

# Ultrafast Unordering\* of Electronic Order\*\*

\*Unordering = coherent and/or incoherent; disordering = incoherent.

\*\*Mostly charge-density waves.

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Ruprecht Haensel Laboratory

Kiel Nano, Surface and Interface Science KiNSIS

Kiel University

Deutsches Elektronen-Synchrotron DESY



# A special memory of a beautiful place

Cargèse, August 2022

Day 1



Day 2



Day 4



Day 4+

**Power failure  
(not only WiFi)**



# Ruprecht Haensel Laboratory

Superposition of DESY & CAU Kiel

$$\frac{1}{\sqrt{2}} |\text{DESY}\rangle + \frac{1}{\sqrt{2}} |\text{CAU}\rangle$$

Nanoscopic & femtostroboscopic soft x-ray spectroscopy



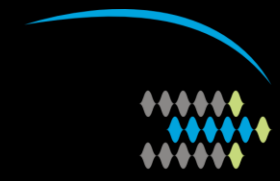
Interdisciplinary nanoscience



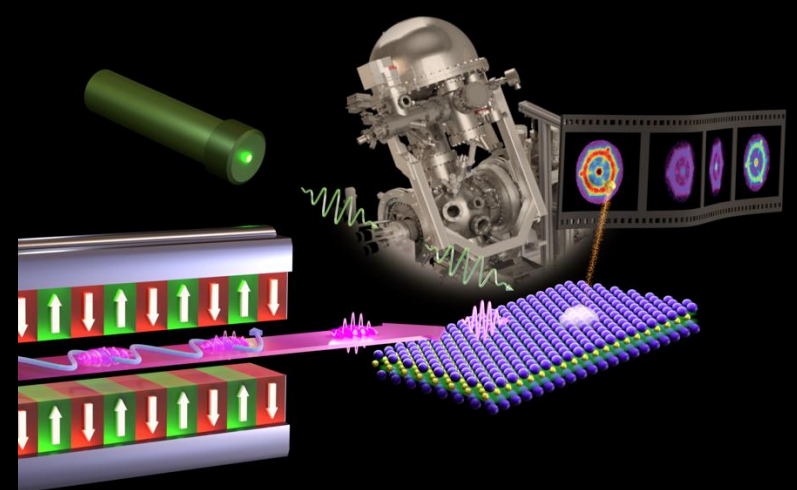
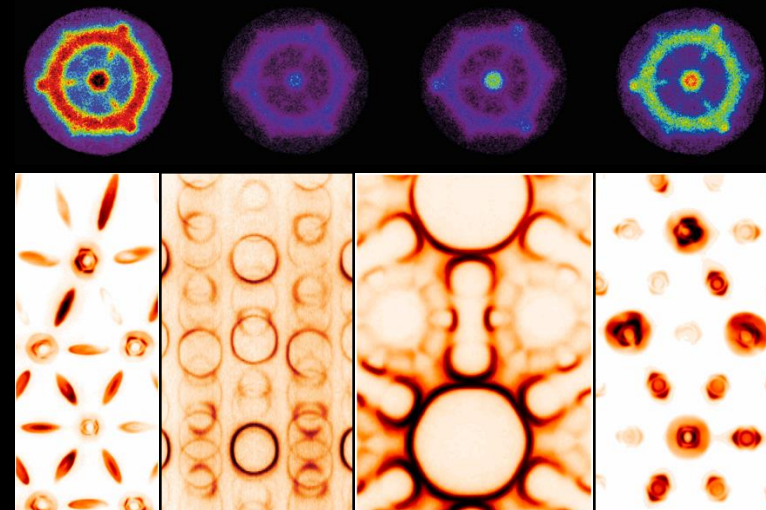
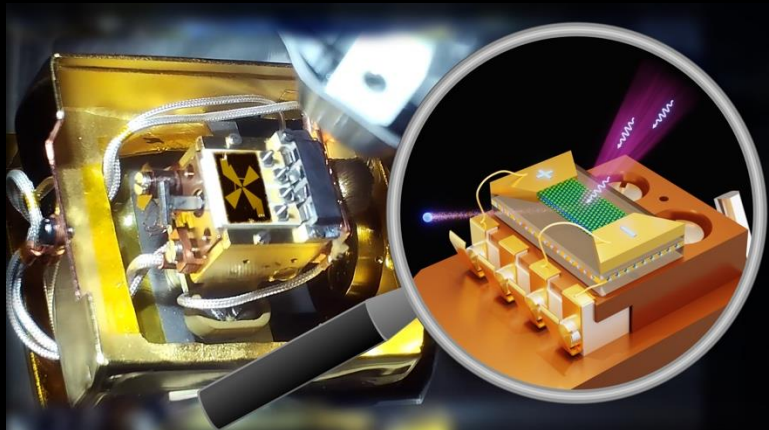
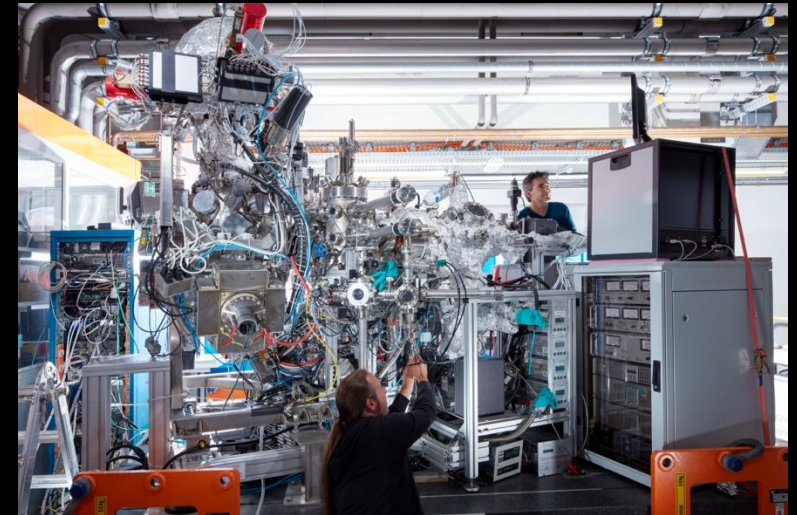
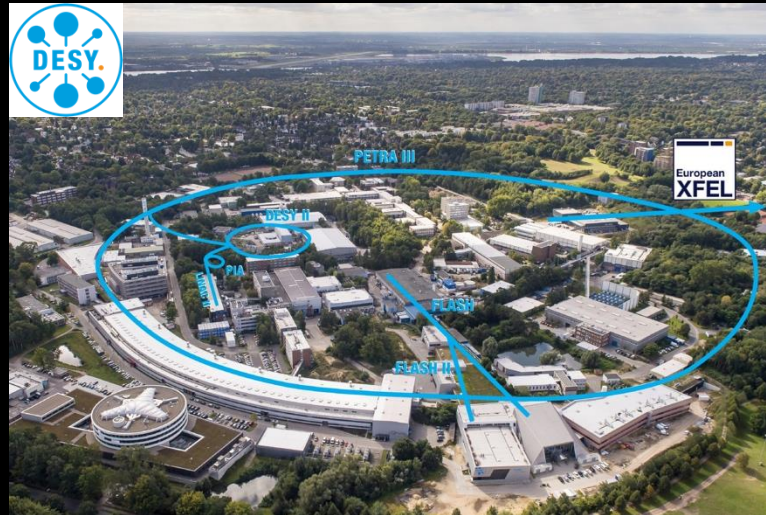
Kiel University  
Christian-Albrechts-Universität zu Kiel

# Soft X-ray Spectroscopy of Quantum Materials

Imaging electrons in action on nanometer length and femtosecond time scales



RUPRECHT  
HAENSEL  
LABOR



# Outline

... a reflection of my (narrow) world view

## Ultrafast Unordering of Electronic Order

### I. Motivation

Quest for a new switch.

### II. Notes on charge-density waves

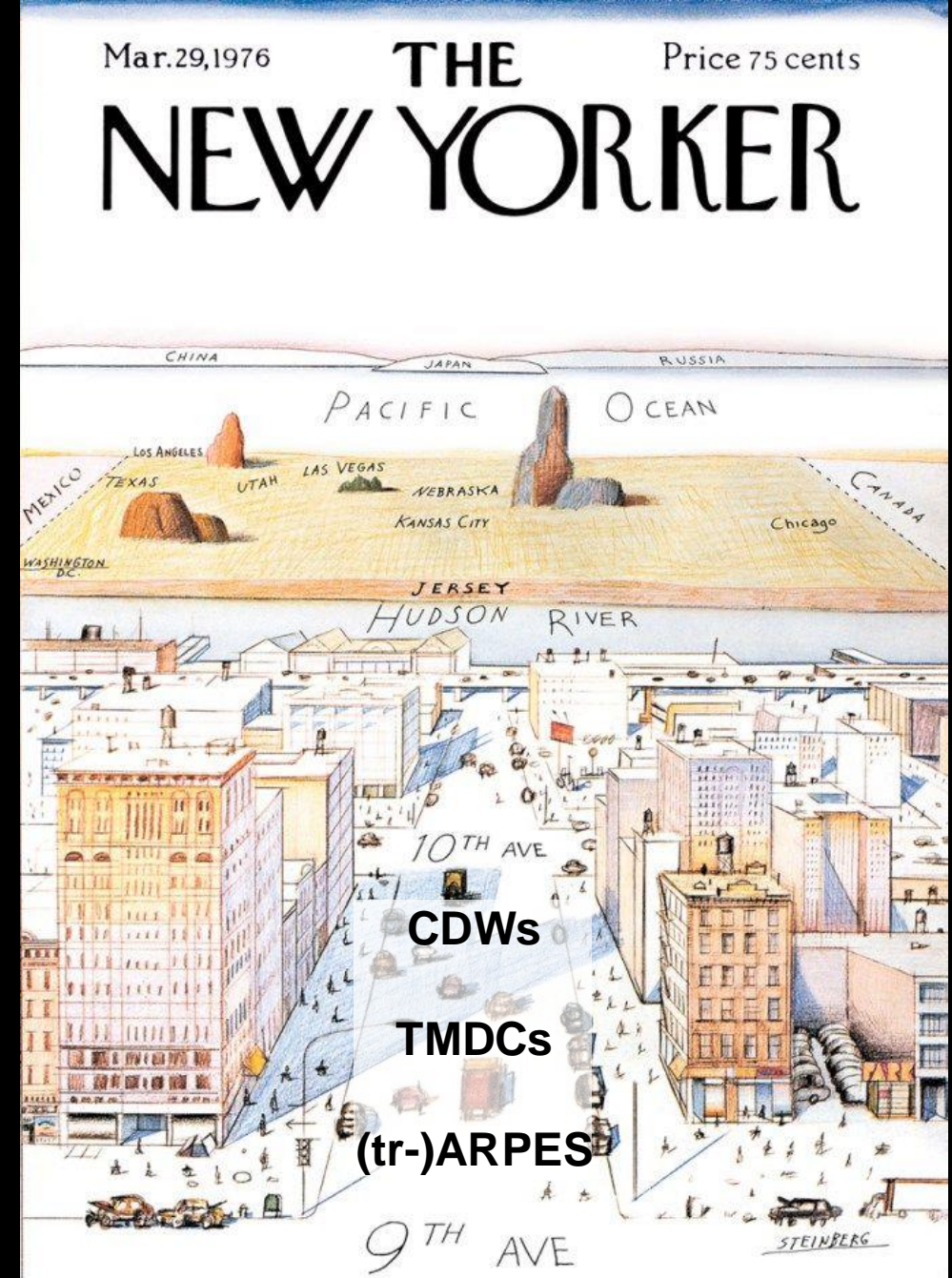
Nature & origin (“types”).

### III. Ultrafast unordering (of CDWs)

Mechanisms. Speeds. Efficiencies.  
Time-domain classification.

### IV. Outlook

Tools & concepts.



# What has humanity produced in the greatest quantity?

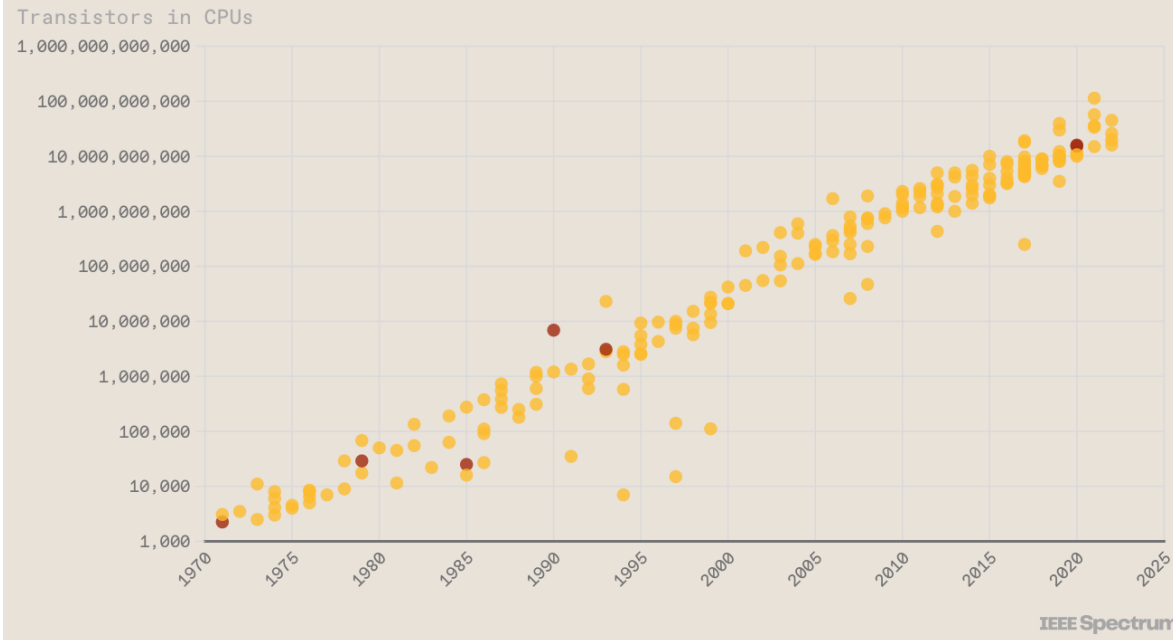
In what quantity?

IEEE Spectrum: *The State of the Transistor in 3 Charts* (2022)

Moore's law: Doubling of transistor count every 1.5-2 years

Semiconductor industry:  $\approx 10^{22}$  transistors / 76 a

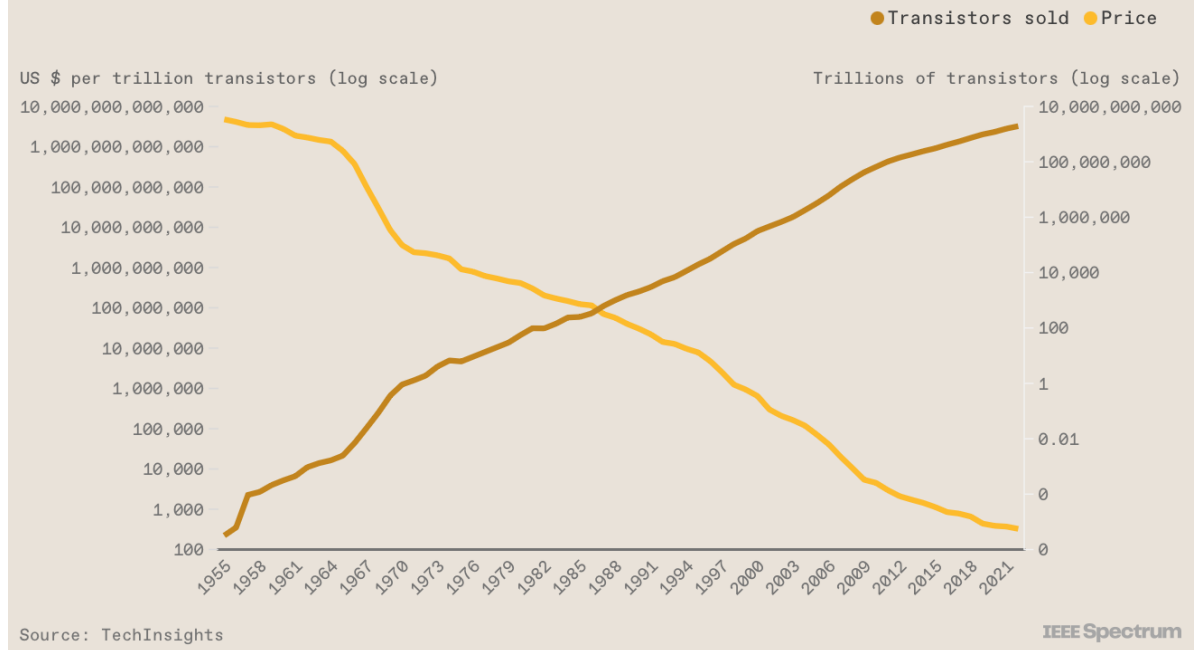
## Transistors per Processor



Average growth rate:  $\approx 40\%$  per year

## Huge Volume, Small Price

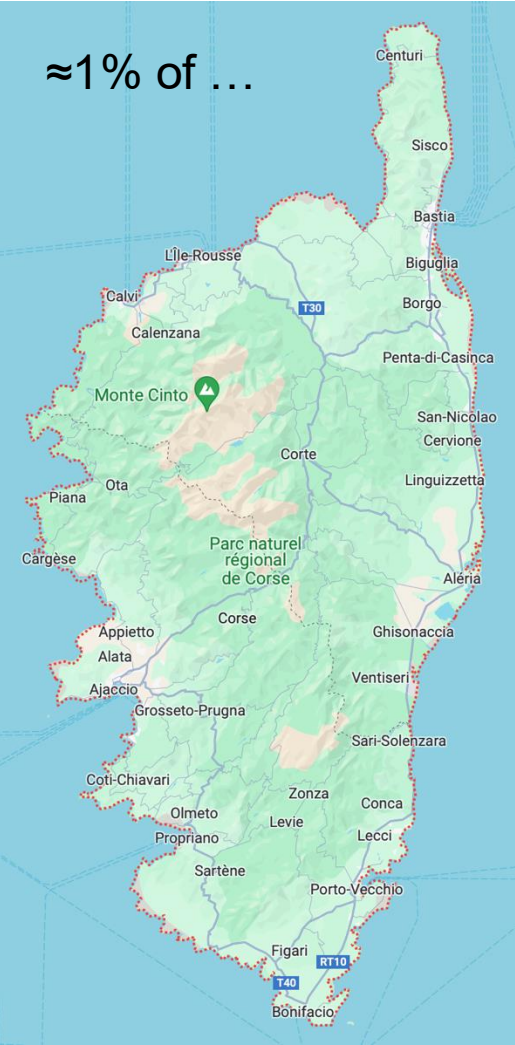
Price per trillion transistors, US \$; transistors sold per year, trillions



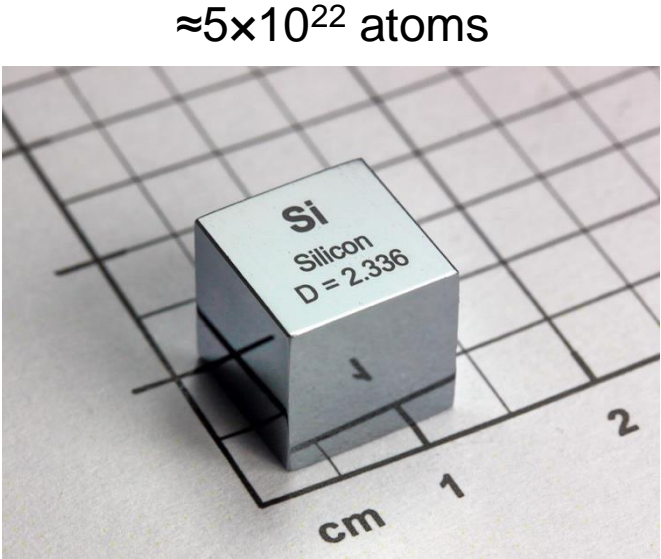
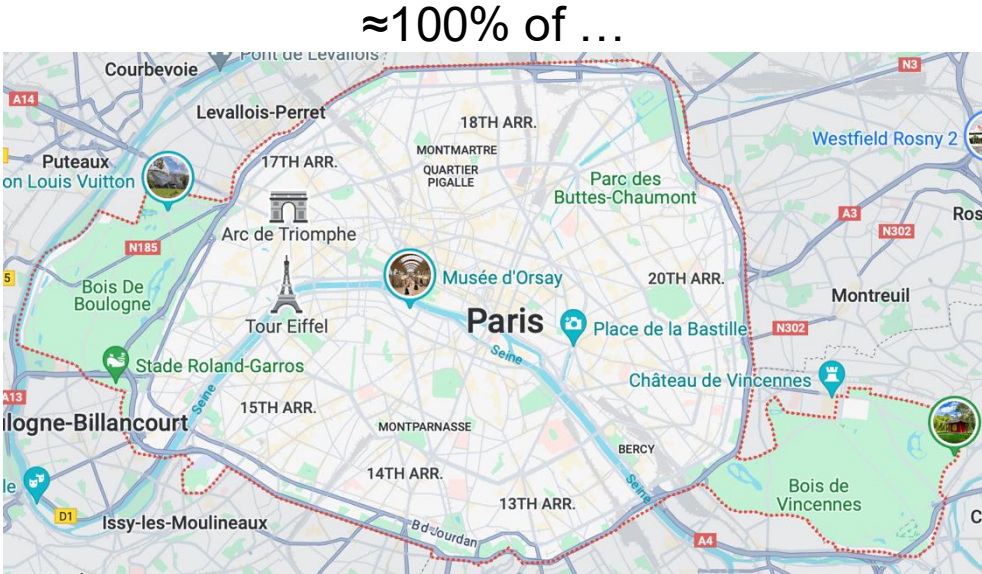
Evolution:  $\approx 10^{40}$  cells / 4 Ga

# Many small things can have a big footprint

## Visualization of $10^{22}$ transistors

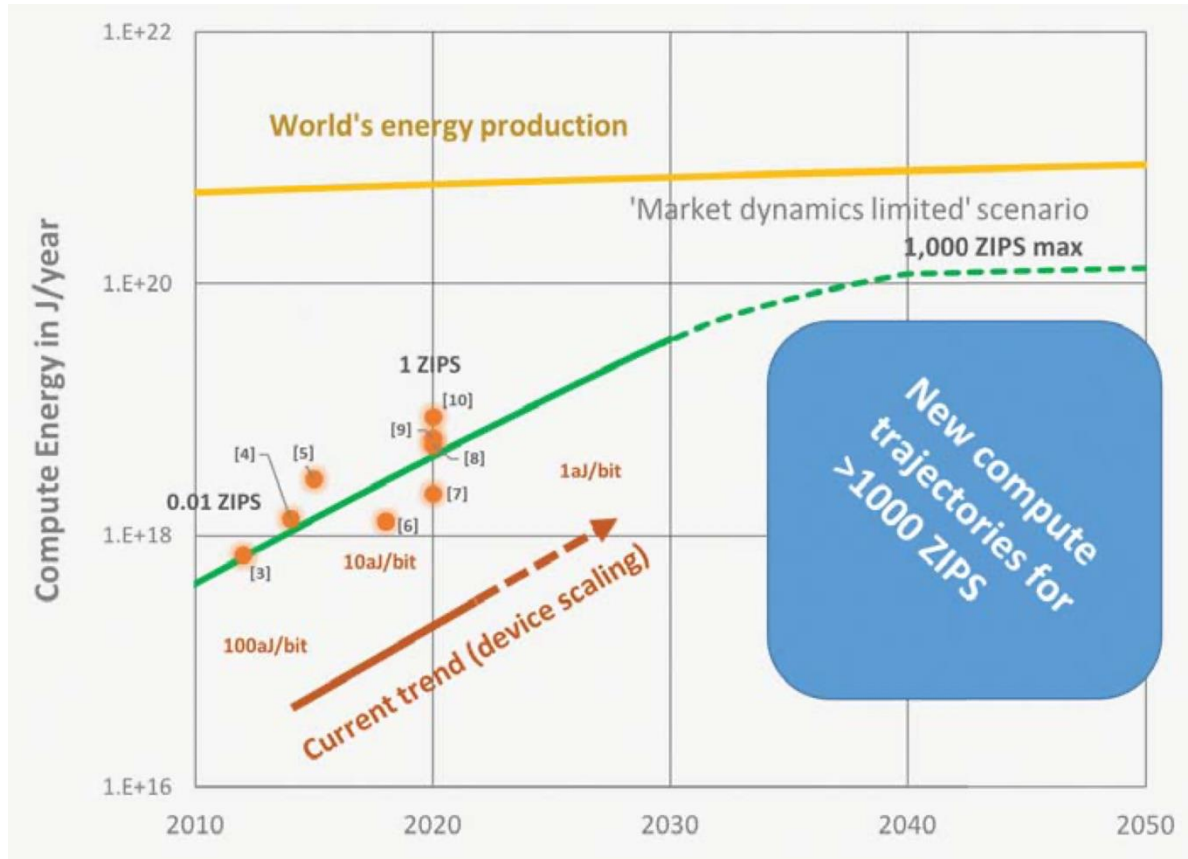


$$10^{22} \text{ transistors} \times (100 \times 100) \text{ nm}^2 \text{ area per transistor} = 100 \text{ km}^2$$



# Grand challenge

How to innovate our way out of informational meltdown?



$$\begin{aligned}
 & 10^{22} \text{ transistors} \times 10^{14} \text{ bit transitions} \times 10^{-17} \text{ J energy per transition} \\
 & = \boxed{10^{19} \text{ J per year}} \\
 & \approx 2\% \text{ of global primary energy production}
 \end{aligned}$$

Semiconductor Research Corporation: *Decadal Plan for Semiconductors* (2021)  
 Scharf, *Commentary: The rule of information*, *Physics Today* **75**(3), 10 (2022)



# Quest for a new switch: How fast?

Speed is relative ... and limited.

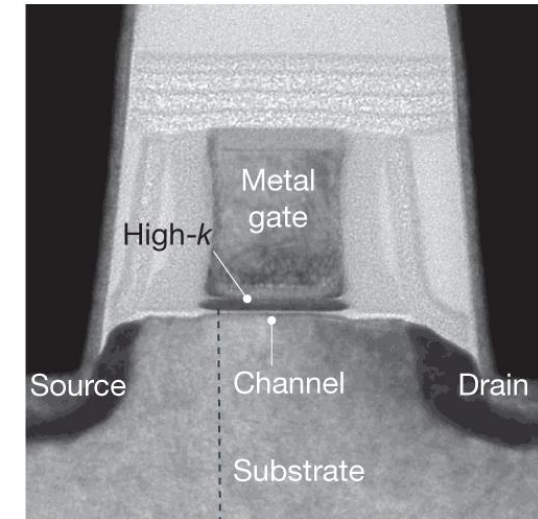
**Arion cronos**  
Disinfection system for soft contact lenses  
No preservatives  
Ultra-fast process (1 hour)  
Color indicator (Vitamin B2)

**Arion cronos**  
Système de désinfection pour lentilles de contact souples  
Sans conservateurs  
Processus ultrarapide (1 heure)  
Indicateur de couleur (Vitamine B2)

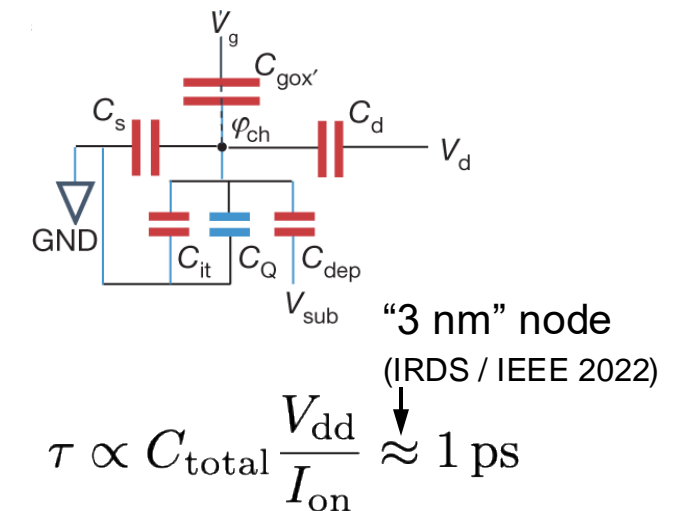
**Arion cronos**  
Desinfektionssystem für weiche Kontaktlinsen  
Ohne Konservierungsstoffe  
Komplette Reinigungszeit (1 Stunde)  
Enthält Vitamin B2 (Indikator)

**Arion cronos**  
Sistema di disinfezione per lenti di contatto morbide  
Senza conservanti  
Processo ultrarapido (1 ora)  
Indicatore di colore (Vitamine B2)

**DISOP**  
360 ml + 36 comp

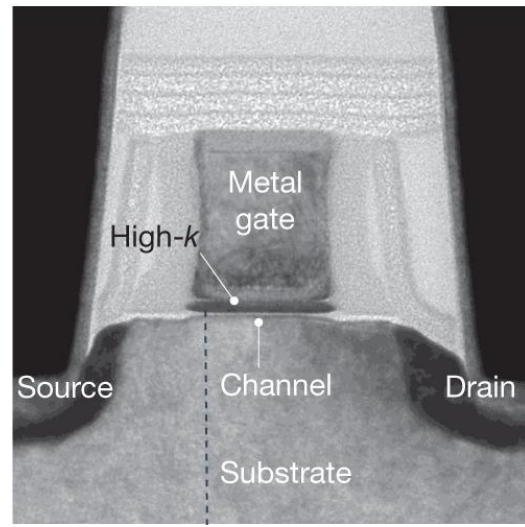
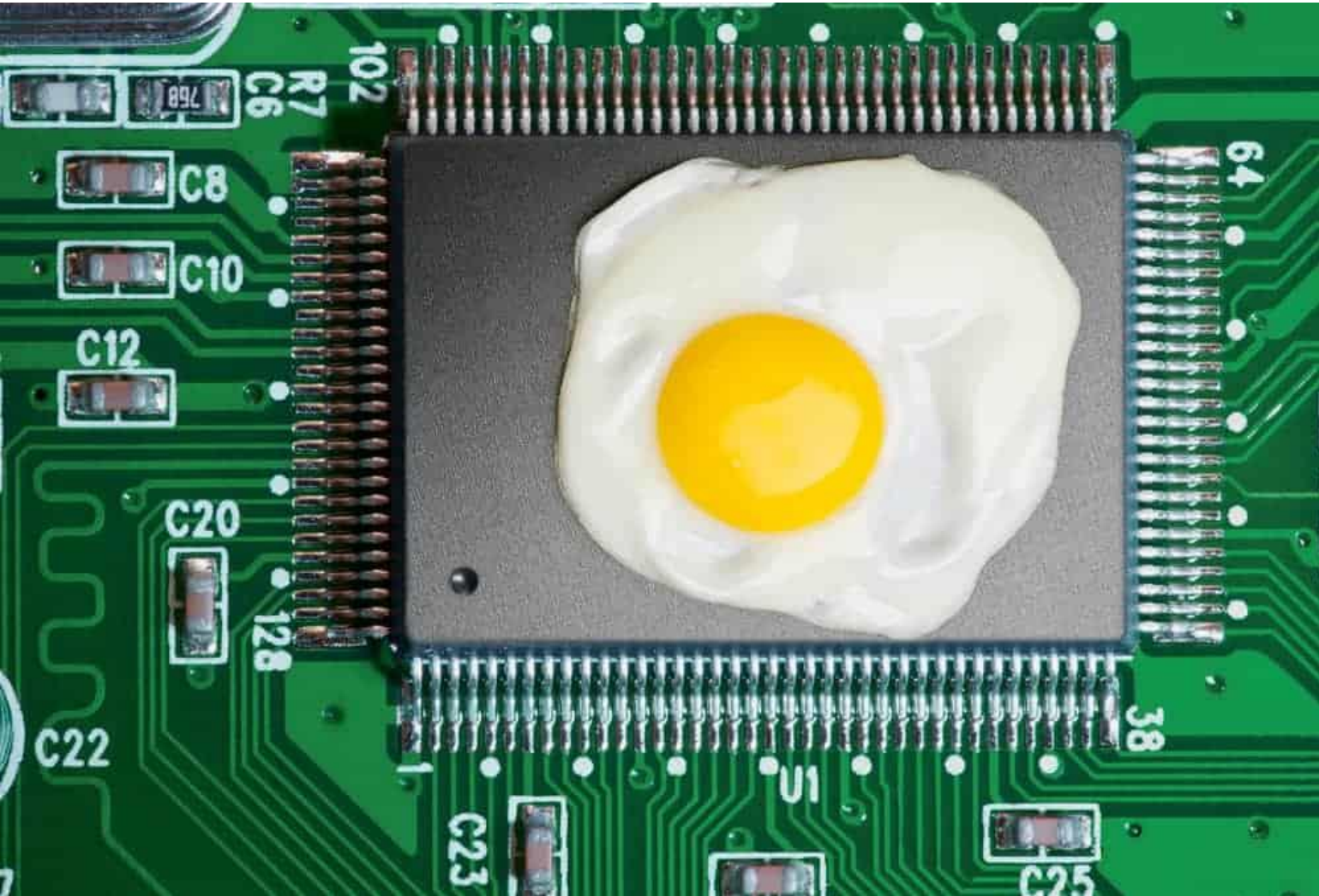


Cao et al., Nature 620, 501 (2023)

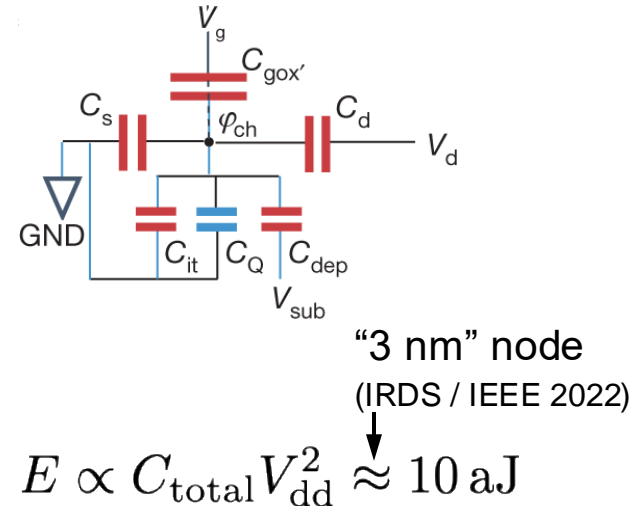


# Quest for a new switch: How efficient?

Efficiency is the avoidance of unwanted excitations.



Cao et al., Nature 620, 501 (2023)

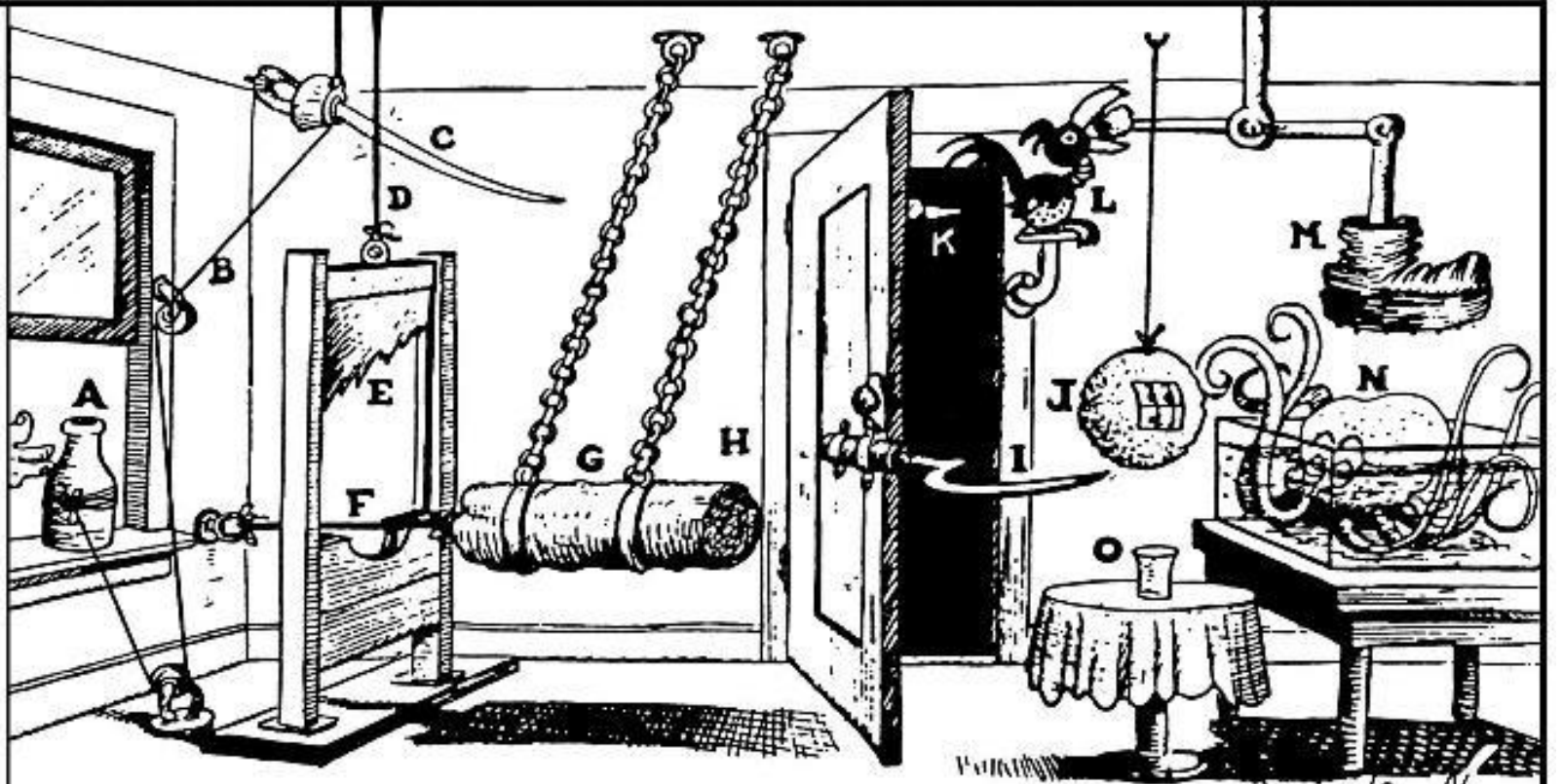


# Quest for a new switch: How exactly?

Mechanisms can be complicated ... and need not be universal.

## Simple Orange Squeezing Machine

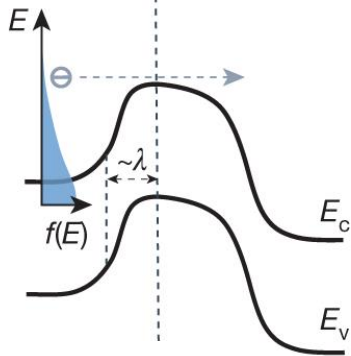
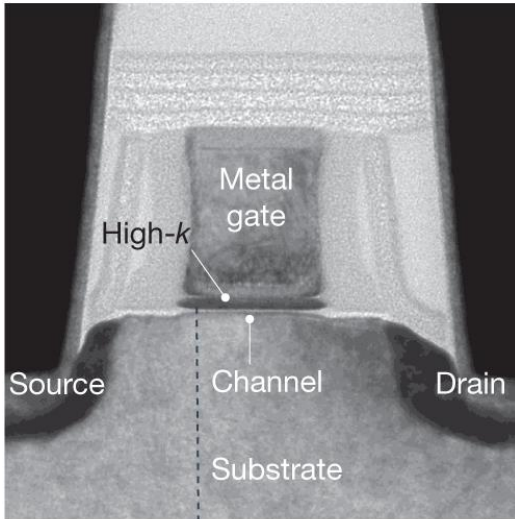
PROFESSOR BUTTS STEPS INTO AN OPEN ELEVATOR SHAFT AND WHEN HE LANDS AT THE BOTTOM HE FINDS A SIMPLE ORANGE SQUEEZING MACHINE. MILK MAN TAKES EMPTY MILK BOTTLE (A) PULLING STRING (B) WHICH CAUSES SWORD (C) TO SEVER CORD (D) AND ALLOW GUILLOTINE BLADE (E) TO DROP AND CUT ROPE (F) WHICH RELEASES BATTERING RAM (G). RAM BUMPS AGAINST OPEN DOOR (H) CAUSING IT TO CLOSE. GRASS SICKLE (I) CUTS A SLICE OFF END OF ORANGE (J) AT THE SAME TIME SPIKE (K) STABS PRUNE HAWK (L). HE OPENS HIS MOUTH TO YELL IN AGONY, THEREBY RELEASING PRUNE AND ALLOWING DIVER'S BOOT (M) TO DROP AND STEP ON SLEEPING OCTOPUS (N). OCTOPUS AWAKENS IN A RAGE AND SEEING DIVER'S FACE WHICH IS PAINTED ON ORANGE, ATTACKS IT AND CRUSHES IT WITH TENTACLES, THEREBY CAUSING ALL THE JUICE IN THE ORANGE TO RUN INTO GLASS (O). LATER ON YOU CAN USE THE LOG TO BUILD A LOG CABIN WHERE YOU CAN RAISE YOUR SON TO BE PRESIDENT LIKE ABRAHAM LINCOLN.



© Rube Goldberg

# Quest for a new switch: How exactly?

## Thermionic electron emission across a barrier



Cao *et al.*, Nature **620**, 501 (2023)

Independent electrons:

$$I_{\text{off}} = I_{\text{on}} e^{-\frac{qV}{k_B T}}$$

$$\left(\frac{d[\log I_{\text{on}}]}{dV}\right)^{-1} = \left(\frac{q}{k_B T} \log e\right)^{-1}$$

$$= \frac{k_B T}{e} \frac{1}{\log e}$$

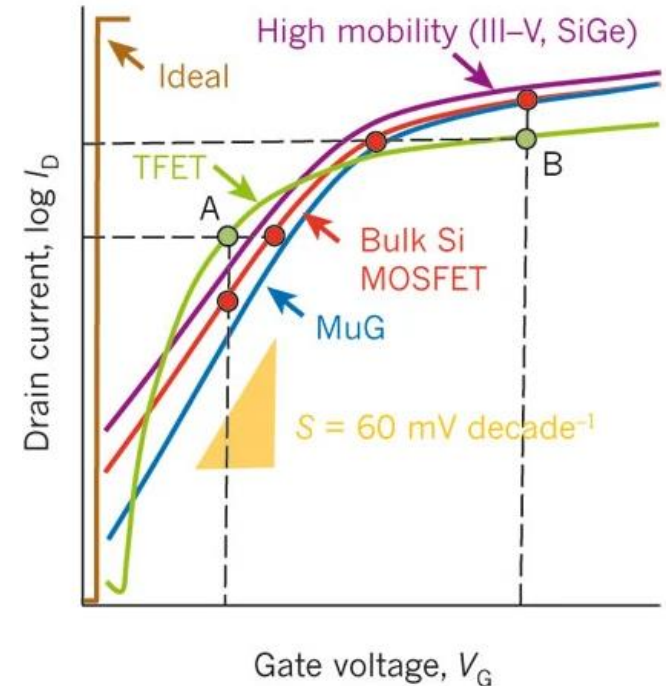
$$\stackrel{300 \text{ K}}{=} 60 \text{ mV/decade}$$

Collective electrons:

$$I_{\text{off}} = I_{\text{on}} e^{-\frac{QV}{2k_B T}}$$

$$\Delta E_{\text{on/off}} = \frac{1}{2} CV^2 = \frac{1}{2} QV, \quad Q \approx 100 e$$

$$\left(\frac{d[\log I_{\text{on}}]}{dV}\right)^{-1} = \frac{k_B T}{\frac{1}{2} Q} \frac{1}{\log e}$$

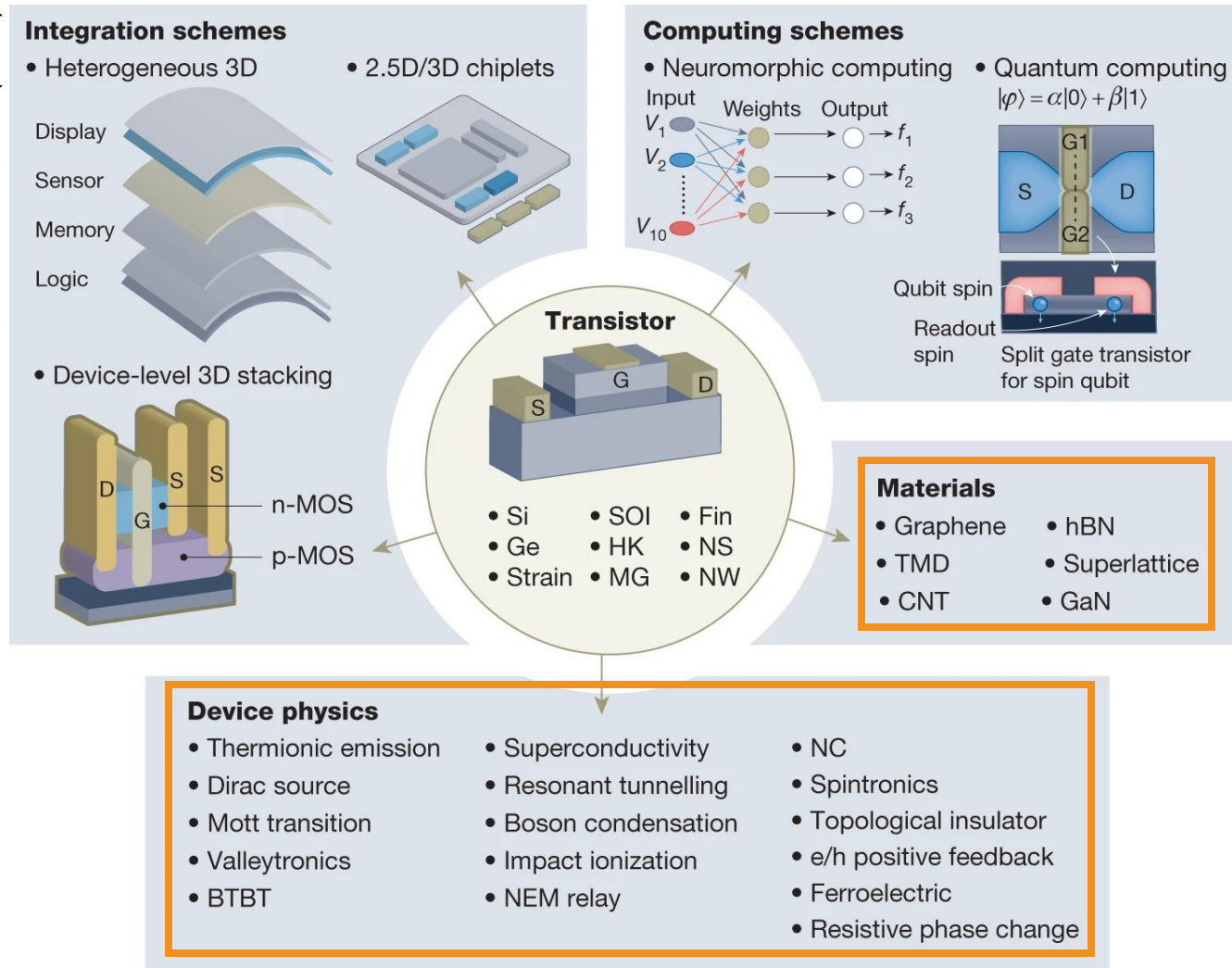


Ionescu & Riel, Nature **479**, 329 (2011)

# The quest for a new ultrafast, energy-efficient switch

A playground for quantum materials & electronic phase transitions

Cao et al., Nature 620, 501 (2023)



## How fast?

Beyond drift-diffusion.  
Sub-vibrational.  
<100 fs.

## How efficient?

Sub-Boltzmann.  
<60 meV/decade.

## How exactly?

Electronic. Correlated.  
Nonthermal.

## Quantum materials

(“where **electrons** do interesting and potentially useful quantum things”).

## Charge-density waves

(archetypal electronic order).

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... a reflection of my (narrow) world view

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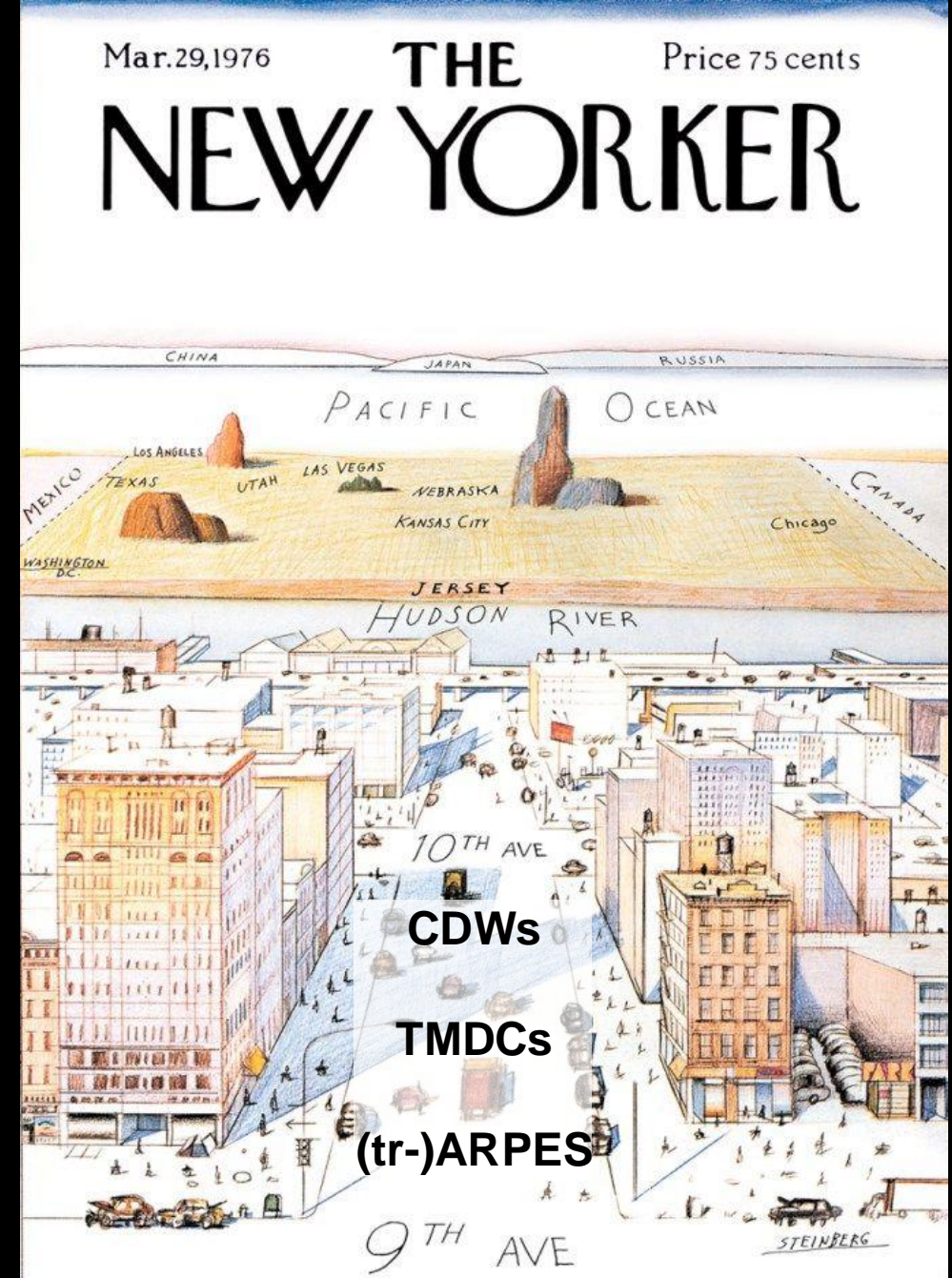
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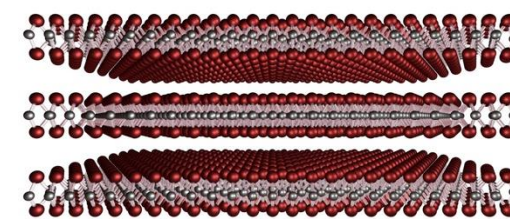
### IV. Outlook

Tools & concepts.

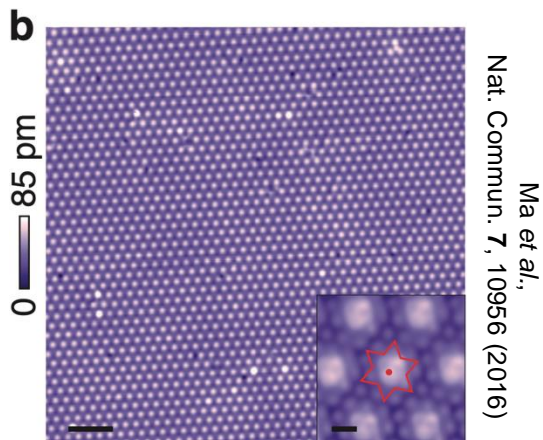


# What's a charge-density wave (CDW)?

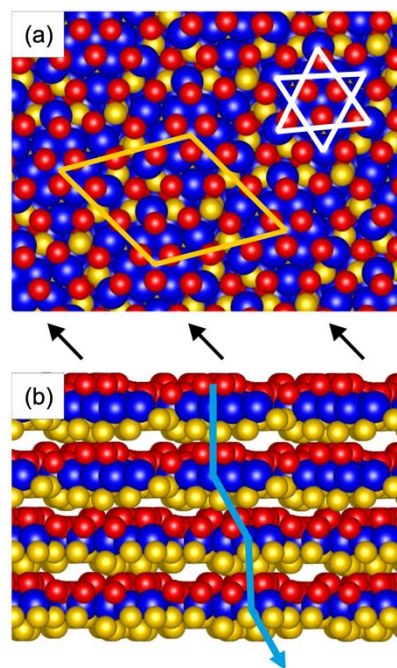
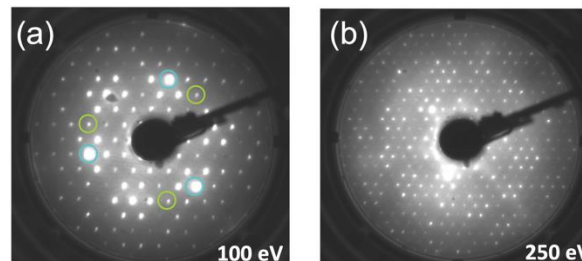
Three characteristic properties. Example: 1T-TaS<sub>2</sub>.



## Charge-density modulation

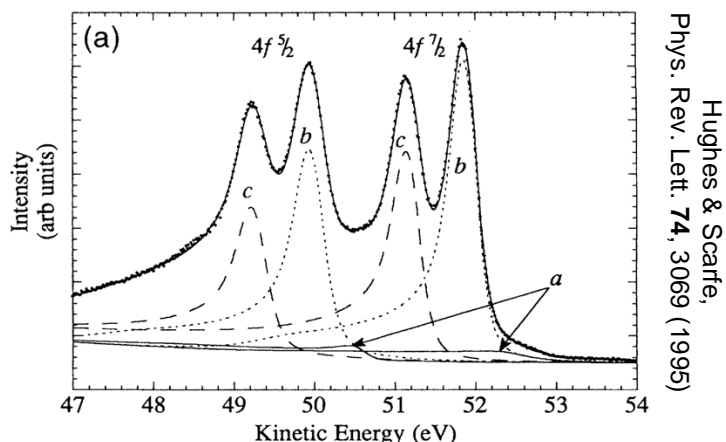
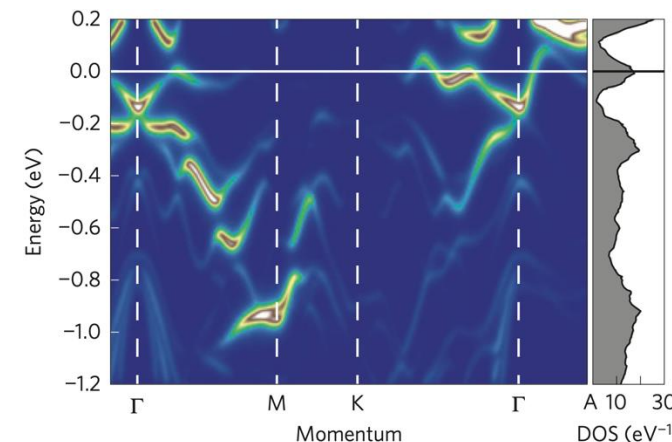
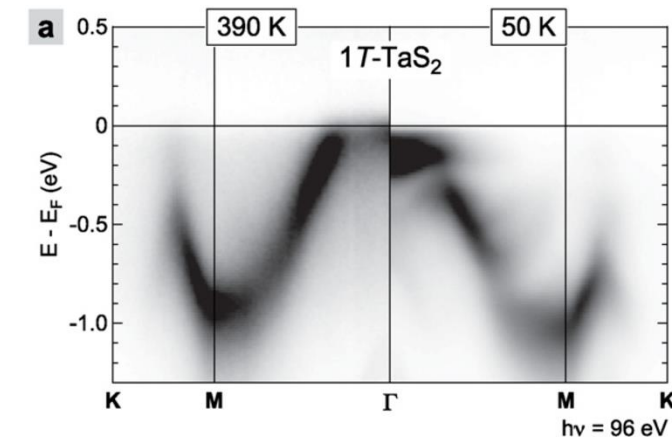


## Periodic lattice distortion



von Witte et al.,  
Phys. Rev. B **100**, 155407 (2019)

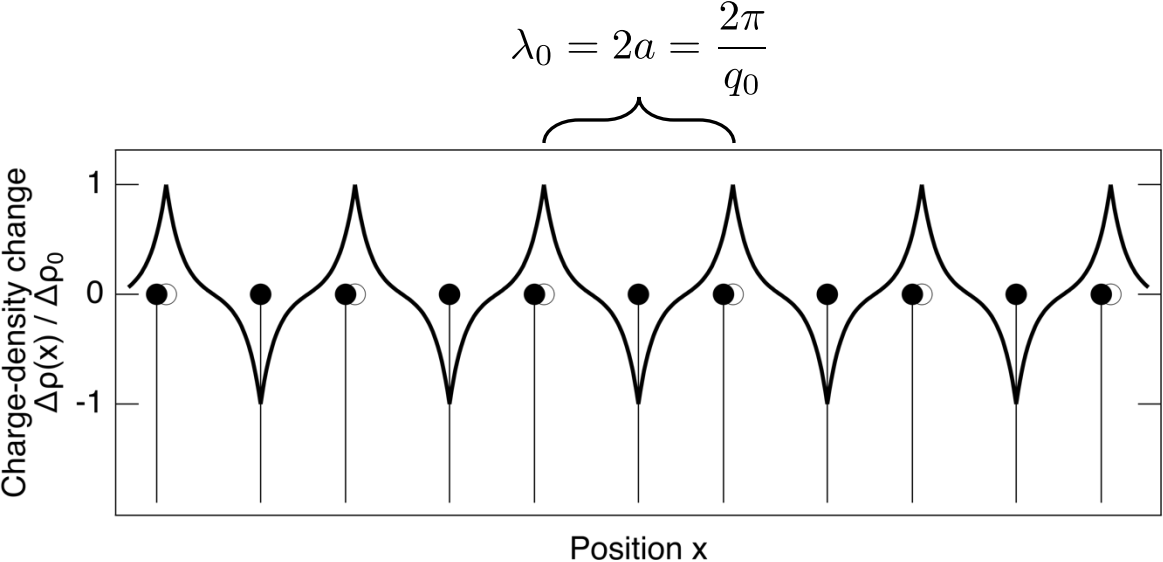
## Energy gap(s)



# Simple physical picture

## Kronig-Penney model with attractive Dirac comb

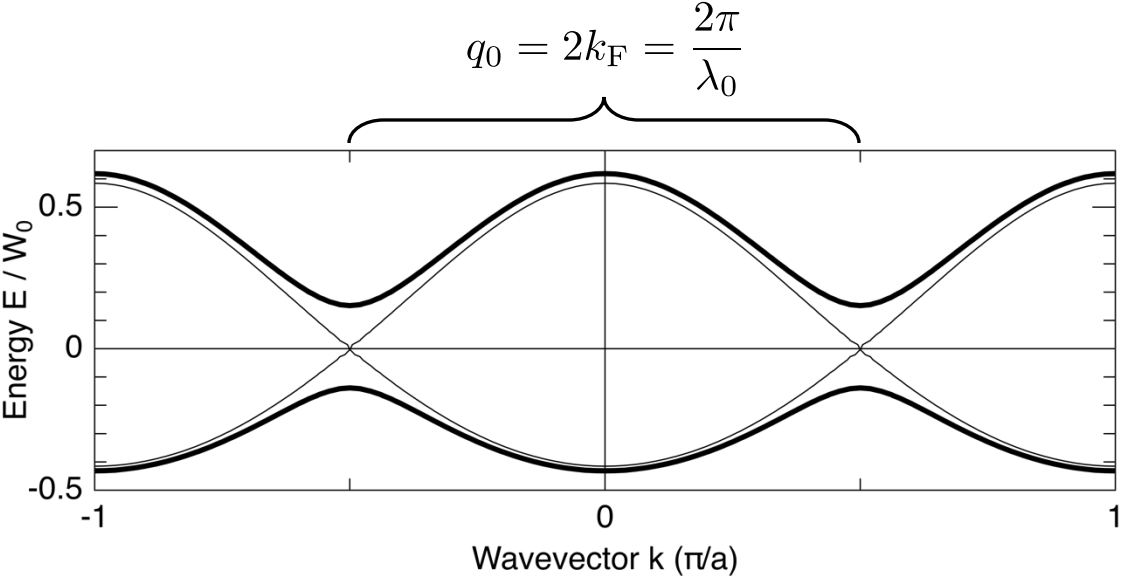
Periodic lattice distortion + charge-density modulation



$$\Delta E_{\text{latt.}} > 0$$

“passive”

Energy gap



$$\Delta E_{\text{electr.}} < 0$$

“active”

$$\Delta E_{\text{latt.}} + \Delta E_{\text{electr.}} < 0$$

Peierls (1930s/1955), Fröhlich (1954), Kohn (1959), Overhauser (1968/71), Chan & Heine (1973), McMillan (1977), ...

# Phase transition

## General energy–entropy considerations

$$\Delta F = \Delta E_{\text{latt.}} + \Delta E_{\text{electr.}} - T (\Delta S_{\text{latt.}} + \Delta S_{\text{electr.}})$$

$\Delta E < 0 (T = 0)$   $\Delta S < 0$

$\Delta F(T = T_0) = 0$ 
 $> 0$   
"passive"
 $< 0$   
"active"
 $< 0$   
(stiffening)
 $< 0$   
(gapping)

$\Delta E_{\text{electr.}}$

{

**1-e<sup>-</sup>**  
Band-structure energy  
Peierls instability

**many-e<sup>-</sup>**  
Exchange-correlation energy  
Excitonic insulator instability

$\Delta E_{\text{electr.}}$

{

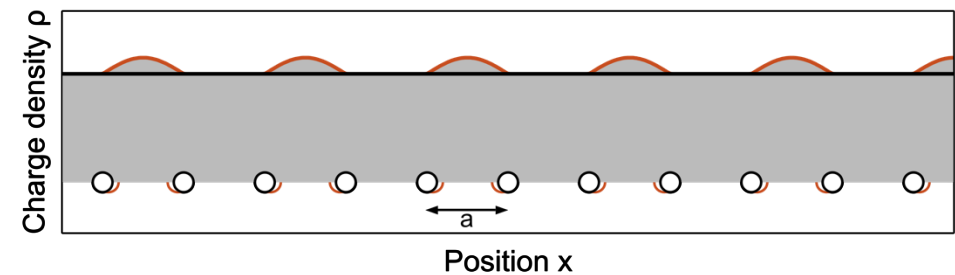
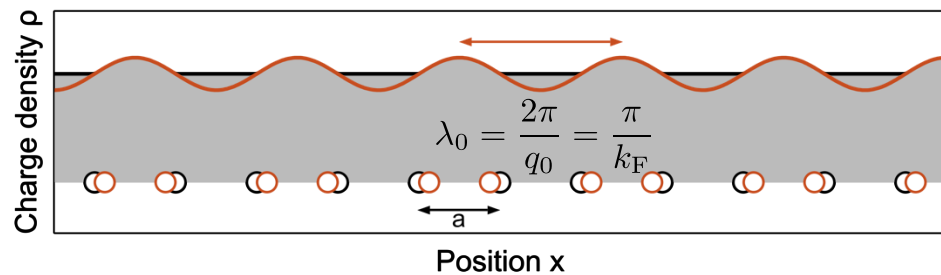
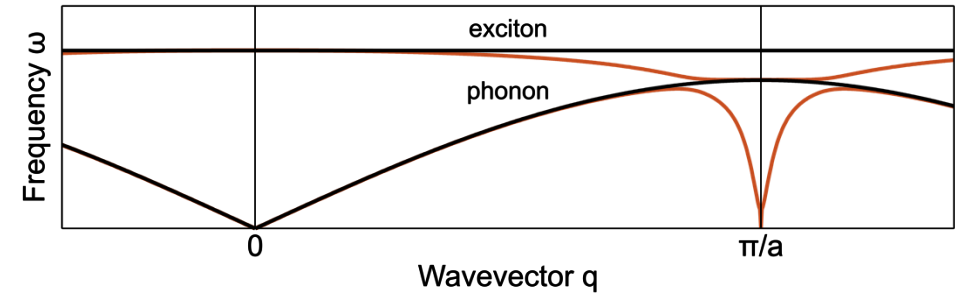
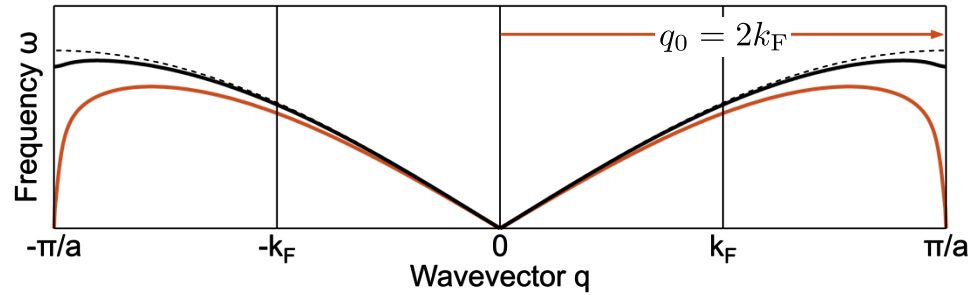
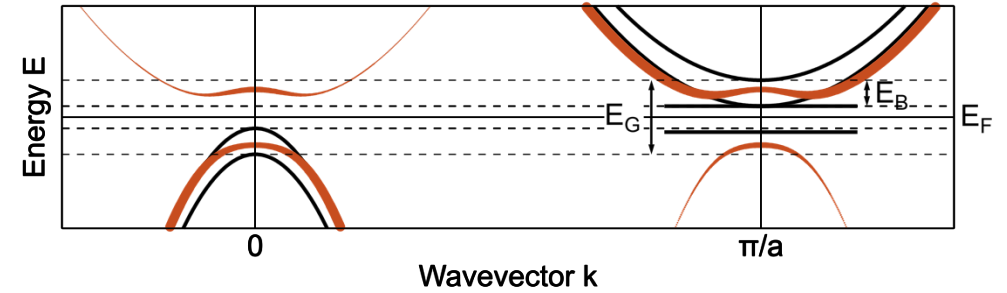
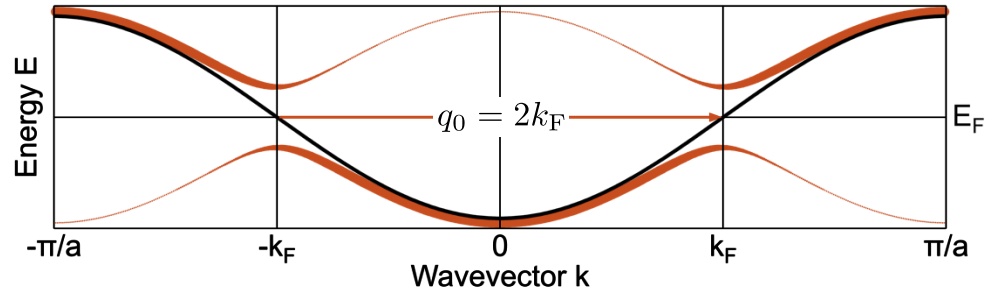
**e<sup>-</sup> near  $E_F$**  (metals)  
Small energy gap at  $E_F$   
Weak-coupling CDW

**e<sup>-</sup> far from  $E_F$**  (metals & insulators)  
Large energy gaps/splittings  
Strong-coupling CDW

Tosatti (1980/1995)

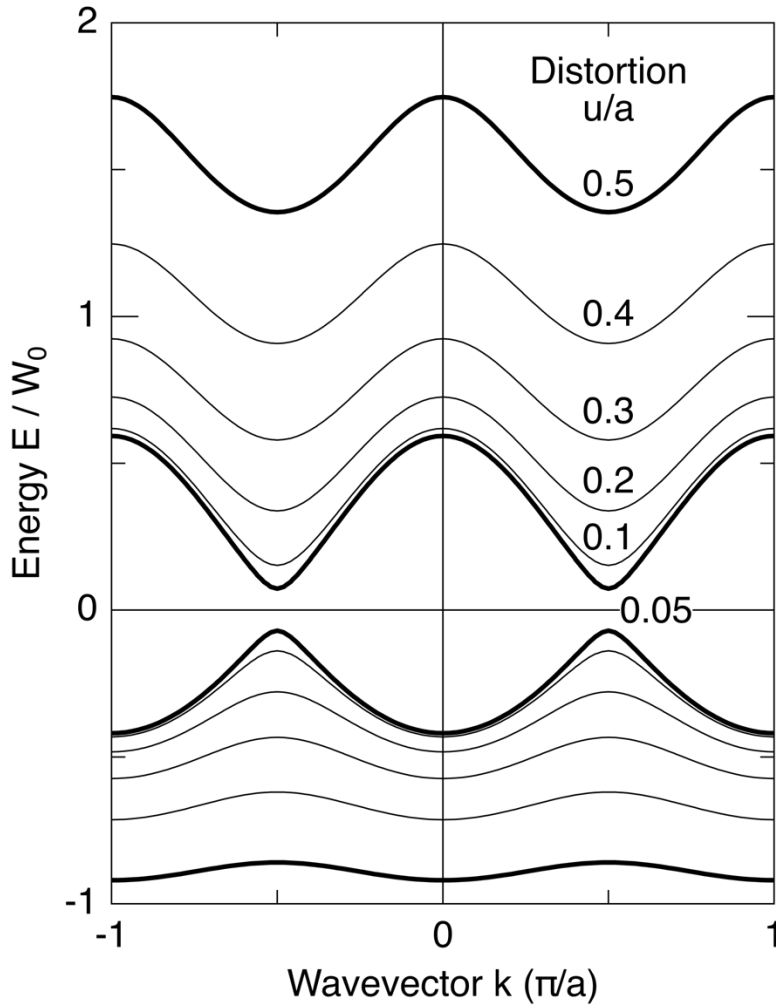
# “Two types” of CDW instability

## Peierls insulator *versus* excitonic insulator



# “Two types” of CDW

Weak coupling *versus* strong coupling

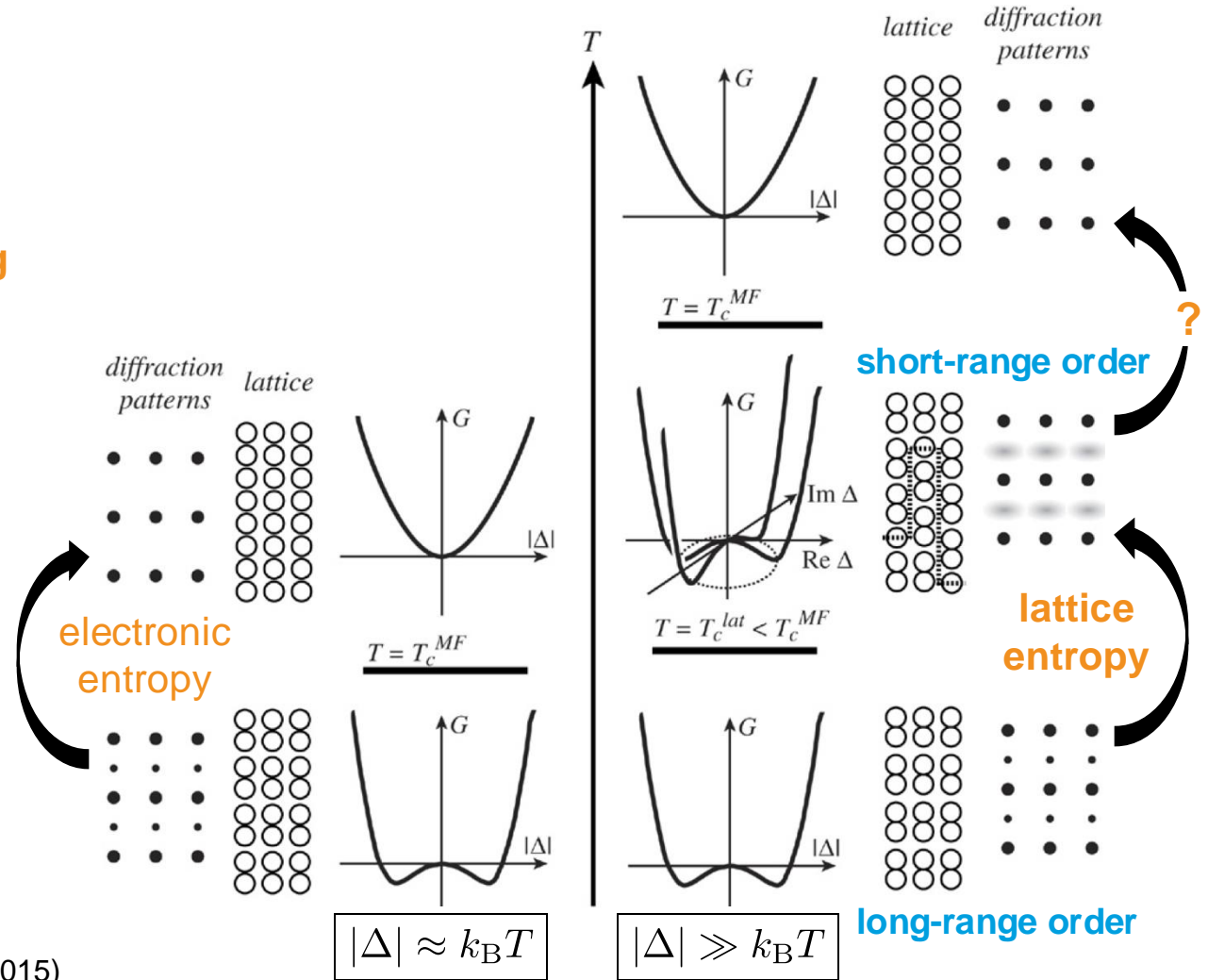


Chemical bond

Strong-coupling CDW

Weak-coupling CDW

McMillan (1977)  
Tosatti (1995)  
Aruga (2006)  
Flicker & van Wezel (2015)



# Microscopic origin

## Simple model

Chan & Heine (1973)

Cowley (1980), Withers & Wilson (1986)

$$\frac{1}{\chi(\mathbf{q})} \leq \frac{|g(\mathbf{q}j)|^2}{\hbar\omega_0(\mathbf{q}j)} - \beta(\mathbf{q})$$

electron–phonon (inelastic)      electron–electron  
 electron–lattice (elastic)      ion–ion      (nearly free electrons, no Umklapp processes)

large! →      large!      small!      small!

- No CDW without **sufficiently strong electron–phonon coupling**.
- No understanding of a CDW without **precise consideration of all four material-dependent functions**.
- No universal CDW mechanism.

**Noninteracting electronic susceptibility:** (almost never shows a peak at  $\mathbf{q}_0$ )

$$\chi(\mathbf{q}) = - \sum_{nn'\mathbf{k}} \frac{f(\mathbf{k}n) - f(\mathbf{k} + \mathbf{q}n')}{E(\mathbf{k}n) - E(\mathbf{k} + \mathbf{q}n')} |\langle \mathbf{k}n | e^{-\mathbf{q}\cdot\mathbf{r}} | \mathbf{k} + \mathbf{q}n' \rangle|^2$$

“Type I”

**“Structured” electron–phonon coupling:** (depends on momentum ( $\mathbf{q}, \mathbf{k}$ ) & orbital)

$$g(\mathbf{q}j) \sim \langle g(\mathbf{k} + \mathbf{q}n', \mathbf{k}n; j) \rangle$$

“Type II”

**Electron–electron interaction:** (prefers larger  $q$ )

$$\beta(\mathbf{q}) \propto \frac{e^2}{\epsilon_0 q^2} \left( 1 - \frac{1}{2} \frac{q^2}{q^2 + \xi^2} \right)$$

bare Coulomb      exchange & correlation

“Type III”

Zhu *et al.*, Adv. Phys.: X **2**, 622 (2017)

# Classification

## Three types of CDWs

	Type I	Type II	Type III
Driving force	Fermi surface nesting	“Structured” electron-phonon coupling	Electron correlations?
Systems	Quasi-1D (TTF-TCNQ <i>et al.</i> )	Quasi-2D (NbSe <sub>2</sub> <i>et al.</i> )	Cuprates

**Table 1.** Comparison of the key features of typical CDW materials with Peierls’ model.

	Peierls’ model (1D atomic chain)	TTF-TCNQ (Quasi-1D material)	NbSe <sub>2</sub> (Quasi-2D material)	Sn/Ge or Pb/Ge (2D films on surface)	Cuprates
Fermi Surface Nesting	√	√ [27]	X [30–32]	√ [37,38]	√ [64,69,70]
Sharp Peak in Lindhard Function	√	√ [23]	X [30–32]	√ [38]	√ [32]
Kohn Anomaly	√	√ [25]	√ [20]	√ [39,40]	X [32]
Structural Transition	√	√ [24]	√ [15–19]	√ [37,38]	√ [67,68]
Metal–insulator Transition	√	√ [22]	X [34]	√ [37]	X [69]

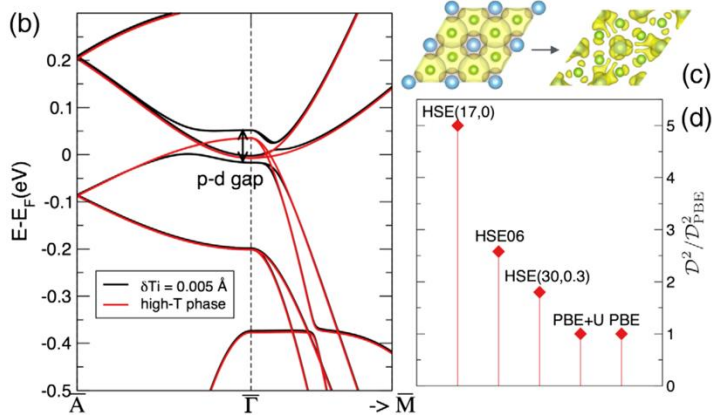
Zhu *et al.*, PNAS **112**, 2367 (2015)

Zhu *et al.*, Adv. Phys.: X **2**, 622 (2017)

# Quantitative microscopic understanding

Combined theoretical–experimental approach. Example: 1T-TiSe<sub>2</sub>.

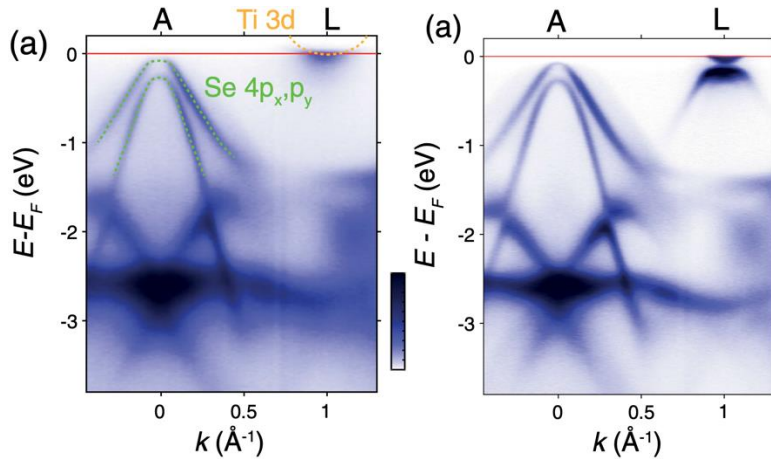
DFT-HSE [Hellgren *et al.*, PRL **119**, 176401 (2017)]



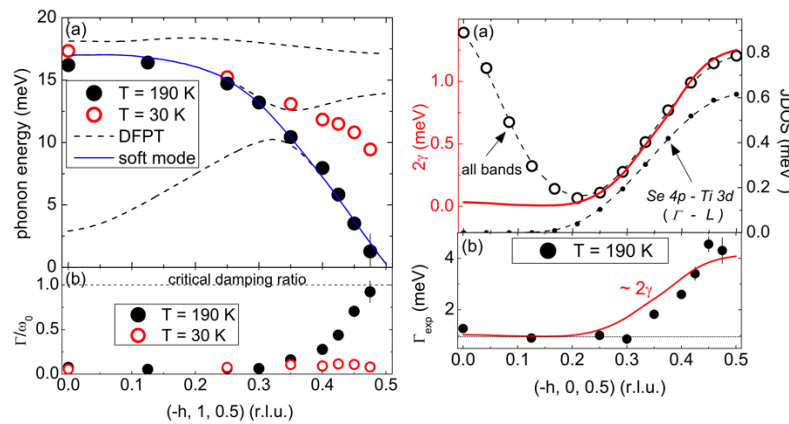
*Ab initio* theory + empirical models  
TB, DFT (e.g., PBE, HSE), DMFT, DFPT, TDDFT, ...

$$\frac{1}{\chi(\mathbf{q})} \leq \frac{|g(\mathbf{q}j)|^2}{\hbar\omega_0(\mathbf{q}j)} - \beta(\mathbf{q})$$

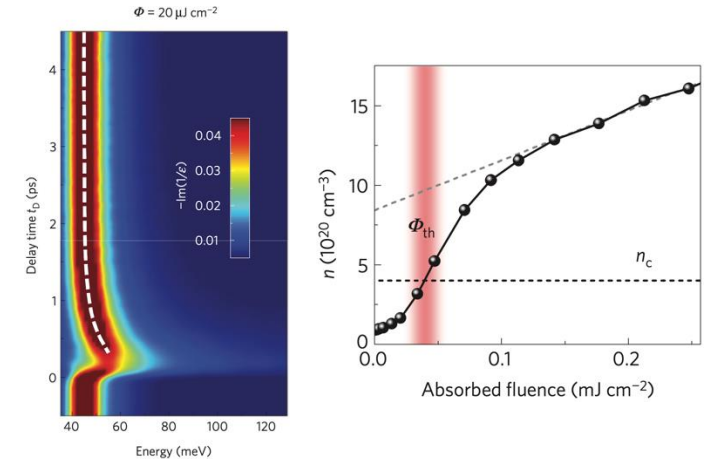
ARPES [Watson *et al.*, PRL **122**, 076404 (2019)]



IXS [Weber *et al.*, PRL **107**, 266401 (2011)]

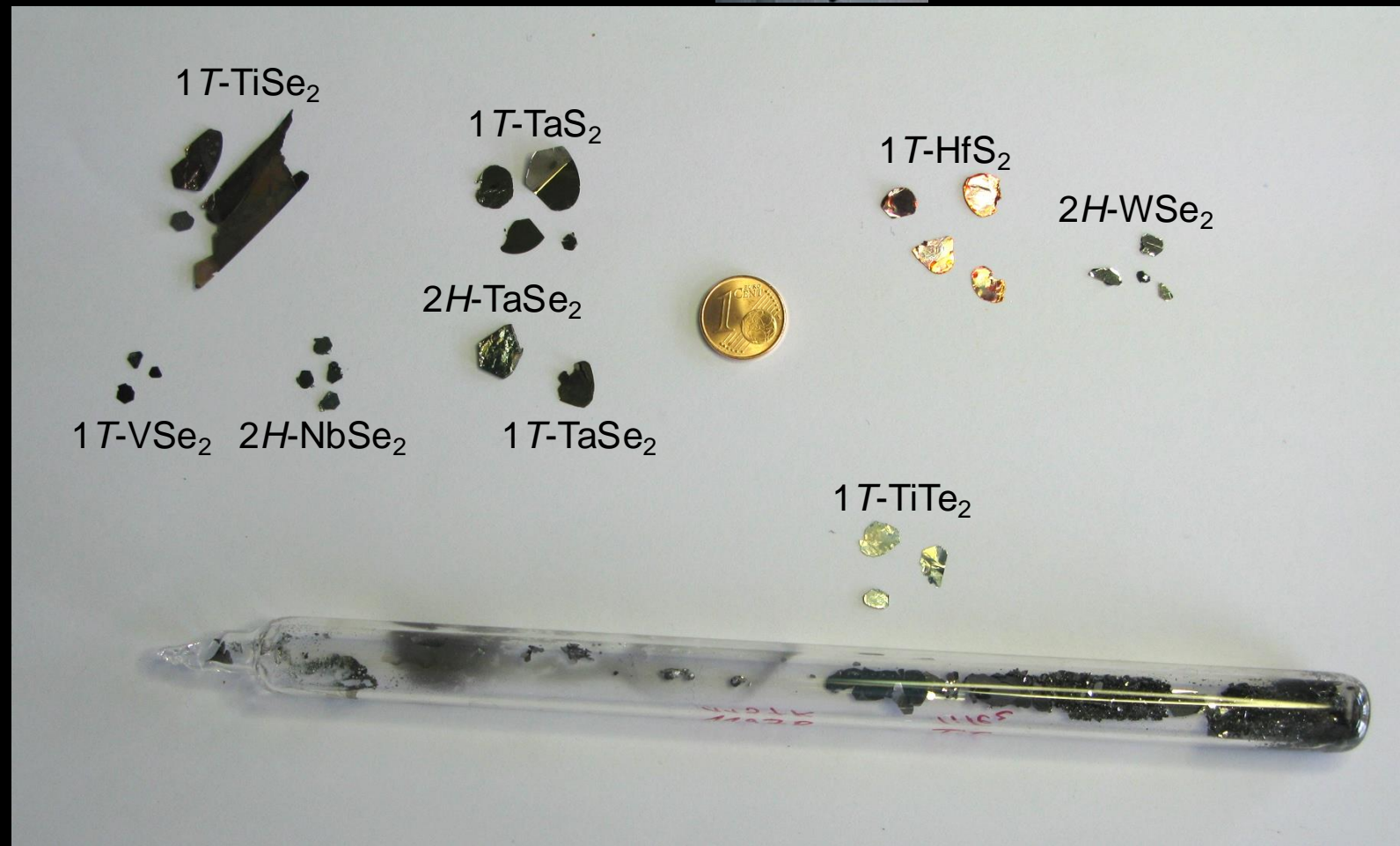
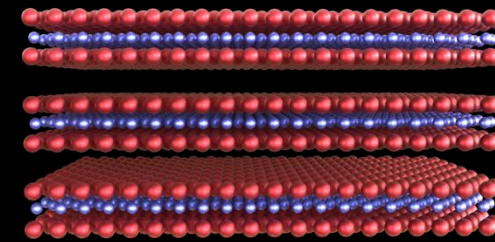


tr-multi-THZ [Porer *et al.*, NMAT **13**, 857 (2014)]



# Commercial break: TMDC crystals

The thinnest fish sandwiches from Kiel. Do you want one?

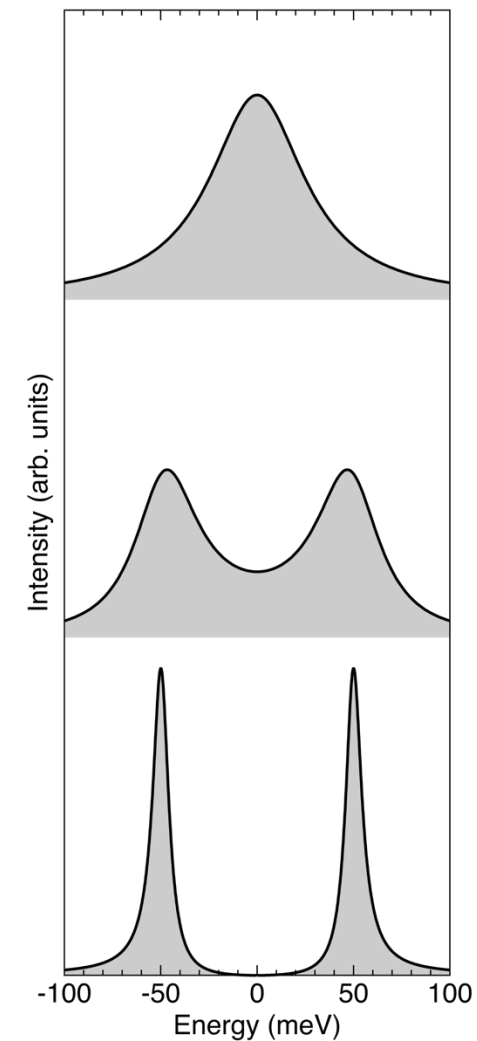
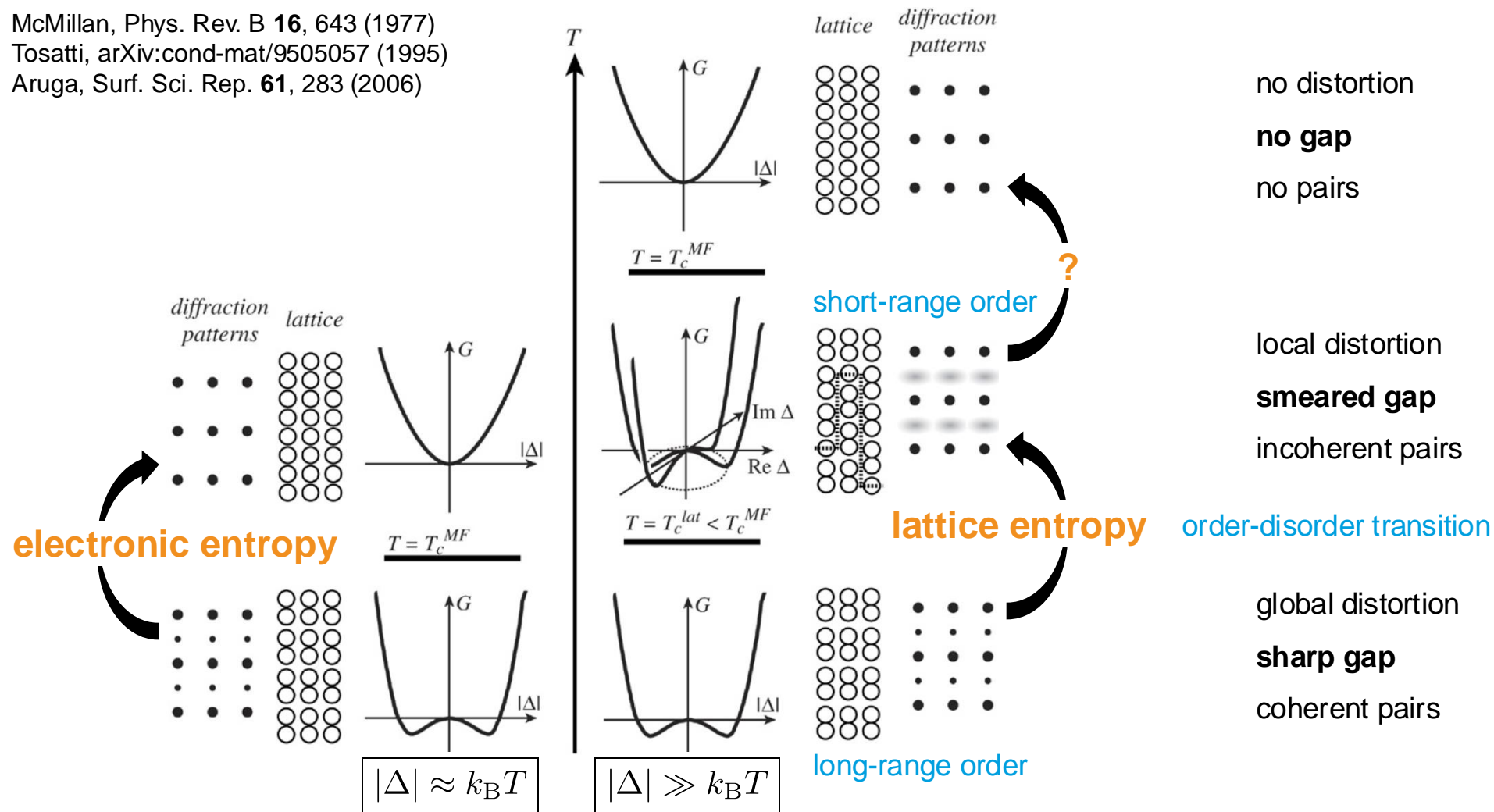


[rossnagel@physik.uni-kiel.de](mailto:rossnagel@physik.uni-kiel.de)

# Thermal disordering

## Weak coupling *versus* strong coupling

McMillan, Phys. Rev. B **16**, 643 (1977)  
 Tosatti, arXiv:cond-mat/9505057 (1995)  
 Aruga, Surf. Sci. Rep. **61**, 283 (2006)



# Outline

... a reflection of my (narrow) world view

## Ultrafast Unordering of Electronic Order

### I. Motivation

Quest for a new switch.

### II. Notes on charge-density waves

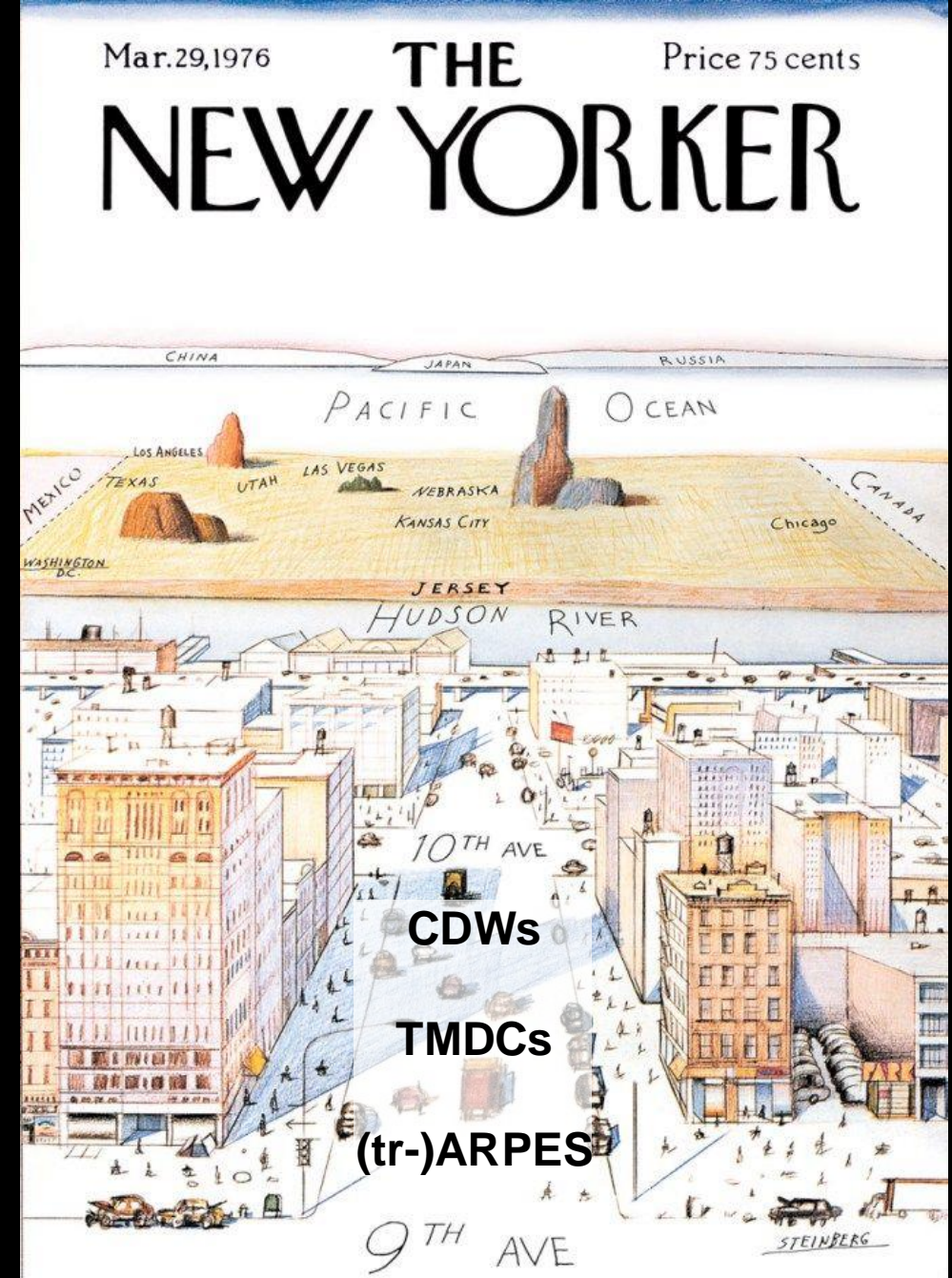
Nature & origin (“types”).

### III. Ultrafast unordering (of CDWs)

Mechanisms. Speeds. Efficiencies.  
Time-domain classification.

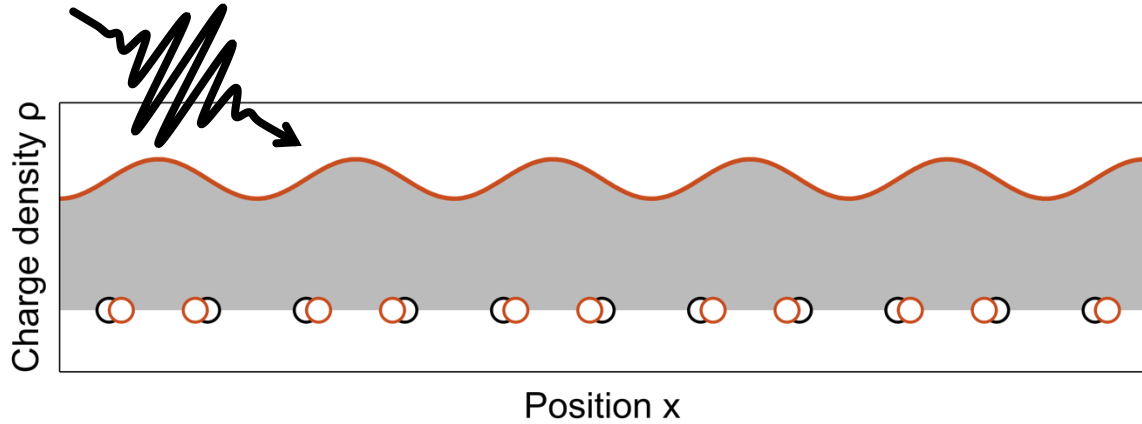
### IV. Outlook

Tools & concepts.



# Ultrafast CDW unordering: Multiplicity of time scales

Three fundamental time scales (speed limits) of the CDW trinity

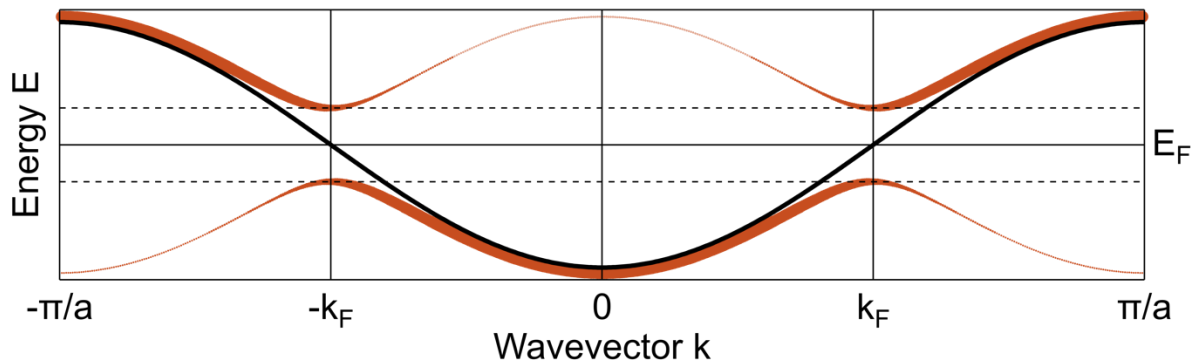


Charge-density wave

$$\tau_e = \frac{h}{W} = \mathcal{O}\left(\frac{h}{1 \text{ eV}}\right) = \mathcal{O}(4 \text{ fs})$$

Periodic lattice distortion

$$\tau_{\text{lat}} = \frac{h}{E_A} = \mathcal{O}\left(\frac{h}{10 \text{ meV}}\right) = \mathcal{O}(400 \text{ fs})$$

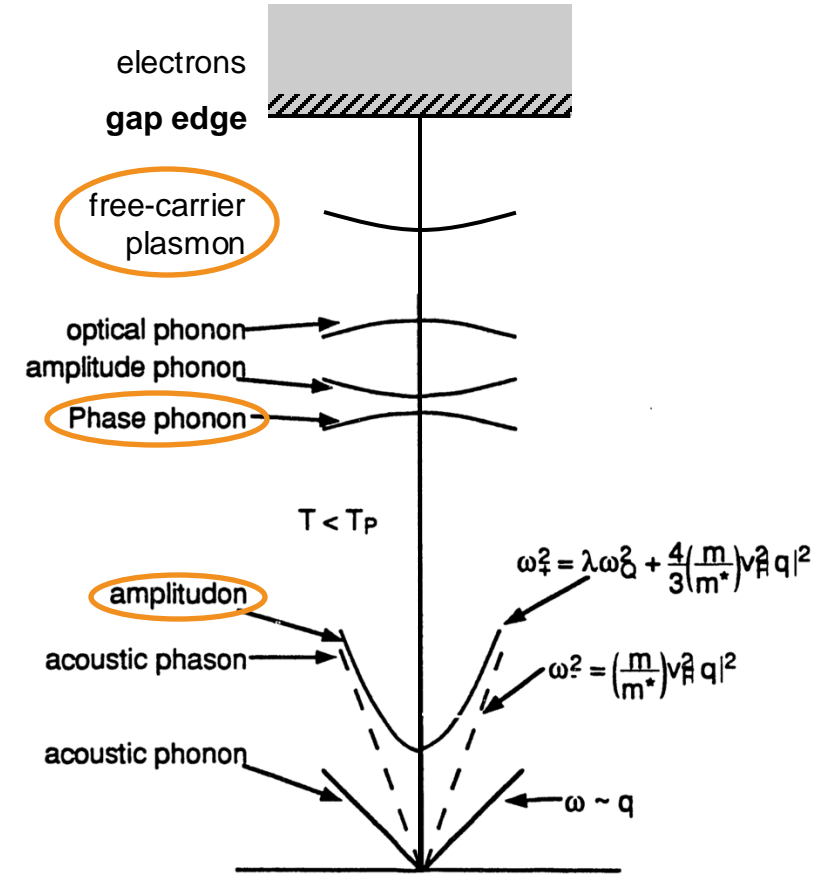
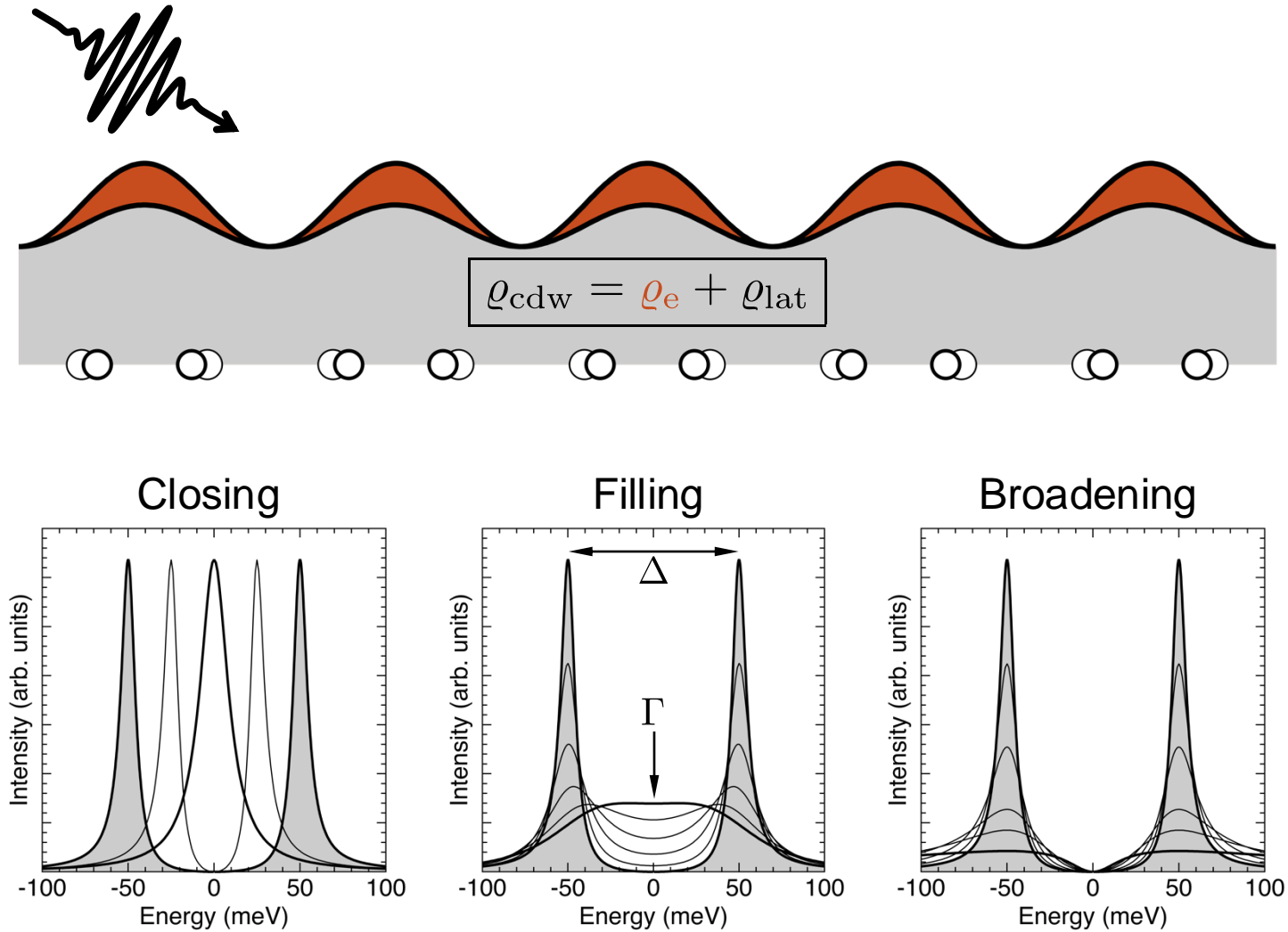


Energy gap

$$\tau_{\Delta} = \frac{h}{2\Delta} = \mathcal{O}\left(\frac{h}{100 \text{ meV}}\right) = \mathcal{O}(40 \text{ fs})$$

# Ultrafast CDW unordering: Spectroscopic view

Three ways to quench an electronic gap

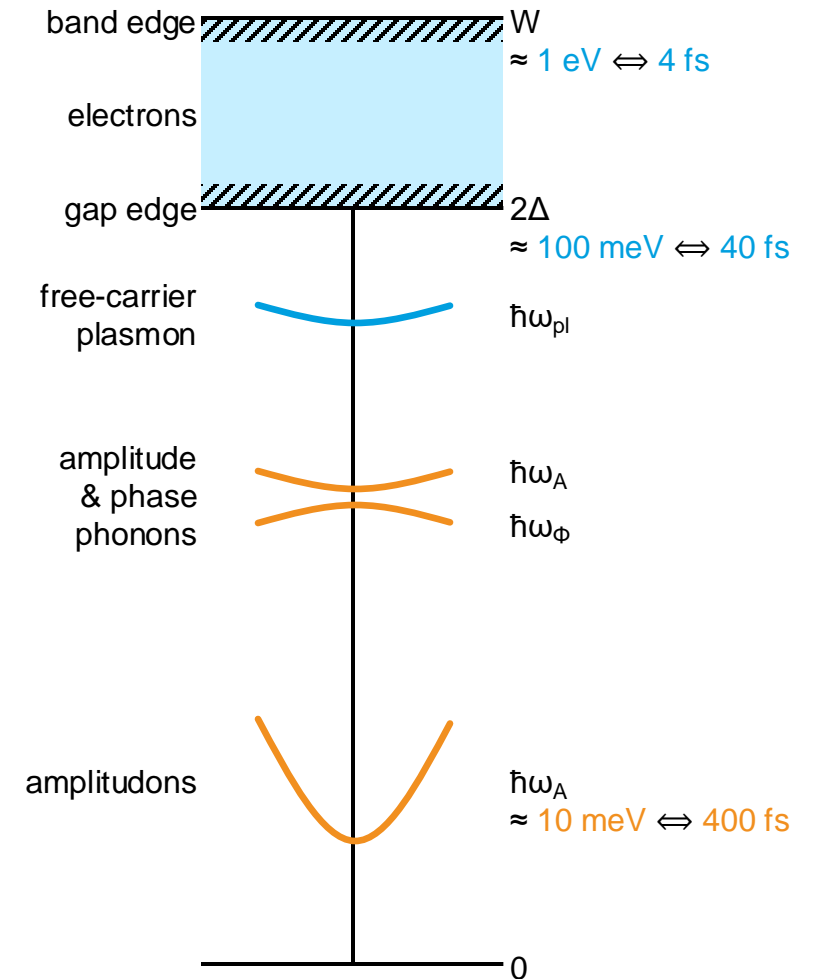


W. N. Creager, Ph.D. Thesis (1991)

# Ultrafast CDW unordering: Overview

## Matrix of materials & dominant excitations

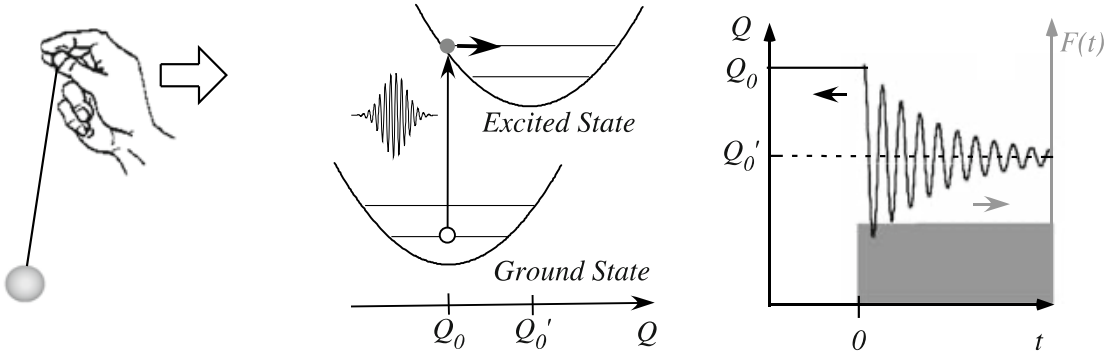
	Coherent Displacive Gap closing ( $\Delta$ )	Incoherent Order-disorder Gap filling ( $\Gamma$ )
<b>Electron-</b> dominated	<p>TiSe<sub>2</sub></p> <p><math>\hbar\omega_{pl}, \hbar\omega_A</math></p> <p>Type III</p>	<p>?</p> <p>W?</p> <p>Type IV</p>
<b>Phonon-</b> dominated	<p>K<sub>0.3</sub>MoO<sub>3</sub>, TbTe<sub>3</sub></p> <p><math>\hbar\omega_A</math></p> <p>Type I</p>	<p>Rb<sub>0.3</sub>MoO<sub>3</sub>, VO<sub>2</sub></p> <p><math>\hbar\omega_\phi, \hbar\omega_A</math></p> <p>Type IIa, IIb</p>



# (I) Acoustic phonon-stabilized CDW

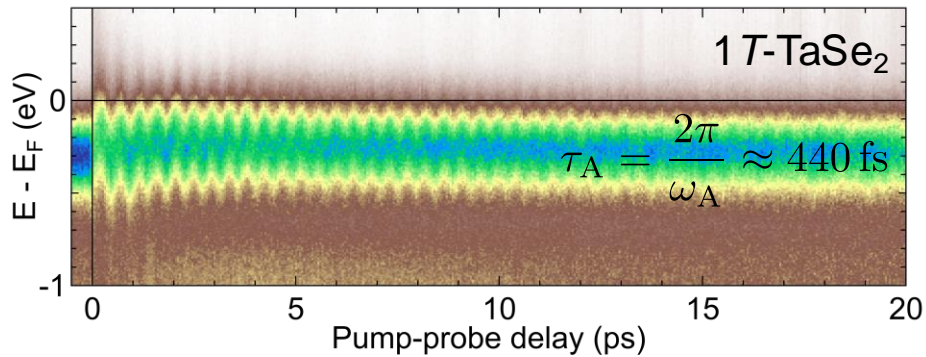
Gap closing via coherent amplitudons (gap follows lattice distortion)

Displacive excitation of coherent phonons



Ishioka & Misochko (2010)

Coherent amplitudons  $\Rightarrow$  coherent gap oscillation



Sohrt *et al.*, Faraday Discuss. **171**, 243 (2014)

$$\tau_A = \frac{2\pi}{\omega_A} \approx 600 \text{ fs}$$

$$\tau_0^{\text{lat}} \approx \frac{\tau_A}{2}$$

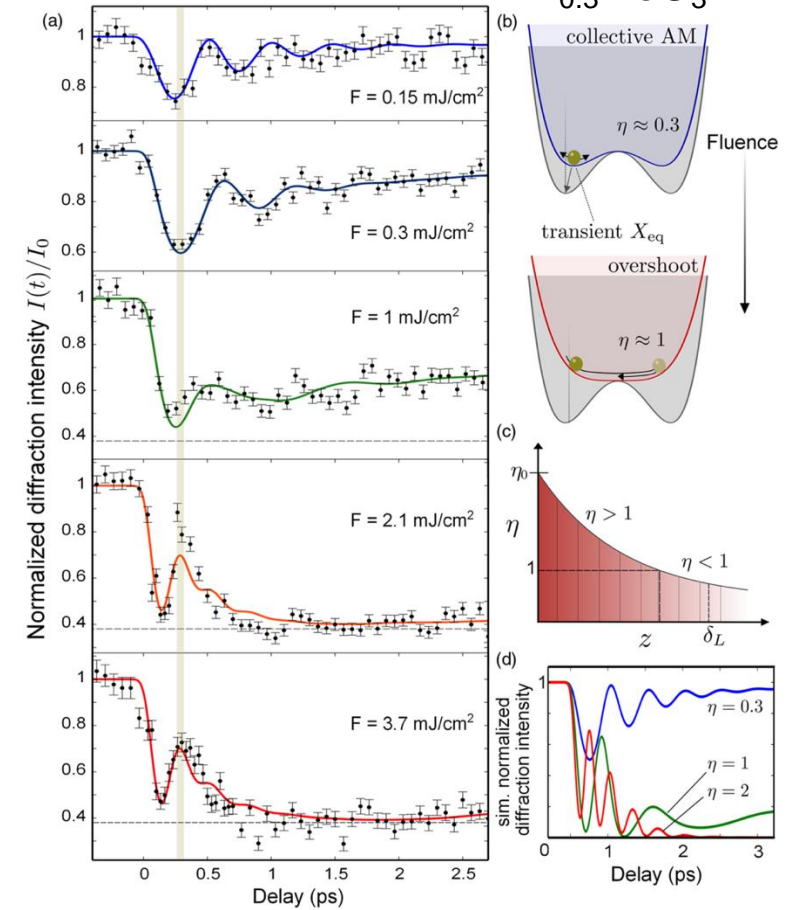
Gap closing

$$\tau_{\Delta} = \tau_A$$

$$\Delta(t) \propto u_A(t)$$

$$\tau_0^{\text{lat}} \approx \frac{\tau_A}{4}$$

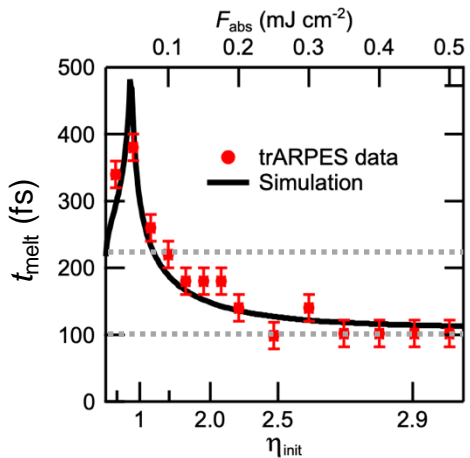
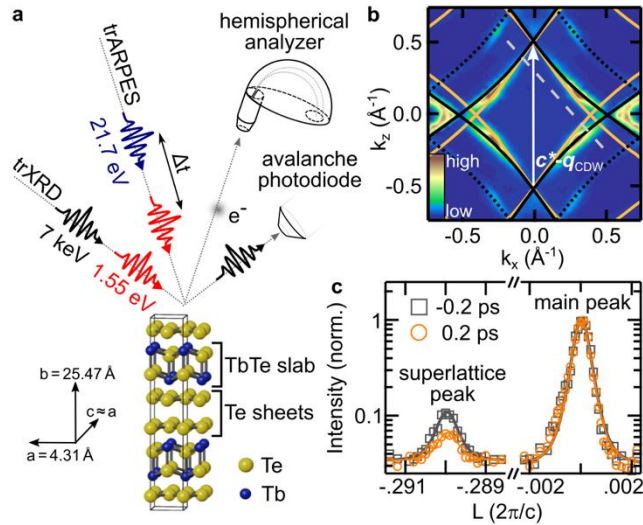
Coherent amplitudons



Huber *et al.*, Phys. Rev. Lett. **113**, 026401 (2014)

# (I) Acoustic phonon-stabilized CDW: $\text{TbTe}_3$

Gap closing via coherent amplitudons (gap follows lattice distortion)



$$\tau_0^{\text{lat}} \approx \frac{\tau_A}{2}$$

$$\tau_0^{\text{lat}} \approx \frac{\tau_A}{4}$$

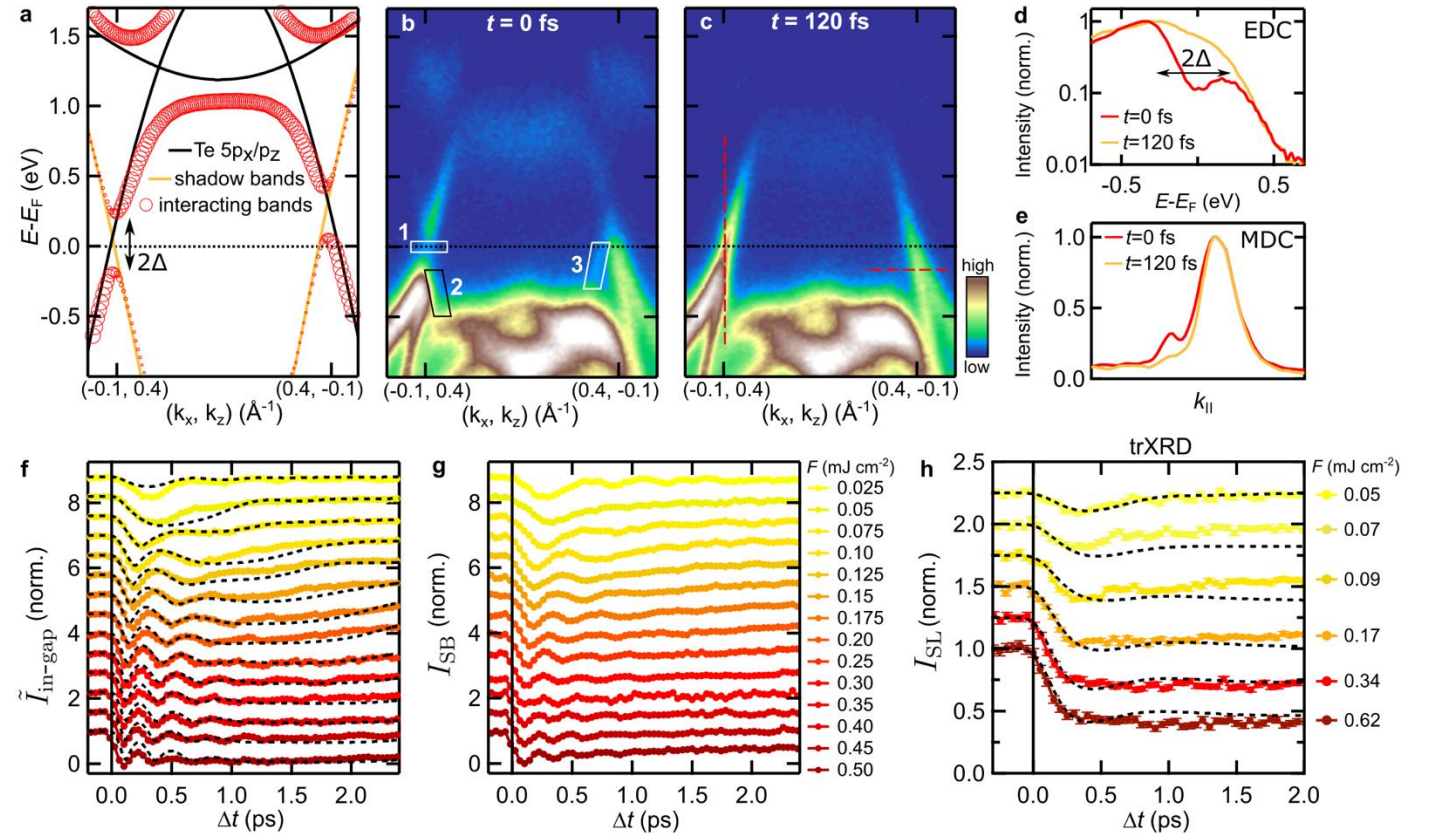
$$\tau_A = \frac{2\pi}{\omega_A} \approx 450 \text{ fs}$$

$$|\Delta_e| \sim |\Delta_{\text{lat}}| \quad \curvearrowright \quad \varrho_{\text{cdw}} = \varrho_{\text{lat}}$$

Gap closing

$$\tau_{\Delta} = \tau_A$$

$$\Delta(t) \propto u_A(t)$$



Maklar *et al.*, Nat. Commun. **12**, 2499 (2021)

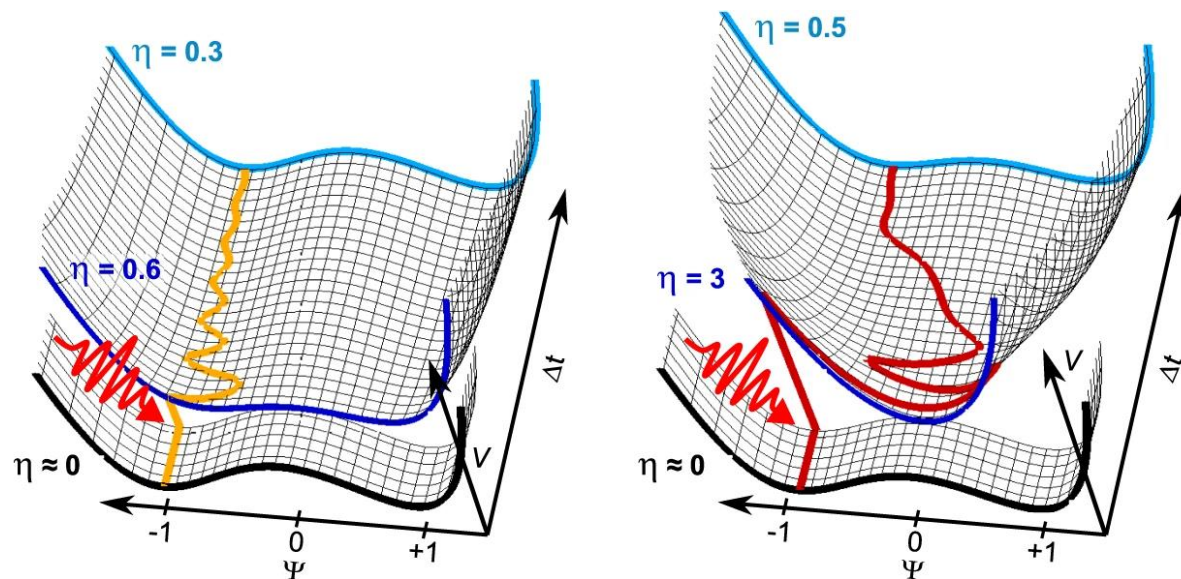
# (I) Acoustic phonon-stabilized CDW

Dynamic energy landscape & maximum unordering speed

Optical excitation

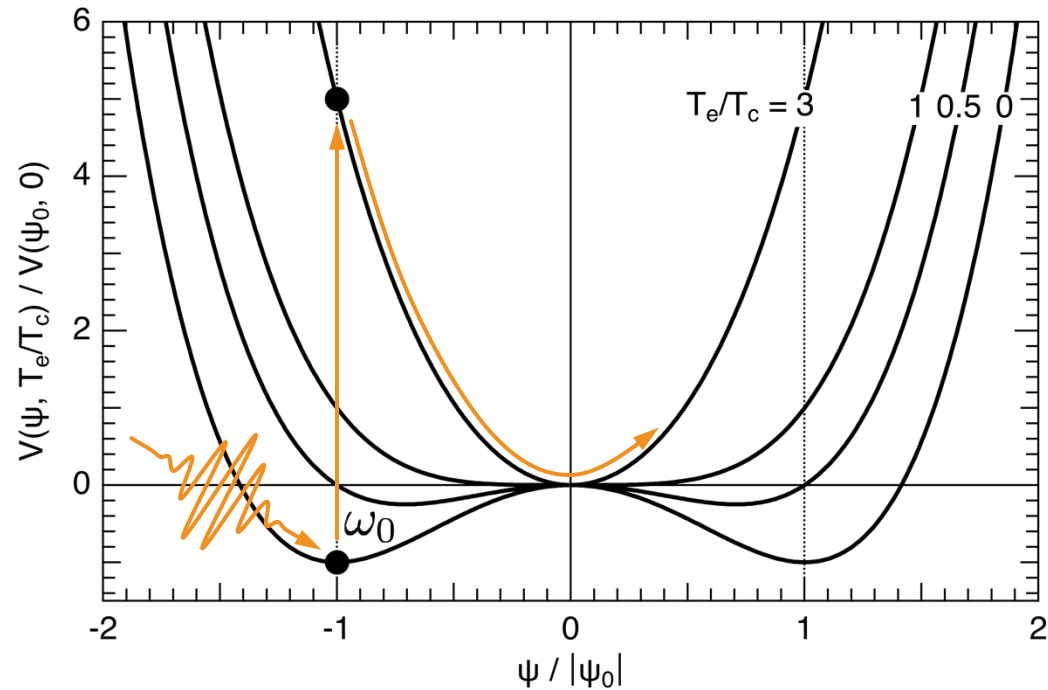
Weak

Strong



Order parameter overshoot

$$\tau(-1 \rightarrow 0) \approx \frac{1}{4} \left( \frac{2\pi}{\omega_0} \right)$$

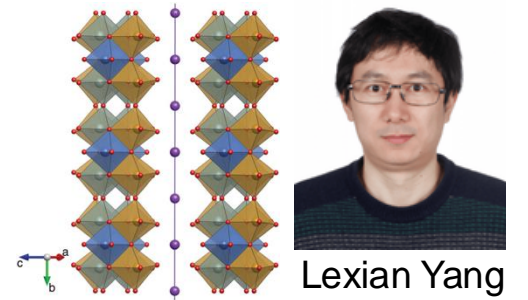


$$V\left(\psi, \frac{T_e}{T_c}\right) = a \left( \frac{T_e}{T_c} - 1 \right) \psi^2 + \frac{1}{2} b \psi^4 + \dots, \quad a = b = 1$$

Maklar *et al.*, Nat. Commun. **12**, 2499 (2021)

# (IIa) Multiphonon-stabilized CDW: $\text{Rb}_{0.3}\text{MoO}_3$

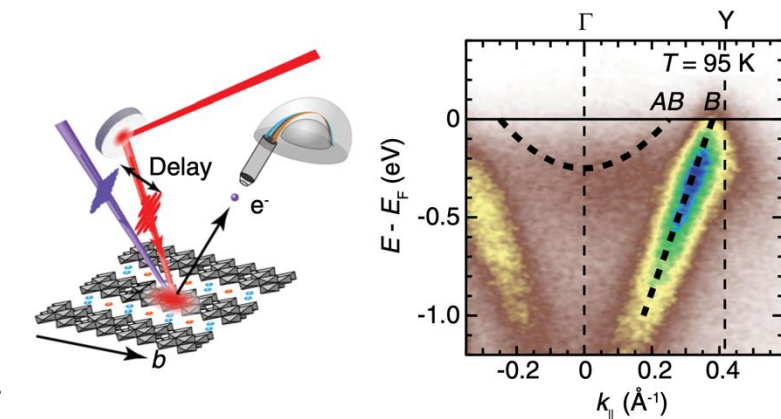
Gap filling via incoherent phase phonons (gap follows lattice order/disorder)



Disorder parameter (lattice fluctuations):

$$\eta = \frac{\hbar}{\tau_{\text{e-ph}}\Delta} = \pi \sum_n \lambda_n \frac{\hbar\omega_n}{2\Delta} \coth\left(\frac{\hbar\omega_n}{2T}\right) > 0.6$$

Degiori *et al.*, Phys. Rev. B **49**, 14754 (1994)



Yang *et al.*, Phys. Rev. Lett. **125**, 266402 (2020)

Gap filling & closing

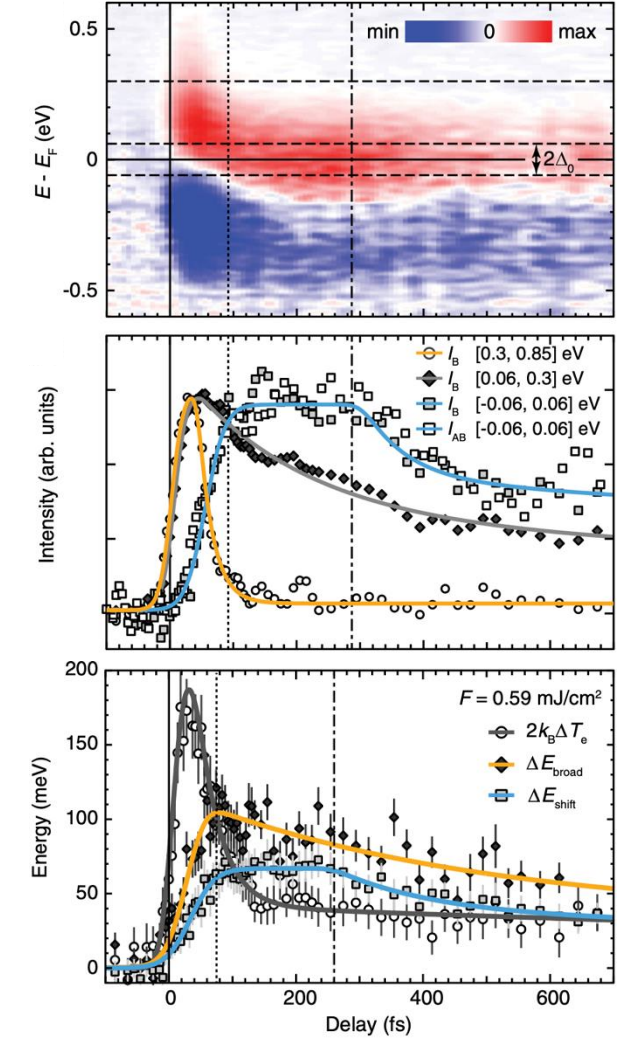
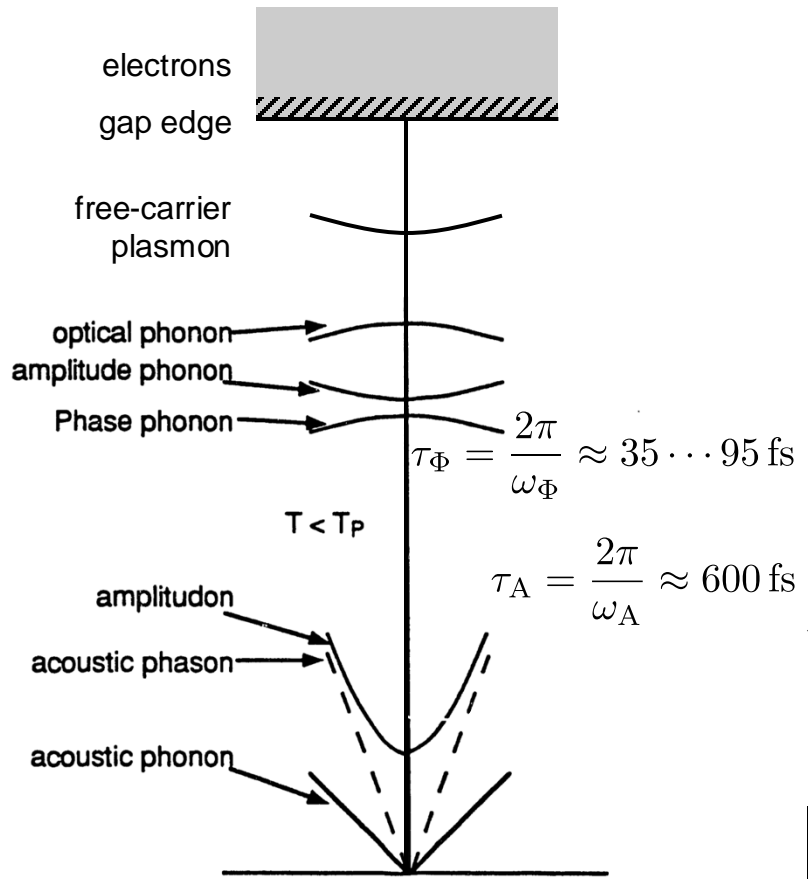
$$\tau_{\Delta} = \tau_{\Phi} \cdots \tau_A$$

$$\leadsto \varrho_{\text{cdw}} = \varrho_{\text{lat}}$$

$$\tau_0^{\text{lat}} \approx \tau_{\Phi} \cdots \frac{\tau_A}{2}$$

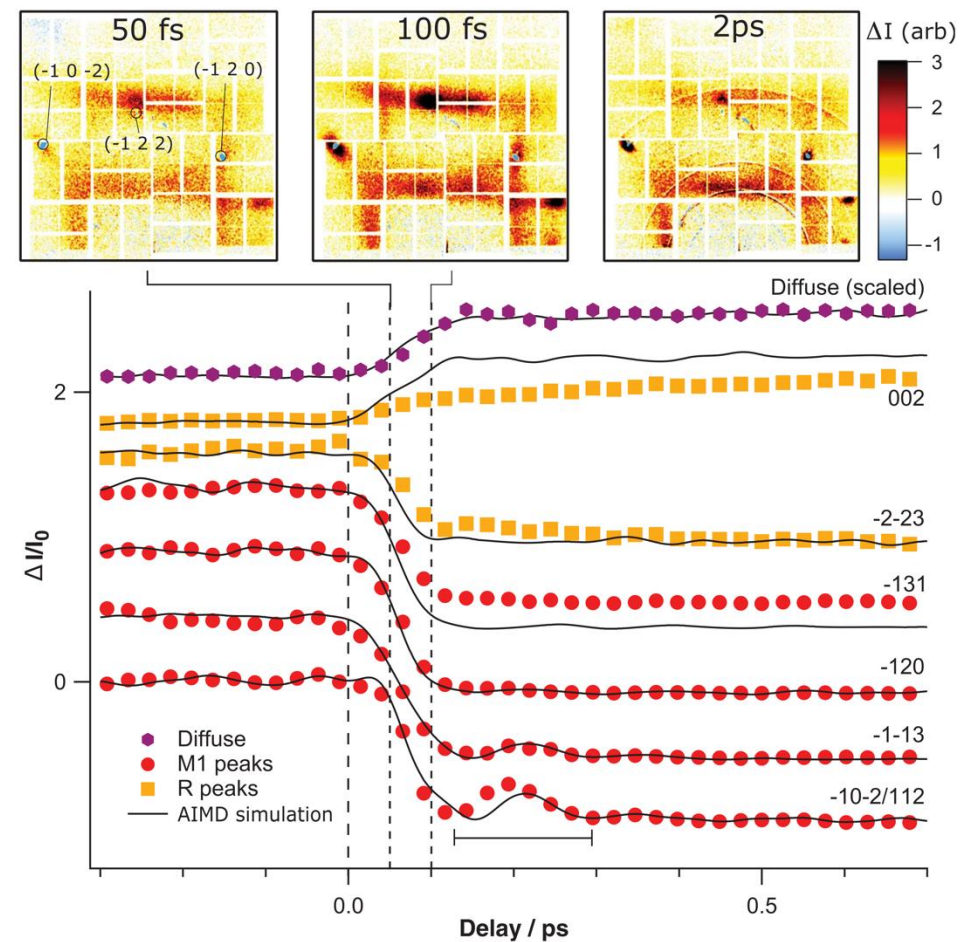
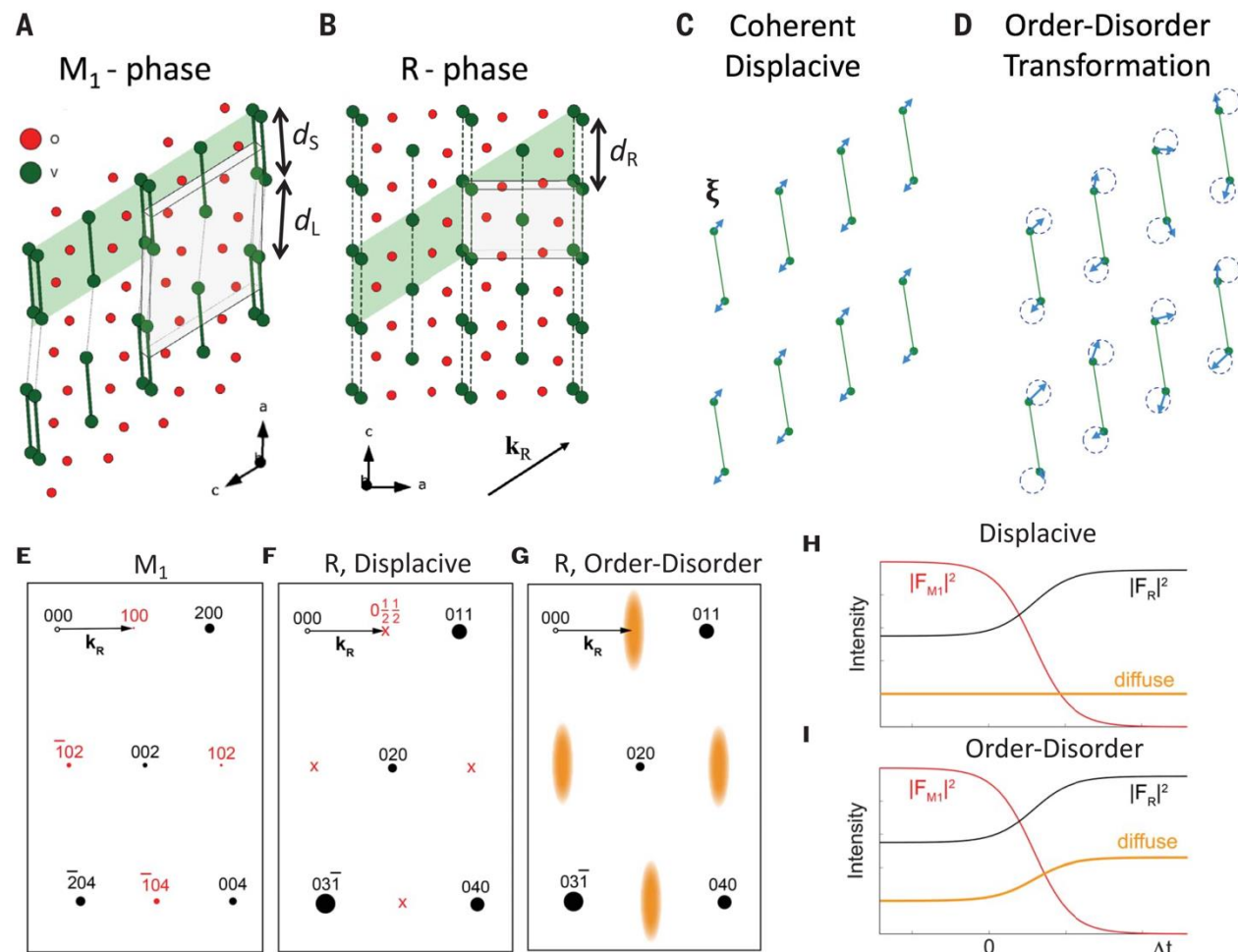
$$\Gamma_{\Phi}(t) \sim T_{\Phi}^*(t)$$

$$\Delta_{\text{lat}}(t) \propto u_A(t)$$



# (IIb) Dimer-stabilized CDW: $\text{VO}_2$

Ultrafast “undimerization” via *incoherent* acoustic phonons



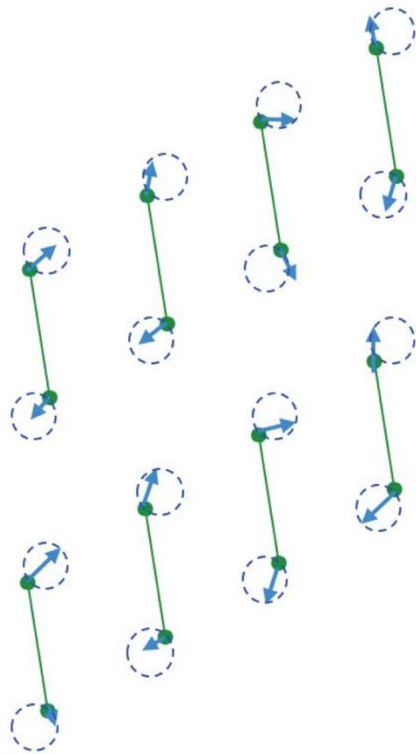
$$\tau_0^{\text{lat}} \approx 150 \text{ fs} \approx \frac{1}{2 \cdot 3 \text{ THz}} \approx \frac{\tau_{\text{ac}}}{2}$$

Wall *et al.*, Science **362**, 572 (2018)

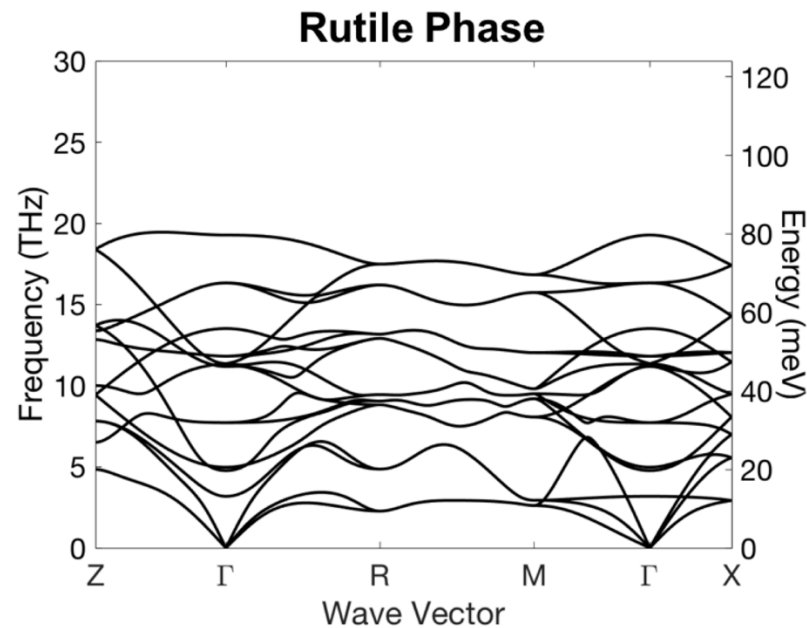
# (IIb) Dimer-stabilized CDW: VO<sub>2</sub>

Ultrafast “undimerization” via *incoherent* acoustic phonons

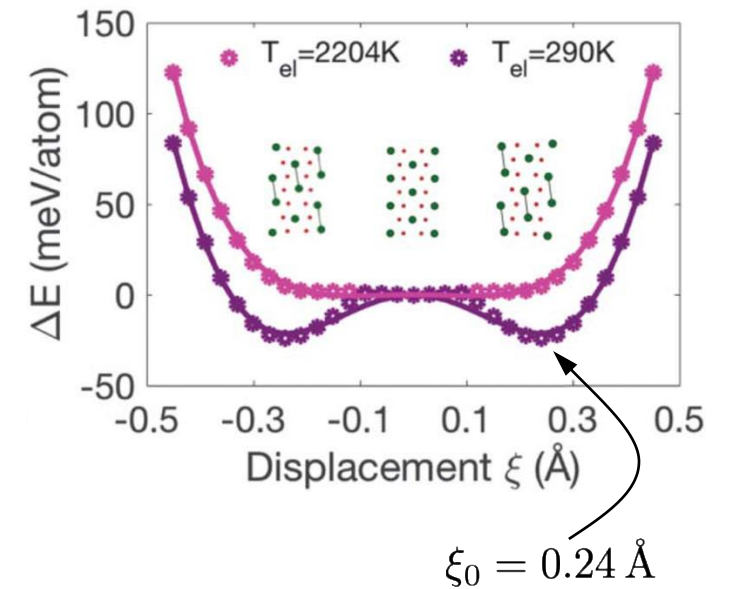
Incoherent ion motion



Flat acoustic phonons



Flat photoexcited potential



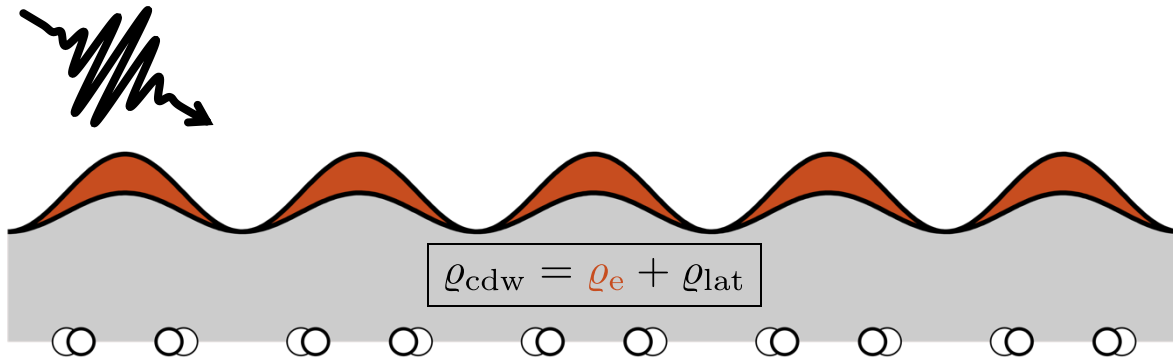
$$v_0 \approx v_{th} = \sqrt{\frac{3k_B T}{M}} \stackrel{300K}{=} 3.8 \frac{\text{Å}}{\text{ps}}$$

Large phonon entropy  
(low energy, no well-defined wavevector)

Inertial motion  
(speed limited by initial velocity)

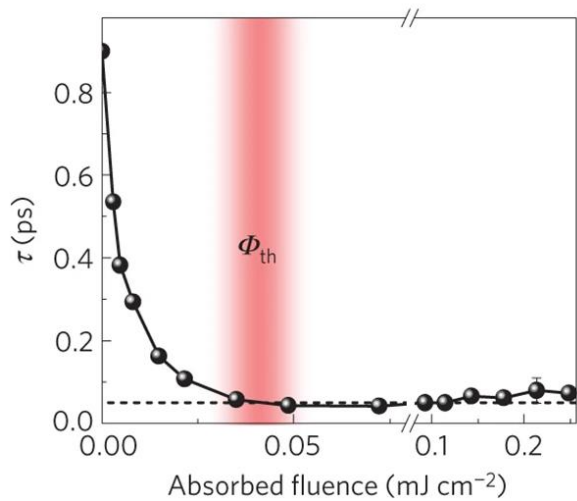
# (III) Phonon-plasmon/exciton-stabilized CDW: $\text{TiSe}_2$

Intertwined but separable electron–lattice order

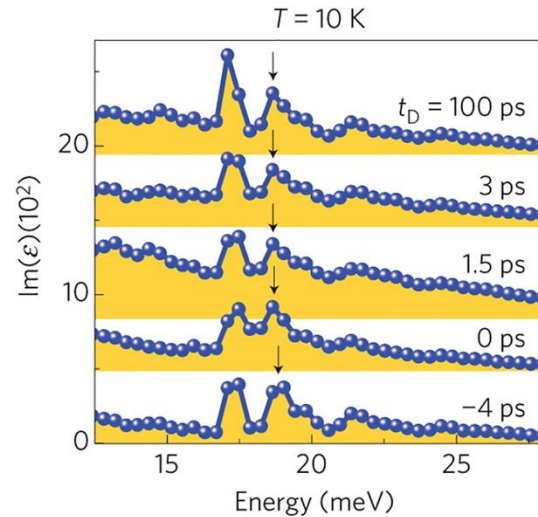


Porer *et al.*, Nat. Mater. **13**, 857 (2014)

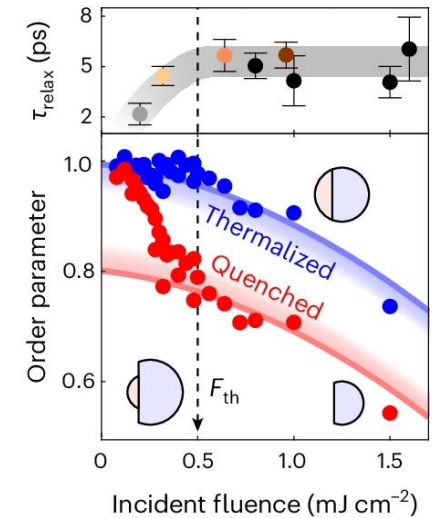
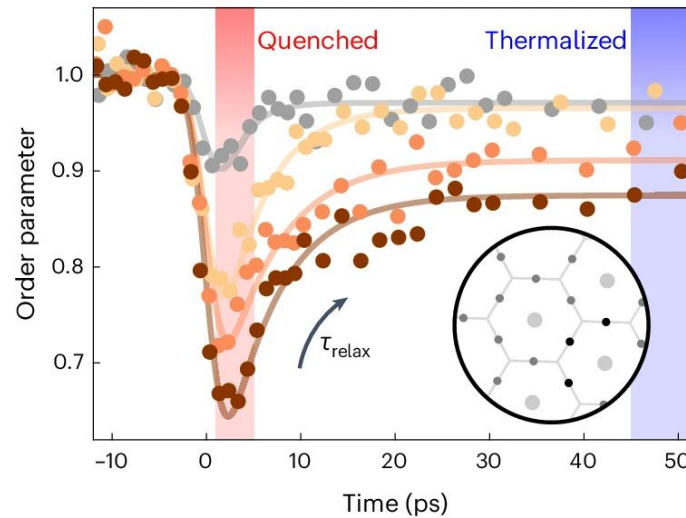
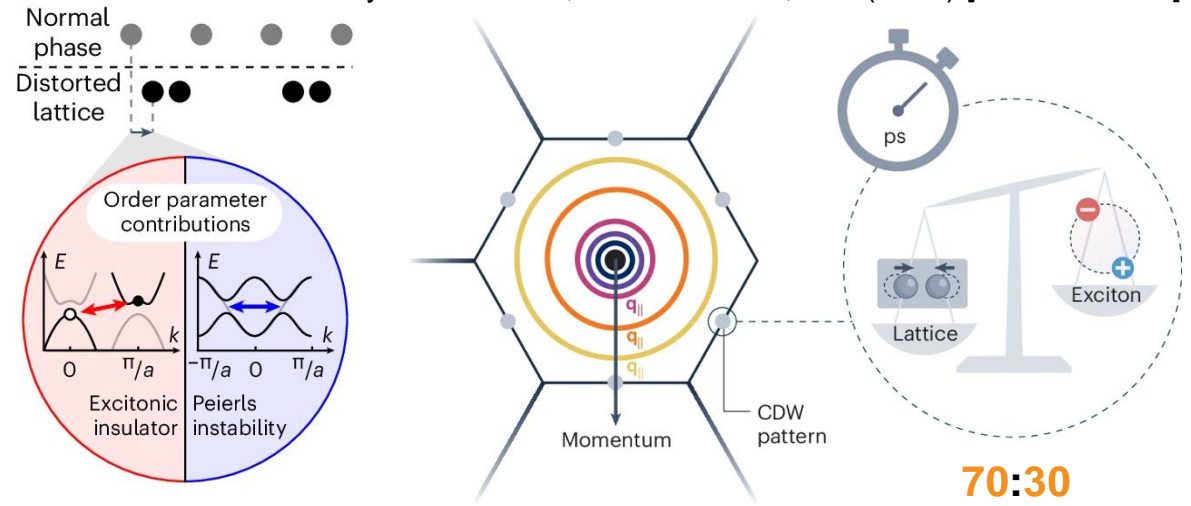
Quenched  $\text{CDW}_e$



Persistent  $\text{CDW}_{\text{lat}}$  (PLD)

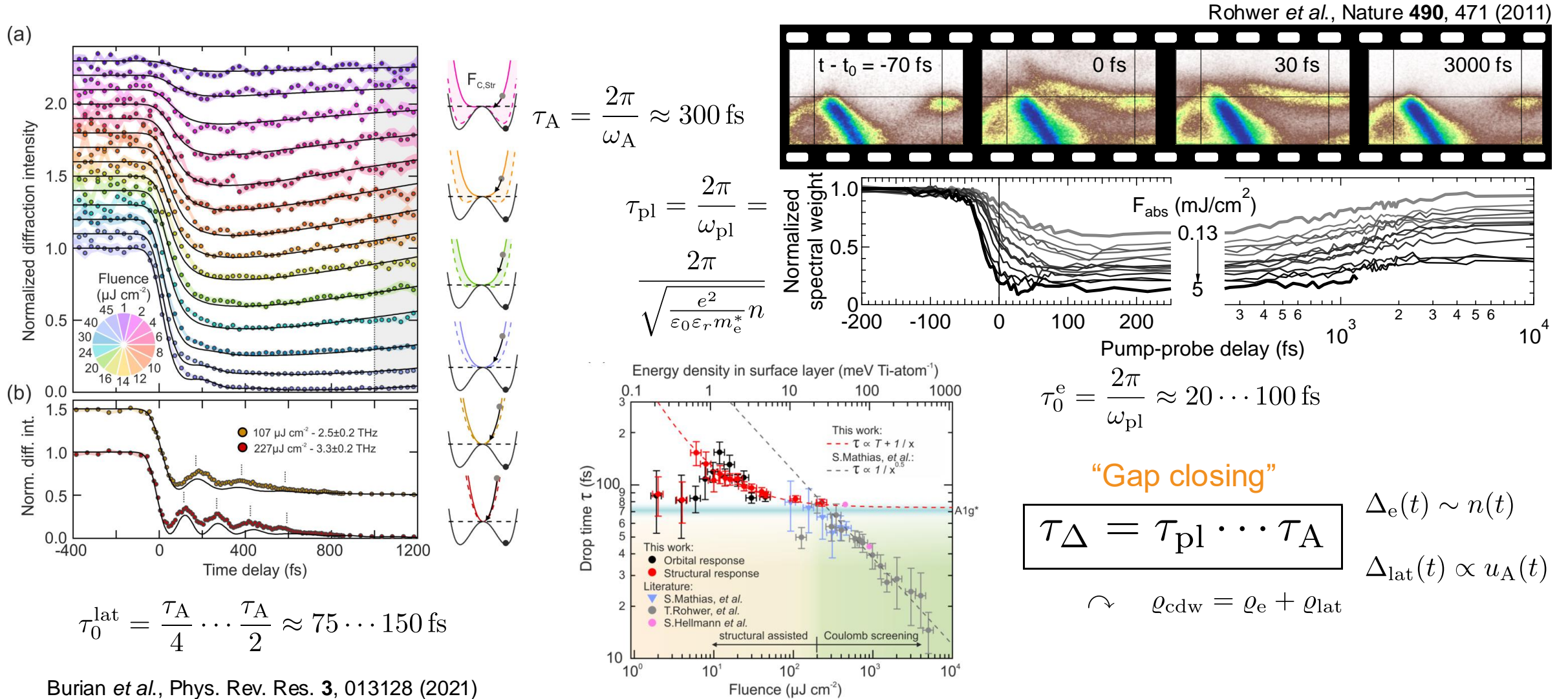


Kurtz *et al.*, Nat. Mater. **23**, 890 (2024)  
Sayers & Cerullo, Nat. Mater. **23**, 870 (2024) [News & views]



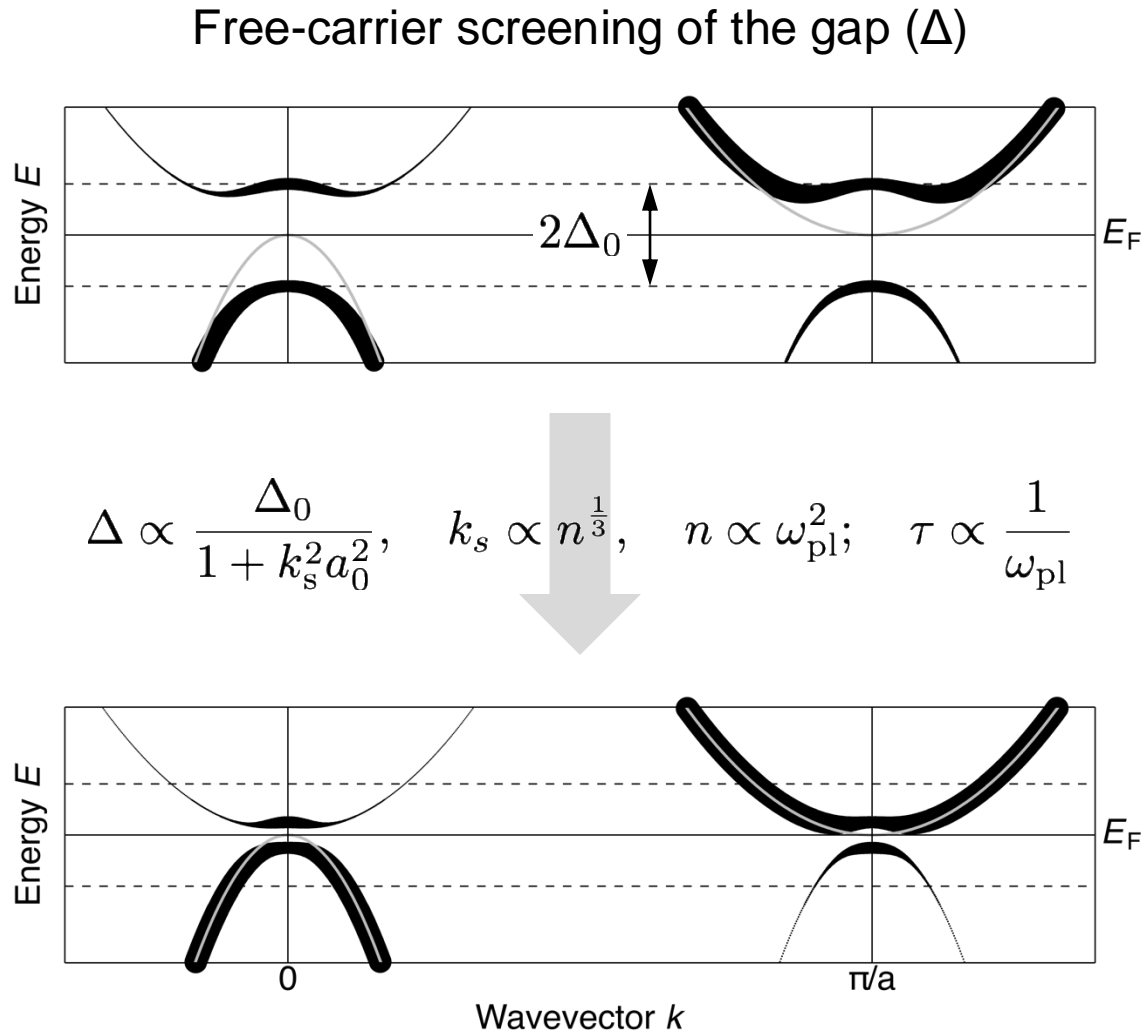
# (III) Phonon-plasmon/exciton-stabilized CDW: $\text{TiSe}_2$

Gap closing via coherent amplitudons & free-carrier screening (gap follows split electron-lattice order)

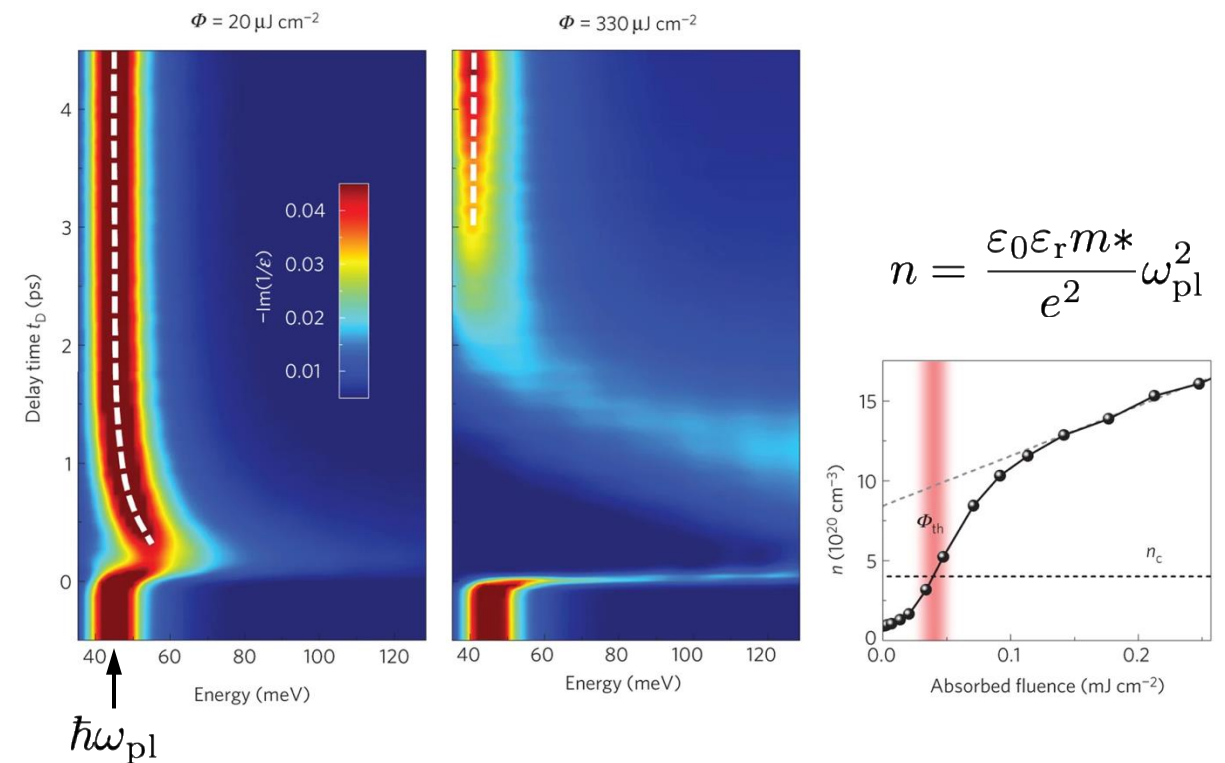


# (III) Phonon-plasmon/exciton-stabilized CDW: TiSe<sub>2</sub>

## Gap closing via free-carrier screening



## Direct tracking of plasmon dynamics



Porer *et al.*, Nat. Mater. **13**, 857 (2014)

# Ultrafast CDW unordering

## Time-domain classification

	Type I	Type IIa	Type IIb	Type III
Stabilization	Acoustic phonon	Multiphonon	Tilted dimerized V-V pairs + correlations	Phonon-plasmon/exciton
Transition	<i>Coherent</i> displacive	<i>Incoherent</i> order-disorder + <i>coherent</i> displacive	<i>Incoherent</i> order-disorder	Electronic + <i>coherent</i> displacive
Driving force	<i>Coherent</i> amplitudons	<i>Incoherent</i> phase phonons + <i>coherent</i> amplitudons	<i>Incoherent</i> flat acoustic phonons	Electronic screening + <i>coherent</i> amplitudons
Ungapping	Closing	Filling + closing	Filling?	Closing?
Speed limit	$\tau_0^{\text{lat}} \gtrsim \frac{\tau_A}{4}$	$\tau_0^{\text{lat}} \gtrsim \tau_\Phi$	$\tau_0^{\text{lat}} \gtrsim \frac{\tau_{\text{ac}}}{2}$	$\tau_0^{\text{e}} \gtrsim \tau_{\text{pl}} \geq \frac{h}{2\Delta}$
System	TbTe <sub>3</sub>	K <sub>0.3</sub> MoO <sub>3</sub>	VO <sub>2</sub>	TiSe <sub>2</sub>

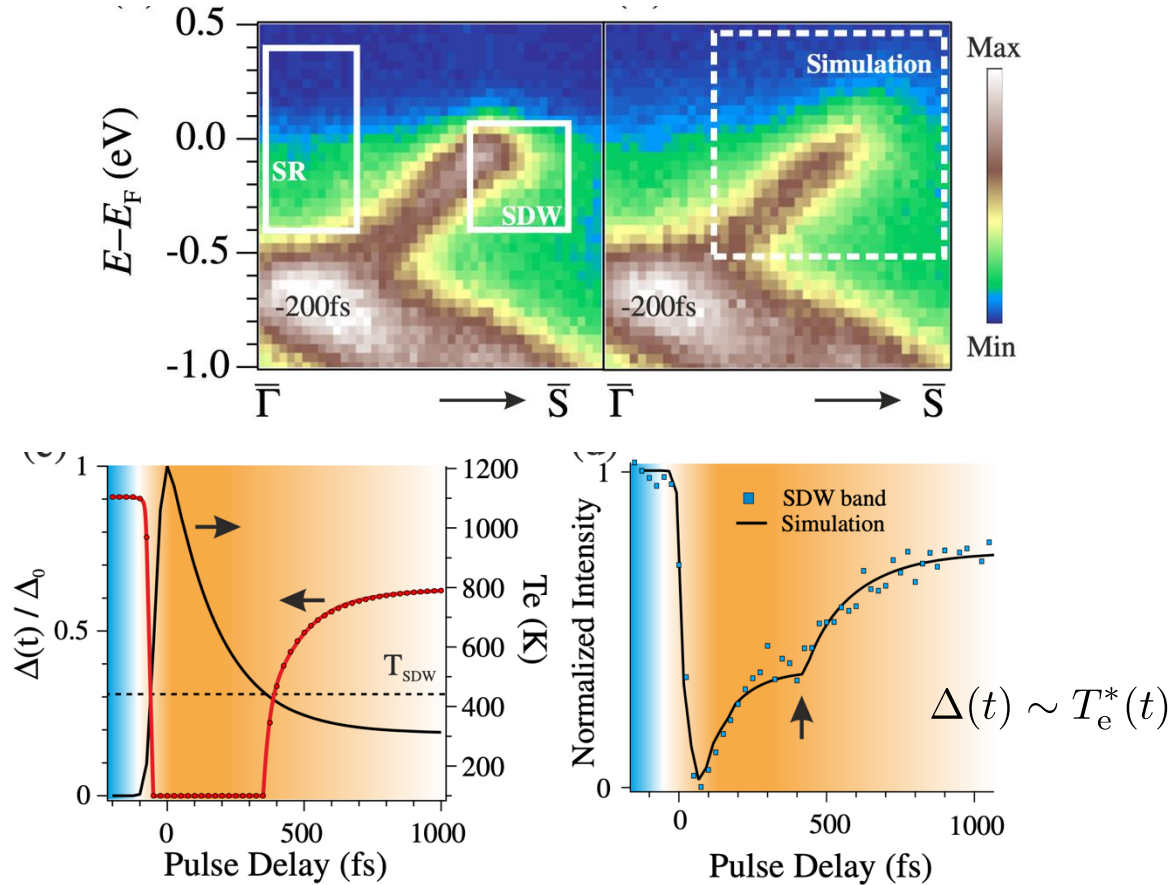
# Other types of electronic order

## Characteristic time scales

Wigner crystal

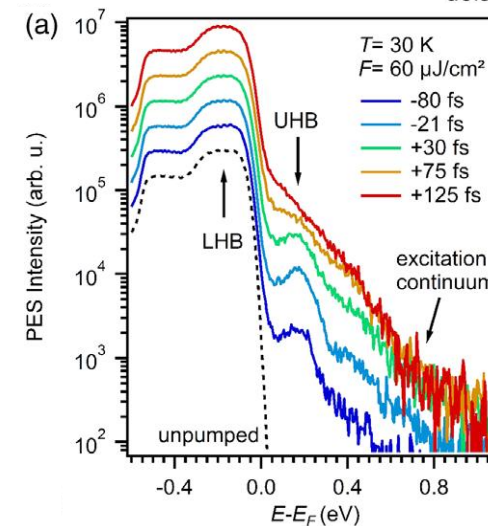
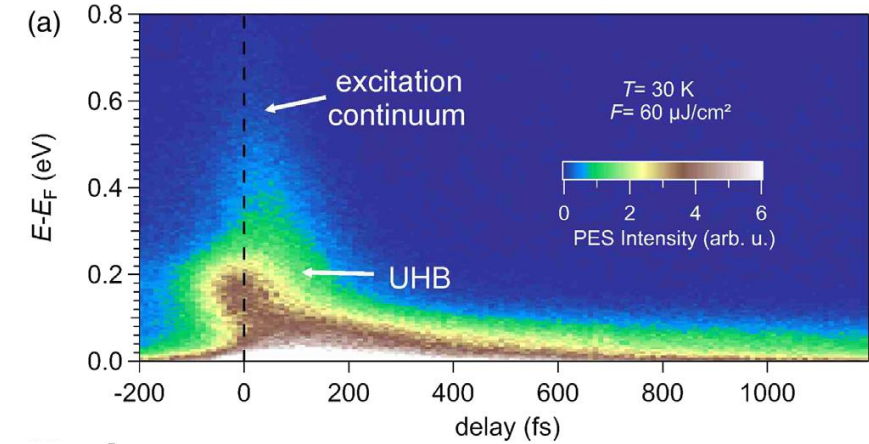
$$\tau_e = ?$$

### Spin-density wave (Cr)



$$\tau_e < 100 \text{ fs}$$

### Mott insulator (1T-TaS<sub>2</sub>)



$$\tau_e = \frac{h}{W} \approx 12 \text{ fs}$$

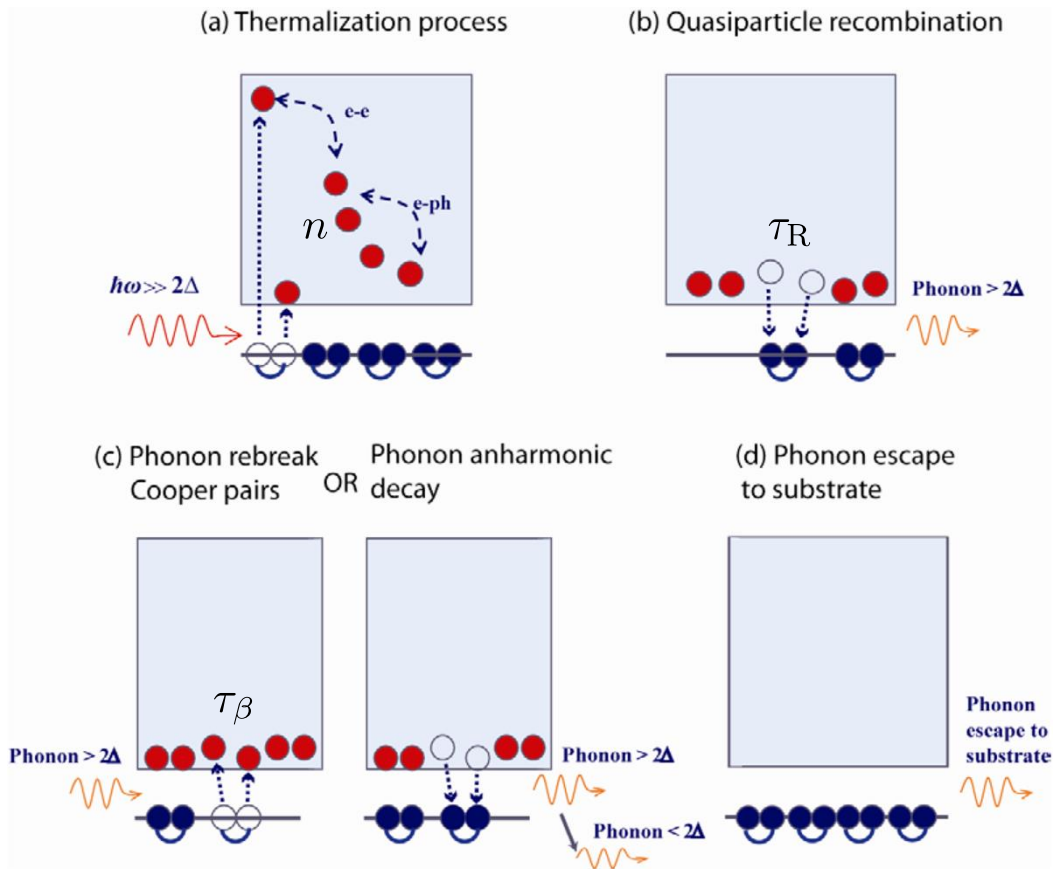
Ligges *et al.*,  
Phys. Rev. Lett. **120**, 166401 (2018)

Nicholson *et al.*,  
Phys. Rev. Lett. **117**, 136801 (2016)

# BCS superconductor: NbN

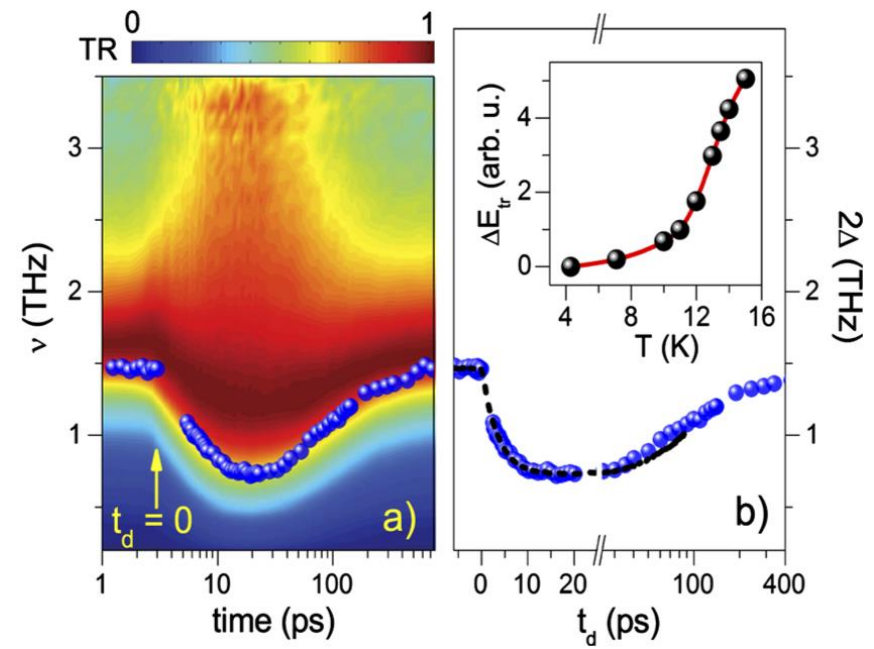
## Gap closing via $2\Delta$ phonons

### Rothwarf-Taylor model



Cross, Ph.D. Thesis (2009)

### Time-resolved THz spectroscopy



$$\tau_\Delta \gtrsim \tau_\beta \quad \Delta(t) \sim n(t)$$

Beck *et al.*, Phys. Rev. Lett. **107**, 177007 (2011)  
 Demsar, J. Low Temp. Phys. **201**, 676 (2020)

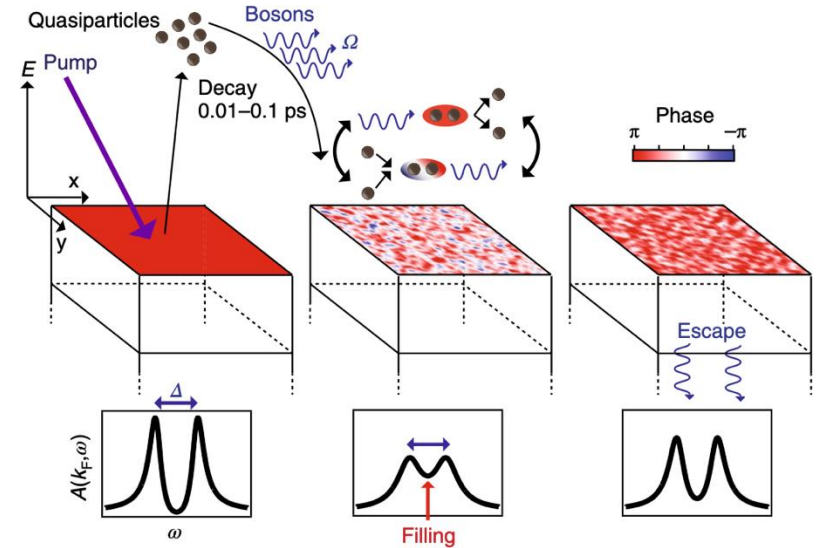
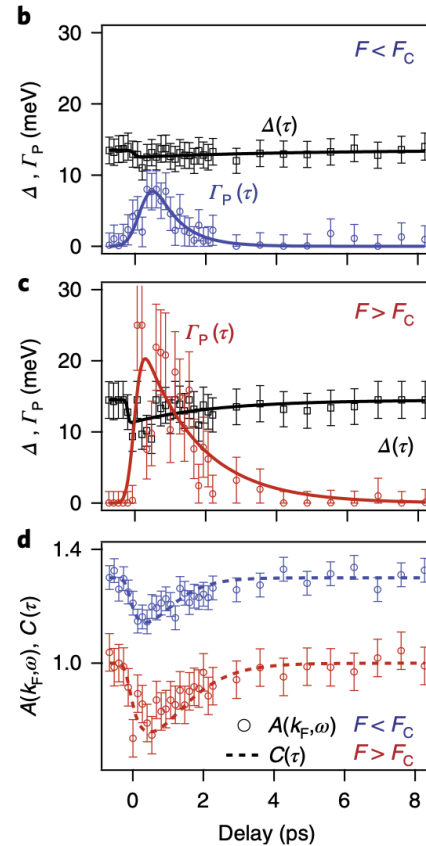
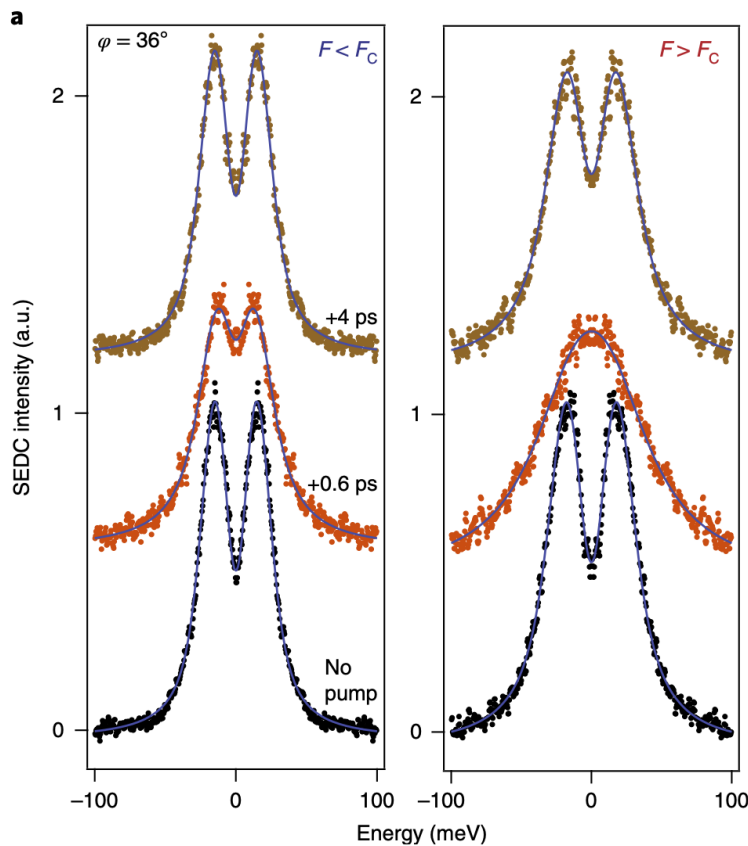
# Cuprate superconductor: $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (underdoped)

## Gap filling via phase fluctuations

Energy scale for phase fluctuations:

$$\hbar\Omega_\Theta = \frac{\hbar^2 n_S(0) a}{2m^*} \approx 15 \text{ meV}$$

$$\tau_\Theta = \frac{2\pi}{\Omega_\Theta} \approx 280 \text{ fs}$$



$$\tau_0 \approx 500 \text{ fs}$$

$$\tau_0 \approx 280 \text{ fs}$$

$$\tau_0 \approx \tau_\Theta \cdots \tau_{e-\Omega}$$

$$\tau_\Delta \approx \tau_\Theta$$

$$\Gamma_p(t) \sim T_\Theta^*(t)$$

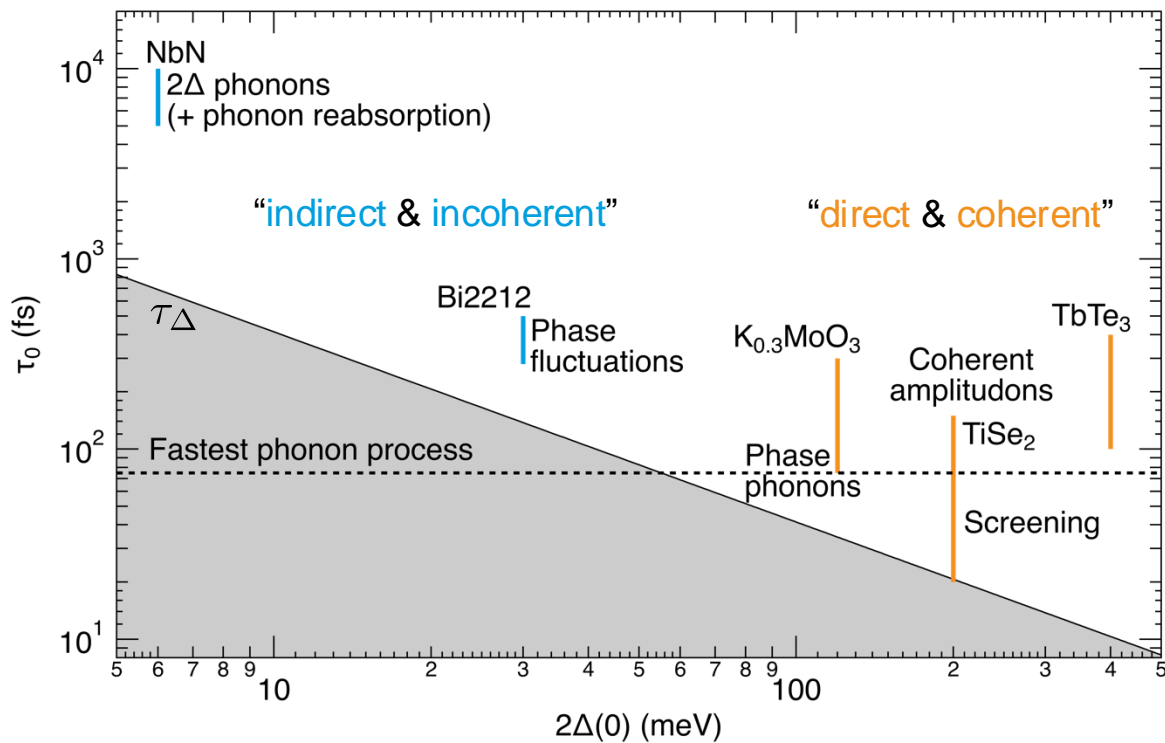
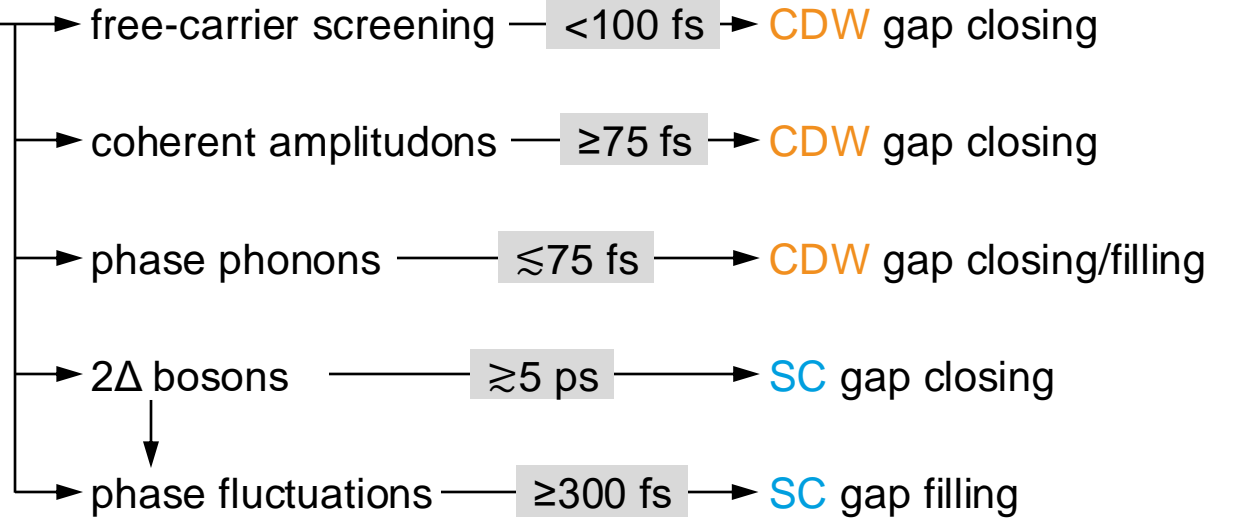
$$\Delta(t) \sim T_e^*(t)$$

Boschini *et al.*, Nat. Mater. **17**, 416 (2018)

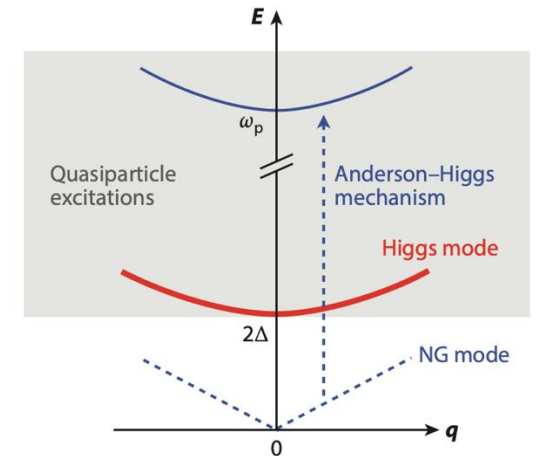
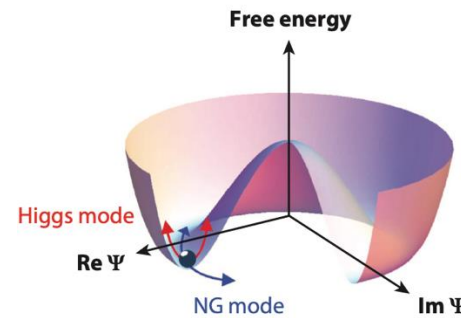
# Speed of ultrafast electronic unordering

(1) The more direct, the faster. (2) Electrons or phase modes for high speed.

Optical pump (nonresonant) → hot electrons



$$\tau_{\Delta} = \frac{h}{2\Delta(0)}$$



Shimano & Tsuji, Annu. Rev. Condens. Matter Phys. **11**, 103 (2020)

# Efficiency of ultrafast electronic unordering

(1) The more direct & coherent, the more efficient. (2) CDW for higher efficiency.

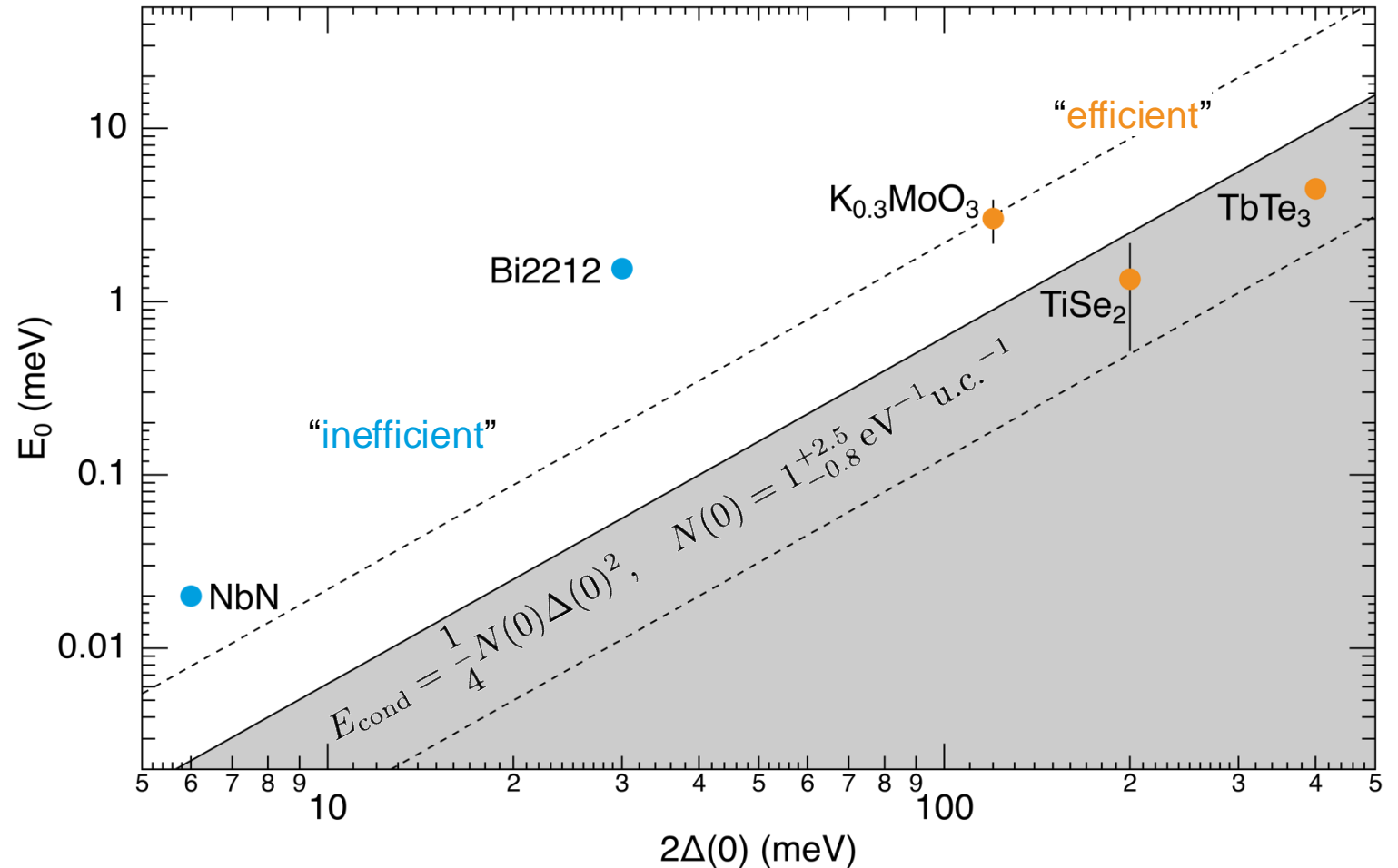
Condensation energy:

$$E_{\text{cond}} = \frac{1}{4} N(0) \Delta(0)^2$$

↑  
per both spin states

“Vaporization energy”:

$$E_0 = \frac{(1 - R) F_{\text{th}}}{\lambda_{\text{opt}}}$$



Stojchevska *et al.*, Phys. Rev. B **84**, 180507(R) (2011)

# Quest for a new switch: CDW switch?

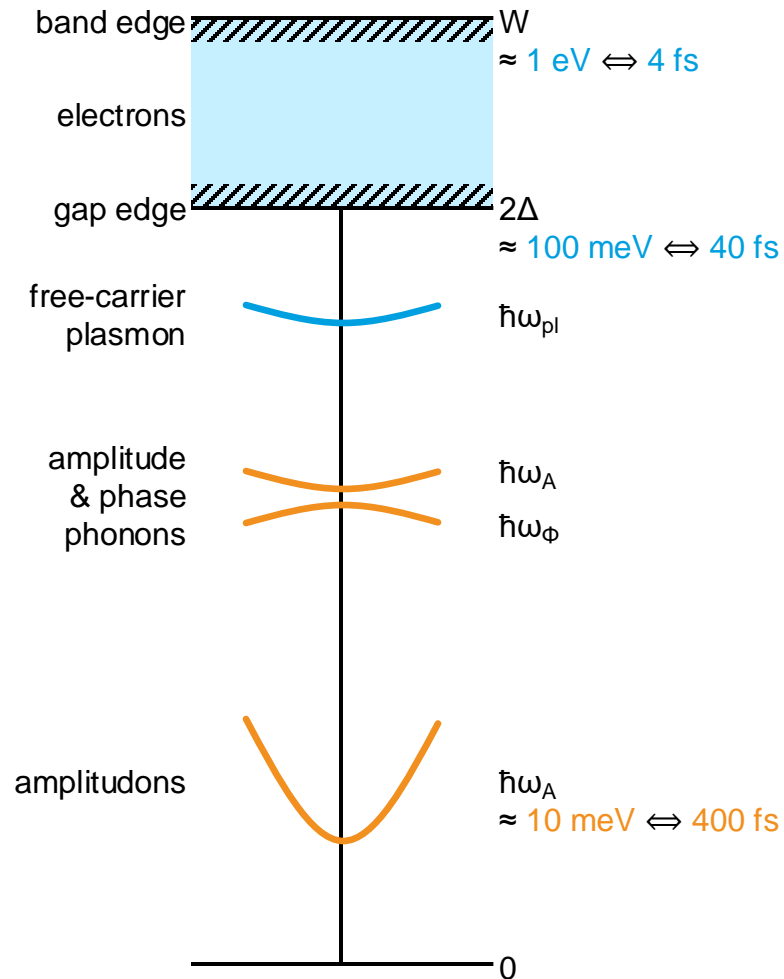
## Energy–time–size comparison

FinFET ("3 nm" node, IRDS / IEEE 2022)	Charge-density-wave switch	Shannon–von Neuman–Landauer limit
$E_{\text{switch}} \approx 10 \text{ aJ} = 62 \text{ eV}$	$E_{\text{switch}} \approx \frac{1}{4} N_{2\text{D}} \Delta(0)^2 \stackrel{!}{=} 53 \text{ meV}$ $N_{2\text{D}} = \frac{m_e}{\pi \hbar^2} = 4.2 \frac{1}{\text{eV nm}^2}$ $\Delta(0) = 1.75 k_{\text{B}} T_c \stackrel{300 \text{ K}}{=} 45 \text{ meV}$	$E_{\text{min}} = k_{\text{B}} T \ln 2 \stackrel{300 \text{ K}}{=} 18 \text{ meV}$
$\tau_{\text{switch}} \approx 1 \text{ ps}$	$\tau_{\text{switch}} \geq \frac{\hbar}{2\Delta(0)} \approx 46 \text{ fs}$	$\tau_{\text{min}} = \frac{\hbar}{E_{\text{min}}} \stackrel{300 \text{ K}}{=} 37 \text{ fs}$
$V_{\text{switch}} \approx 4000 \text{ nm}^3$	$A_{\text{switch}} \approx (5 \times 5) \text{ nm}^2$ $[\approx (15 \times 15) \text{ unit cells}]$ <p>(liquid-N operation)</p>	$x_{\text{min}} = \frac{\hbar}{\sqrt{2m_e E_{\text{min}}}} \stackrel{300 \text{ K}}{=} 1.5 \text{ nm}$ <p>Zhirnov <i>et al.</i>, Proc. IEEE <b>91</b>, 1934 (2003)</p>

# Ultrafast CDW unordering: Mechanisms & energy (time) scales

## Tabular summary & open challenges

	Coherent Displacive Gap closing ( $\Delta$ )	Incoherent Order-disorder Gap filling ( $\Gamma$ )
<b>Electron-</b> dominated	Free-carrier screening ( $\omega_{pl}$ )  Coherent gap oscillation ( $\Delta$ )?	Electron hopping ( $W$ )?
<b>Phonon-</b> dominated	Coherent amplitudons ( $\omega_A$ )	Incoherent amplitude phonons ( $\omega_A$ )  Incoherent phase phonons ( $\omega_\phi$ )



- See how **incoherent electrons** hop ( $W^{-1}$ ) and fill the gap.
- See how **coherent electrons** oscillate ( $\Delta^{-1}$ ) and close the gap.
- See how **coherent amplitudons and coherent electrons** oscillate simultaneously ( $\omega_A^{-1}, \Delta^{-1}$ ).

# Outline

... a reflection of my (narrow) world view

## Ultrafast Unordering of Electronic Order

### I. Motivation

Quest for a new switch.

### II. Notes on charge-density waves

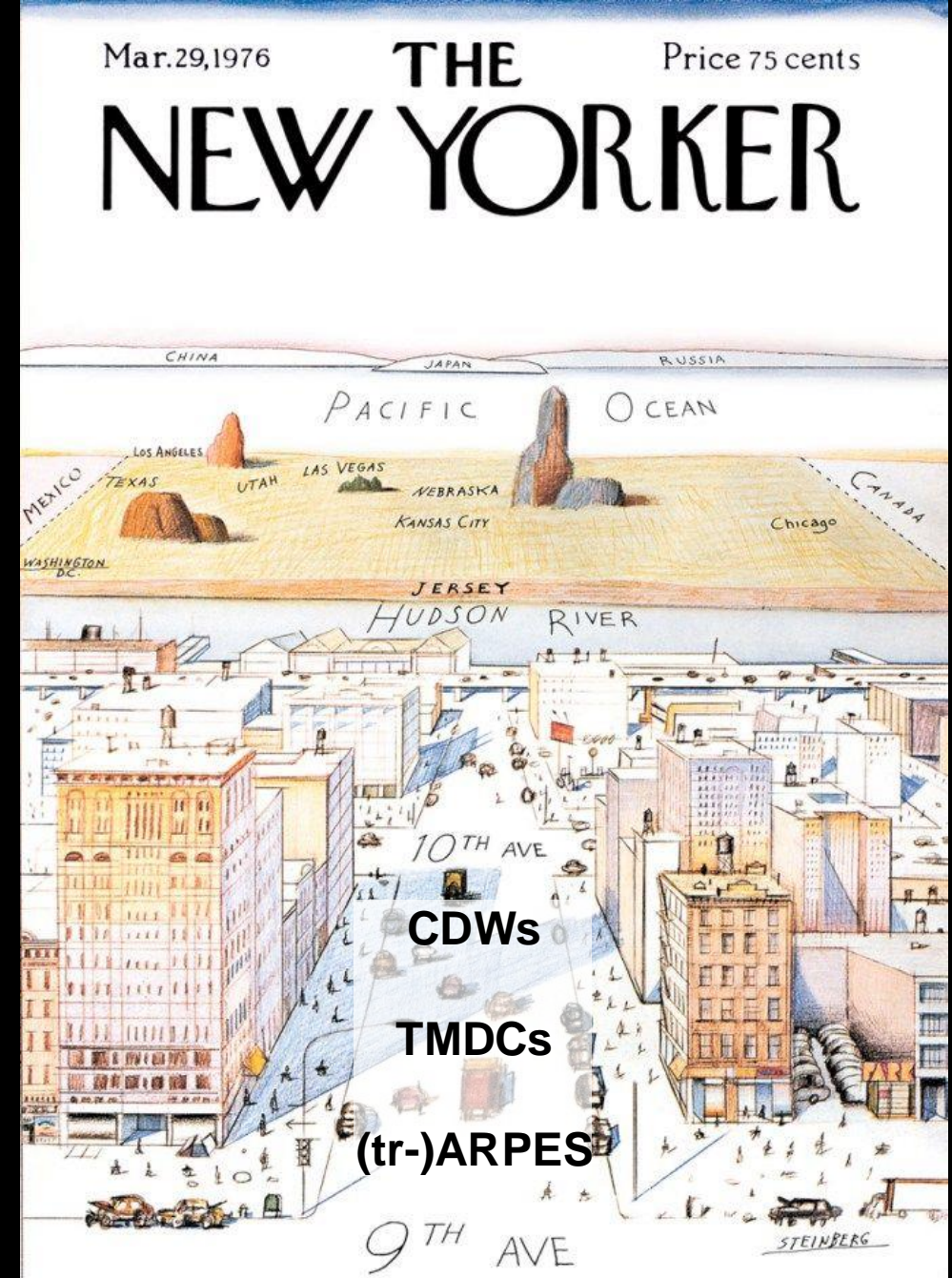
Nature & origin (“types”).

### III. Ultrafast unordering (of CDWs)

Mechanisms. Speeds. Efficiencies.  
Time-domain classification.

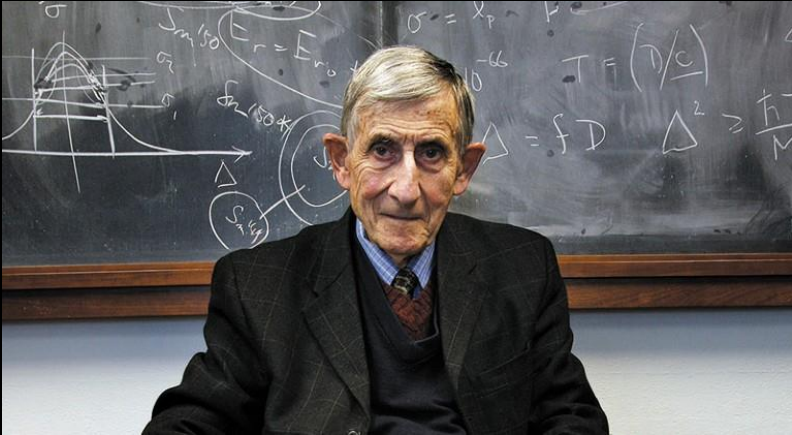
### IV. Outlook

Tools & concepts.



# Outlook

## Tools & concepts



Freeman Dyson

Physics World 6(8), 33 (1993)

Scientific revolutions are more often driven by **new tools** than by new concepts.

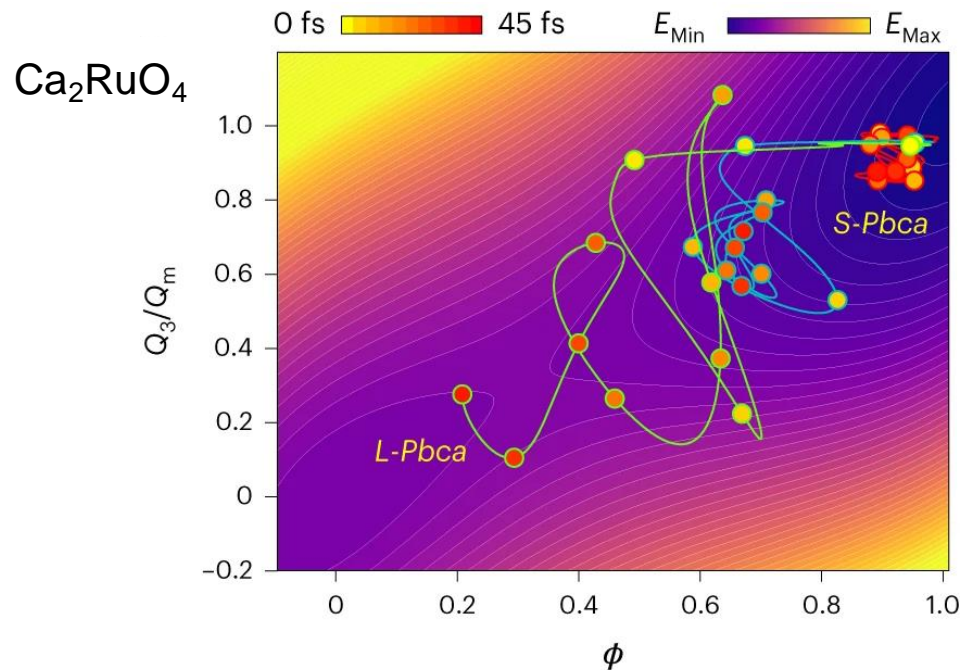
The effect of a concept-driven revolution is to explain old things in new ways.

The effect of a tool-driven revolution is to **discover new things** that have to be explained.

# Concepts

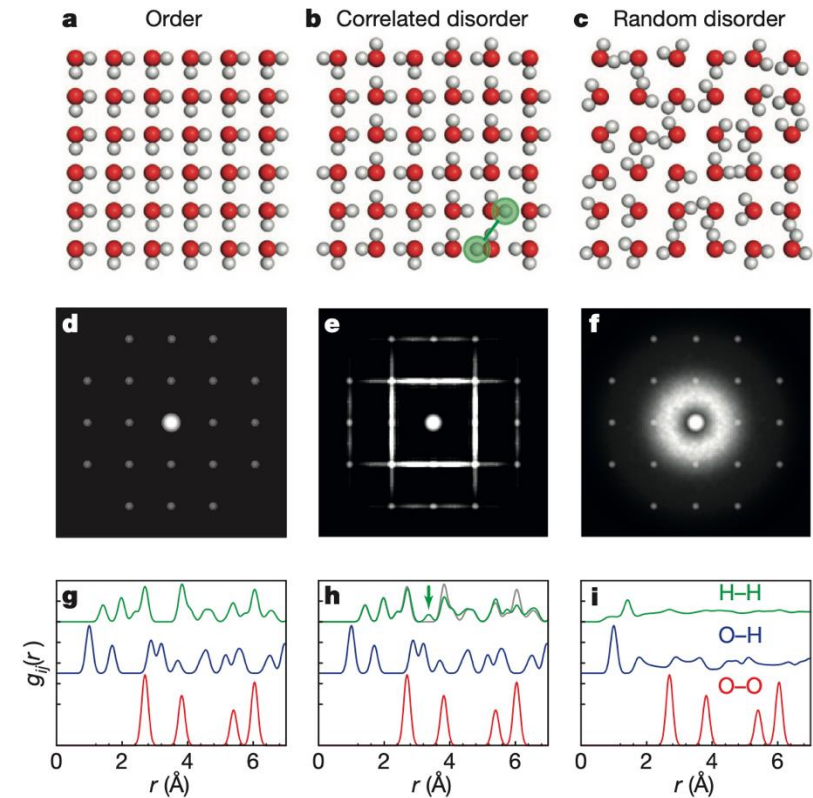
... can explain complex things in an intuitive way

Motion in (dynamic) energy landscapes  
("coherent", quantitative)



Verma *et al.*, Nat. Phys. **20**, 807 (2024)  
Georgescu & Millis, Commun. Phys. **5**, 135 (2022)

Correlated (dynamic) disorder  
("partially incoherent", qualitative)



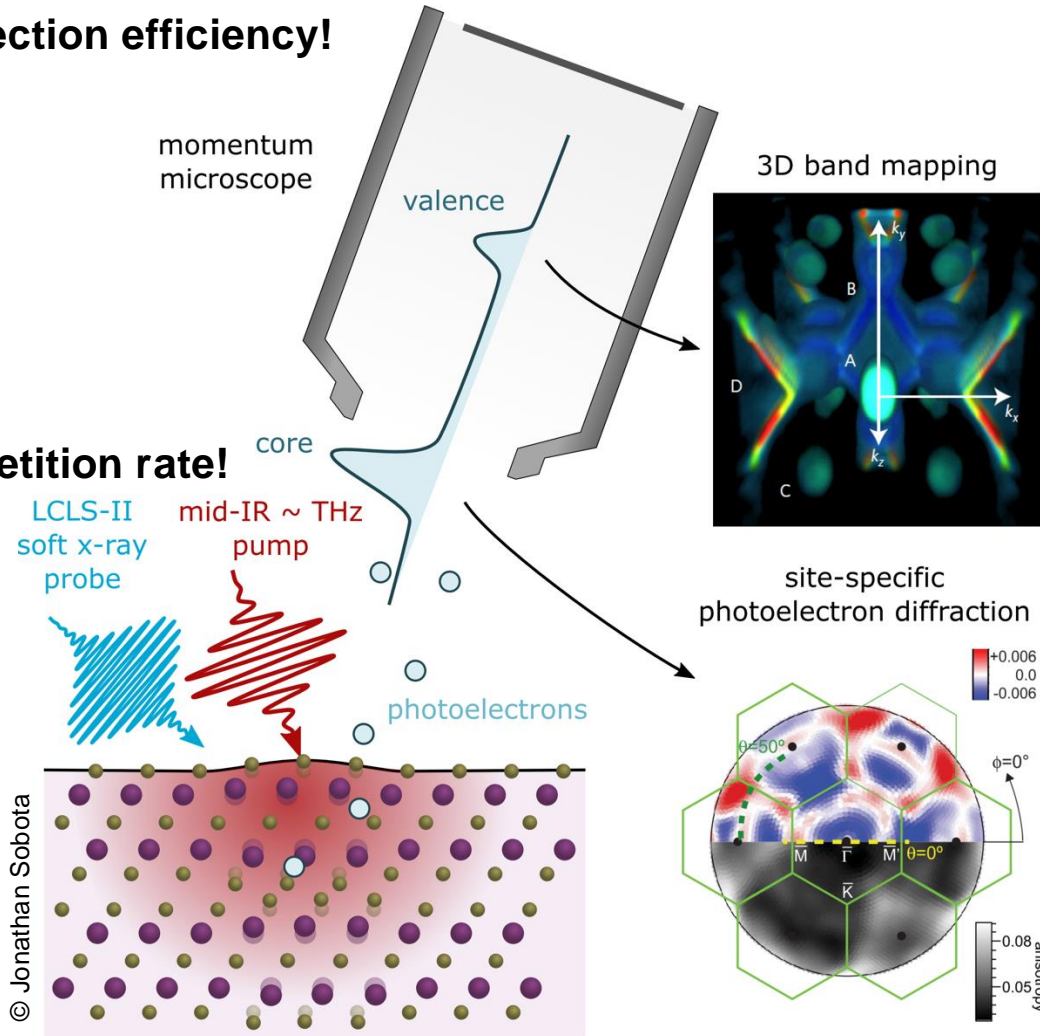
Keen & Goodwin, Nature **521**, 303 (2015)  
Simonov & Goodwin, Nat. Rev. Chem. **4**, 657 (2020)

# Novel tool: *All-in-one* ultrafast photoelectron spectroscopy

Simultaneous multimodal observation of *electron & lattice (& spin) dynamics*

Detection efficiency!

Repetition rate!



© Jonathan Sobota

femtosecond time resolution (via pump-probe)

+

tunable & shapable THz-MIR-NIR pump

- low-energy (1 meV...1.6 eV) resonances
- spin & orbital angular momentum
  - strong fields

+

tunable & shapable soft x-ray probe

- interface sensitivity
- 3D momentum selectivity
  - core resonances
  - forward scattering

+

3D electron energy-momentum imaging (without slit)

+

micrometer spatial resolution (PEEM mode, field aperture)

+

2D spin detection (spin filter)

=

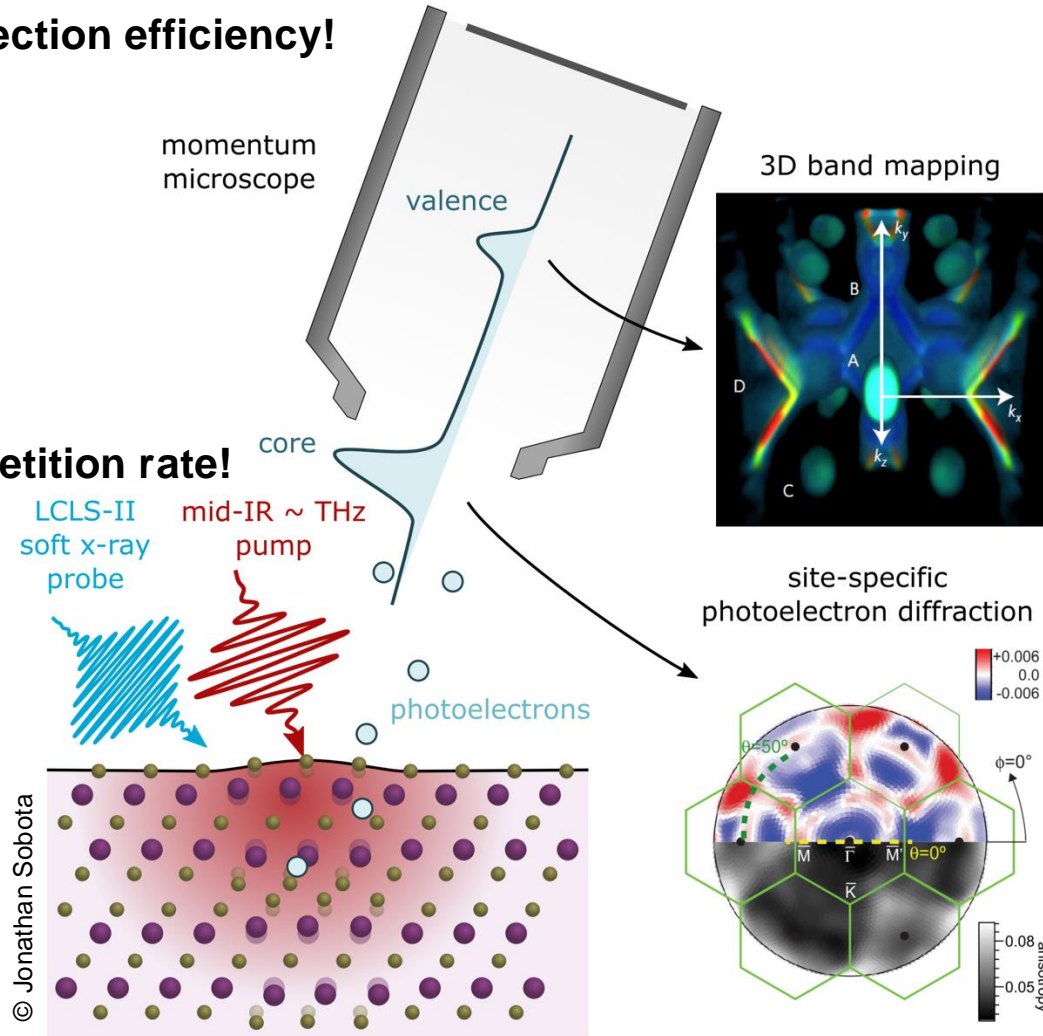
**complete ultrafast soft x-ray PES**

# Novel tool: *All-in-one* ultrafast photoelectron spectroscopy

Simultaneous multimodal observation of *electron & lattice (& spin) dynamics*

Detection efficiency!

Repetition rate!



NEWS | 18 September 2023

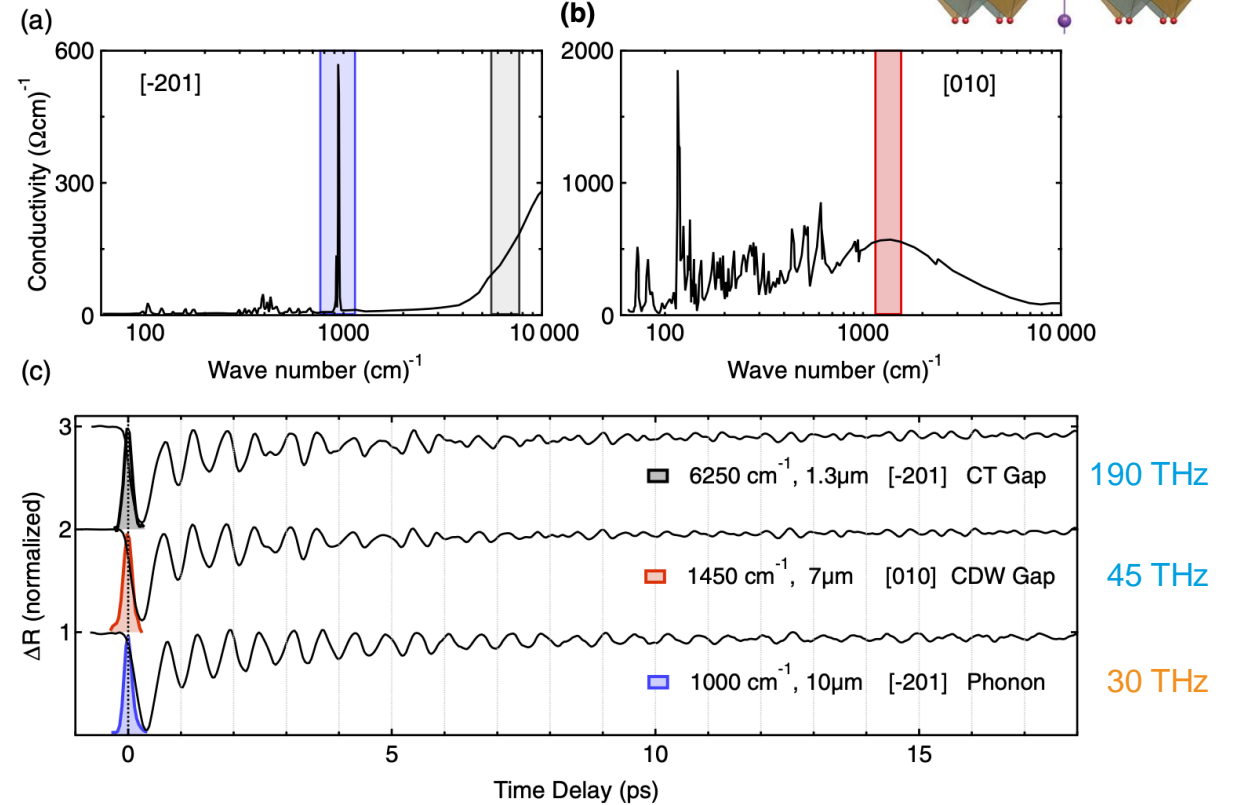
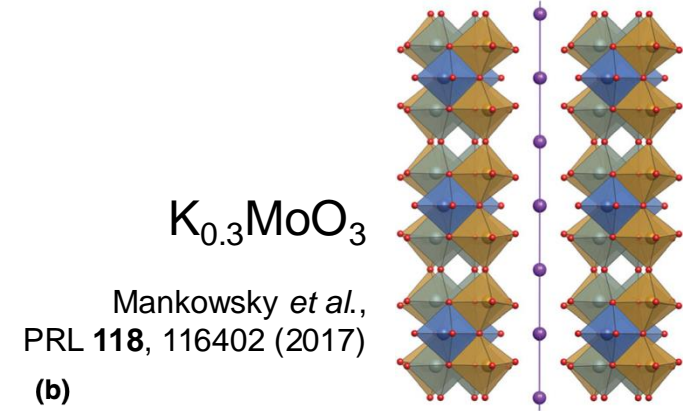
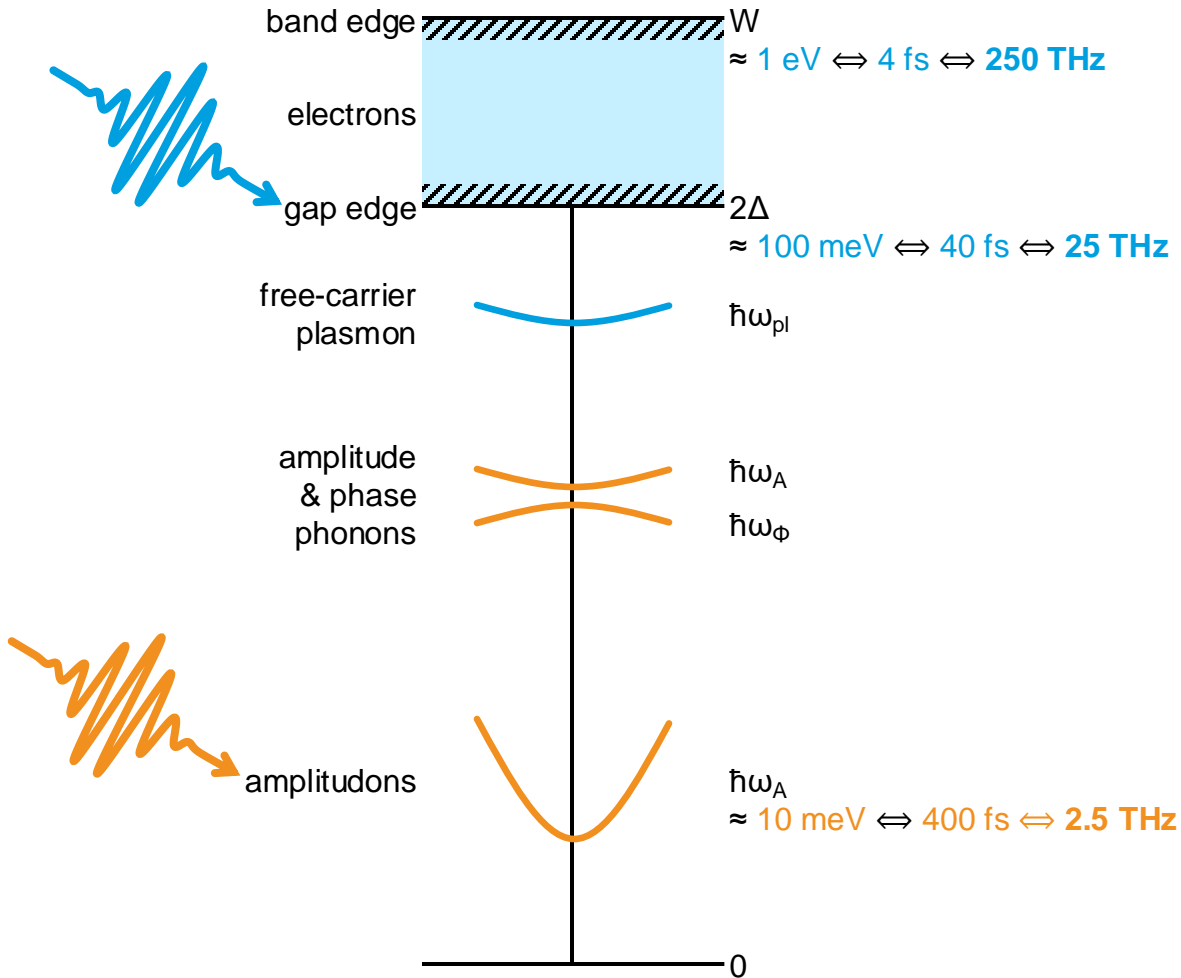
## World's most powerful X-ray laser will 'film' chemical reactions in unprecedented detail

Upgraded laser in California will produce one million X-ray pulses per second to study ultrafast processes at the atomic level.



# “Novel” tool: Photon scalpel instead of hammer

Toward selective & efficient electronic order unordering

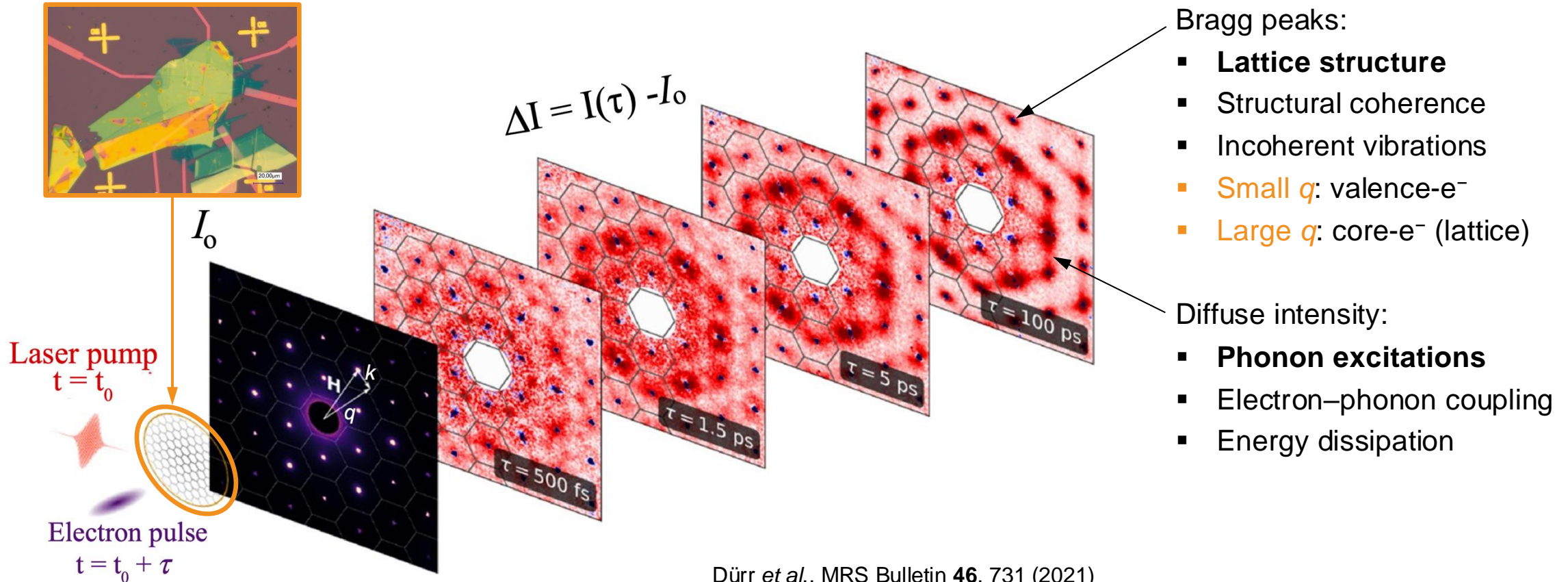


# “Novel” tool: MeV ultrafast electron diffraction

## Femto-tomography of intertwined *structural & electronic* dynamics

MeV electron pulses:

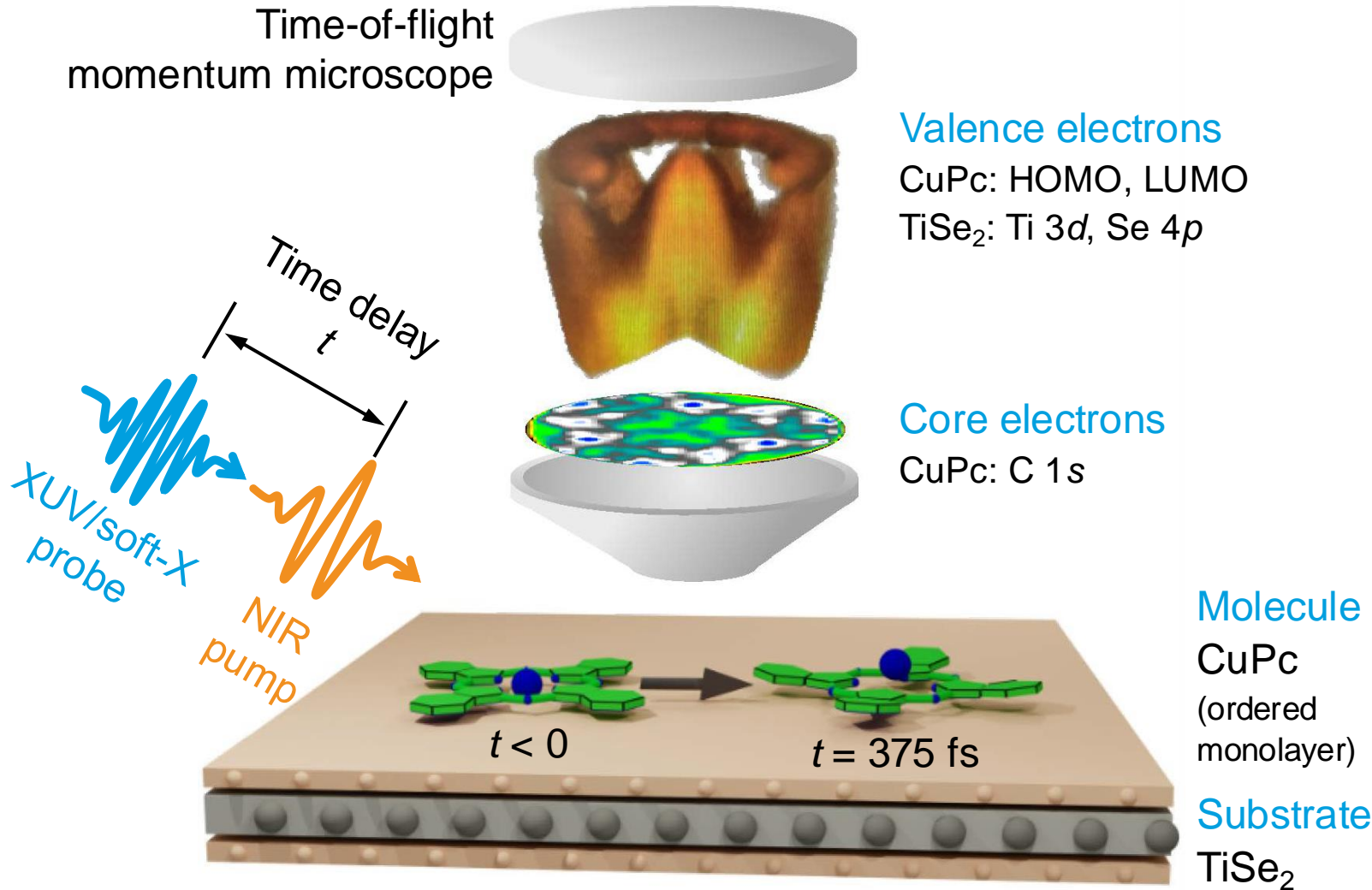
- <50 fs pulse duration
- >100 nm penetration depth



Dürr *et al.*, MRS Bulletin **46**, 731 (2021)

# Pump-probe “all-in-one” soft x-ray femto-PES

Ultrafast multiplexed electron cinematography of a molecule / 2D material interface



Femtosecond dynamics of

molecular orbitals  
band structure  
charge carriers

+

chemical states  
local structure



Kiana Baumgärtner

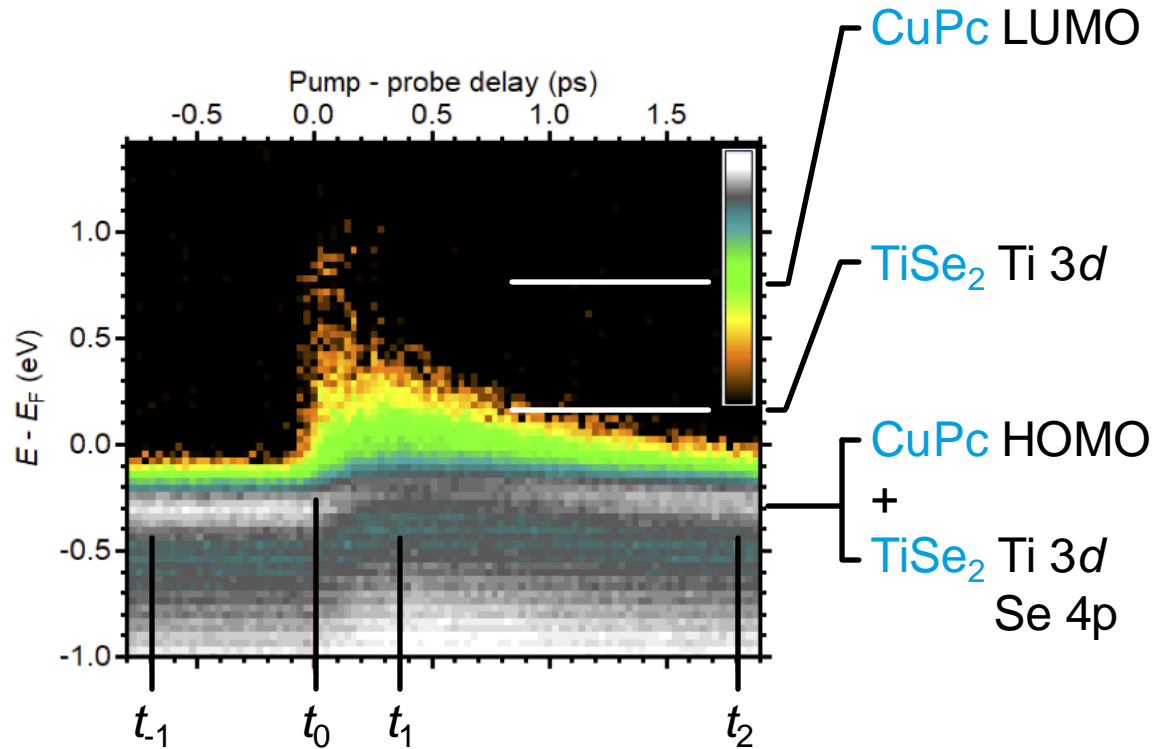


Markus Scholz

# Carrier & electronic structure dynamics

## Time-resolved momentum microscopy (ARPES)

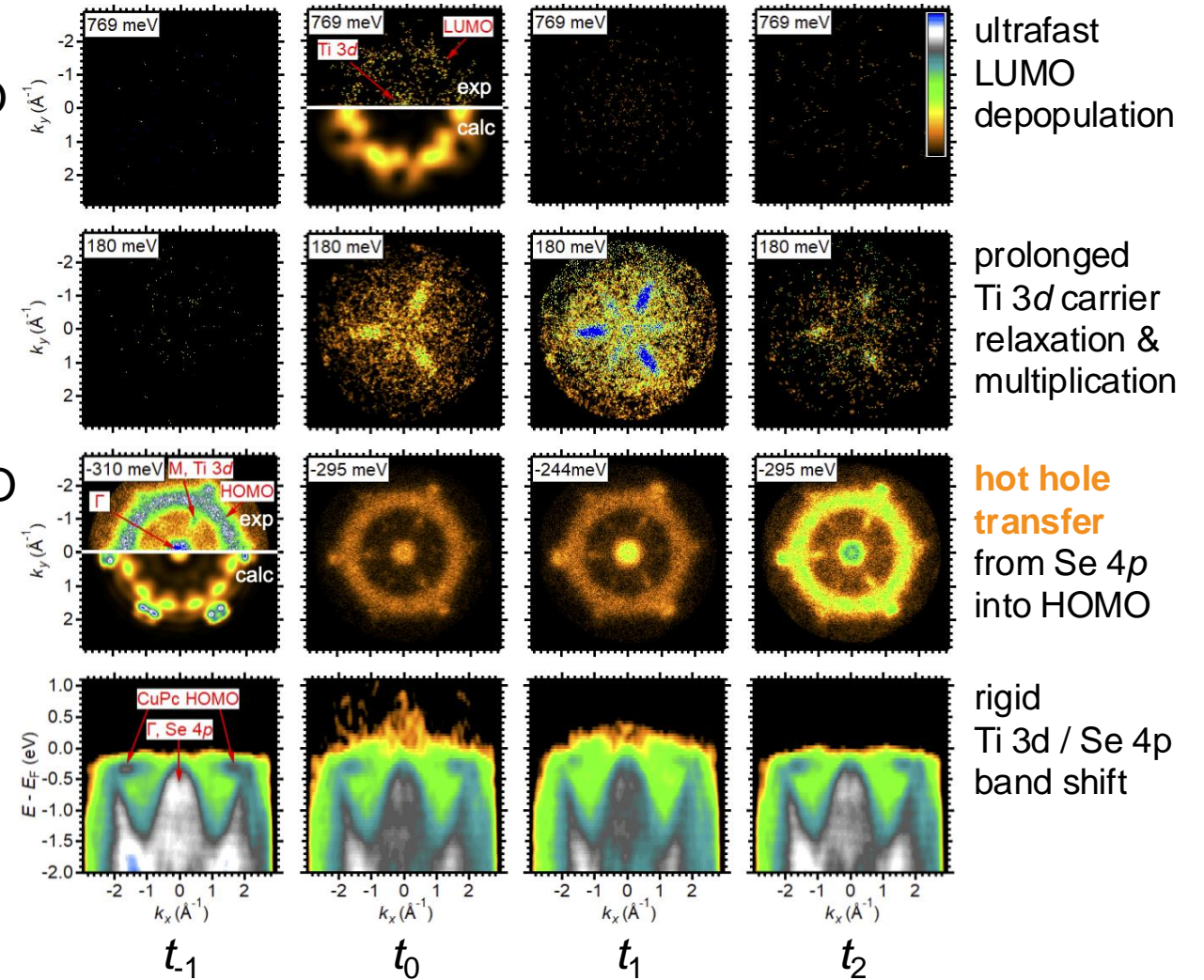
Baumgärtner *et al.*, arXiv:2305.07773



$$h\nu_{\text{probe}} = 36.3 \text{ eV}$$

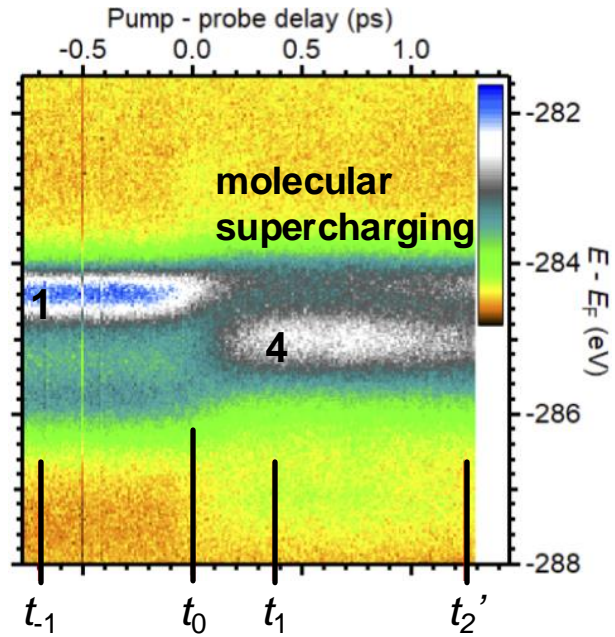
$$h\nu_{\text{pump}} = 1.6 \text{ eV}$$

$$F \approx 3.5 \text{ mJ/cm}^2$$



# Chemical & structural dynamics

## Time-resolved C 1s XPS & XPD

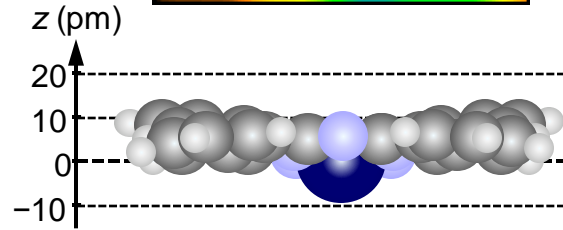
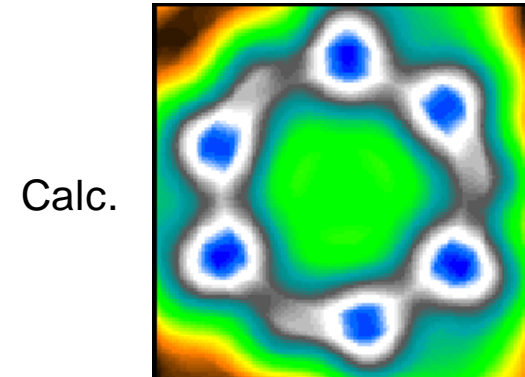
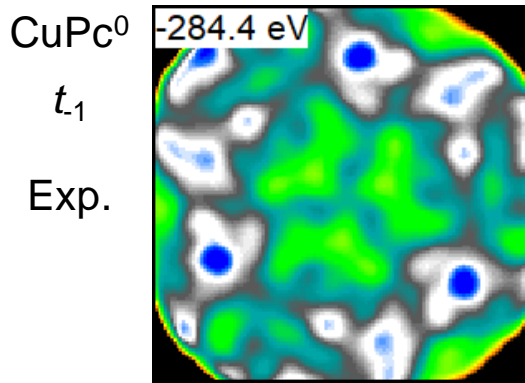
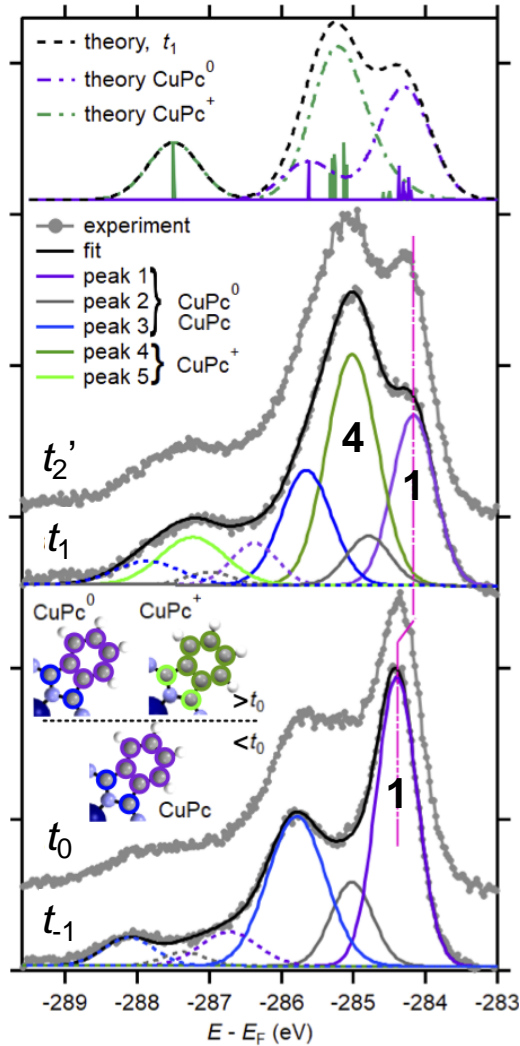


Theory:  
 XPS: Marvin Reuner, Daria Gorelova  
 XPD: Kaori Niki, Masato Nozakami,  
 Misa Nozaki

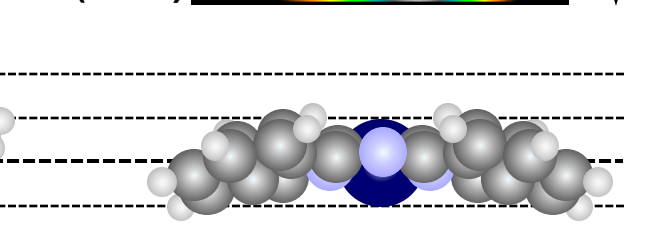
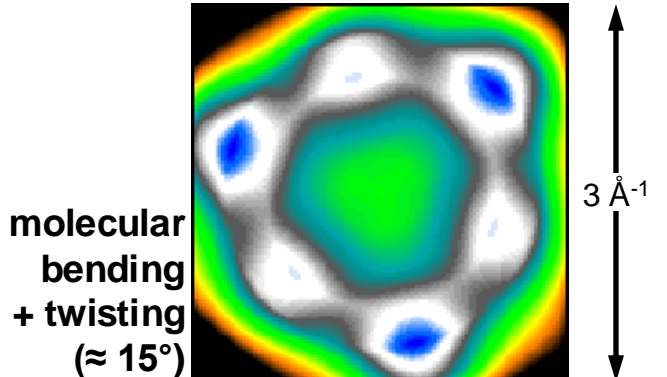
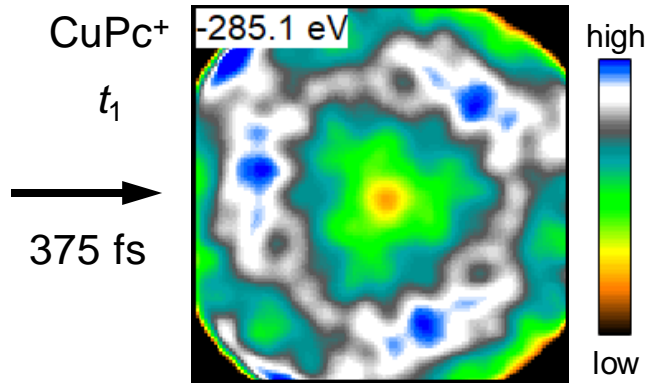
$$h\nu_{\text{probe}} = 371 \text{ eV}$$

$$h\nu_{\text{pump}} = 1.6 \text{ eV}$$

$$F \approx 3.5 \text{ mJ/cm}^2$$



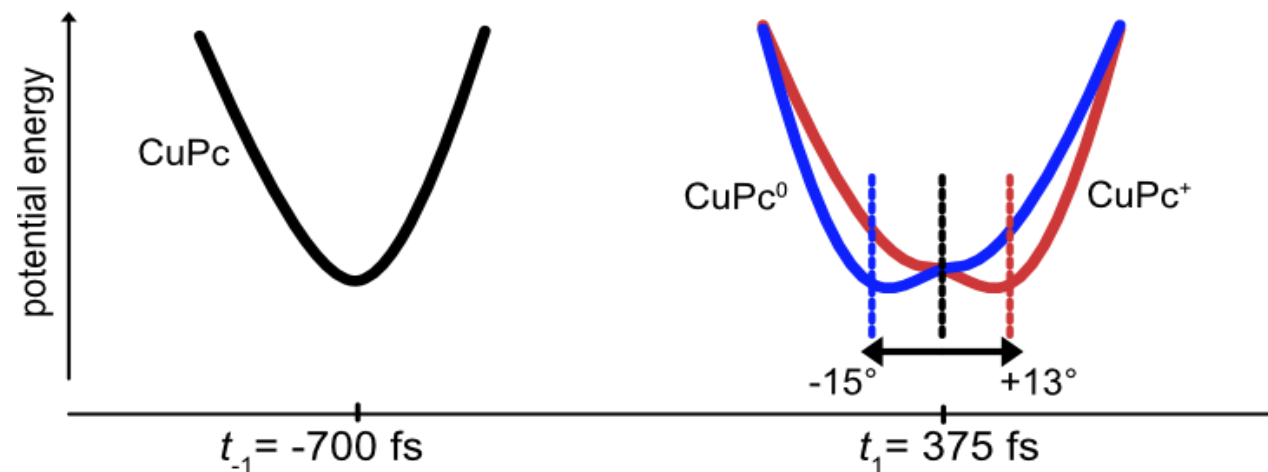
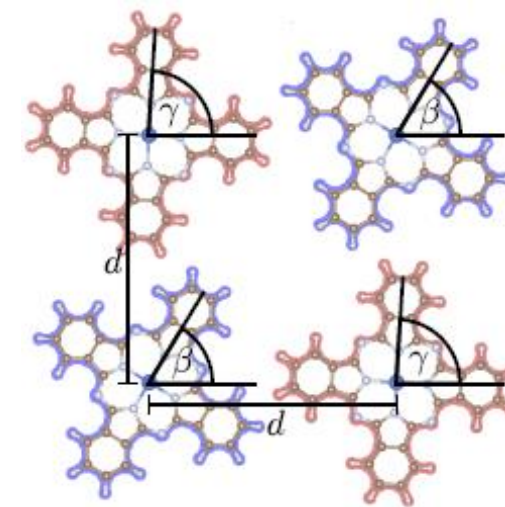
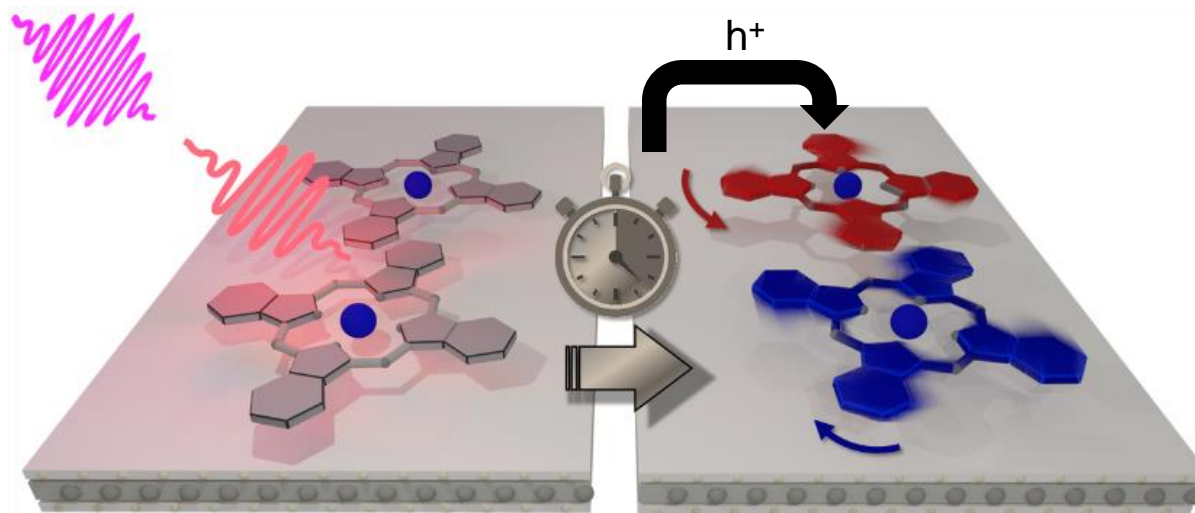
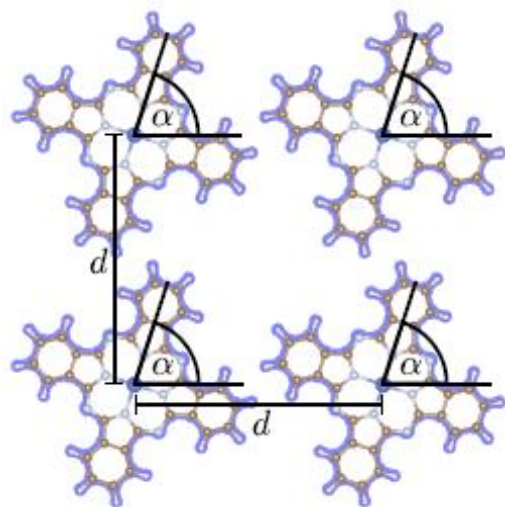
Baumgärtner *et al.*, arXiv:2305.07773



# Charge-driven concerted rotation of molecules

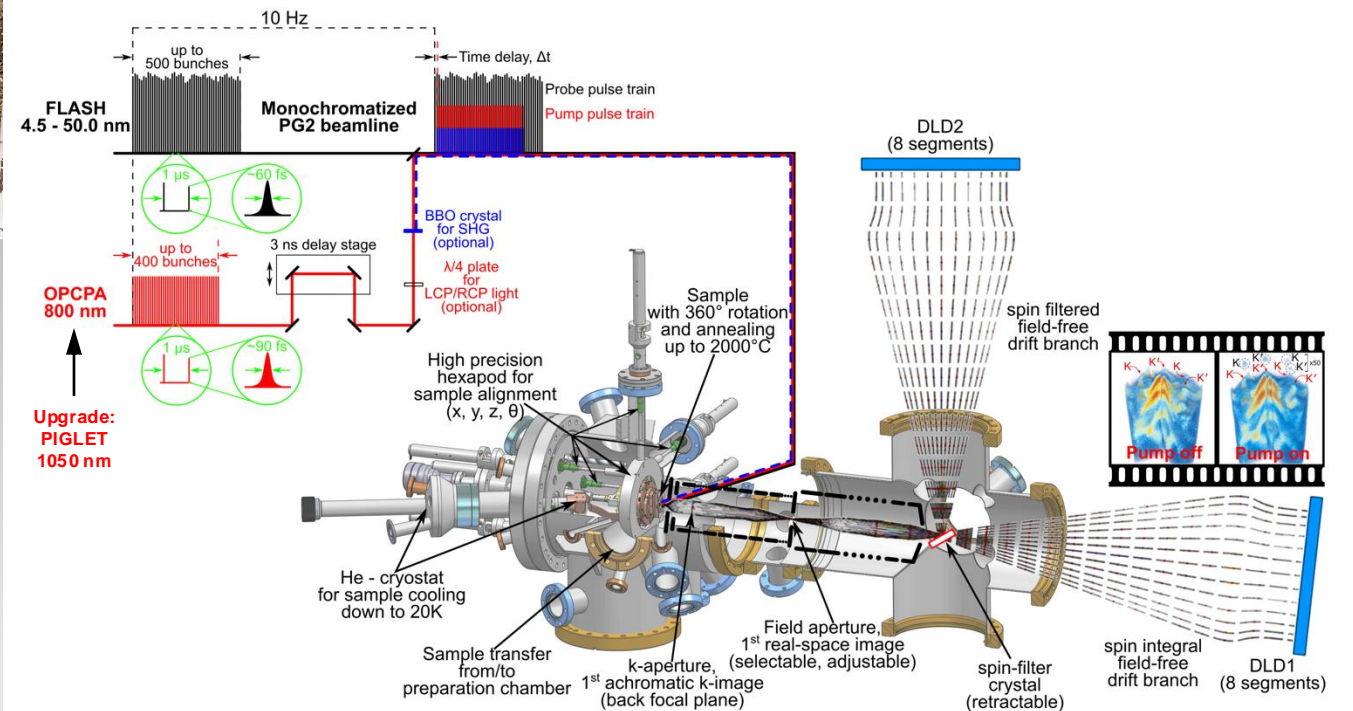
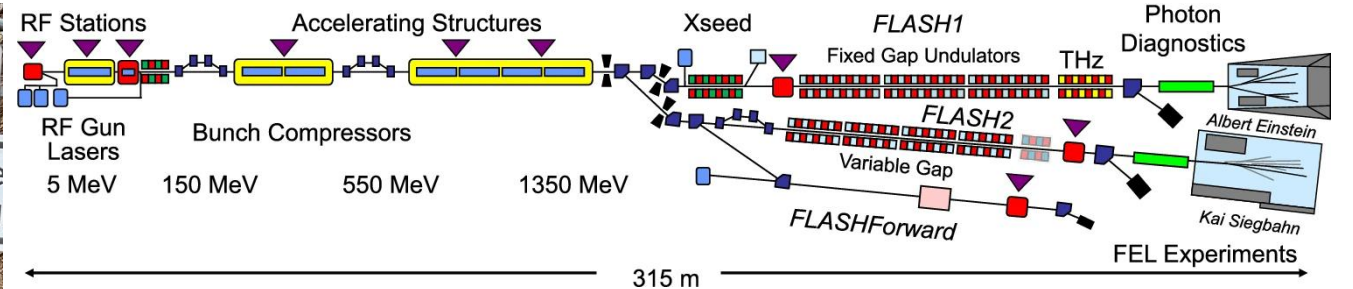
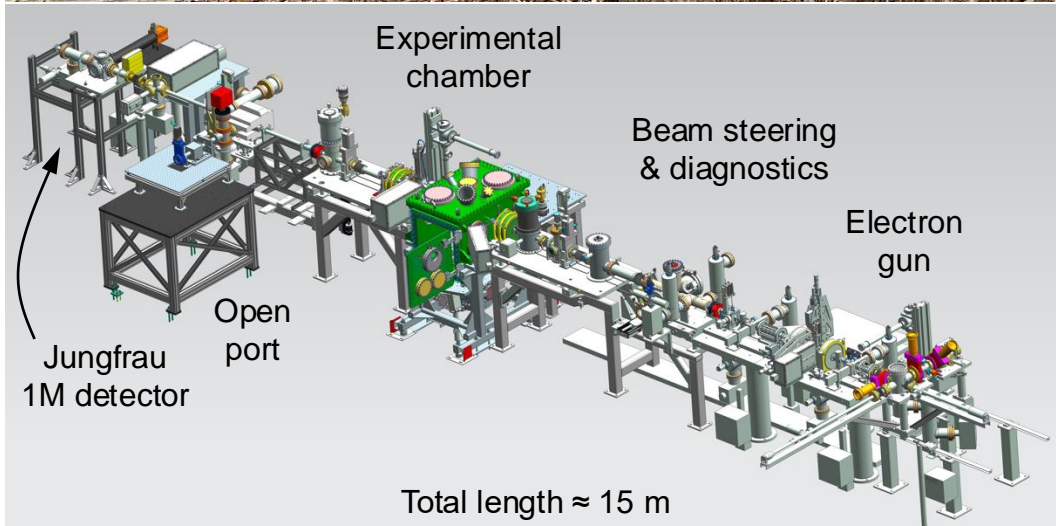
Molecular charging  $\rightarrow$  modification of potential energy surface  $\rightarrow$  ultrafast molecular gear motion

Baumgärtner *et al.*, arXiv:2305.07773



# Commercial break: MeV-UED + FEL k-microscopy @ DESY

Complementary observation of ultrafast *lattice & electronic* structure dynamics: REGAE + FLASH



Kutnyakhov *et al.*, Rev. Sci. Instrum. **91**, 013109 (2020)

# Heading

Subheading, optional