Time- and Angle-Resolved Photoemission Techniques for Understanding Correlated Electron States

Charlotte Sanders

UK Central Laser Facility, Artemis Program

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Acknowledgements

Artemis

- Y. Zhang, R. T. Chapman, A. S. Wyatt, E. Springate

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And also

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- DESY FLASH
- Elettra Sincrotrone Trieste

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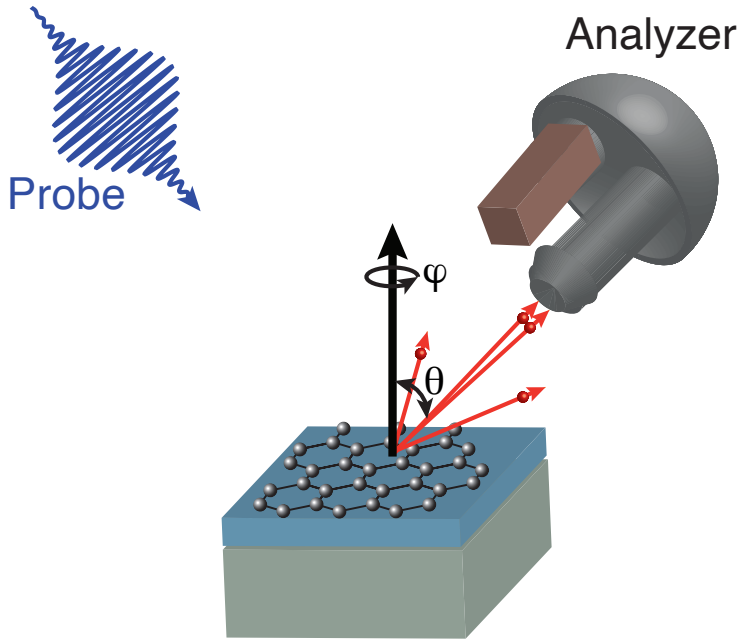
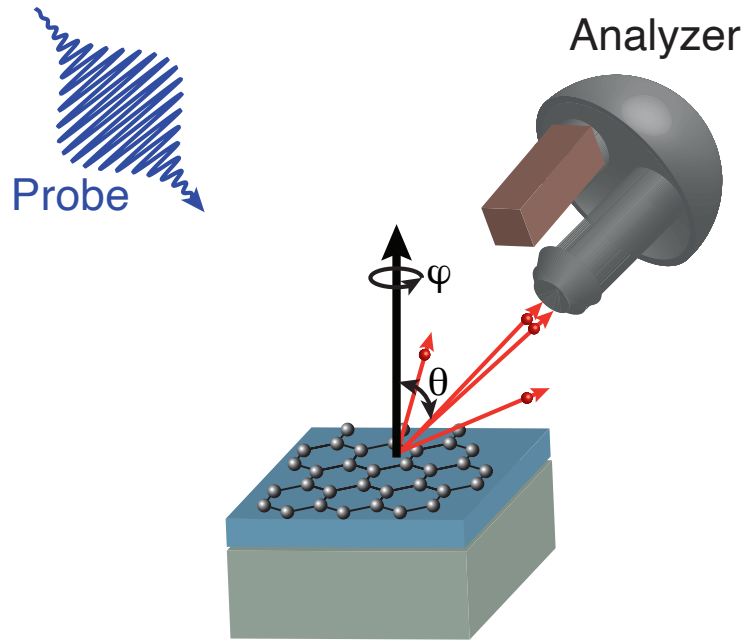
- EU Laserlab-Europe 871124
- VILLUM FONDON 11744
- EU CALIPSOplus 730872

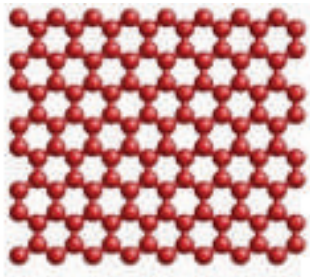
“Static” Photoemission Spectroscopy

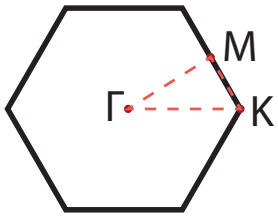


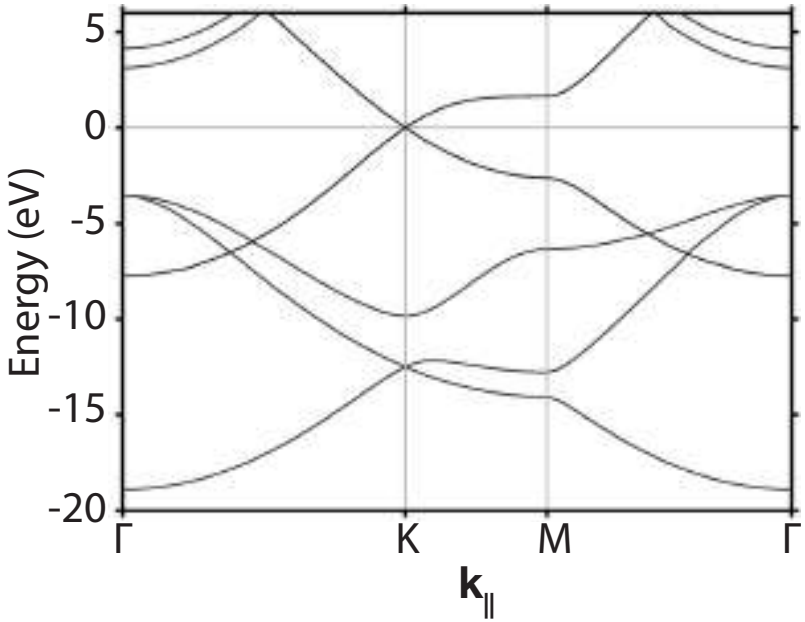
Angle-Resolved Photoemission Spectroscopy (ARPES)



Example: Graphene







Katsnelson, *Physics of Graphene* (2020)

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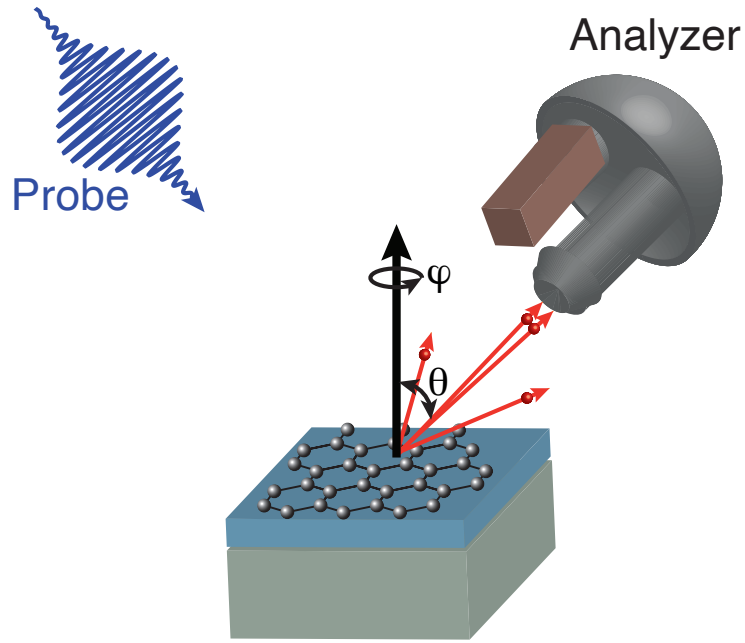

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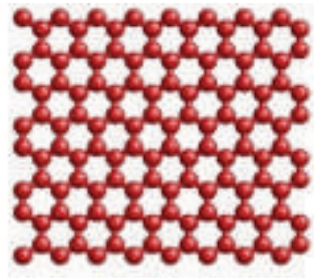

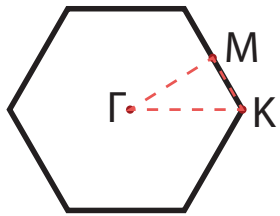

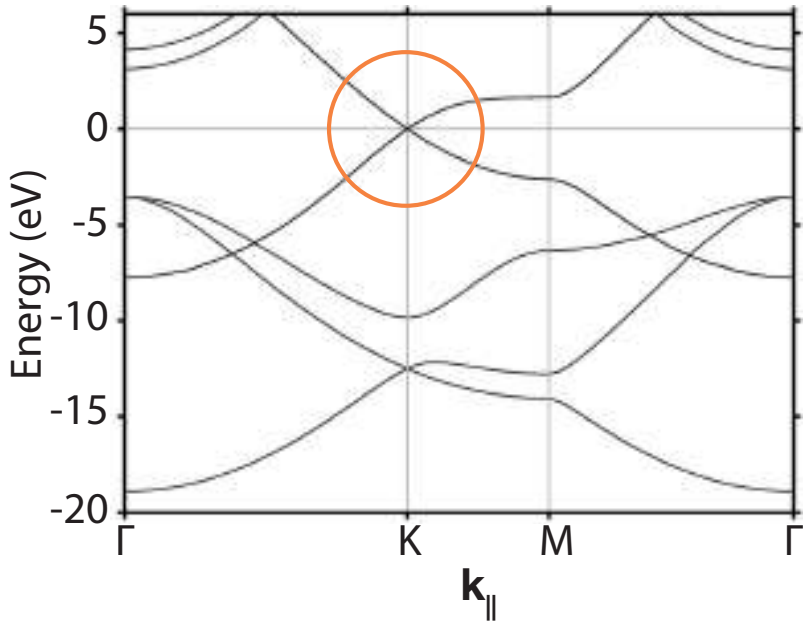

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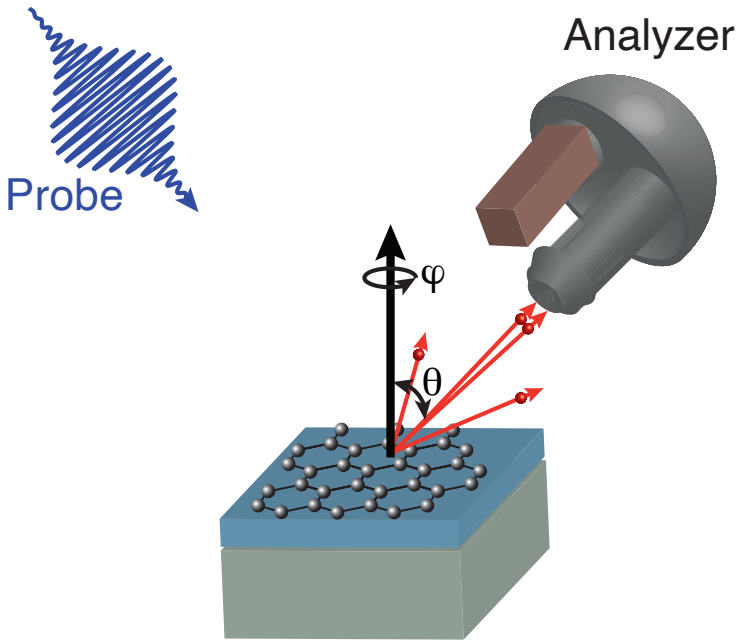

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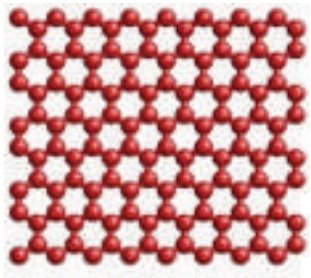

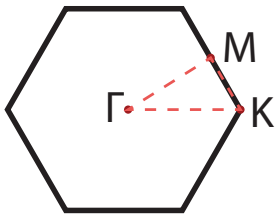

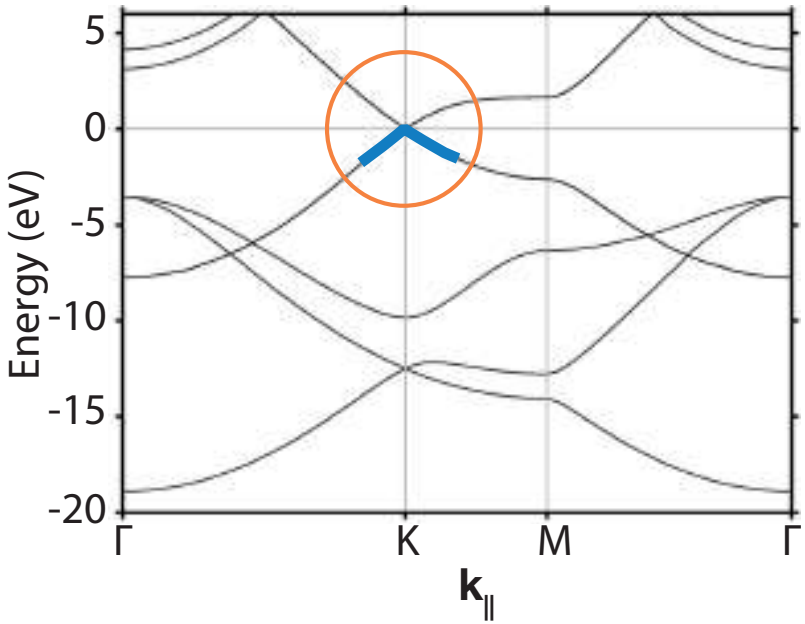

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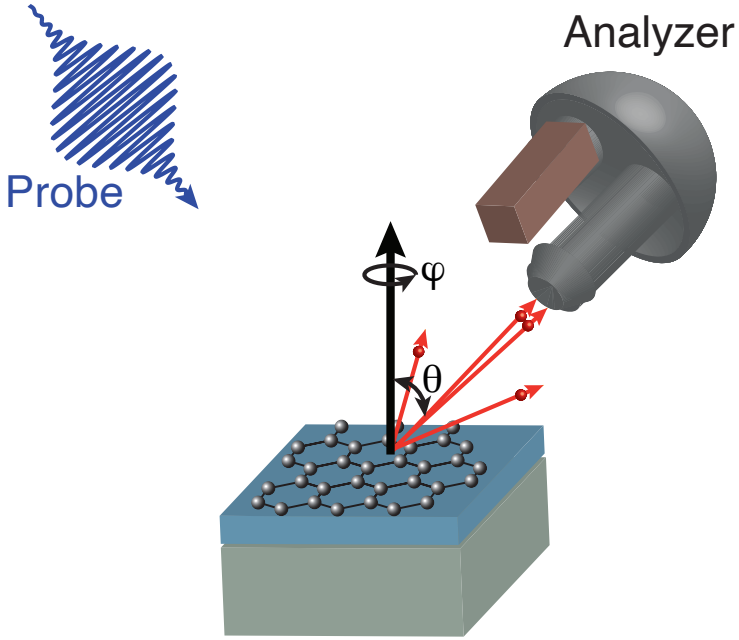

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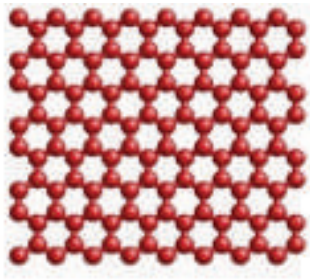
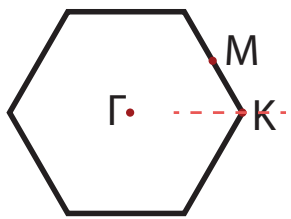
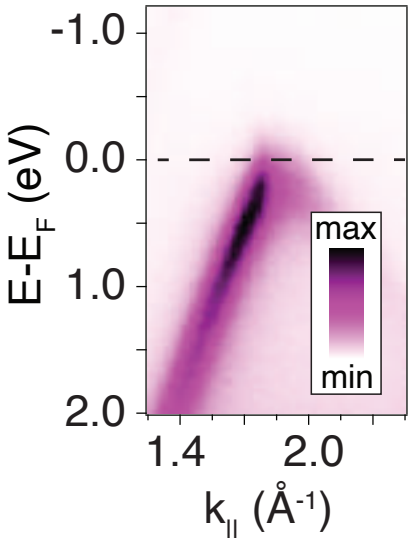
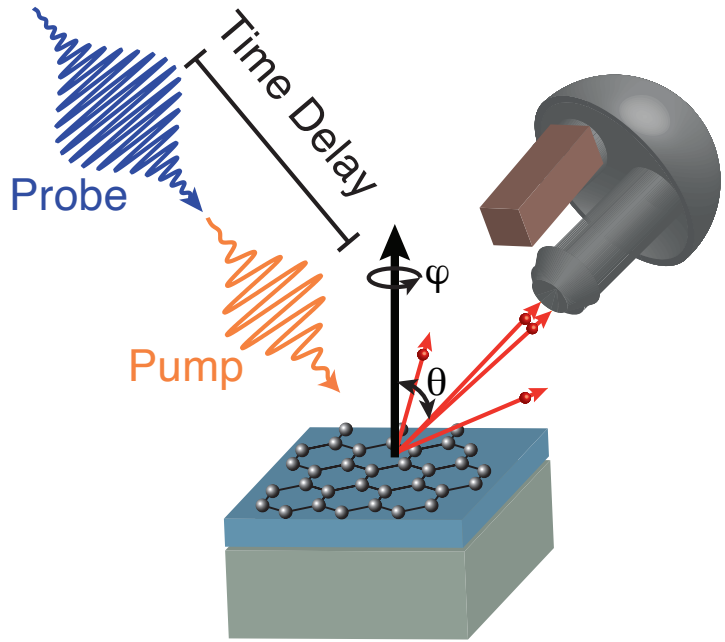






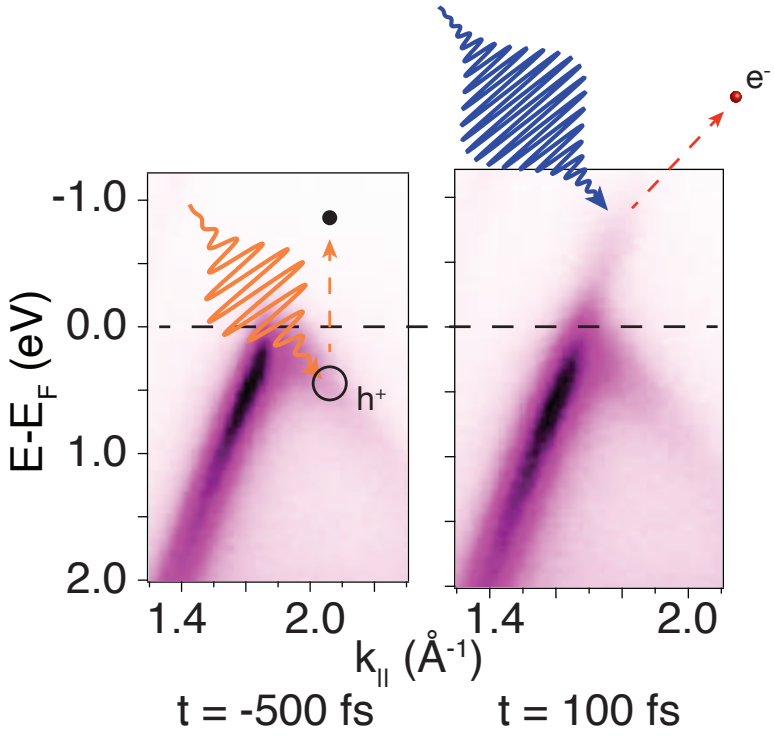
PRL **111** (2013) 027403

Pump-Probe: Time Resolution (TR)



Pump-Probe: Time Resolution (TR)

- Tune **pump photon energy**
- Tune **pump-probe delay**: measure **system dynamics**
- Matrix elements
  - Boschini, *et al., New J. Phys.* **22** (2020) 023031
  - Volckaert, *et al., PRB* **100** (2019) 241406(R)



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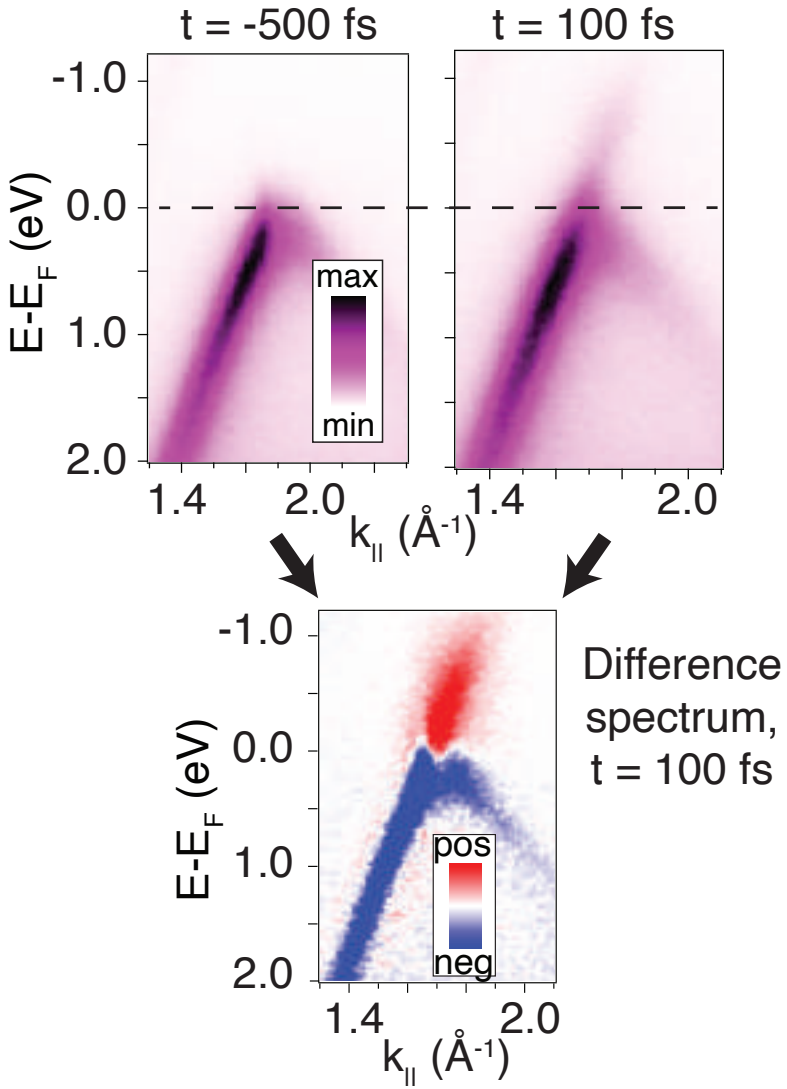
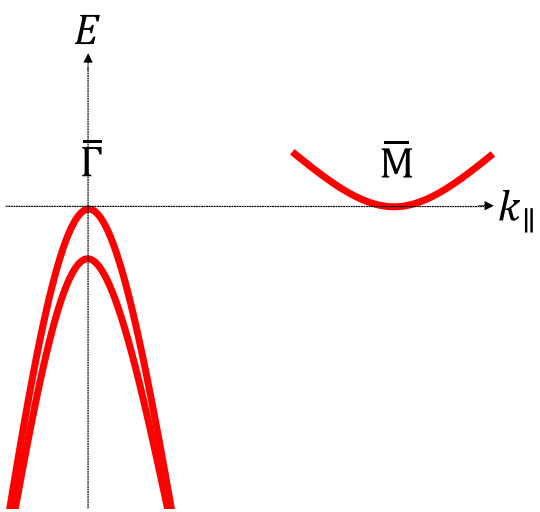
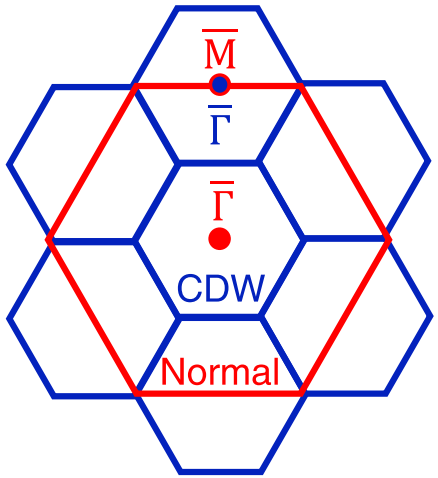


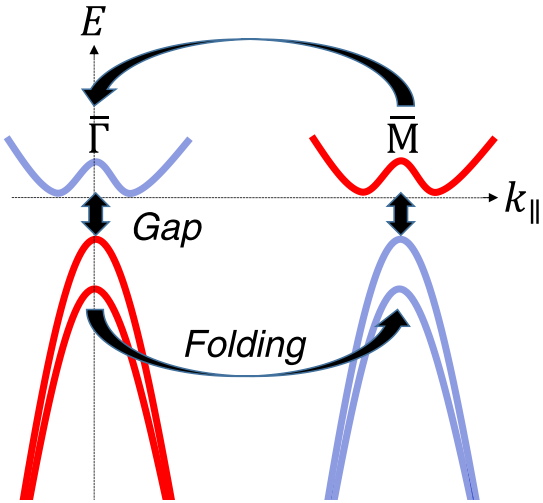
Dynamical Changes to Electronic Dispersion

Bulk 1T-$TiSe_2$: normal state



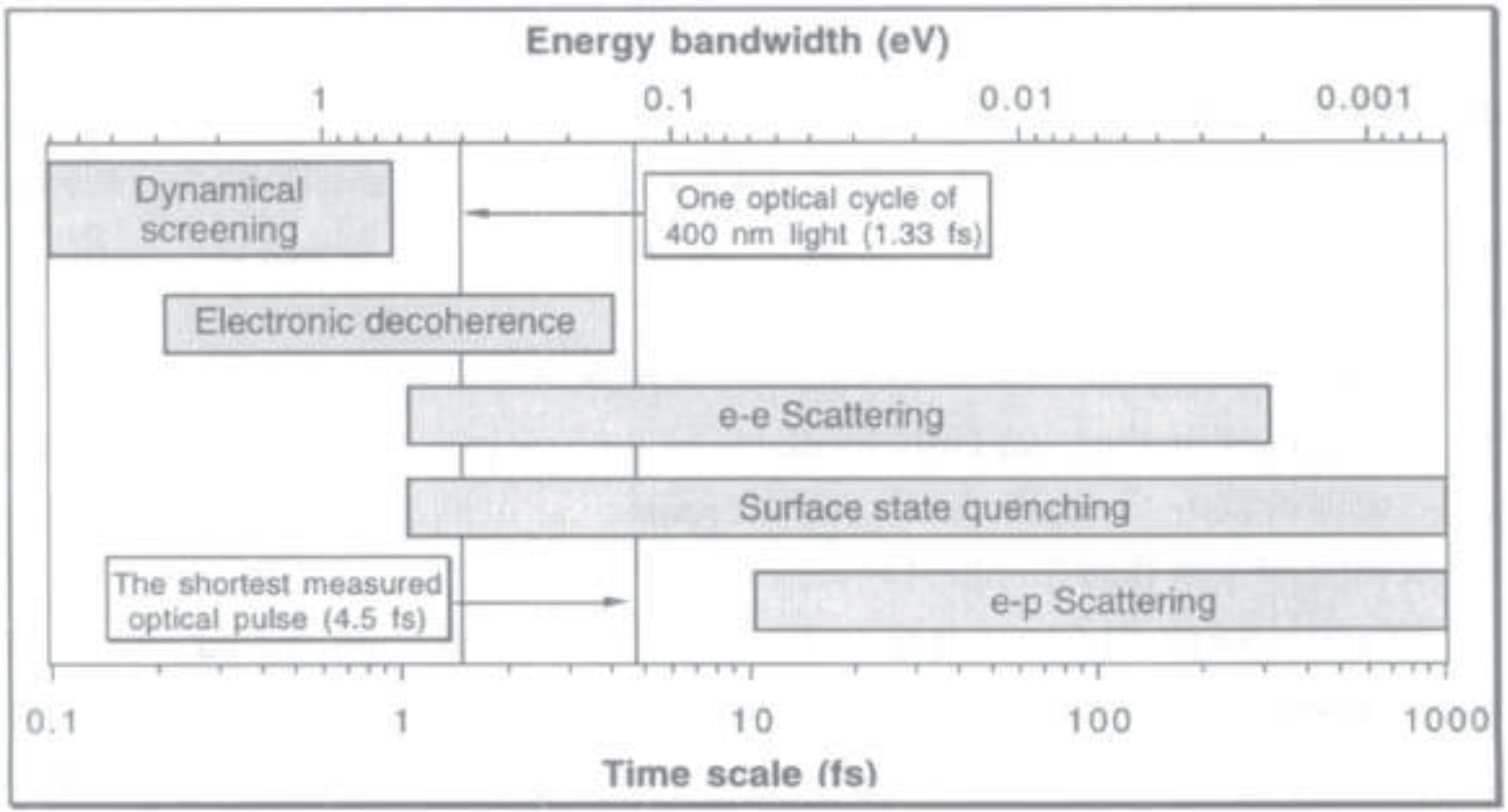


Charge density wave (CDW) state



Hedayat, *et al., Phys. Rev. Res.* **1** (2019) 023029

Time Scales



Petek & Ogawa, *Prog. Surf. Sci.* **56** (1997) 239

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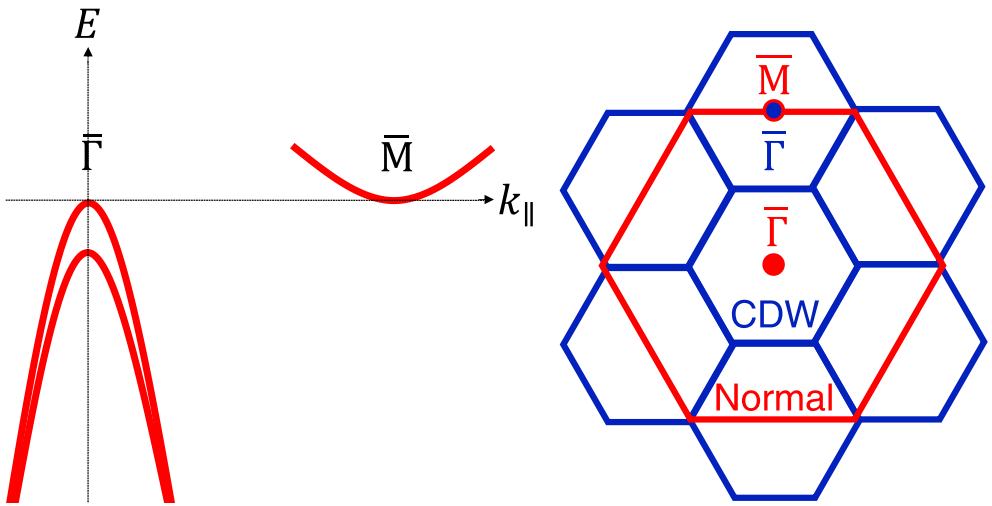
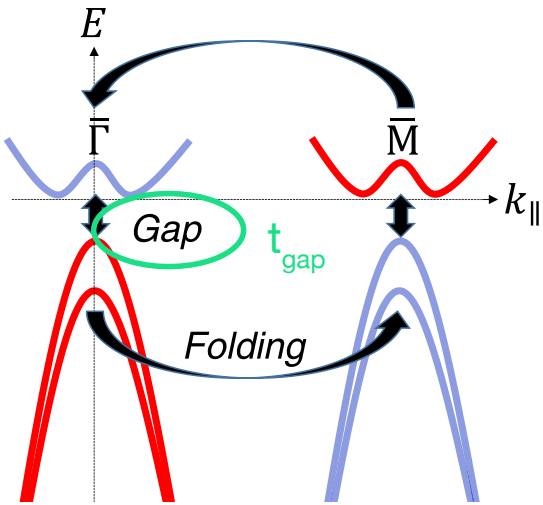


CDW state



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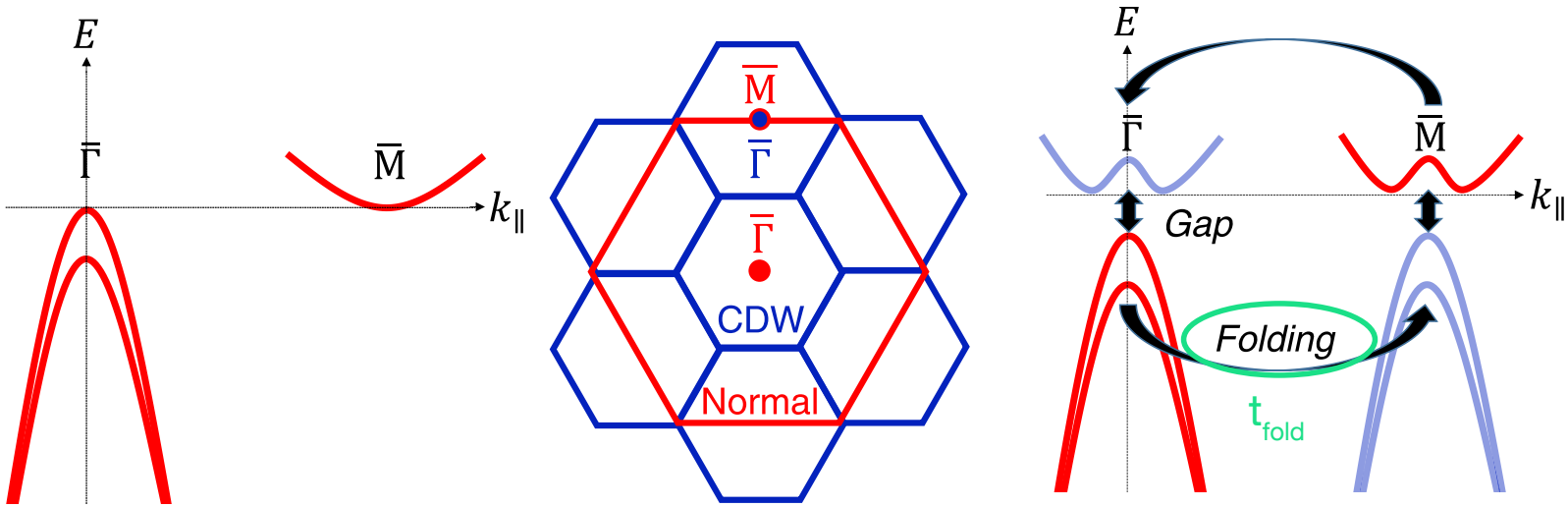

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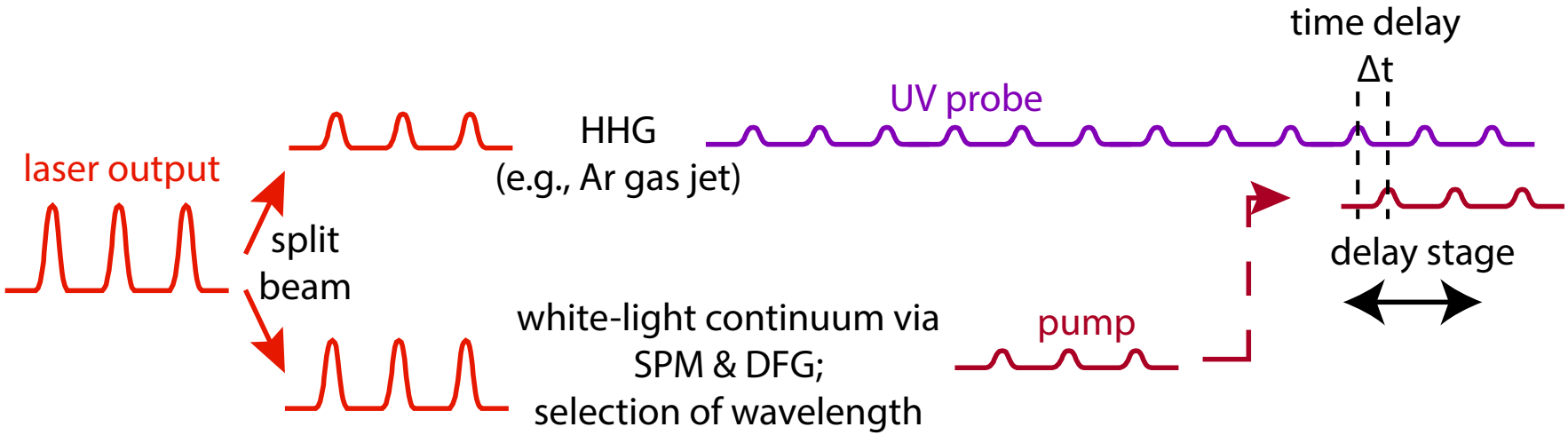
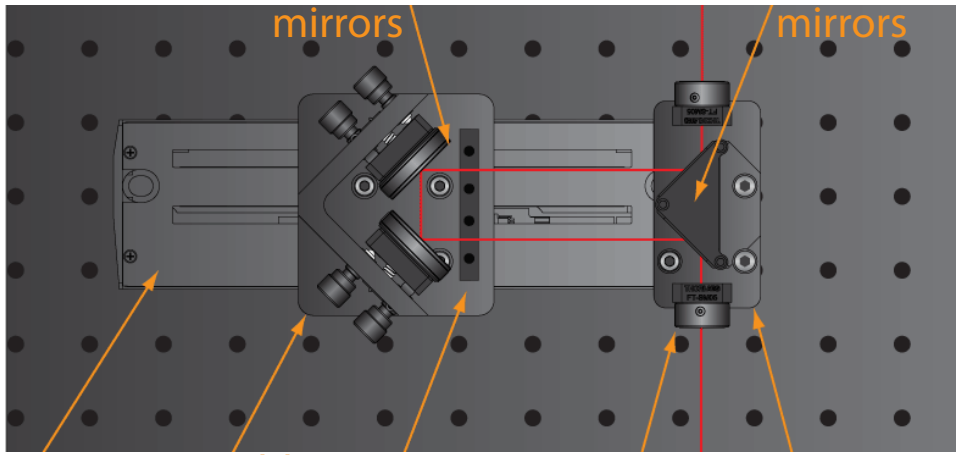
Generating Pump & Probe



delay stage:



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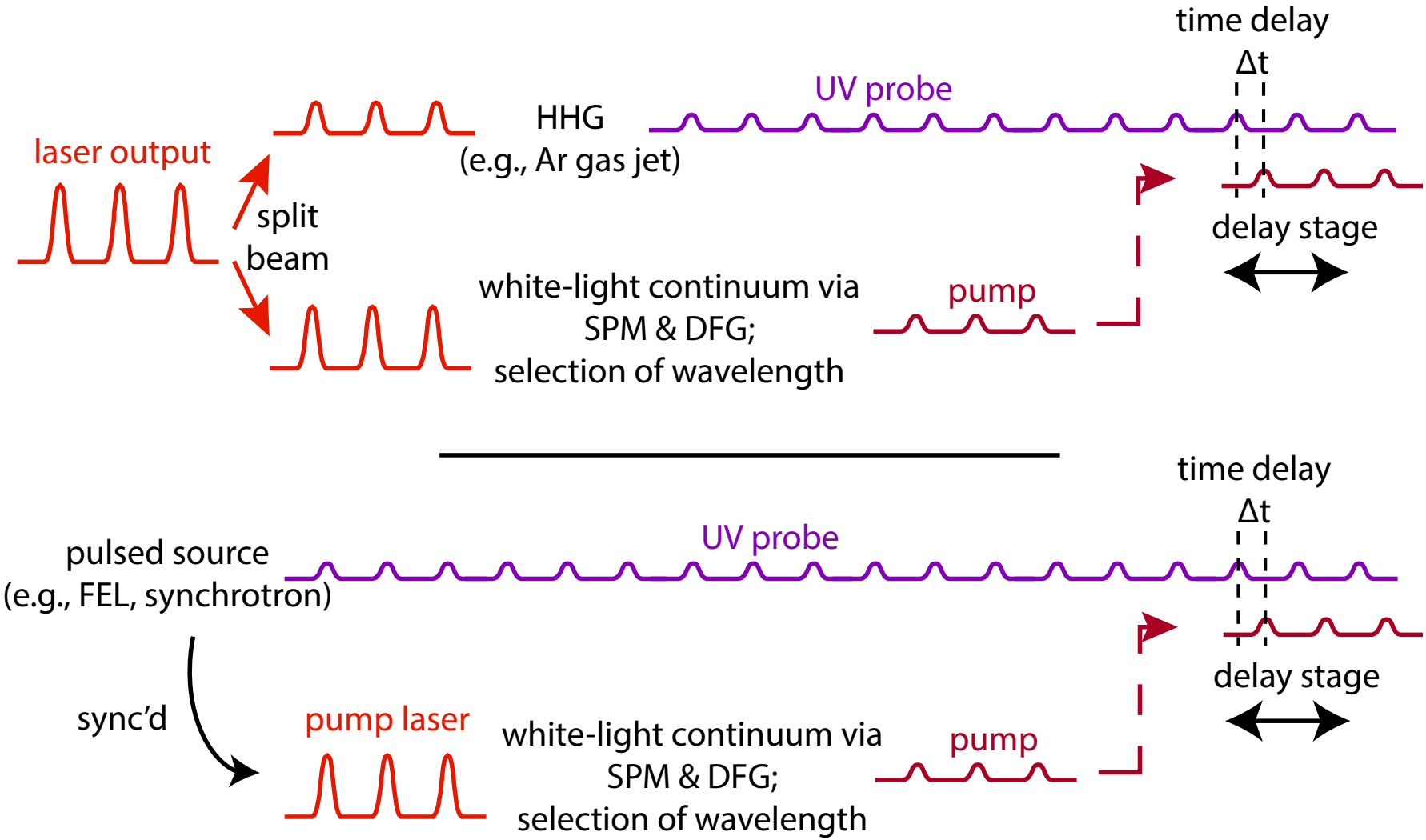
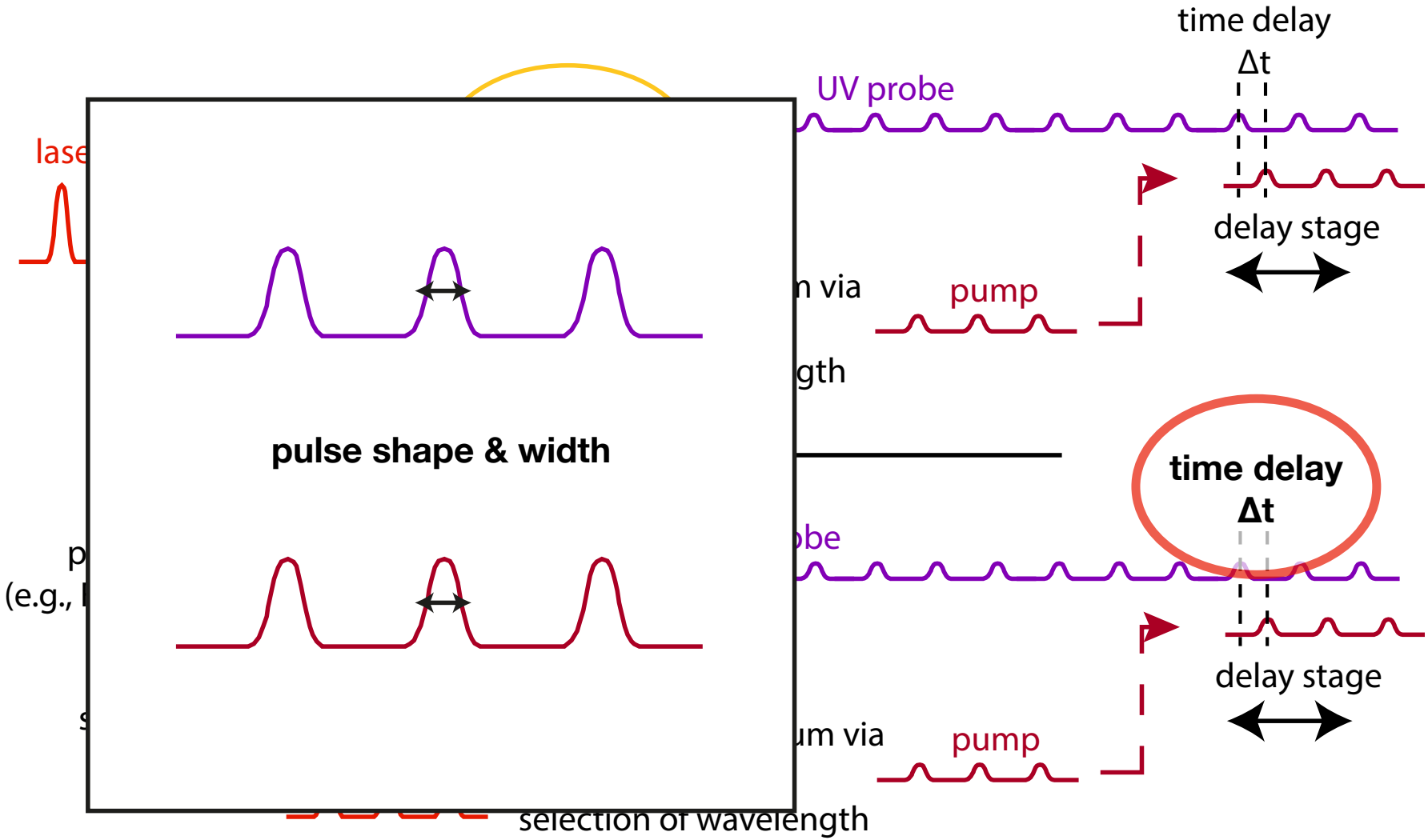


Time & Energy Resolution



Light Sources

Tabletop HHG Setups

- Good time resolution, e.g. $\sim$ tens of fs
  - (Short, well-synchronized pulses, low jitter)
- Probe energies in range 15 $\sim$ 80 eV “easily” achievable
  - Full Brillouin zone accessible
- Repetition rates $\sim$ hundreds of kHz available, even up to MHz
  - (High repetition rates improve signal-to-noise and acquisition times)

Free electron lasers (FELs)

- Probe energies up to hundreds of eV, or higher

CDWs & Mottism in 1T Transition Metal Dichalcogenides (TMDCs)

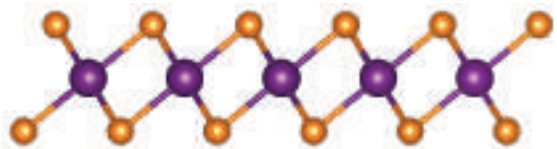
H																He
Li	Be											B	C	N	O	F	Ne
Na	Mg		IV	V								Al	Si	P	**S**	Cl	Ar
K	Ca	Sc	**Ti**	**V**	Cr	Mn	Fe	**Co**	**Ni**	Cu	Zn	Ga	Ge	As	**Se**	Br	Kr
Rb	Sr	Y	**Zr**	**Nb**	**Mo**	**Tc**	Ru	**Rh**	**Pd**	Ag	Cd	In	Sn	Sb	**Te**	I	Xe
Cs	Ba	La - Lu	**Hf**	**Ta**	**W**	**Re**	Os	**Ir**	**Pt**	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac - Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fl	Uup	Lv	Uus	Uuo

MX_2
M = transition metal
X = chalcogen

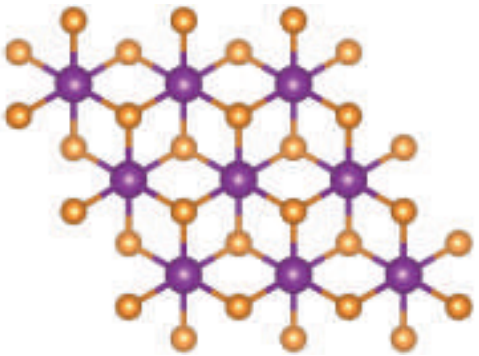
1T (octahedral) layer structure:

Top

Side

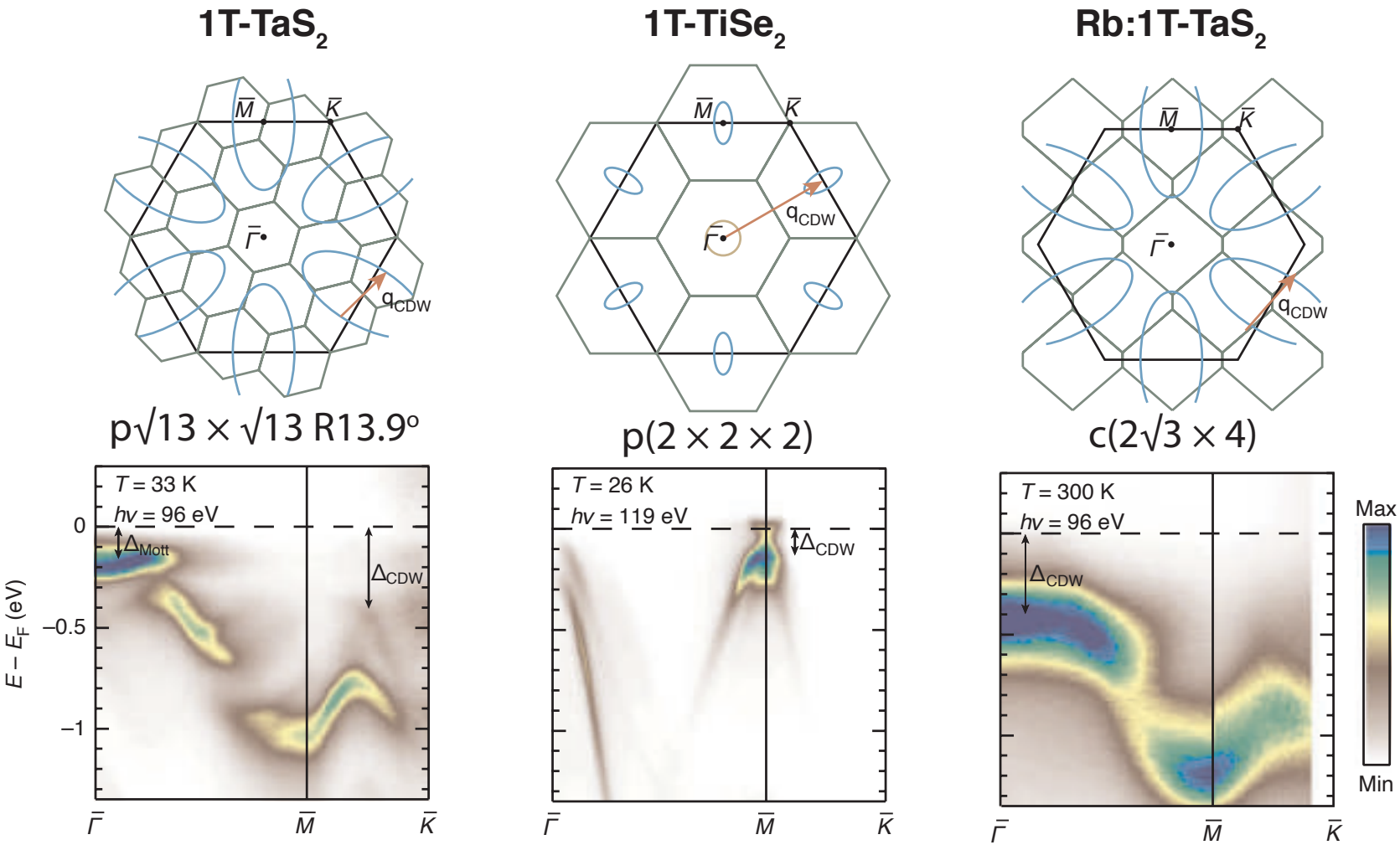


transition metal
chalcogen



Nat. Chem. **5** (2013) 263

Multiple Competing Instabilities: Bulk TaS_2, $TiSe_2$



Hellmann, *et al., Nat. Comm.* **3** (2012) 1069

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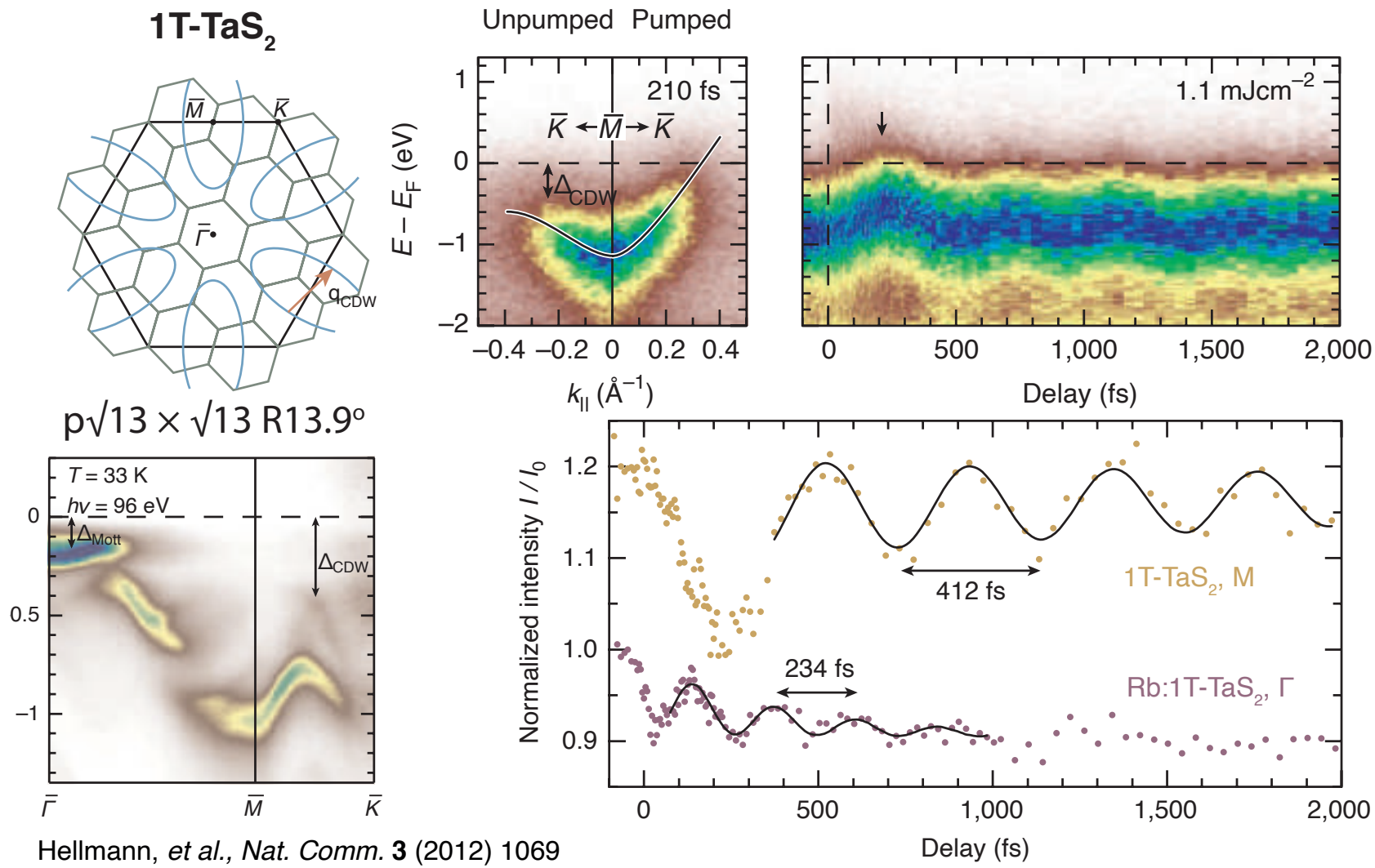

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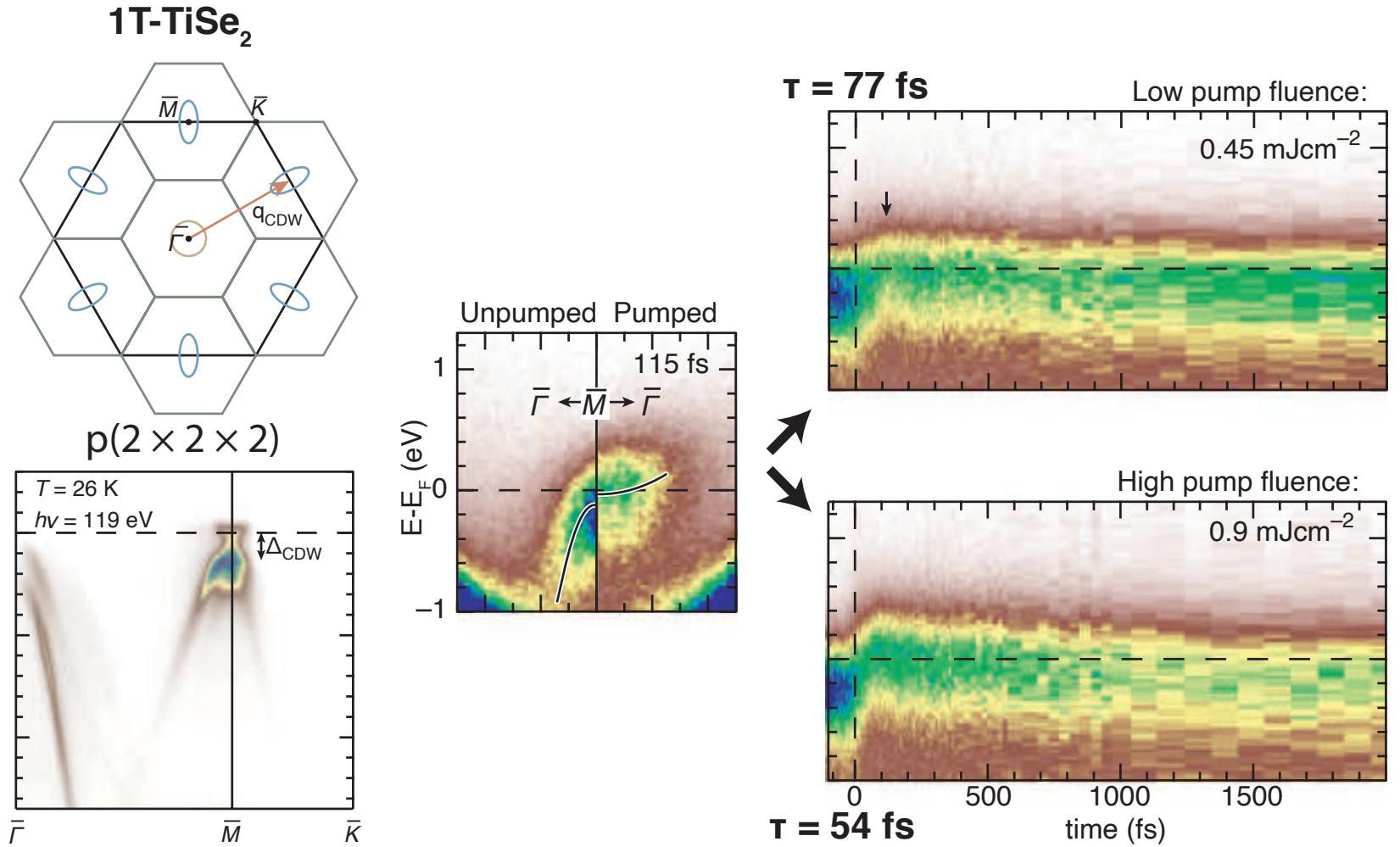

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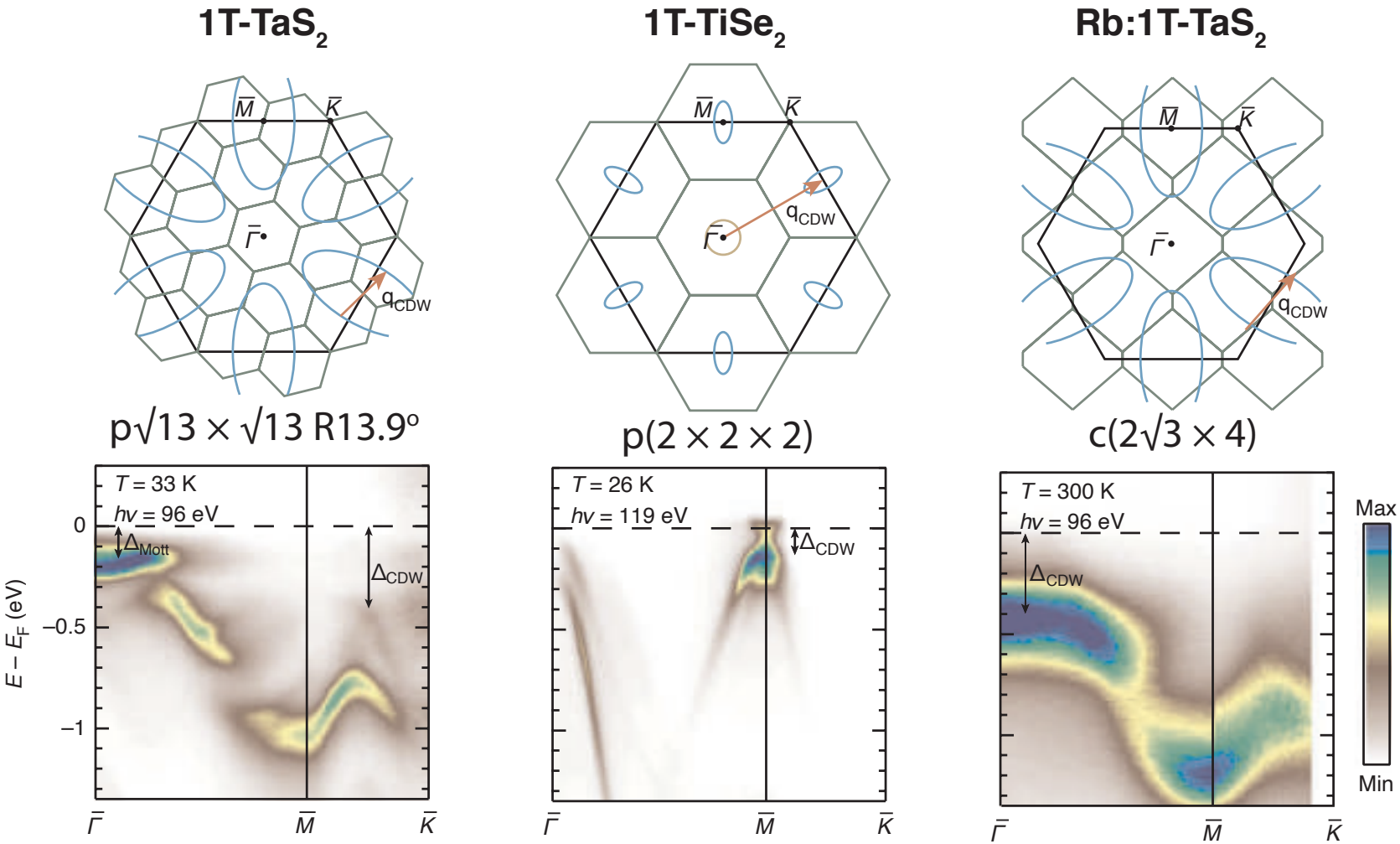

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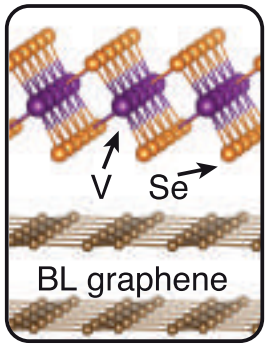
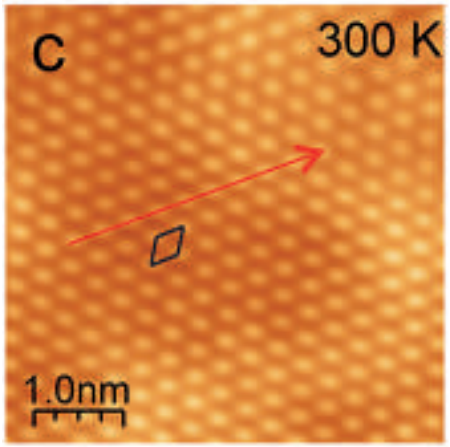
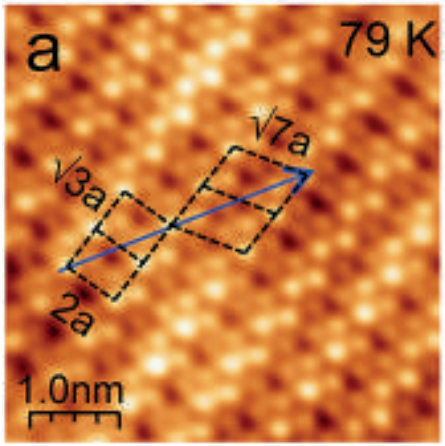
Long-Time Dynamics & Metastable States: Single-Layer VSe_2

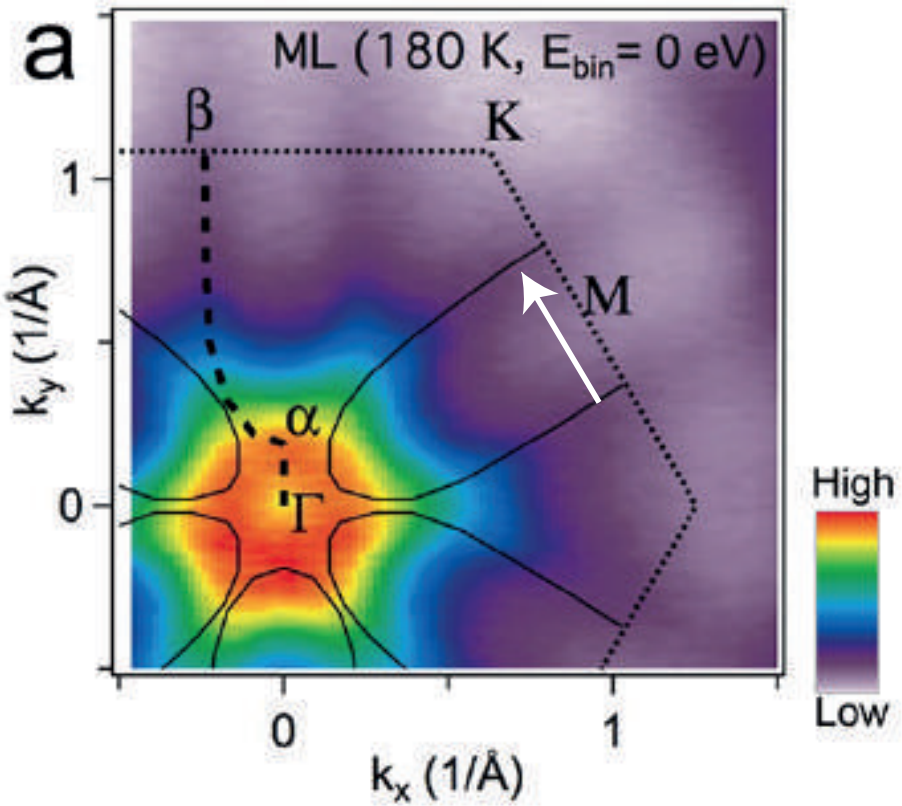




← weak 4a CDW distortion up to 350K (?)
(*cf. 4ax4ax3c in bulk, T_{CDW}=105K*)



further phase change at 135K, with new distortion(s) and full gapping of Fermi surface



Duvjir, *et al., Nano Lett.* **18** (2018) 5432

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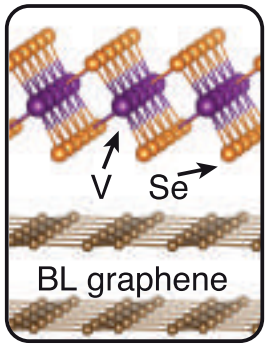

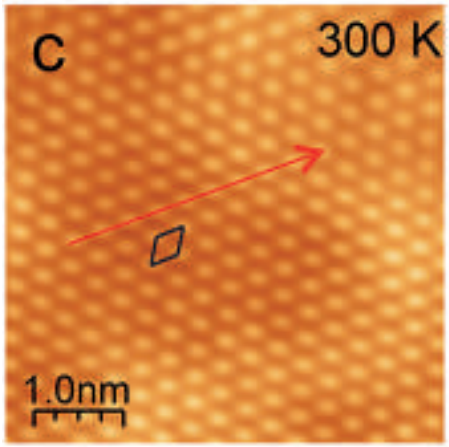

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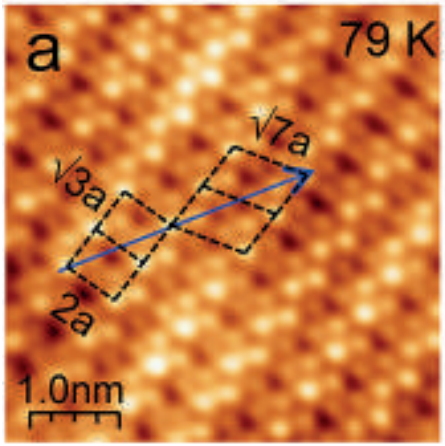

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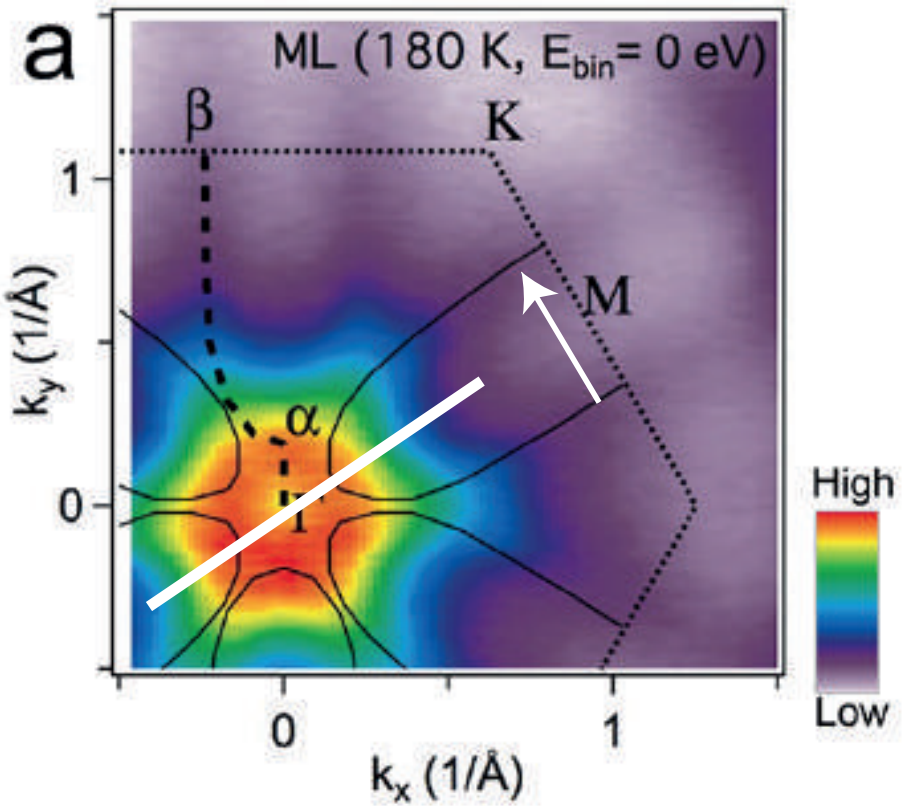

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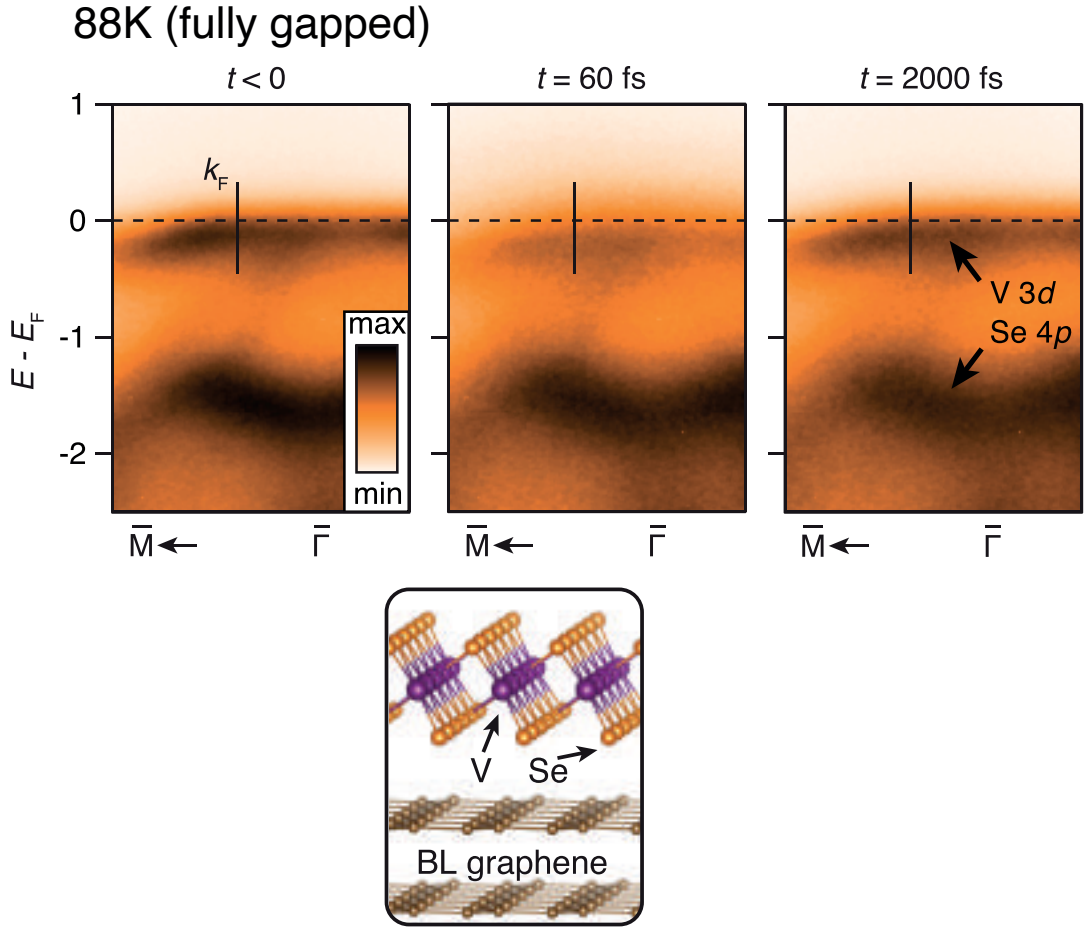

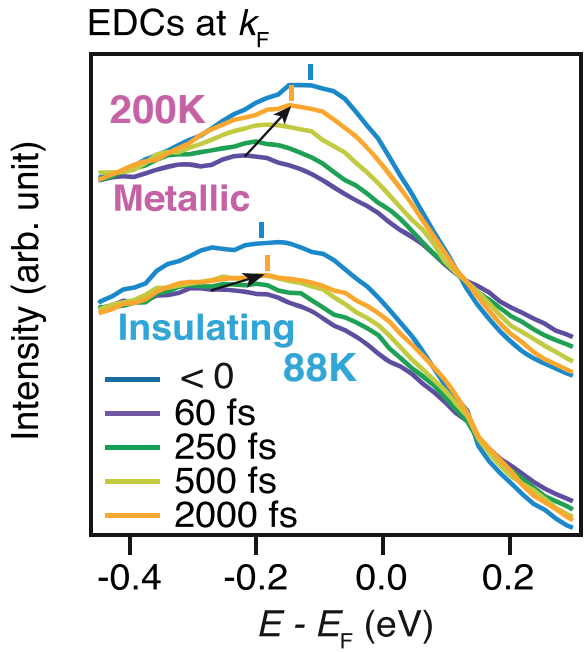

Biswas, *et al., Nano Lett.* **21** (2021) 1968

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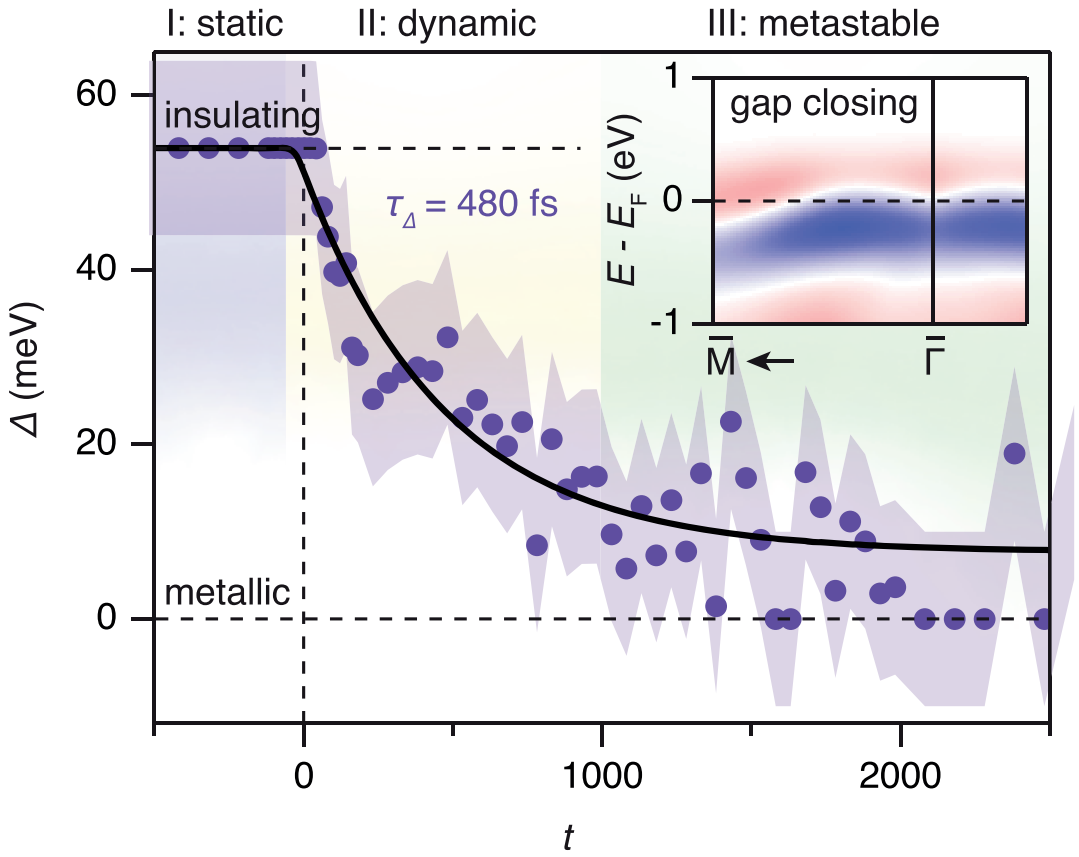
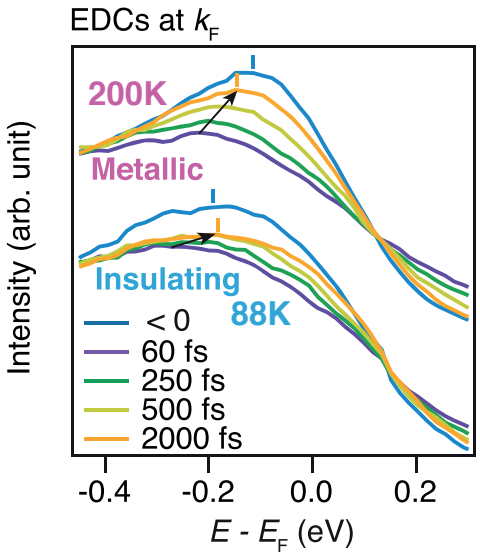
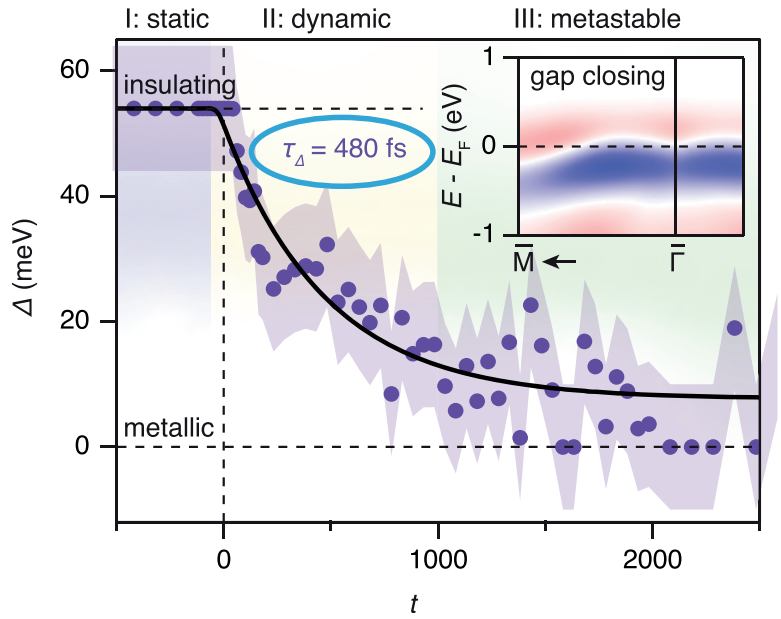




Long-Time Dynamics & Metastable States: Single-Layer VSe_2

Not an electronic process:

Mott physics excluded



1. Hot electrons decay via high-energy optical phonons
2. Optical phonons decay anharmonically
3. Lattice thermalizes at new temperature higher than the T_C of the collective ground state

What is the current state of the art?

Pump-probe “time-resolved” photoemission is a powerful tool for understanding correlated states and their mechanisms

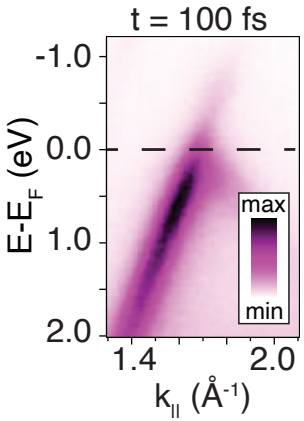
What is the direction of the technological development?

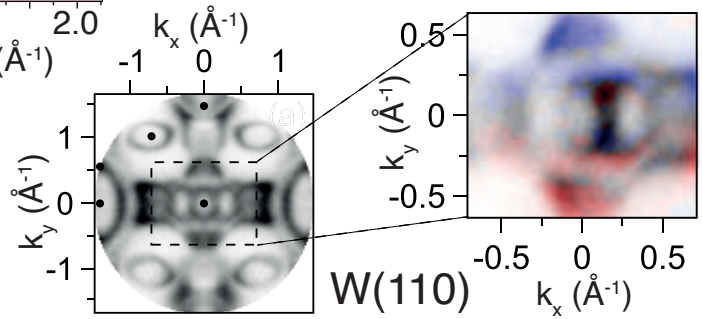
Spectrometer Technology

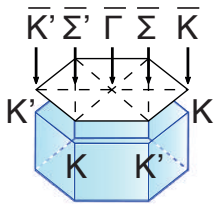
Hemispherical

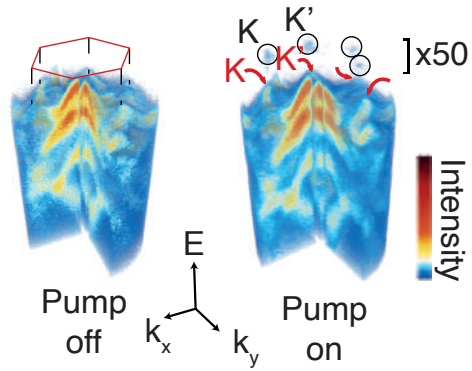
FeSuMa

Momentum microscopy/
photoelectron emission microscopy









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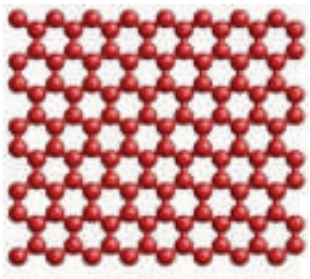
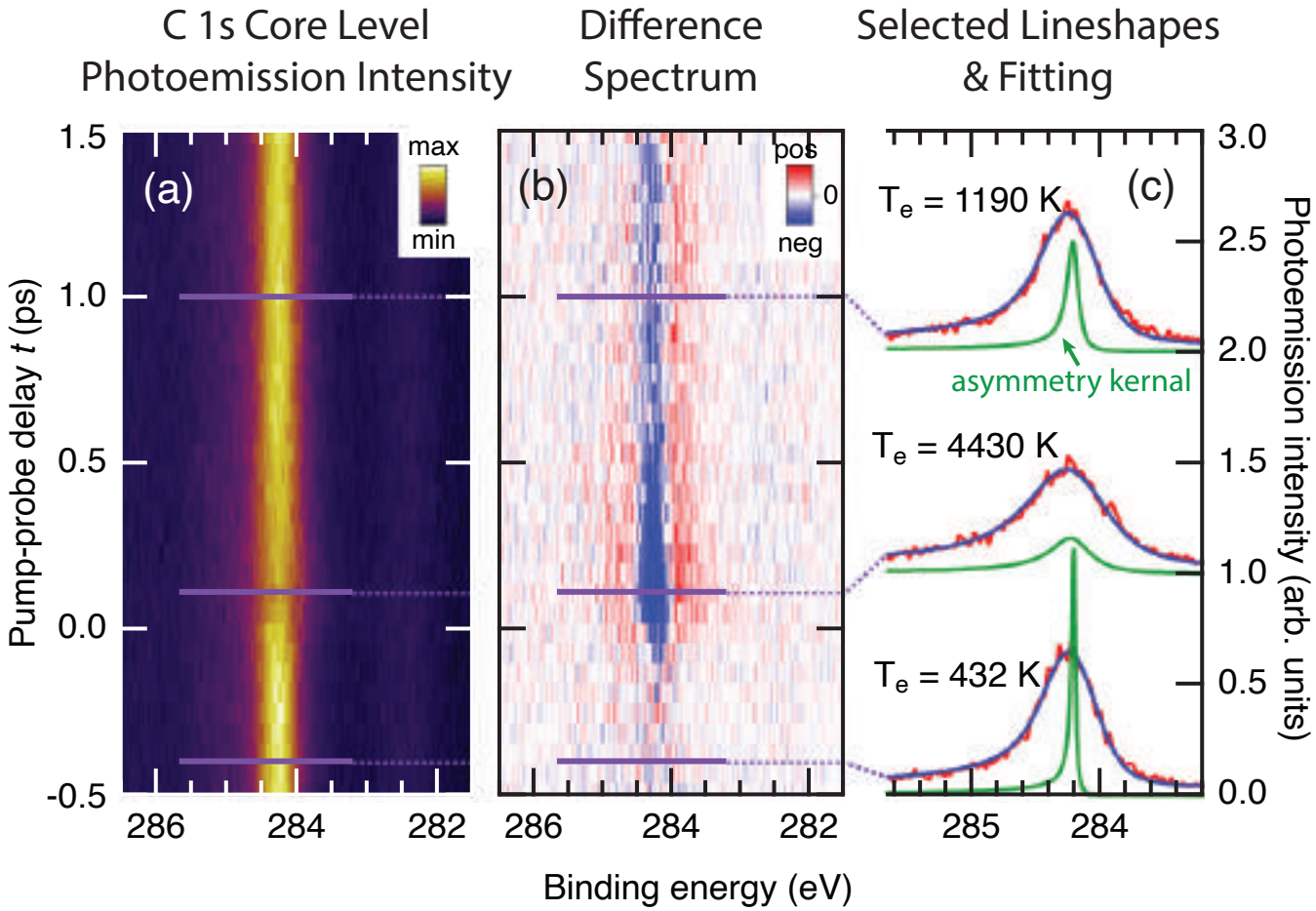
Pulsed-Laser Technology

- High repetition rates
- Increasing stability and ease of use
- Pulse lengths down to attosecond regime
- Expanded probe energy range
  - Challenge for tabletop HHG generation: energy resolution
  - Soft x-ray ARPES, x-ray photoelectron spectroscopy (XPS), x-ray photoelectron diffraction (XPD)

Pump-Probe XPS

Graphene





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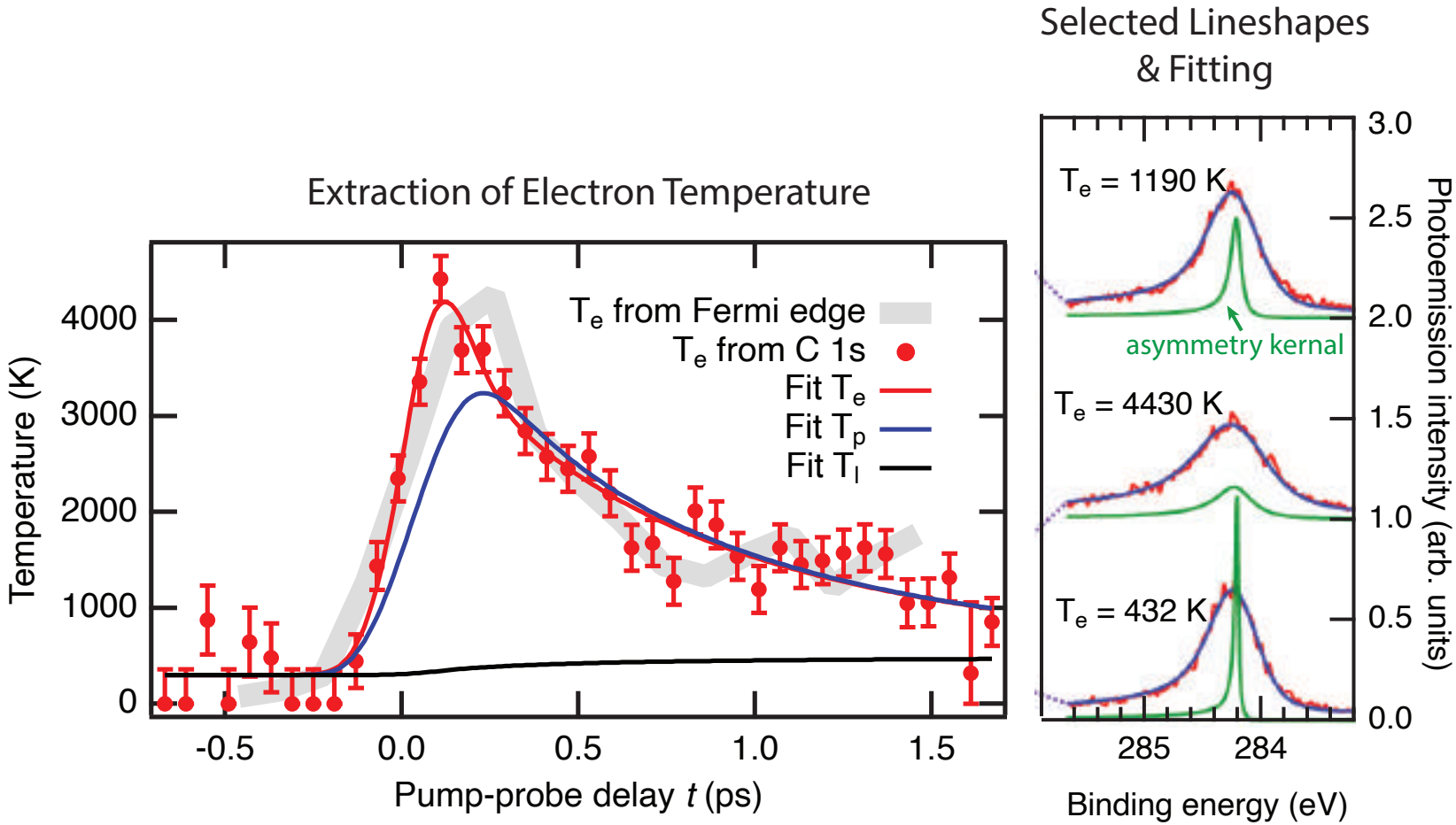
Pump-Probe XPS



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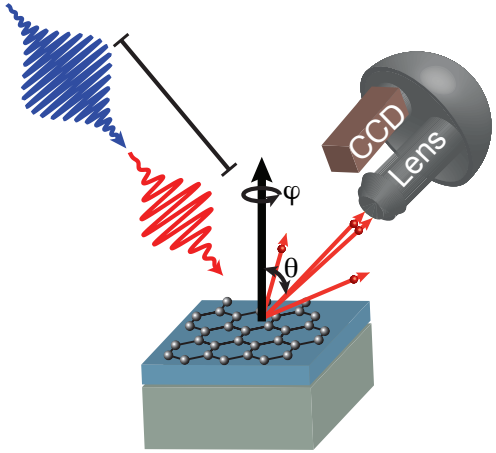
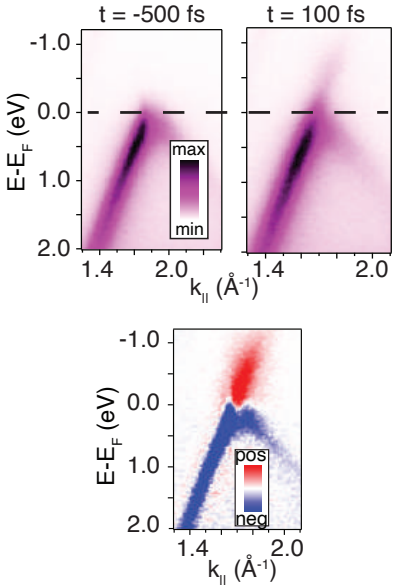
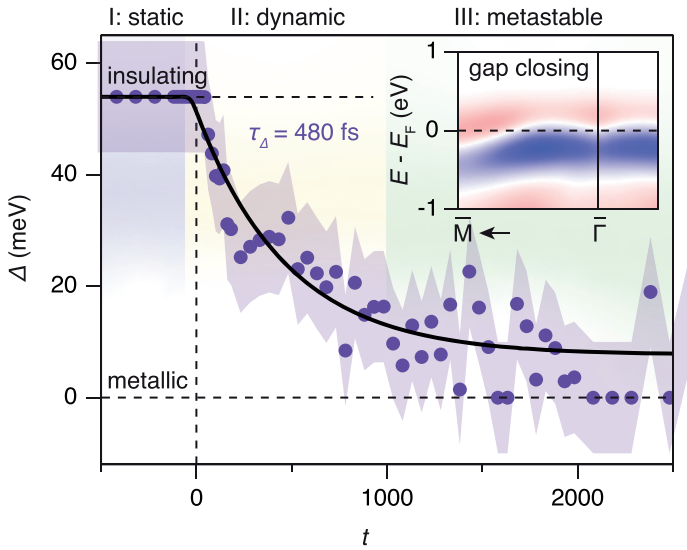
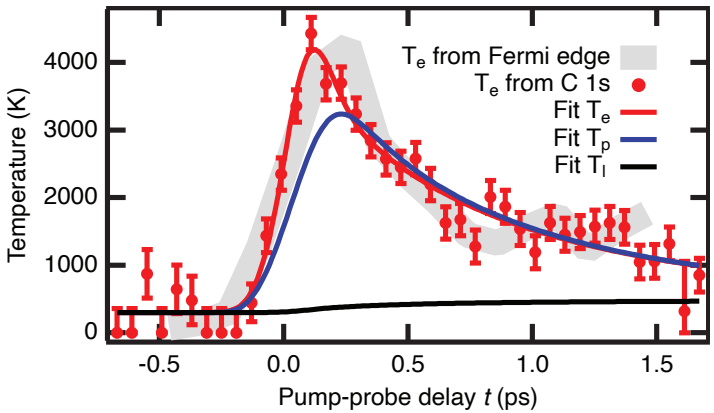
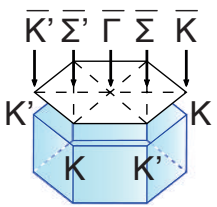
Summary

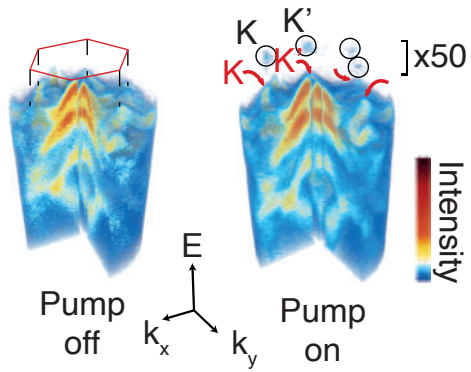

CCD
Lens
φ
θ


t = -500 fs
t = 100 fs
E-E_F (eV)
max
min
pos
neg
k_∥ (Å^-1)


I: static
II: dynamic
III: metastable
insulating
metallic
τ_Δ = 480 fs
gap closing
Δ (meV)
E - E_F (eV)
M̄
Γ̄
t


T_e from Fermi edge
T_e from C 1s
Fit T_e
Fit T_p
Fit T_l
Temperature (K)
Pump-probe delay t (ps)


K̄' Σ̄' Γ̄ Σ̄ K̄
K'
K
K
K'


K
K'
K
K'
x50
Intensity
E
k_x
k_y
Pump off
Pump on