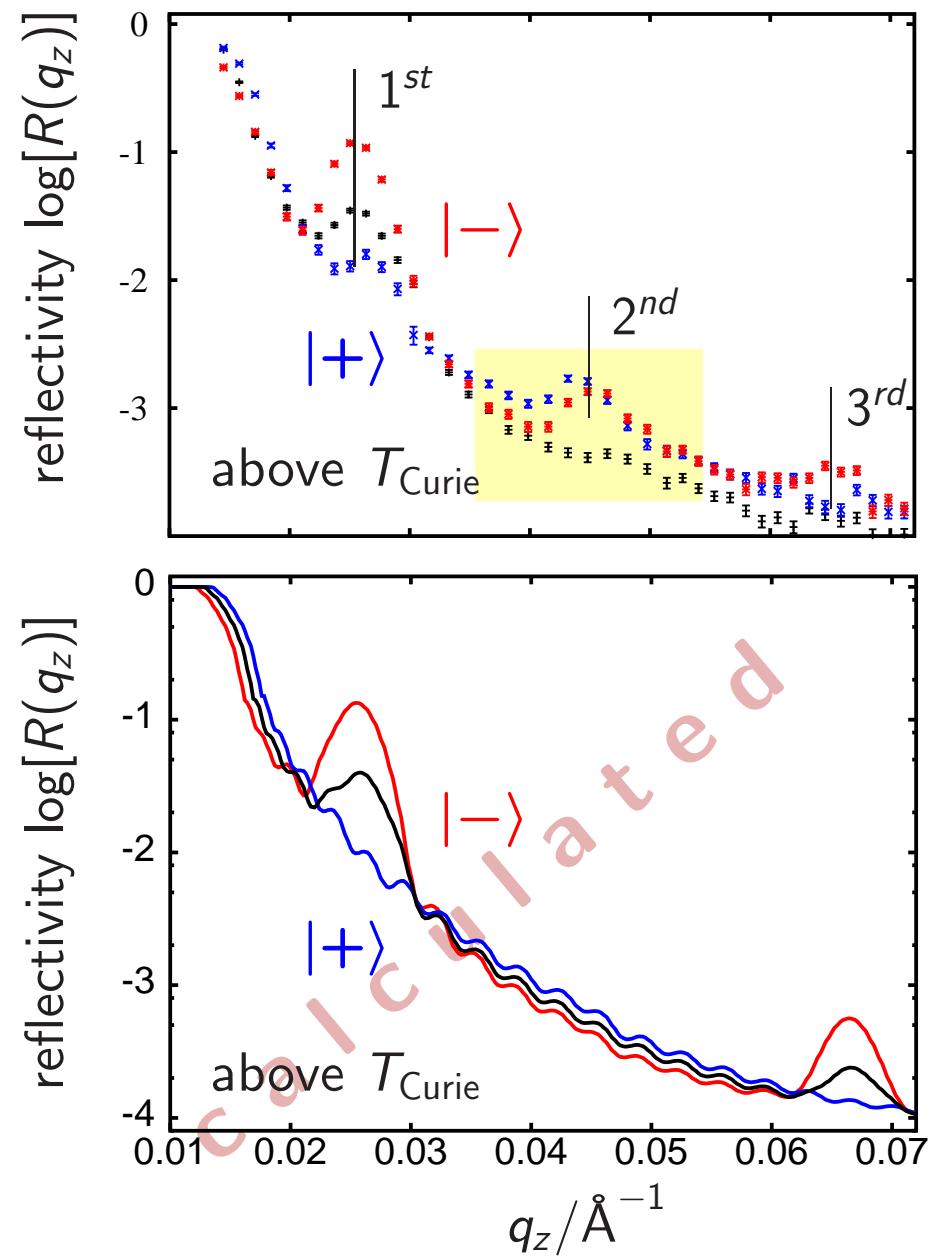
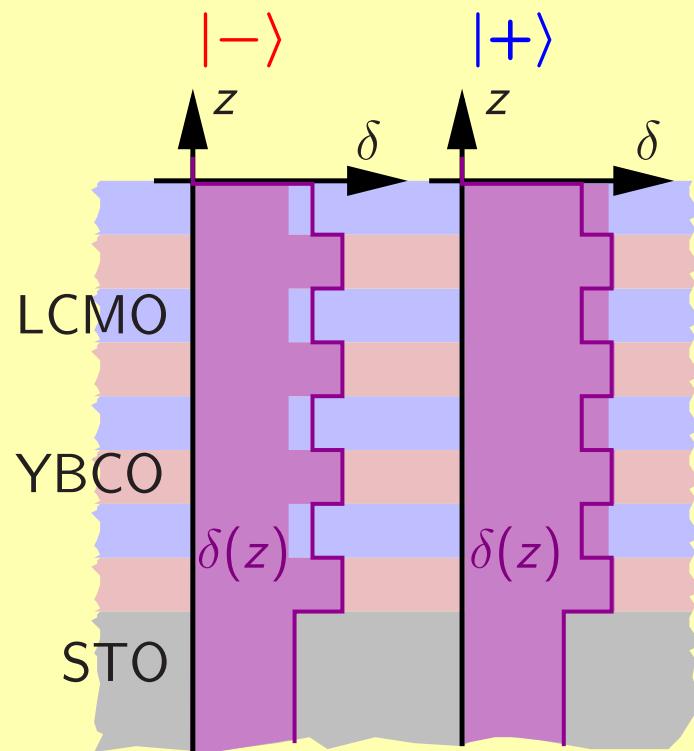
decrease of layer thickness towards the borders taken into account



# reflectometry simulation specular, polarised

sample:

[YBCO(150 Å)/LCMO(140 Å)]<sub>5</sub>

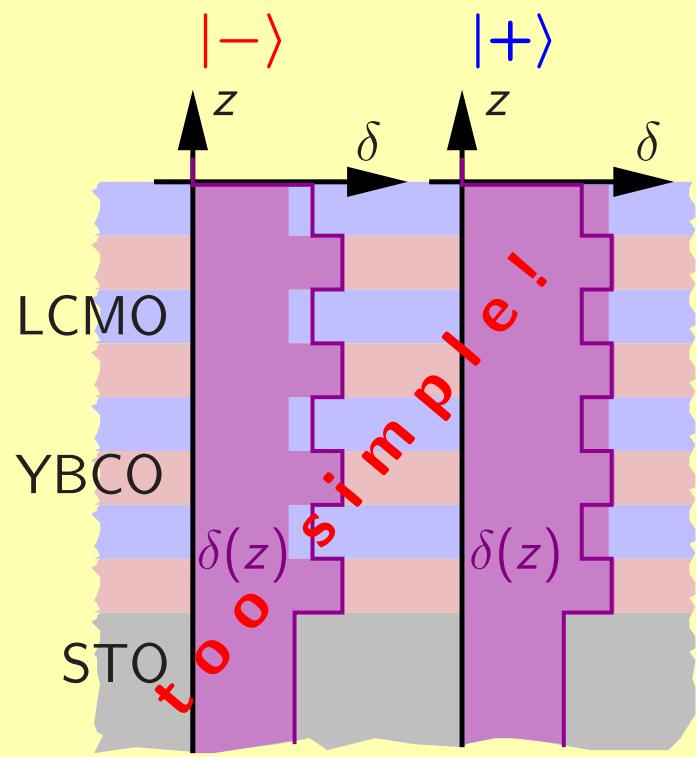


## reflectometry

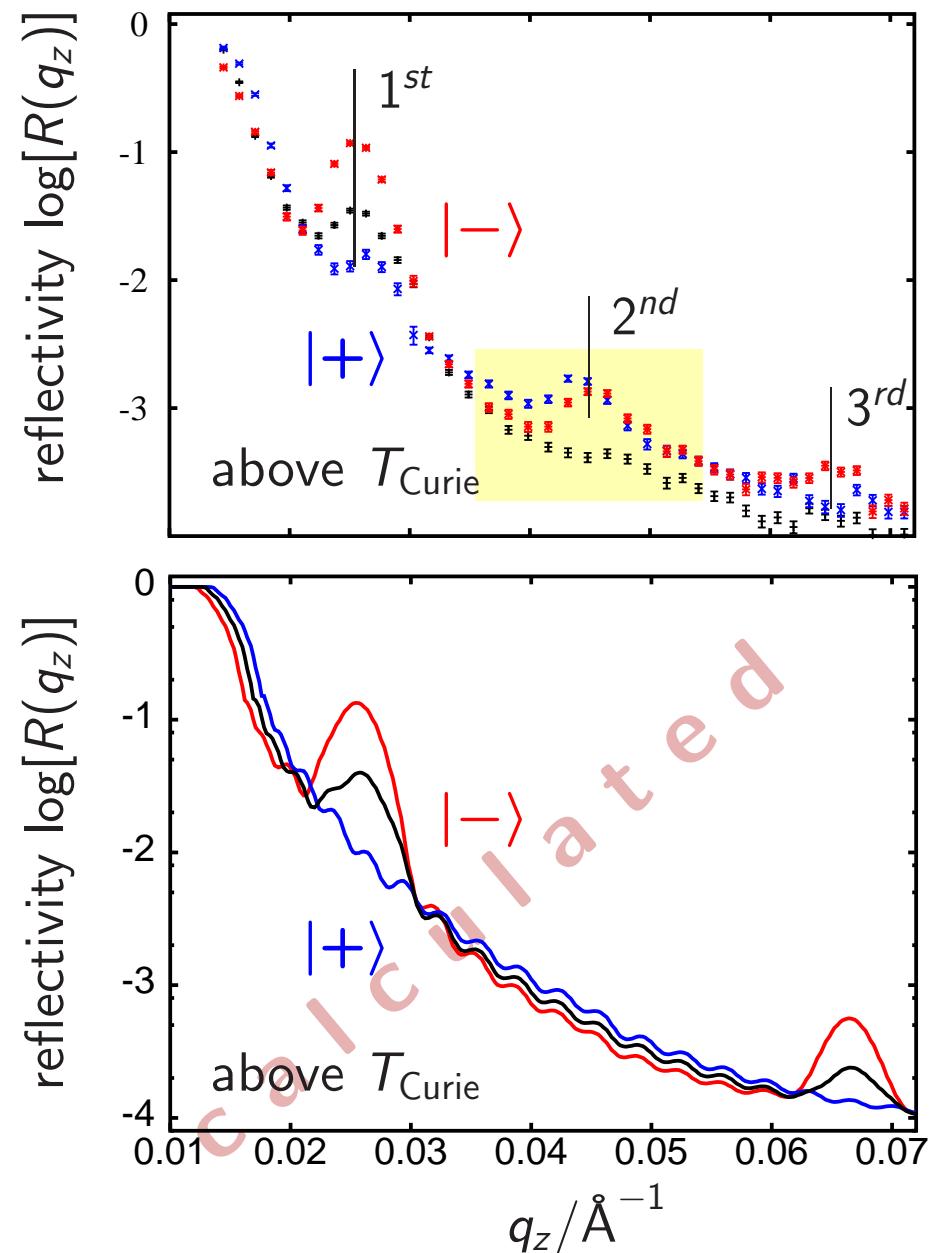
specular, polarised

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[YBCO(150 Å)/LCMO(140 Å)]<sub>5</sub>



$$\delta_{\text{mag}}(z) \neq \delta_{\text{nuc}}(z) \times \begin{cases} 0 & \text{for YBCO} \\ \text{const} & \text{for LCMO} \end{cases}$$



# modelling magnetic profile at the interfaces

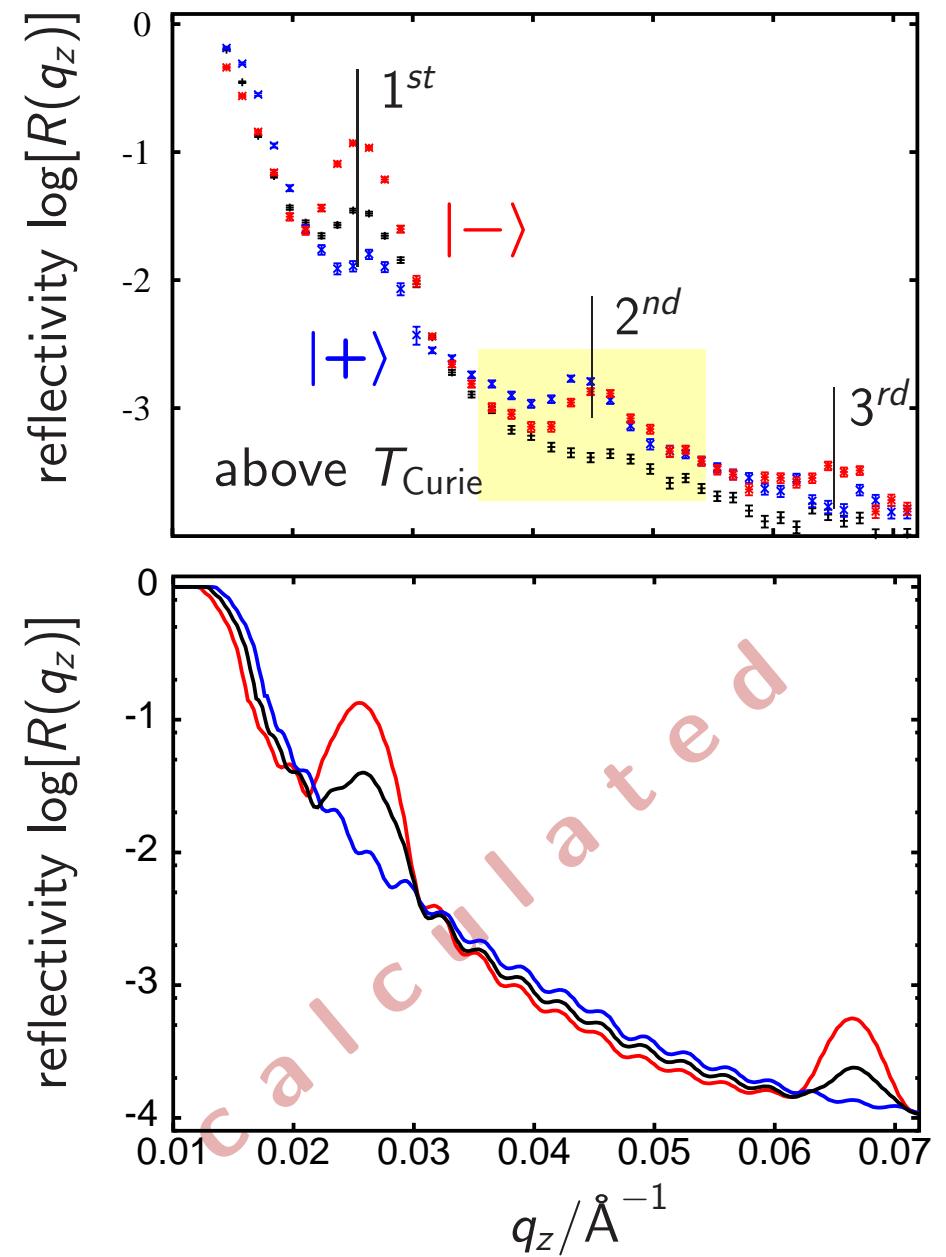
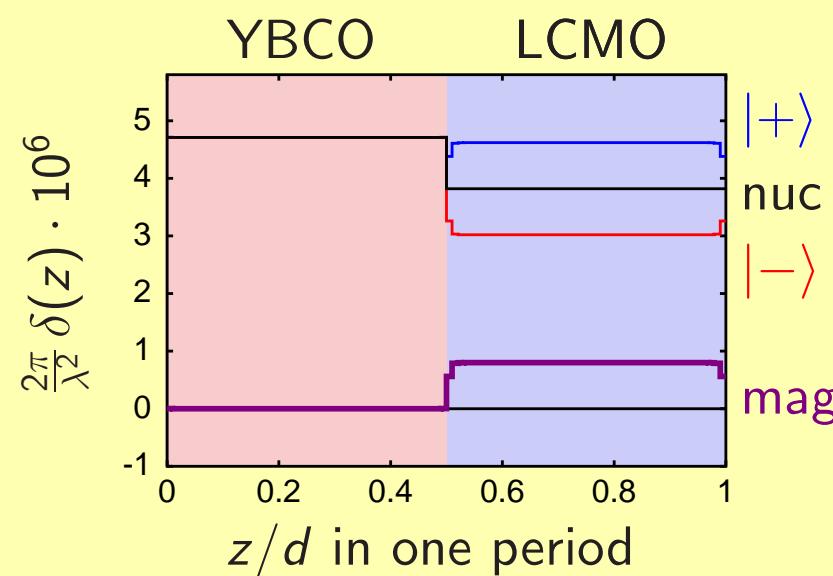
sharp contrast at the interface

exponential decay into YBCO

AFM exponential decay into YBCO

penetration into YBCO

magnetically dead layer in LCMO



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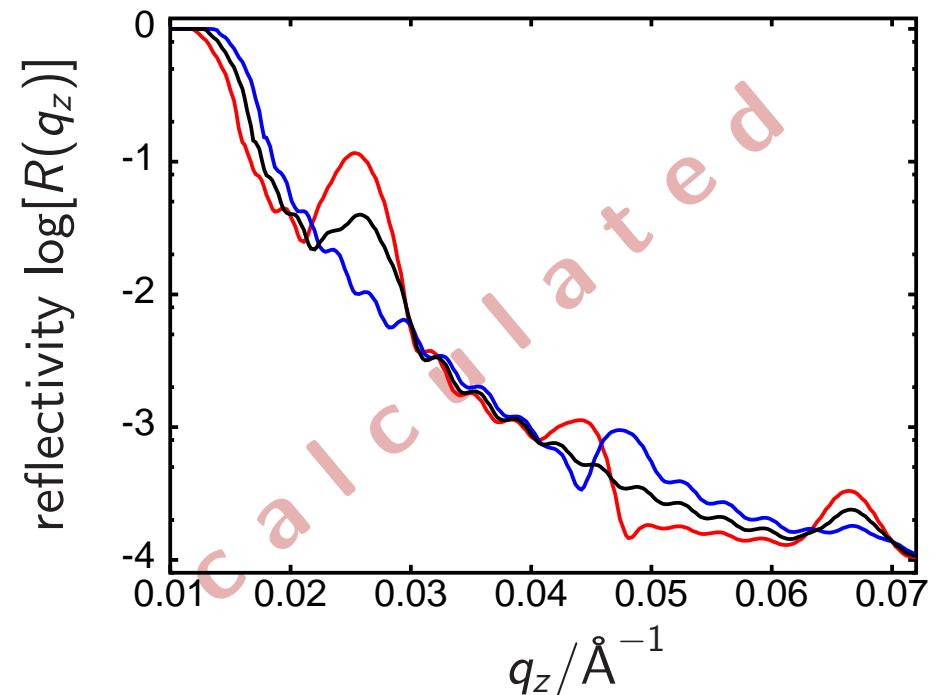
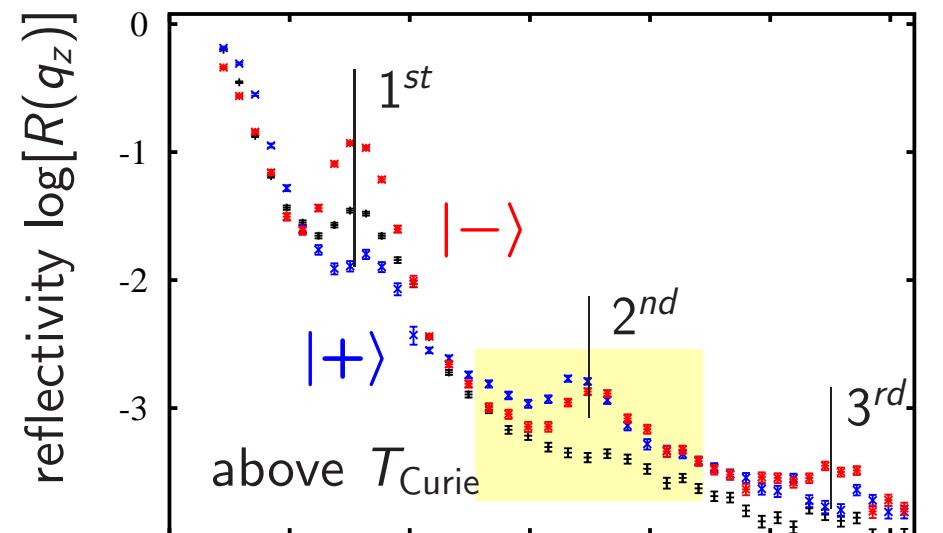
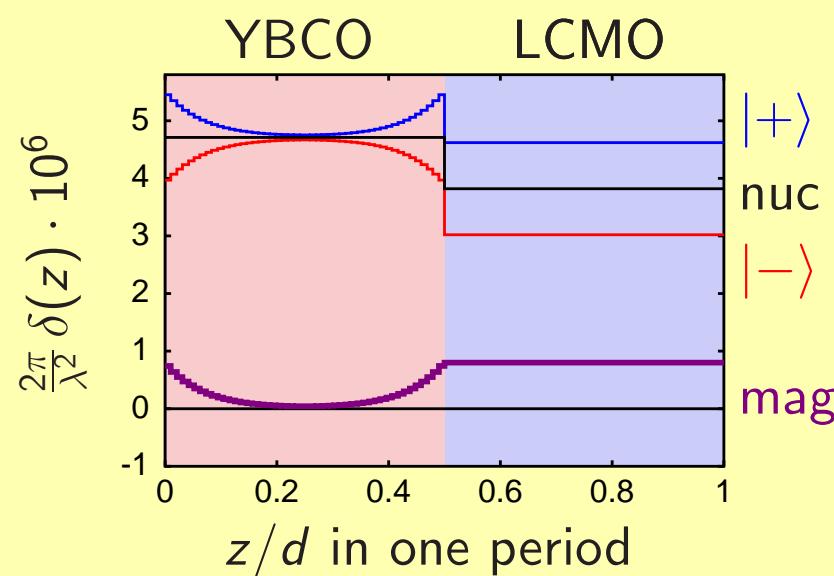
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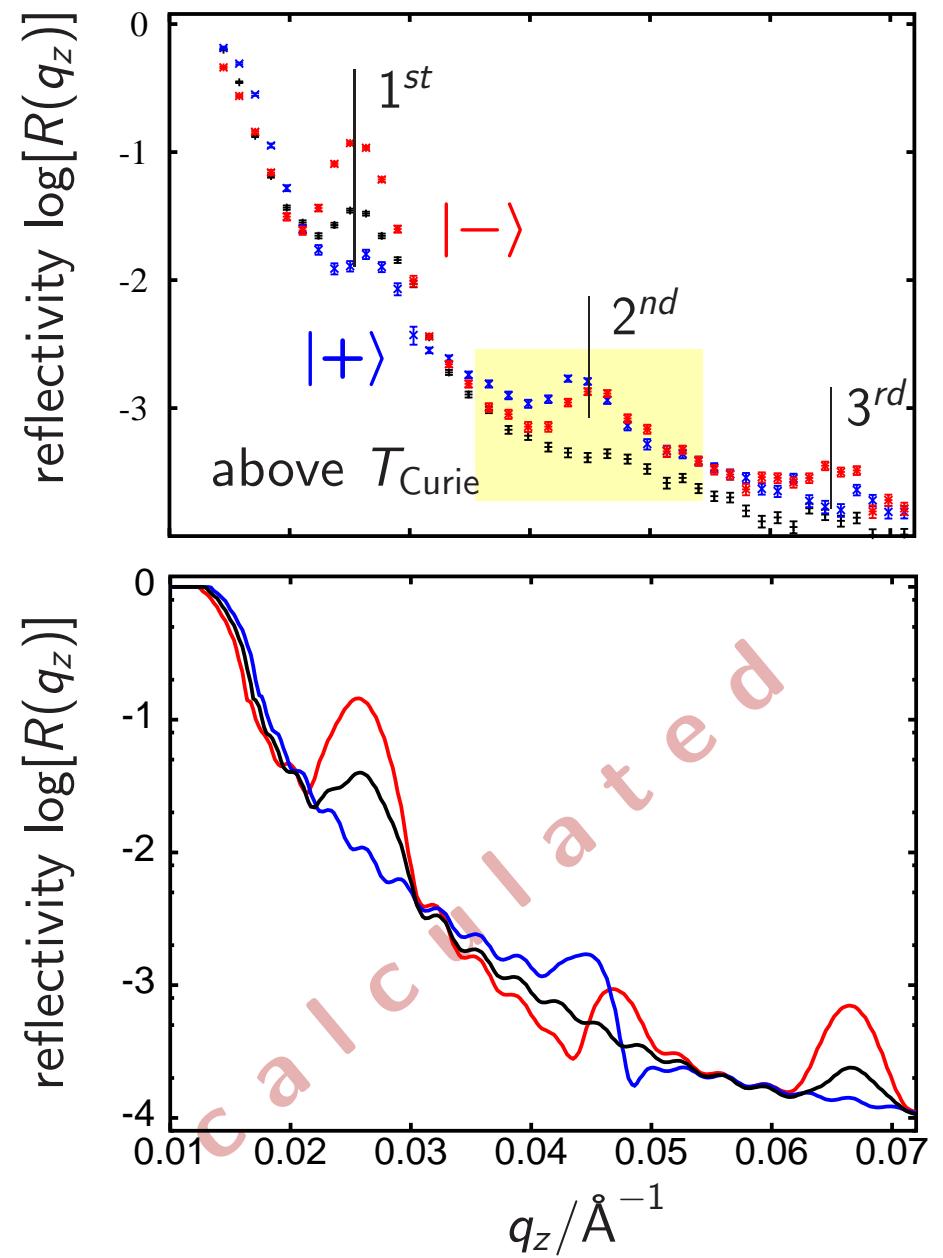
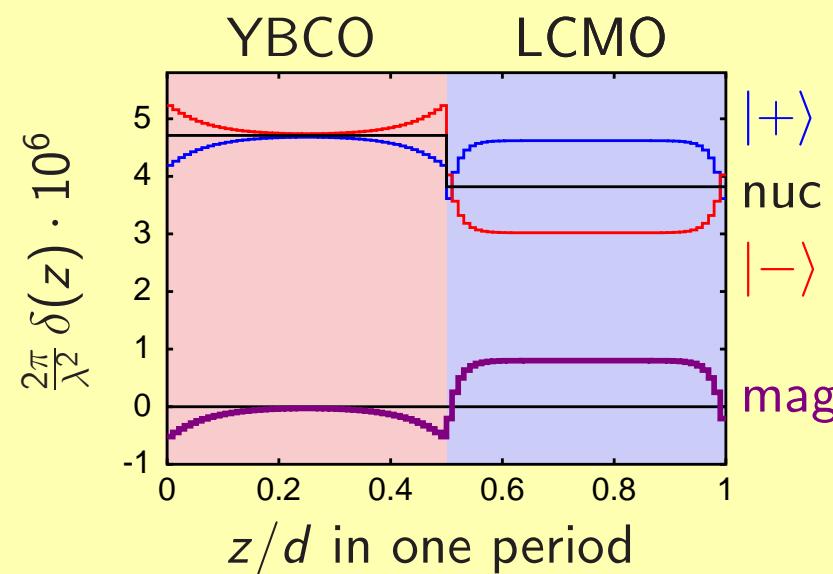
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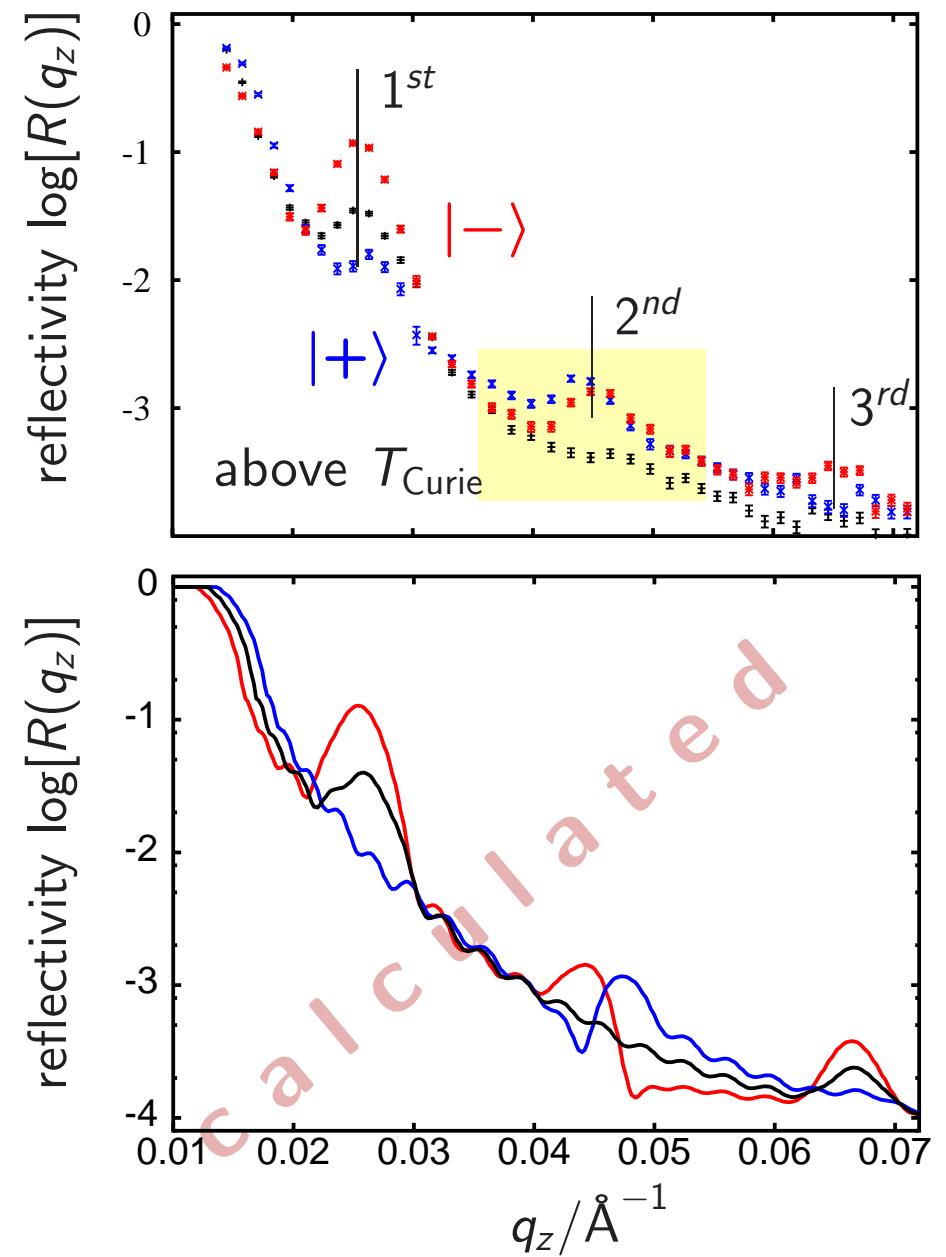
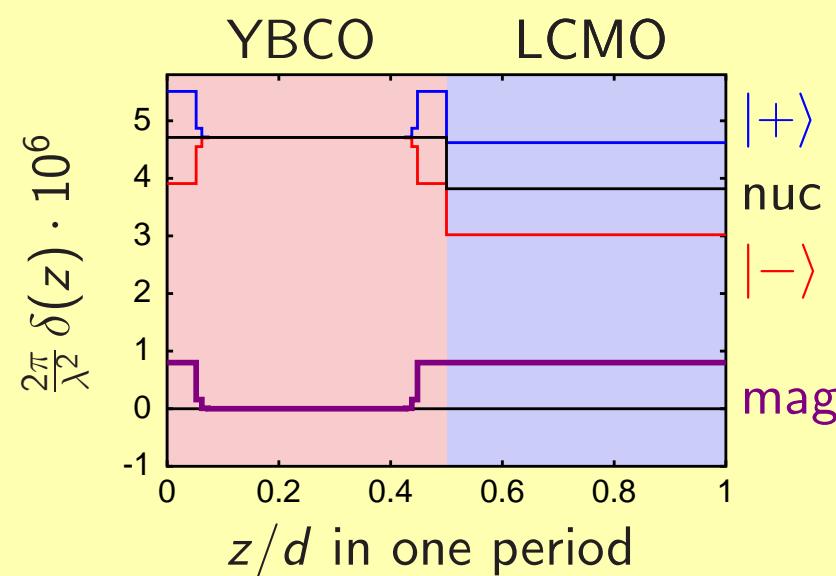
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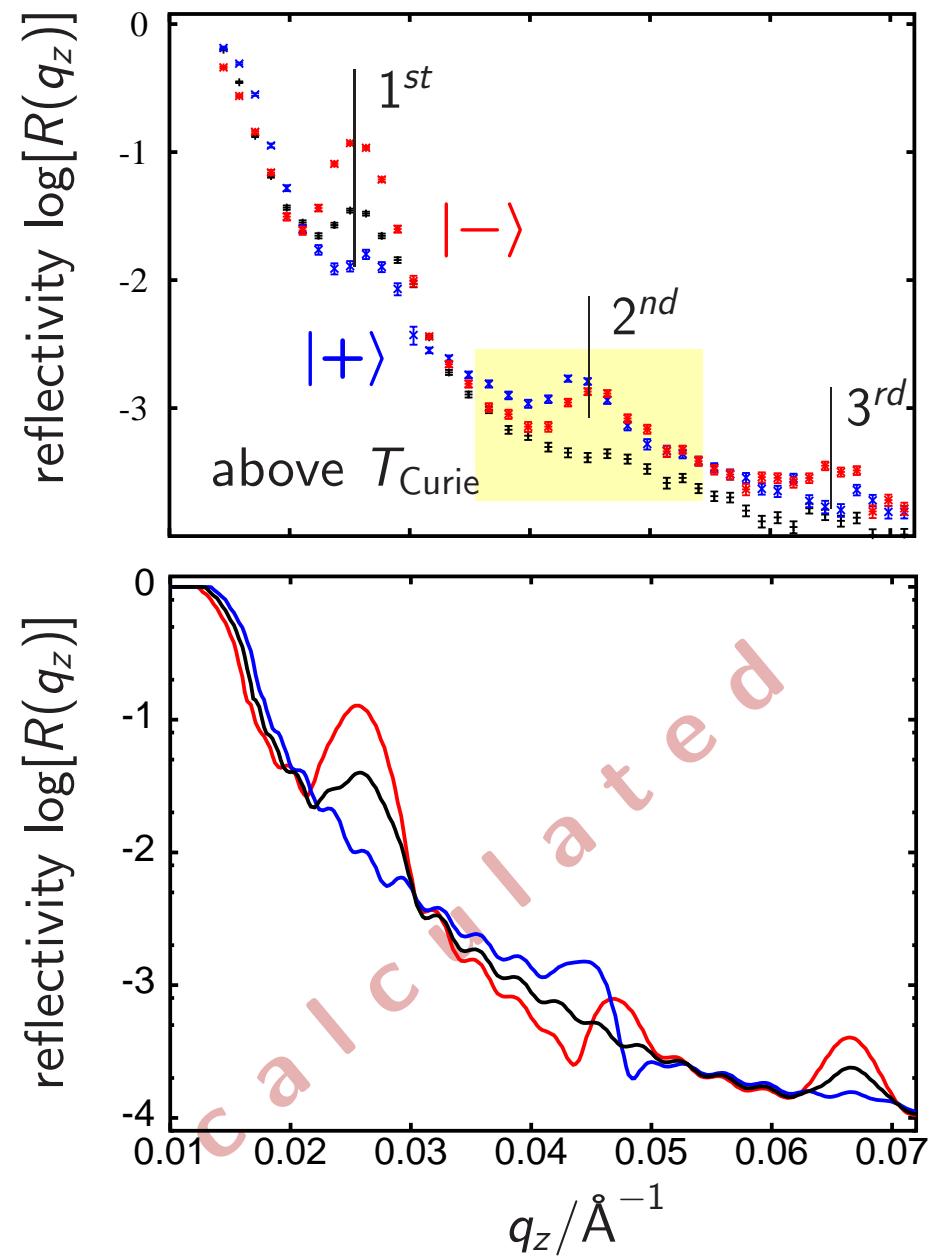
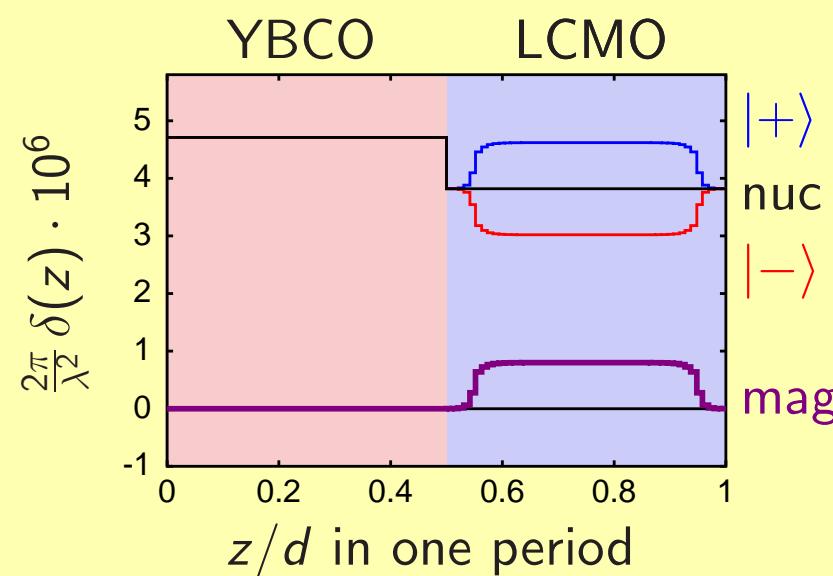
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## résumé

PNR at RT and below  $T_{\text{Curie}}$  and  $T_c$   
exclude *all* models besides

### AFM-region within LCMO

charge-injection from YBCO leads to  
a doping of LCMO and thus to an  
AFM ground state

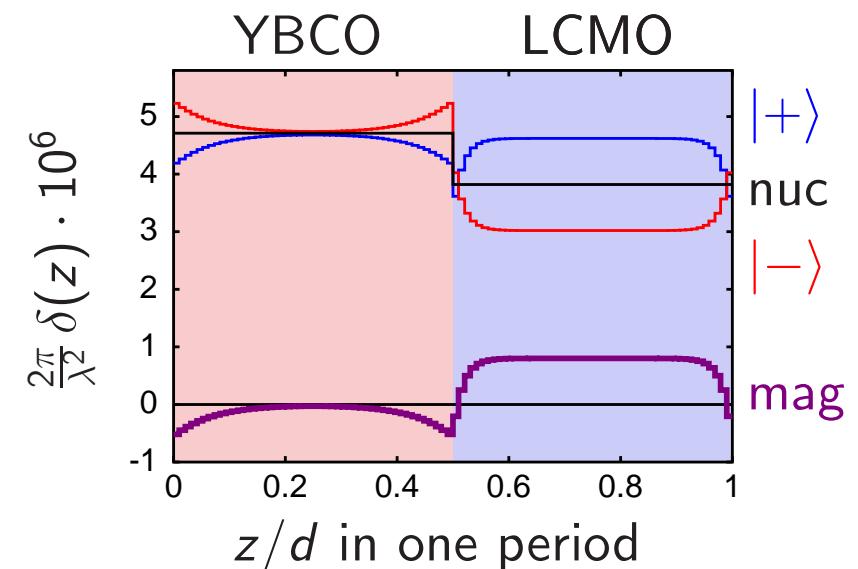
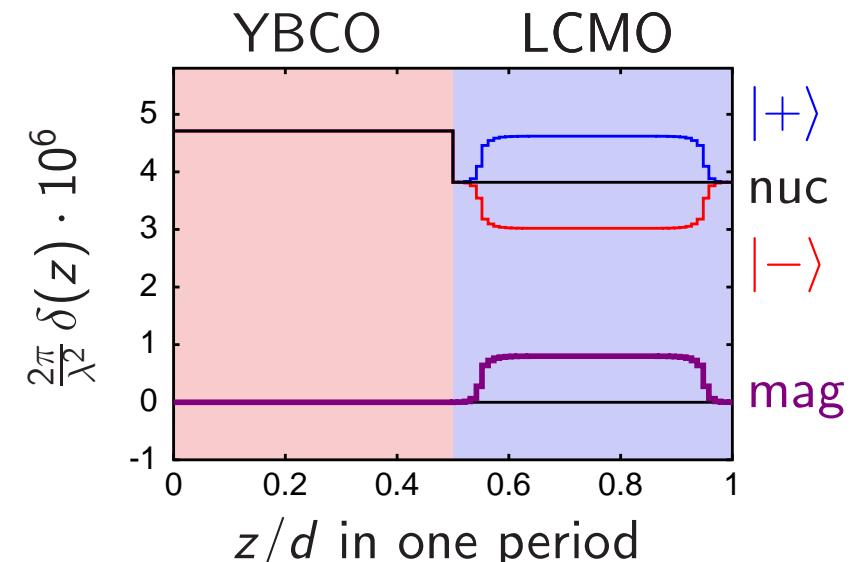
### antiphase magnetic proximity effect

AF coupling of Mn and Cu moments  
through oxygen

or

Cooper pairs penetrate into LCMO  
and are *polarised*

⇒ antiparallel magnetisation in YBCO



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PRB **69**, 174504 (2004):

F. S. BERGERET, A. F. VOLKOV AND K. B. EFETOV

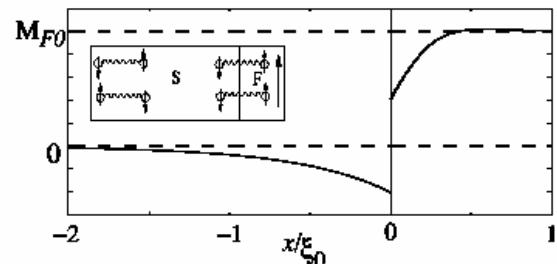
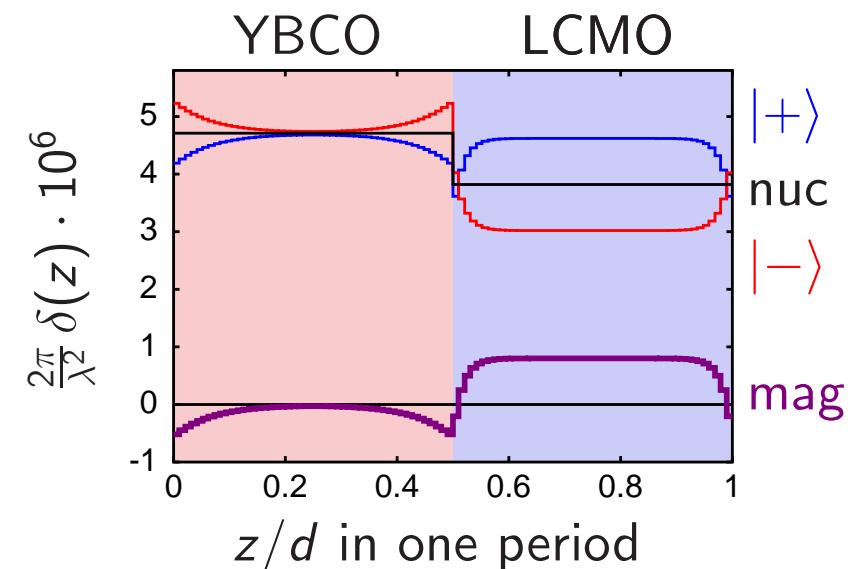
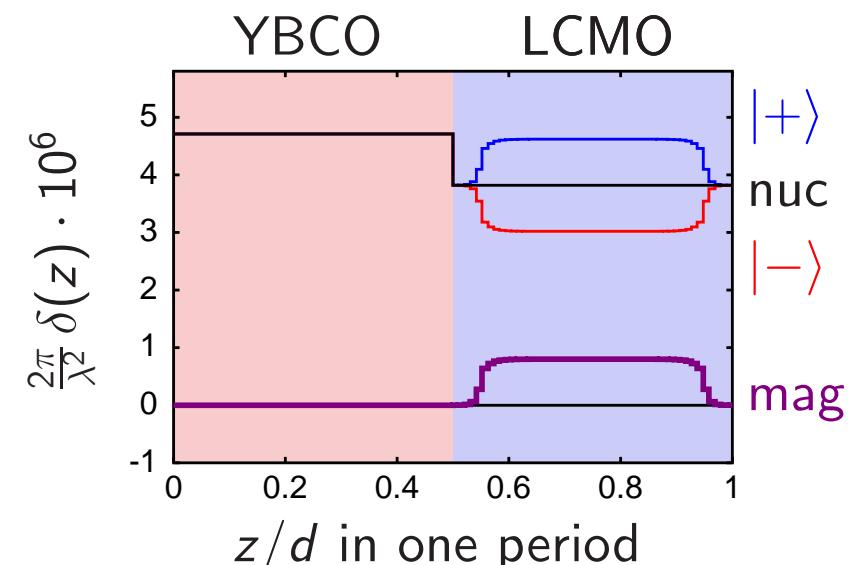


FIG. 1. Spatial dependence of the magnetization in the whole system. Here  $\gamma_F/\gamma_s = 0.5$ ,  $\bar{\gamma}_F = \gamma_F/\xi_0 = 0.1$  ( $\xi_0 = \sqrt{D_s/2T_c}$ ),  $J/T_c = 15$ , and  $d_F/\xi_0 = 1$ . Inset: Schematic view of the inverse proximity effect in a S/F system (for discussion see text).



## magnetometry

SQUID measurements by F. Treubel, Konstanz

$$T = 5 \text{ K}$$

cooled in  $H = 100 \text{ Oe}$

coercitive field  $H_{\text{co}} \approx \pm 400 \text{ Oe}$

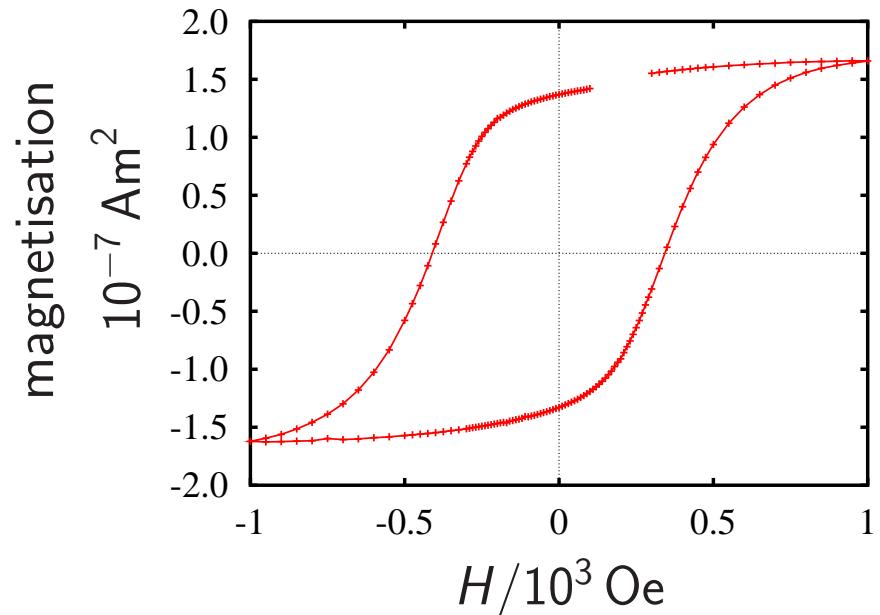
exchange bias  $H_{\text{eb}} \approx -60 \text{ Oe}$



presence of an AFM coupling  
at the FM-interface

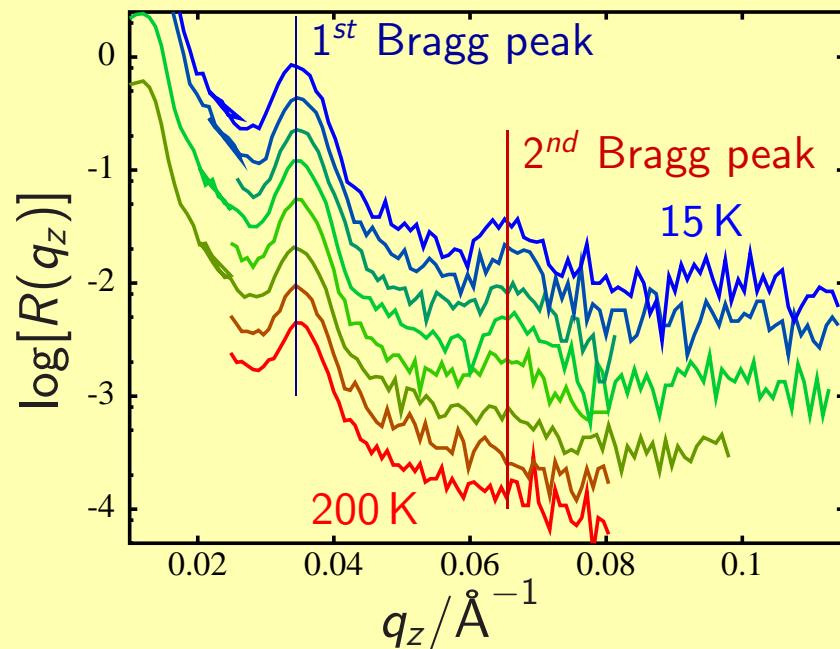
but:

- *magnetically dead layer* might be an AFM
- B in YBCO might be an AFM with net magnetic moment

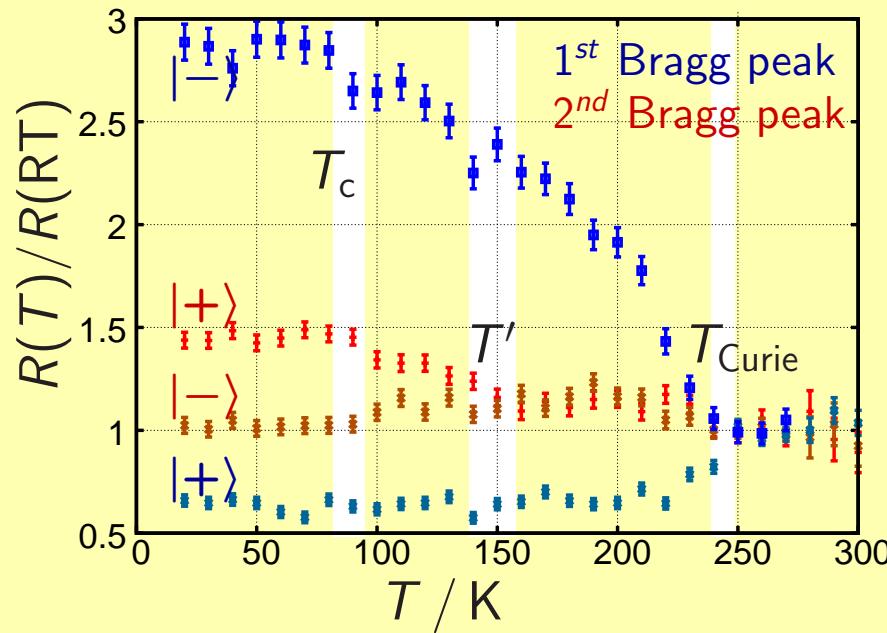


## $T$ dependence of $R(q_z)$

$[\text{YBCO}(100\text{\AA})/\text{LCMO}(100\text{\AA})]_7$



$[\text{YBCO}(200\text{\AA})/\text{LCMO}(200\text{\AA})]_8$



$T_{\text{Curie}}$  ( $160 \rightarrow 270 \text{ K}$ )

onset of FM: changed contrast

$T'$  ( $\approx 140 \text{ K}$ )

formation of  $2^{\text{nd}}$  peaks

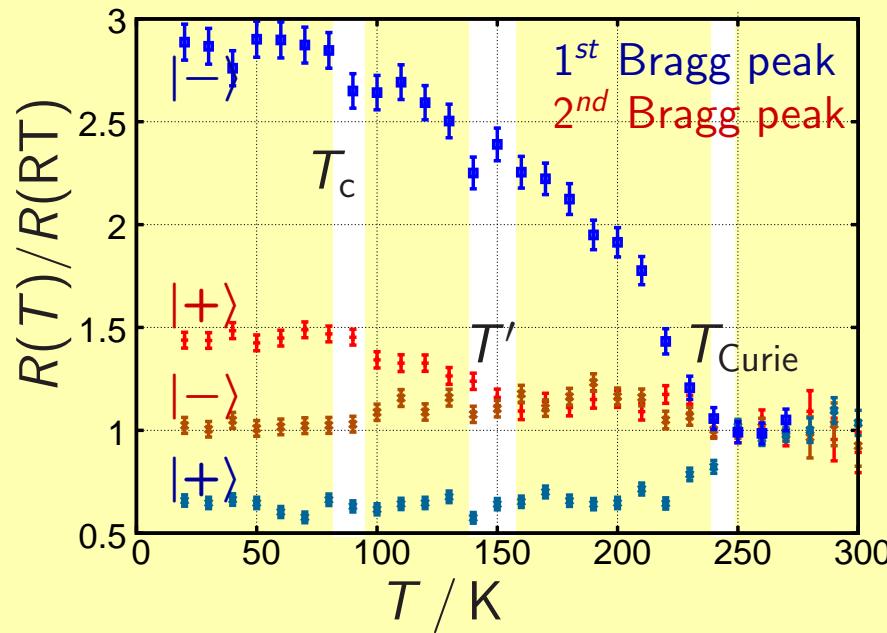
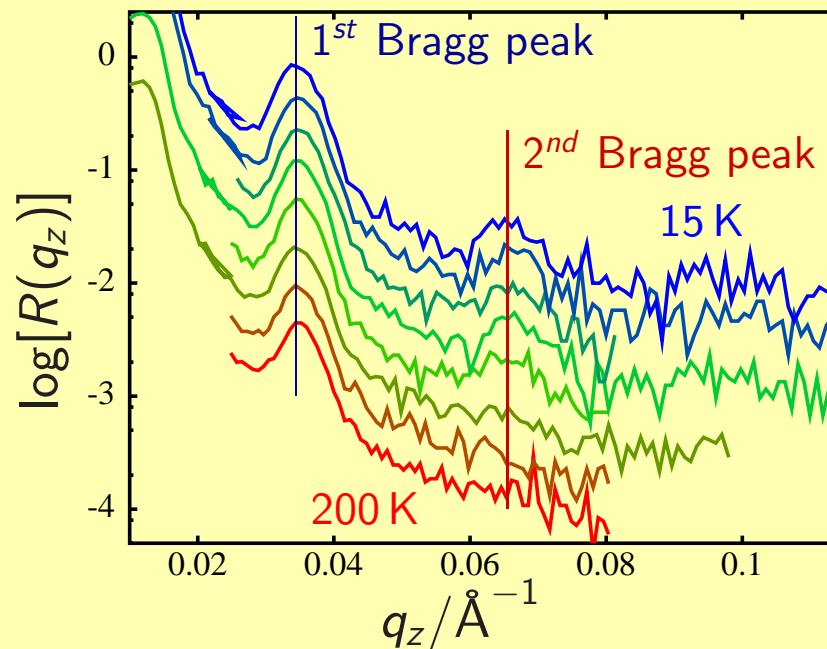
$B(z)$  and  $V_{\text{nuc}}(z)$  differ

$T_c$  ( $60 \rightarrow 90 \text{ K}$ )

onset of SC

## $T$ dependence of $R(q_z)$

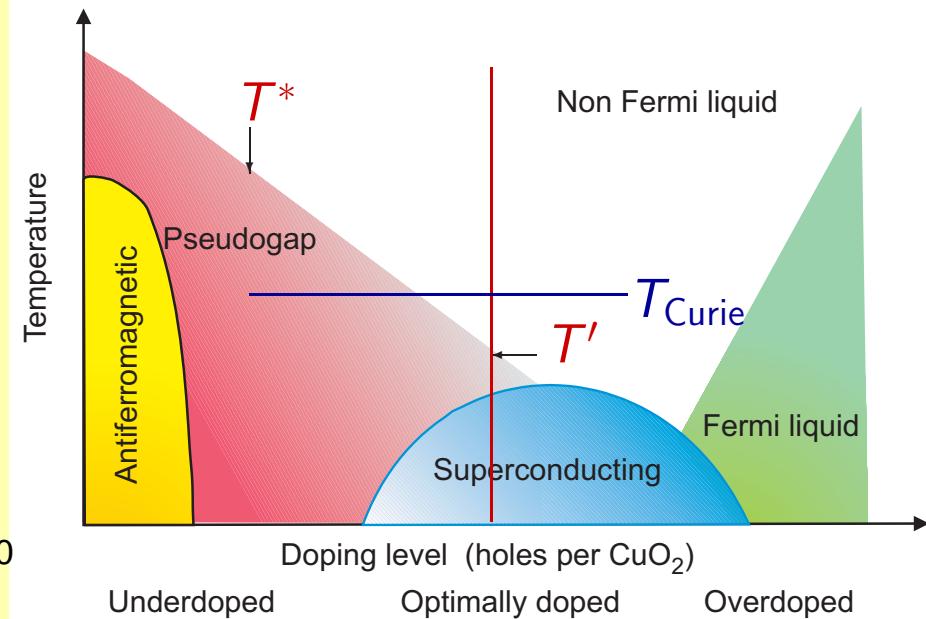
[YBCO(200 Å)/LCMO(200 Å)]<sub>8</sub>



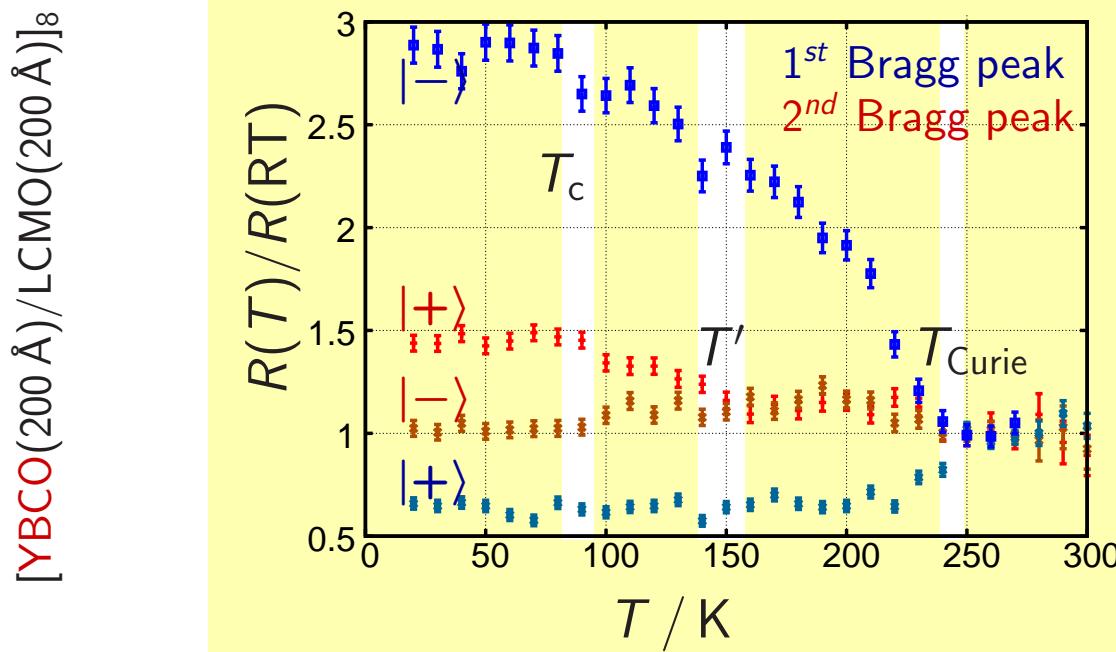
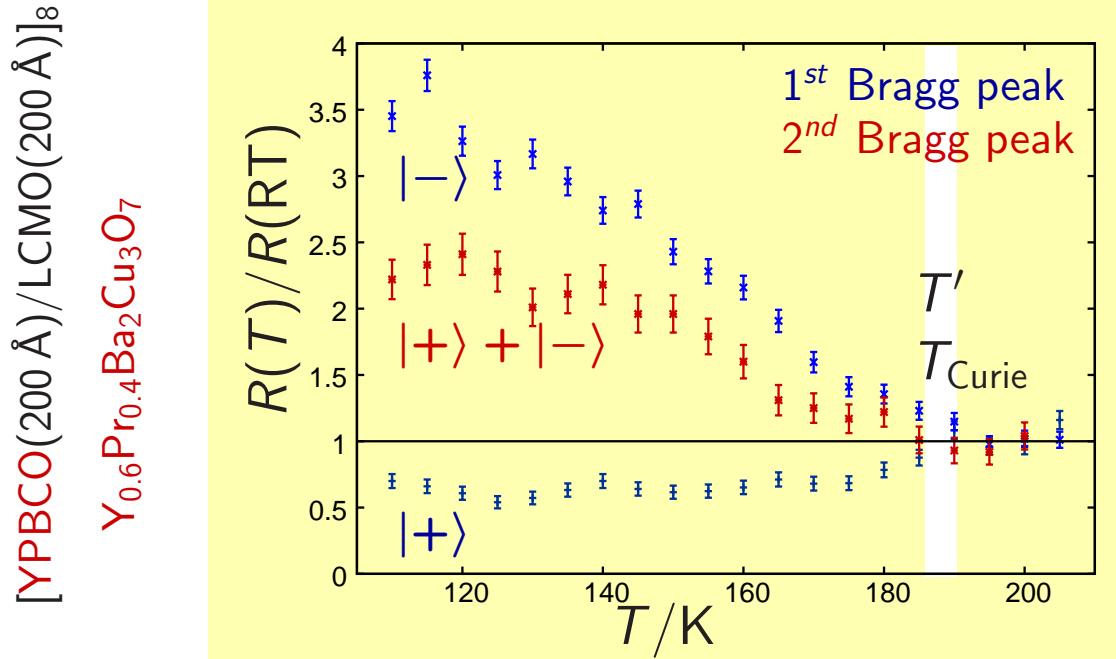
$T_{\text{Curie}}$  ( $160 \rightarrow 270 \text{ K}$ )  
onset of FM: changed contrast

$T' \approx T^* (\approx 140 \text{ K})$   
formation of 2<sup>nd</sup> peaks  
 $B(z)$  and  $V_{\text{nuc}}(z)$  differ

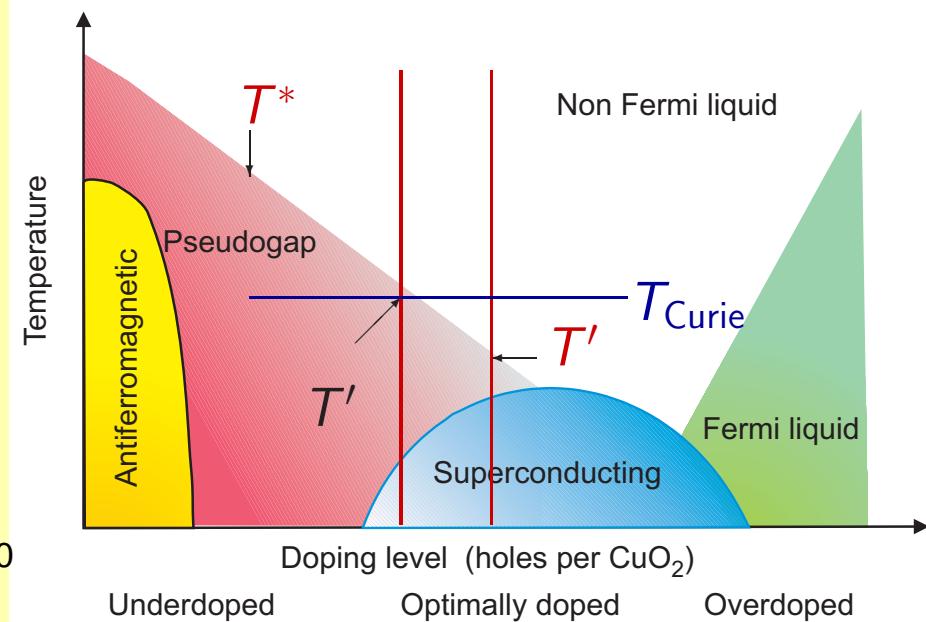
$T_c$  ( $60 \rightarrow 90 \text{ K}$ )  
onset of SC



## $T$ dependence of $R(q_z)$



$T' \approx T^*$  varies with doping!



## overview

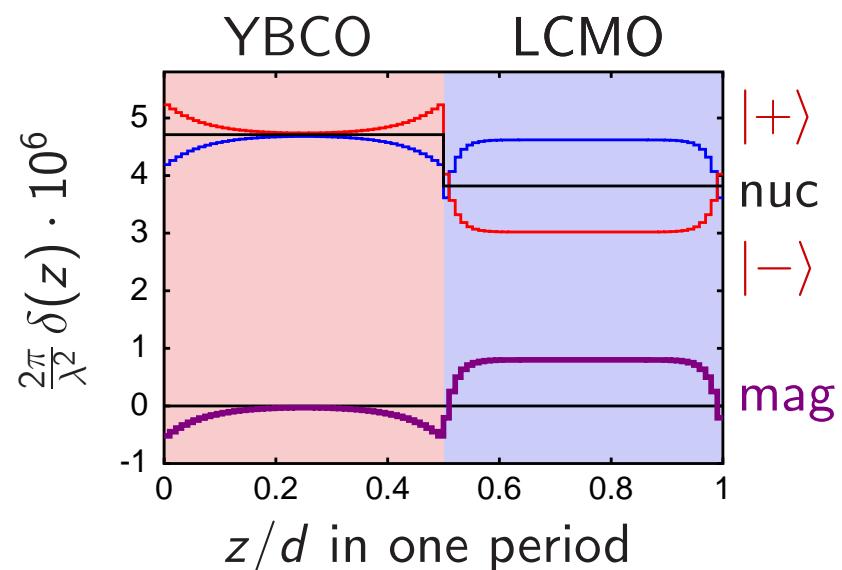
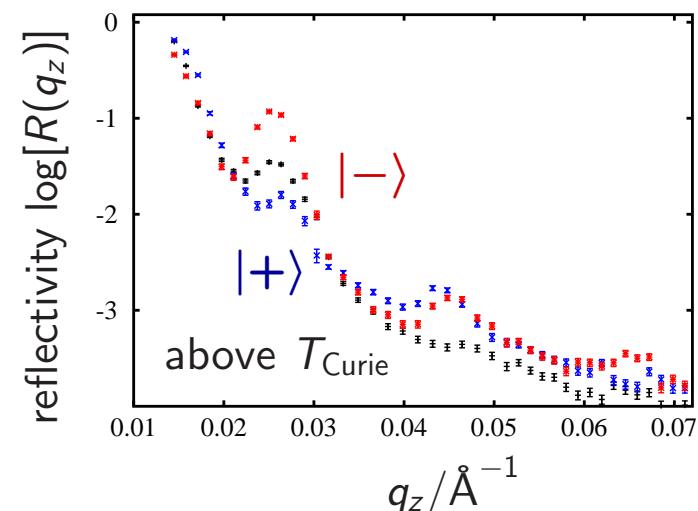
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## off-specular scattering principle

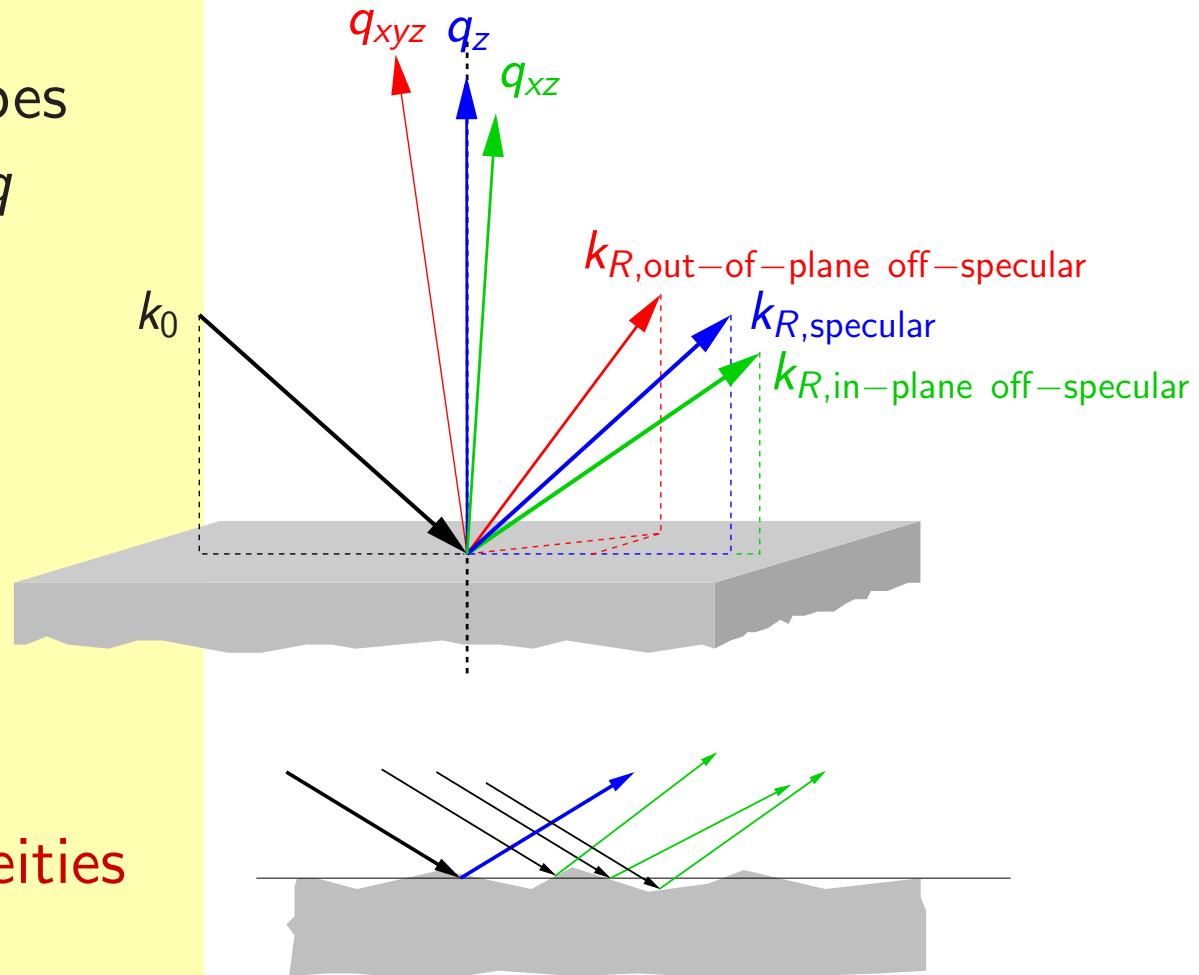
neutron scattering probes  
a potential parallel to  $q$

if  $q \neq q(z)$  also lateral  
structure is accessible

here:

lateral and vertical  
correlation length of  
(magnetic) inhomogeneities

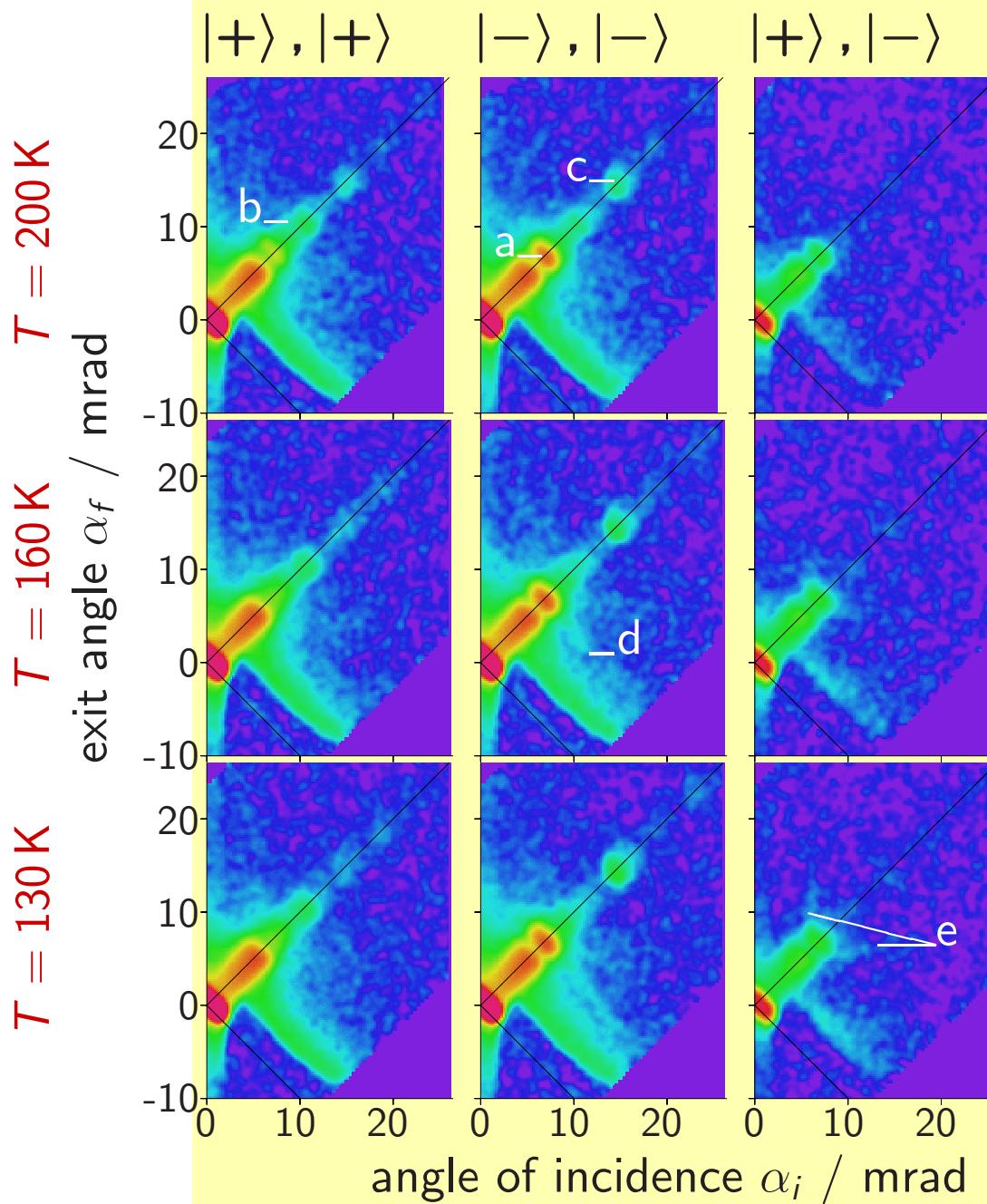
in our cases:  
resolution in  $x$ :  $\approx 0.01^\circ$   
resolution in  $y$ :  $> 1^\circ$   
 $\Rightarrow$  integrated over  $y$



inclined surface facets  $\Rightarrow \Delta\omega$   
height-variation  $\Rightarrow$  phase-shifts in  $k_R$   
 $\Rightarrow$  damping of  $R(q_z > q^c)$

## off-specular scattering

## measured on HADAS@Jülich



No off-specular sheets at RT or 200 K  
 $\Rightarrow$  no structural roughness detectable

Increase of the Bragg sheet at 1<sup>st</sup>  
 Bragg peak (d) below 160 K  
 $\Rightarrow$  magnetic roughness, correlated  
 vertically

Appearance of sheets in the spin-flip  
 channel (e)  
 $\Rightarrow$  magnetic moments not parallel to  
 the neutron spins

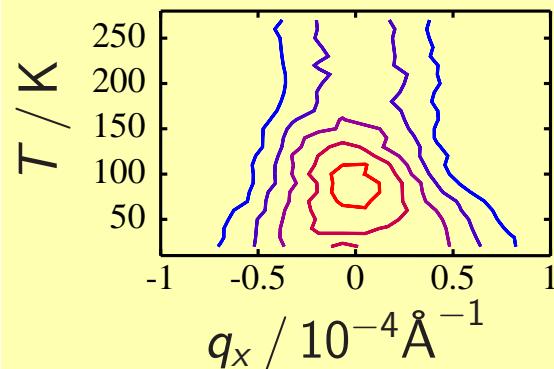
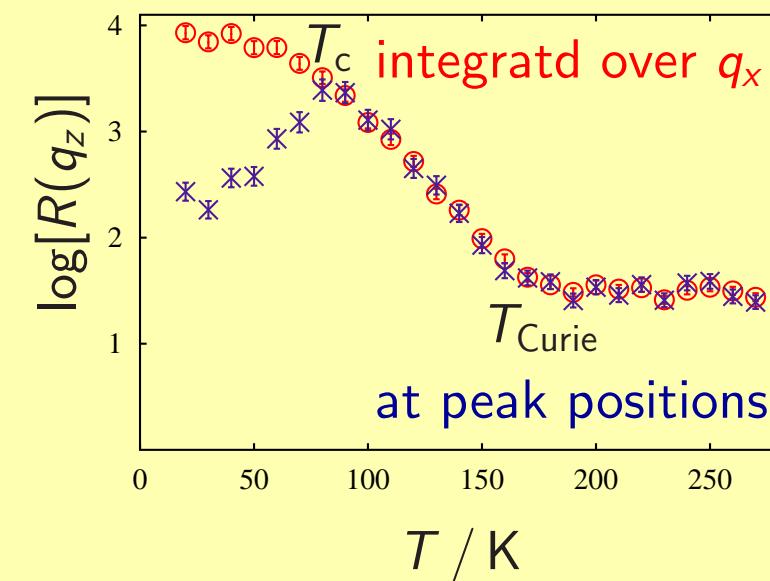
Interpretation (of all measurements):  
 Magnetic domains of similar size ( $\approx 5$   
 to  $10\mu\text{m}$ ) are formed in the LCMO  
 layers. These are correlated through  
 YBCO over the whole stack.

## off-specular scattering

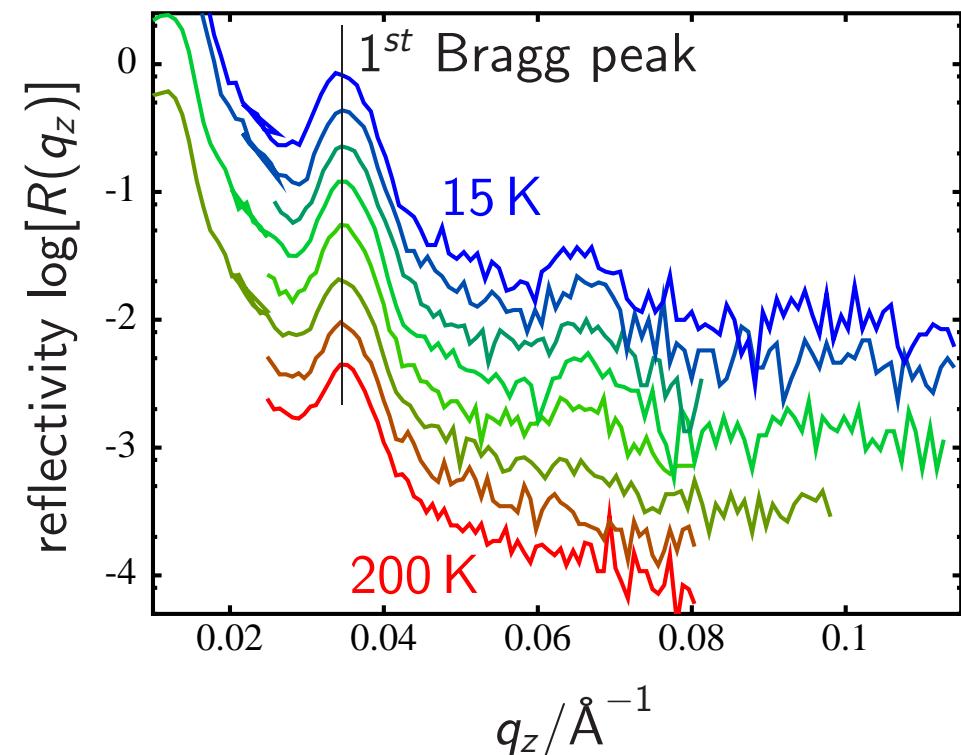
$\omega$ -scans

non-polarised, various  $T$

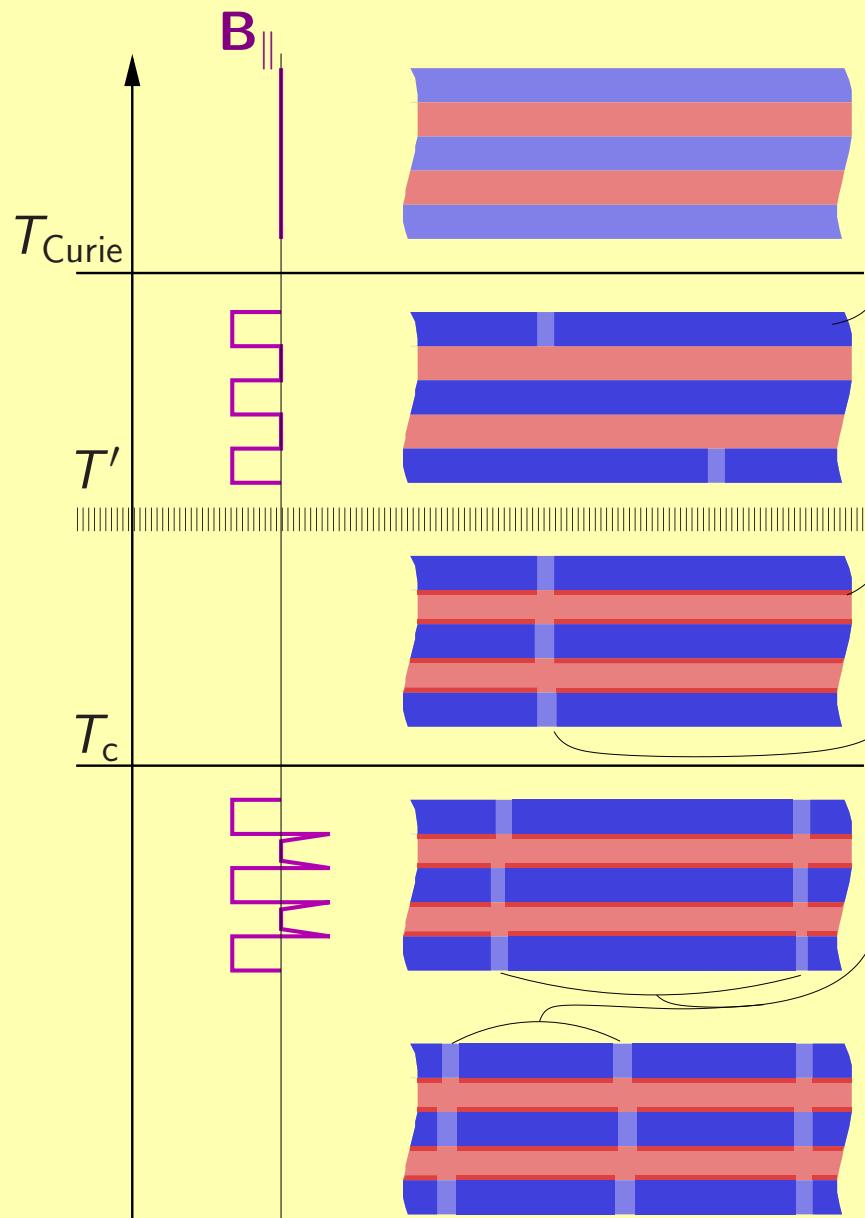
sample: [YBCO(100 Å)/LCMO(100 Å)]<sub>7</sub>



magnetic domains shrink below  $T_c$   
from  $10 \mu\text{m}$  to  $5 \mu\text{m}$  when cooling



## conclusion:



- all LCMO layers are magnetised parallel
- interface effect of  $\mathbf{B}(z)$  of the order of  $10 \text{ \AA}$  is measured at  $T_c < T' \approx 140 \text{ K} < T_{\text{Curie}}$
- *magnetic dead layer or antiphase proximity effect*
- simultaneous appearance of Bragg-sheets
- *vertical correlation of magnetic domains*
- increase of off-specular scattering below  $T_c$
- shrinking of magnetic domains / characteristic lengthscale
- correlation of domain size with  $T < T_c$  and XMCD measurements support the *antiphase proximity effect*

## essence

question: What is the magnetic induction (profile) in HTSC / FM multilayers?

method: polarised neutron reflectometry allows for the determination of  $\rho(z)$  and  $B_{\parallel}(z)$

answers: FM layers magnetised parallel net magnetic moment in SC at the interfaces, antiparallel to FM magnetisation

SC creates and aligns domain walls in FM

