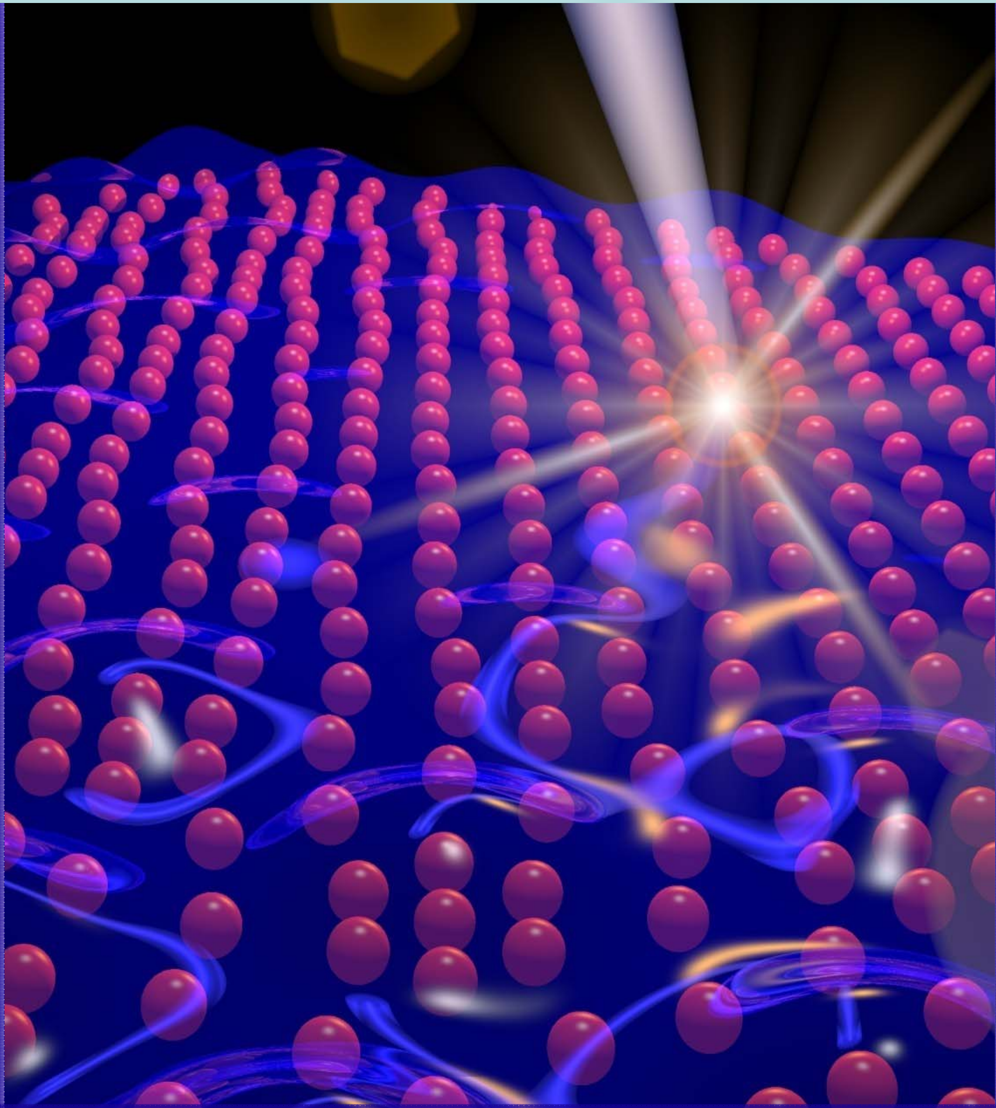
Photoinduced phase transitions in CDW's probed by ultrafast electron diffraction



M. Eichberger, H. Schäfer, G. Birmuske, C. Klose, M. Krumova (Konstanz)

G. Sciaini, G. Moriena, R.J.D. Miller (Toronto / Hamburg)

N. Erasmus, K. Haupt, H. Boshoff, G. Kassier, H. Schwörer (Stellenbosch)

K. Vorobeva, T. Huber, P. Beaud, G. Ingold, S. Johnson (ETH Zürich / PSI Villigen)

H. Berger (EPFL)

K. Biljakovic (Zagreb)

V.V. Kabanov, (*Ljubljana*)

Jure Demsar

FB Physik & CAP, Universität Konstanz

IMPACT, 11.9. 2012

Outline

- Motivation

- Observing a Peierls distortion on the few-hundred-femtosecond timescale over which it occurs (1T-TaS_2)

- Order parameter dynamics and PI C-IC phase transition in 4Hb-$TaSe_2$

- Summary

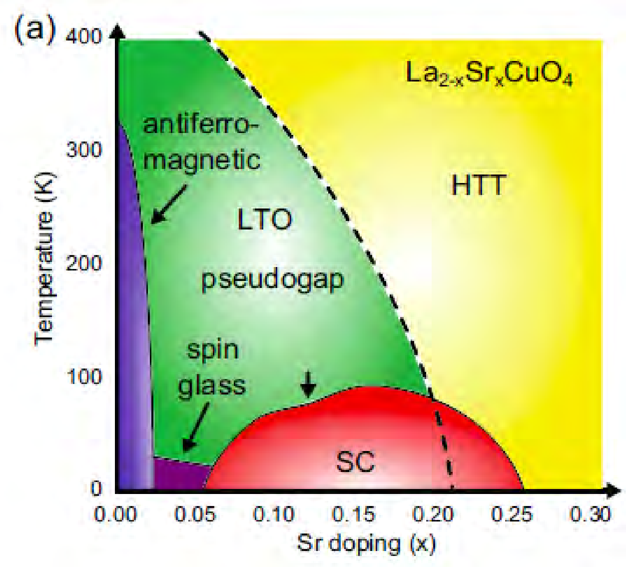
Motivation

Advanced quantum solids (e.g. high-T_c superconductivity, colossal magneto-resistance, multiferroicity):

- functional properties due to a delicate balance among different interactions between electrons, phonons, and spins on the nanoscale.
- Their **equilibrium properties** (e.g. electronic structure) **not well understood**!

Prime example: cuprate high-T_c SC:

- Undoped: CT Insulator
- Nature of excitations?
- Doping - new states within CT gap?
- Origin of states giving rise to SC?
- AFM vs. SC: competition or cooperation?
- Origin of normal state pseudogap?



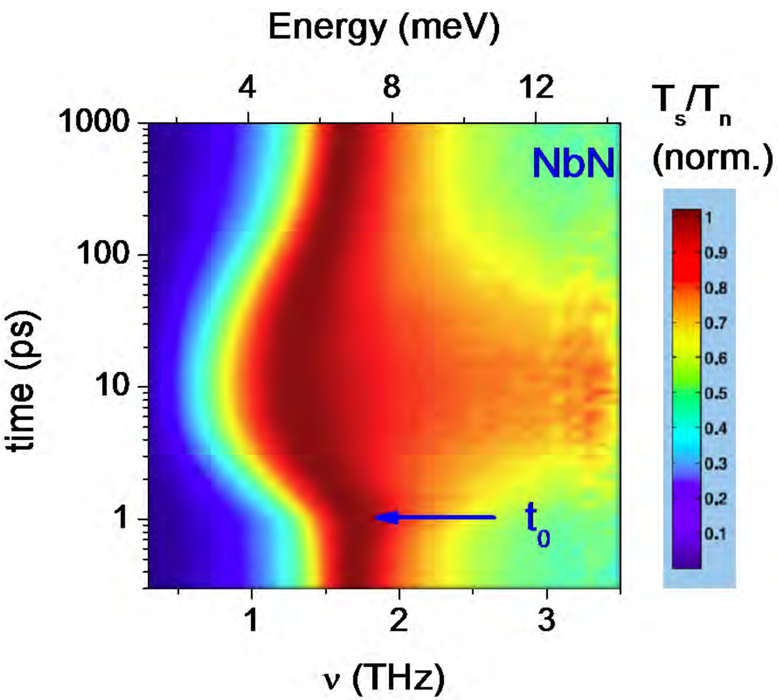
Wells, et al., *Science* **277**, 1067 (1997).

Approach:

- Drive the system far from equilibrium and observe its re-assembly on the **elementary time and atomic scale**.
- Determine the **interplay and coupling strengths** between the different degrees of freedom -> **manipulation**!

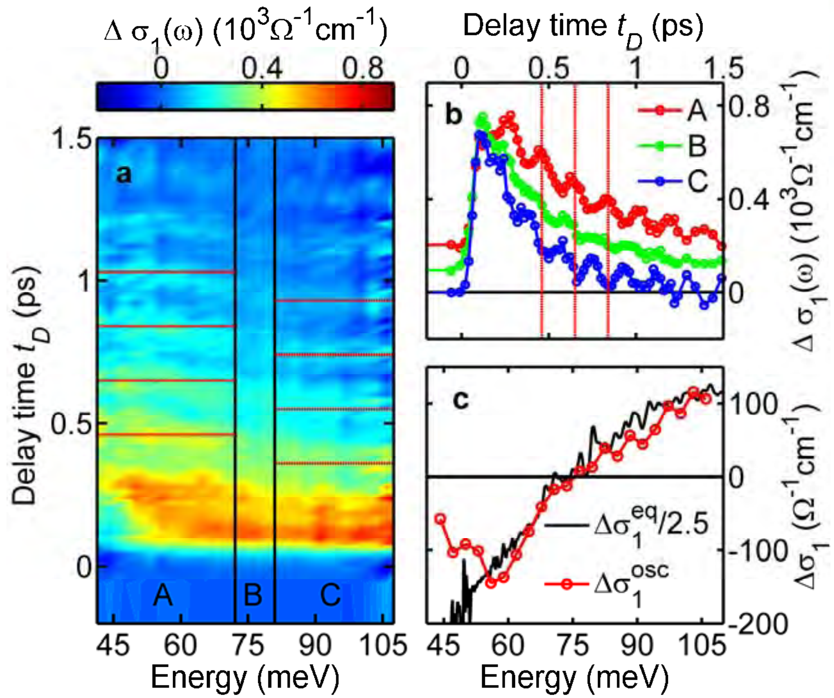
Examples: Gap dynamics in SC or SDW's

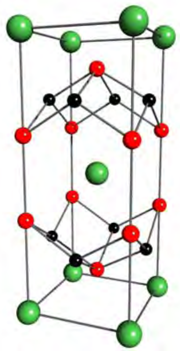
NbN

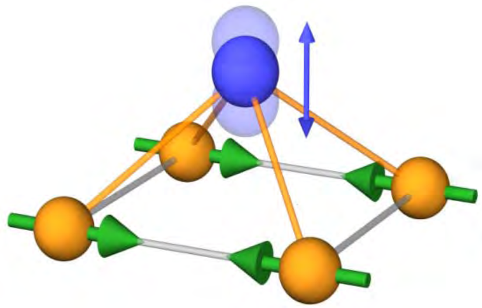


Beck et al., *PRL* 107, 177007 (2011).

$BaFe_2As_2$

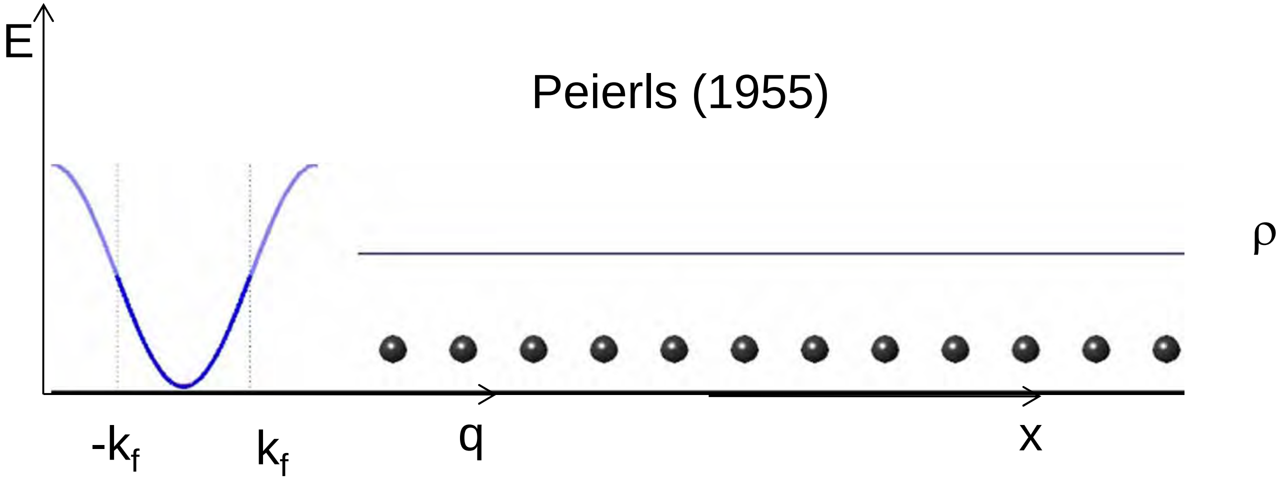


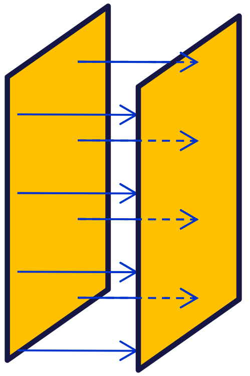




Kim et al., *Nature Mat.* **11**, 497 (2012).

Charge Density Waves





Response of electron gas to potential

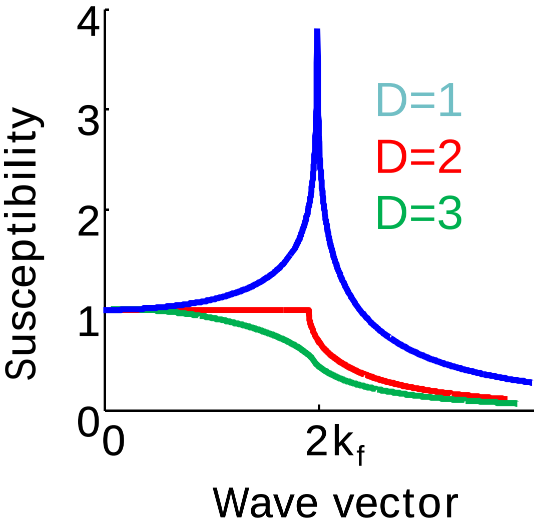
$$\rho_{\text{ind}}(\mathbf{q}) = \chi(\mathbf{q})\varphi(\mathbf{q})$$

Lindhard response function (1D):

$$\chi(\mathbf{q}) = \frac{1}{2\pi}\int d\mathbf{k}\,\frac{f_{\mathbf{k}} - f_{\mathbf{k+q}}}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k+q}}} = -\frac{e^2}{\pi\hbar v_F}\ln\left|\frac{\mathbf{q}+2\mathbf{k}_F}{\mathbf{q}-2\mathbf{k}_F}\right|$$

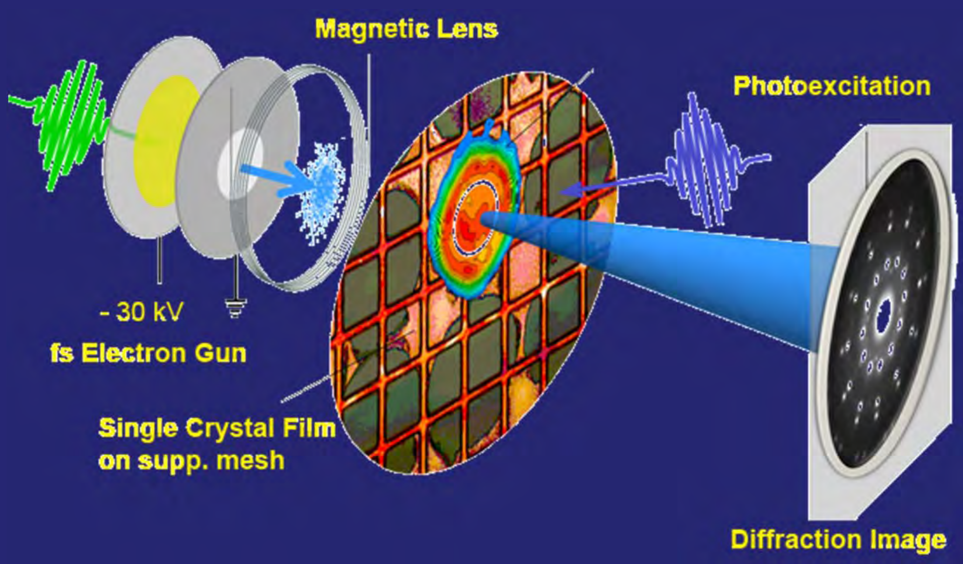
Divergence at $\mathbf{q} = 2\mathbf{k}_f$

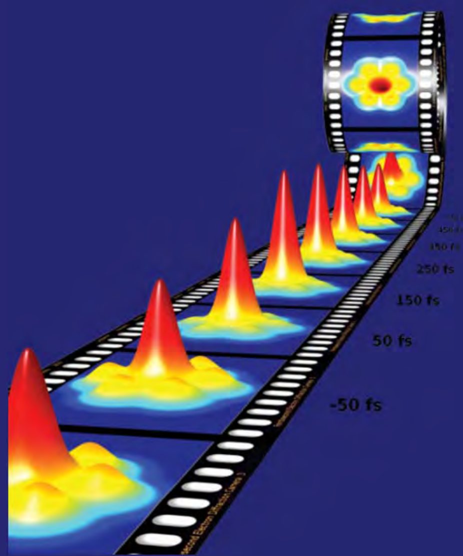
Lindhard-Response Function



Making molecular movies with a table top FED:

“Observing a Peierls distortion on the few-hundred-fs timescale over which it occurs”



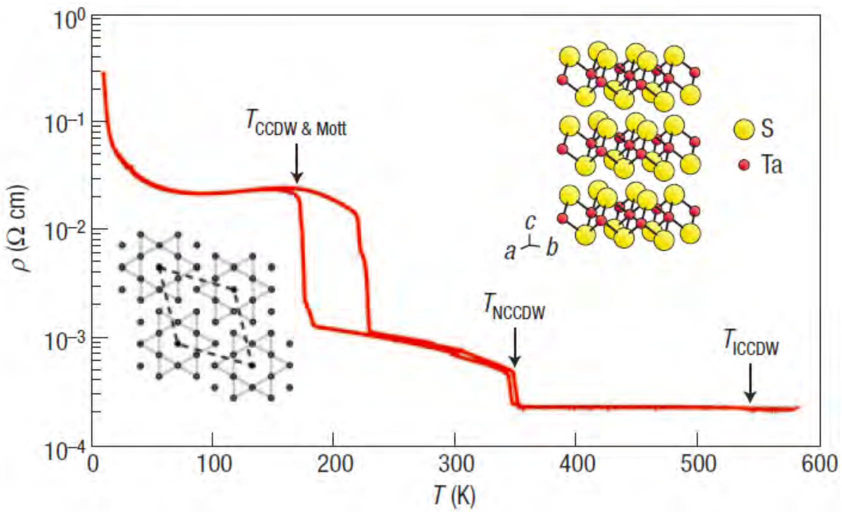


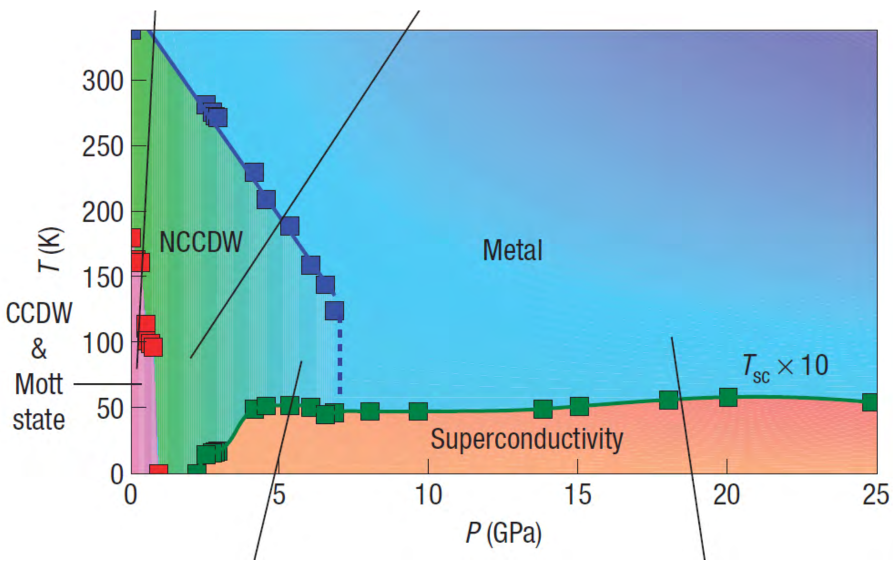
<u>Charge density waves</u>

Open questions:

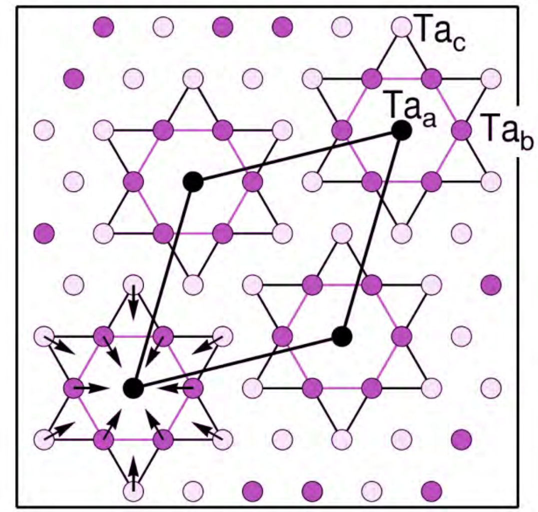
- CDW mechanism – Peierls vs. strong e-ph interaction
- Existence of pure 2D CDW (Berezinskii-Kosterlitz-Thouless type)?
- CDW’s and superconductivity

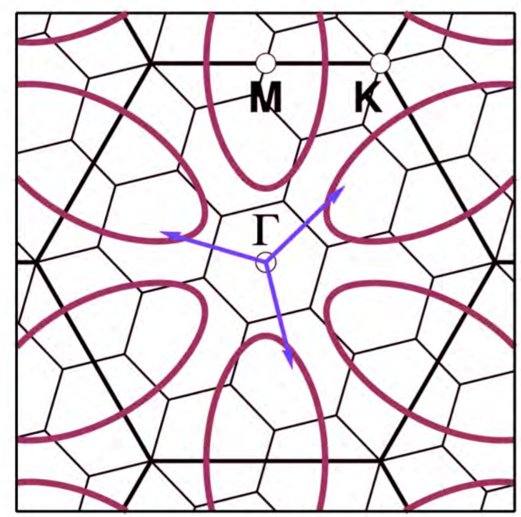
1T-TaS_2: Electronic Structure, CDW phases





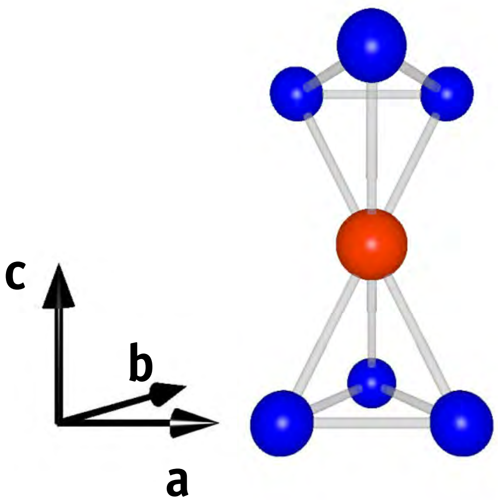
Sipos *et al.*, Nature Mat. **7** 960-965 (2008)



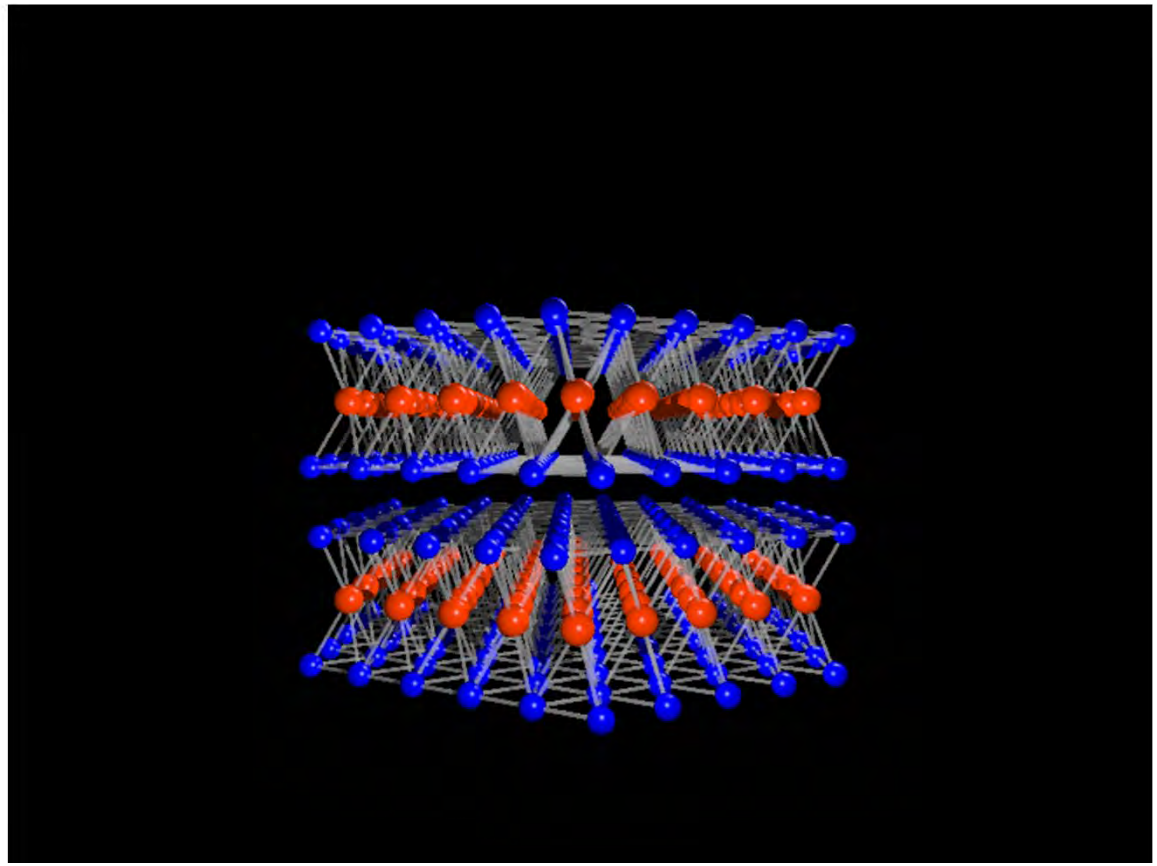


Rossnagel, J. Phys.: Cond. Matt. 23 (2011) 213001

1T-TaS_2 : quasi-2D CDW compound

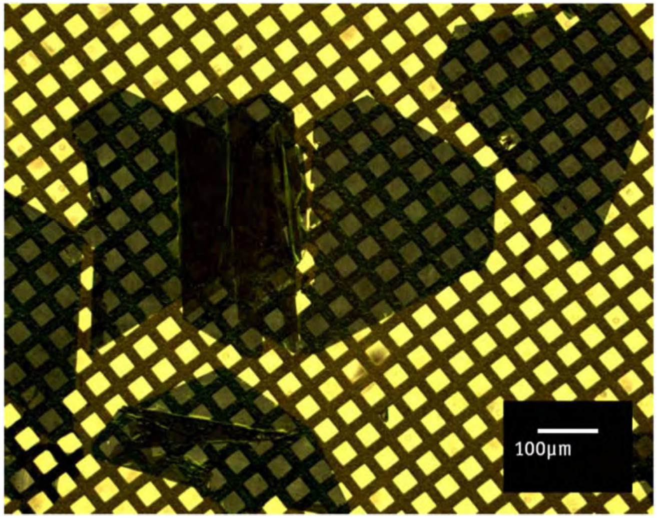


a_0=b_0=3.36 Å,
c_0=5.89 Å

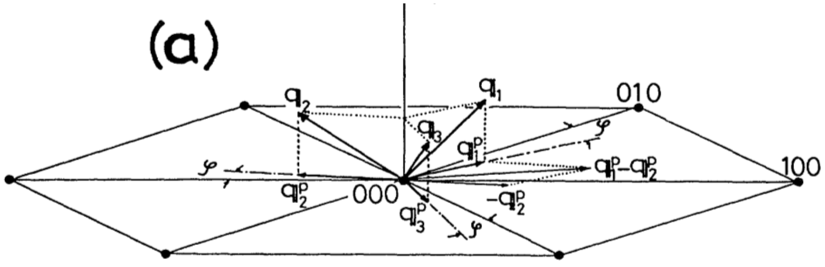


Highly exaggerated: **maximum displacement ~ 0.1 Å**

$1T\text{-}TaS_2$: free standing nm films

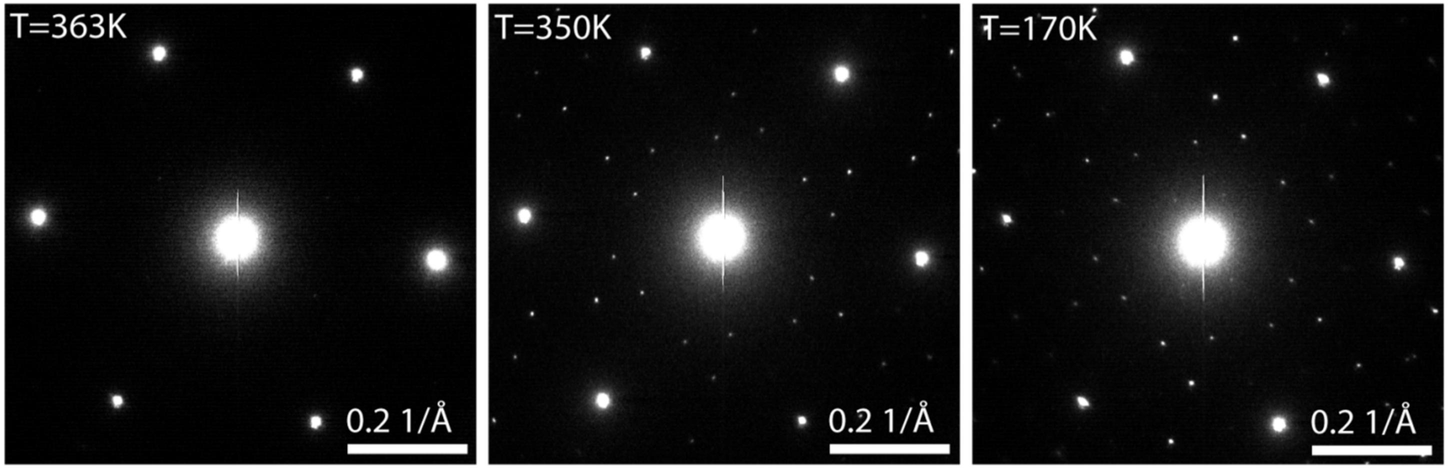


~ 30 nm single crystalline free standing films (ultramicrotome)



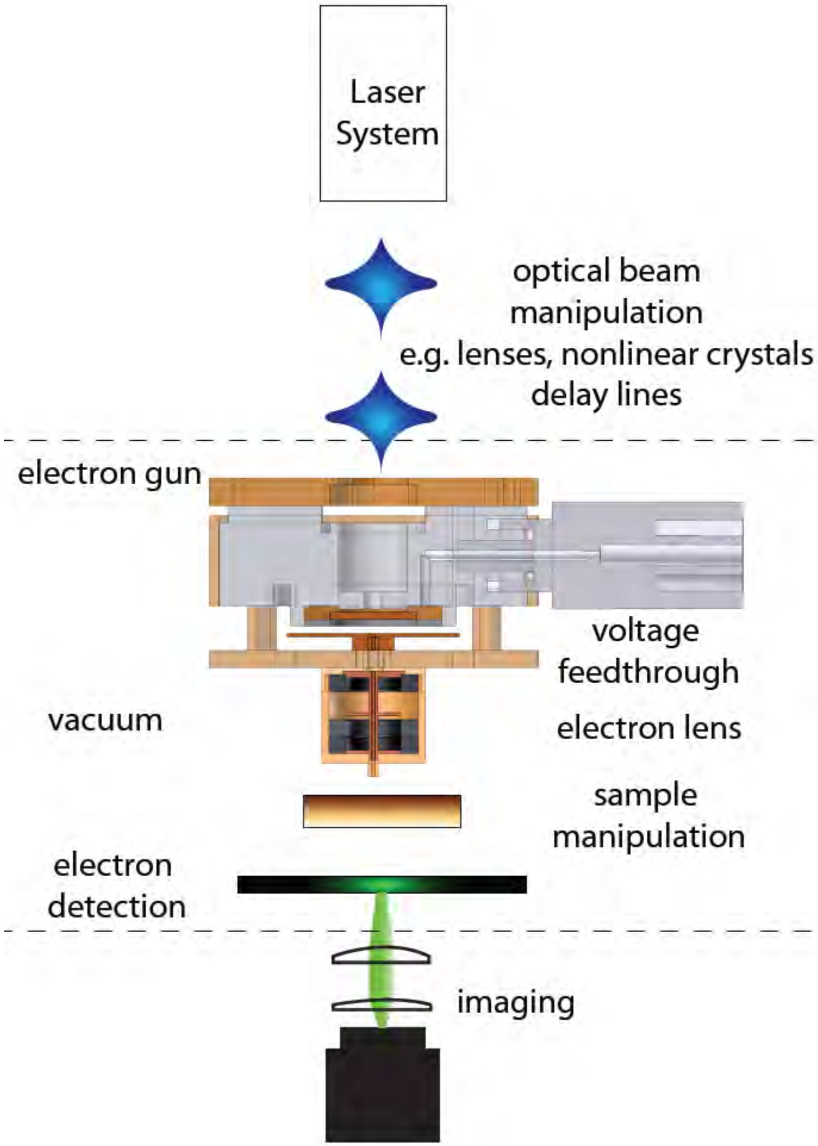
T. Ishiguro et al, Phys. Rev. B 52, 759-765 (1995)

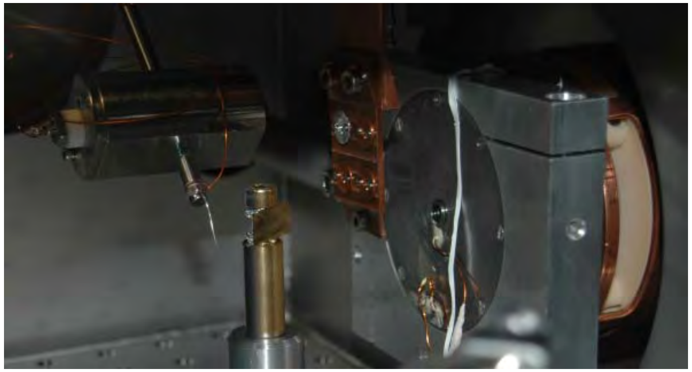
TEM characterization

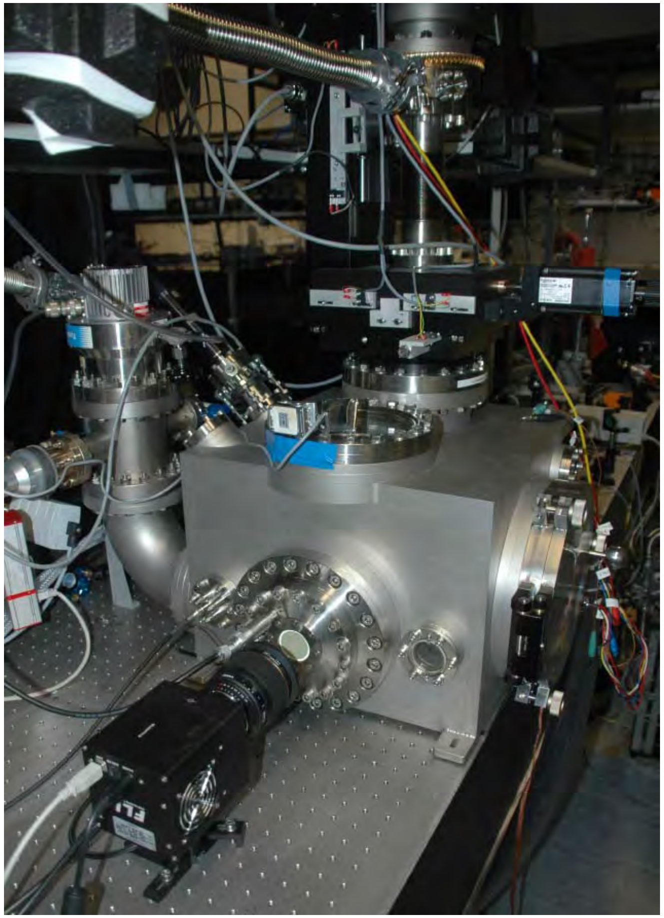


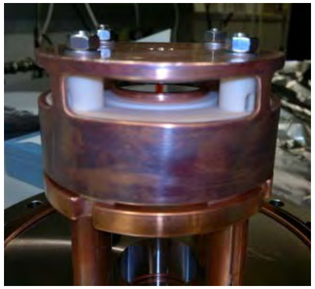
Eichberger et al., Ultramicroscopy (2012).

Femtosecond electron diffraction set-up

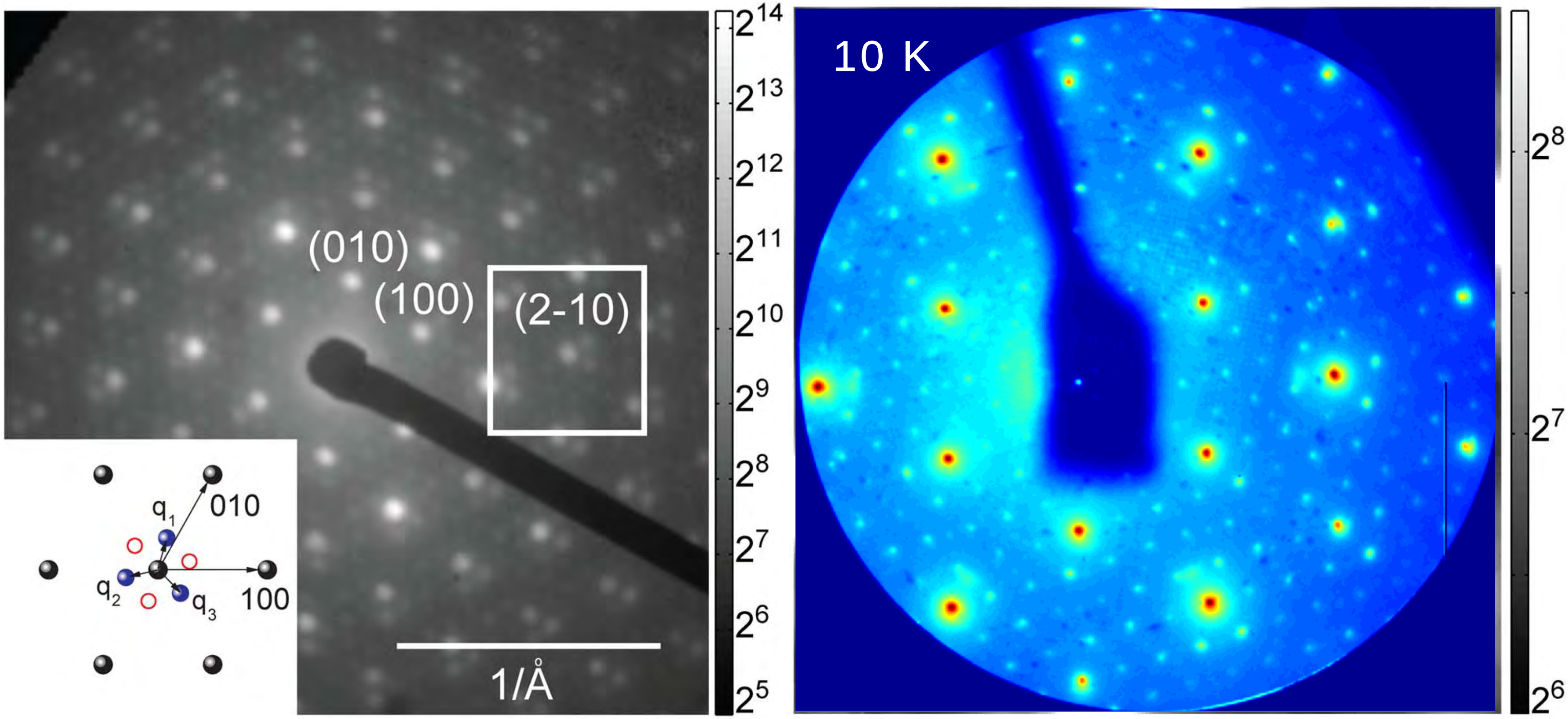




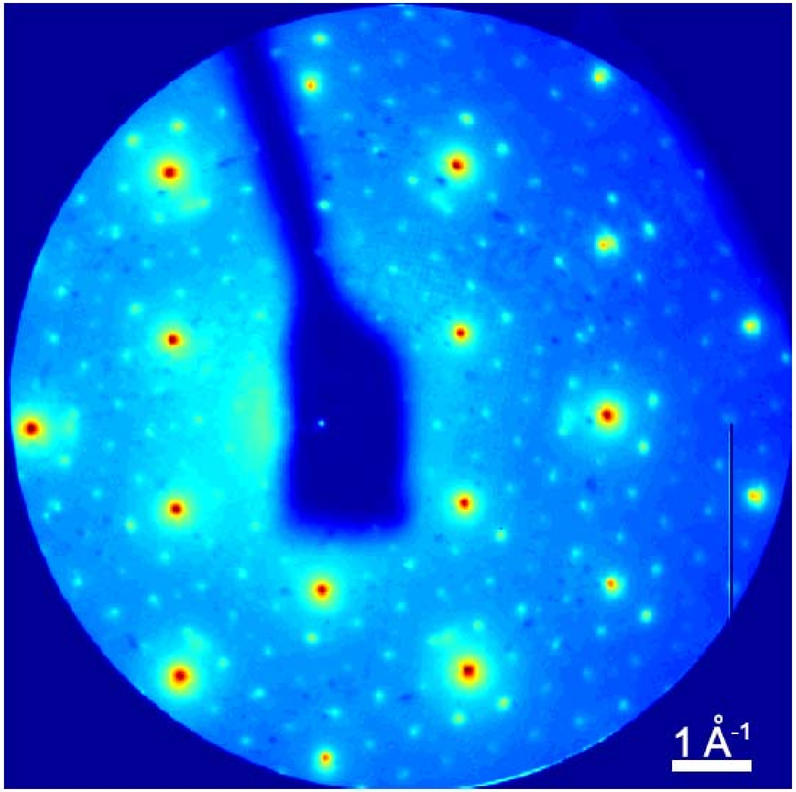




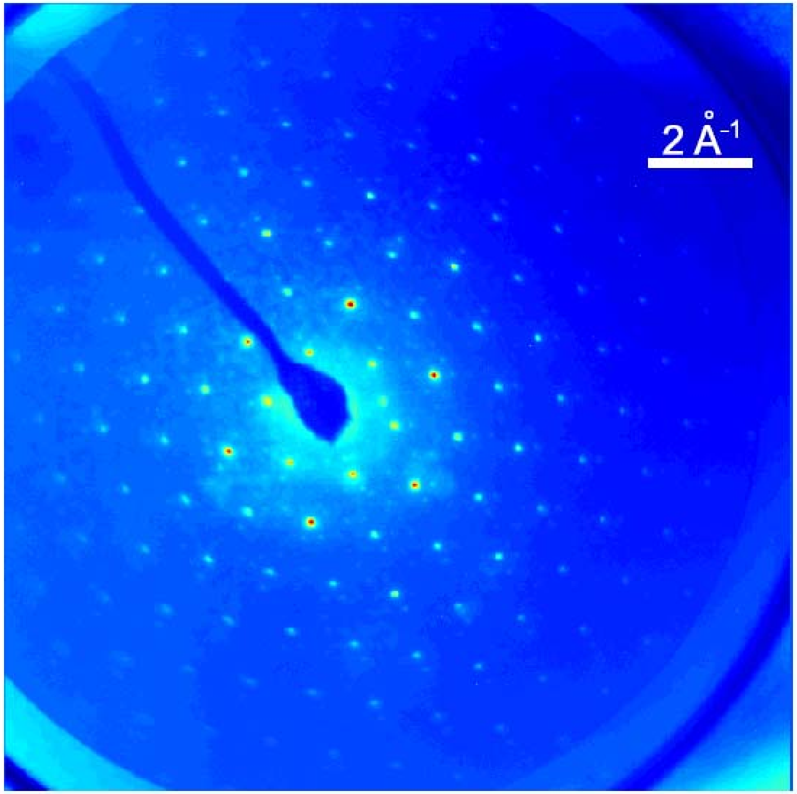
1T-TaS_2 : static diffraction (200 K)



FED setup: static diffraction (10 K)

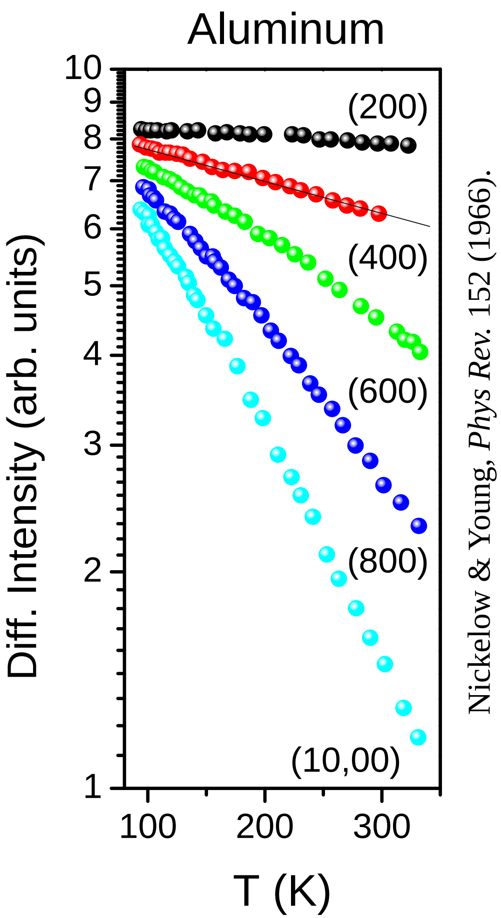


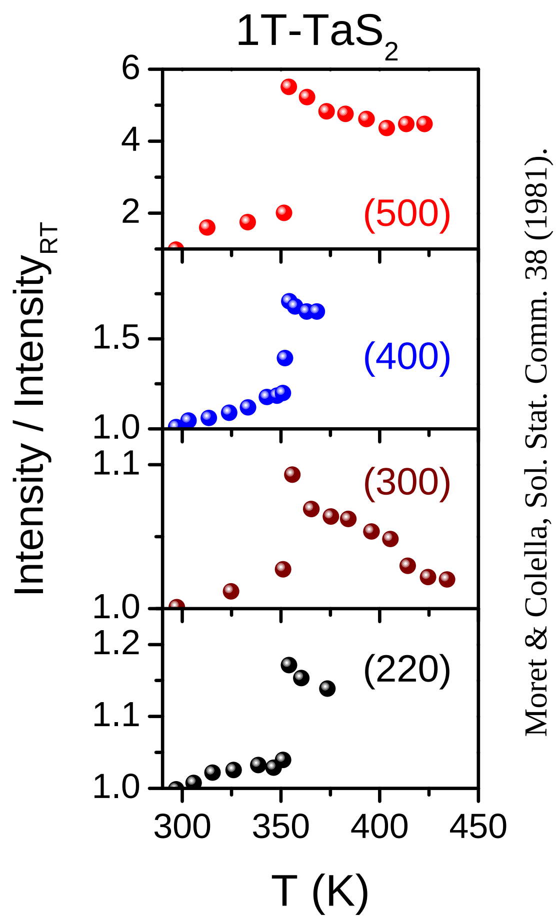
$1T\text{-}TaS_2$

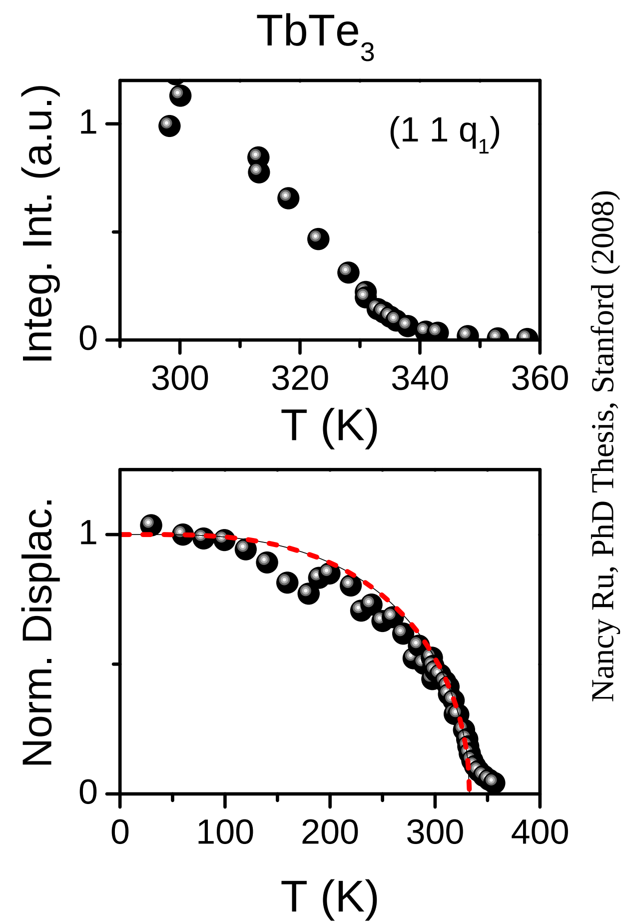


$4Hb\text{-}TaSe_2$

Manifestation of CDW order in thermal equilibrium







Intensity of **Bragg (Lattice) reflections** follows the Debye-Waller law:

$$I_Q = I_Q^0 \exp\left(-\tfrac{1}{3} Q^2 \langle u_Q^2 \rangle\right) = I_0 \exp\left(-Q^2 \frac{k_B T}{M\omega^2}\right)$$

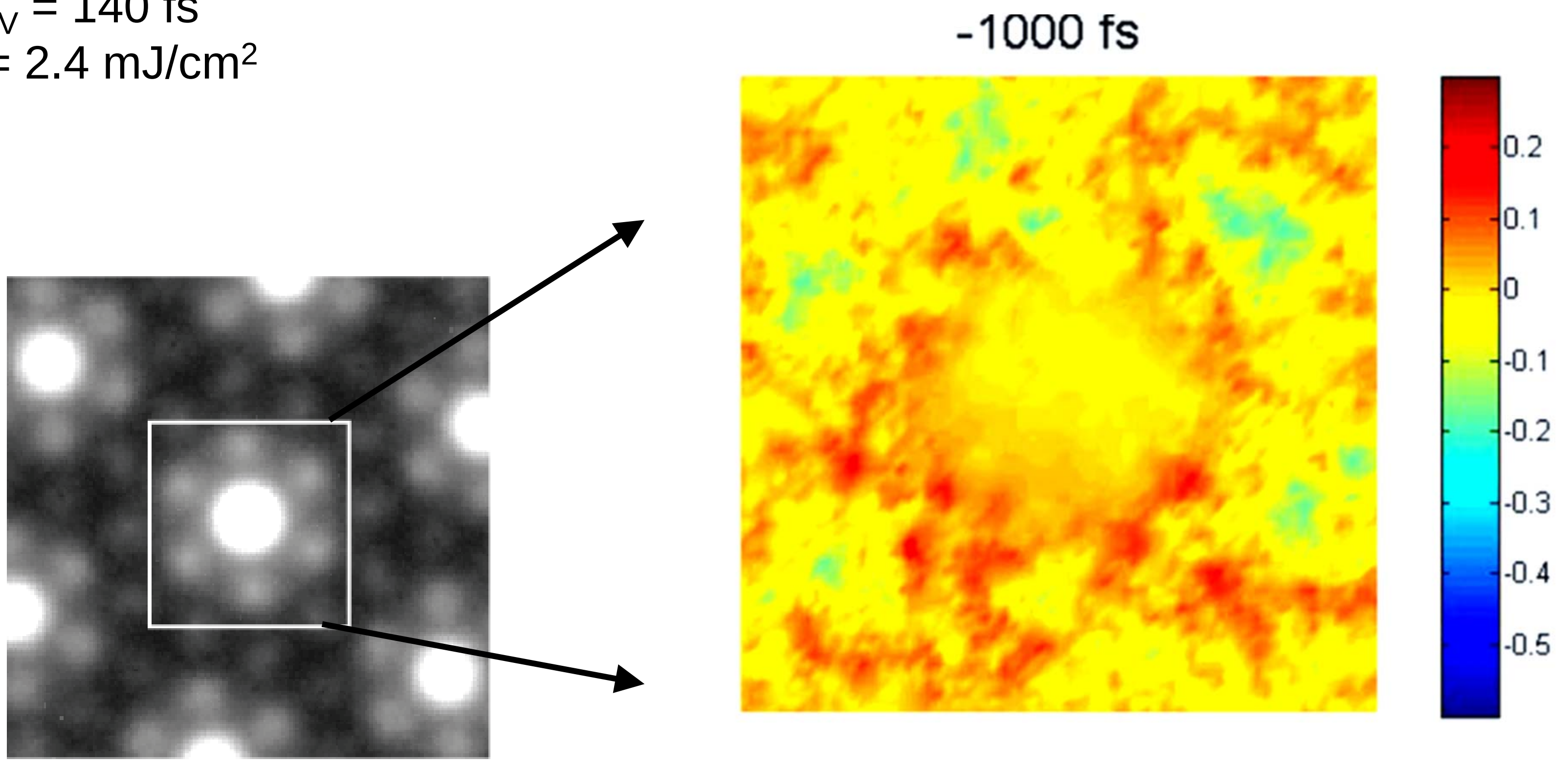
CDW's: Intensity of **Bragg (Lattice) reflections** affected by the presence of PLD!

Intensity of the **CDW (superlattice) reflections**:

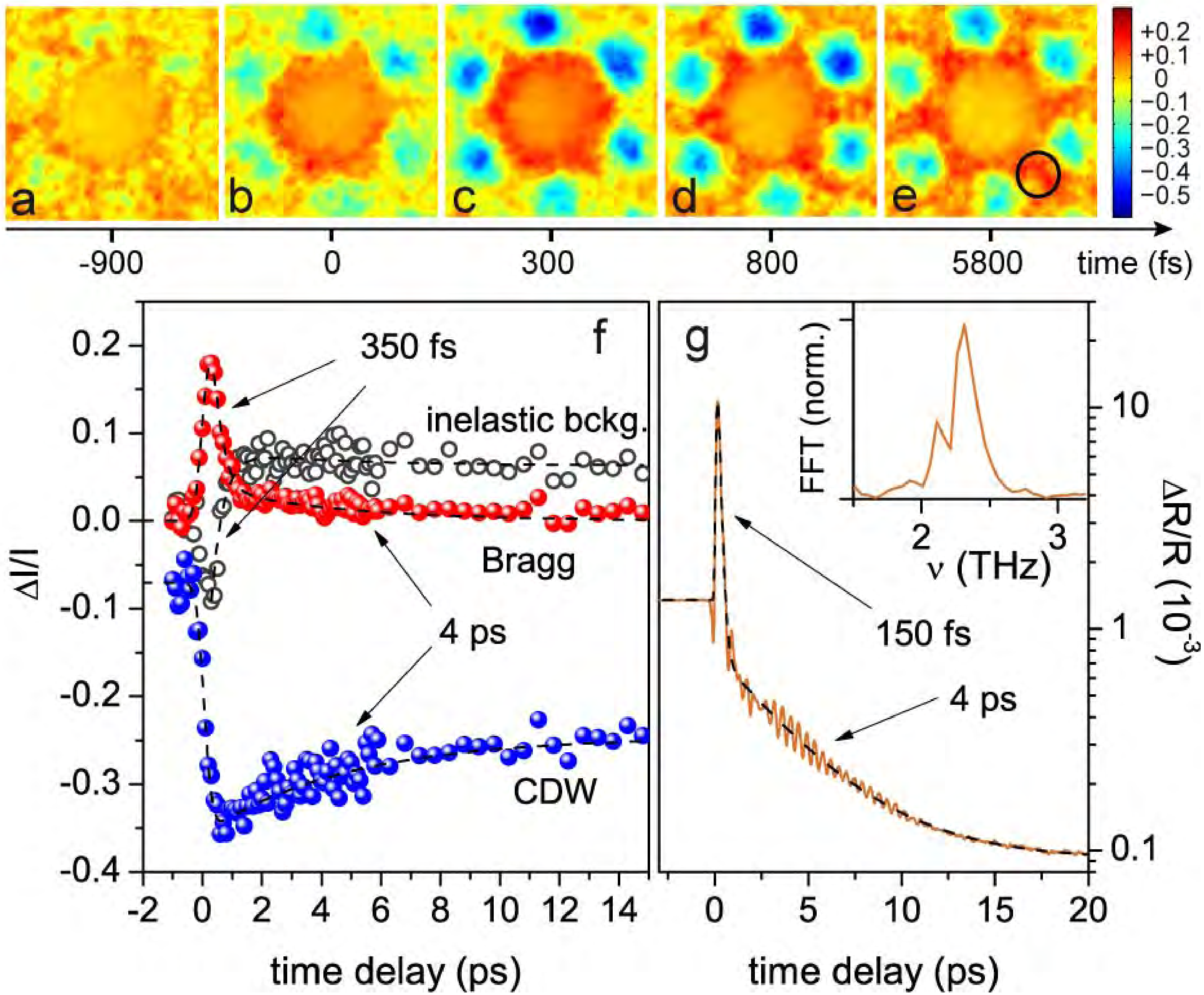
$$I_{CDW} \propto Q_{CDW}^2 \cdot A^2$$

Time-evolution of the diffraction on 1T-TaS_2

T = 200 K
τ_{el} = 200 fs
τ_{3eV} = 140 fs
F = 2.4 mJ/cm^2

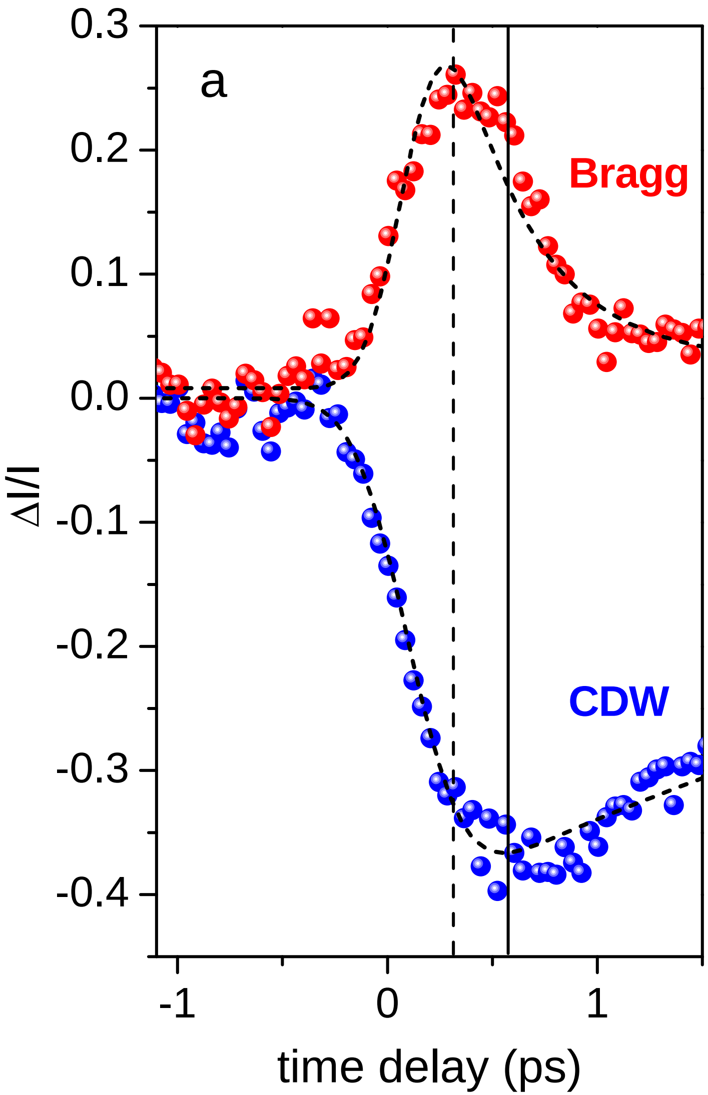


Time evolution of Bragg, CDW and inelastic bckg.

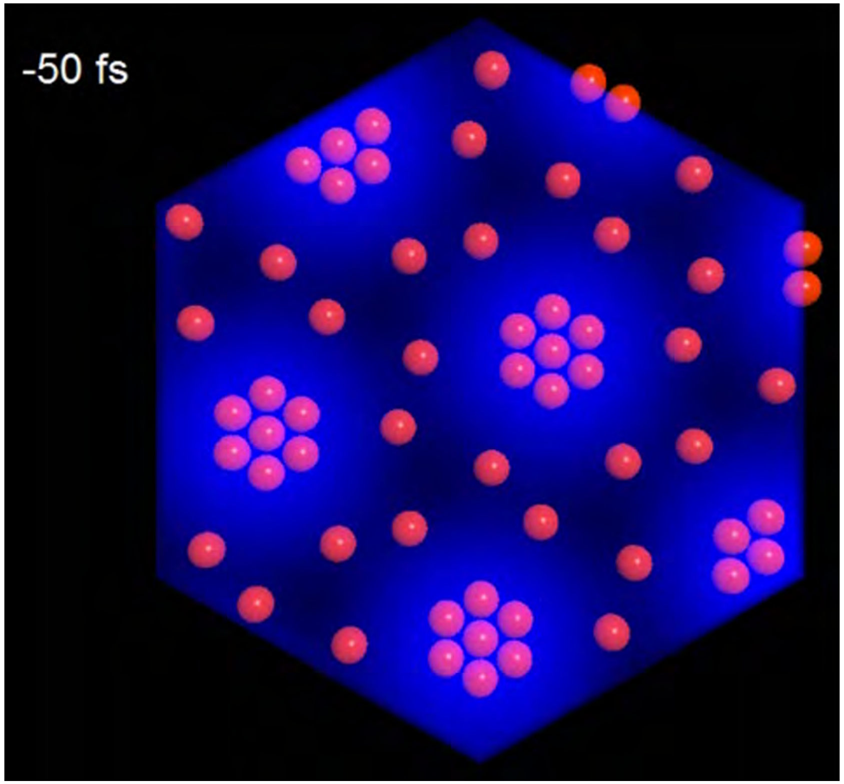


Eichberger et al., Nature 468, 799 (2010).

1T-TaS_2 - Processes and characteristic timescales



Eichberger et al., Nature 468, 799 (2010).

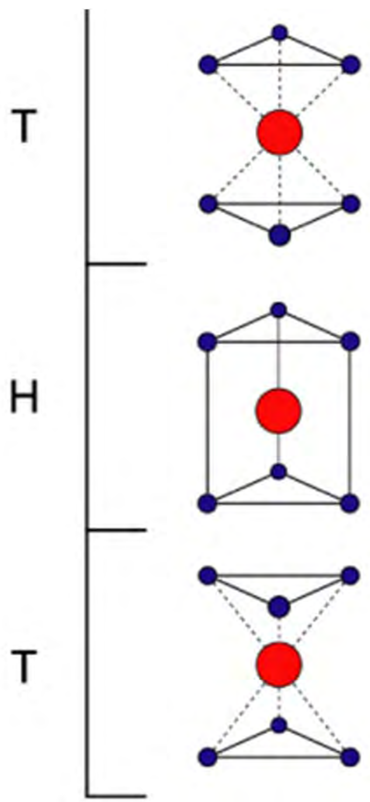


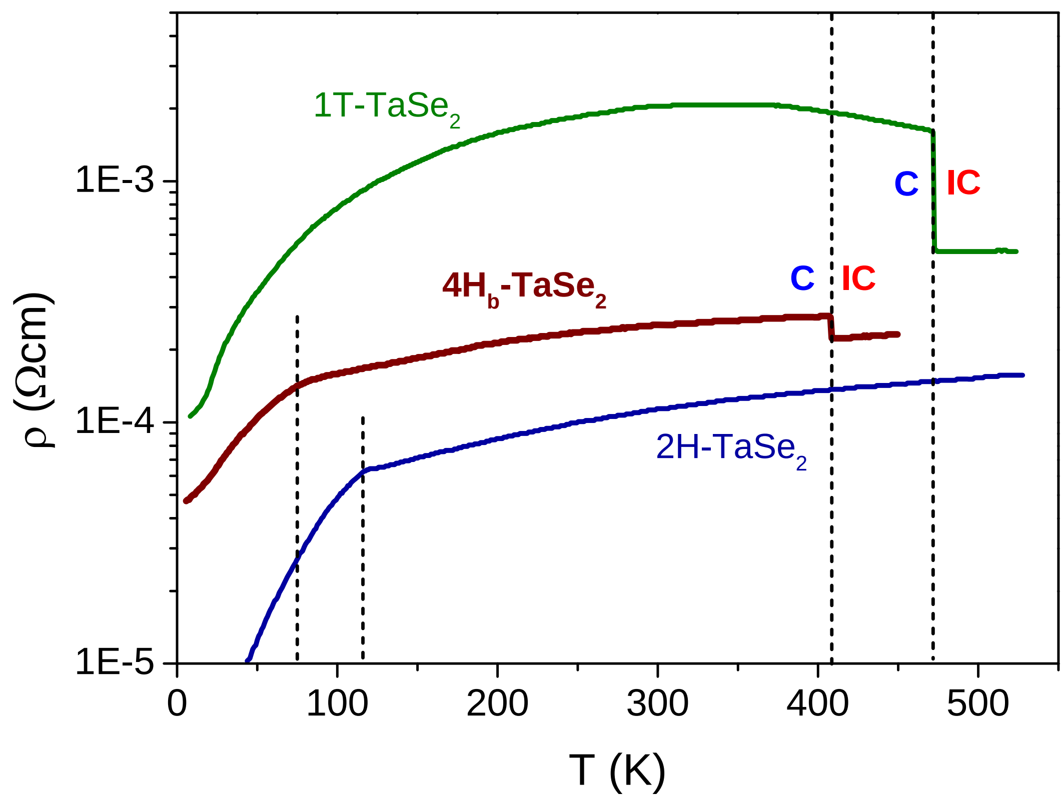
Disentangling electronic and thermal effects!

CDW extremely robust. Energy required to drive the NC-IC phase transition comparable to energy required to heat up the sample to T_c.

Charge Density Waves in 4Hb-$TaSe_2$

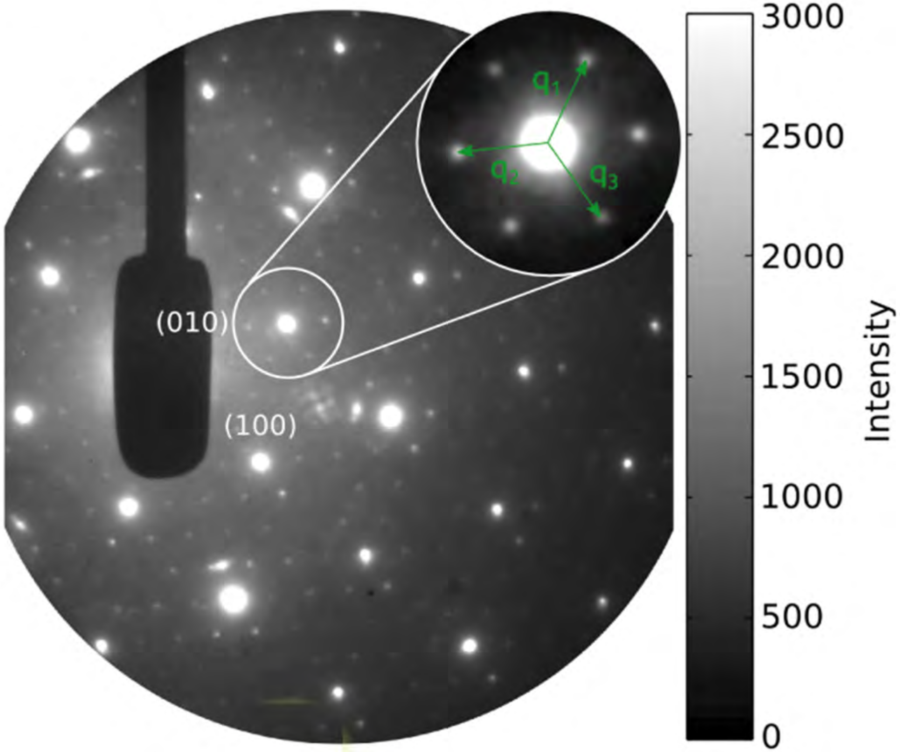
T (K)	CDW state	Layers involved	Modulation wave vector
>600	Normal		
410–600	Incommensurate	Octahedral	$\mathbf{q}^T = 0.265\mathbf{a}^*$
75–410	Commensurate	Octahedral	$q^T = 0.277a^*$; α, β superlattices
<75	Incommensurate	Trigonal prismatic	$\mathbf{q}^H = \mathbf{a}^*/3(1+\delta)$, at 10 K, $\delta = 0.04$
	Commensurate	Octahedral	$q^T = 0.277a^*$; α, β superlattices

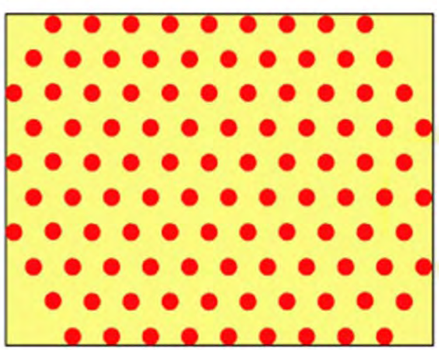


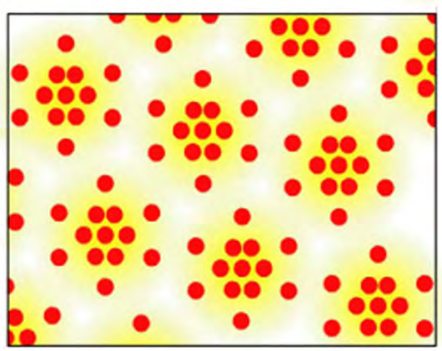


Di Salvo et al., PRB 14, 1543 (1976)

fs Electron Diffraction in 4Hb-$TaSe_2$

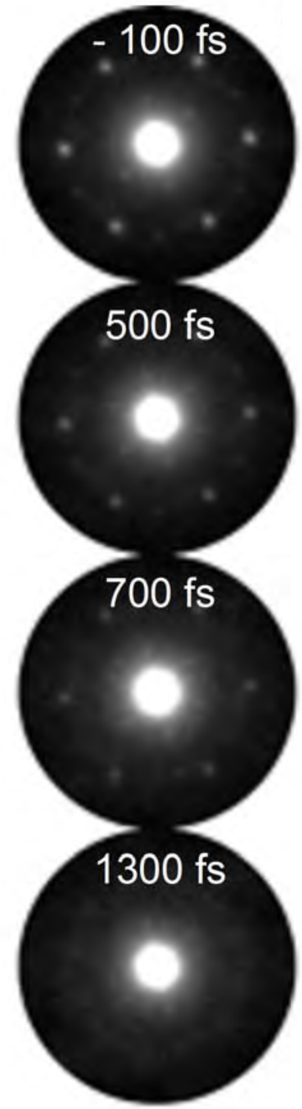






Commensurate CDW:

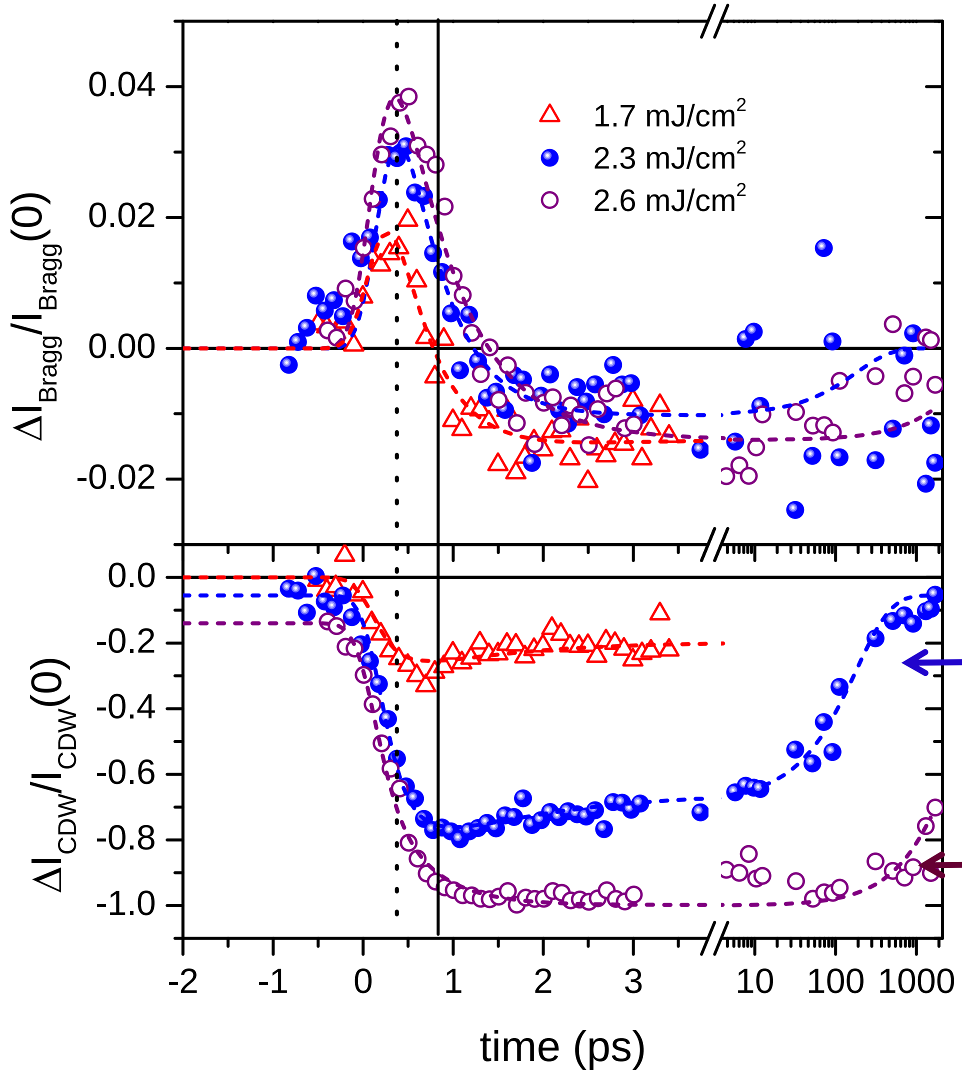
$$\left(\sqrt{13}\times\sqrt{13}\right)R13.9$$

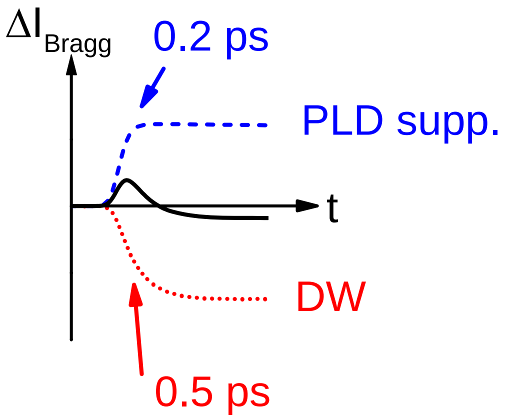


T= 300 K,
F = 2.5 mJ/cm^2

Erasmus et al., to appear in Phys. Rev. Lett. (2012).

Structural dynamics in 4Hb-$TaSe_2$



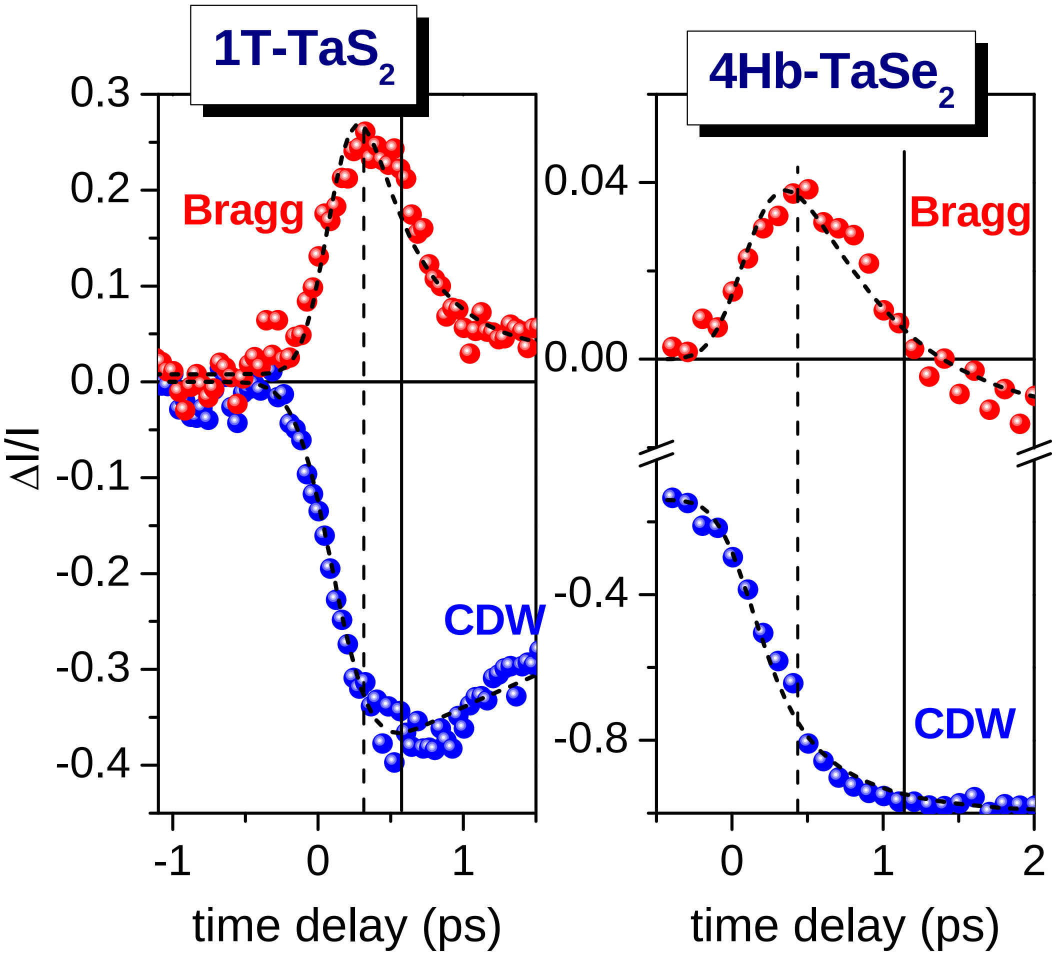


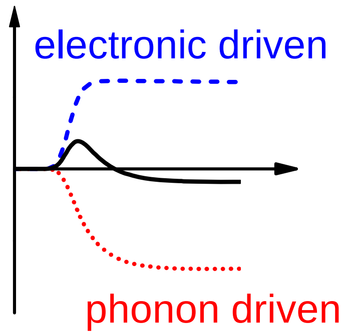
pre-phase transition regime:
CDW recovery time ~ 200 ps

Photoinduced C-IC phase transition:
recovery via heat diffusion

Erasmus et al., to appear in Phys. Rev. Lett. (2012).

Transient CDW melting in 1T-TaS_2 and 4Hb-$TaSe_2$





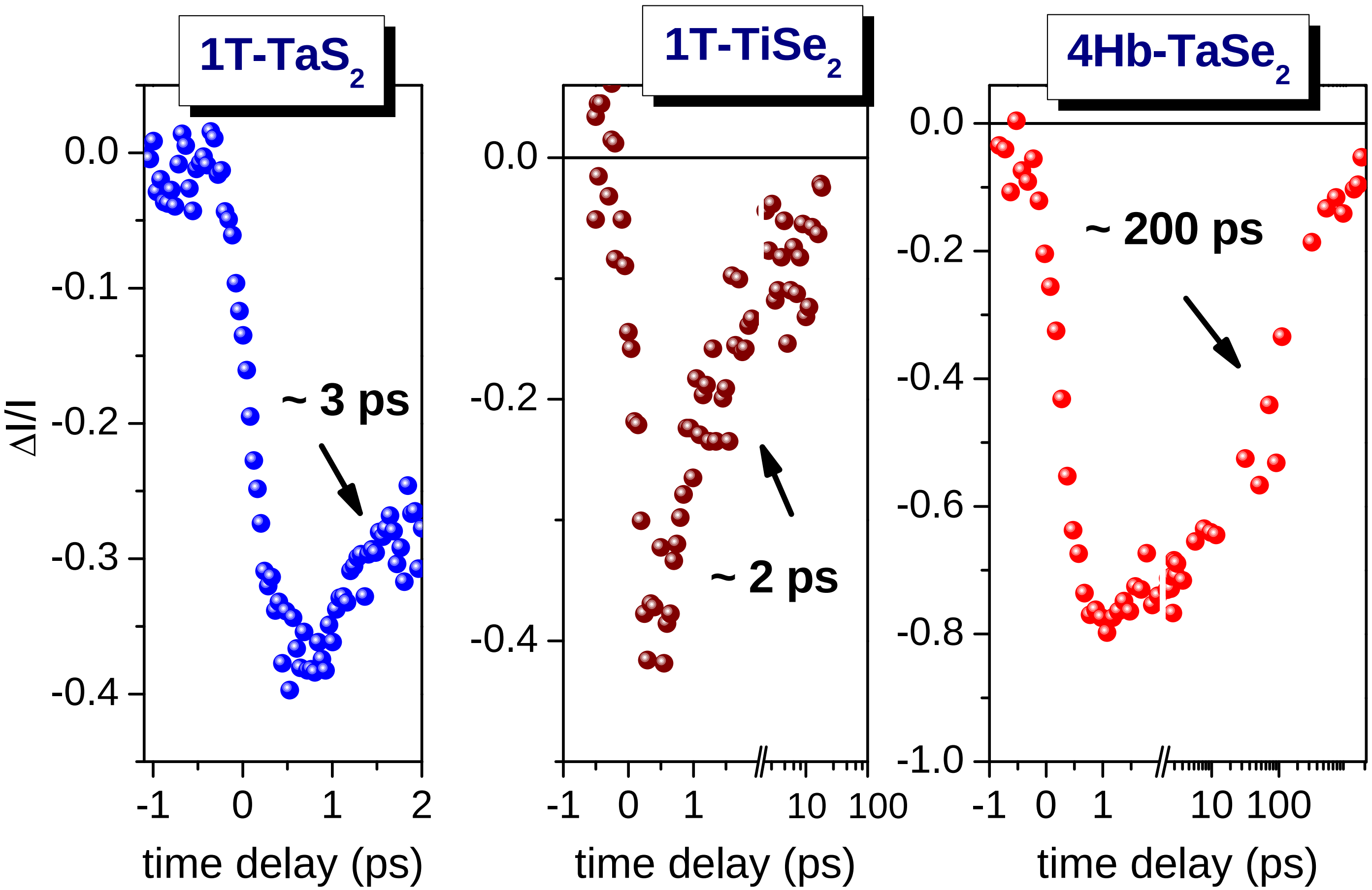
PI CDW melting proceeds via a two step process:

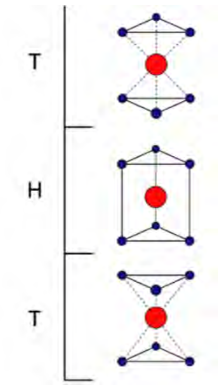
Step 1: electronically launched, "coherent" motion (τ < AM cycle)

Step 2: further destabilization by phonons

Energy required to drive the phase transition comparable to thermal energy!

Timescales of CDW build-up vs. interplane coupling





- Picosecond recovery times found in all CDW systems studied thus far:
$K_{0.3}MoO_3$, $TbTe_3$, $DyTe_3$, 2H-$TaSe_2$, o-TaS_3, $NbSe_3$ (tr-optics, tr-ARPES)

CDW recovery in 4Hb-$TaSe_2$ two orders of magnitude slower – weak interlayer coupling!

Summary

Dynamics on elementary time and atomic scale:

- new insights into the ground state of novel quantum materials!
- coupling strengths (hierarchy) between different degrees of freedom

Density Waves:

- Disentanglement of the electronic and lattice parts of the order parameter
  PRL 102, 066404 (2009), Nature 468, 799 (2010).
- The nature of collective modes PRL 105, 066402 (2010)
- Peierls vs. excitonic CDW mechanism in $TiSe_2$ PRL107, 036403 (2011)
- Strong phonon-spin coupling in ferropnictides Nature Phys. (2012)
- Dichalcogenides: importance od interlayer coupling for existence of CDW!
  to appear in PRL (2012)