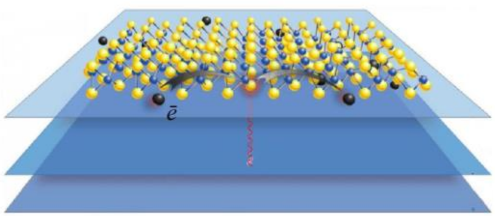
Ultrafast dimerization melting in the Peierls-Mott insulator 1T-TaS_2

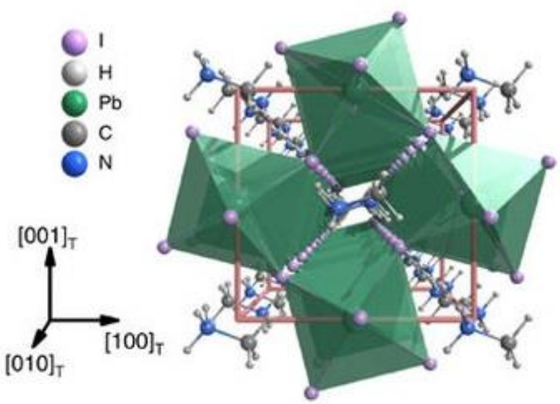
Luca Perfetti *Ecole Polytechnique*

2D Electrons Gases



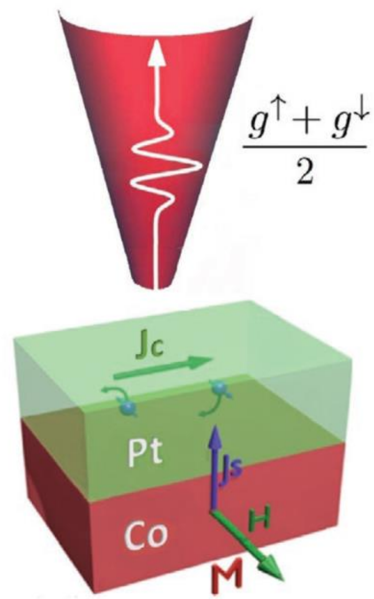
*Z. Chen et al., **PNAS 117,** 21962 (2020)*

Hybride Pérovskites



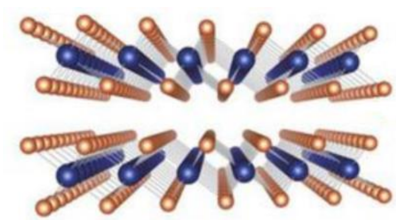
*E. Jung et al., **ACS energy Lett.*** 5, *785 (2020)*

Spintronic Interfaces



J. Hawecker, ***Adv. Opt. Mater.*** 9, *2100412 (2021)*

Correlated materials



*N. Nilforoushan, **PNAS** 8, e2108617118 (2021)*

Collaborations

Transition Metal dichalcogenide

1T - TaS_2

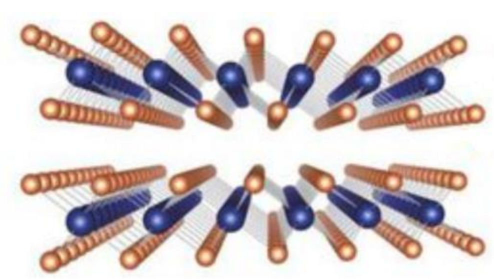
Jingwei Dong
Evangelous Papalazarou
Romain Grasset

Ernest Pastor

Angel Rubio
Dongbin Shin

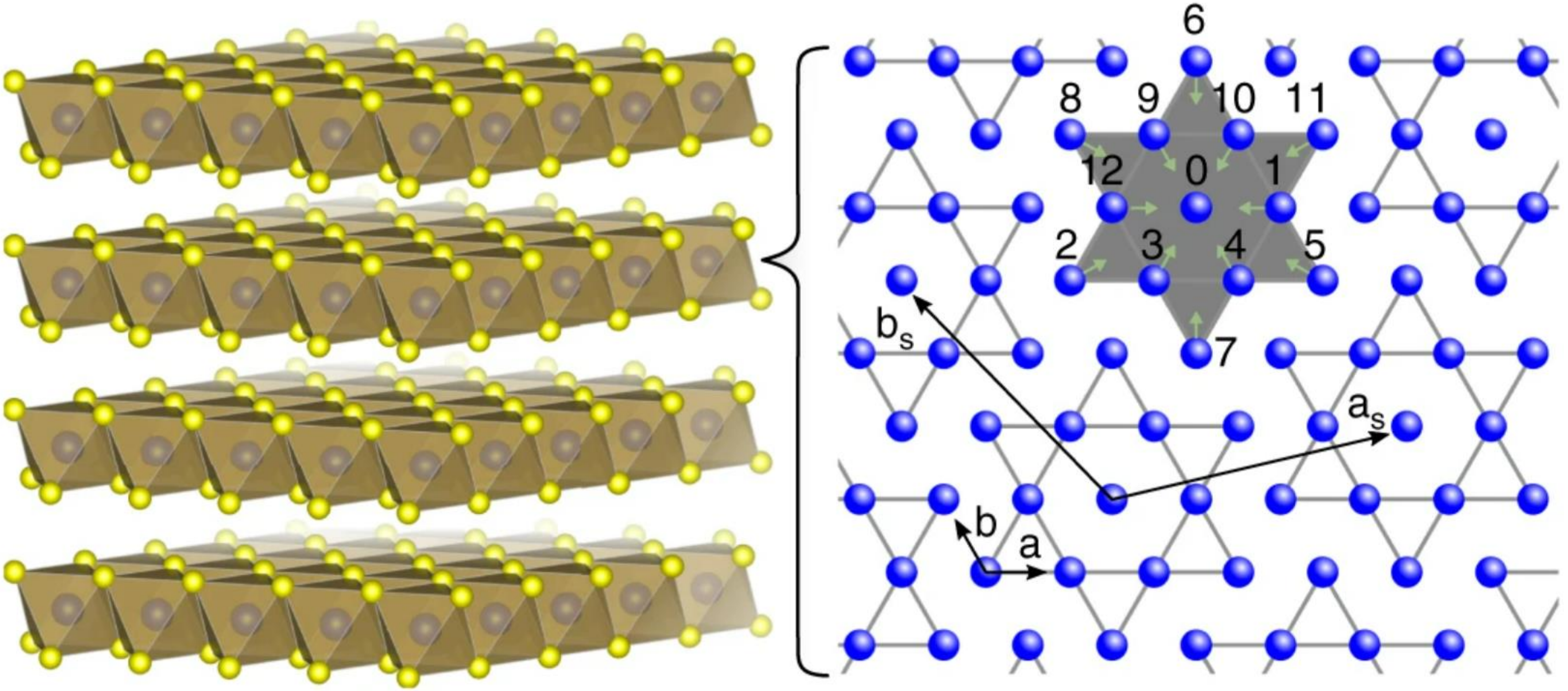
Laurent Cario

Tobias Ritschel



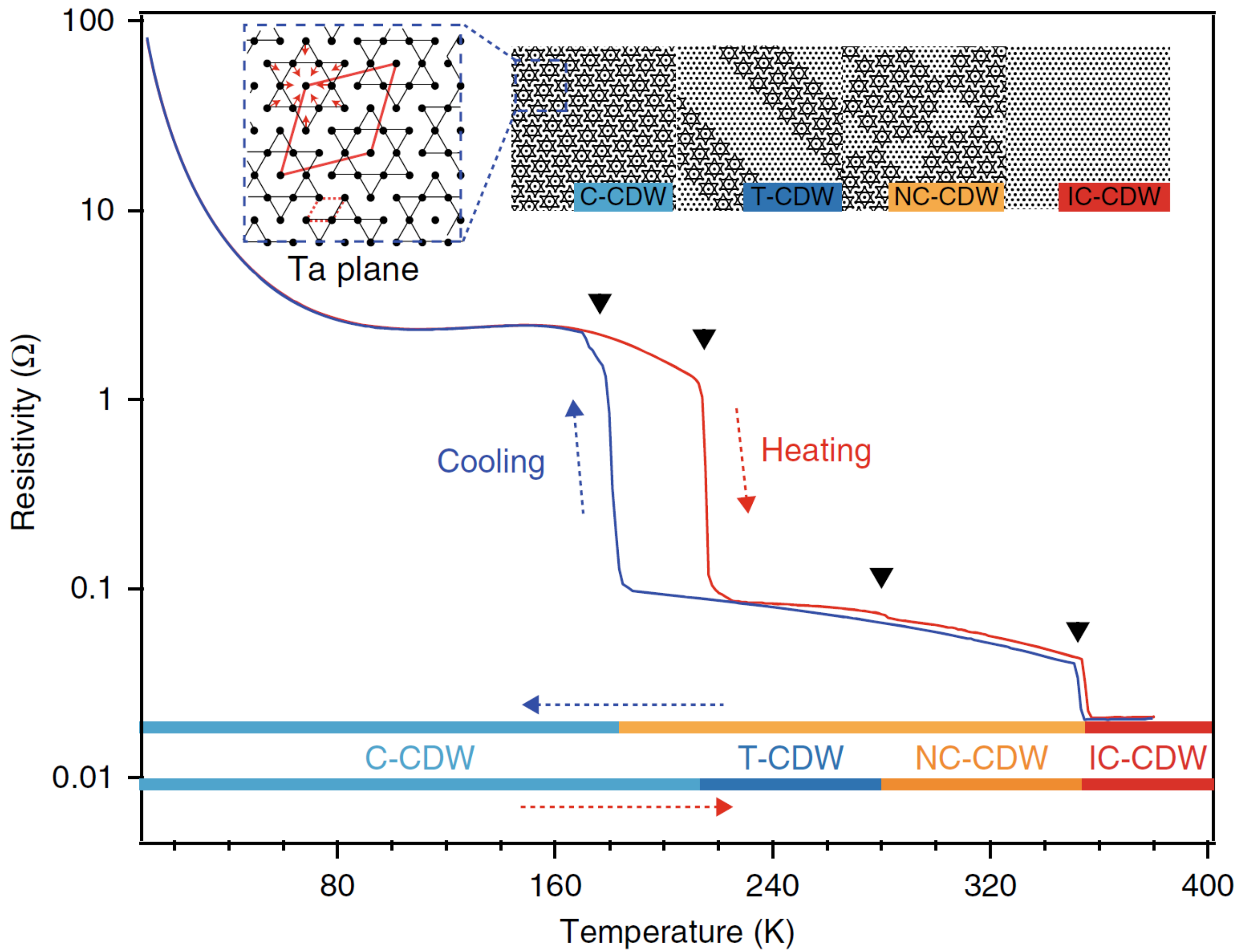
Structure

Trigonal antiprismatic structure

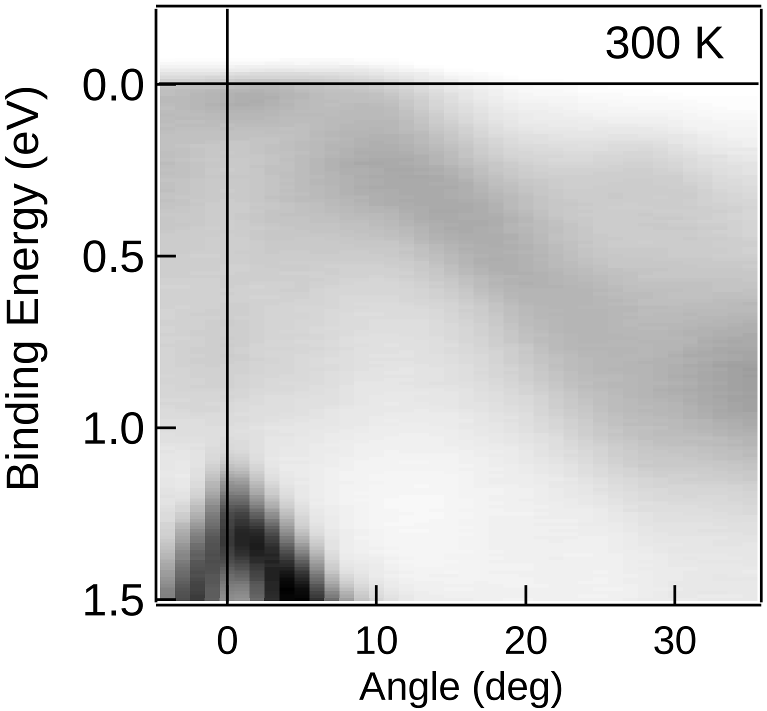


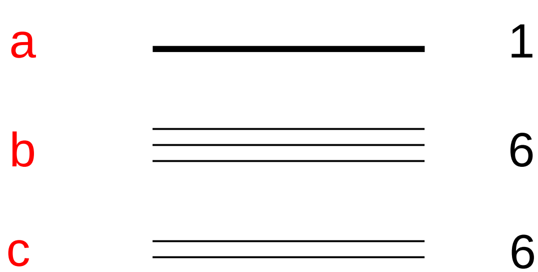
Large CDW distortion already at room temperature

Phase transition



CDW and Electronic states

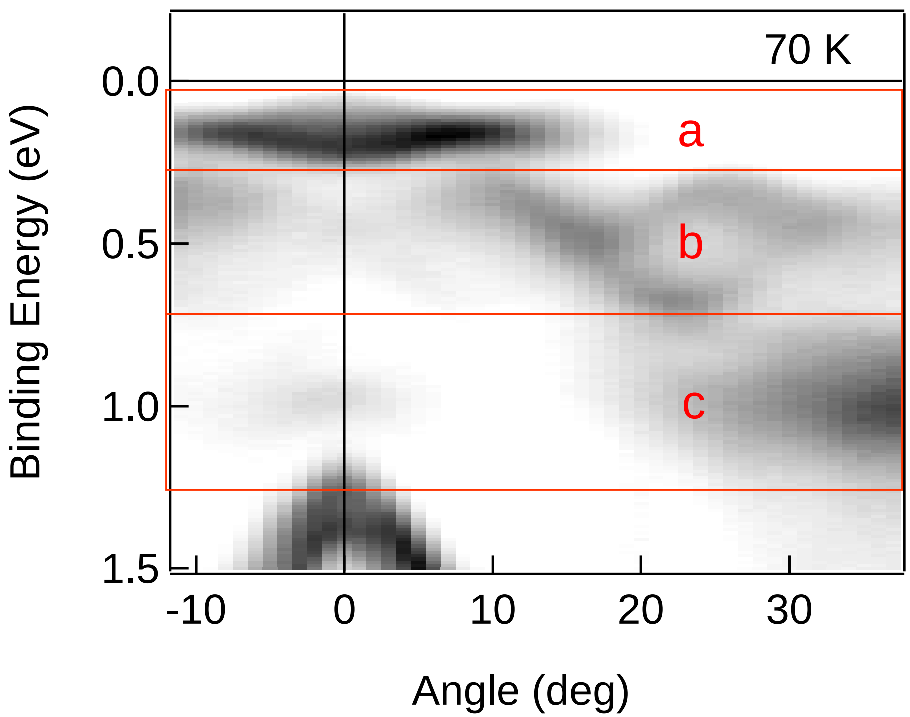




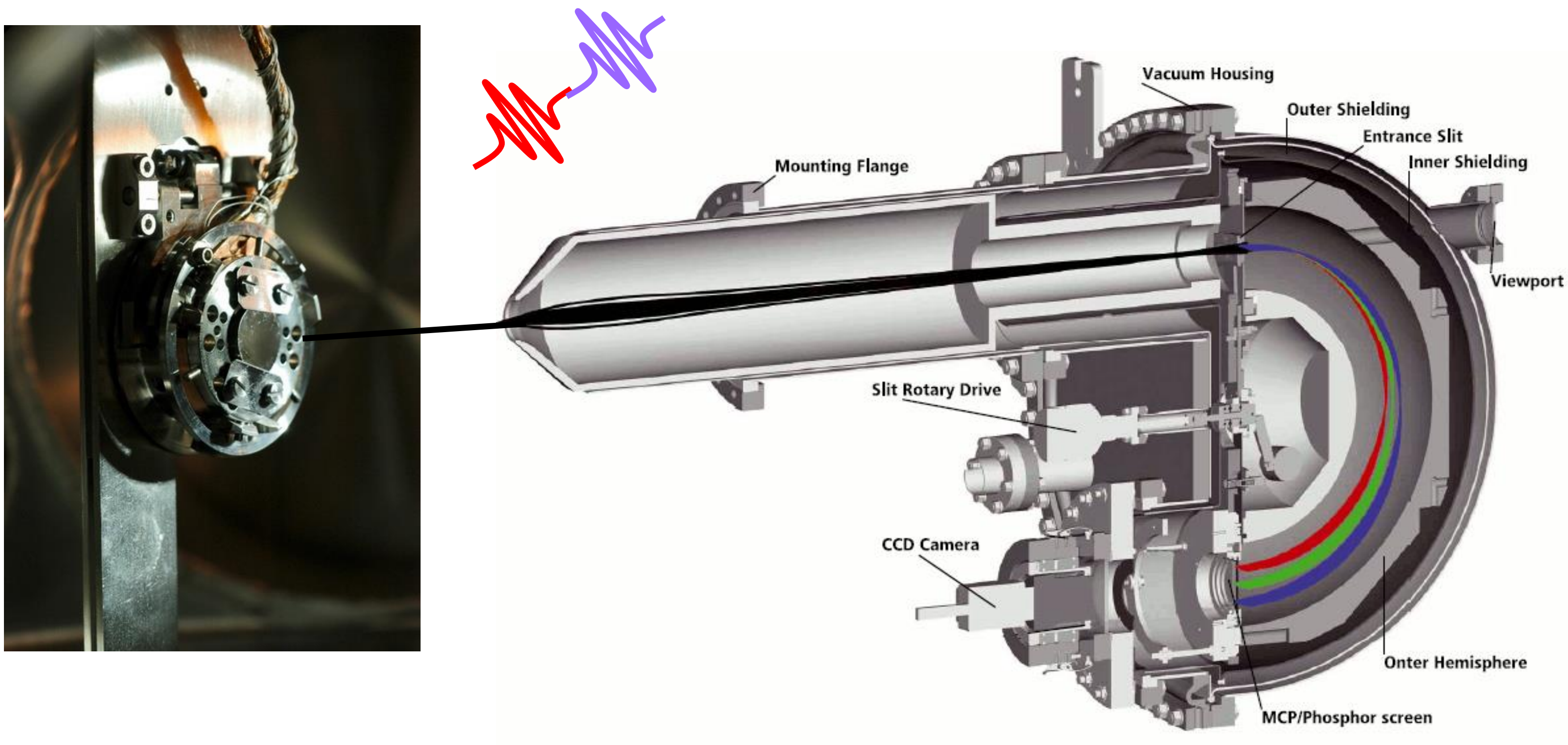
3 many-folds induced by the CDW.

Gapped groundstate

L. Perfetti et al., ***Phys. Rev. B*** *71, 153101 (2005)*

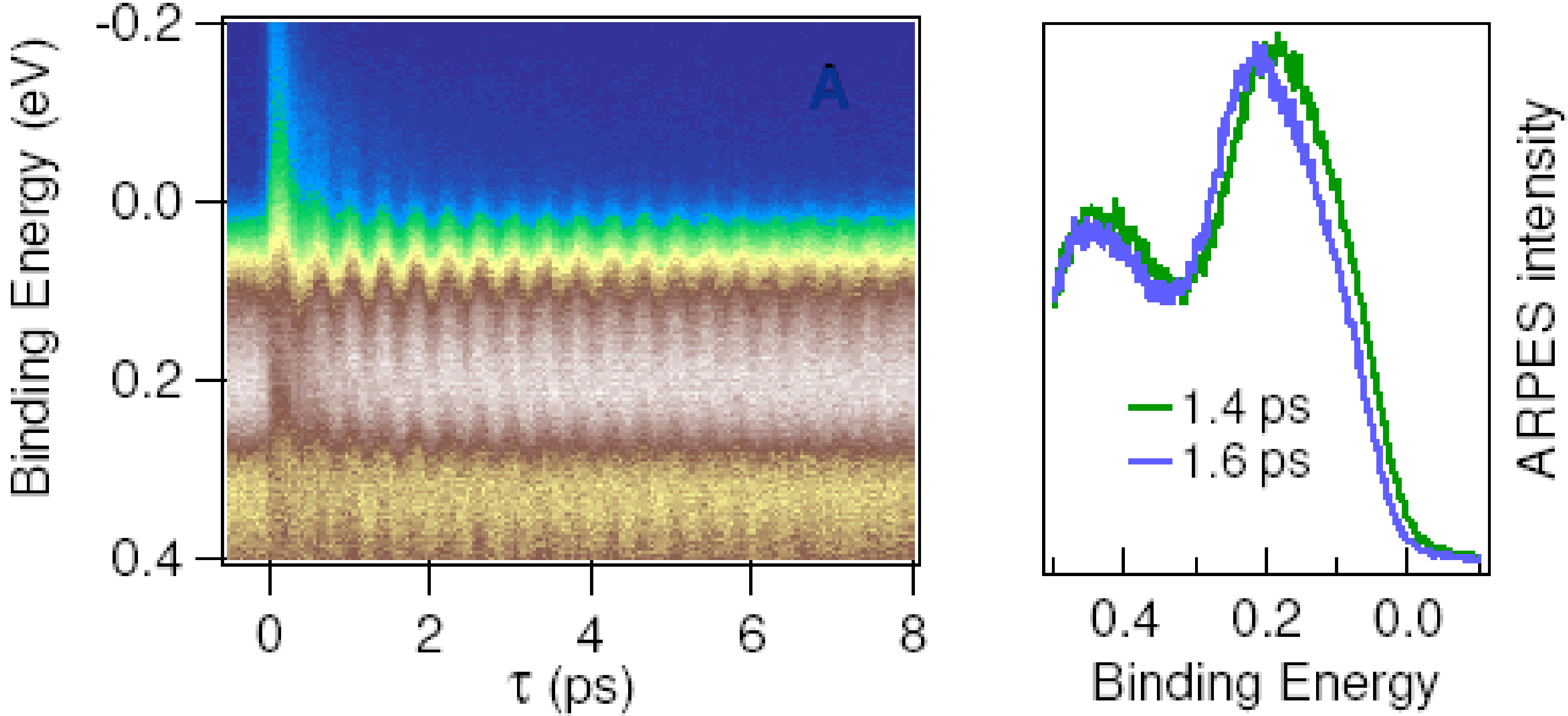


Time and Angle resolved ARPES



Coherent Oscillations of CDW amplitude

L. Perfetti et al., ***Phy Rev Lett.*** *97, 067402 (2006)*



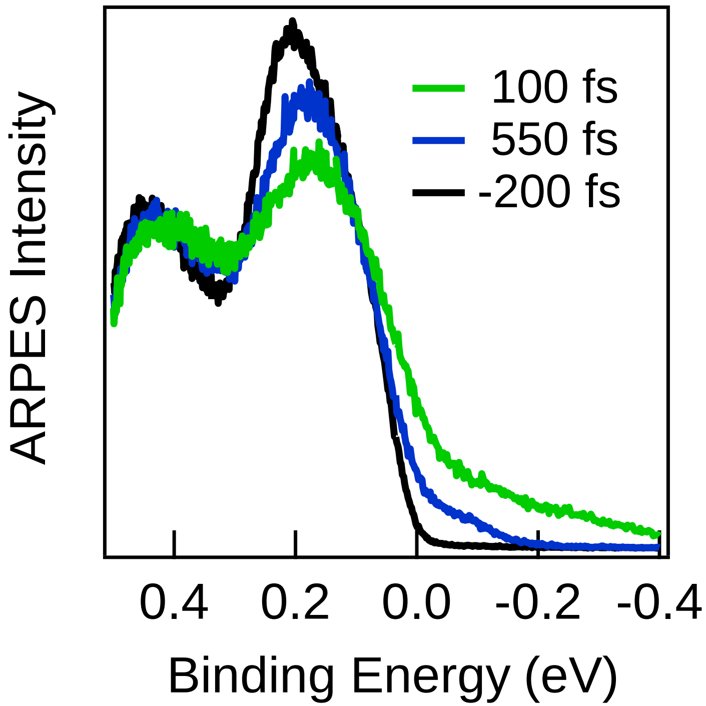
Large oscillations lasting longer than 10 ps

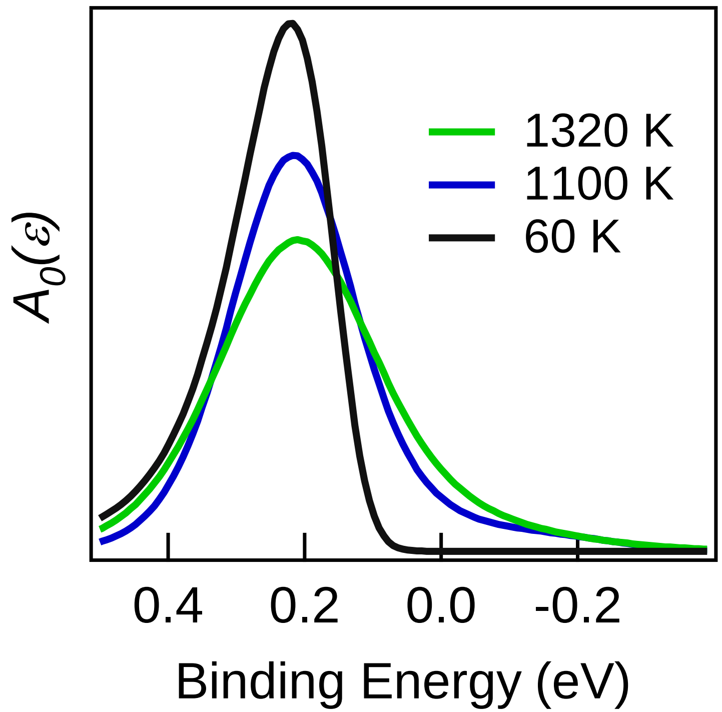
Rigid spectral shift of 18 meV

C-CDW $T_l = 30$ K

Electronic Gap Melting

Large electronic temperature near half filling



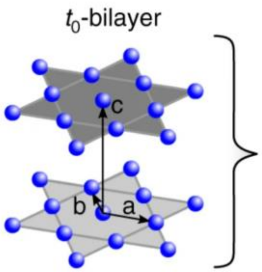


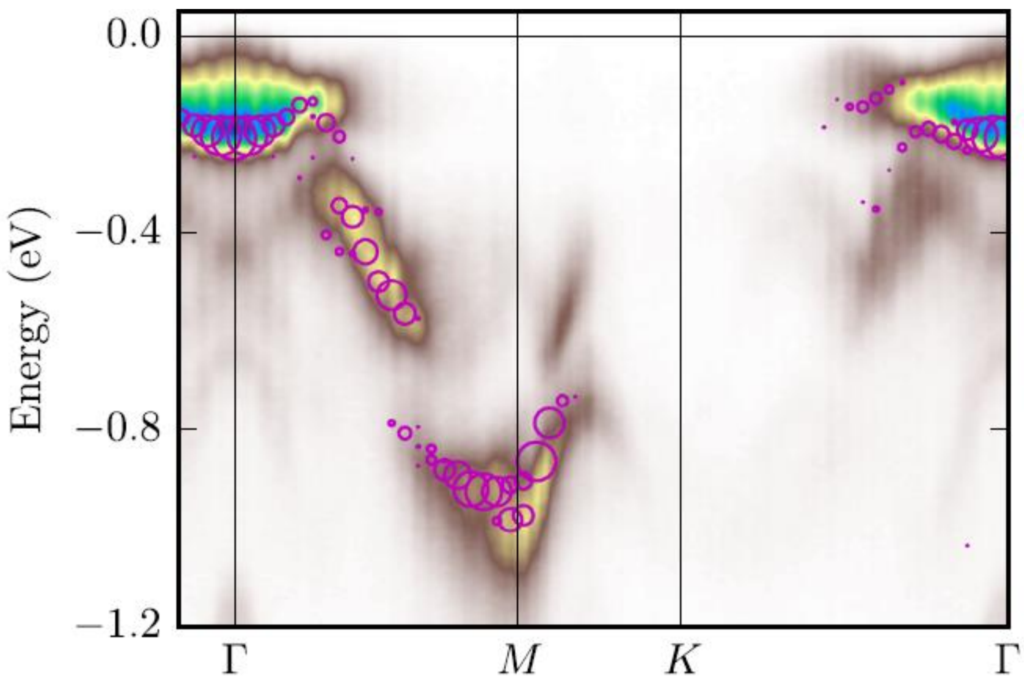
Dynamical mean field theory

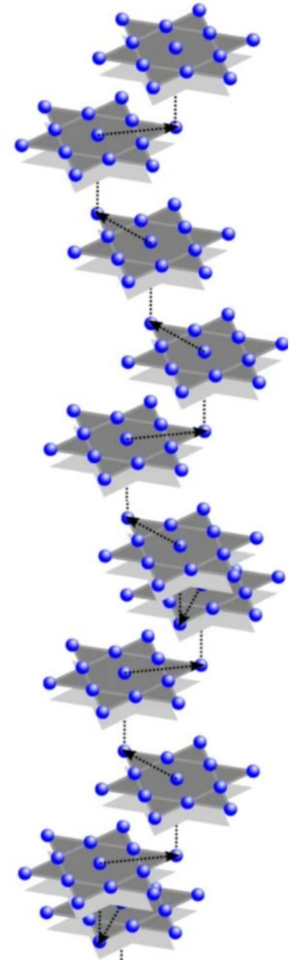
S. Birmann and A. Georges

Stacking Order and ARPES

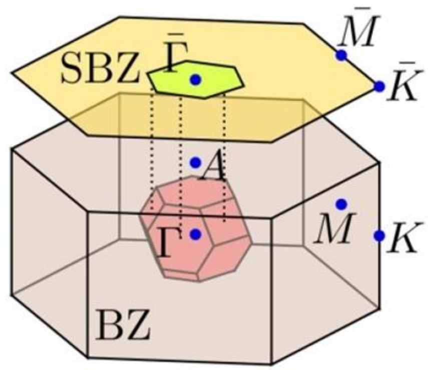
T. Ritschel et al., ***Nature Phys.*** *11, 328 (2015)*

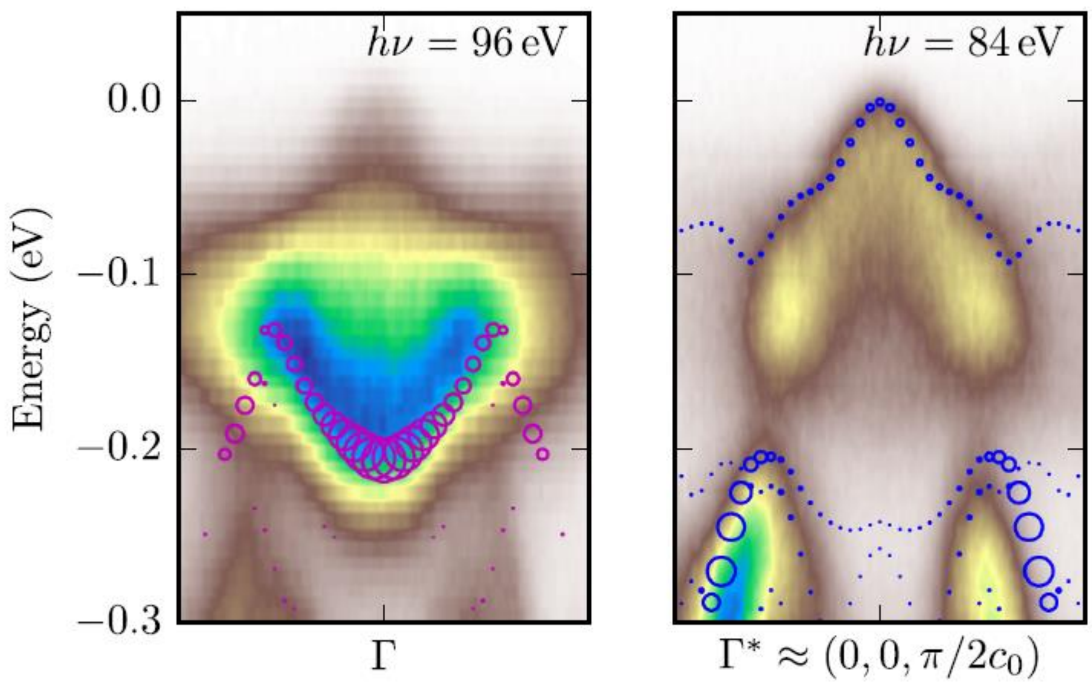


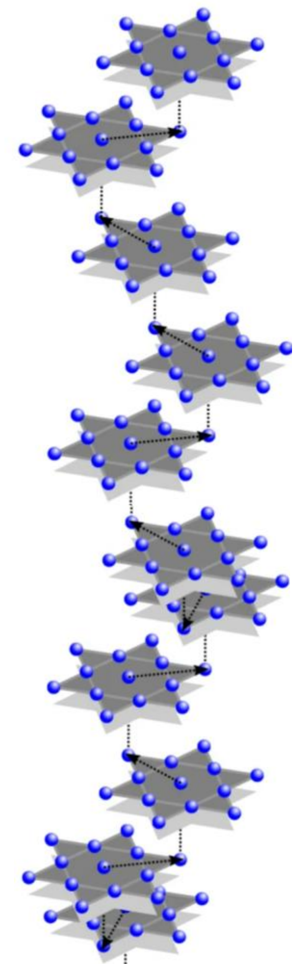




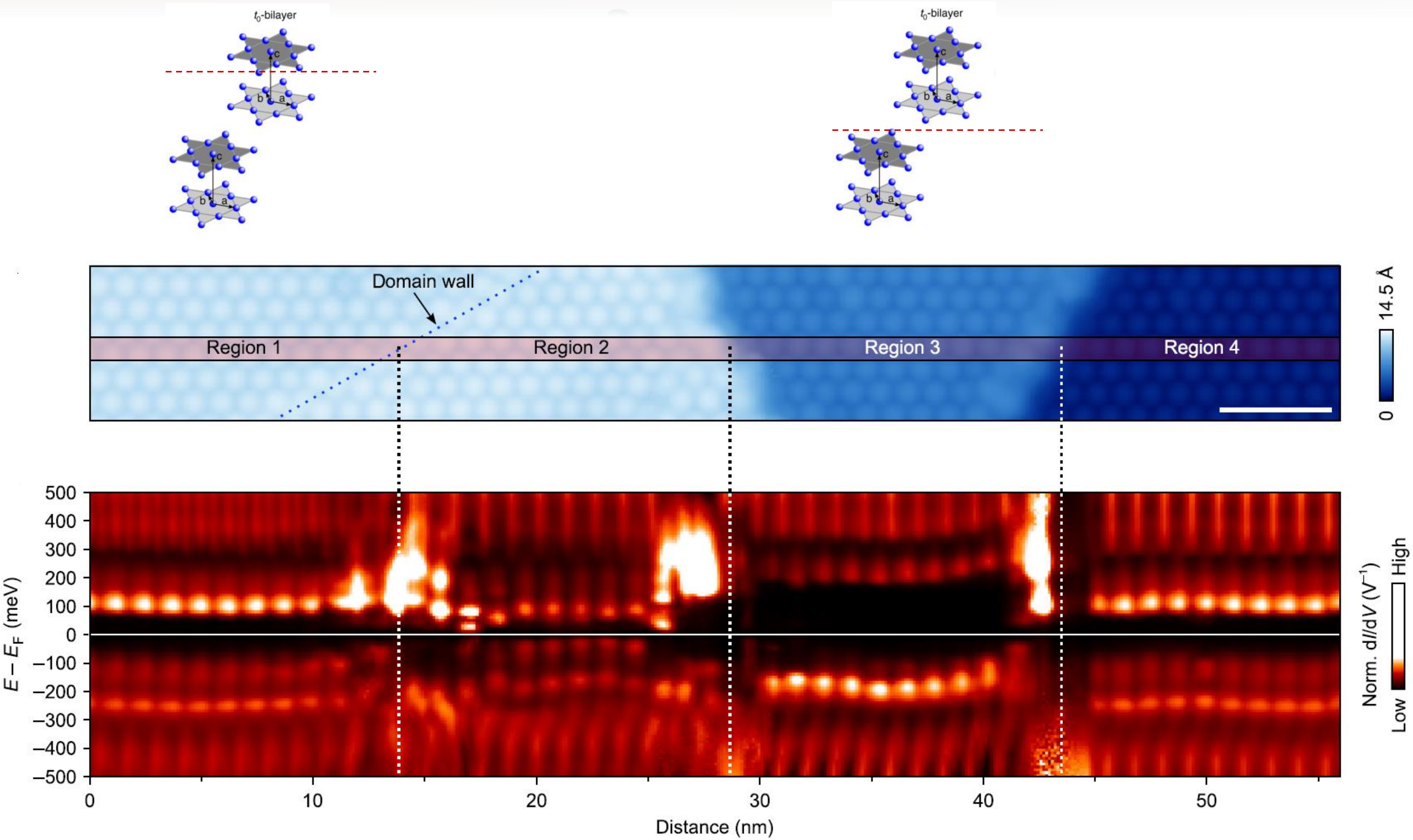
Stacking Order and ARPES







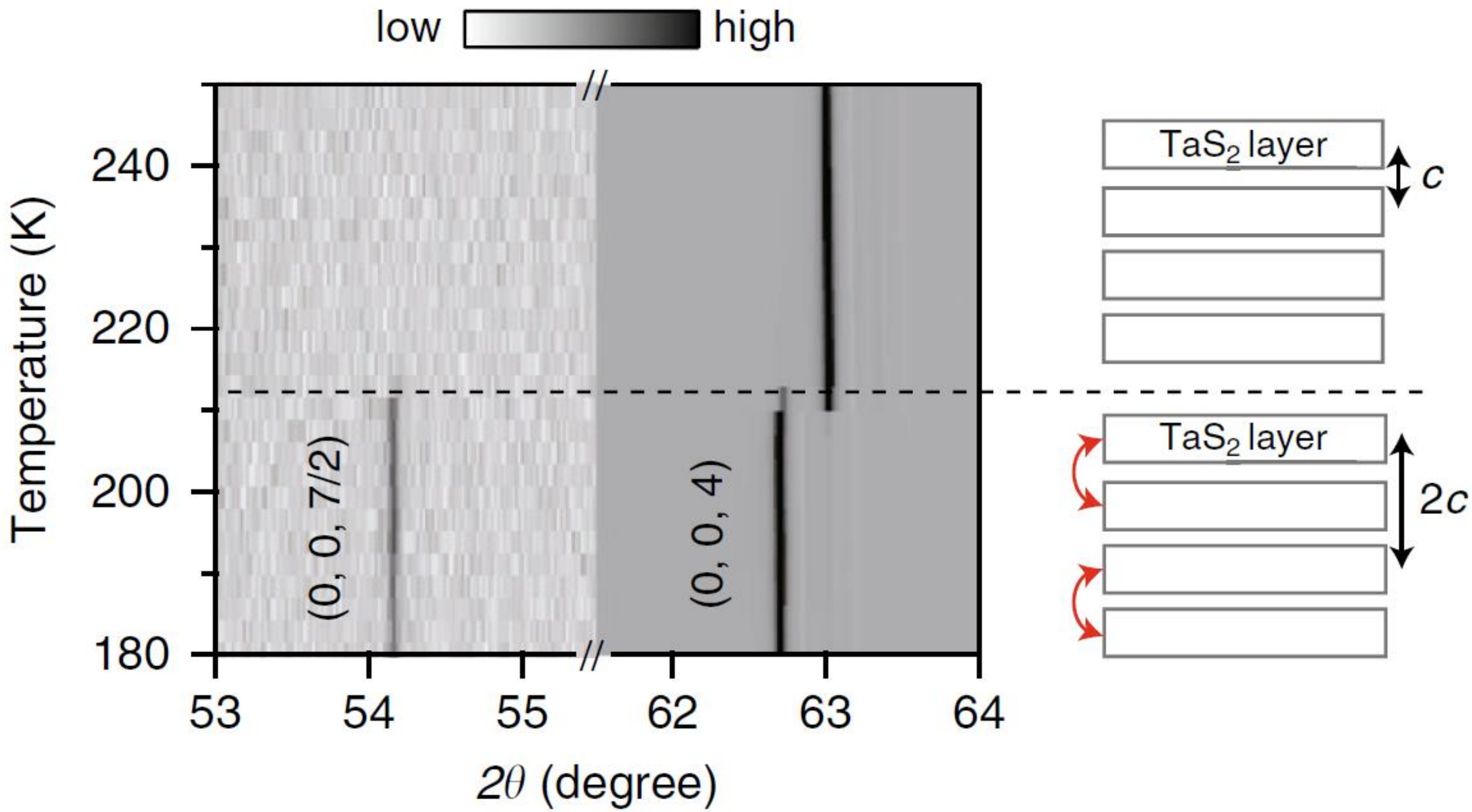
Stacking Order and STM



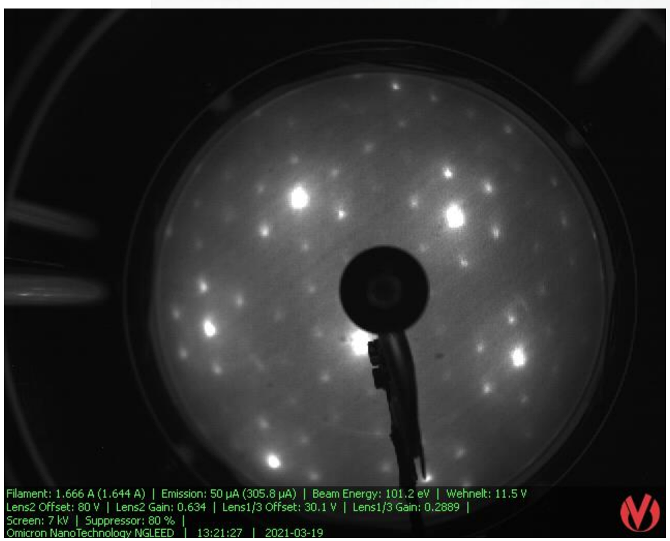
C. J. Butler et al., ***Nature Comm.*** *11, 2477 (2020)*

Stacking Order and XRD

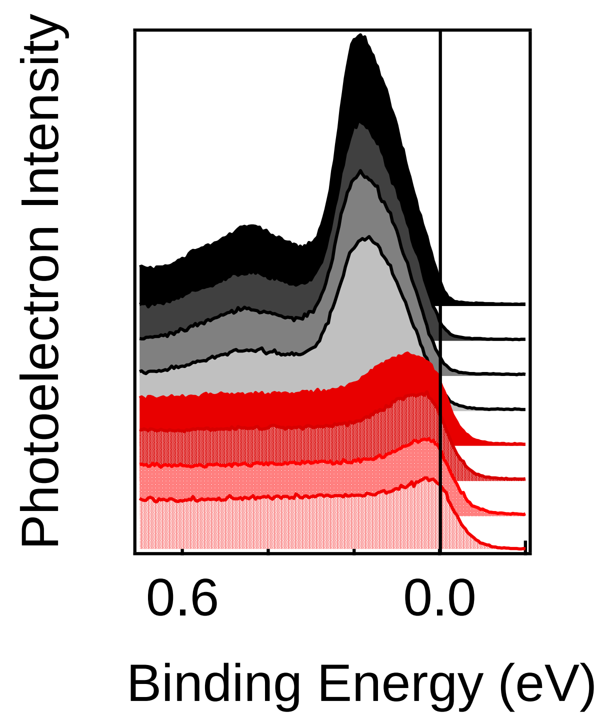
Y. D. Wang *et al.*, ***Nature Comm.* 11**, 4215 (2020)



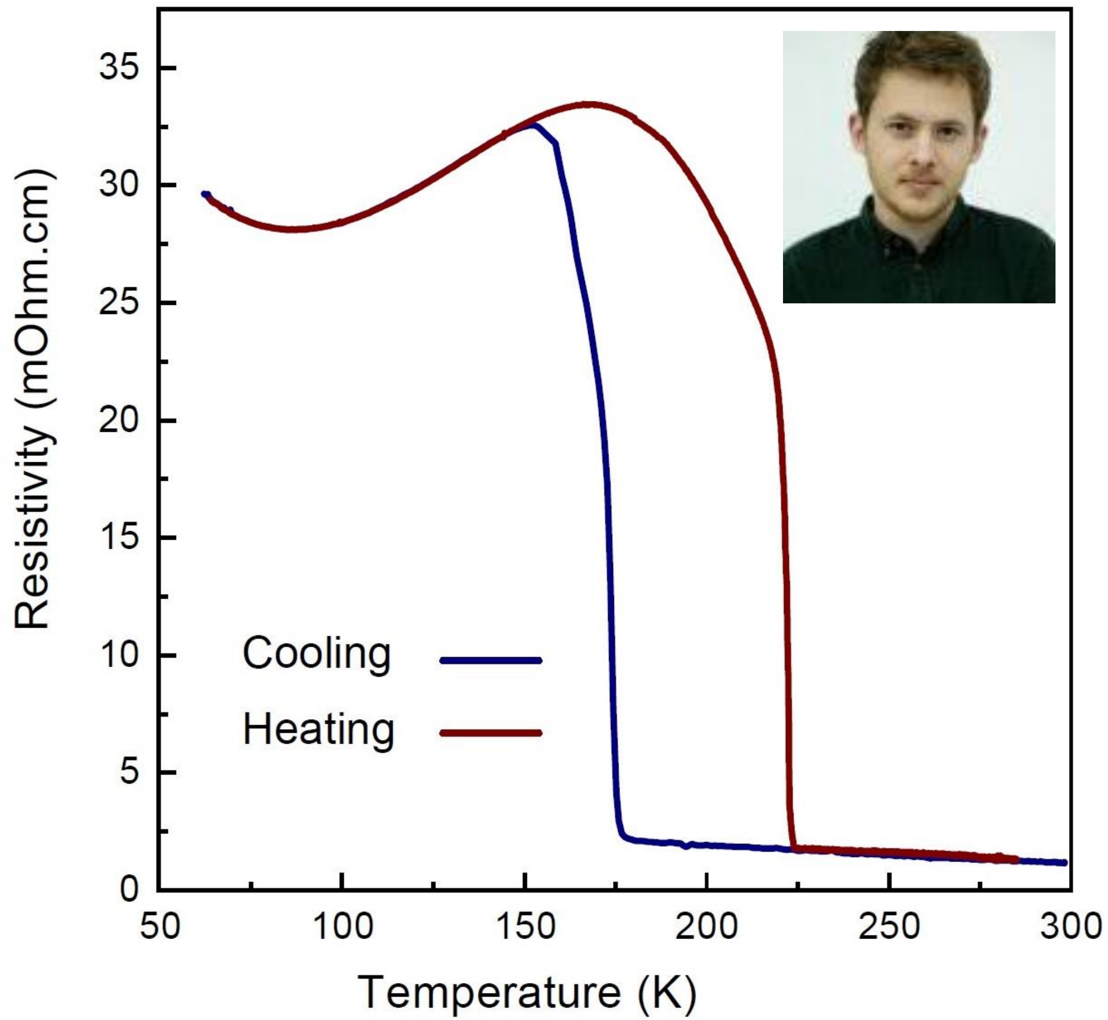
High Quality samples



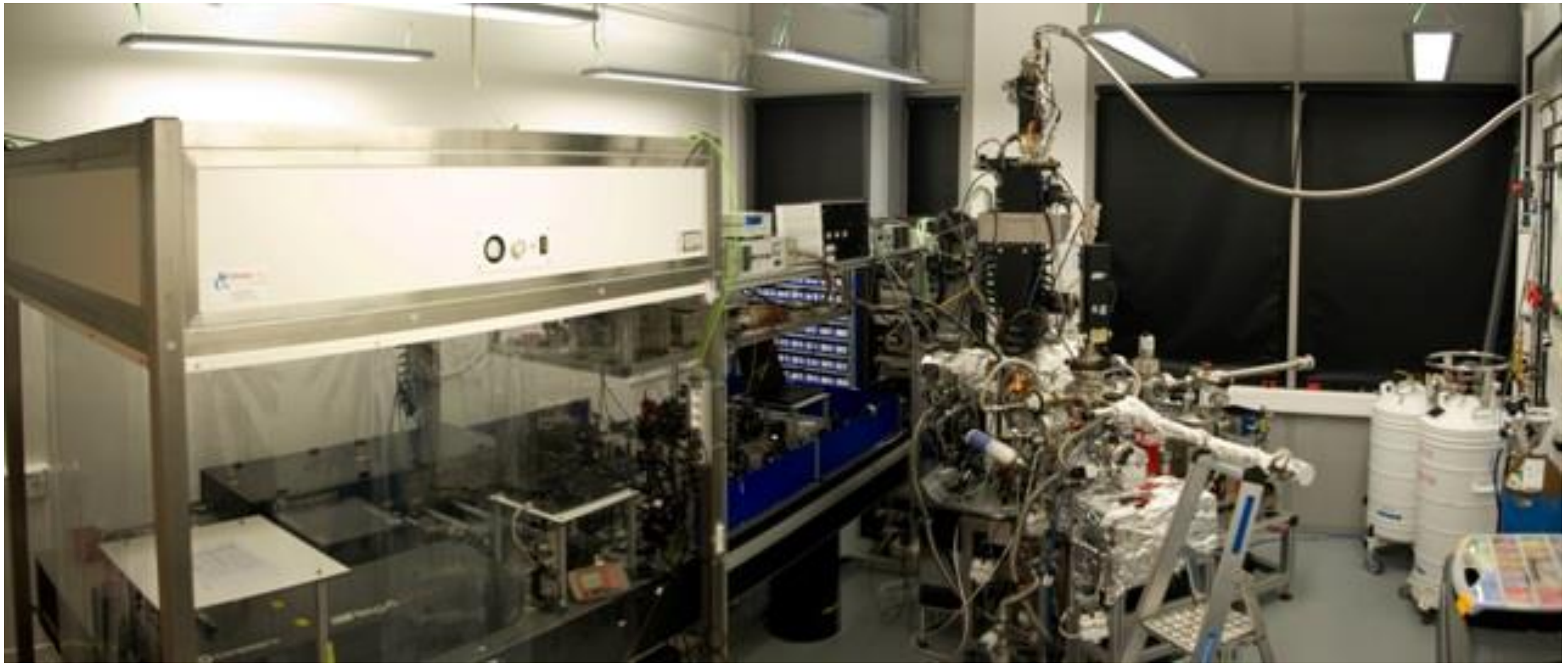
Classes for M2 students

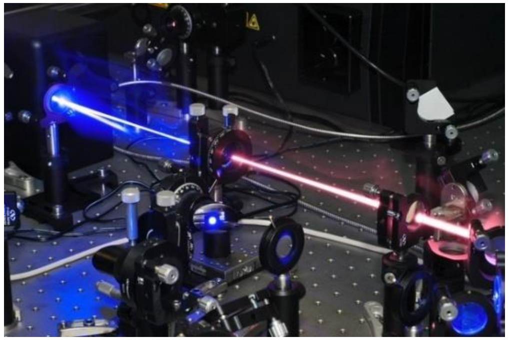


Resistivity



FemtoARPES laboratory





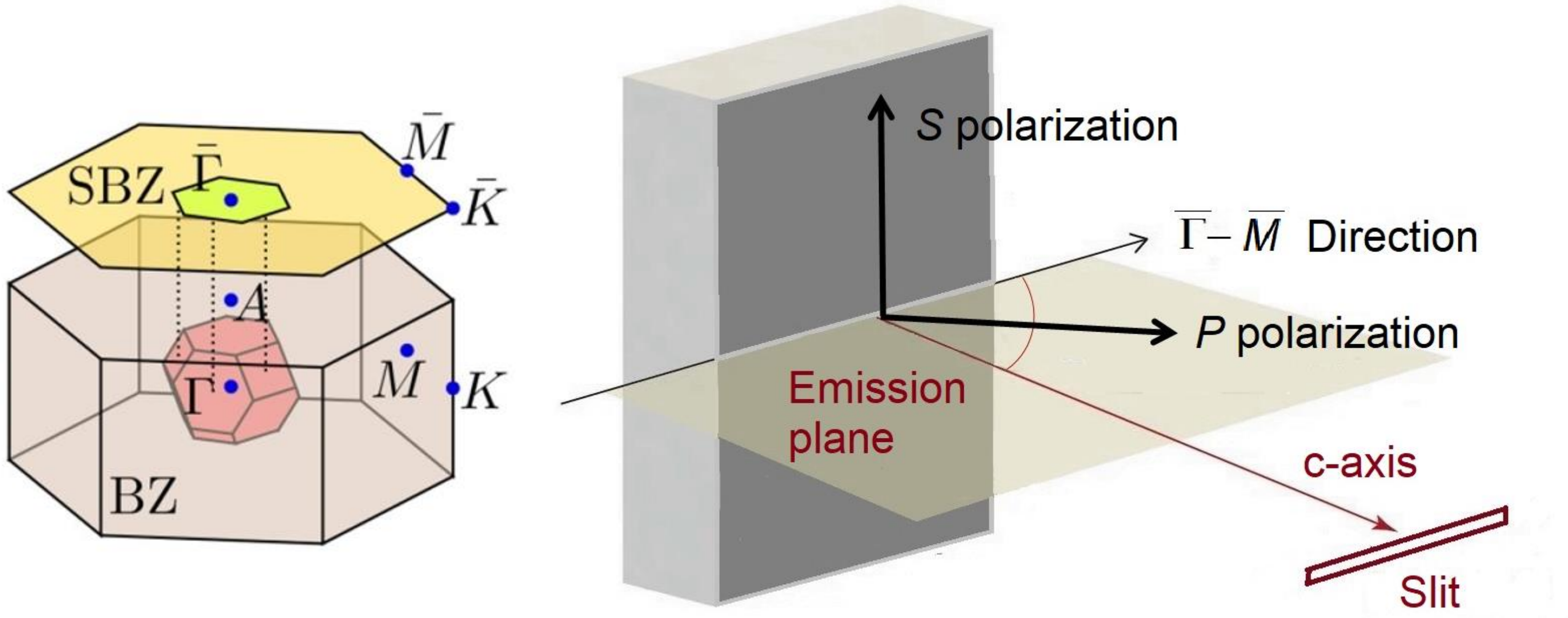
ω = 1.55 eV

4ω = 6.3 eV

Cross correlation

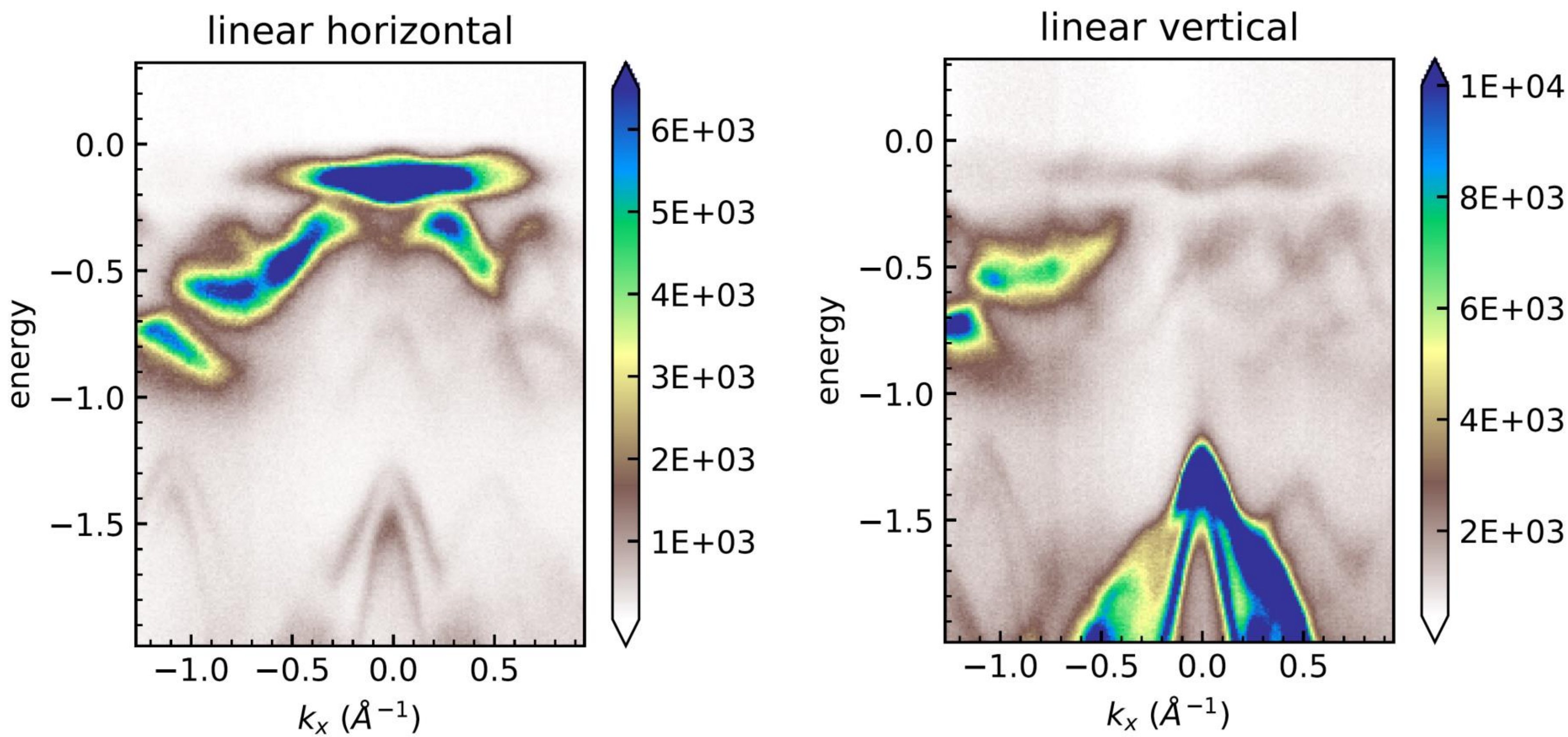
100 fs

Polarization Geometry

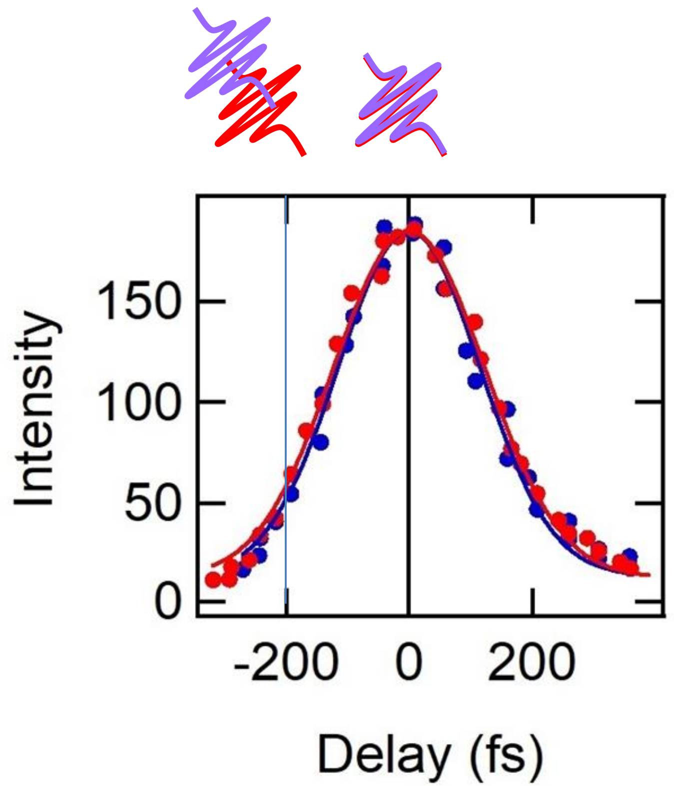


Polarization at Synchrotron

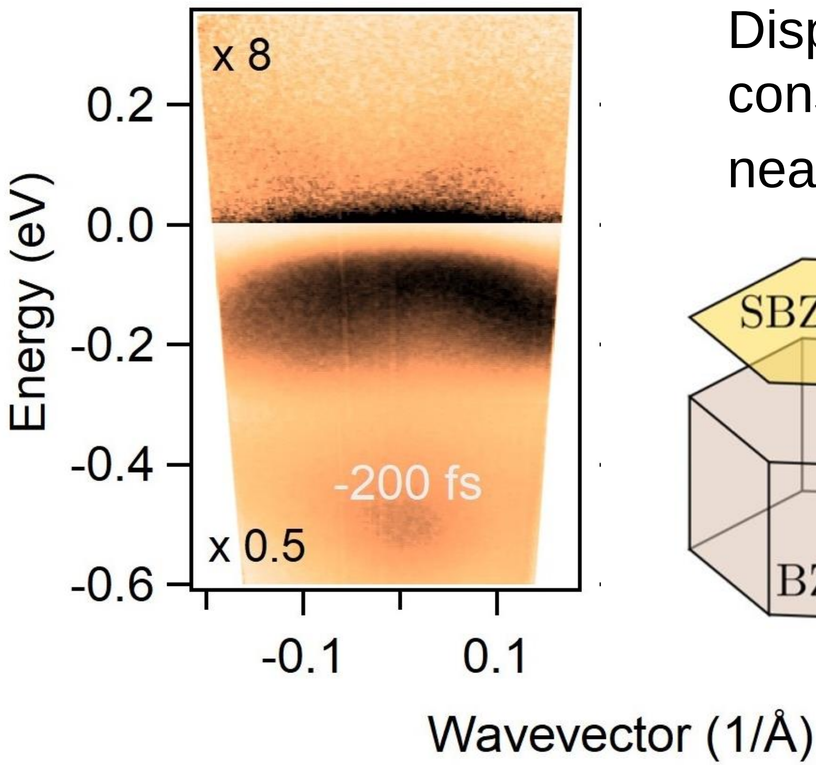
Tobias Ritschel



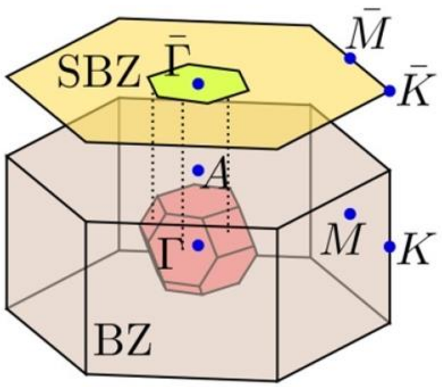
Time resolved ARPES



P Polarization

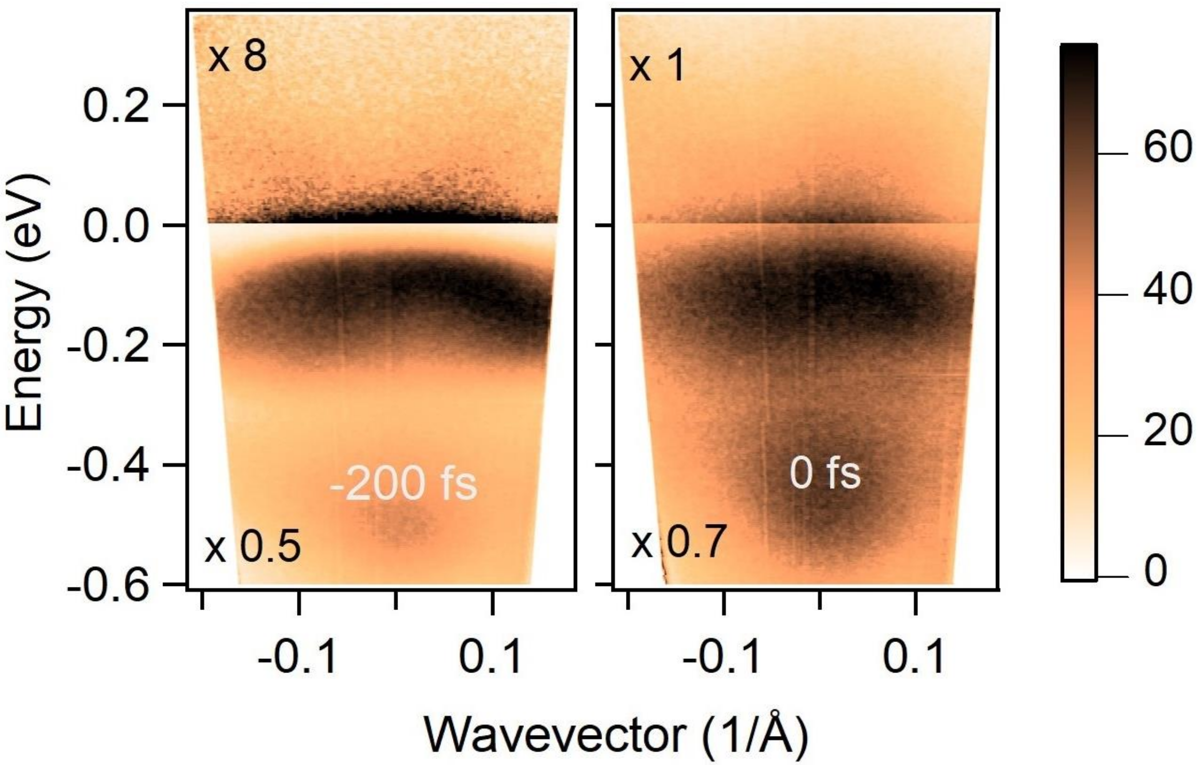


Dispersion consistent with k_z near to $\pi/2c_0$



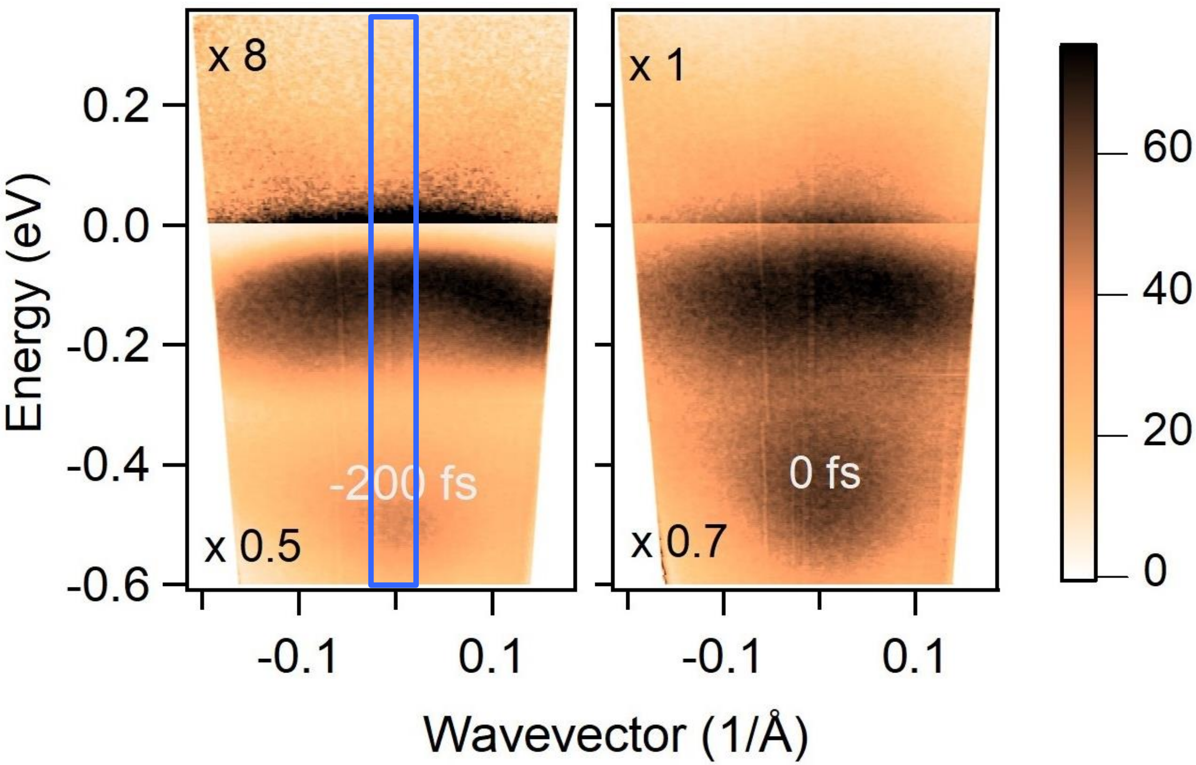
P Polarization

Structureless background fills the gap

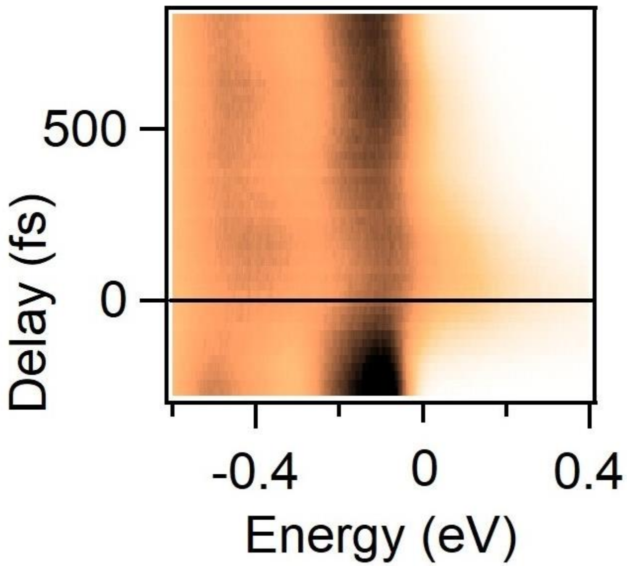


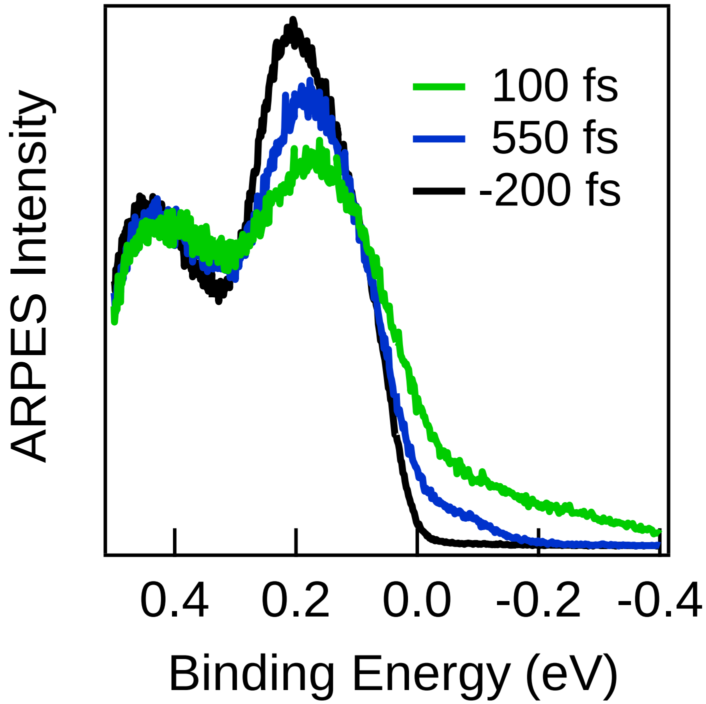
P Polarization

Structureless background fills the gap



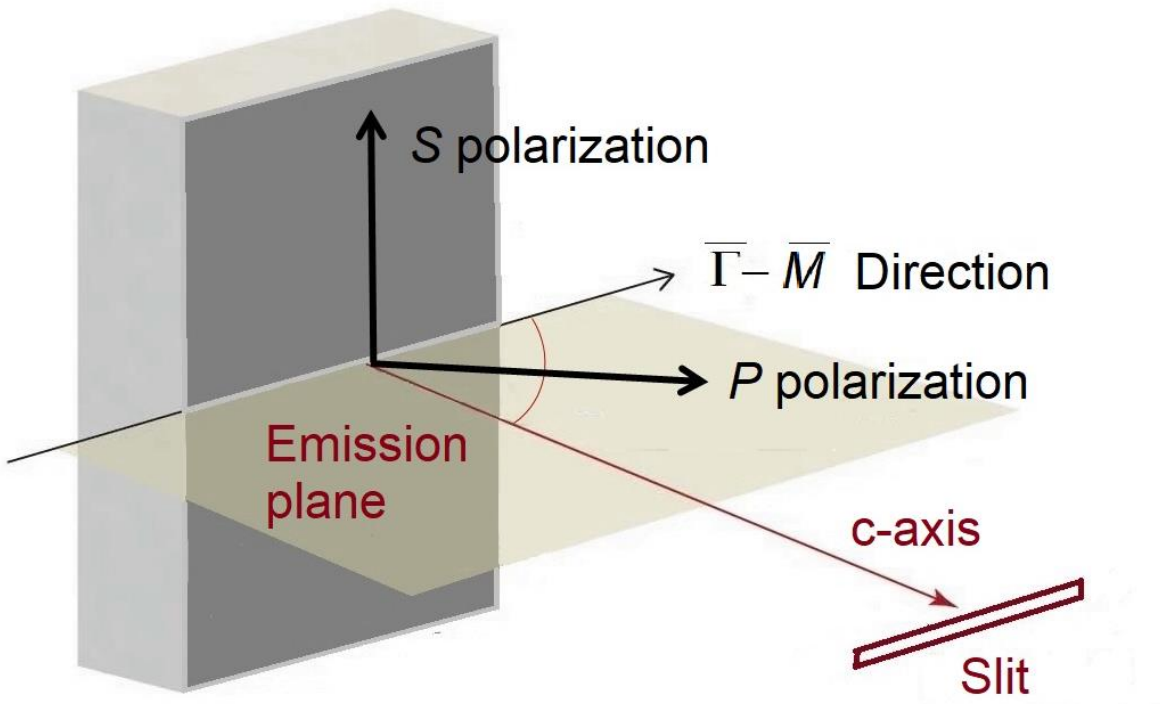
P Polarization





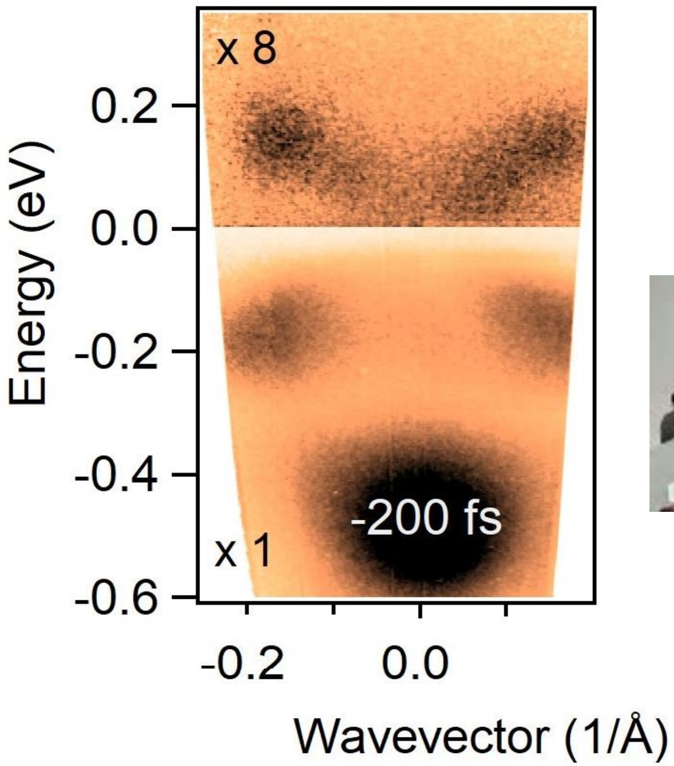
L. Perfetti et al., ***Phy Rev Lett.*** *97, 067402 (2006)*

Polarization Geometry



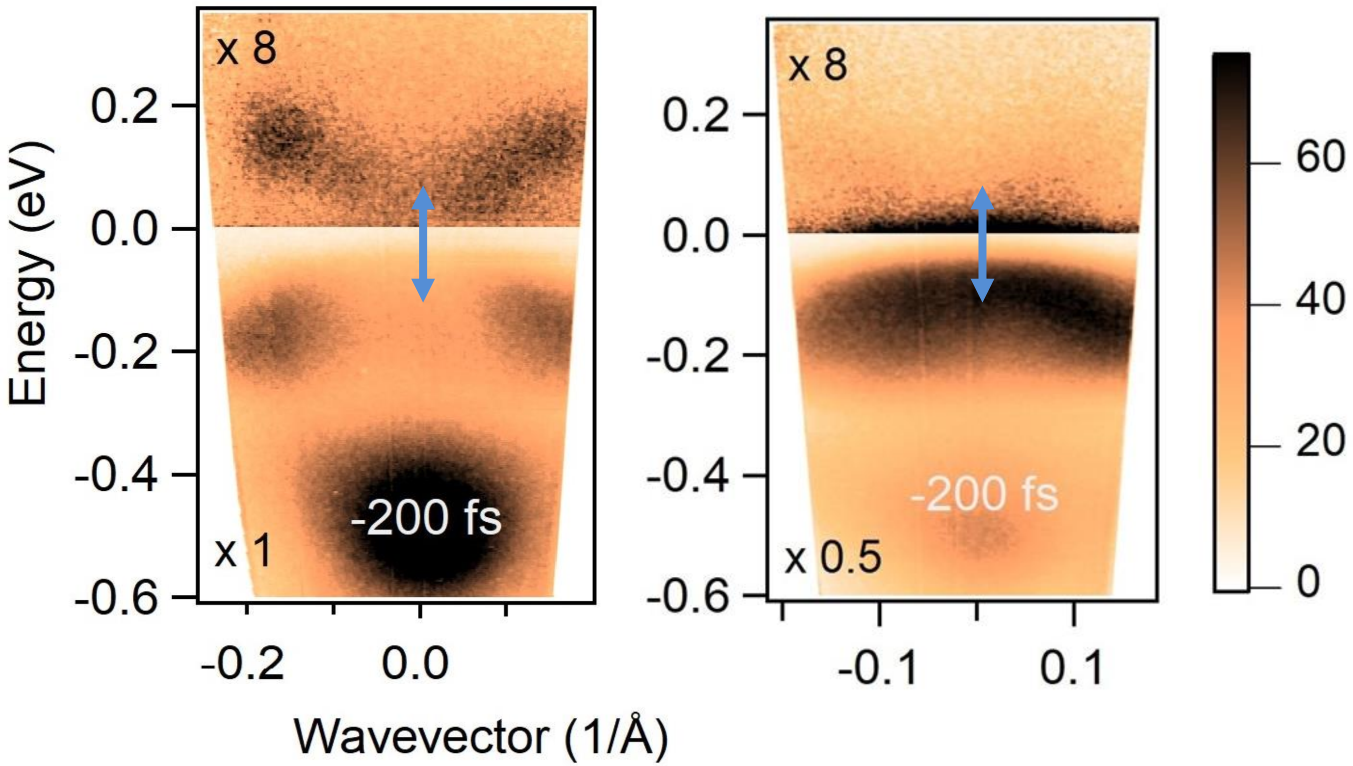
S Polarization

Dispersion of the states above μ



S Polarization

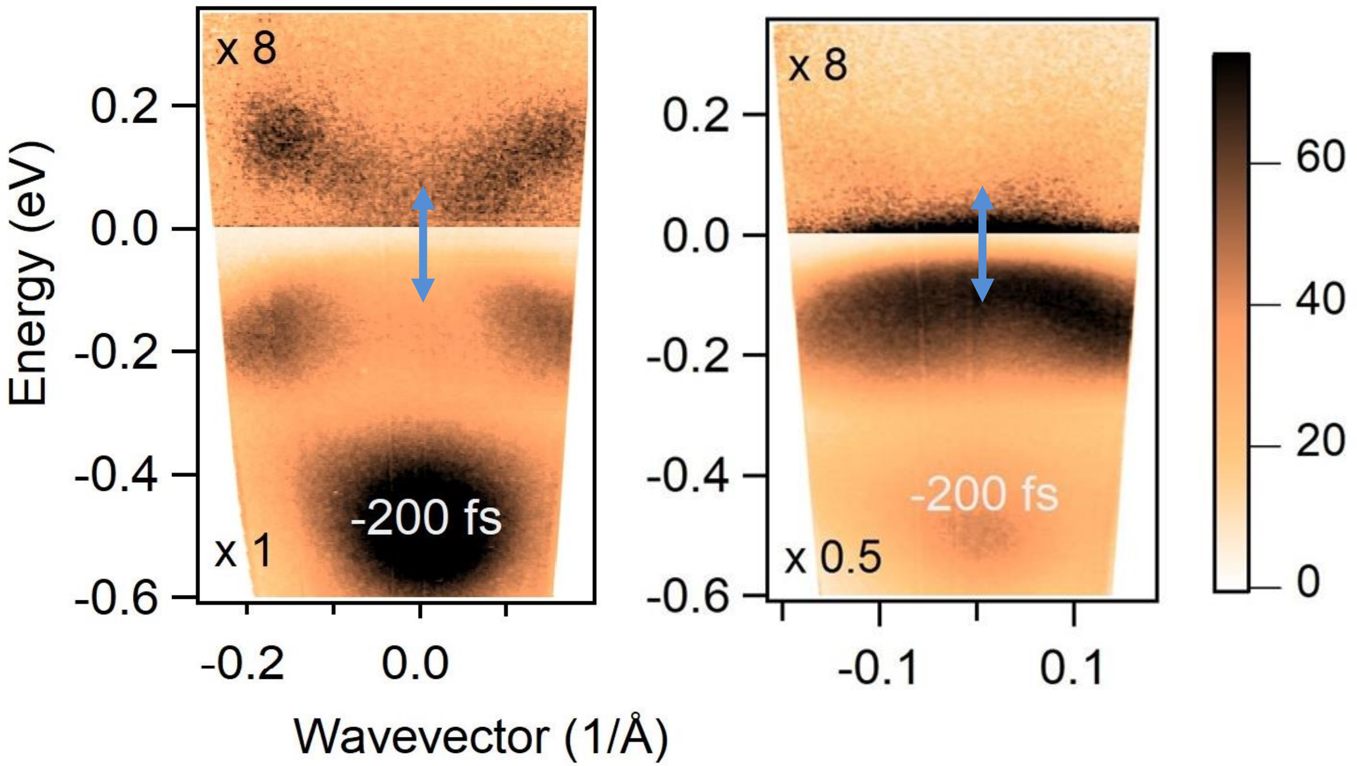
Gap 0.19 eV



Different orbitals define the gap

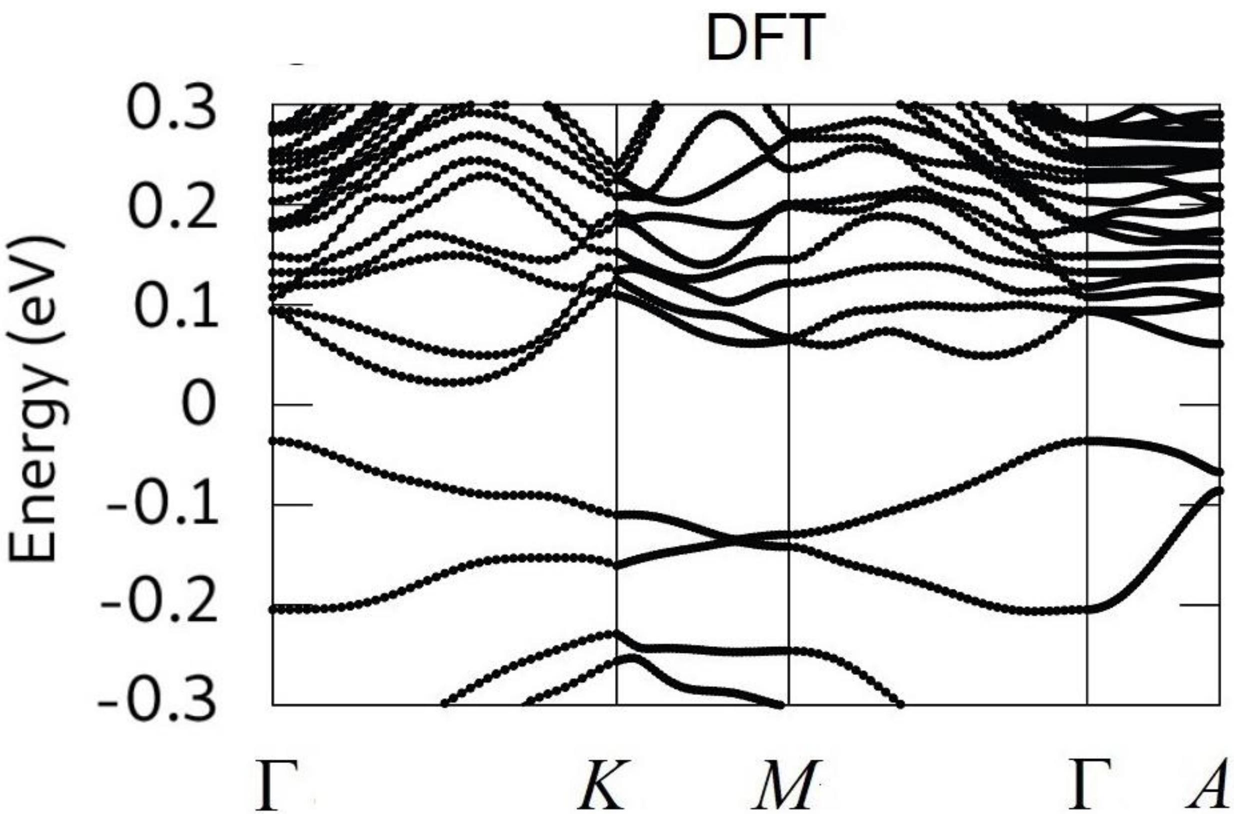
S Polarization

Gap 0.19 eV



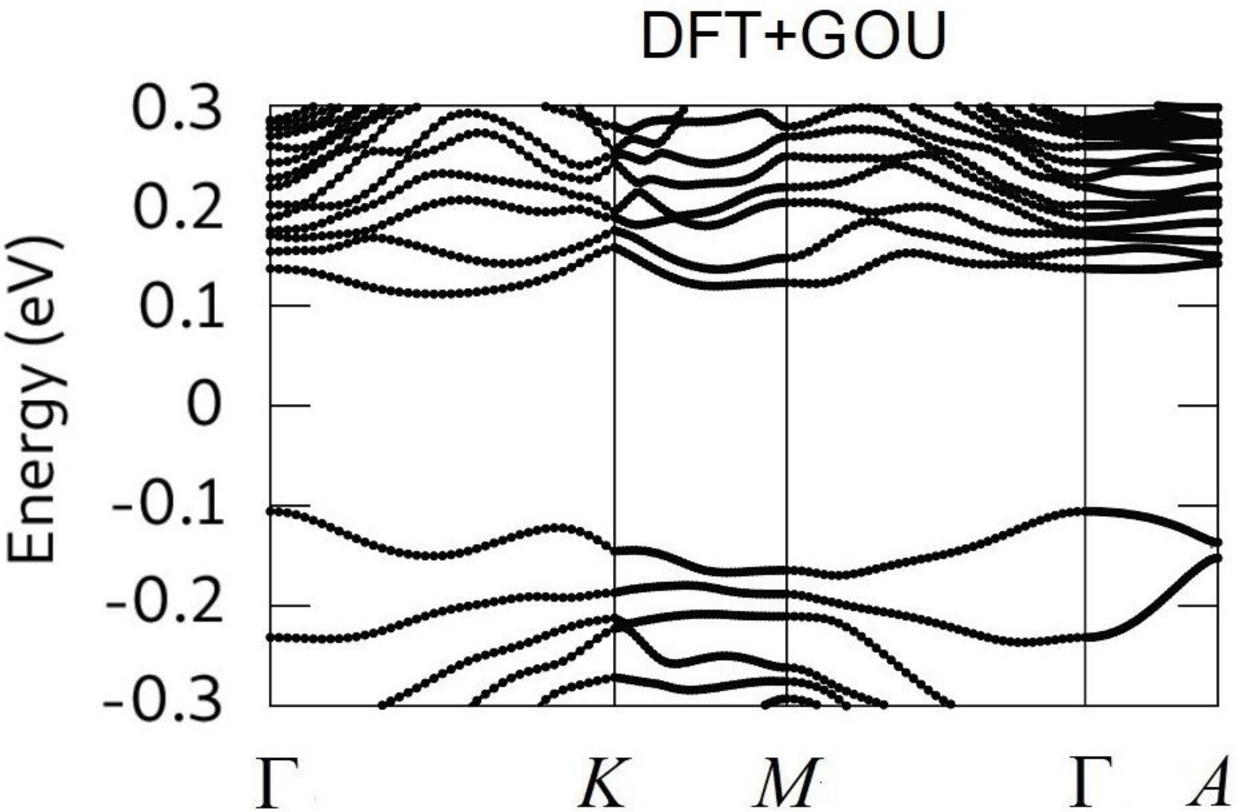
Simple Mott scenario breaks down

Density Functional Theory



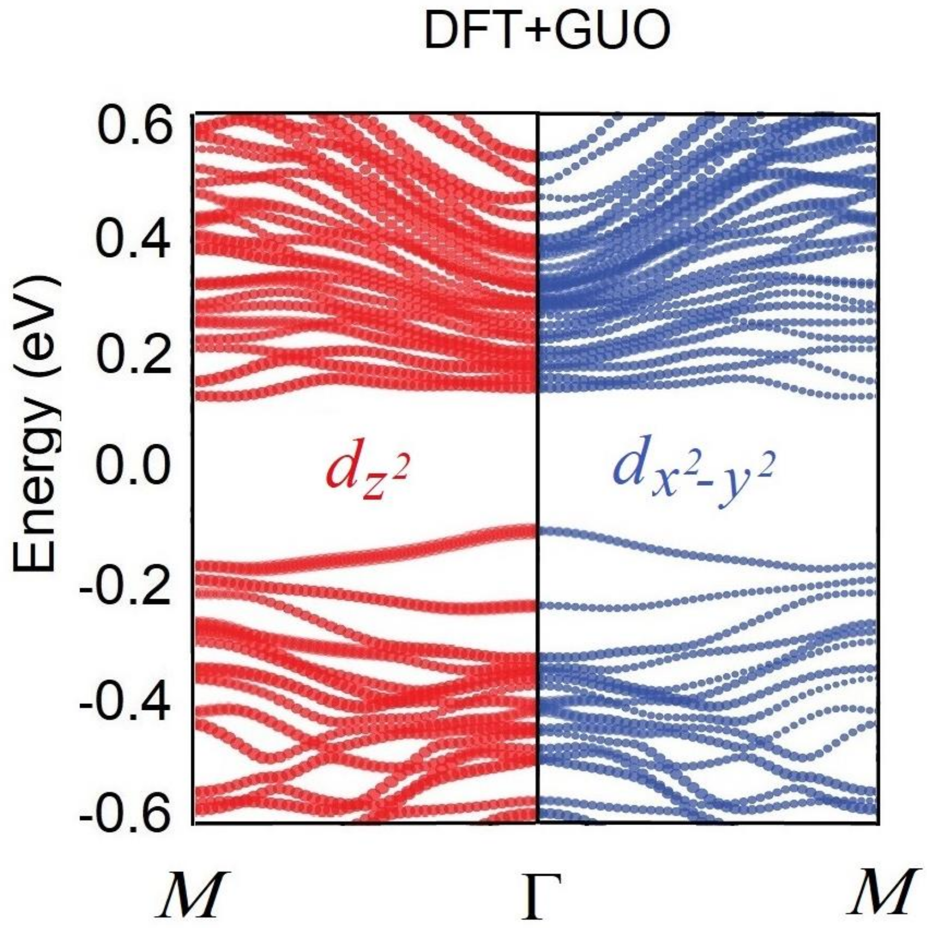
Gap
0.08 eV

Generalized Orbital U

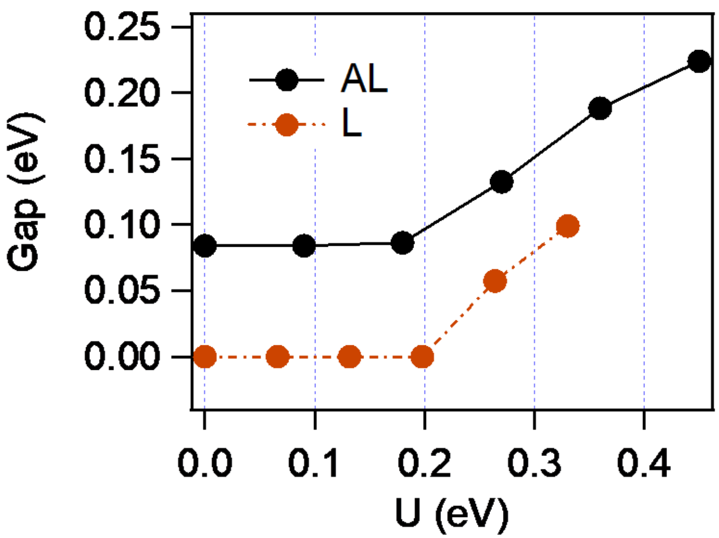


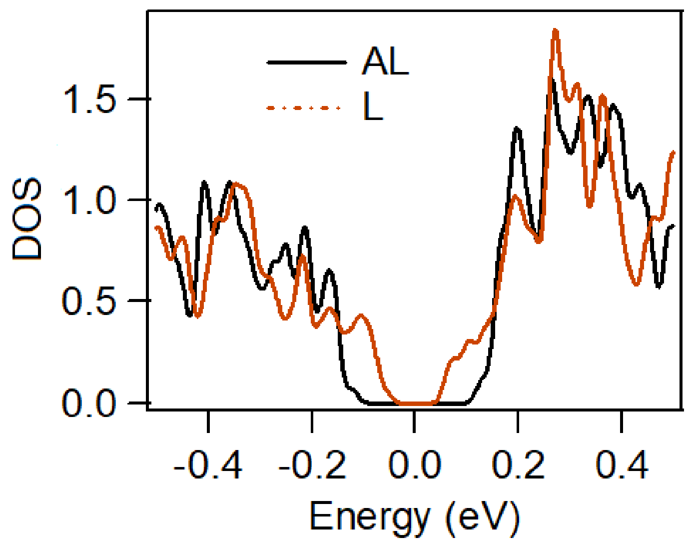
Gap
0.22 eV

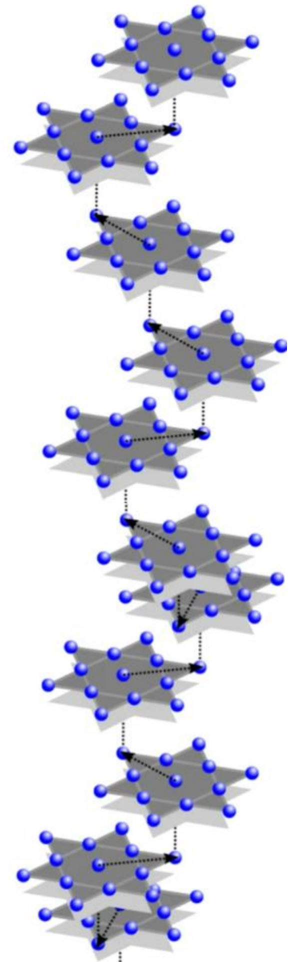
Generalized Orbital U



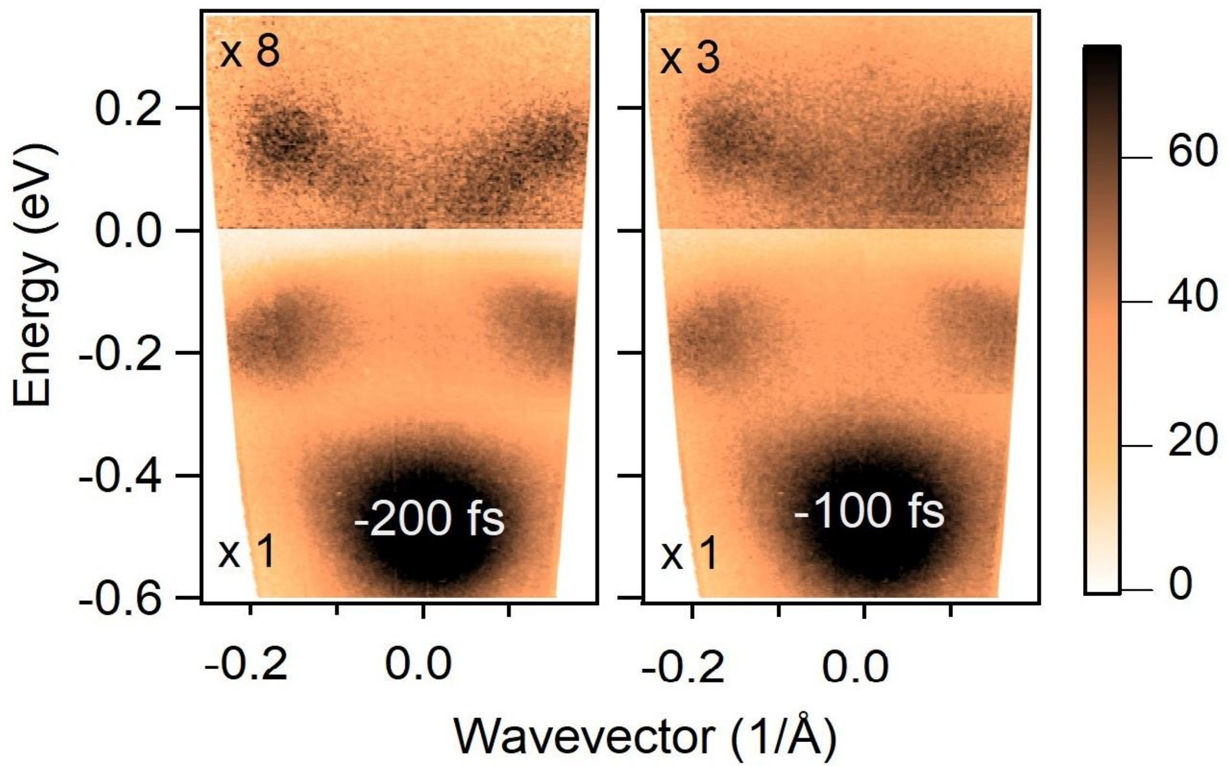
Dimerization and correlations



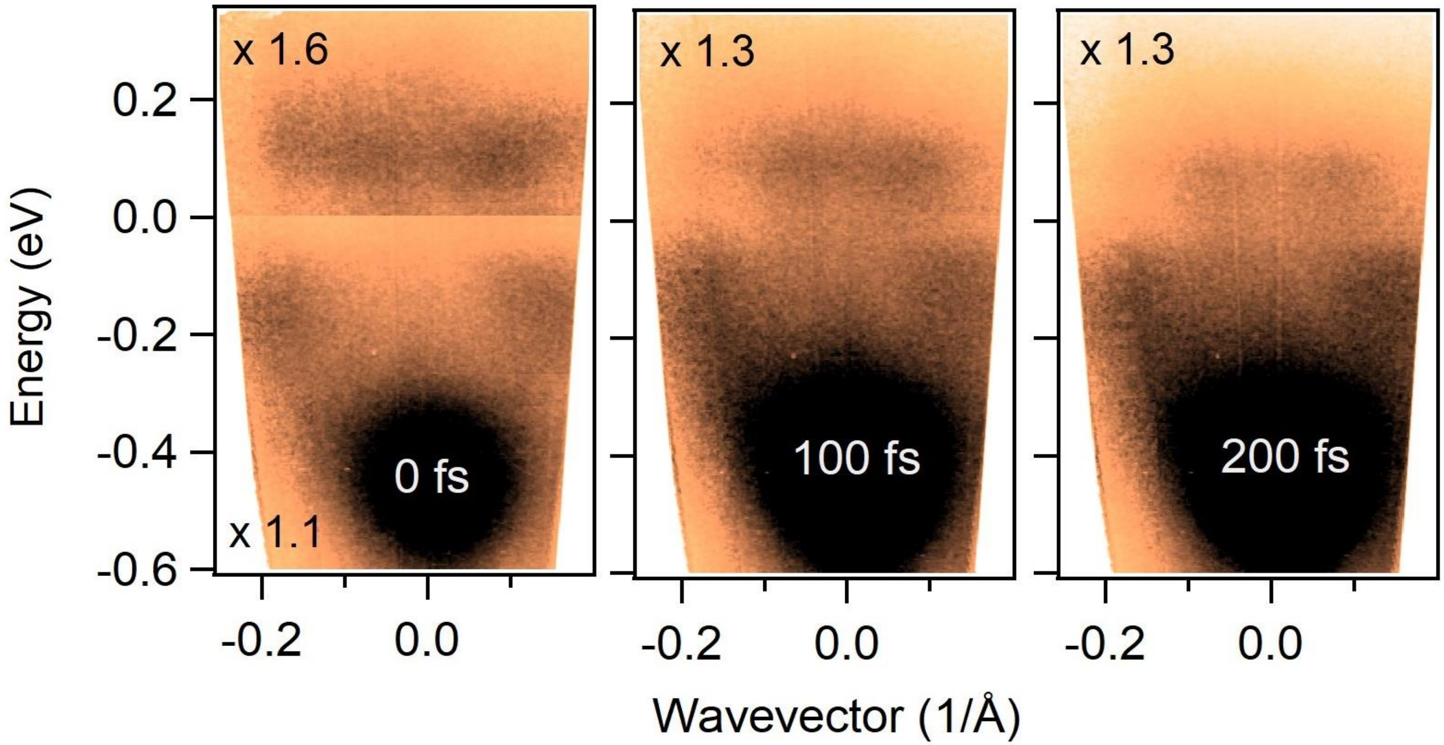




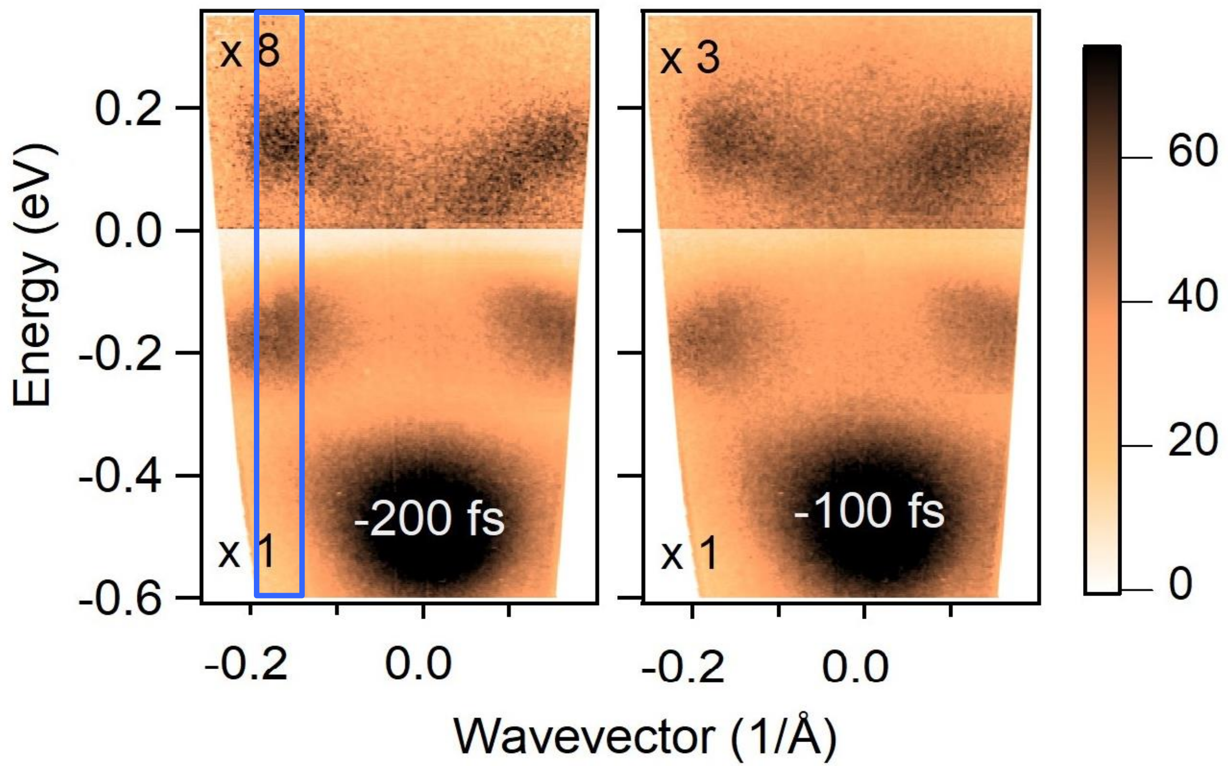
Collapse of the CDW



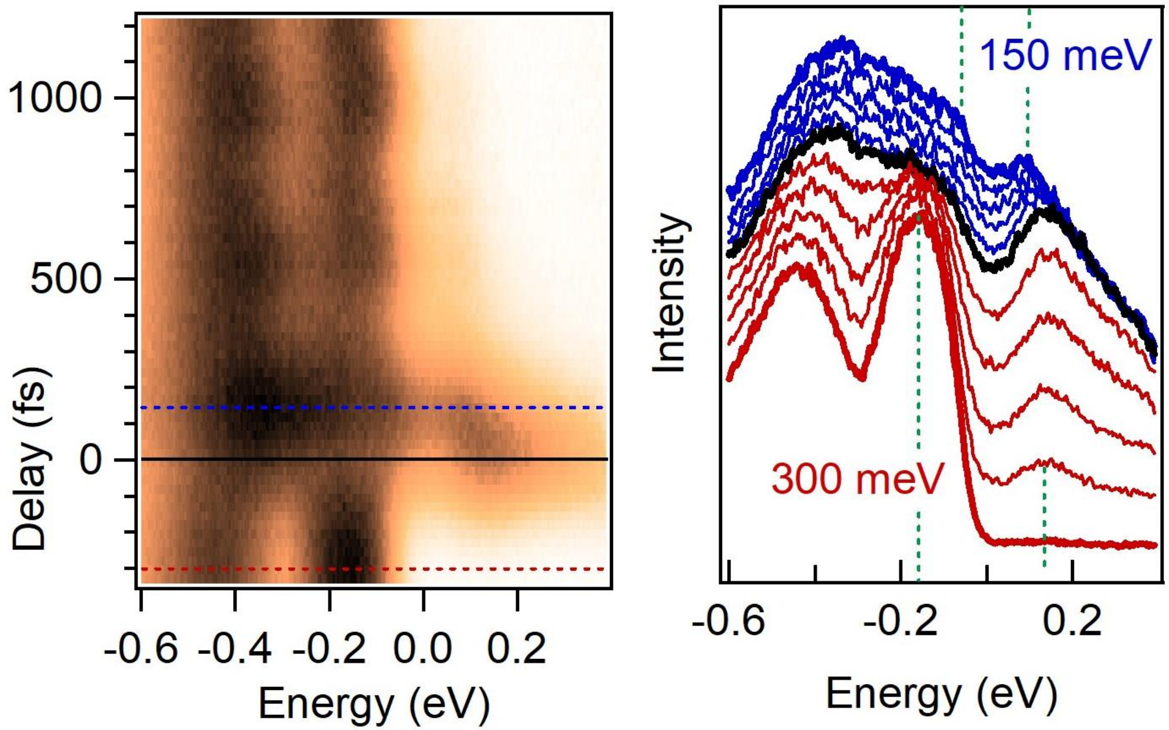
Collapse of the CDW



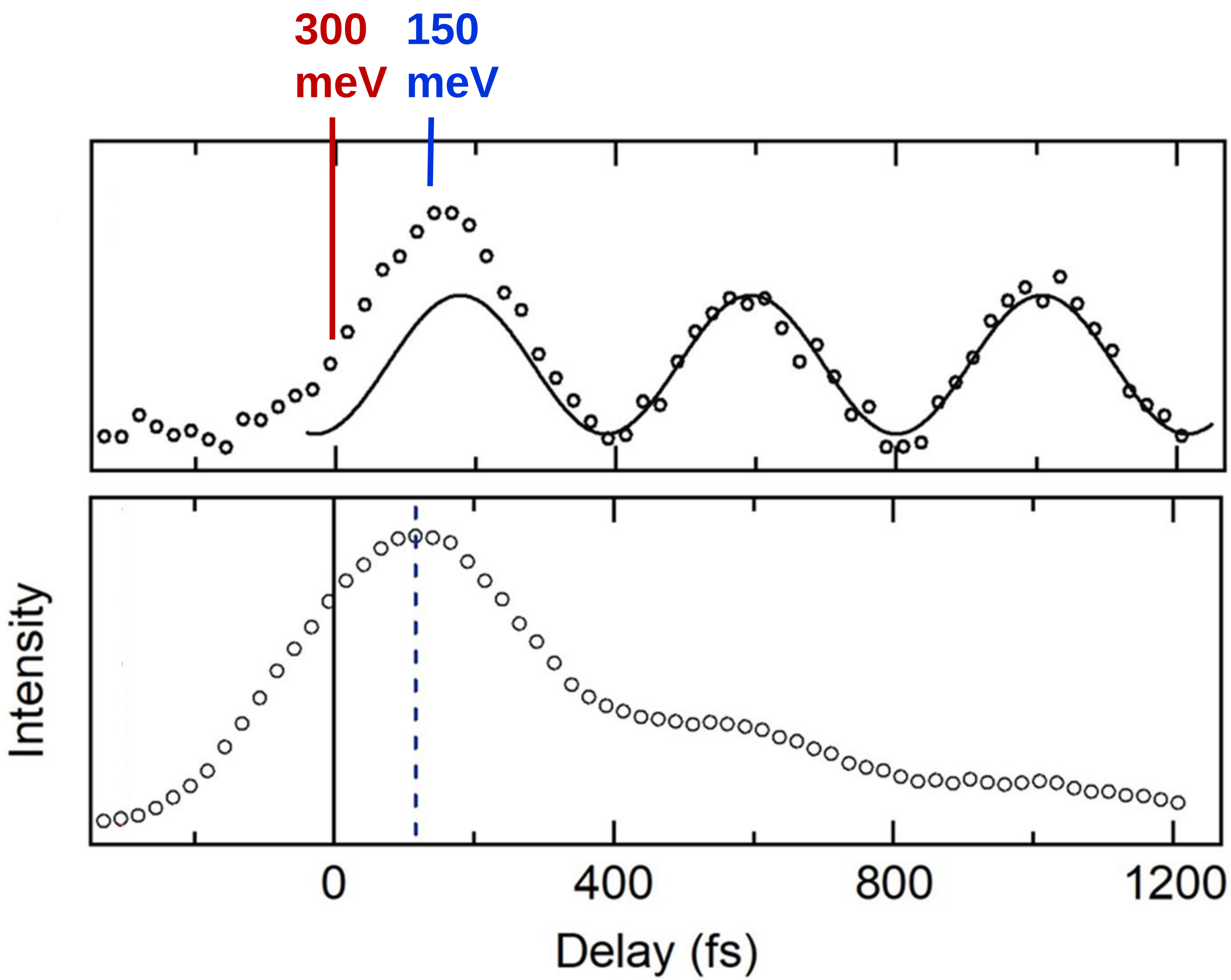
Collapse of the CDW



Gap dynamics



Fluctuations and Amplitude



Coherent motion of CDW amplitude mode

Shrinking of gap size

CDW fluctuations
Shorter coherence length

Filling of the gap

Conclusion

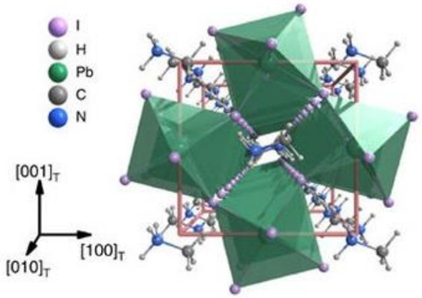
Observation of Dispersing states above the chemical potential

Comparison with DFT: indication of a Peierls-Mott insulator

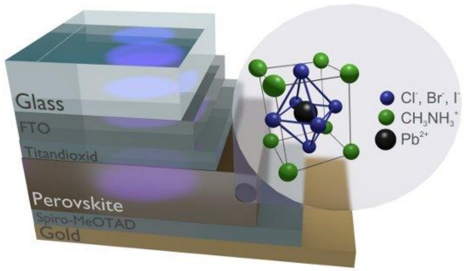
Collapse of the charge gap: strong and ultrafast fluctuations on top of a CDW amplitude oscillations

Collaborators

Hybrid perovskite
$CH_3NH_3PbI_3$



Solar cells



LSI - Ecole Polytechnique

Marie CHERASSE
Zhesheng CHEN
Jingwei DONG

LPS - Paris Sud

Evangelos PAPALAZAROU
Marino MARSI

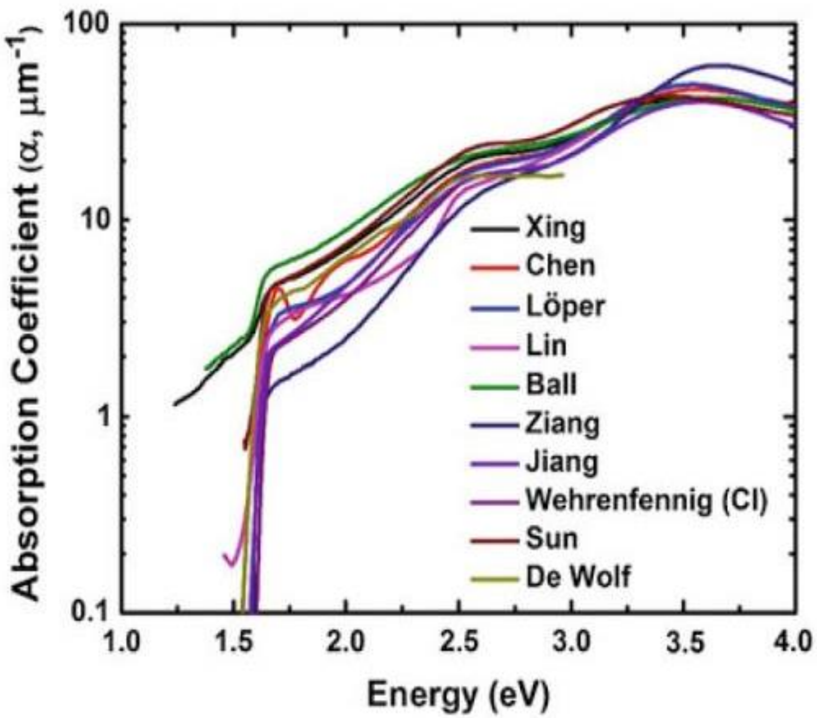
LPS - Paris Sud

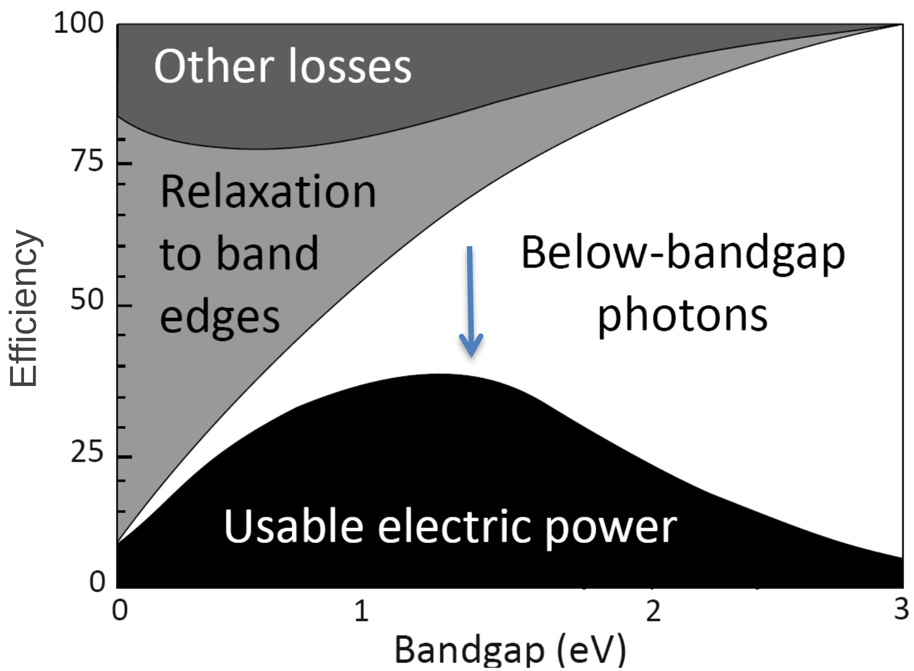
Emmanuelle Deleporte
Gaelle trippé Allard
Damier Garrot

Lumière, Matière et Interfaces

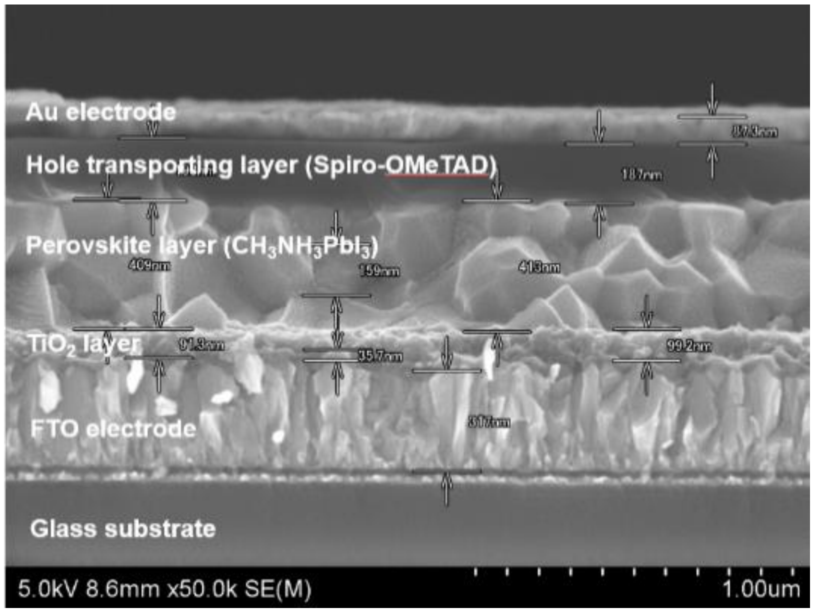
High carriers mobility

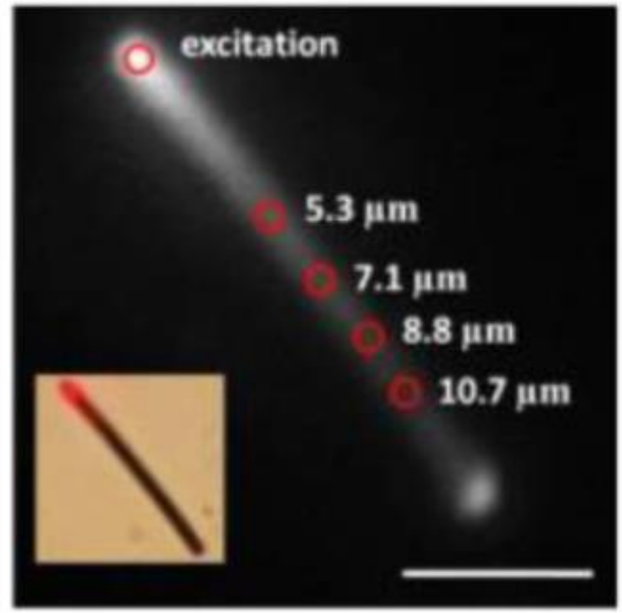
Ideal gap size, high absorption coeffient





High carriers mobility

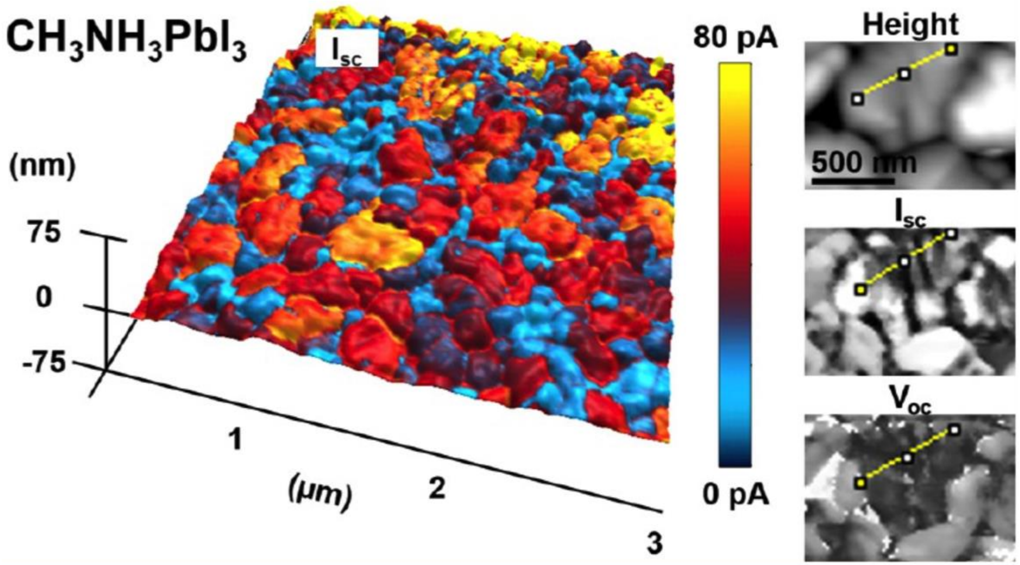




Time and spatially resolved photoluminescence:

D (cm^2 s^{-1})	μ (cm^2 V^{-1} s^{-1})
2.05 ± 0.02	79.7 ± 0.7

Internal fields

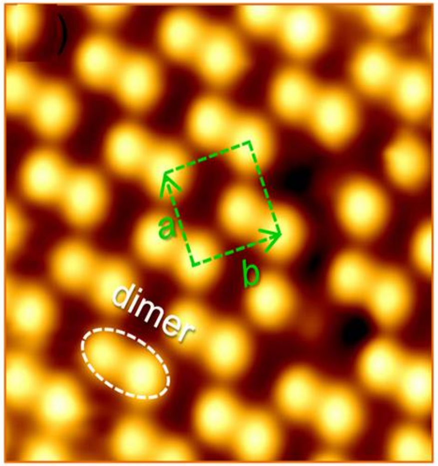


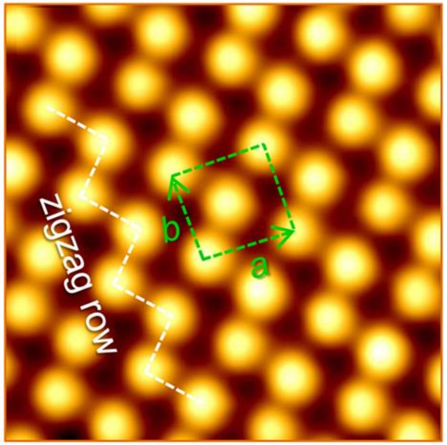
Interface of multi-grains

S. Y. Leblebici, et al., Nature Energy 1, 16093 (2016)
Y. Kutes, et al., Nano Lett. 16, 3434 (2016)

Internal field exist at the interface of grains
Spatial variation of open circuit voltage and local conversion efficiency

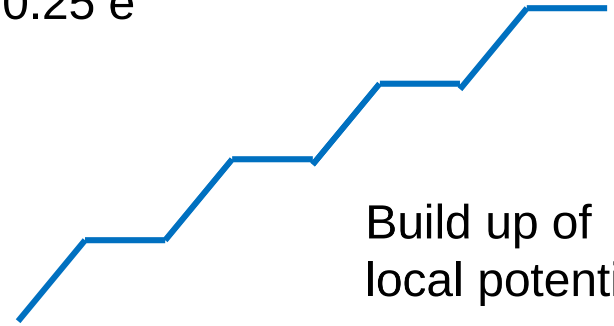
Ionic Surface Terminations



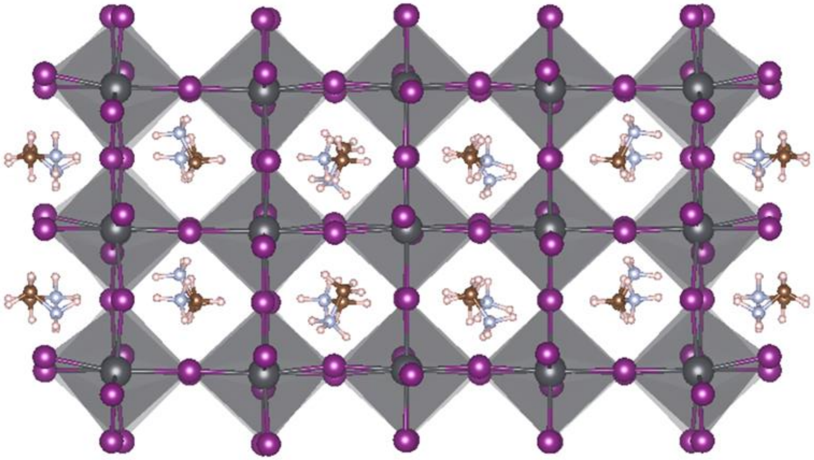


L. Shi, et al., ACS Nano 10, 1126 (2016)

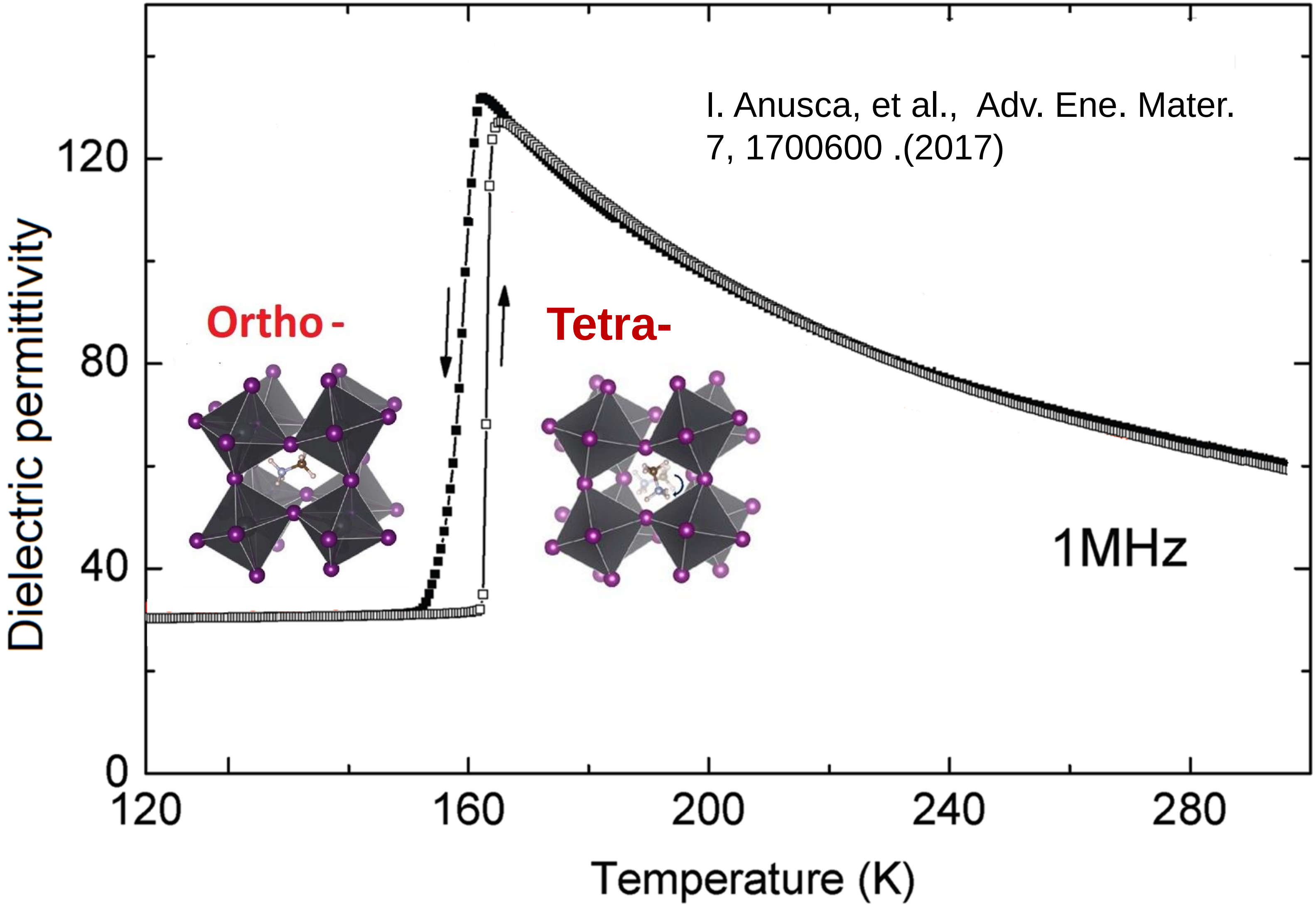
+ 0.25 e



Build up of
local potential

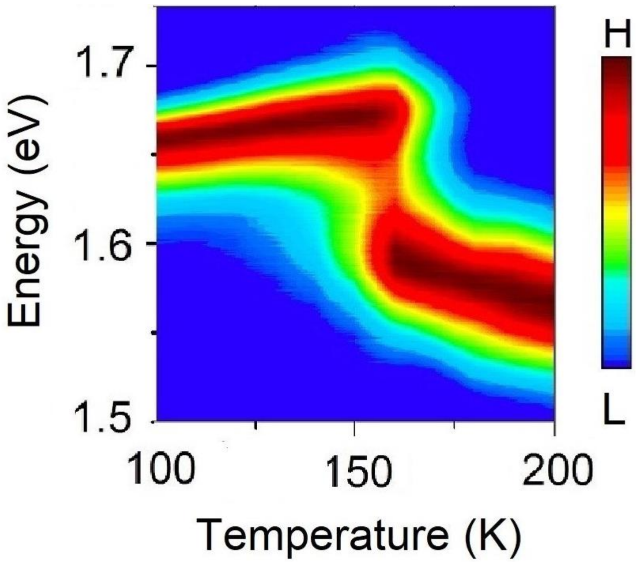


Dielectric Properties

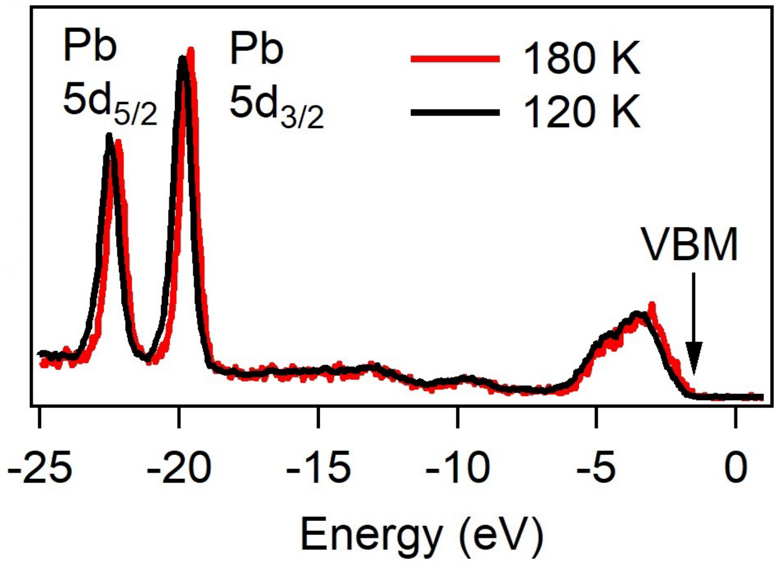


Phase transition

Photoluminiscence

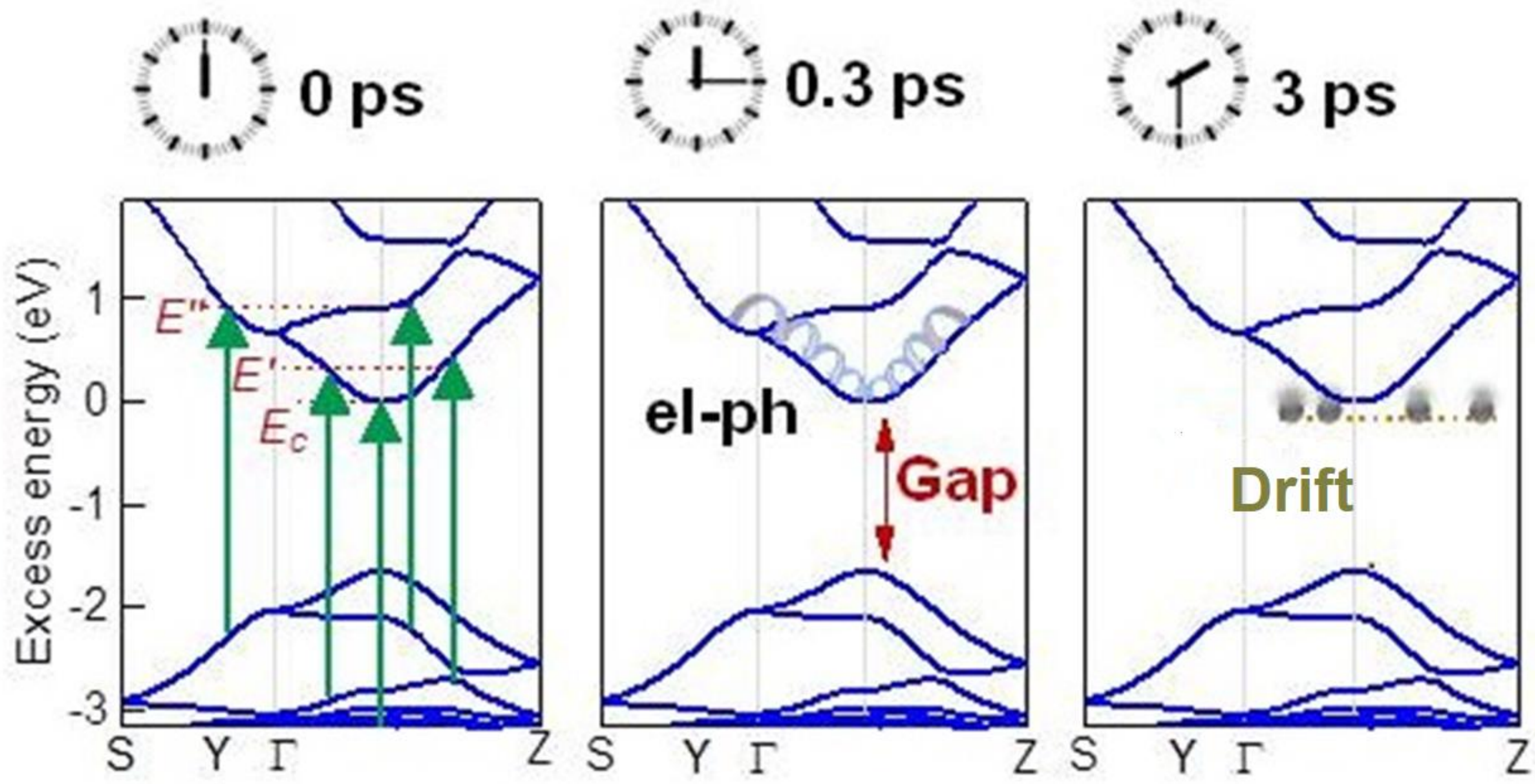


Hard X-rays Phtoemission

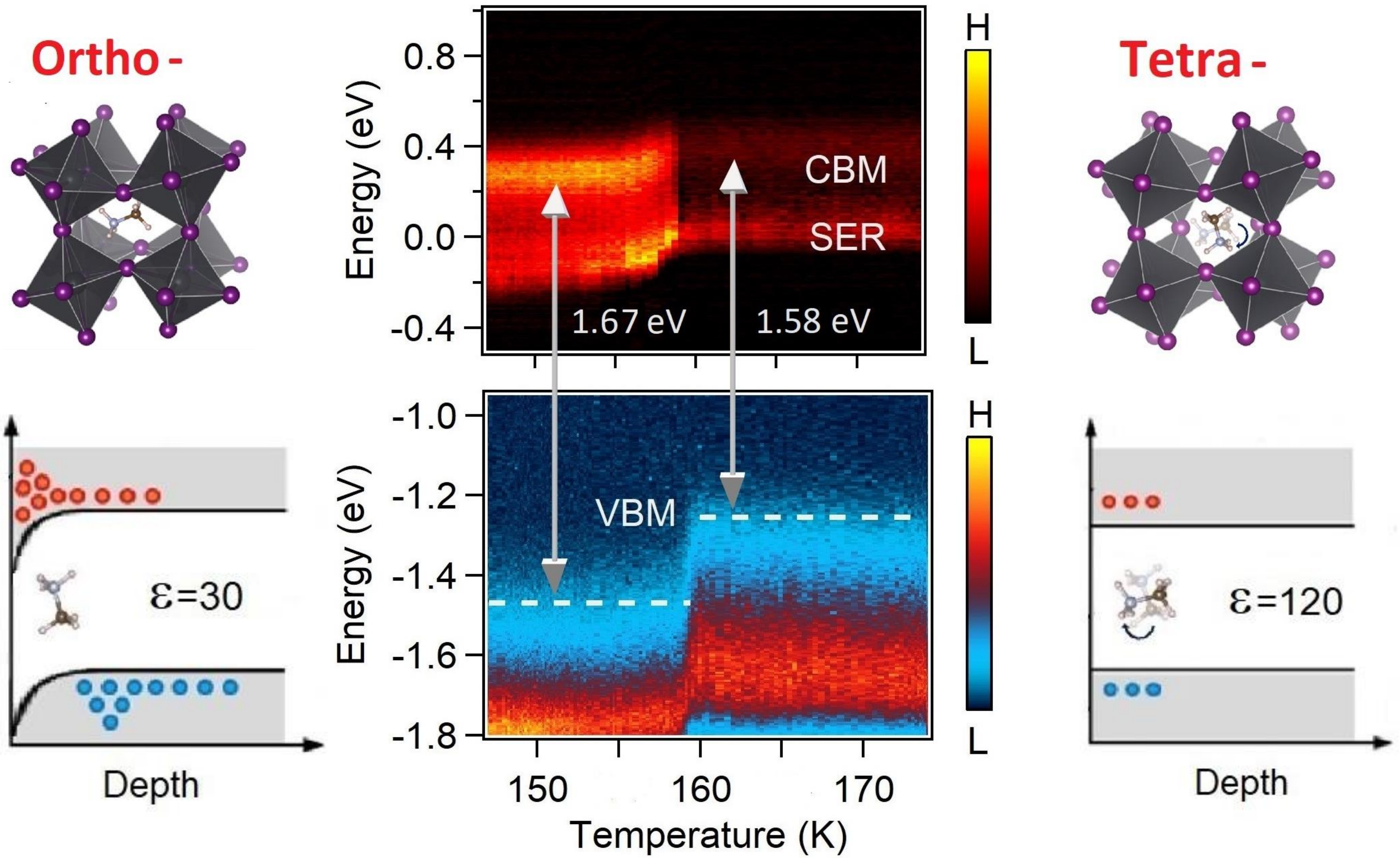


Energy Shift of the valence band: internal fields depends on the structural phase

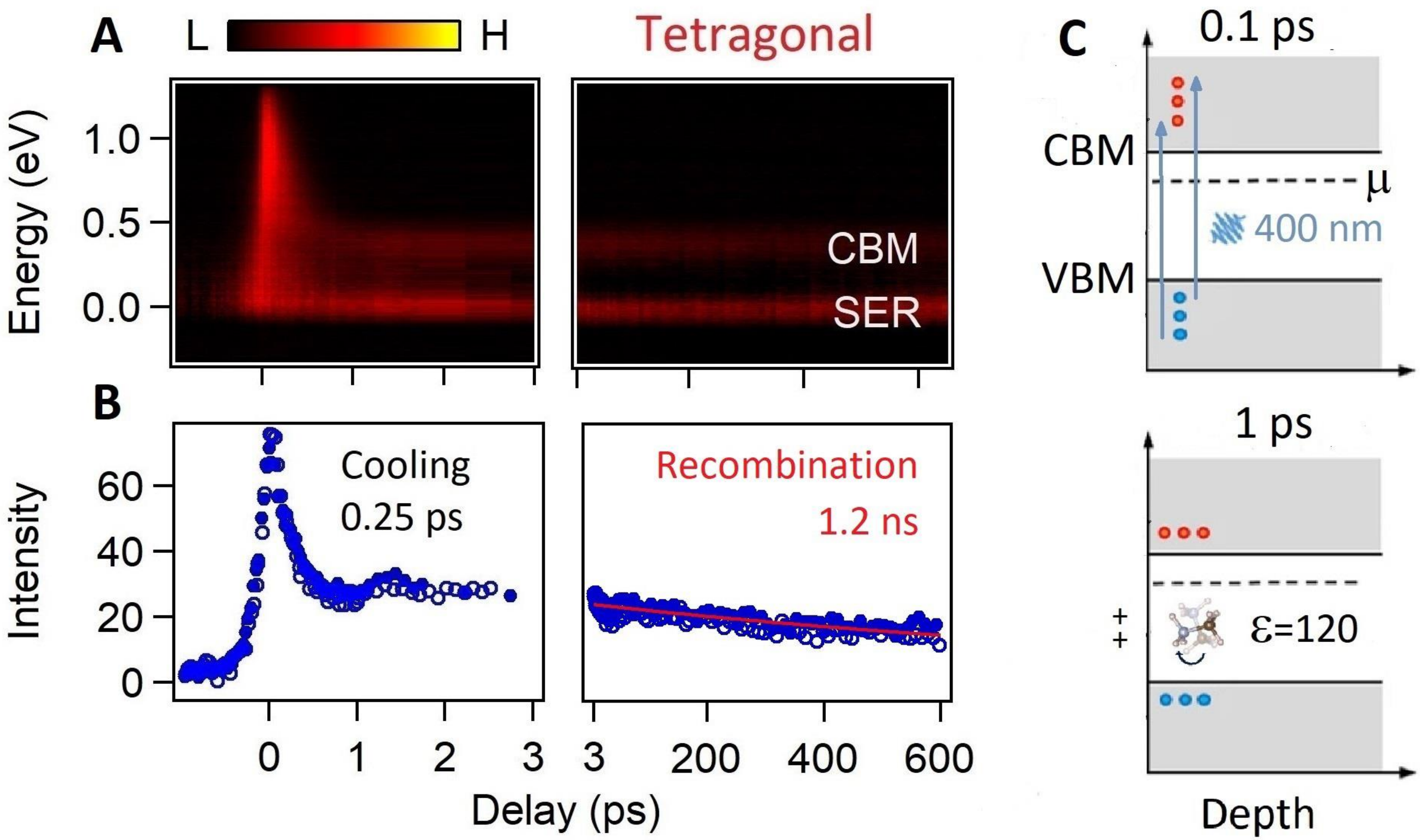
Photoexcited state



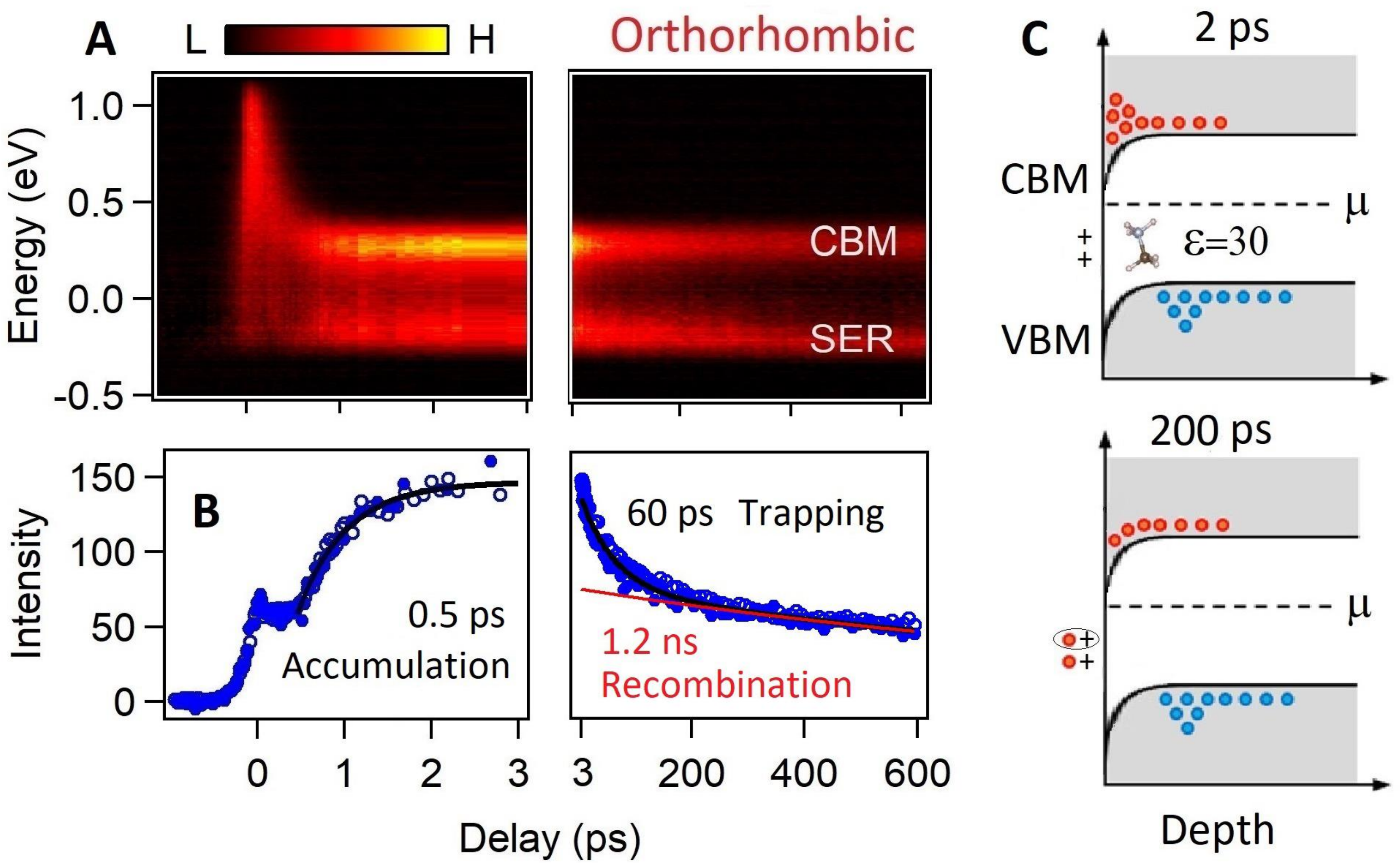
Type II band offset



Dynamics, high temperature



Dynamics, low temperature



Conclusions

Freely moving organic cations are capable of screening local fields in hybrid perovskites.

Type two band offset between the orthorombic and tetragonal phase of $CH_3NH_3PbI_3$