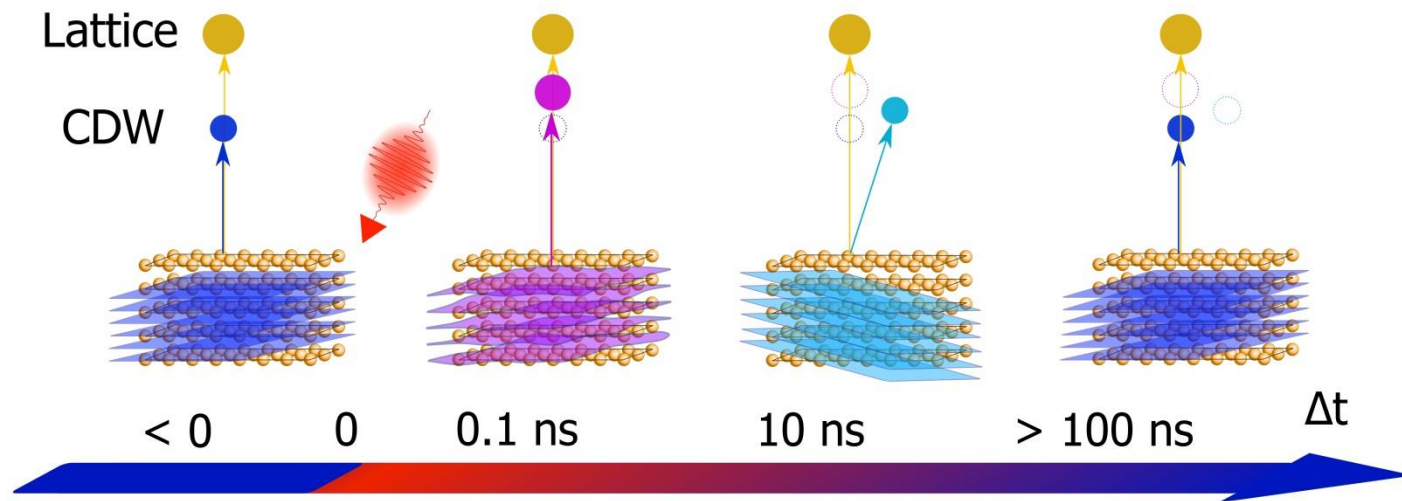


Spin and charge density waves in chromium through the spin-flip transition : statics and ultrafast dynamics



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¹Laboratoire de Physique des Solides, CNRS, Univ.Paris-Sud, Université Paris-Saclay, 91405 Orsay, France

²Synchrotron SOLEIL, L'Orme des Merisiers, Saint-Aubin, 91192 Gif-sur-Yvette, France

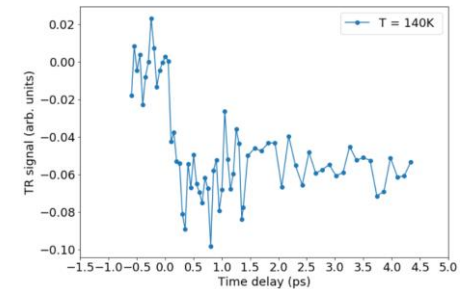
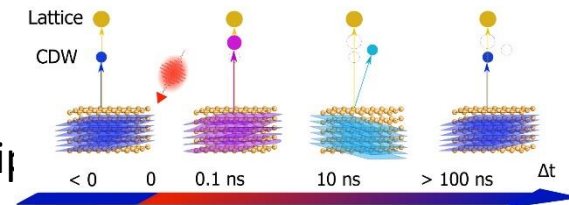
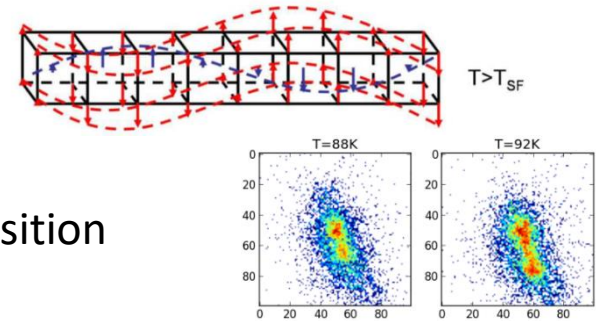


ECRYS2022 – Cargèse – 11/08/2022



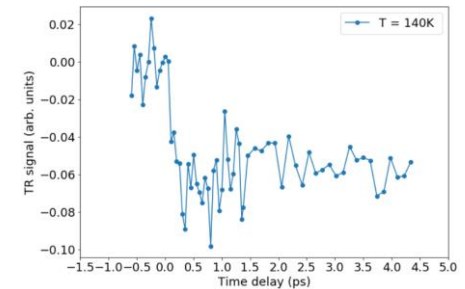
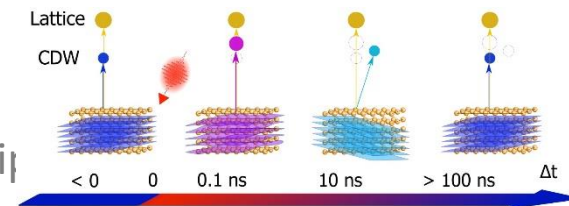
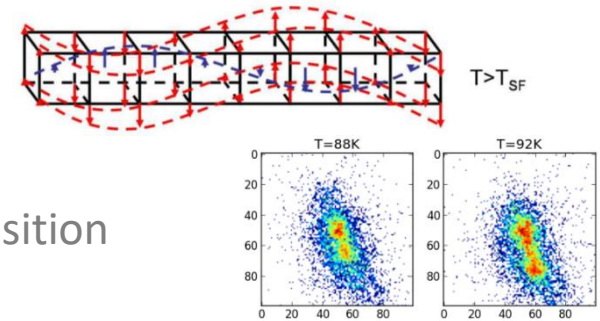
Outline

- Introduction to the density waves of chromium
- SDW and CDW static orders through the spin-flip transition
- CDW behavior after laser-pulse excitation from fs to ns timescales
 - CDW decoupling at ns timescale
 - Dynamics of the CDW through the spin-flip
 - Threshold effect for CDW excitation
- Summary and perspectives



Outline

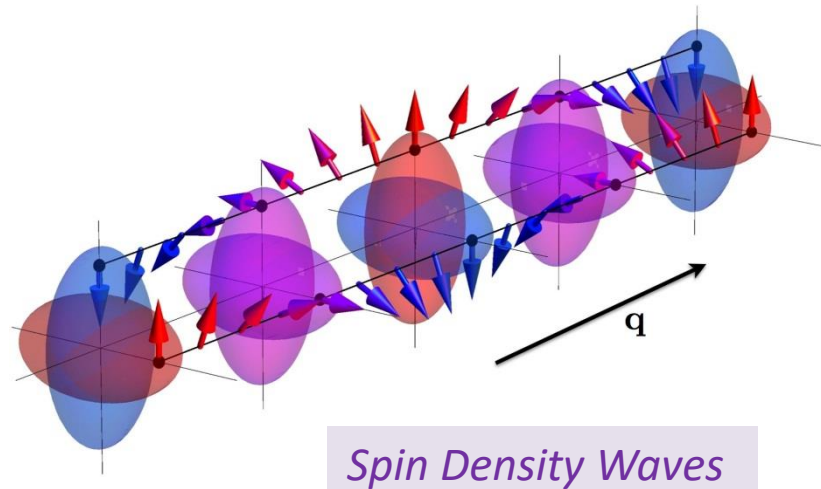
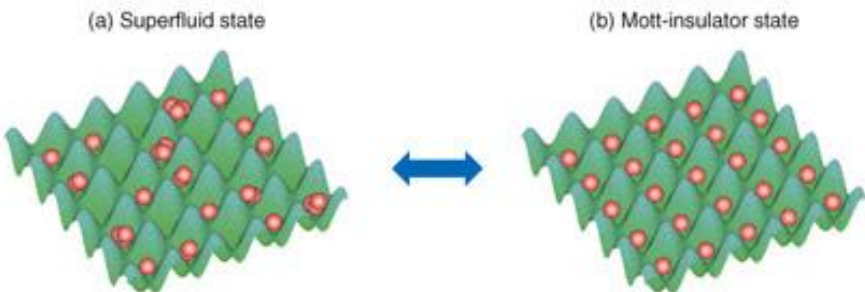
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Correlated electron systems: a rich world

Correlations → drive physical properties

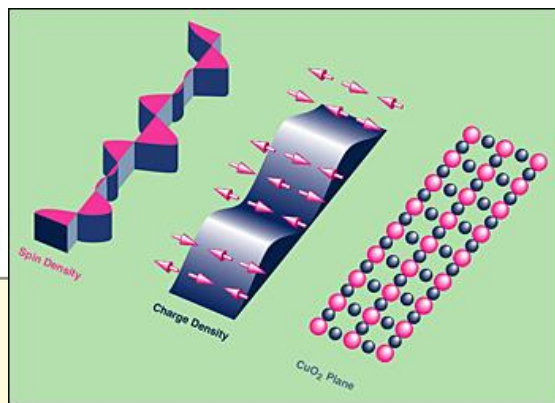
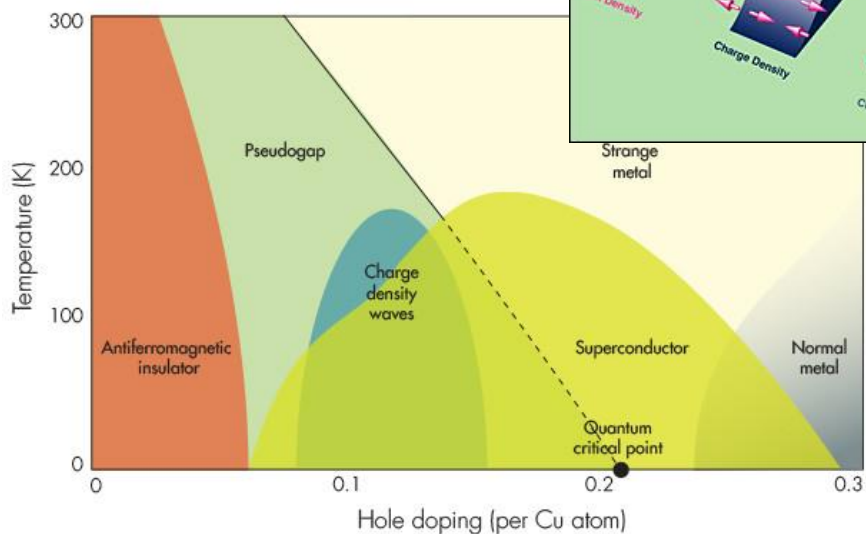
Ex: metal → insulator



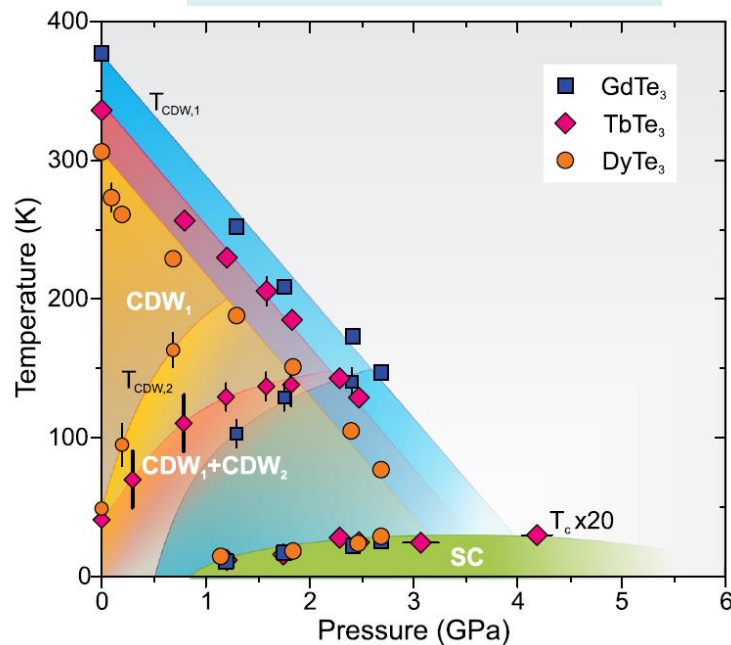
Mott insulators

Superconductors

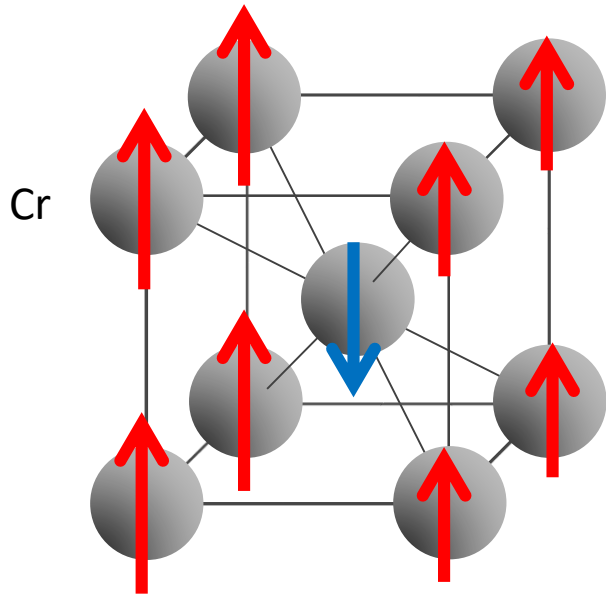
CUPRATE PHASE DIAGRAM



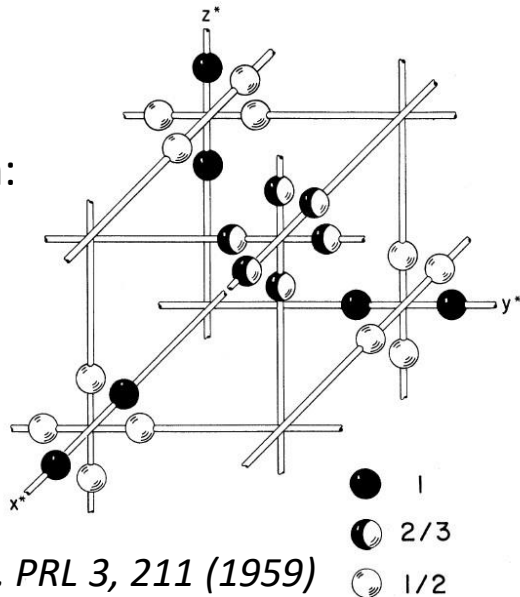
Charge Density Waves



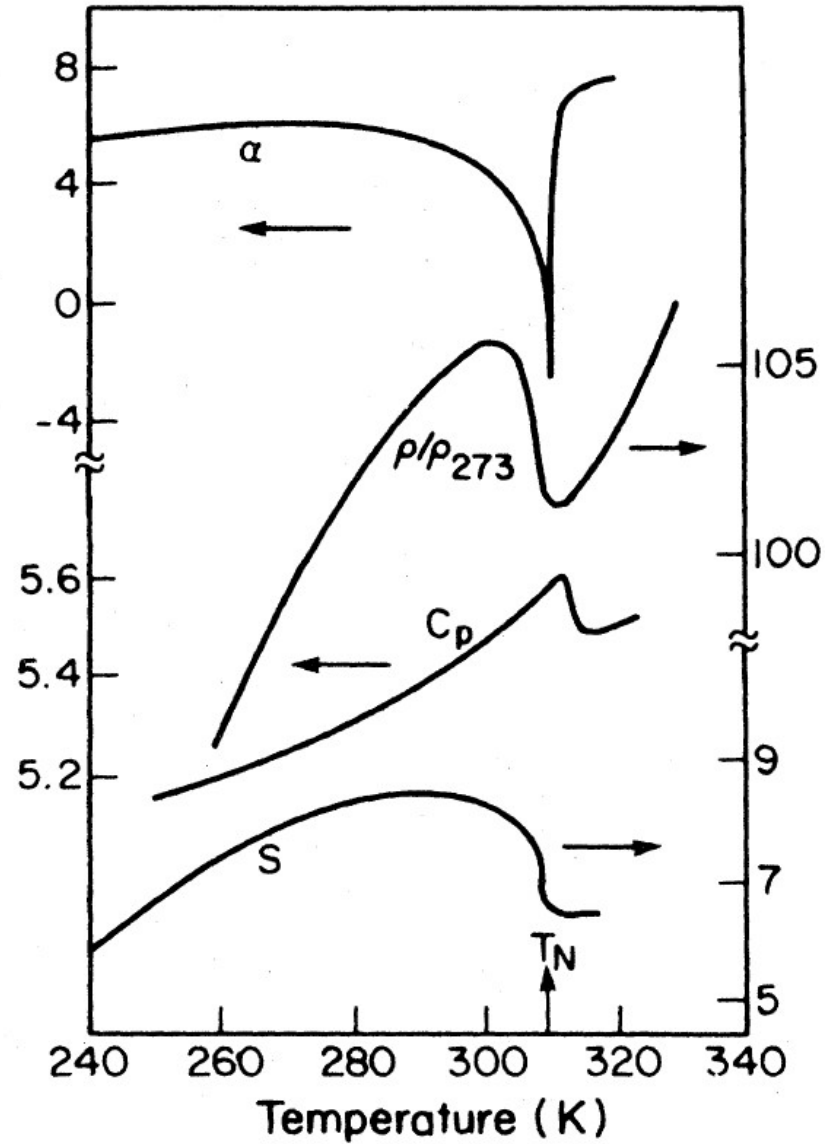
A SDW transition in a simple metal



Neutron
Diffraction:

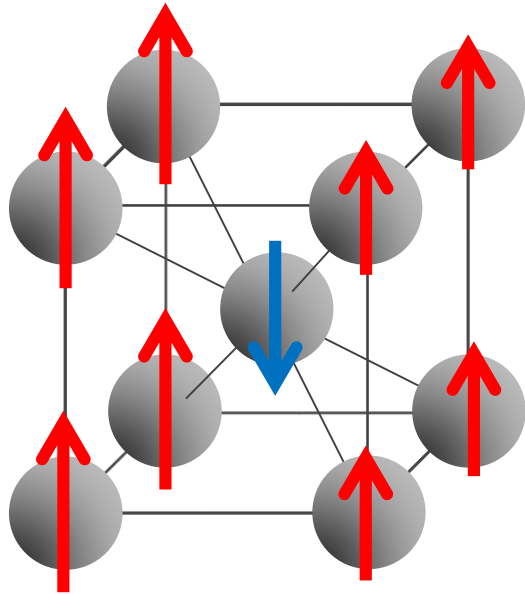


Corliss et al. PRL 3, 211 (1959)



Fawcett, Rev. Mod. Phys. 60, 209 (1988)

SDW in a 3D metal: a peculiar Fermi surface nesting

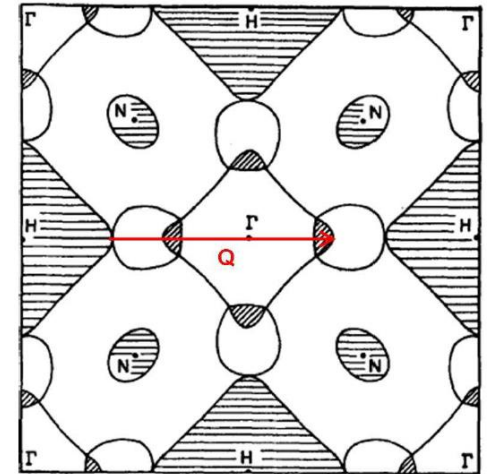


For $T < T_N$:
 AFM + SDW along $\langle 001 \rangle$:
 → Periodic modulation of $|\mathbf{m}|$

Two transitions :

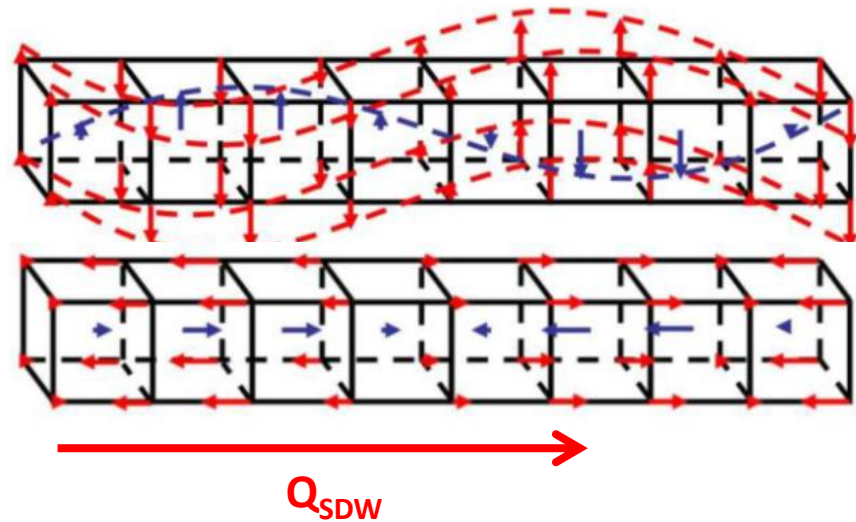
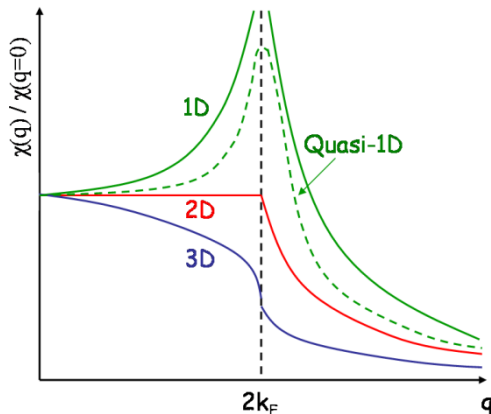
$$T_N = 311\text{K}$$

$$T_{SF} = 123\text{K}$$



Cr Fermi surface

$$\chi(\vec{q}) = \int \frac{d\vec{k}}{(2\pi)^d} \frac{f_{\mathbf{k}} - f_{\mathbf{k}+\mathbf{q}}}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}}}$$



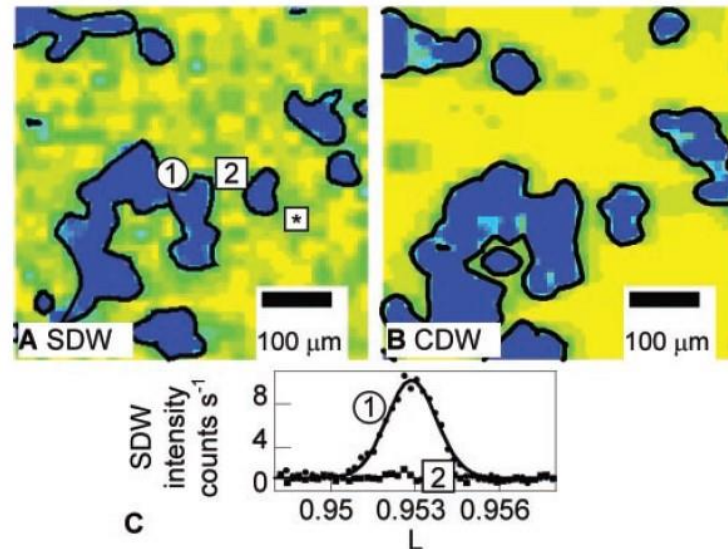
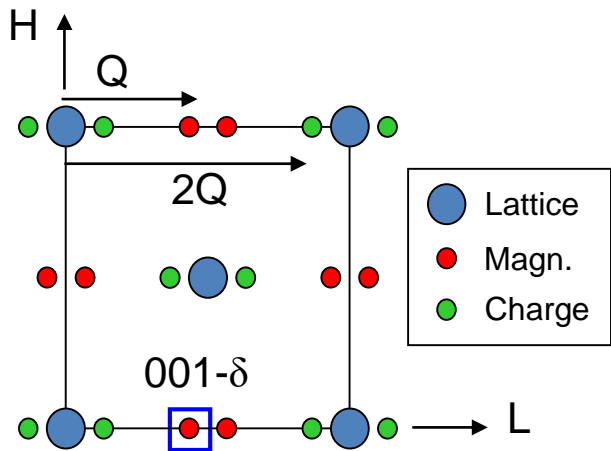
$T > T_{SF}$

$T < T_{SF}$

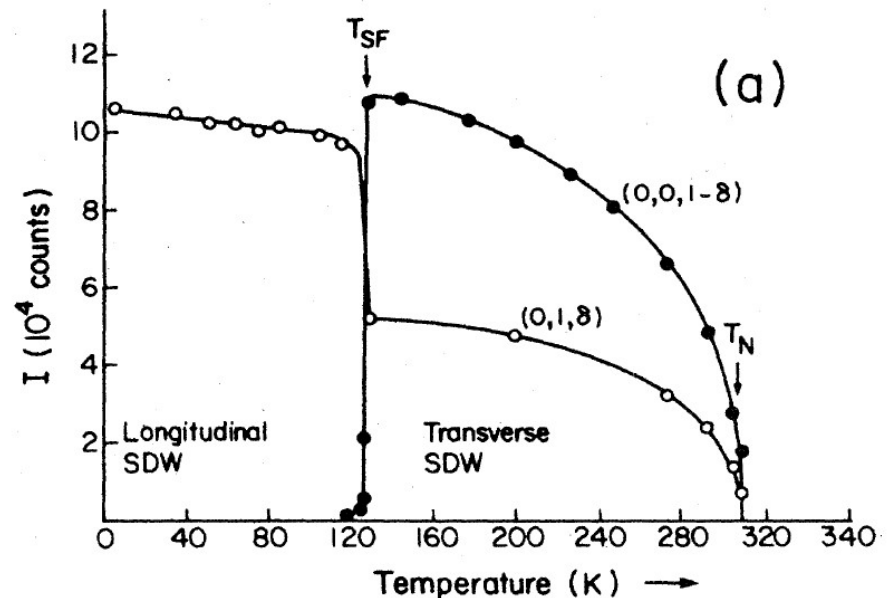
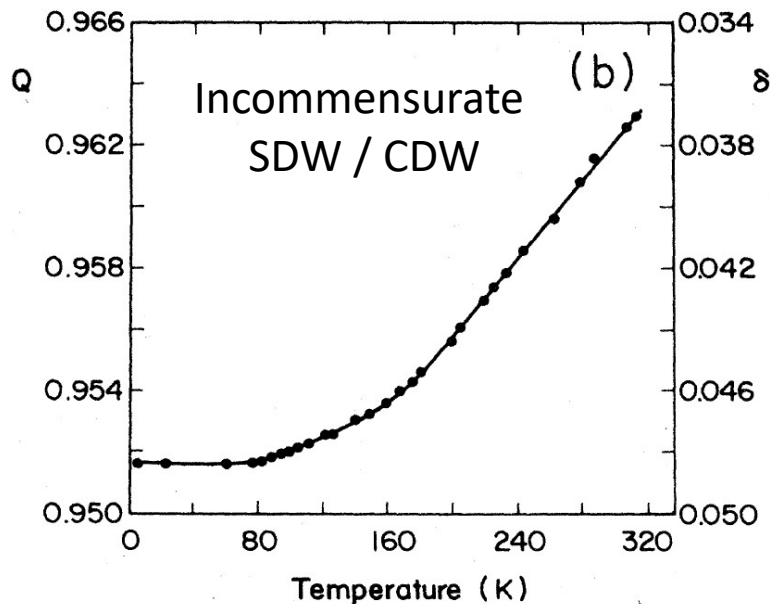
Q_{SDW}

SDW does not come alone : CDW also appears at 2Q

In x-ray diffraction (for single-Q sample):



Evans et al., Science **295**, 1042 (2002)



How does the CDW appear?

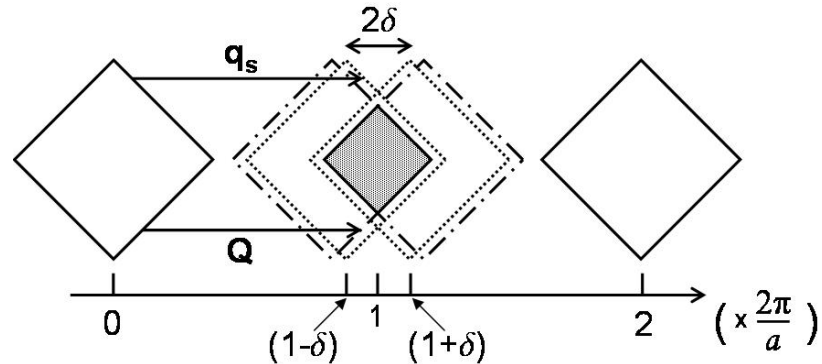
SDW: modulation with wavevector Q + harmonics nQ .

- n even: charge component
- n odd: spin component

band model

2 successive nesting:

- electron-hole \rightarrow SDW
- hole-hole \rightarrow CDW



Young and Sokoloff, J. Phys. F: Metal Phys., **4**, 1304 (1974)

Magnetostriction

The SDW creates a strain wave at $2Q$

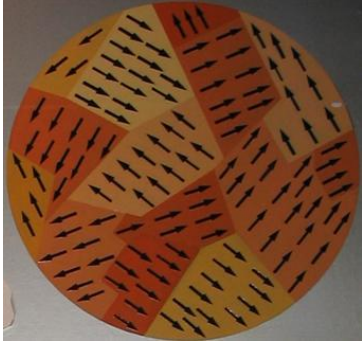
It explains the Spin-Flip Transition at $T=123\text{K}$

Cowan, J. Phys. F: Metal Phys., **8**, 423 (1978)

We use novel techniques to get a new vision of CDW/SDW coupling in Cr:
coherent x-ray diffraction and ***time-resolved x-ray diffraction***

X-ray beam properties at large scale instruments

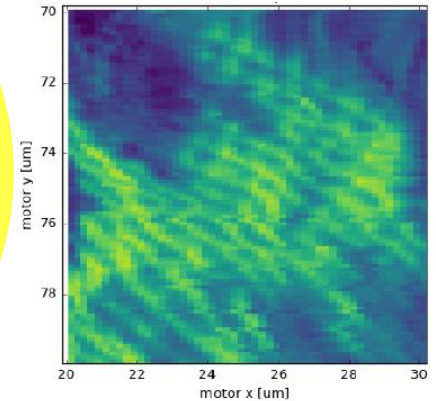
Imaging
Phase Objects/Defects



COHERENCE

NANOFOCUS

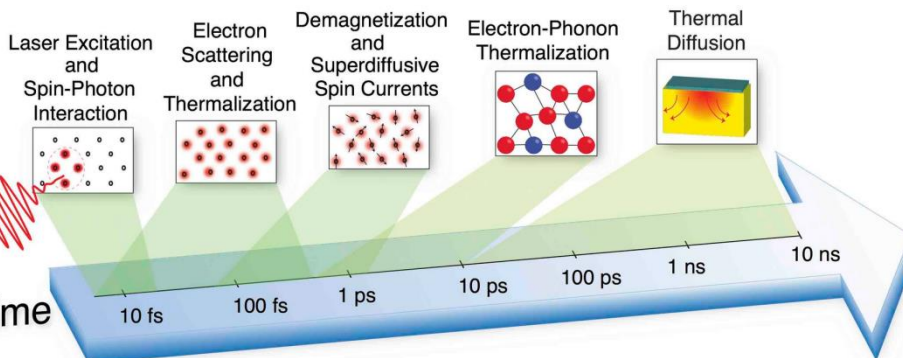
Micro/Nano Structure



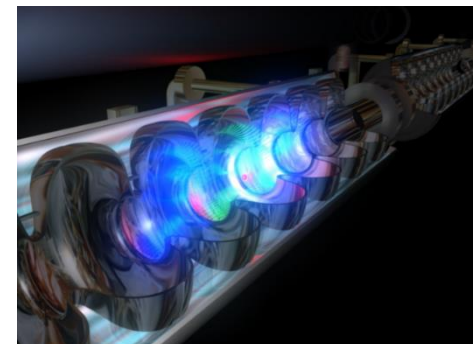
**ULTRASHORT
PULSES**

Ultrafast Dynamics

Synchrotron
SOLEIL

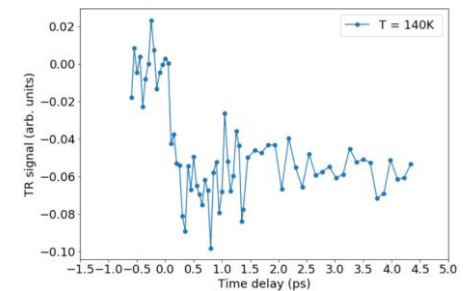
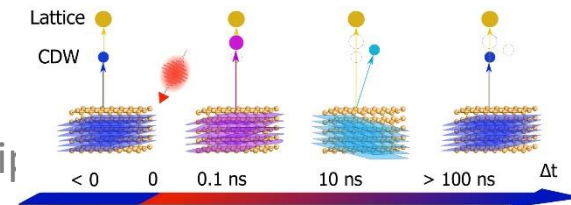
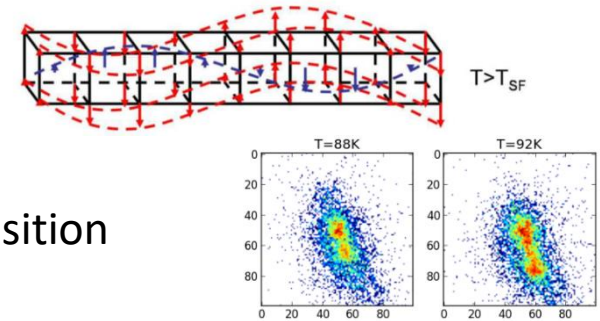


European
XFEL



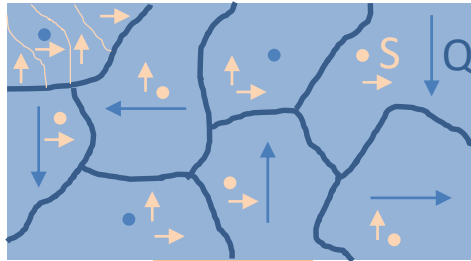
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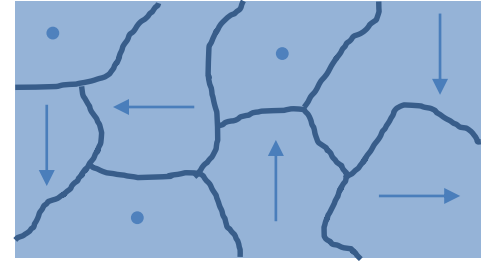


SDW and CDW through the spin-flip transition

Evans et al., Science **295**, 1042 (2002)

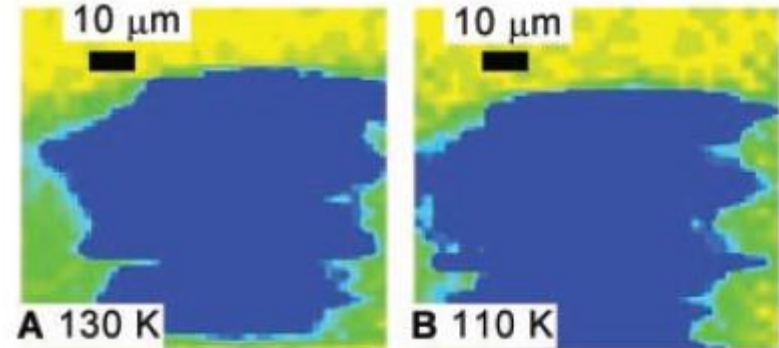
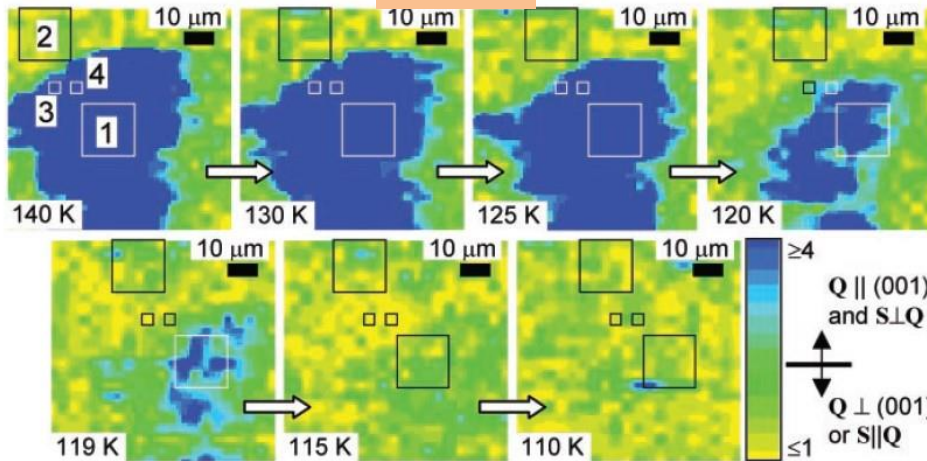


$$T > T_{SF}$$

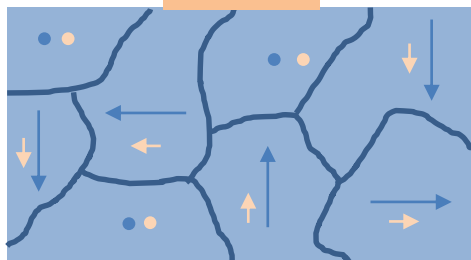


TSDW

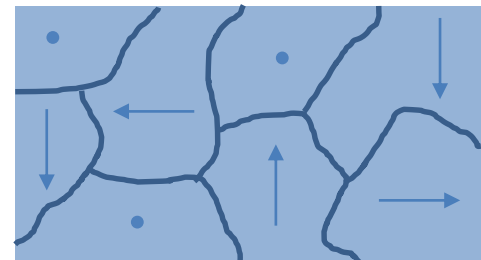
CDW



LSDW

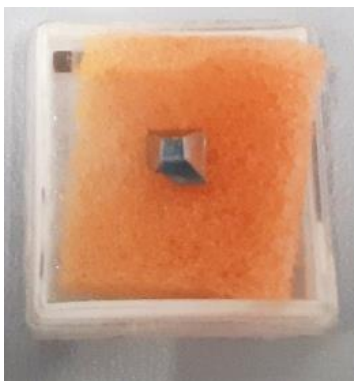


$$T < T_{SF}$$

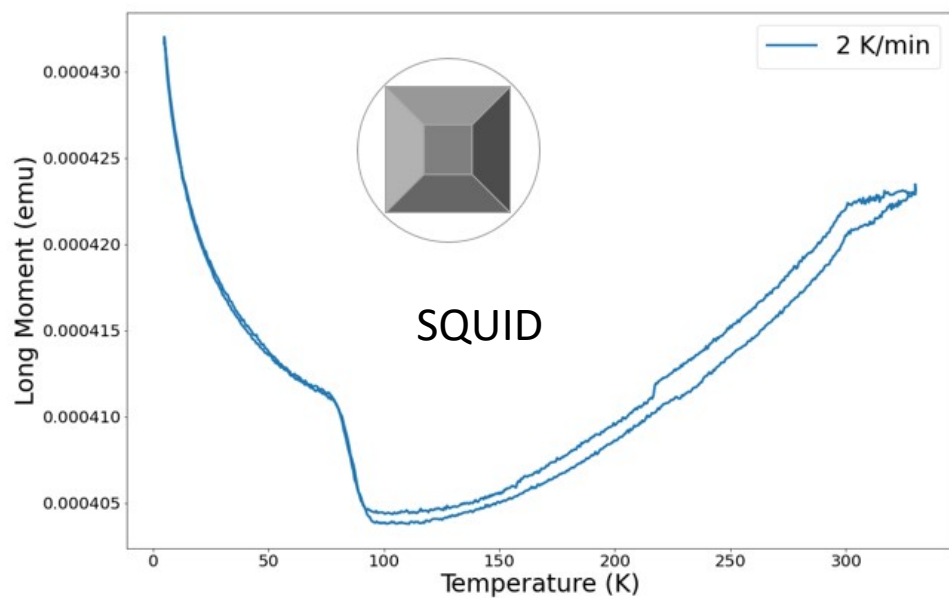
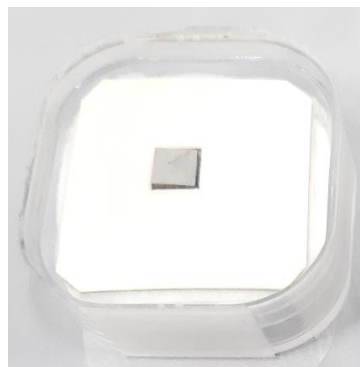


Bulk samples used in our studies

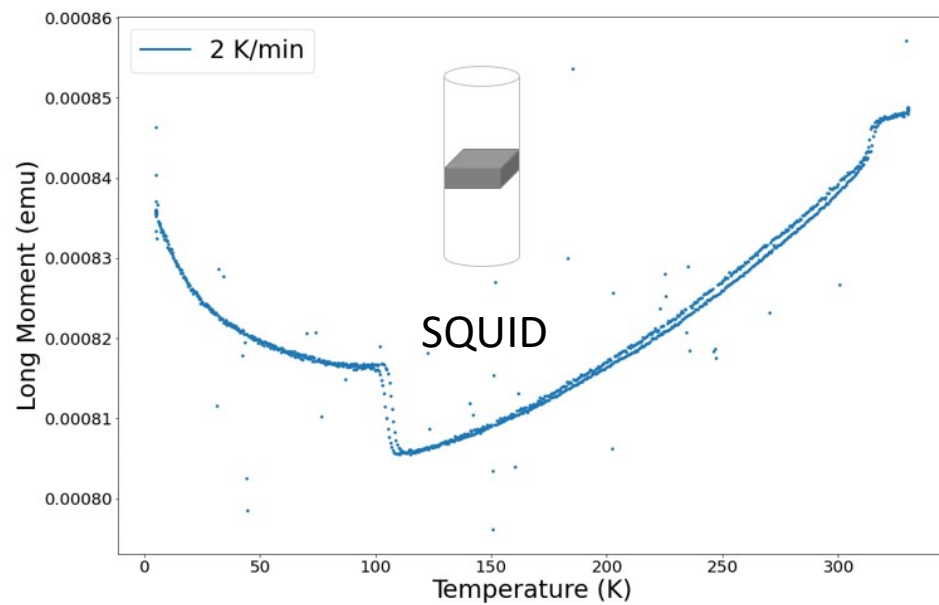
Single-Q
truncated pyramid



Multi-Q
square sample



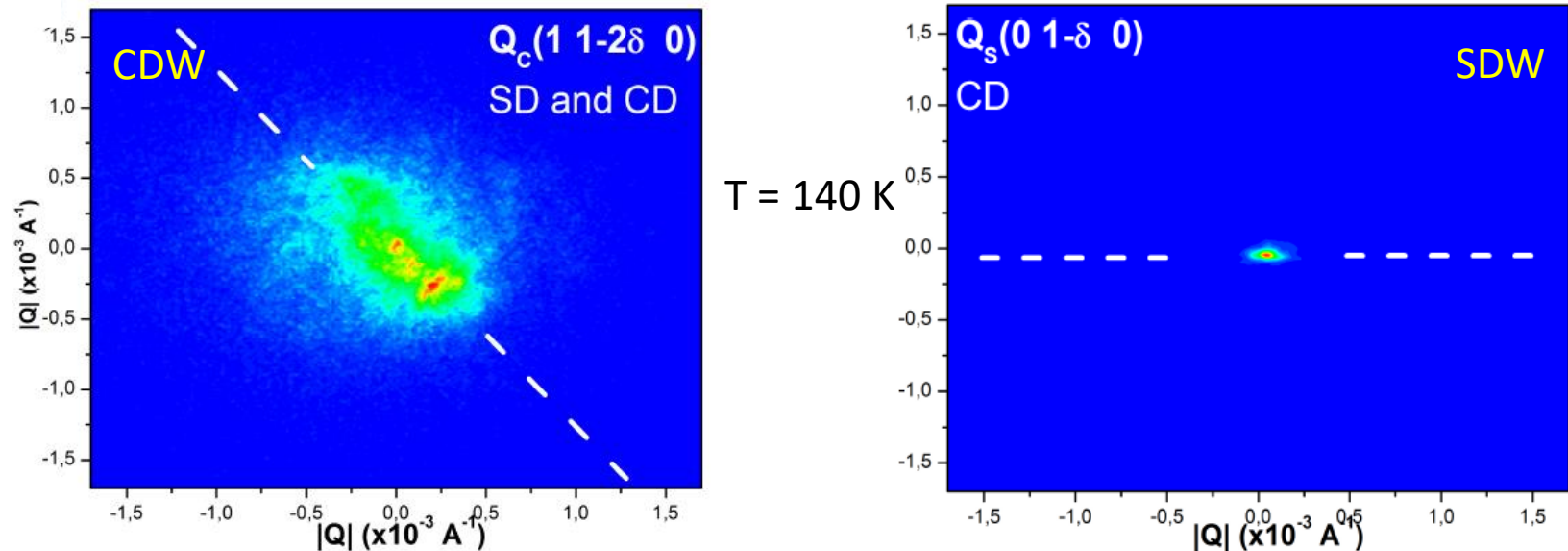
$$T_N = 300 \text{ K}$$
$$T_{SF} = 85 \text{ K}$$



$$T_N = 314 \text{ K}$$
$$T_{SF} = 105 \text{ K}$$

Static SDW and CDW : two very different orders

Coherent and simultaneous x-ray diffraction on CDW and SDW reflections



SDW : perfect order with infinite correlation lengths

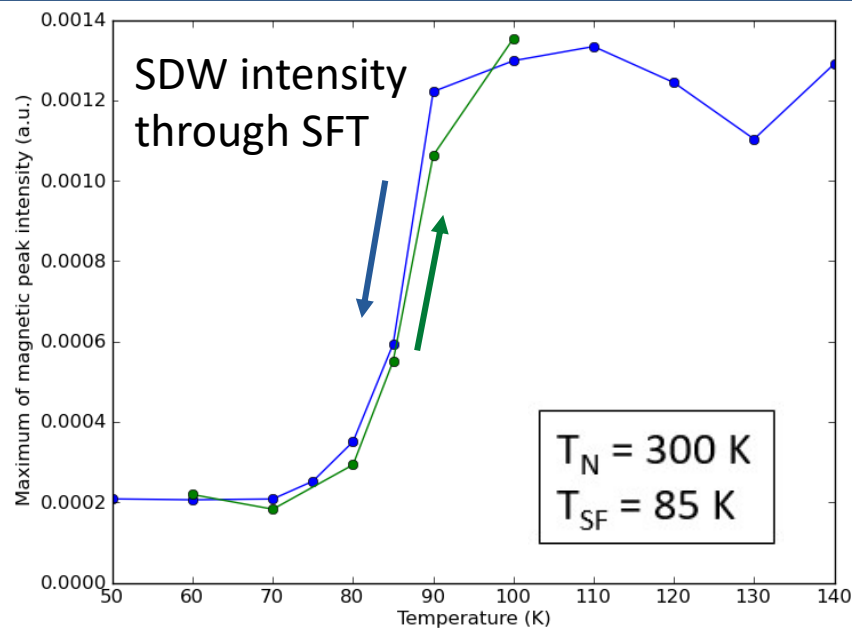
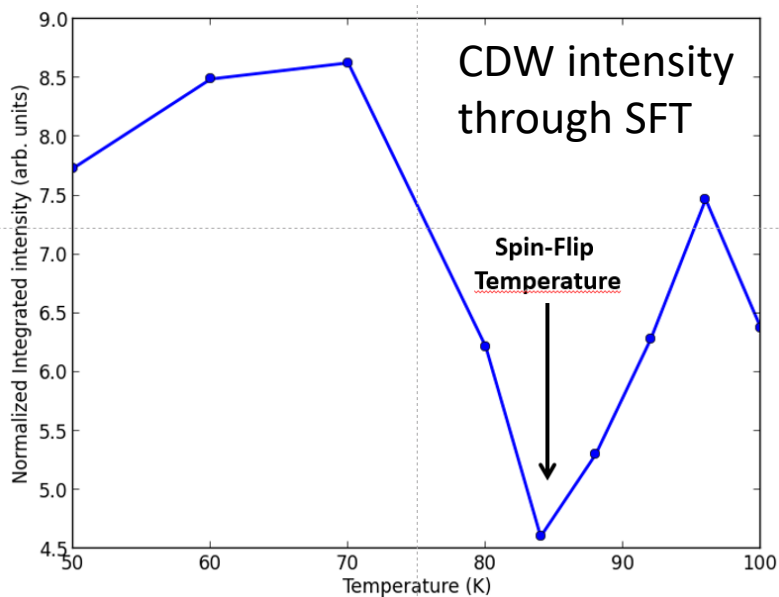
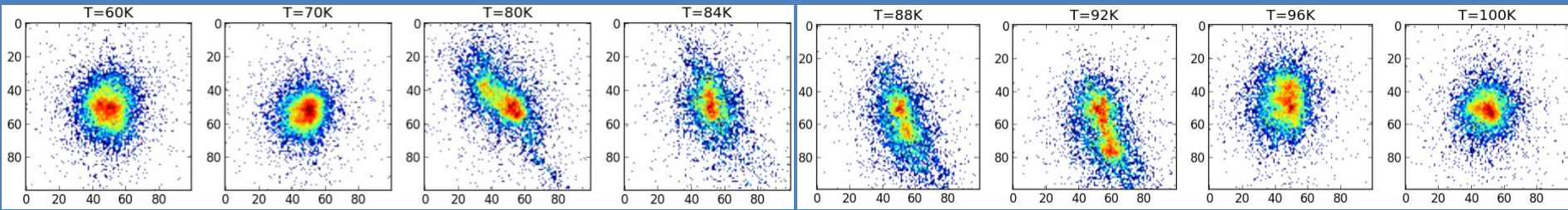
CDW : many phase defects with sub-micrometer correlation lengths

Not the same amount of defects on CDW and SDW:

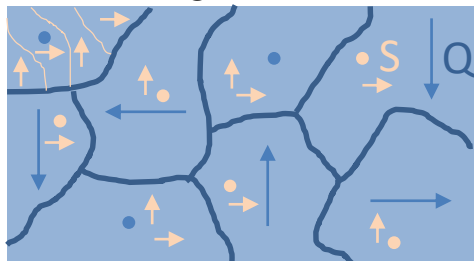
Not in agreement with CDW formation through magnetostriuctive coupling

- ➔ To get more information on the coupling: compare their dynamics of formation
- ➔ Laser Pump – x-ray probe diffraction experiment to perform this

CDW through the spin-slip transition – a clear structural change



TSDW



CDW sensitive to magnetic domain wall reorganization

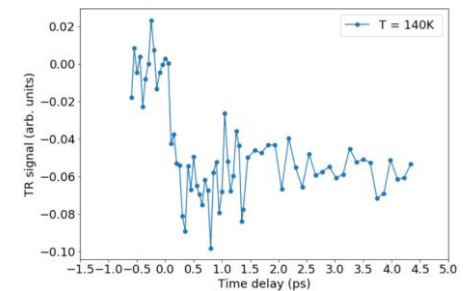
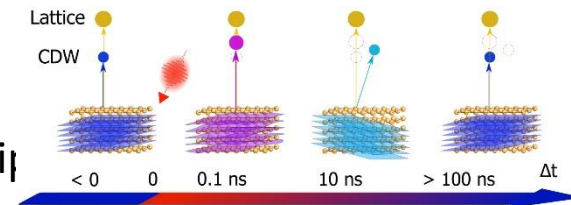
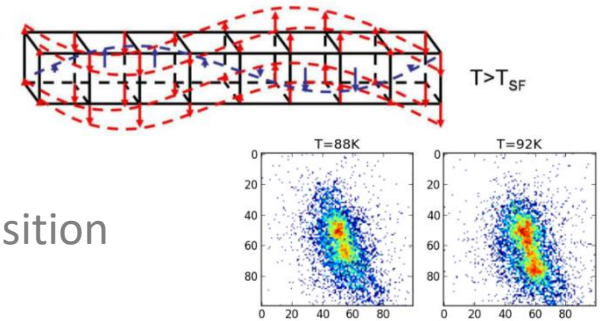


LSDW

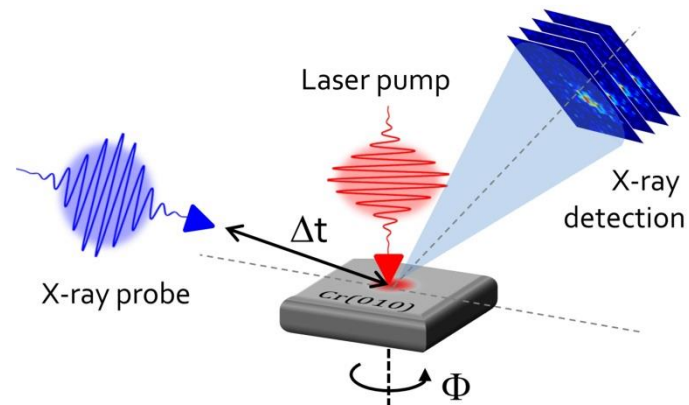
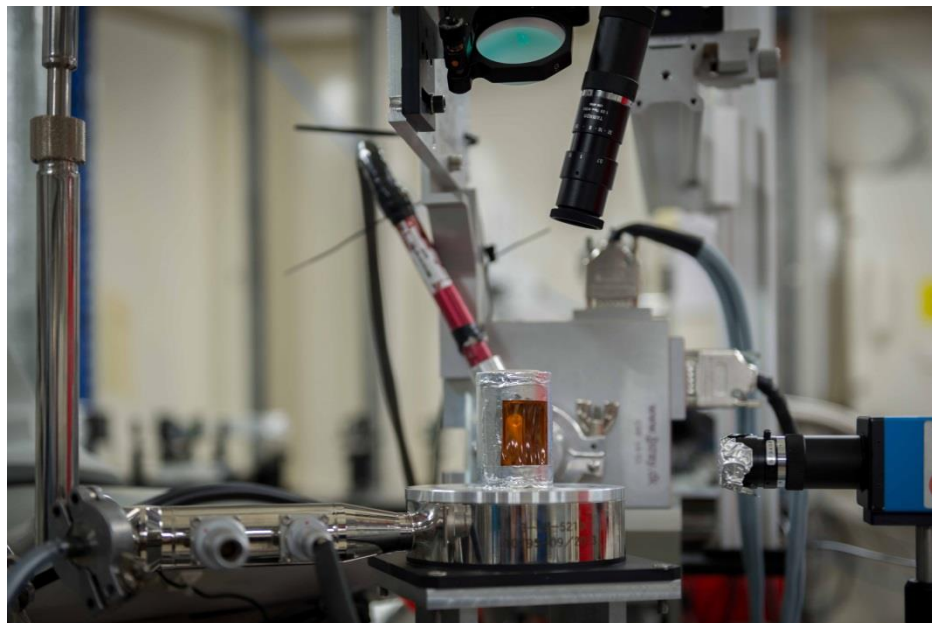


Outline

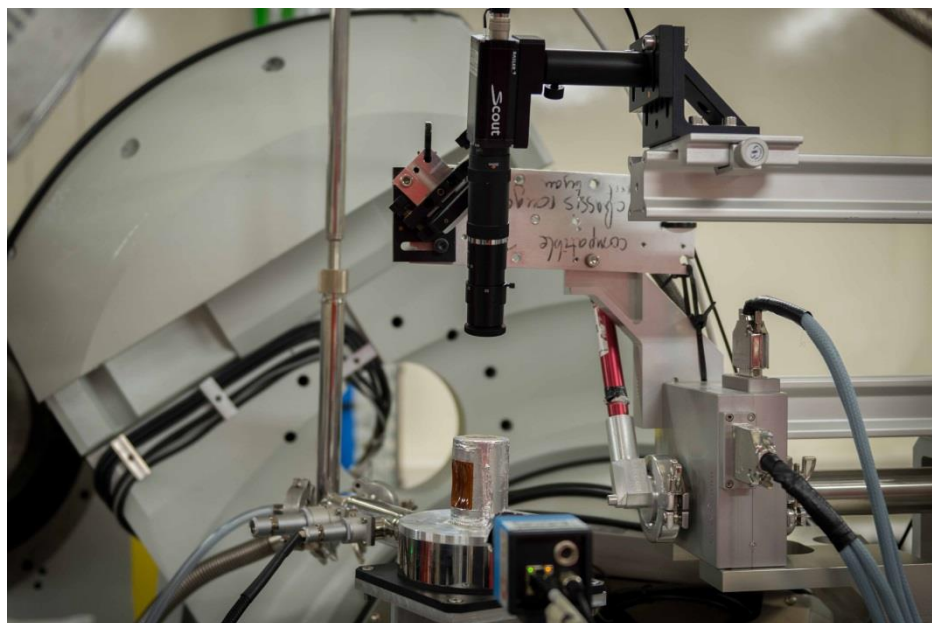
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ps time-resolved x-ray diffraction @ CRISTAL, Soleil Synchrotron



Multi-Q chromium sample



X-rays:

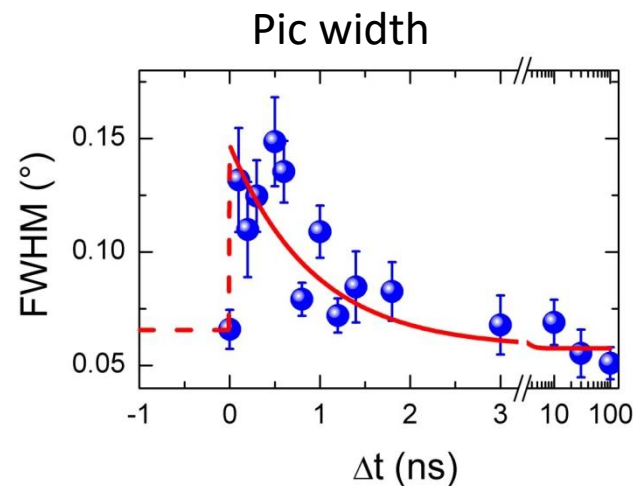
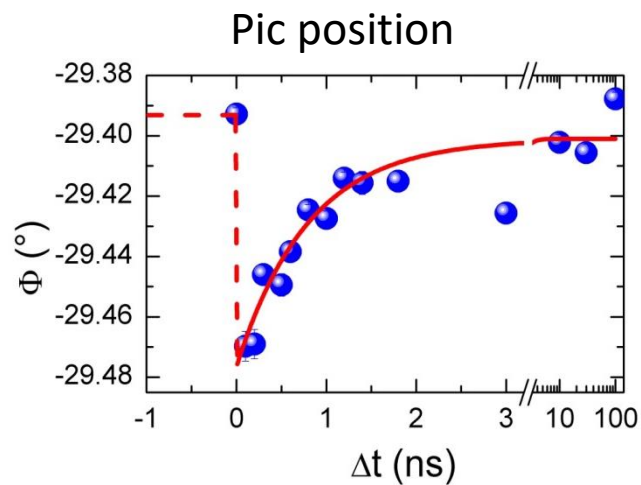
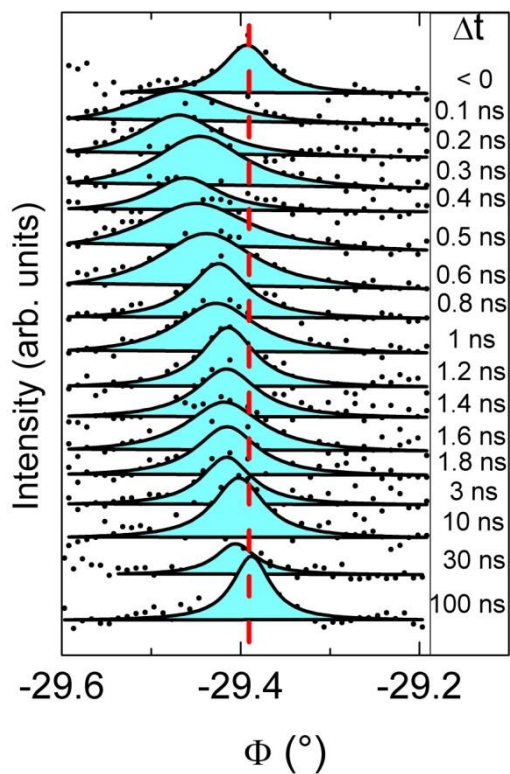
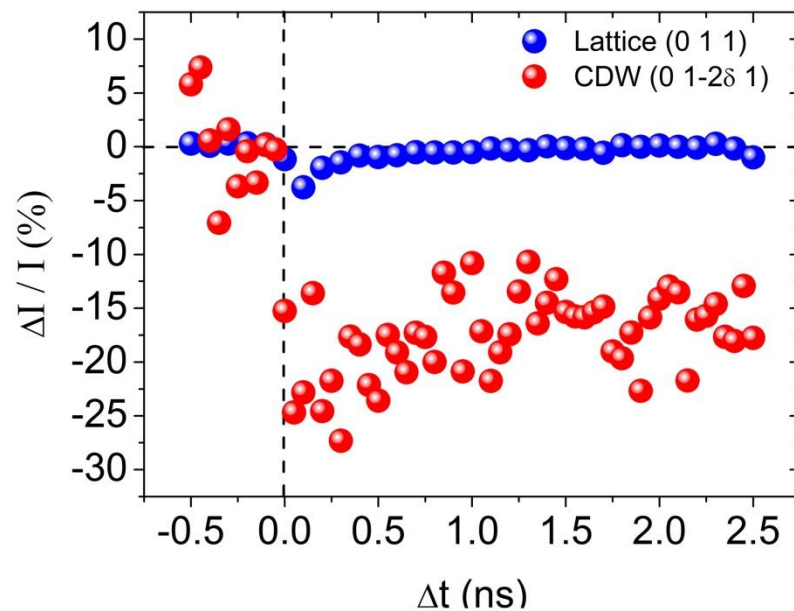
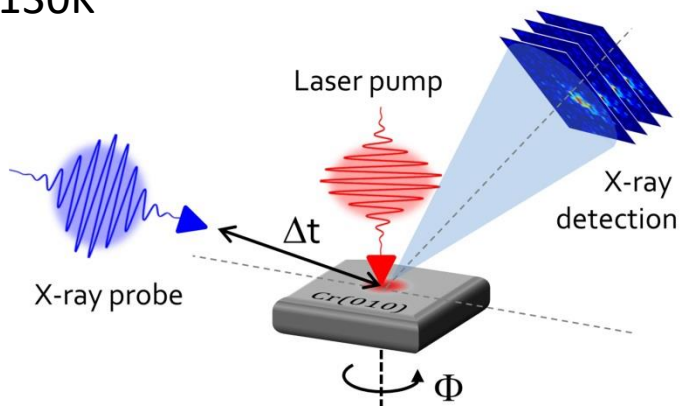
- 7keV
- 100nm penetration
- Detection: XPAD 3.2

Laser:

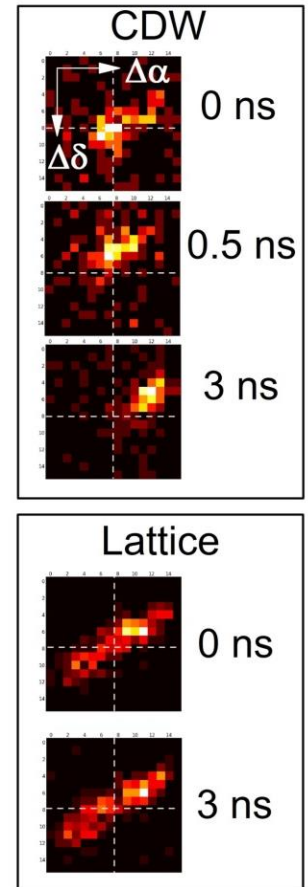
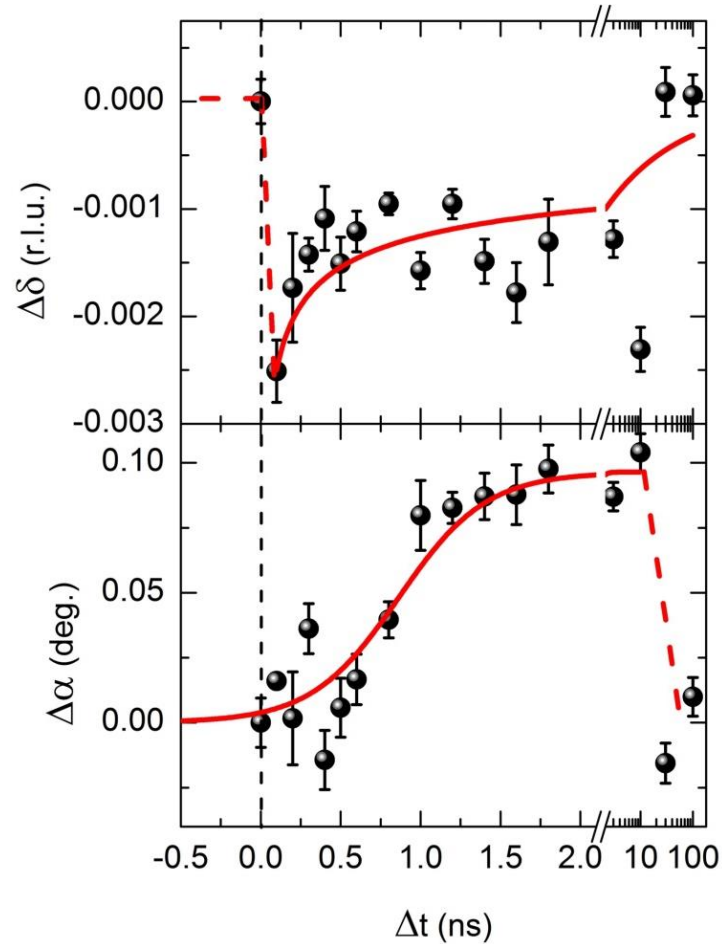
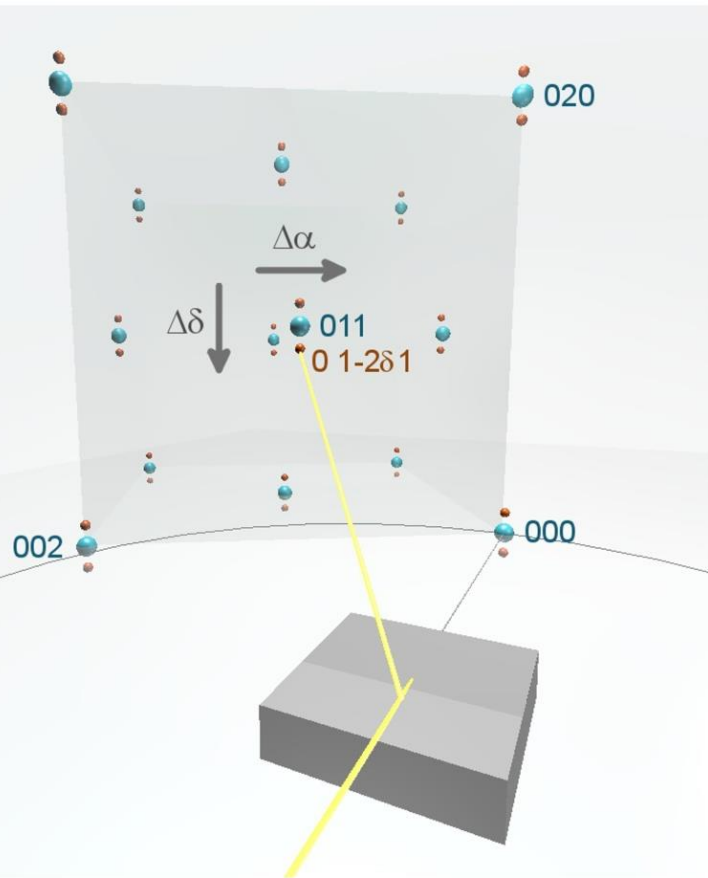
- 800nm, 30fs
- Up to 6.2mJ/pulse
- spot diameter: 2mm
- Rep rate: 1kHz

Time evolution of CDW and atomic lattice

T = 130K

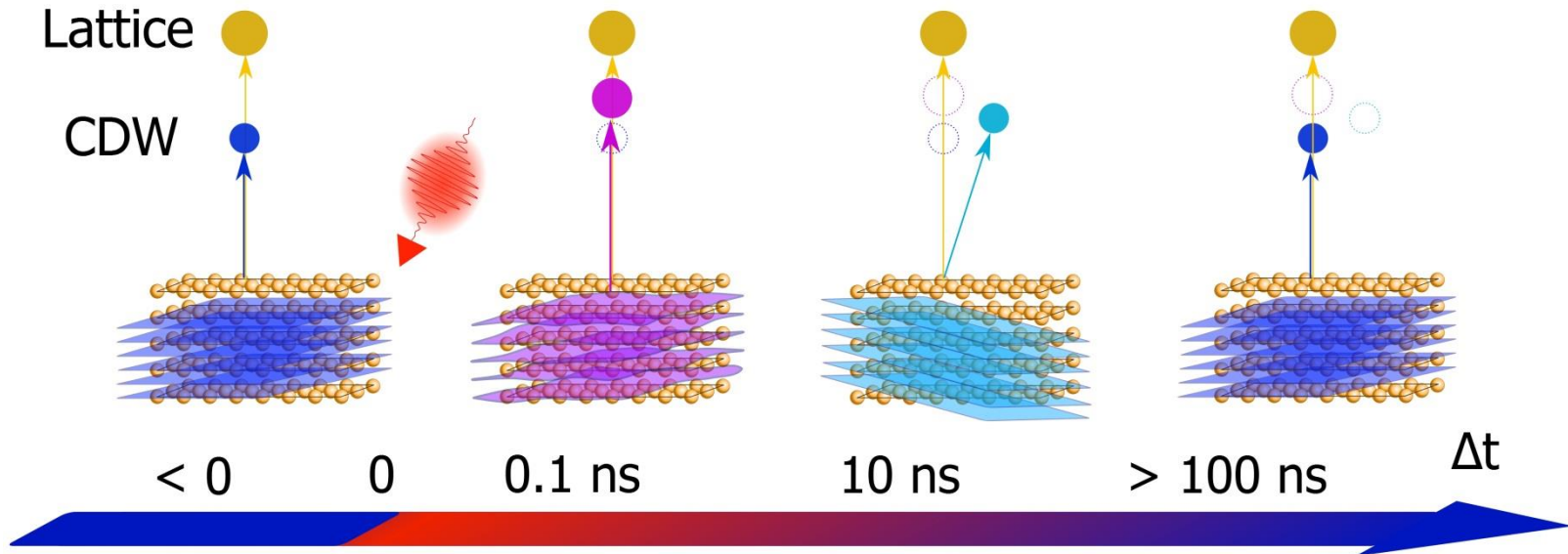


A different temporal evolution of the rotational component



Real space picture of out-of-equilibrium system

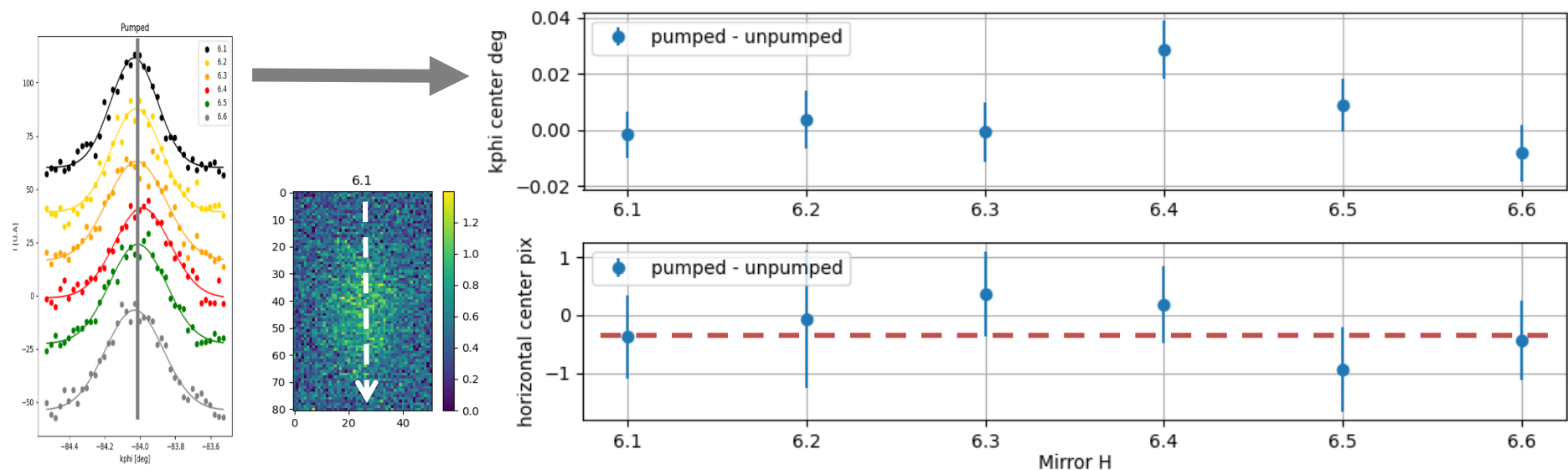
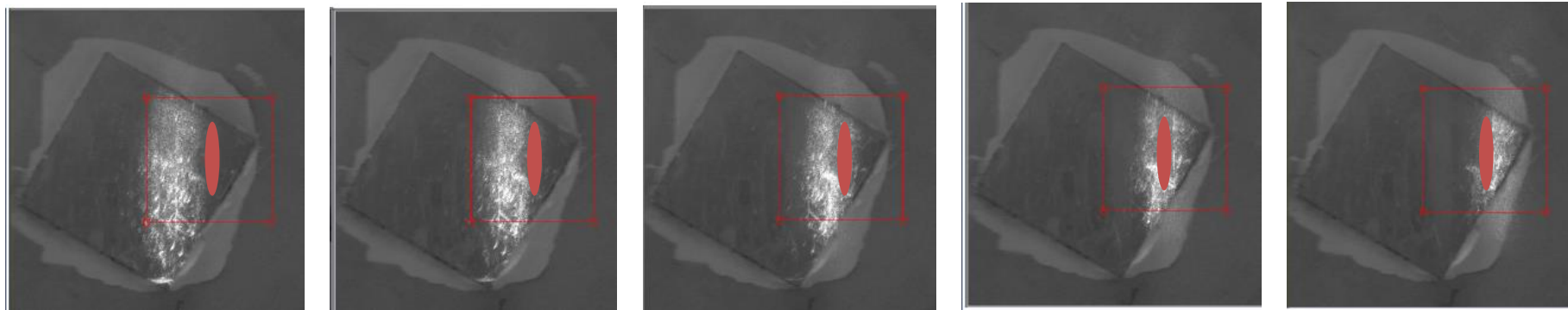
V. Jacques et al., PRL **117**, 156401 (2016)



- CDW expands within time resolution and recontracts after 2ns
- Tilt takes place at longer timescales (2-100 ns)

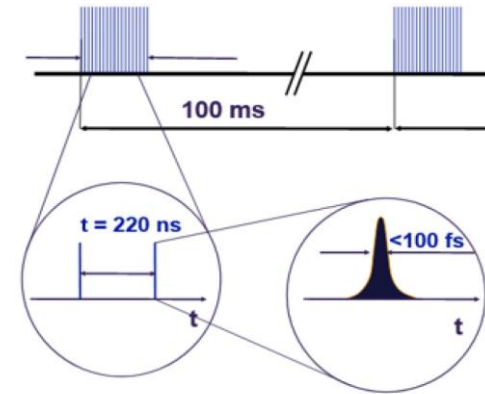
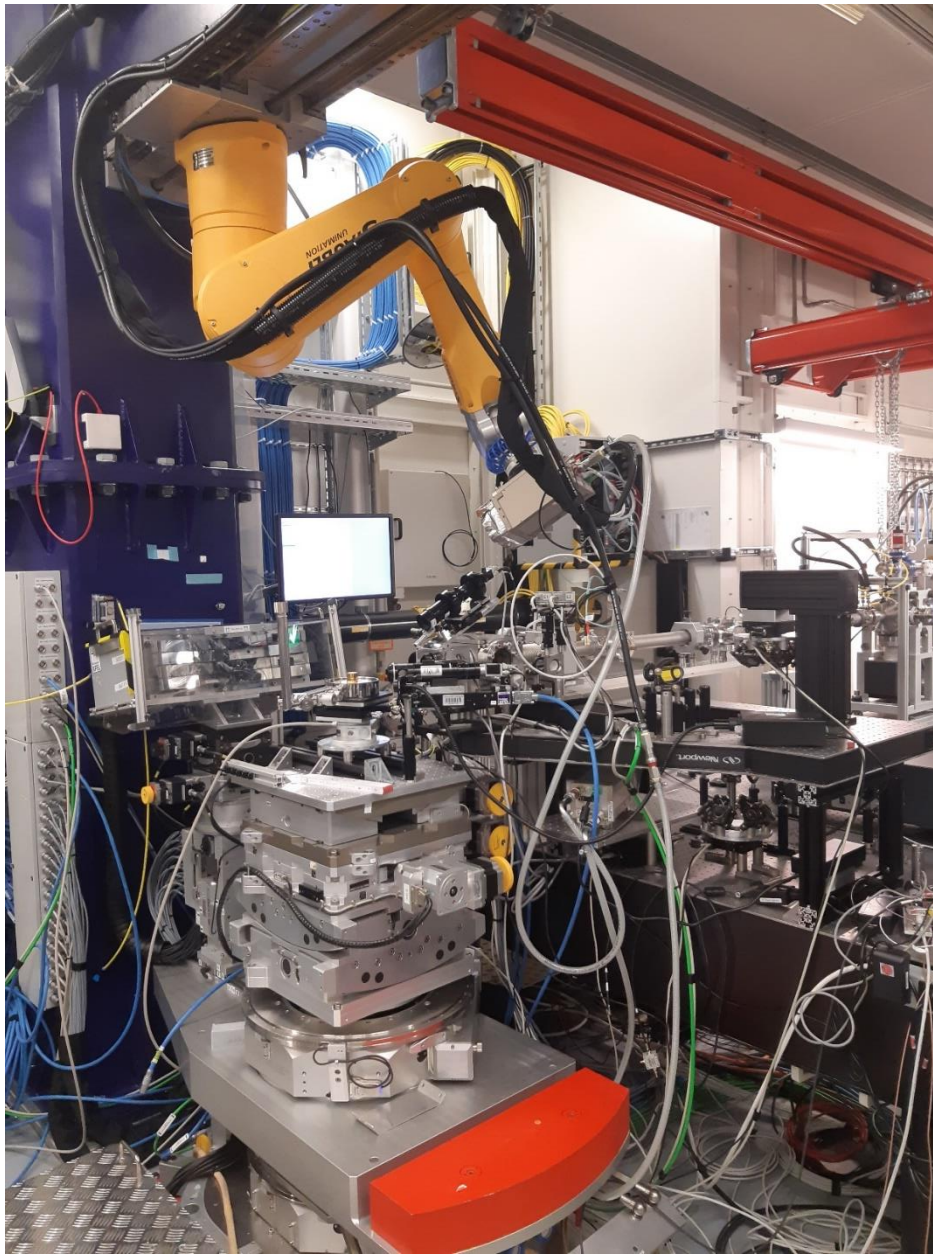
Tilt \rightarrow CDW decouples from atomic lattice \rightarrow CDW able to slide in chromium?

ps time-resolved x-ray diffraction @ CRISTAL, Soleil Synchrotron



➔ Tilt of the CDW due to strain gradient depending on laser position

fs time-resolved x-ray diffraction @ FXE, European XFEL



X-rays

- 1 bunch/train
- 30fs x-ray pulses
- $E = 5.989\text{ keV}$
- $10\mu\text{m}$ spot

Laser

- 30fs pulses
- 800 nm
- $10\mu\text{m}$ spot

Nearly - Colinear and grazing incidence of x-ray and laser beams

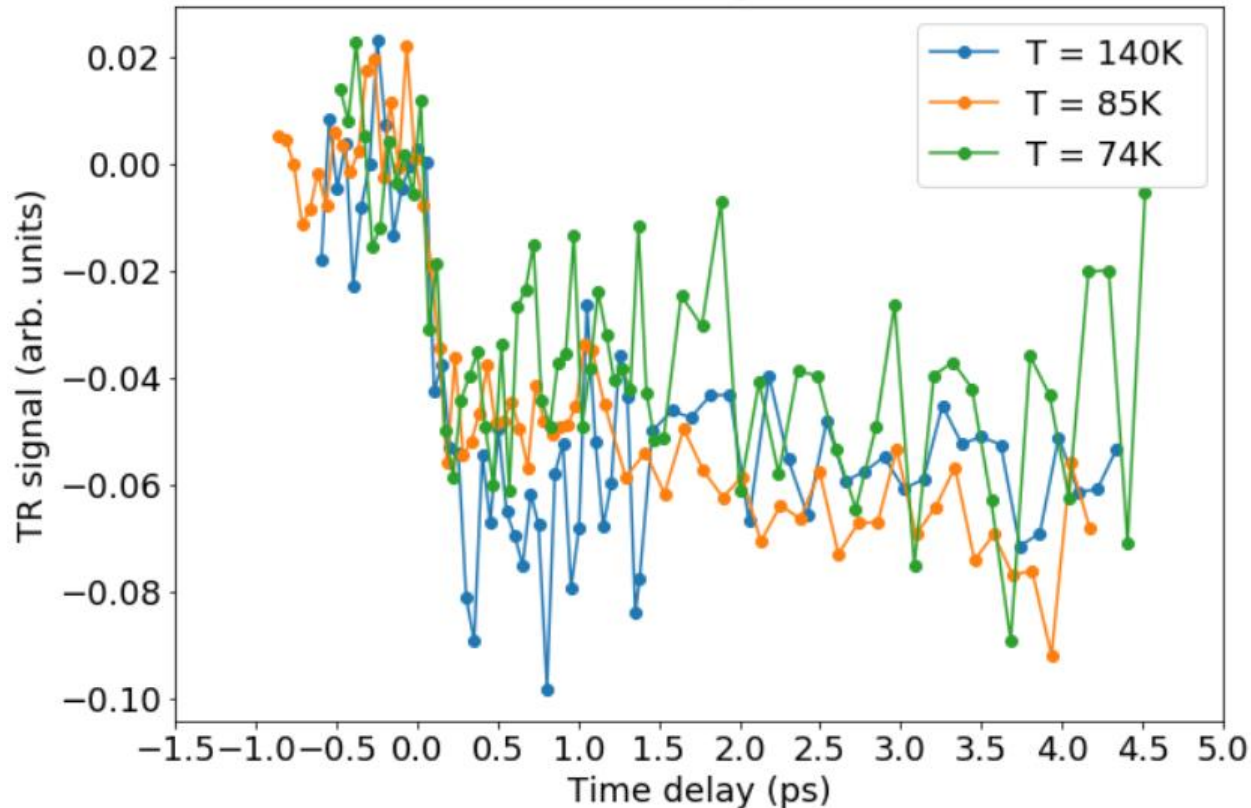


X-ray/laser entrance diamond window



X-ray exit window kapton ($125\mu\text{m}$)

Time-resolved measurement of the CDW through the spin-flip



- 3 successive steps :
 1. Ultrafast decay ($\sim 100\text{fs}$) \rightarrow CDW excitation
 2. Partial recovery ($\sim 1\text{ps}$) \rightarrow CDW reapparition
 3. Longer-time decay ($>1\text{ps}$) \rightarrow start of thermal transfer

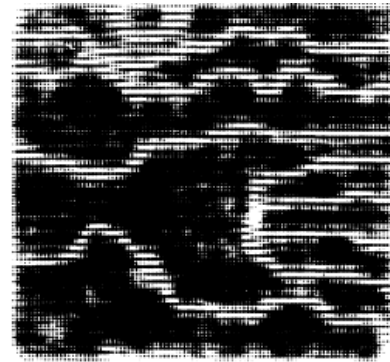
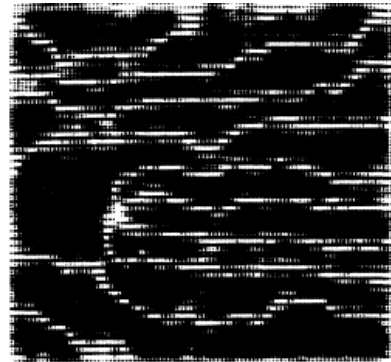
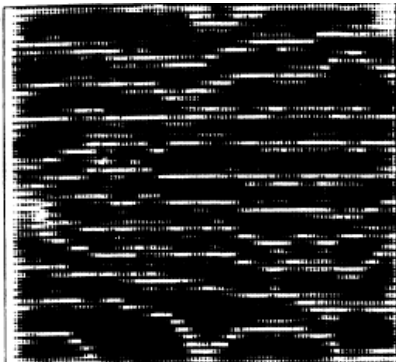
Dynamics of step 2 (CDW reapparition) is different (longer) near Spin-Flip temperature

Time-sequence: excitation process, dislocations, decoupling

1. Excitation process < 100 fs: most probably athermal excitations above gap (150 meV)
2. CDW partial recovery > 1 ps
3. Few ps: thermal effects : Debye-Waller + heat diffusion.

Debye Waller $\rightarrow \Delta a/a \sim 2 \cdot 10^{-4} <$ experimental resolution $\Delta q/q = 3.94 \cdot 10^{-4}$

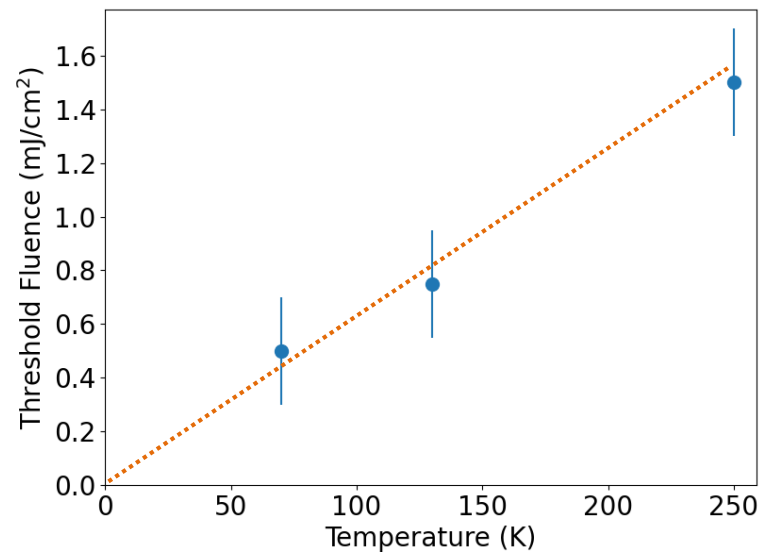
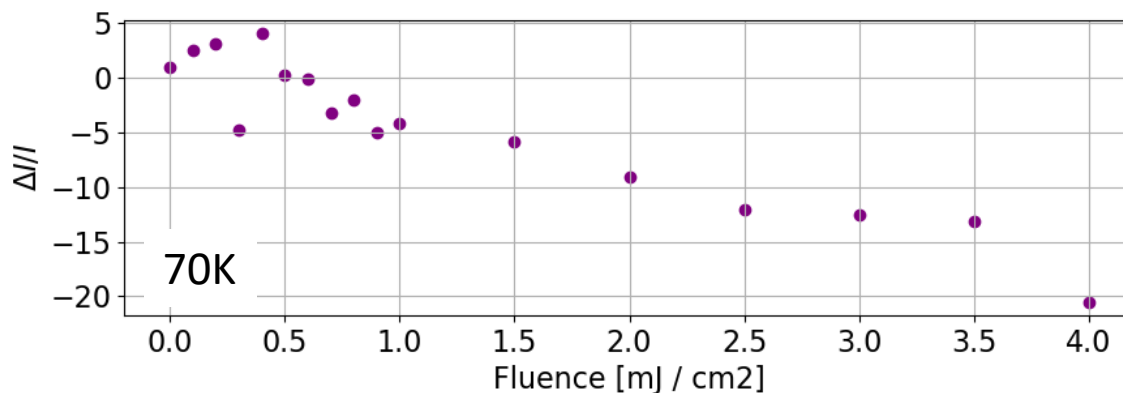
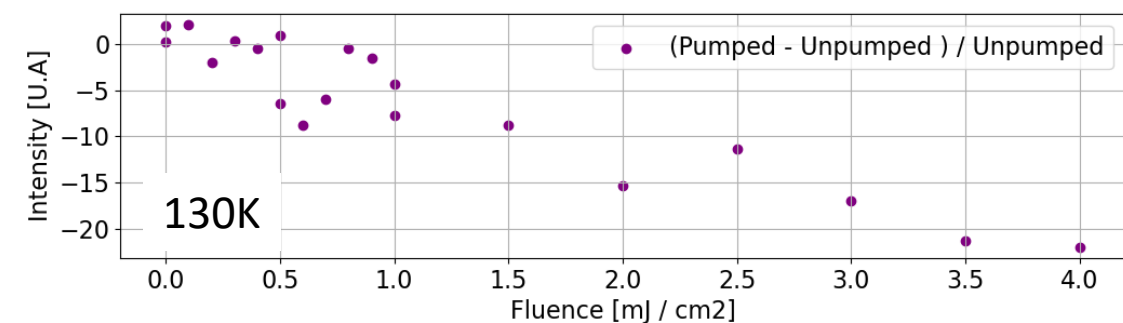
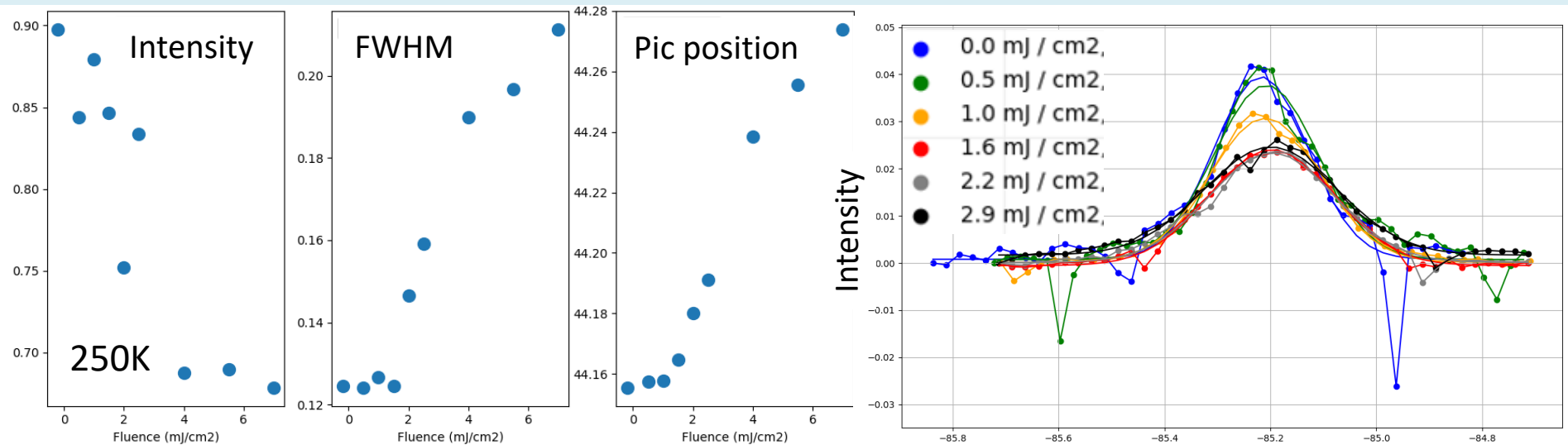
4. Within 500 ps: q_{cdw} change \rightarrow Dislocations involved?



Parlinski, PRB 35, 8680 (1987)

5. Tilt above 1 ns most probably to introduction of a strain-gradient by laser pulse

Threshold fluence for CDW excitation in multi-Q sample



Summary and perspectives

- Static SDW and CDW have **very different correlation length ξ**
 - SDW : $\xi_{SDW} \rightarrow \infty$
 - CDW : $\xi_{CDW} < 1\mu m$
 - CDW through the spin-flip transition :
 - **CDW domains are preserved** but **CDW ‘feels’ the transition** :
 - correlation length decreases
 - Amplitude decreases
 - After laser excitation : CDW partial recovery dynamics is different at SFT
 - **CDW decoupling from lattice** $\sim 2\text{ns}$ after laser excitation, due to strain gradient
 - **Threshold fluence** for excitation of the CDW
- Need to probe the SDW dynamics in the same conditions as CDW.
- Get more XFEL beamtime !

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