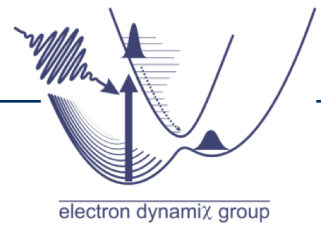




MAX-PLANCK-GESELLSCHAFT



Ultrafast structural dynamics and electronic band gap control in Ta_2NiSe_5

Selene Mor

Fritz Haber Institut of the MPG, Berlin, Germany

Università Cattolica del Sacro Cuore, Brescia, Italy

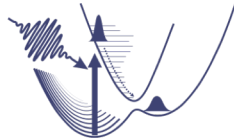
Materials in Quantum Cavities and Excitonic Insulators

CCQ-Flatiron Institute, 12th July 2019

Acknowledgements

Ultrafast spectroscopies:

Julia Stähler



electron dynamics group
(Fritz Haber Institut Berlin)

Claude Monney (Univ. Fribourg)

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Philipp Werner (Univ. Fribourg)

Martin Eckstein (Univ. Erlangen)

ARPES:

Takashi Mizokawa (Univ. Waseda)

Raman Spectroscopy:

Johannes Noack (FHI Berlin)

Annette Trunschke (FHI Berlin)

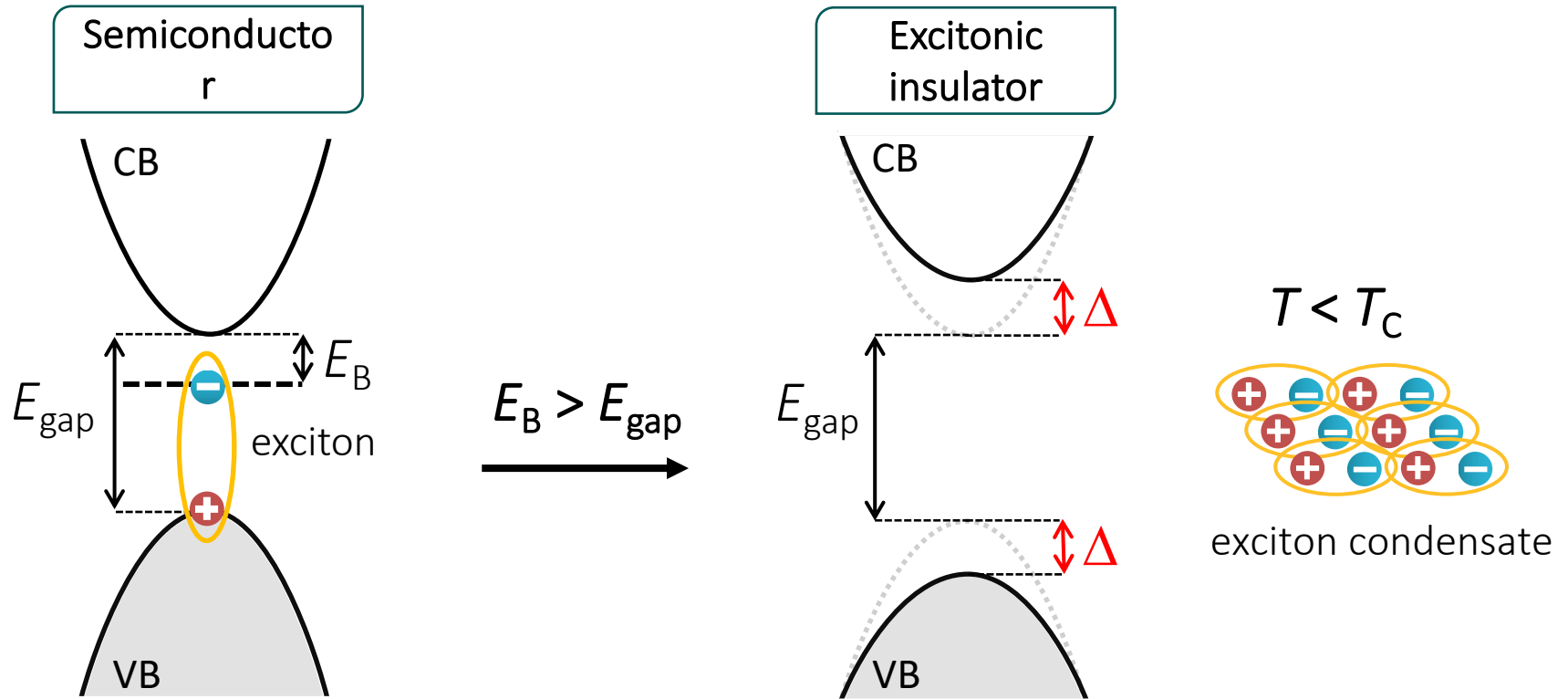
Sample:

Naoyuki Katayama (Univ. Nagoya)

Minoru Nohara (Univ. Okayama)

Hide Takagi (MPI Stuttgart)

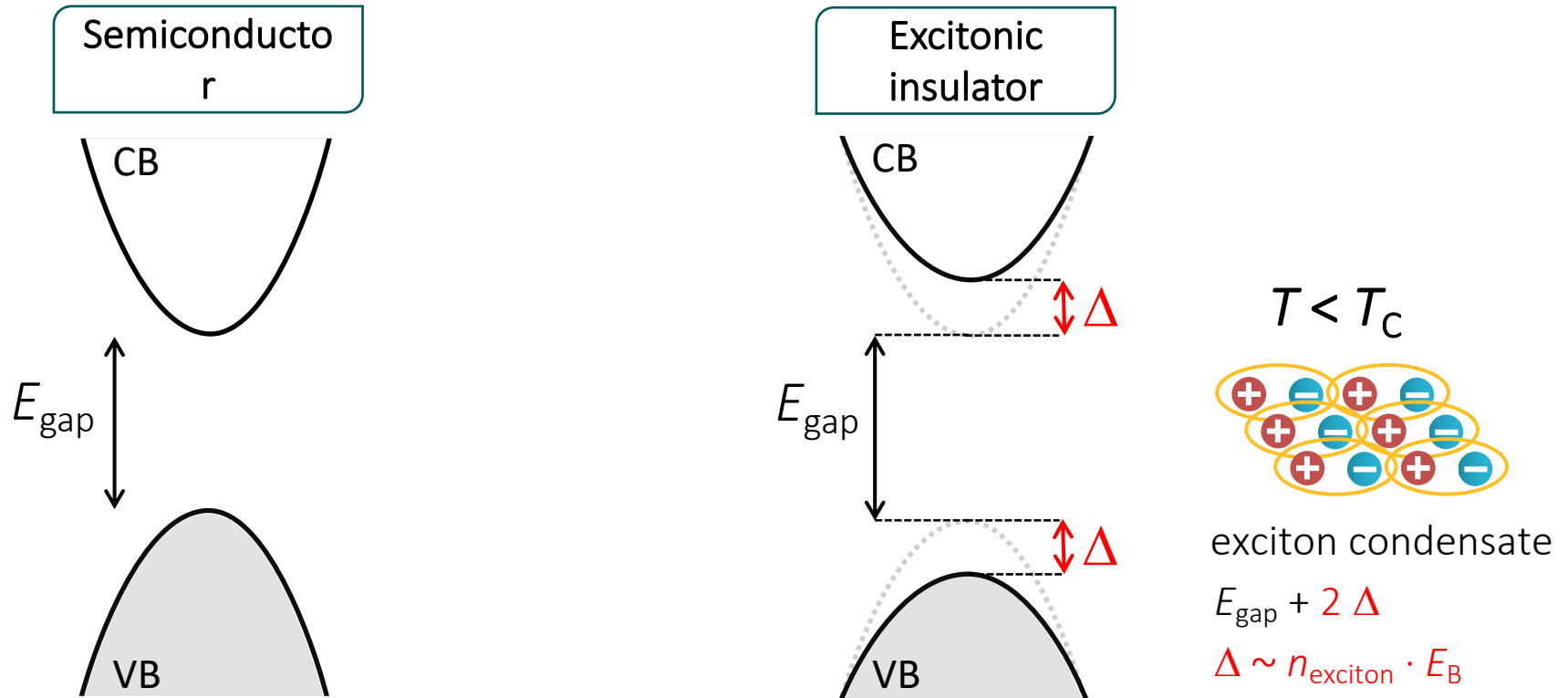
The excitonic insulator phase



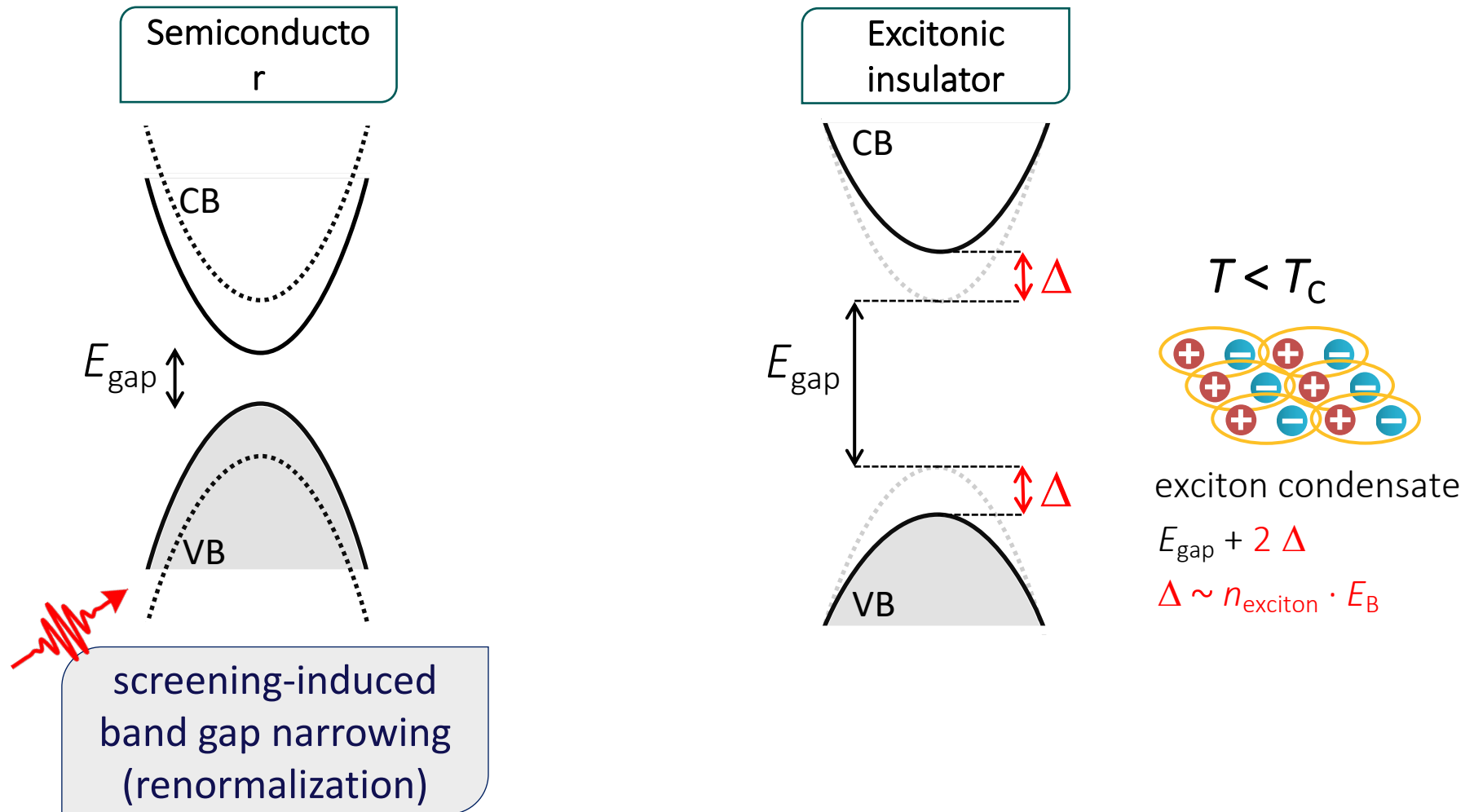
- $E_{\text{gap}} + 2 \Delta$
- $\Delta \sim n_{\text{exciton}} \cdot E_B$ (order parameter)
- combined lattice distortion

Halperin, *Rev. Mod. Phys.* 40, 4, 755 (1968)
 Monney, *New J. of Phys.* 12, 125019 (2010)

Photoexcitation of an excitonic insulator?

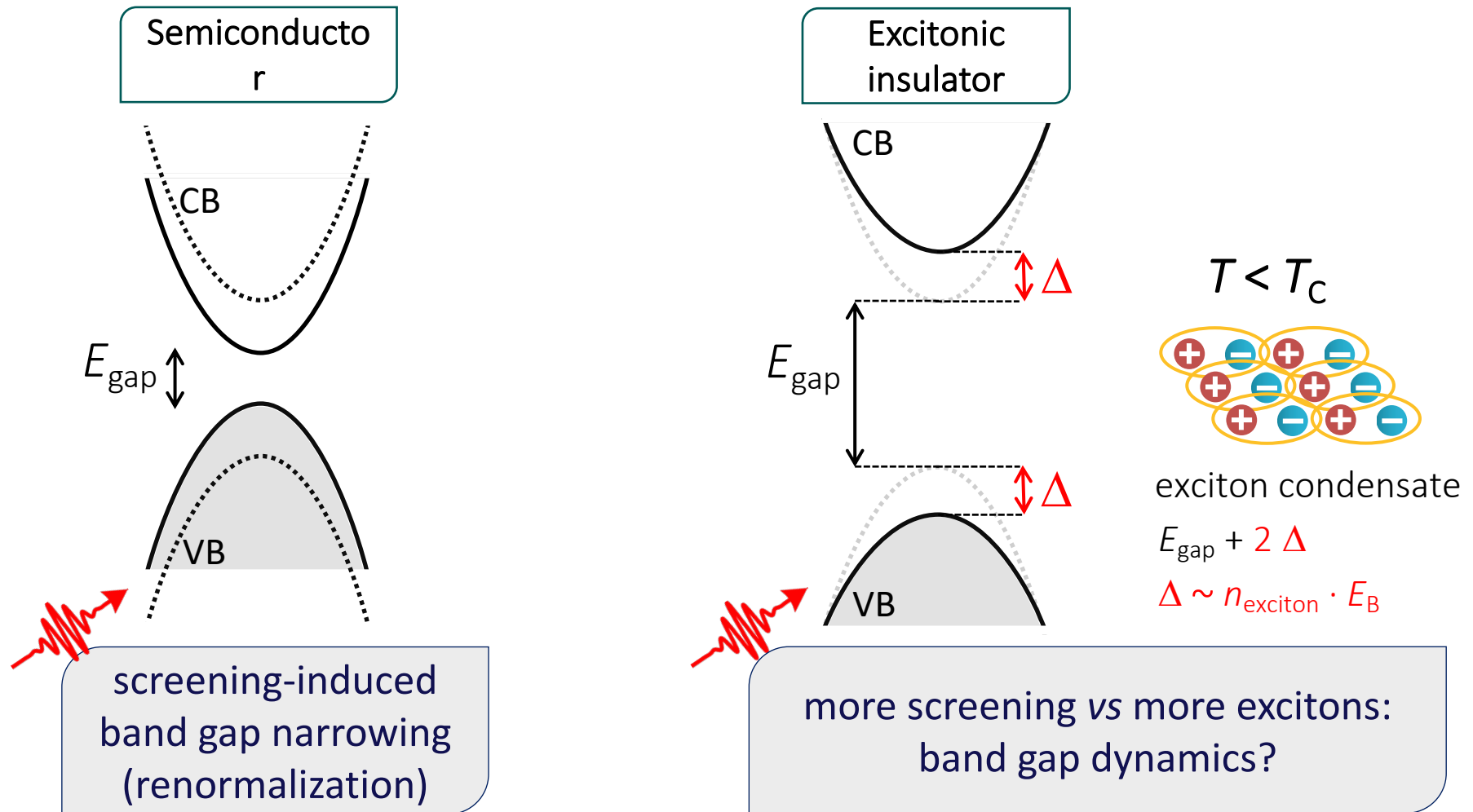


Photoexcitation of an excitonic insulator?



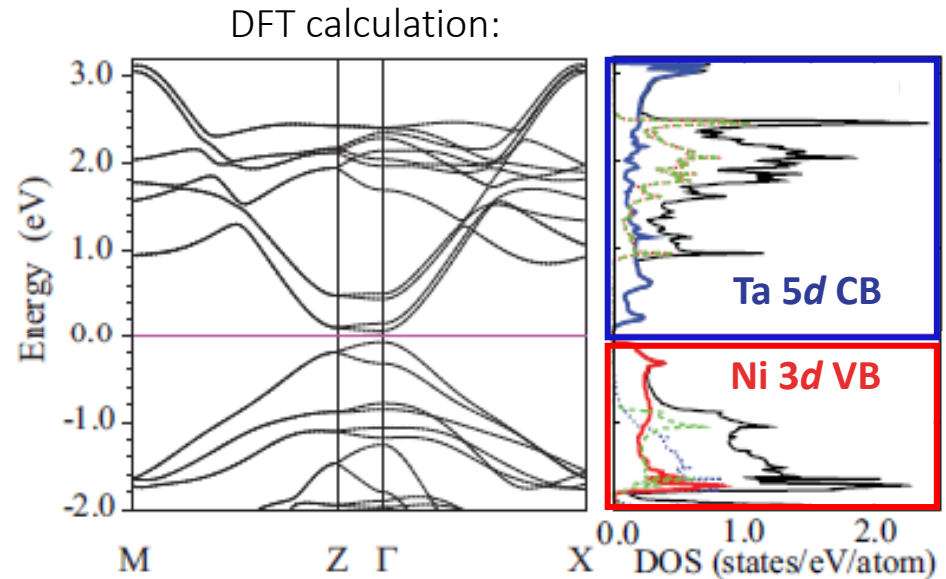
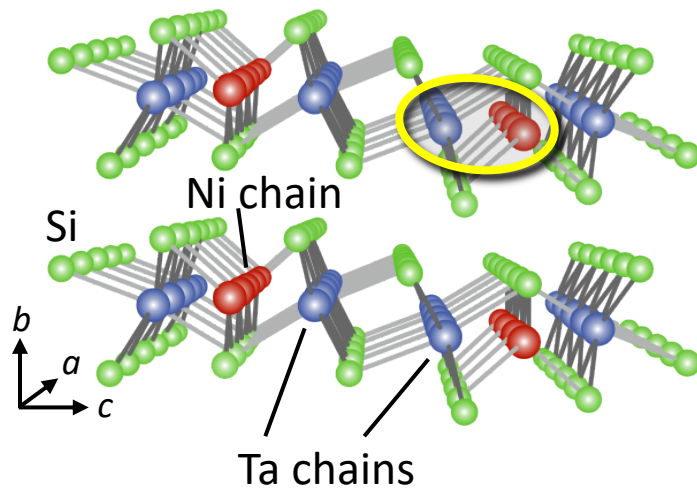
Si and Ge: Berggren, *PRB* 24, 1971 (1981)
 Graphite: Pagliara, *JACS* 133, 6318 (2011)
 WS₂: Chernikov, *Nat. Photonics* 9, 466 (2015)
 MoS₂: Pogna, *AC Nano* 10, 1182 (2016) ...

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Si and Ge: Berggren, *PRB* 24, 1971 (1981)
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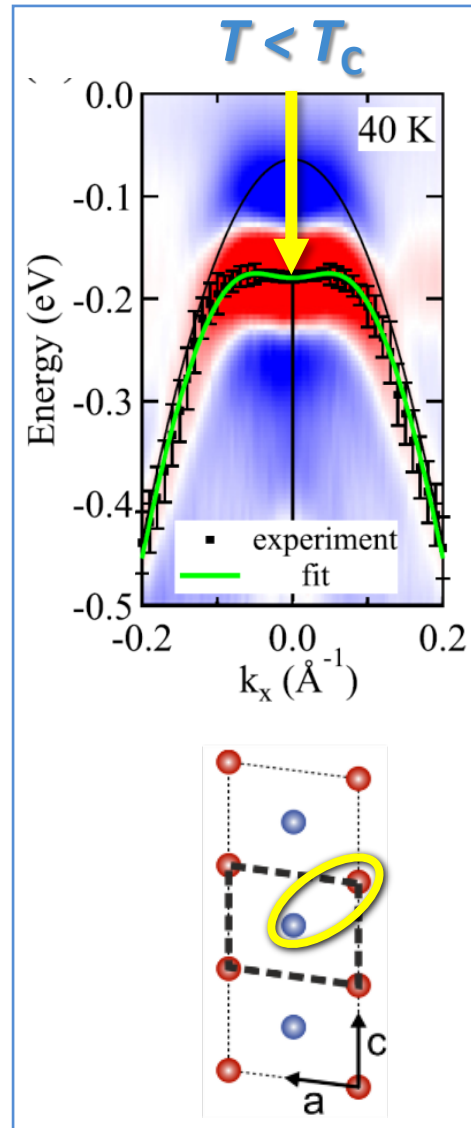
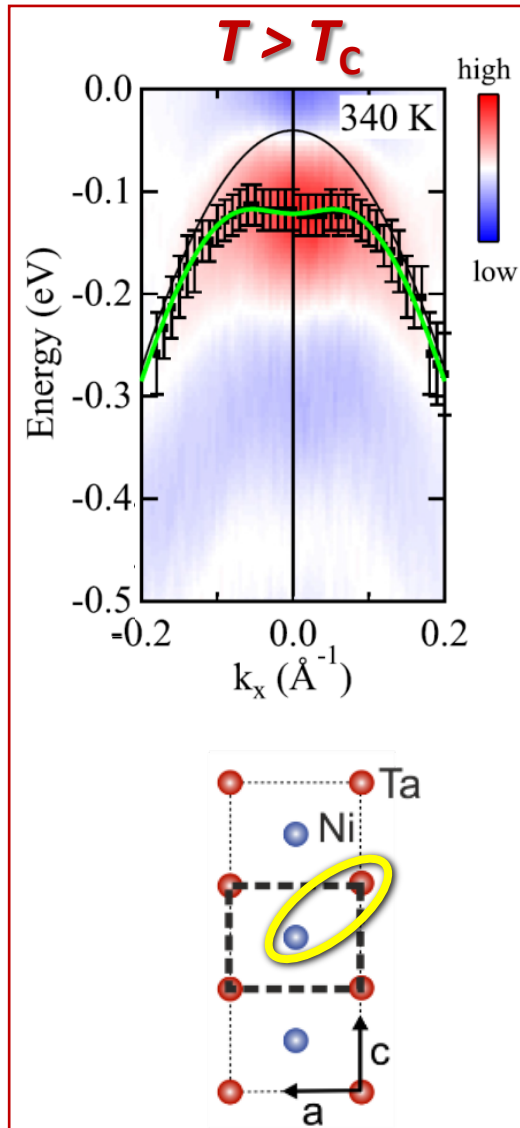
The quasi-one dimensional Ta_2NiSe_5



- exciton formation across atomic chains
- excitonic coupling sensitive to interchain distance?

Kaneko, *PRB* 87, 035121 (2013))

The electronic and structural phase transition in Ta₂NiSe₅



$$T < T_c = 328 \text{ K:}$$

- at Γ : excitonic insulator gap

$$H = H_0 + H_{int}$$

$$H_{int} = U \sum_i n_{i, \text{CB}} n_{i, \text{VB}} \neq 0$$

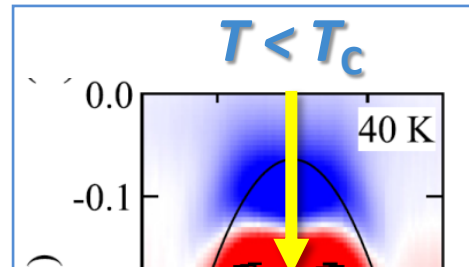
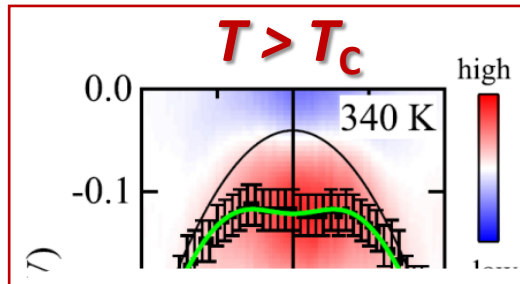
interorbital e-h
Coulomb attraction

- orthorhombic to monoclinic

lattice distortion favors
the excitonic coupling

Seki, *PRB* 90, 155116 (2014)

The electronic and structural phase transition in Ta₂NiSe₅



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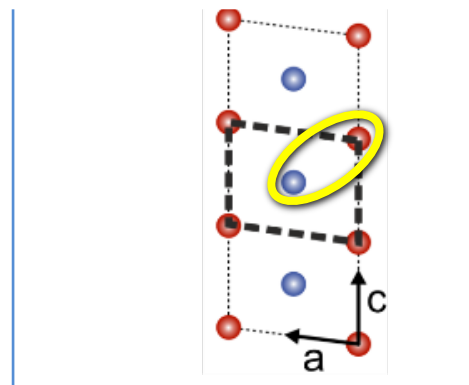
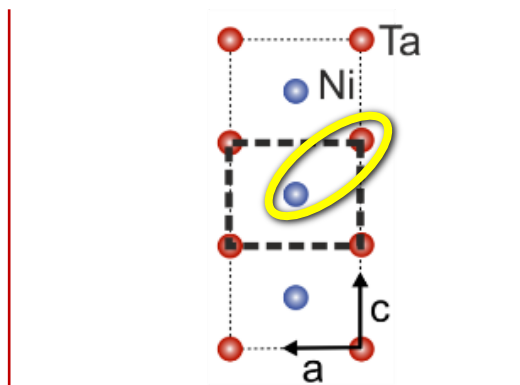
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- ① How do the photoinduced carrier & lattice dynamics connect with each other?
- ② How does the Coulomb interaction govern the *nonequilibrium* electronic structure?

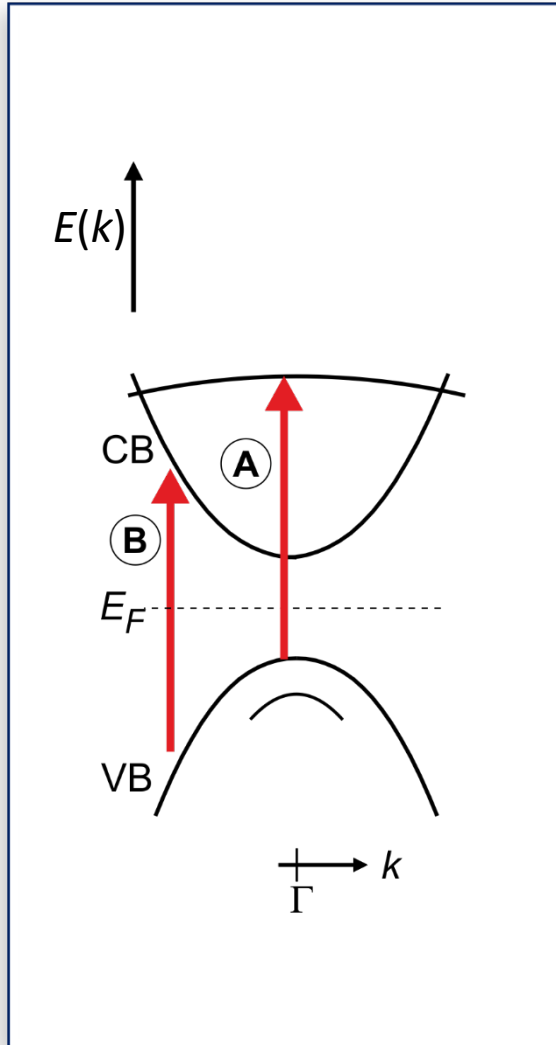


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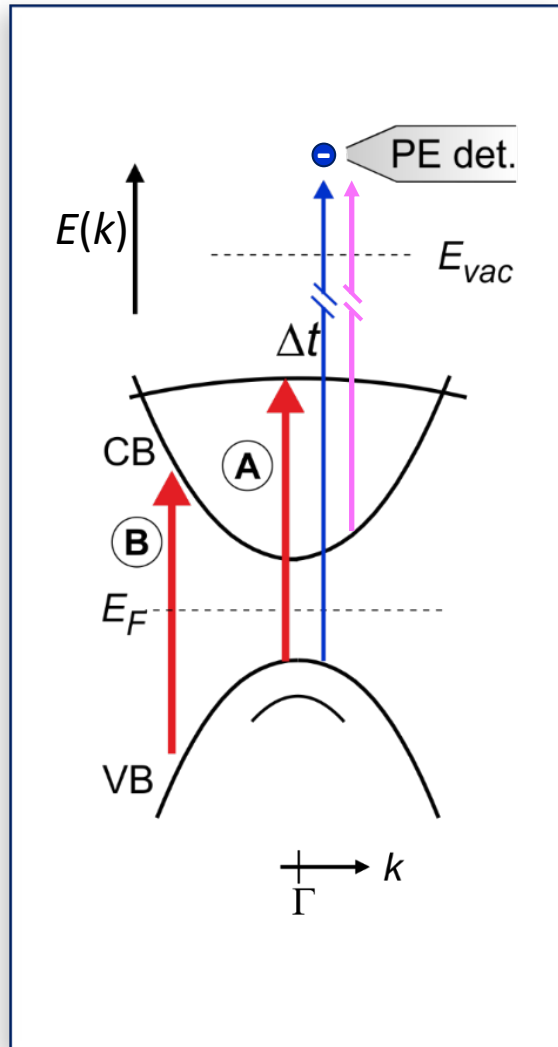
Seki, *PRB* 90, 155116 (2014)

The work tools: time-resolved photoemission and optical spectroscopy



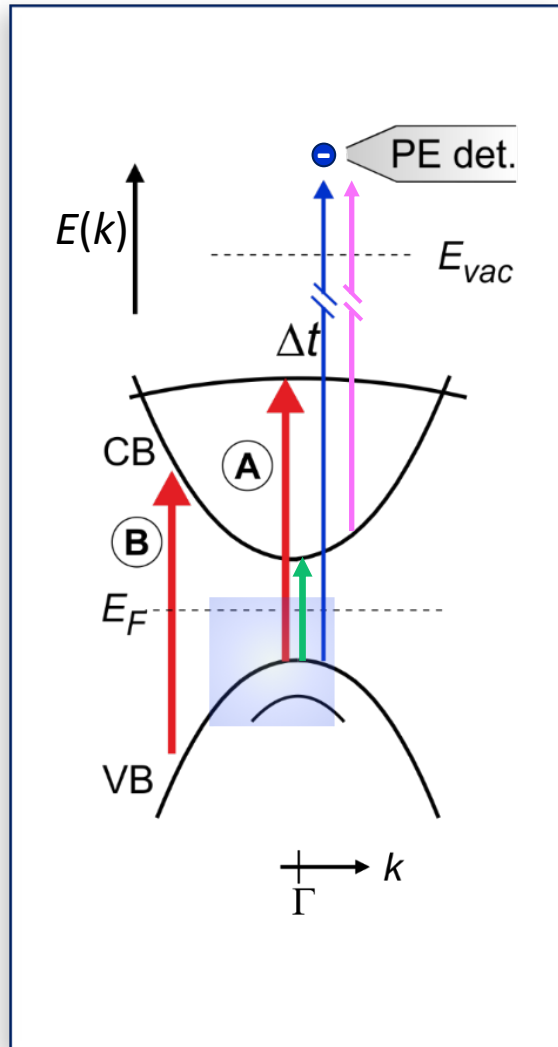
- free carrier excitation by 1.55 eV pump:
 - Ⓐ at Γ
 - Ⓑ at $k \neq 0$

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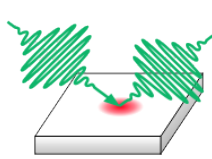


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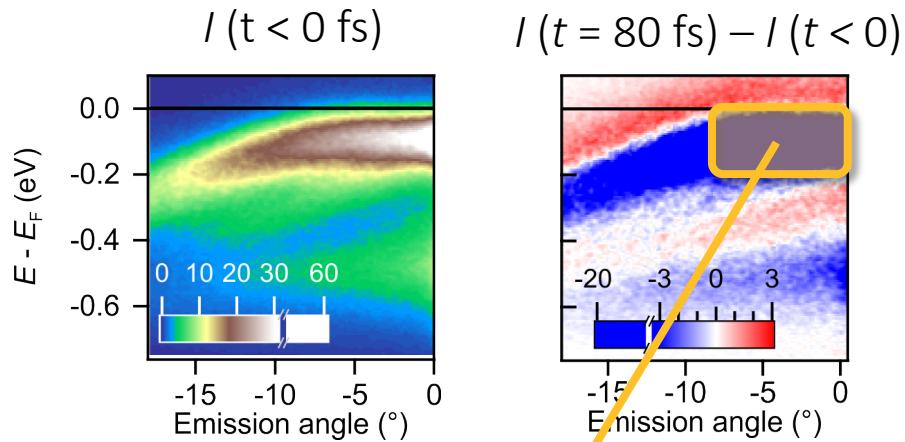
- free carrier excitation by **1.55 eV pump**:
 (A) at Γ
 (B) at $k \neq 0$
- time-resolved ARPES & 2PPE: photoinduced dynamics of the **occupied** & **unoccupied** electronic bands
- midIR transient reflectivity: photoinduced **carrier** and **lattice** dynamics



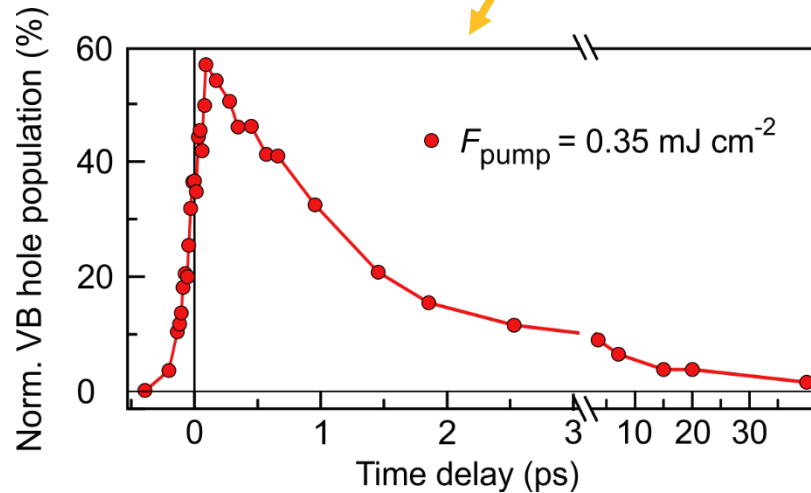
$$\Delta R(t) = \left(\frac{\partial R}{\partial N_c} \right) \Delta N_c(t) + \left(\frac{\partial R}{\partial Q_{ph}} \right) \Delta Q_{ph}(t)$$

carrier
lattice (phonons)

Photoexcitation at Γ

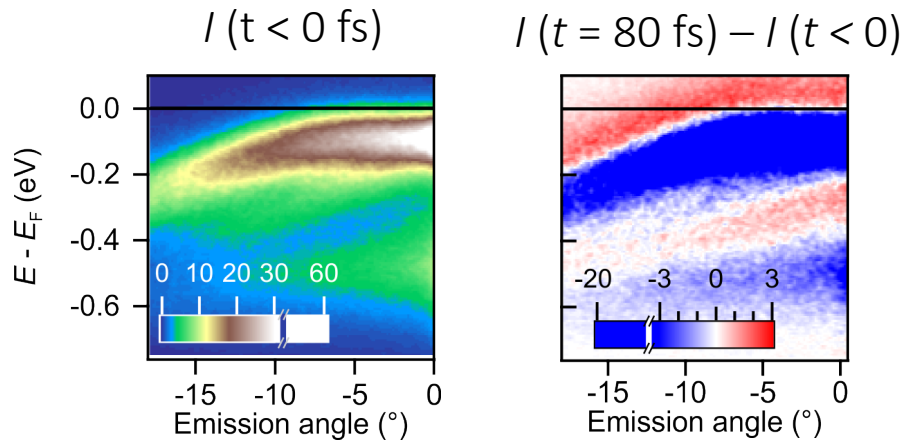


- strong VB depopulation and re-filling within few ps

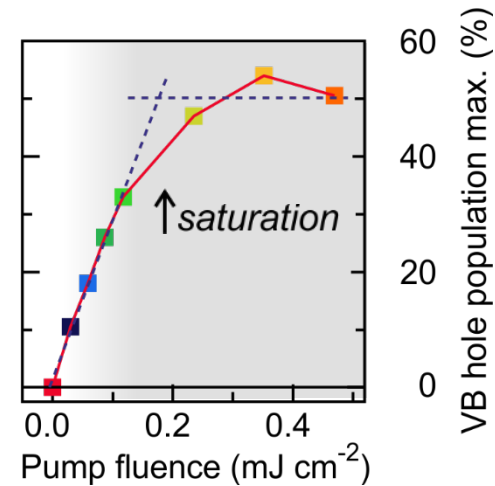
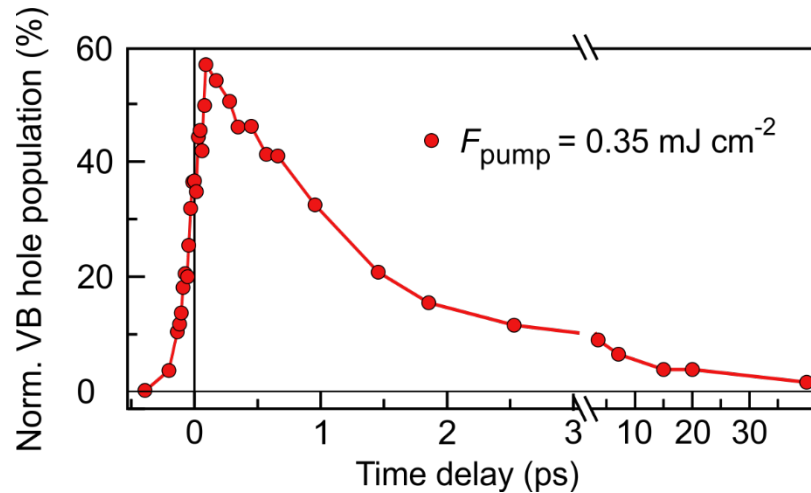


S. Mor *et al.*, PRL 119, 086401 (2017)

Photoexcitation at Γ

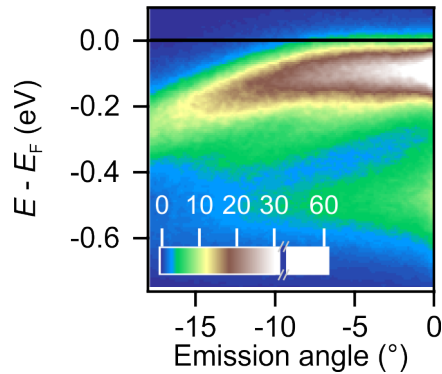


- strong VB depopulation and re-filling within few ps
- hole population saturates at 50 % for $F > F_C \approx 0.2 \text{ mJ cm}^{-2}$

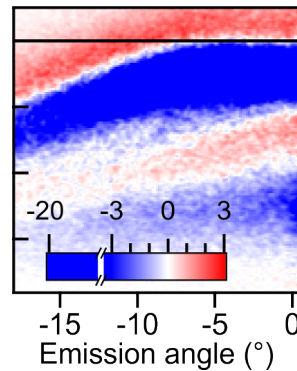


Photoexcitation at Γ

$I(t < 0 \text{ fs})$

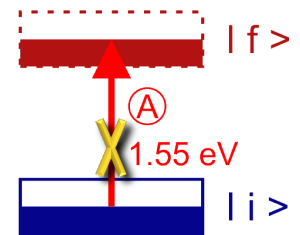
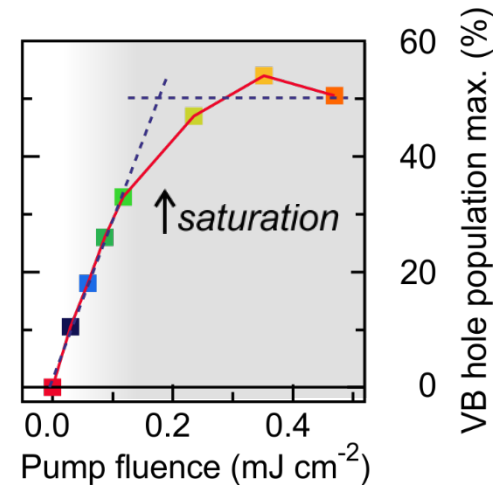
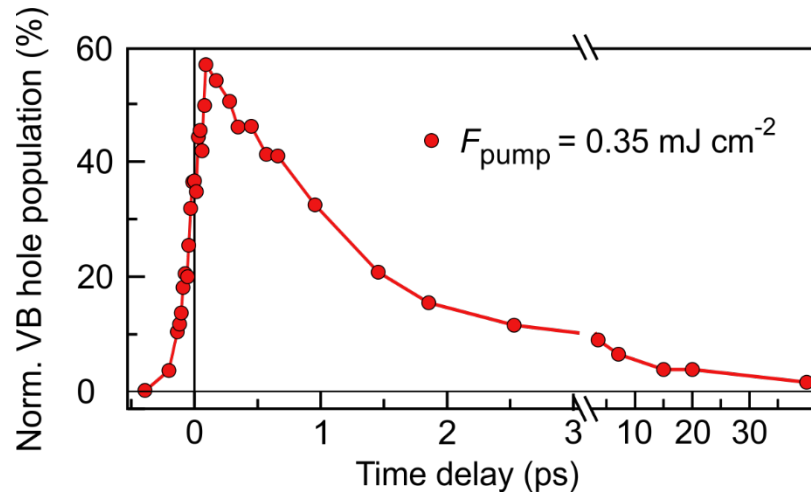


$I(t = 80 \text{ fs}) - I(t < 0)$

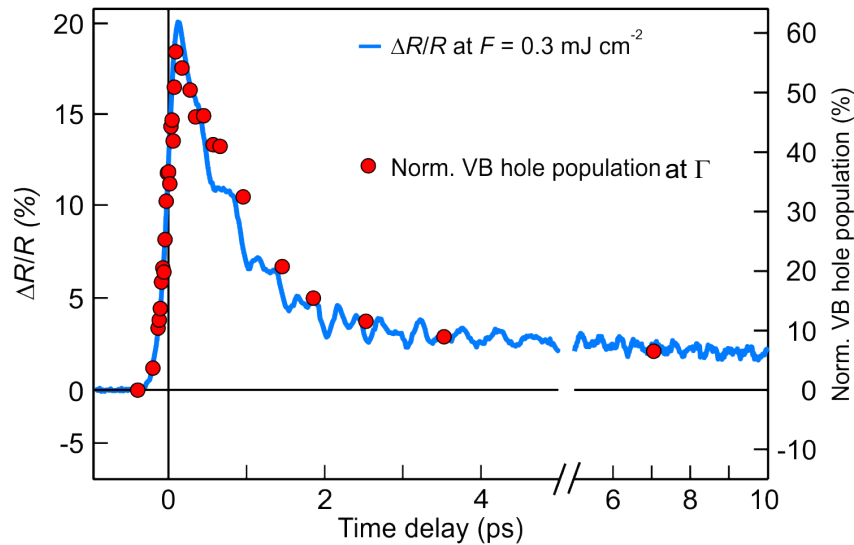


- strong VB depopulation and re-filling within few ps
- hole population saturates at 50 % for $F > F_C \approx 0.2 \text{ mJ cm}^{-2}$

pump absorption saturation
at Γ above F_C



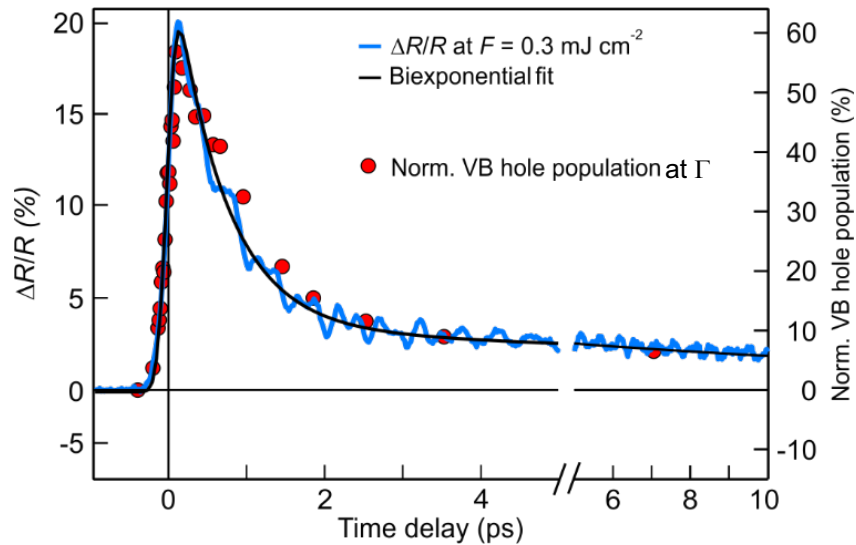
MidIR transient reflectivity probing the hole dynamics at Γ



$$T = 120 \text{ K} \ll T_C$$

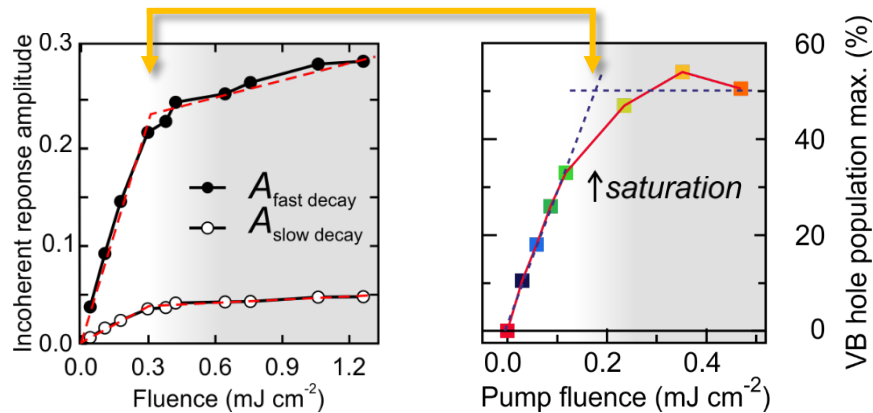
- midIR incoherent $\Delta R/R(t)$ probes the dynamics of photoholes at Γ

MidIR transient reflectivity probing the hole dynamics at Γ

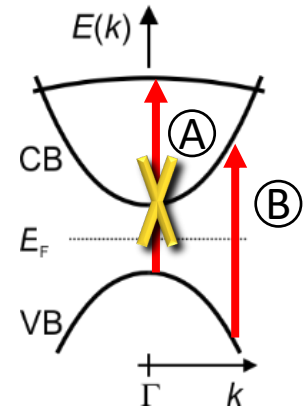


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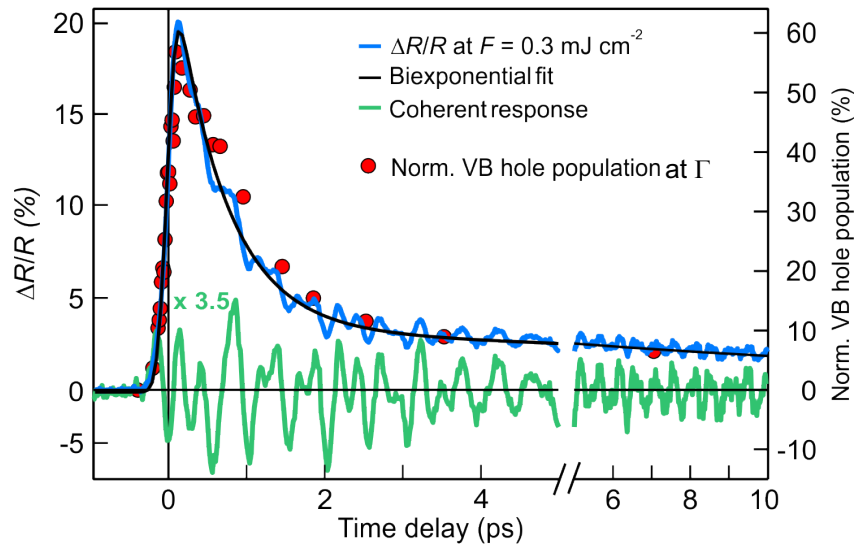
- midIR incoherent $\Delta R/R(t)$ probes the dynamics of photoholes at Γ



- partial saturation above $F_C \sim 0.3 \text{ mJ cm}^{-2}$



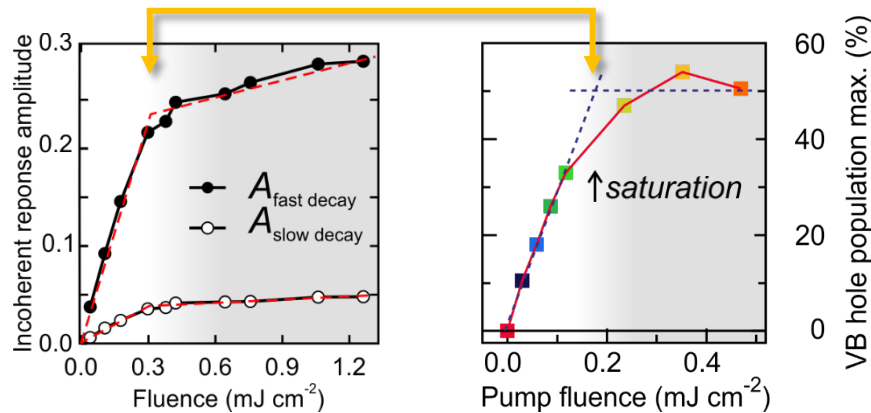
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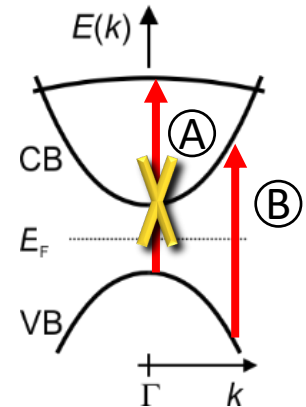
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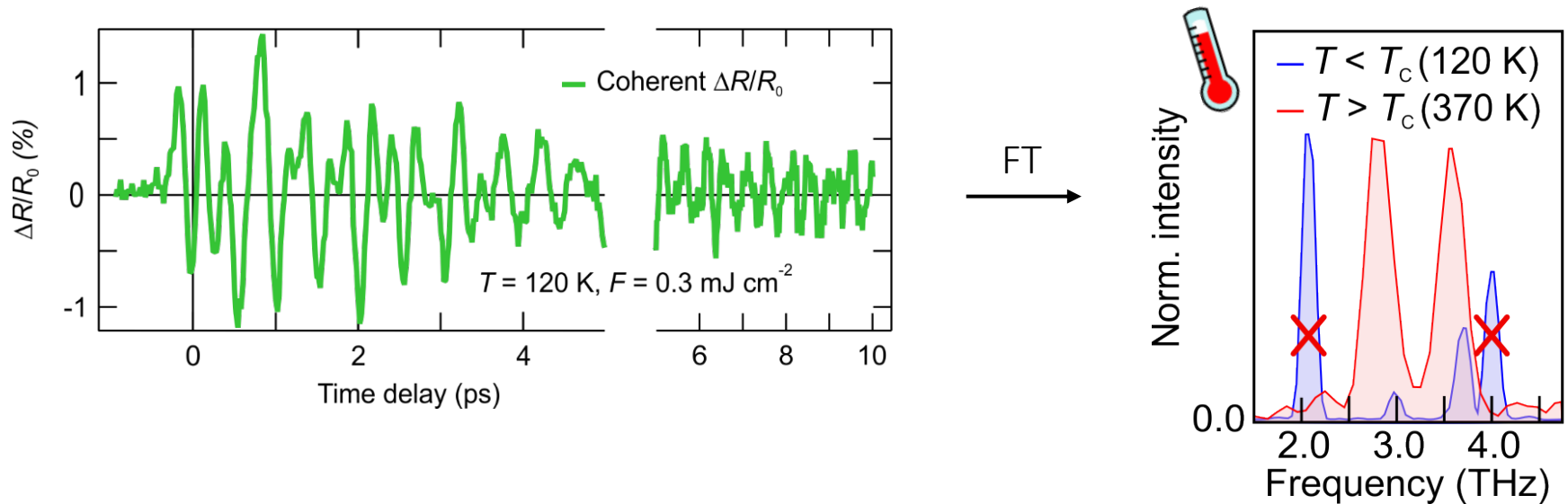
Photoinduced structural dynamics?



- partial saturation above $F_C \sim 0.3 \text{ mJ cm}^{-2}$

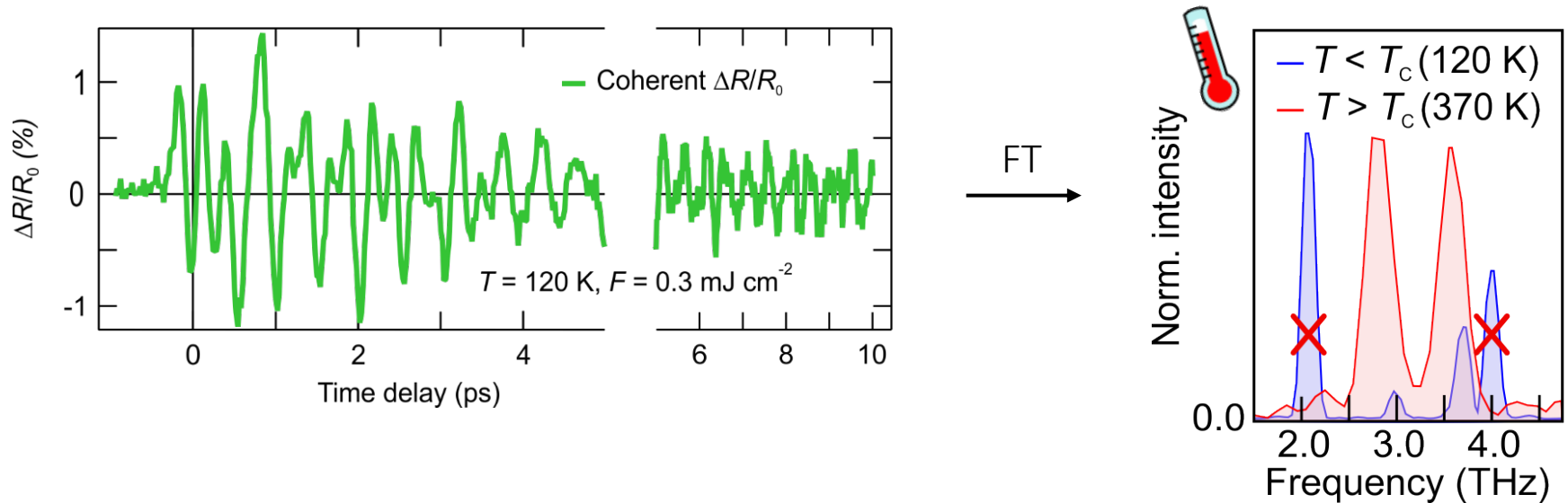


Tracking the thermal structural phase transition



- 2.1 and 4.0 THz phonon modes markers of the low- T structural phase

Tracking the thermal structural phase transition

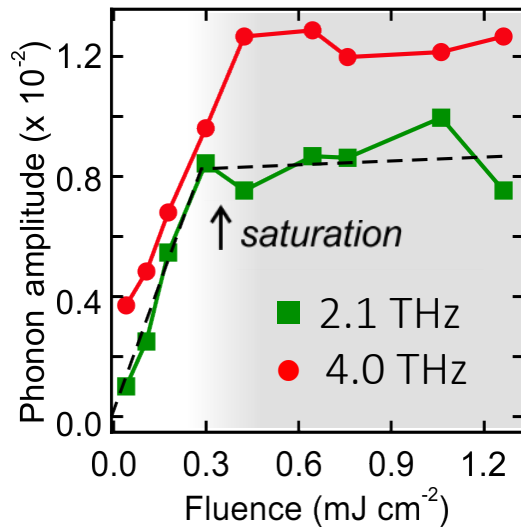


- 2.1 and 4.0 THz phonon modes markers of the low- T structural phase

Photoinduced switching of the structural phase?

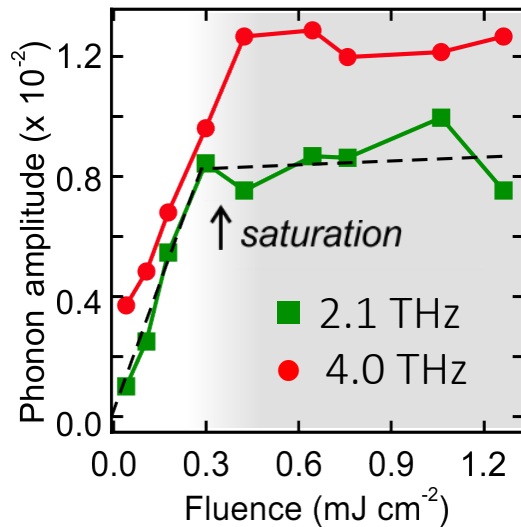


Coherent phonons coupled to carrier excitation at Γ

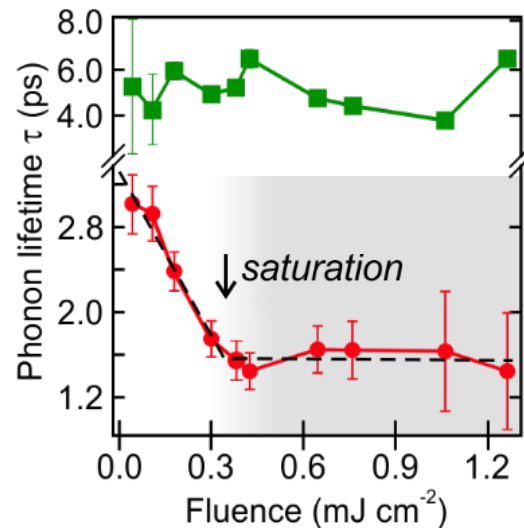


- Amplitudes: complete saturation above F_C
→ no abrupt structural phase transition (PT)

Coherent phonons coupled to carrier excitation at Γ

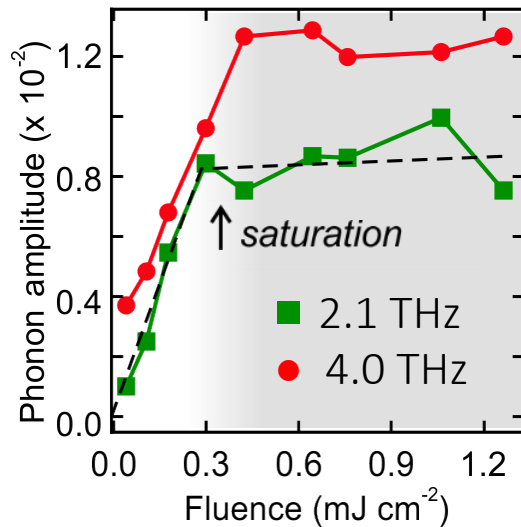


- Amplitudes: complete saturation above F_C
→ no abrupt structural phase transition (PT)



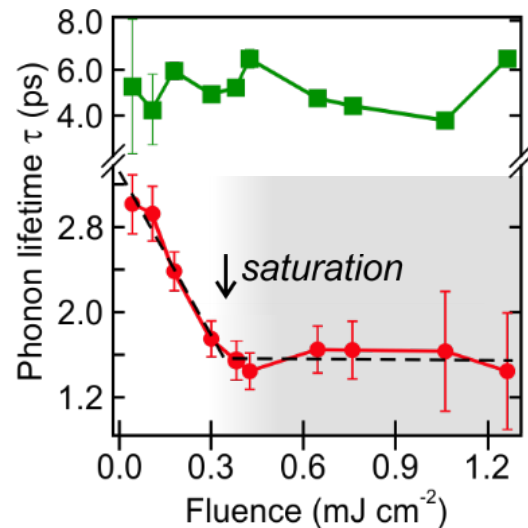
- Lifetimes: $\sim$ few ps
- 4.0-THz-phonon decay coupled to carriers at Γ
→ no delayed *nonthermal* structural PT

Coherent phonons coupled to carrier excitation at Γ



- Amplitudes: complete saturation above F_C
- no abrupt structural phase transition (PT)

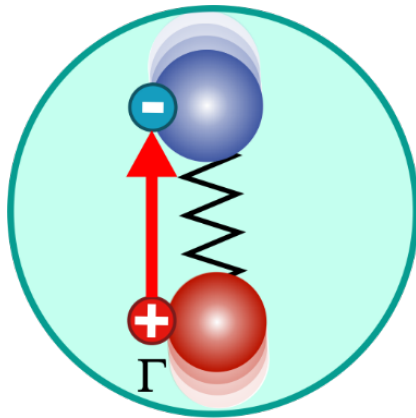
Absorption saturation at Γ inhibits the *nonthermal* structural change



- Lifetimes: $\sim$ few ps
- 4.0-THz-phonon decay coupled to carriers at Γ
- no delayed *nonthermal* structural PT

① How do the photoinduced carrier & lattice dynamics connect with each other?

- Coherent phonons coupled to displacement of charges excited at Γ

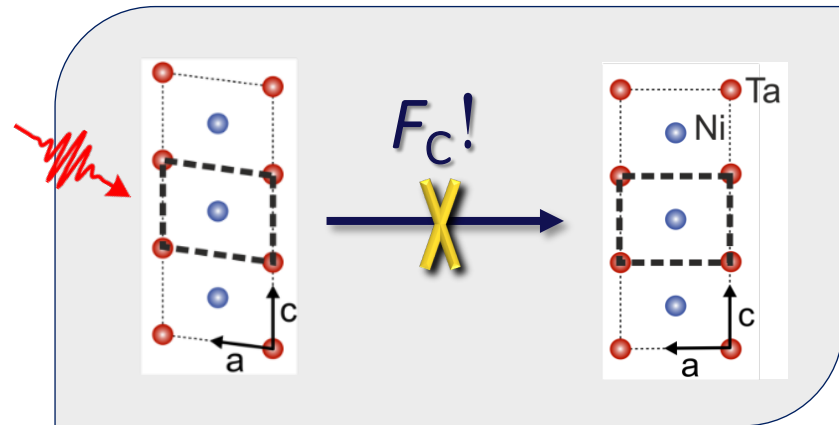
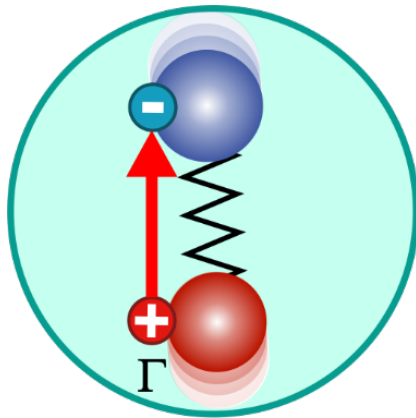


S. Mor *et al.*, PRB 97, 115154 (2018)

S. Mor *et al.*, PRL 119, 086401 (2017)

① How do the photoinduced carrier & lattice dynamics connect with each other?

- Coherent phonons coupled to displacement of charges excited at Γ
- Inhibited photoinduced structural change through optical absorption saturation at Γ

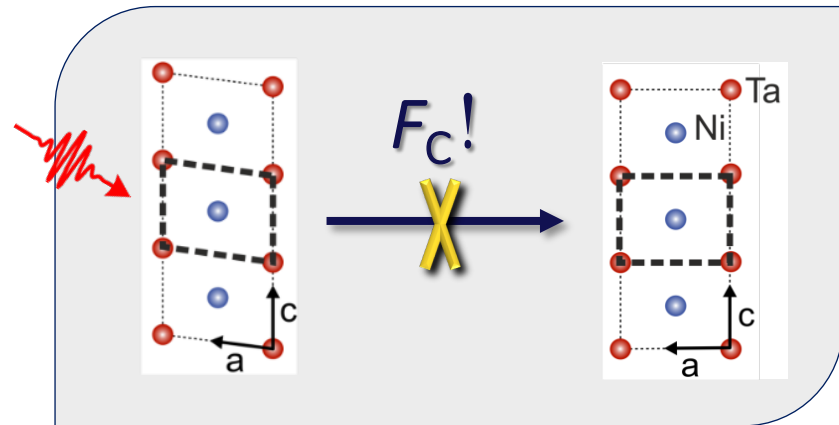
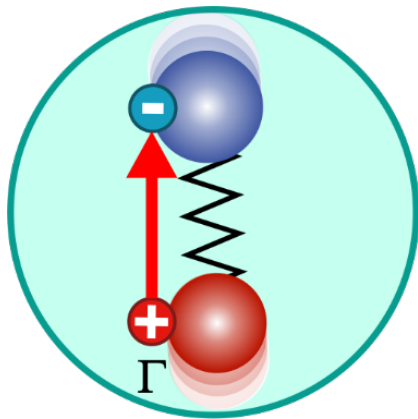


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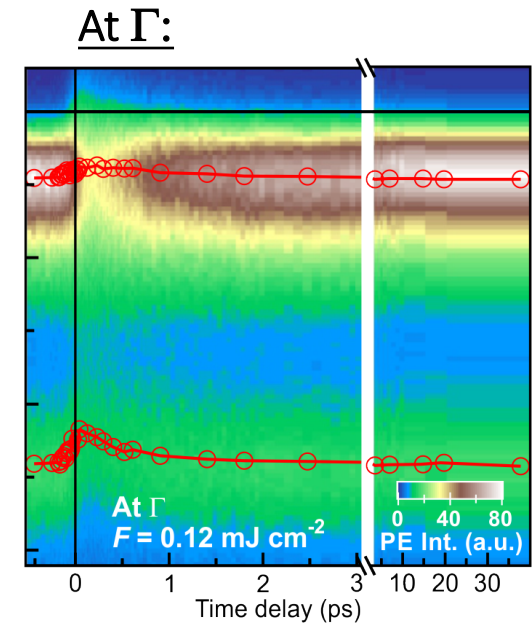
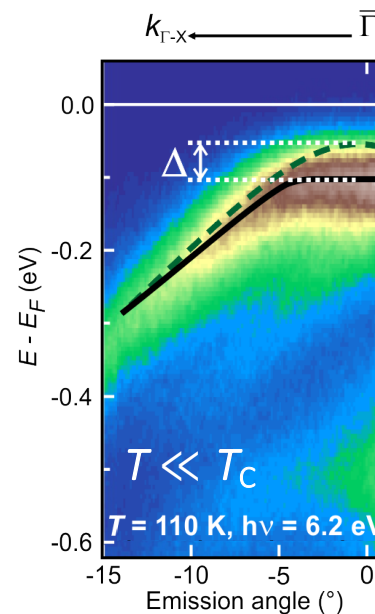
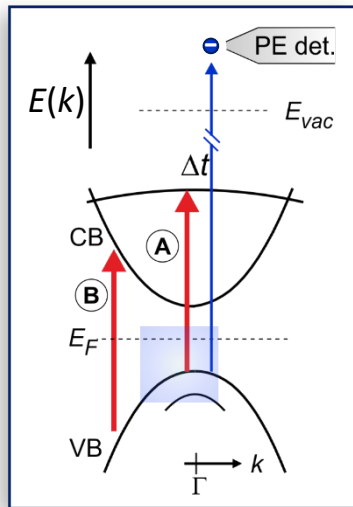
① How do the photoinduced carrier & lattice dynamics connect with each other?

- Coherent phonons coupled to displacement of charges excited at Γ
- Inhibited photoinduced structural change through optical absorption saturation at Γ



... Lattice unperturbed, let's look at the nonequilibrium electronic band structure...

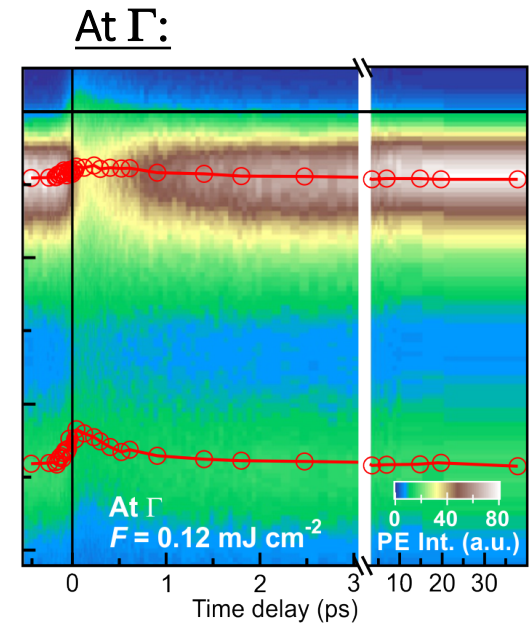
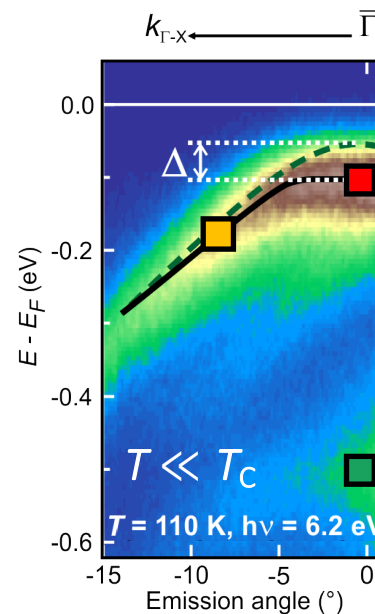
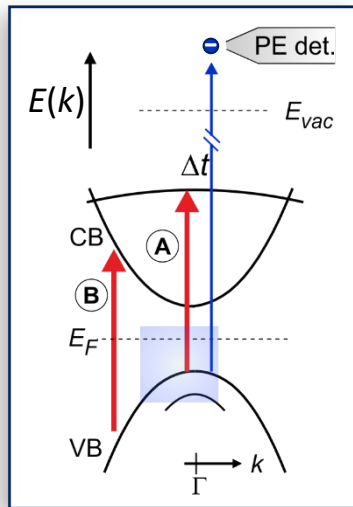
② How does the Coulomb interaction govern the *nonequilibrium* electronic structure?



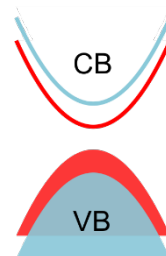
- 1.55 eV pump
 - Ⓐ at Γ
 - Ⓑ at $k \neq 0$
- 6.2 eV probe
occupied bands
around Γ

S. Mor *et al.*, PRL 119, 086401 (2017)

② How does the Coulomb interaction govern the *nonequilibrium* electronic structure?



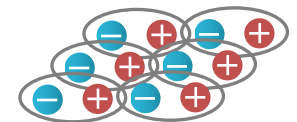
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- 6.2 eV probe
occupied bands
around Γ



screening-
induced
BGR

“vs”

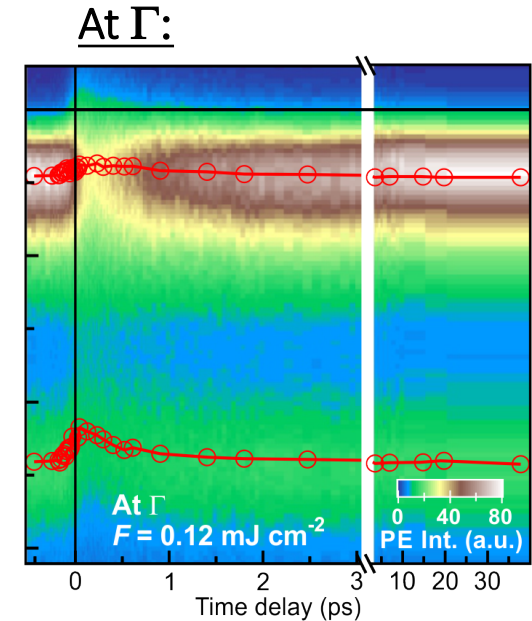
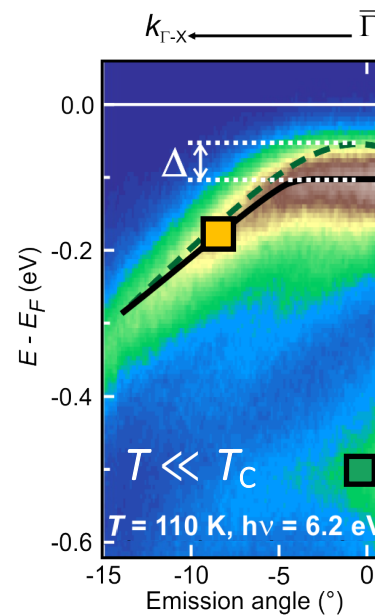
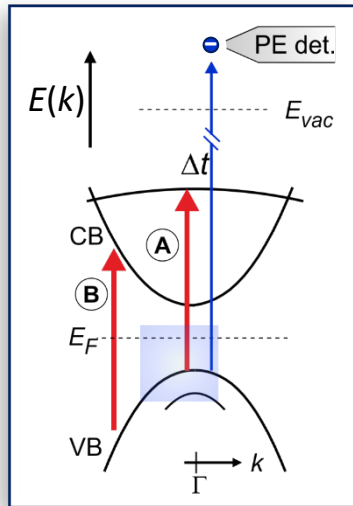
$$\Delta \sim n_{\text{exciton}} \cdot E_B$$



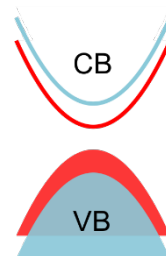
photoinduced band gap dynamics?

S. Mor *et al.*, PRL 119, 086401 (2017)

② How does the Coulomb interaction govern the *nonequilibrium* electronic structure?



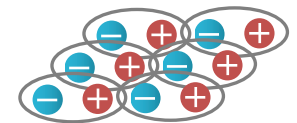
- 1.55 eV pump
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occupied bands
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induced
BGR

“vs”

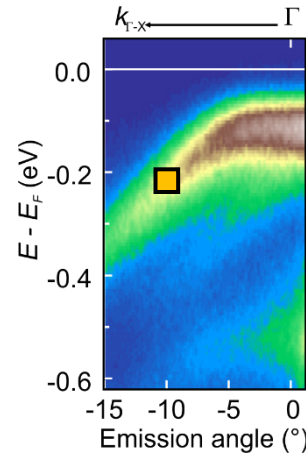
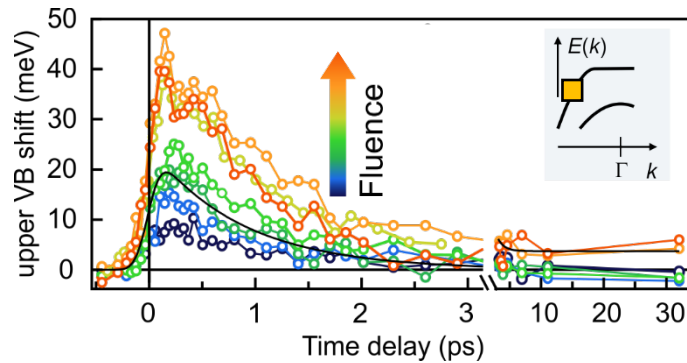
$$\Delta \sim n_{\text{exciton}} \cdot E_B$$



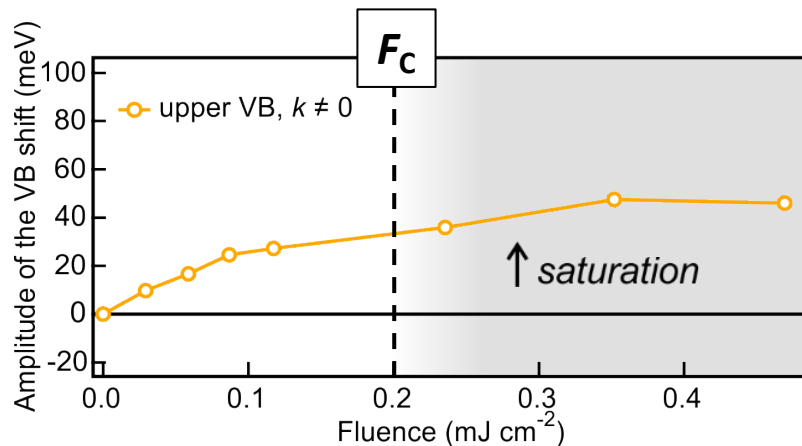
photoinduced band gap dynamics?

S. Mor *et al.*, PRL 119, 086401 (2017)

Photoinduced narrowing of the electronic band splitting

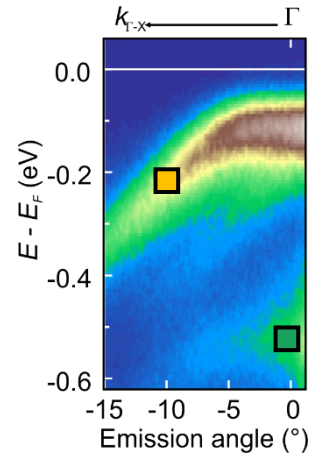
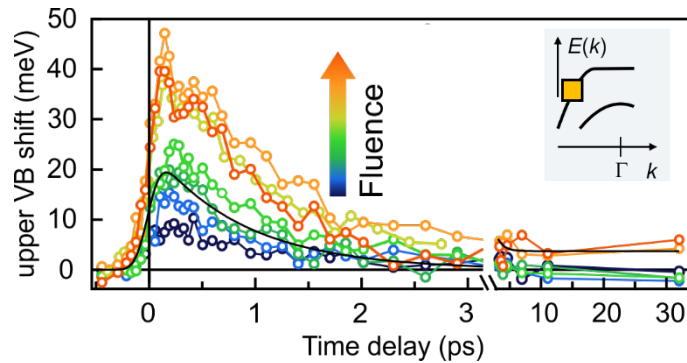


- $k \neq 0$: upshift connected to the density of carriers photoexcited at Γ

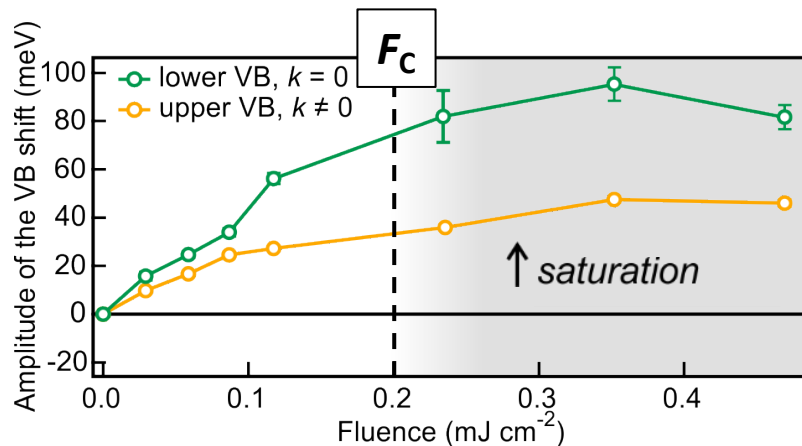


S. Mor *et al.*, PRL 119, 086401 (2017)

Photoinduced narrowing of the electronic band splitting

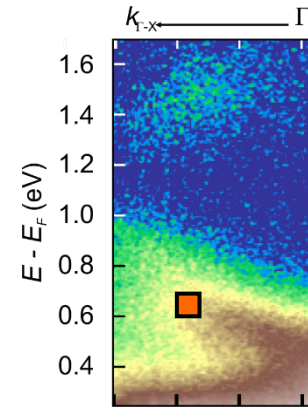
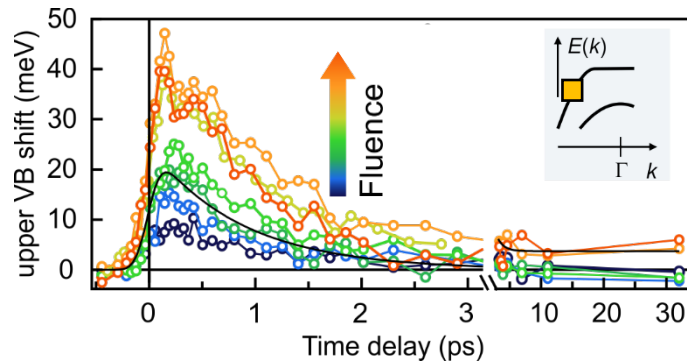


- global upshift connected to the density of carriers photoexcited at Γ

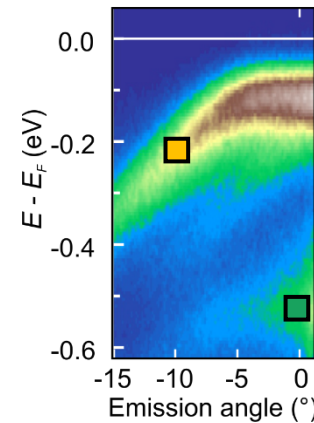
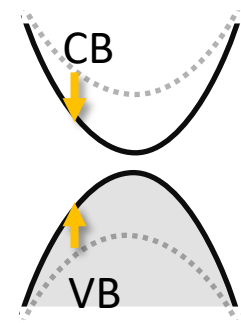


S. Mor *et al.*, PRL 119, 086401 (2017)

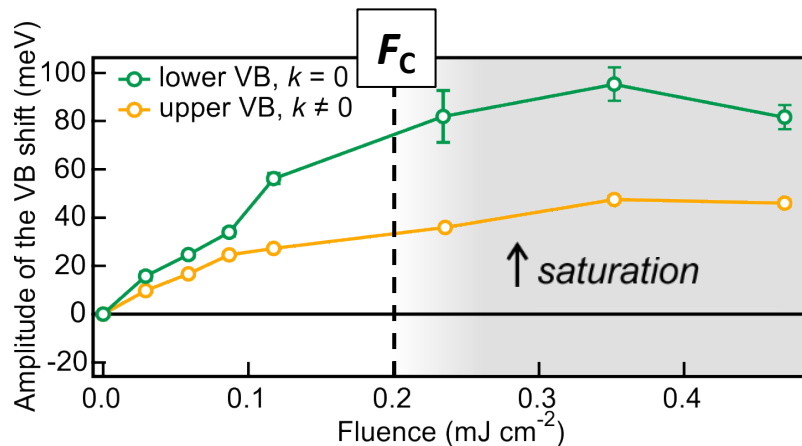
Photoinduced narrowing of the electronic band splitting



- CB does *not* shift in the same direction!

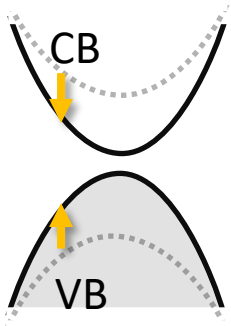
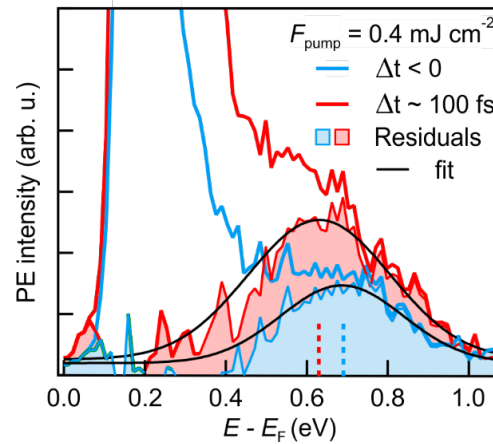
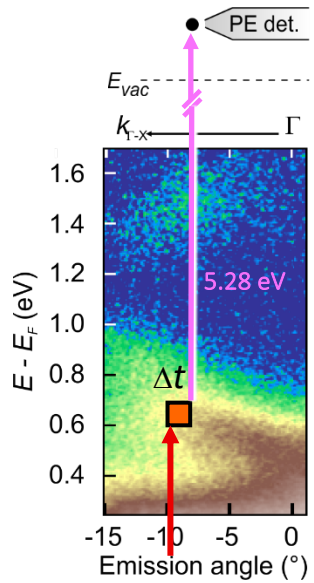


- global VB upshift connected to the density of carriers photoexcited at Γ



S. Mor *et al.*, PRL 119, 086401 (2017)

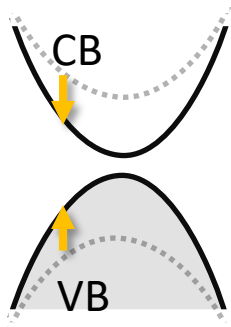
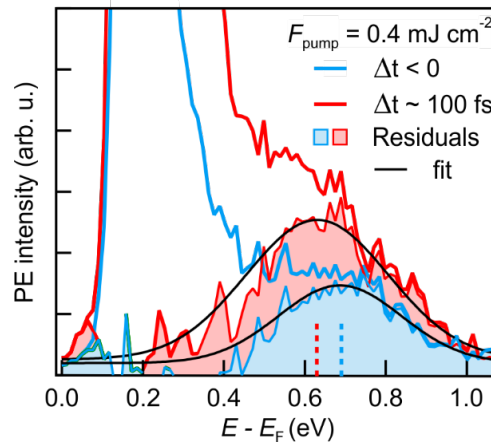
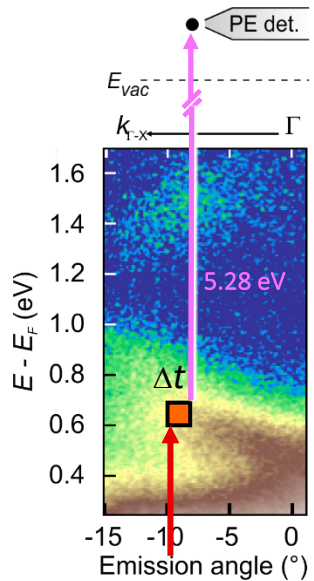
Photoinduced narrowing of the electronic band splitting



- CB at $k \neq 0$: downshift of spectral intensity

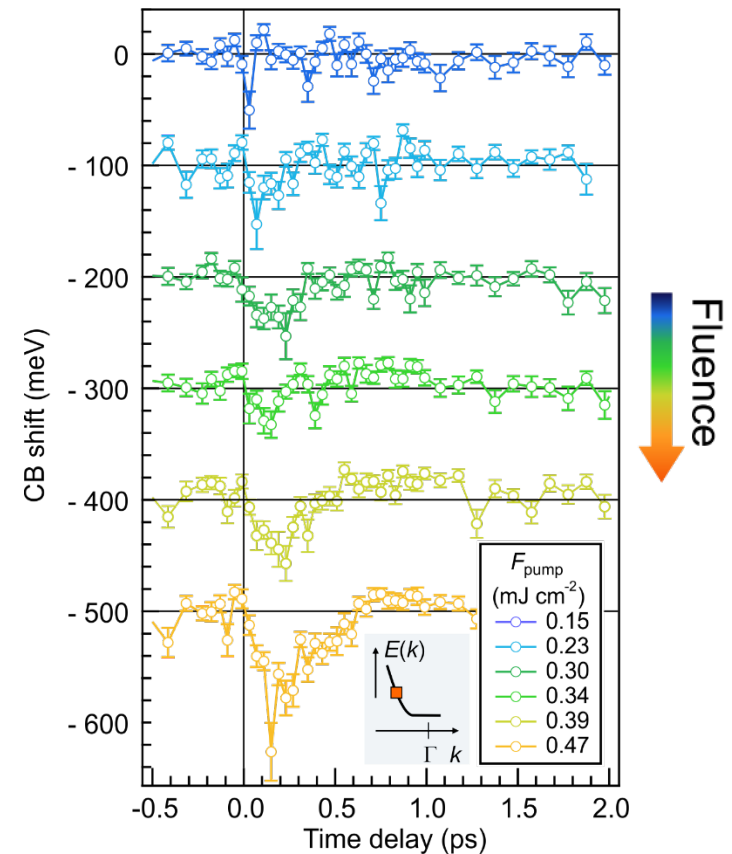
S. Mor *et al.*, in preparation

Photoinduced narrowing of the electronic band splitting



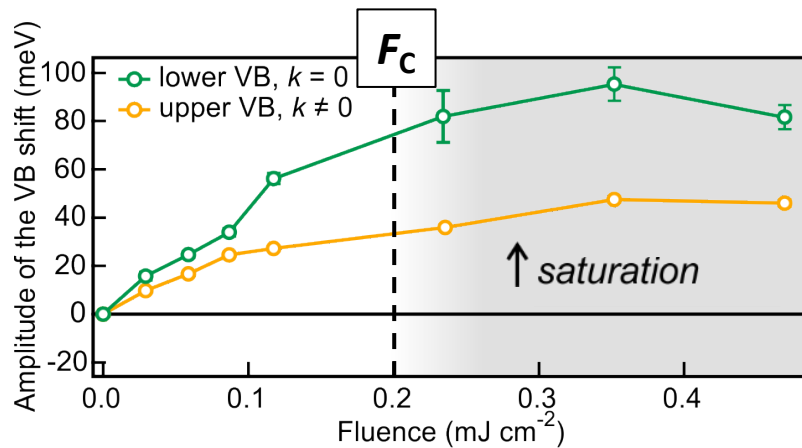
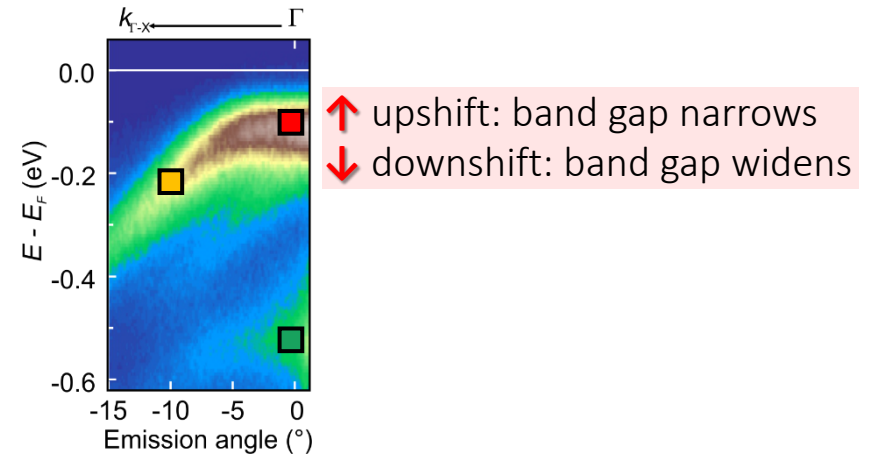
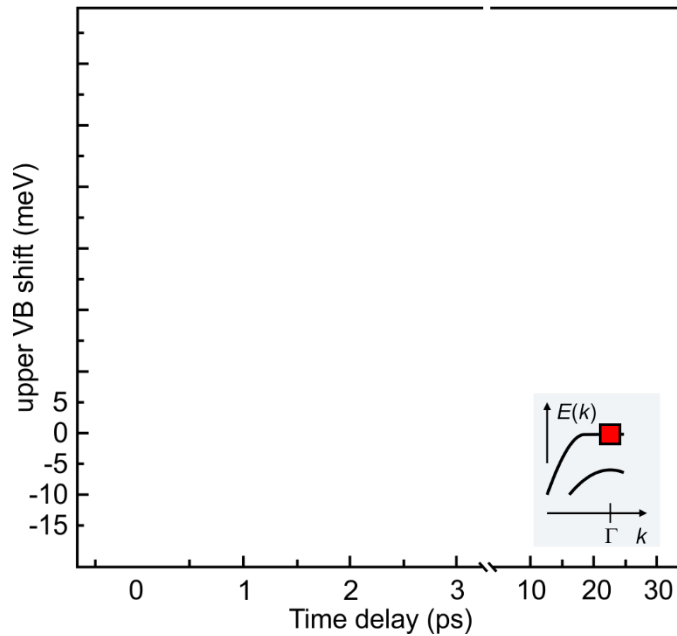
- CB at $k \neq 0$: downshift of spectral intensity connected to density of photoexcited carriers

Fluence-dependent band splitting reduction at $k \neq 0$



S. Mor *et al.*, in preparation

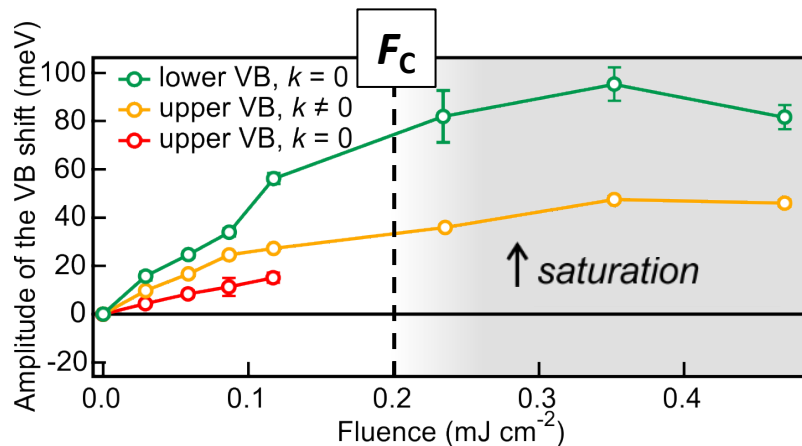
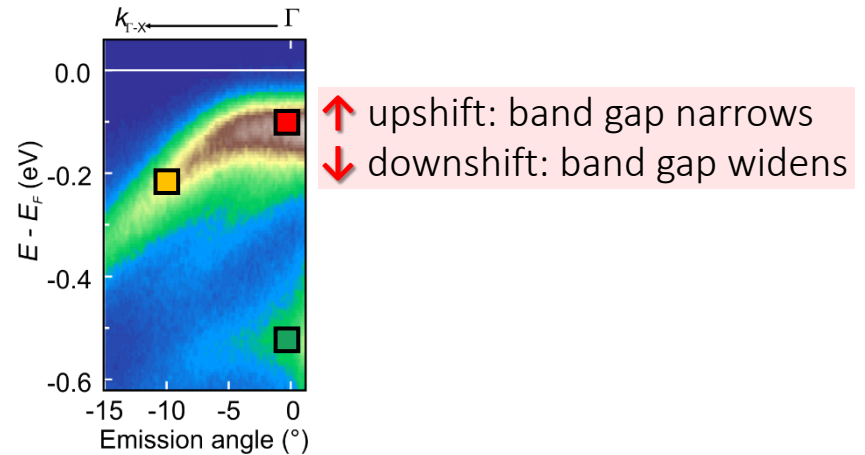
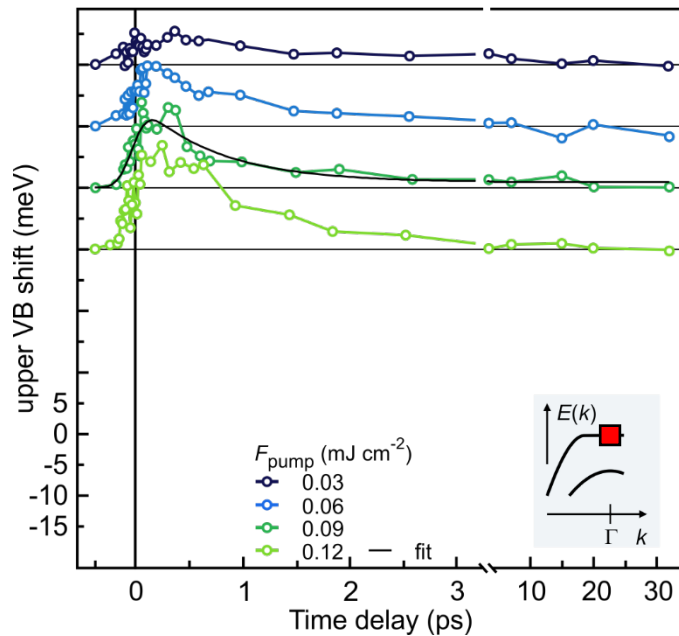
Two dynamics of the band gap at Γ



At Γ :

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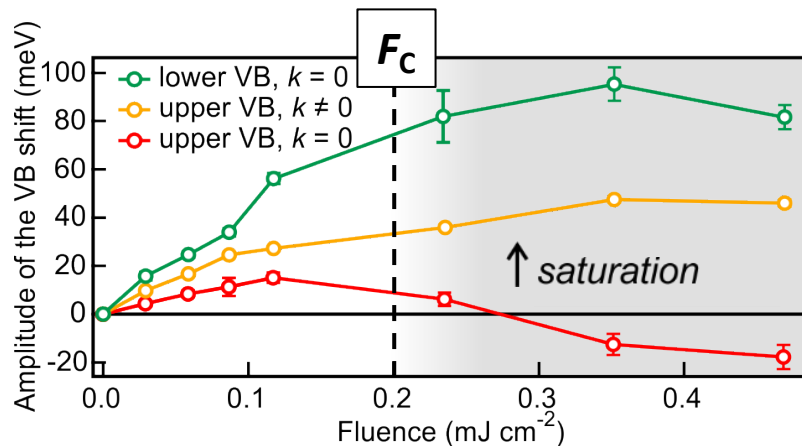
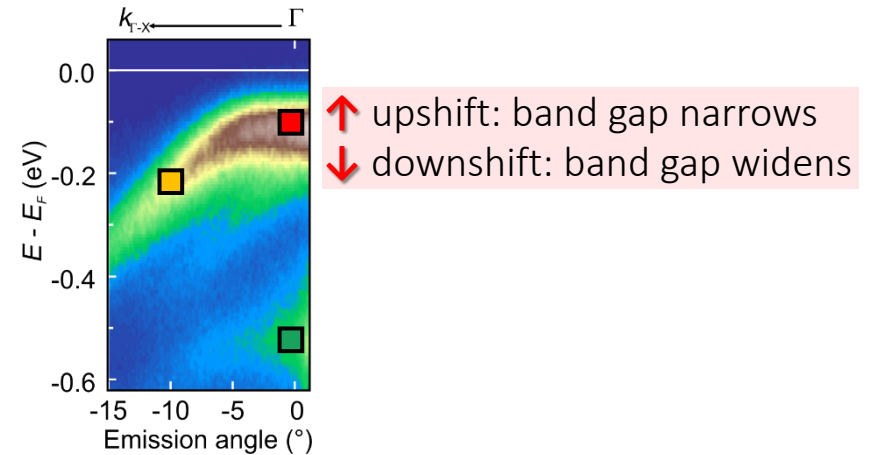
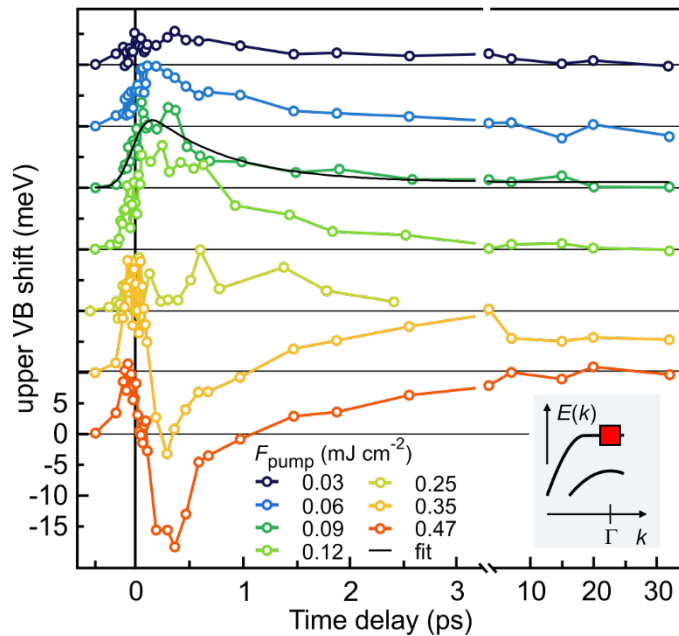
Two dynamics of the band gap at Γ



- At Γ :
- $F < F_C$, screening-induced BGR: analogous to semiconductors

S. Mor *et al.*, PRL 119, 086401 (2017)

Two dynamics of the band gap at Γ

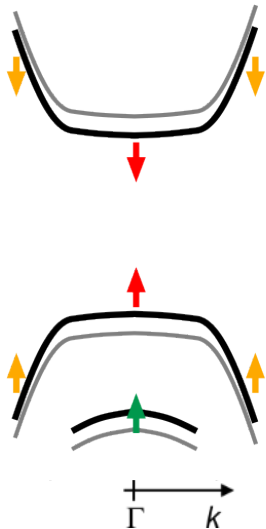


- At Γ :
- $F < F_C$, screening-induced BGR: analogous to semiconductors
 - $F > F_C$, band gap widening: opposite to semiconductors!

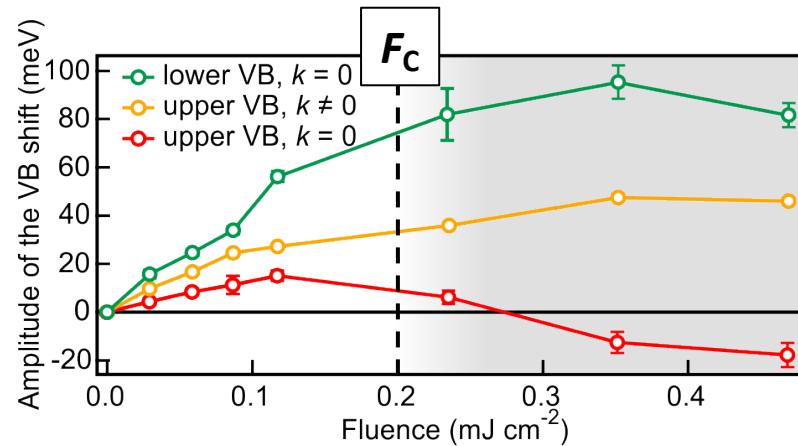
S. Mor *et al.*, PRL 119, 086401 (2017)

Photoinduced modulation of the electronic band gap

semiconductor
band structure
renormalization



governed by
free carriers at Γ

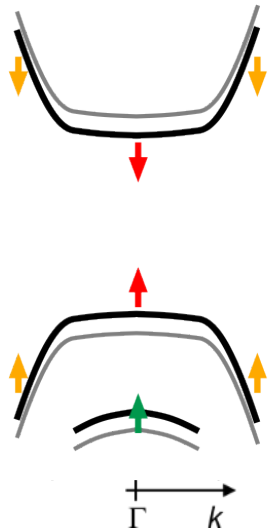


absorption
saturation at Γ

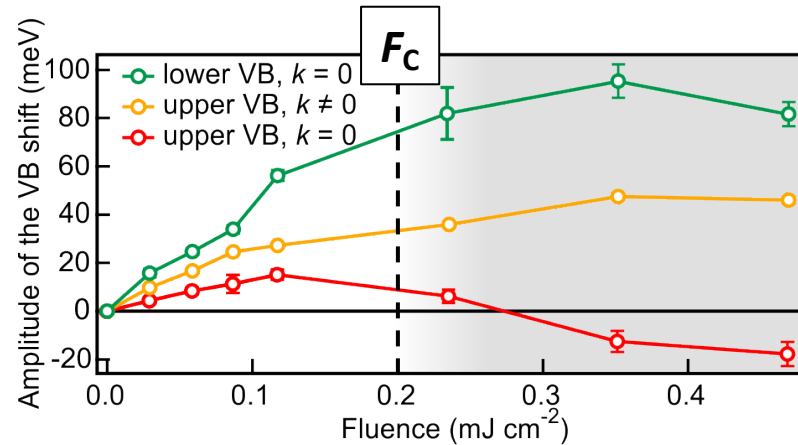
S. Mor *et al.*, PRL 119, 086401 (2017)

Photoinduced modulation of the electronic band gap

semiconductor
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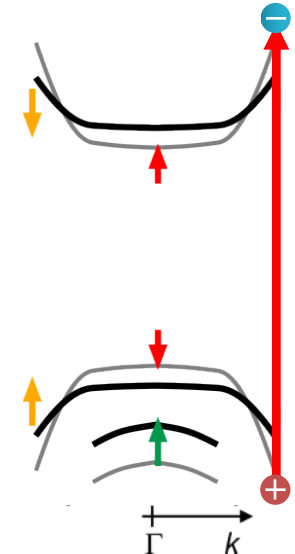


governed by
free carriers at Γ



absorption
saturation at Γ

band gap
widening
at Γ



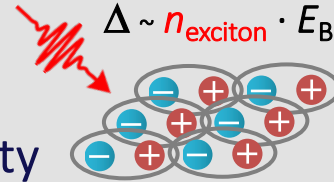
further excitations
at $k \neq 0$

S. Mor *et al.*, PRL 119, 086401 (2017)

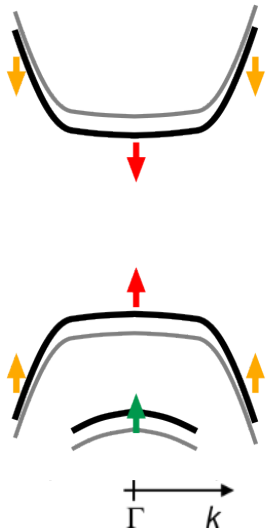
Photoinduced modulation of the electronic band gap

semiconductor
band structure
renormalization

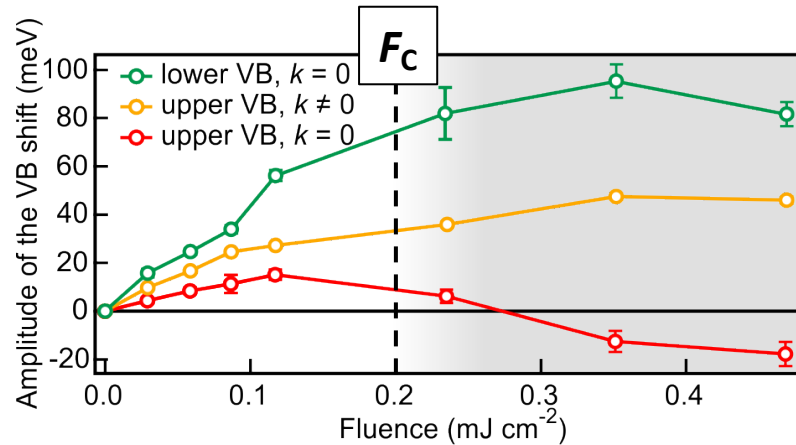
enhancement
of the exciton
condensate density



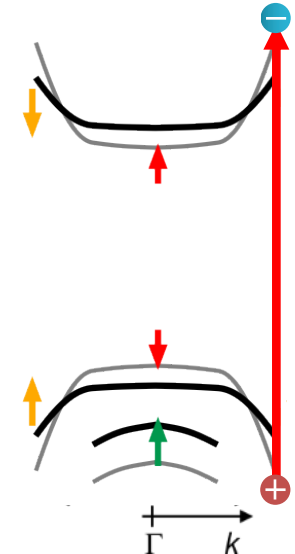
band gap
widening
at Γ



governed by
free carriers at Γ



absorption
saturation at Γ

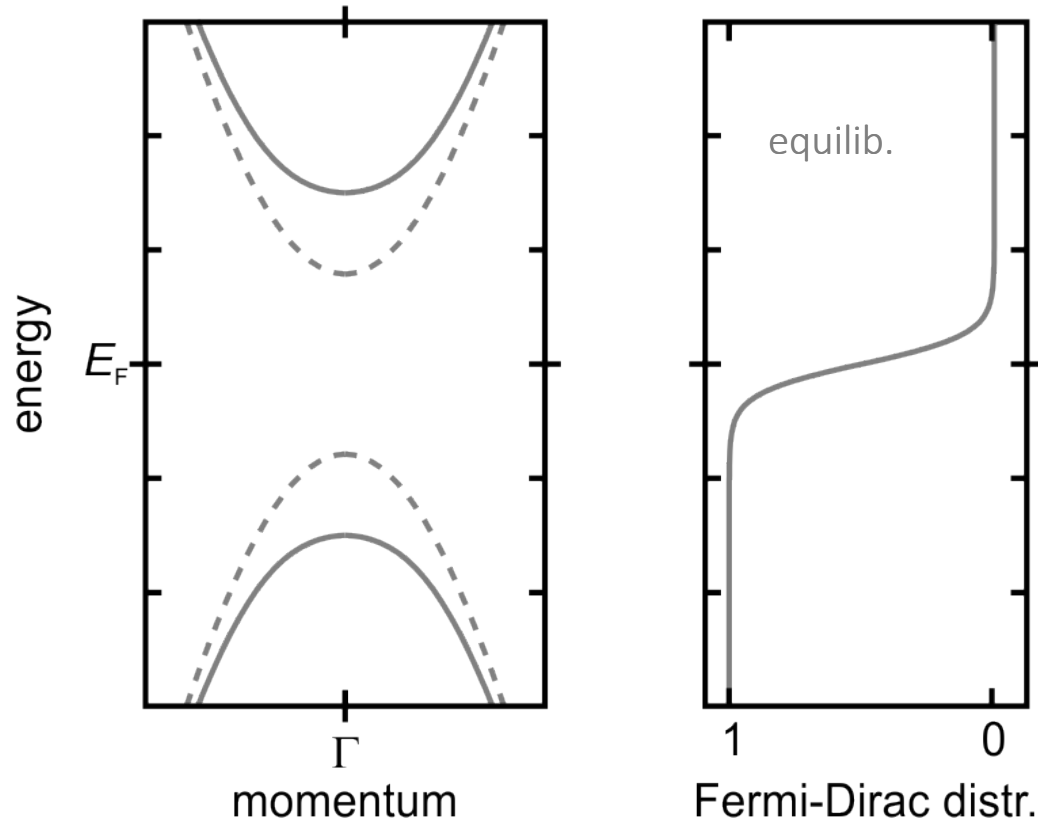


further excitations
at $k \neq 0$

S. Mor *et al.*, PRL 119, 086401 (2017)

Photoenhancement of the excitonic insulator order parameter

Two-band semiconductor with interband interaction $H_{int} = U \sum_i n_{i,\text{CB}} n_{i,\text{VB}}$

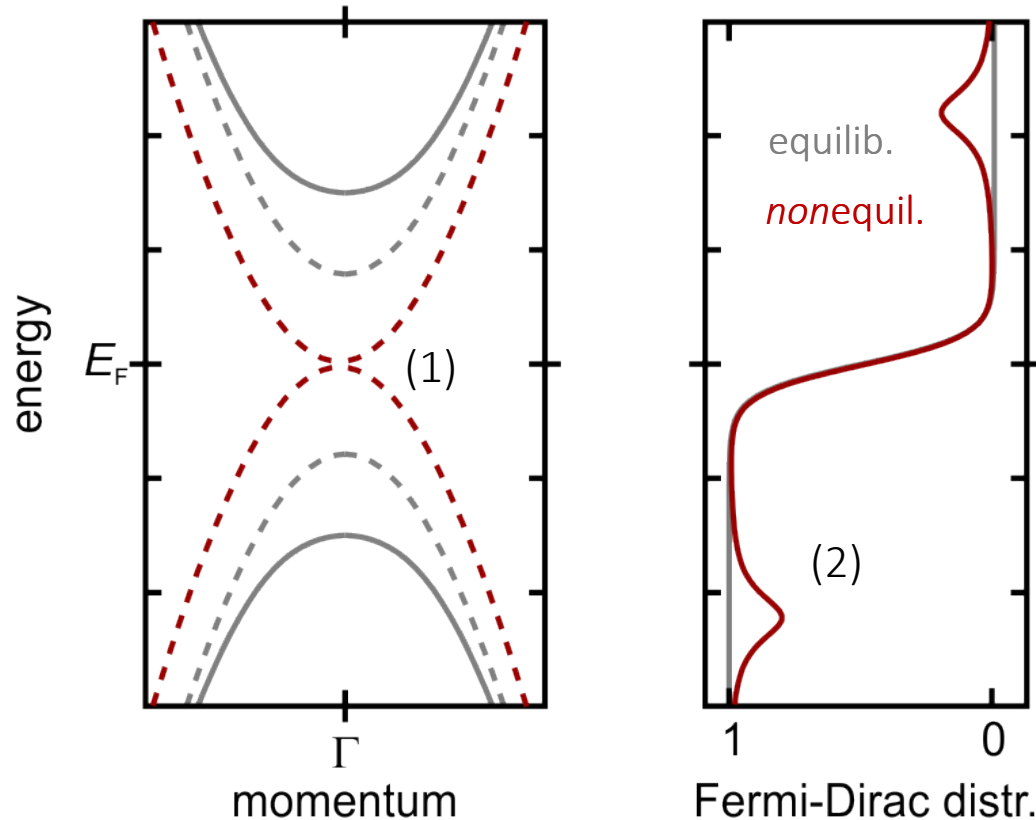


Hartree Fock calculations:
D. Golež, P. Werner, M. Eckstein

S. Mor *et al.*, PRL 119, 086401 (2017)

Photoenhancement of the excitonic insulator order parameter

Two-band semiconductor with interband interaction $H_{int} = U \sum_i n_{i,CB} n_{i,VB}$



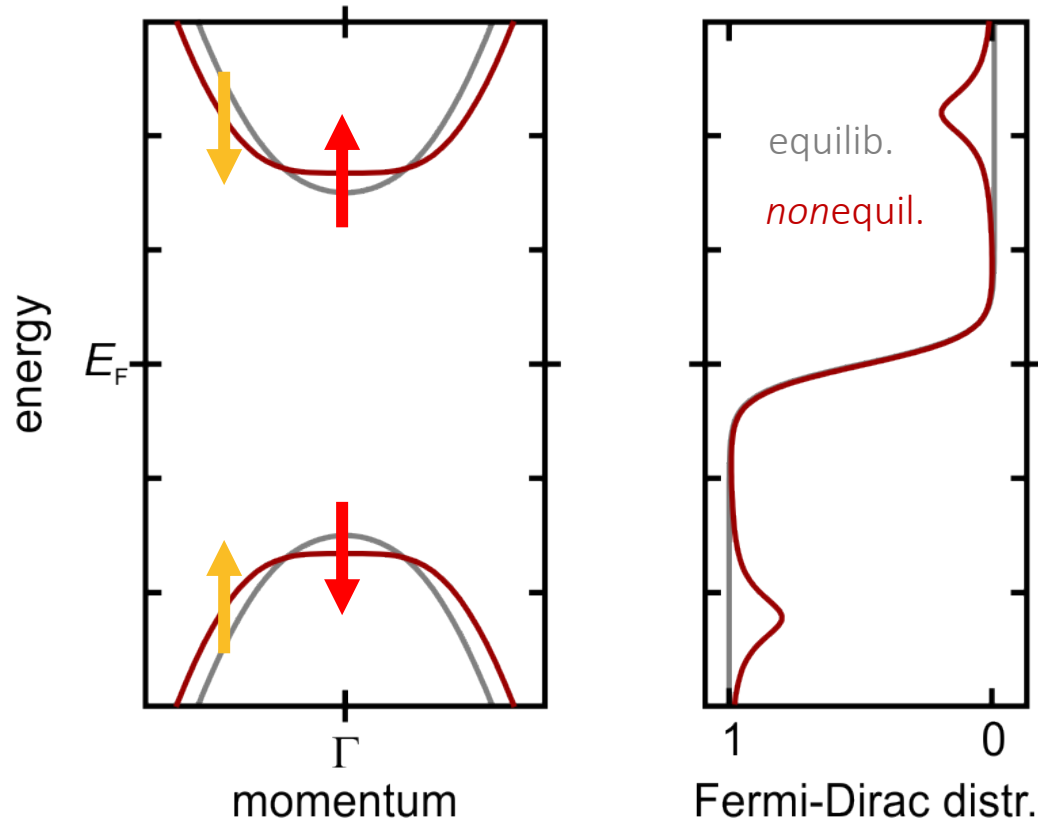
- (1) mimicked BGR
- (2) nonequilibrium carrier population

Hartree Fock calculations:
D. Golež, P. Werner, M. Eckstein

S. Mor *et al.*, PRL 119, 086401 (2017)

Photoenhancement of the excitonic insulator order parameter

Two-band semiconductor with interband interaction $H_{int} = U \sum_i n_{i,CB} n_{i,VB}$



- (1) mimicked BGR
- (2) *nonequilibrium* carrier population

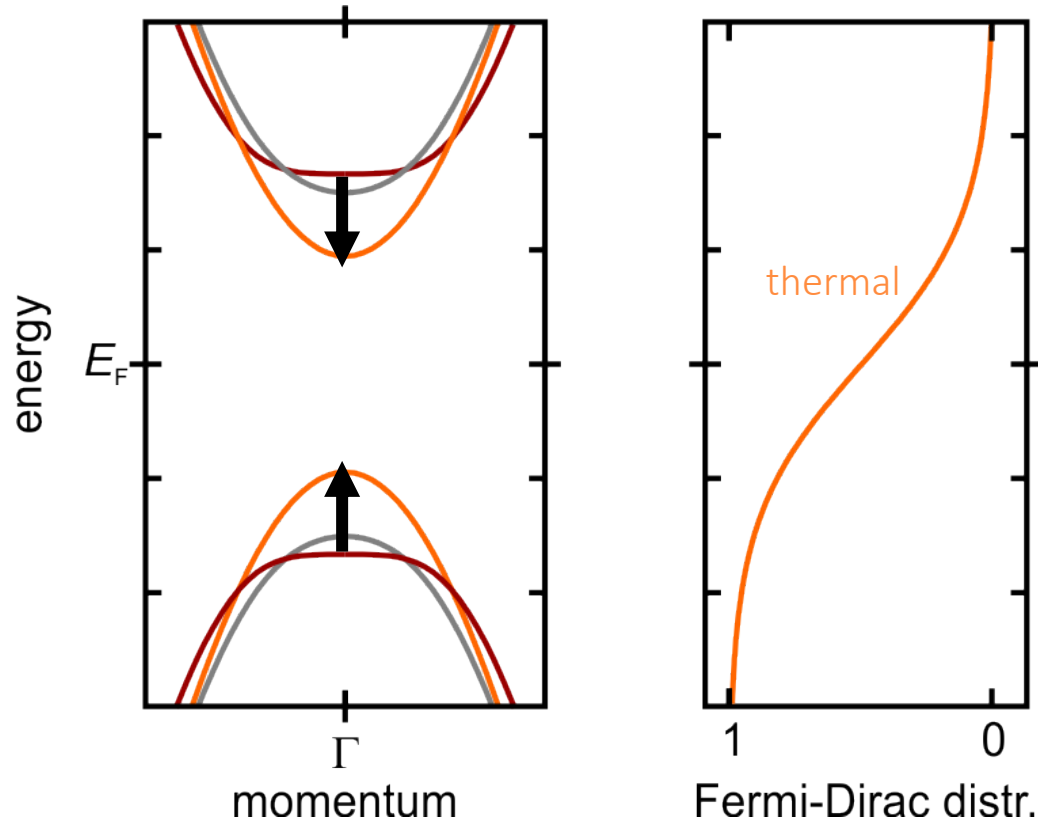
→ enhancement of the order parameter $\Delta \sim n_{\text{exciton}} \cdot E_B$

Hartree Fock calculations:
D. Golež, P. Werner, M. Eckstein

S. Mor *et al.*, PRL 119, 086401 (2017)

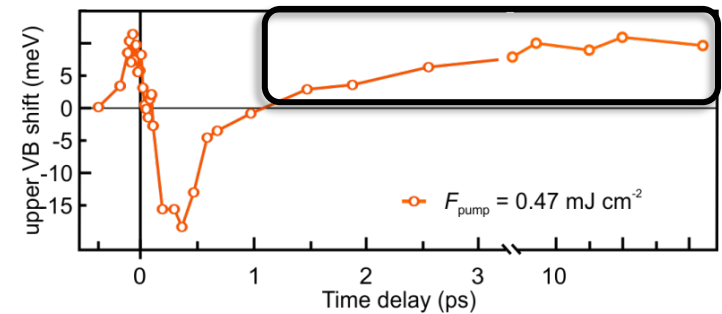
Photoenhancement of the excitonic insulator order parameter

Two-band semiconductor with interband interaction $H_{int} = U \sum_i n_{i,CB} n_{i,VB}$



- (1) mimicked BGR
- (2) *nonequilibrium* carrier population

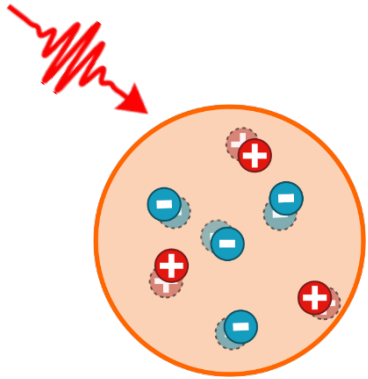
→ enhancement of the order parameter $\Delta \sim n_{\text{exciton}} \cdot E_B$



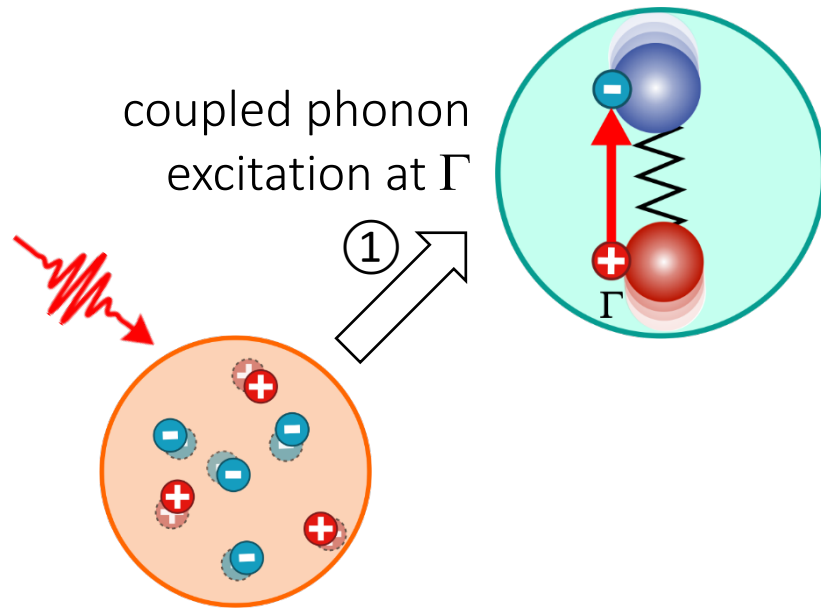
Hartree Fock calculations:
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S. Mor *et al.*, PRL 119, 086401 (2017)

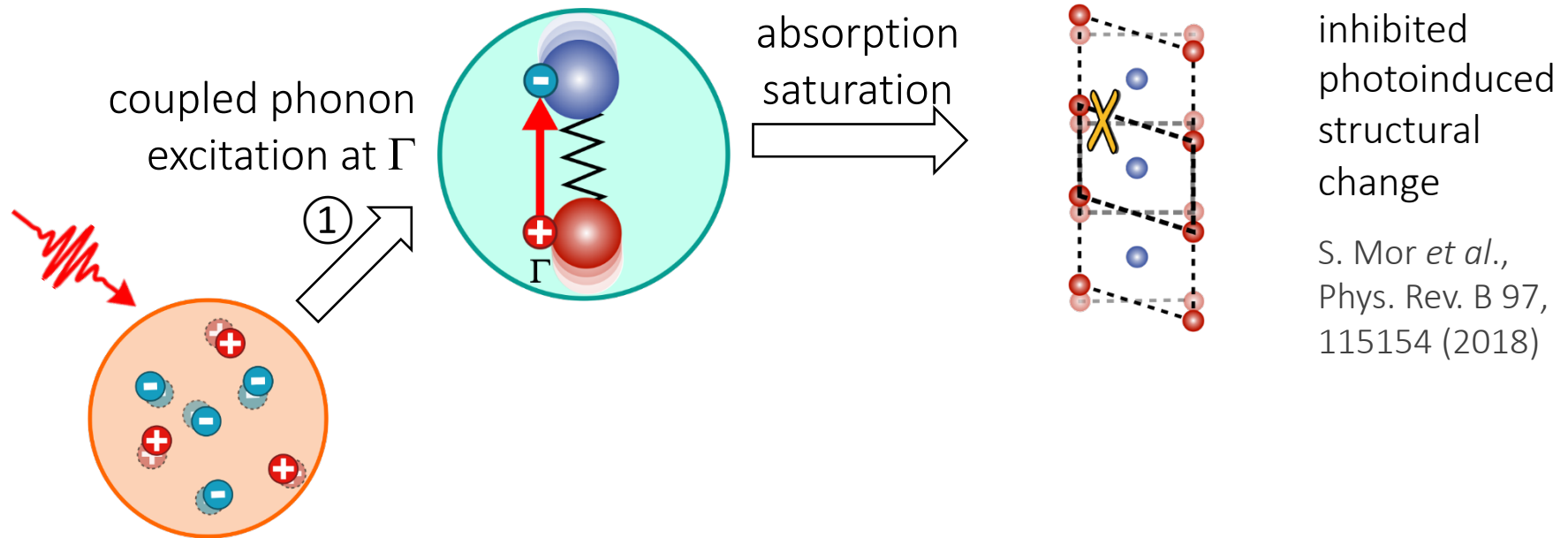
Summary: Ultrafast electronic and structural dynamics of Ta₂NiSe₅



Summary: Ultrafast electronic and structural dynamics of Ta_2NiSe_5

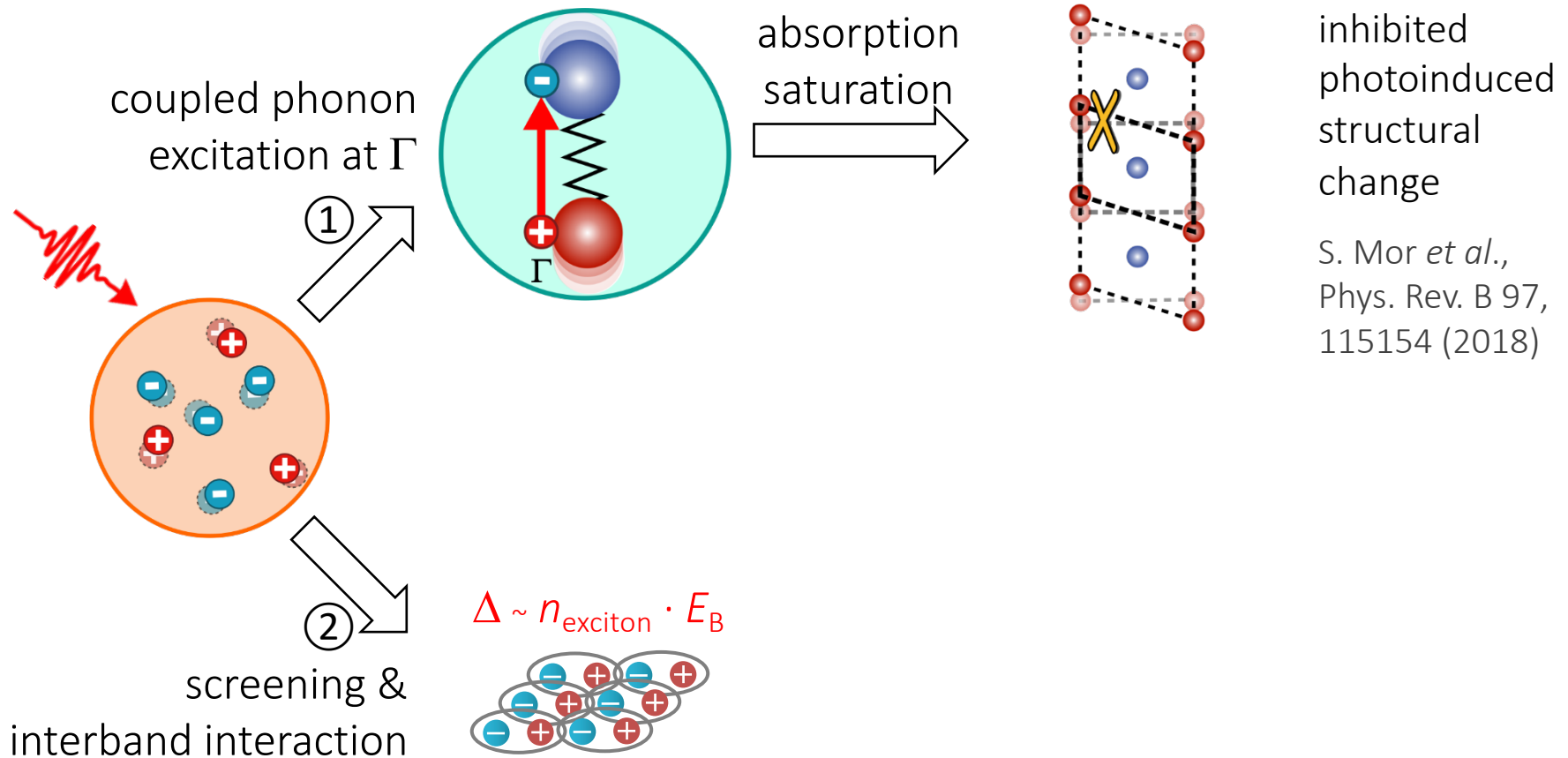


Summary: Ultrafast electronic and structural dynamics of Ta_2NiSe_5

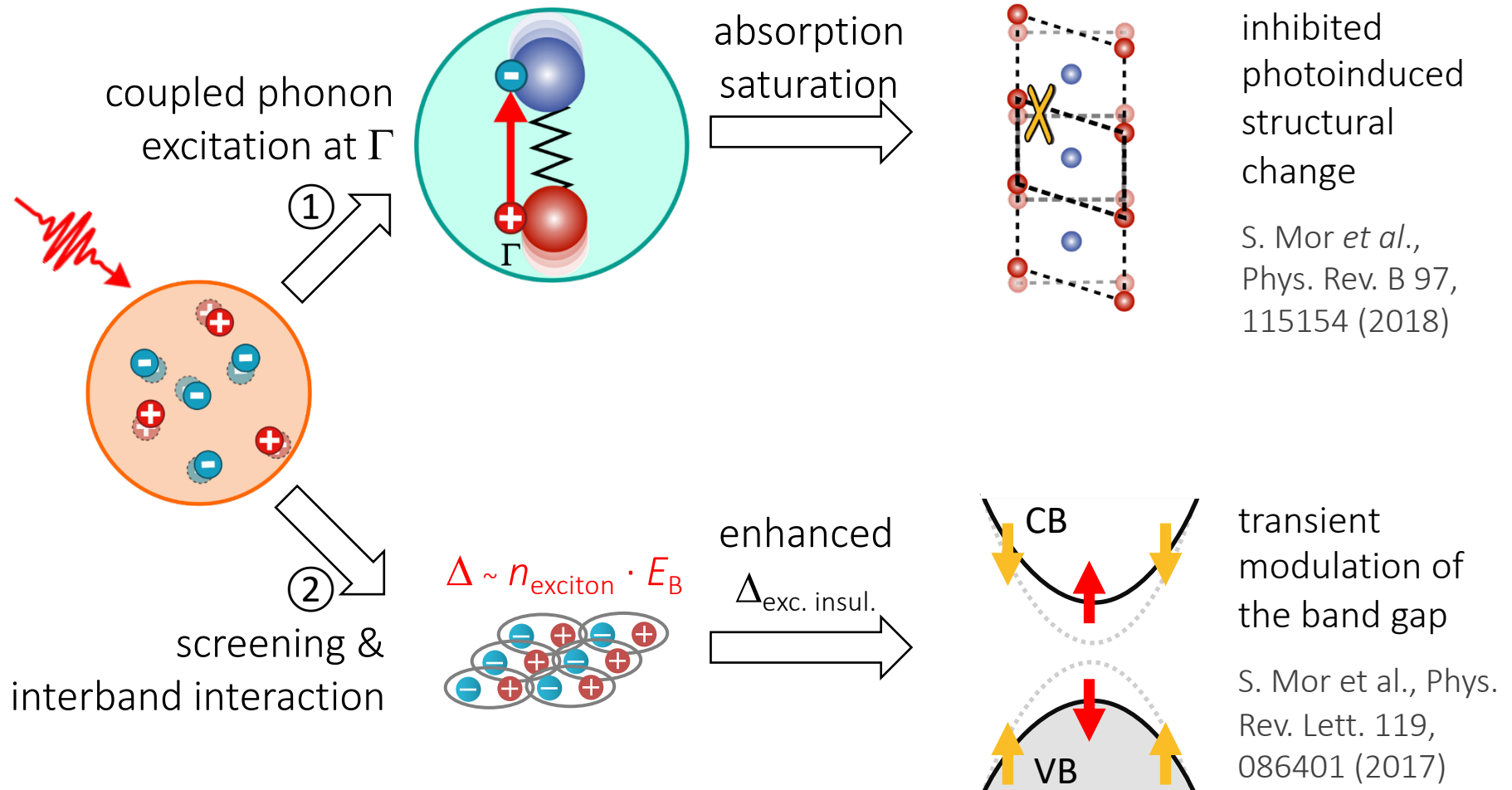


S. Mor *et al.*,
Phys. Rev. B 97,
115154 (2018)

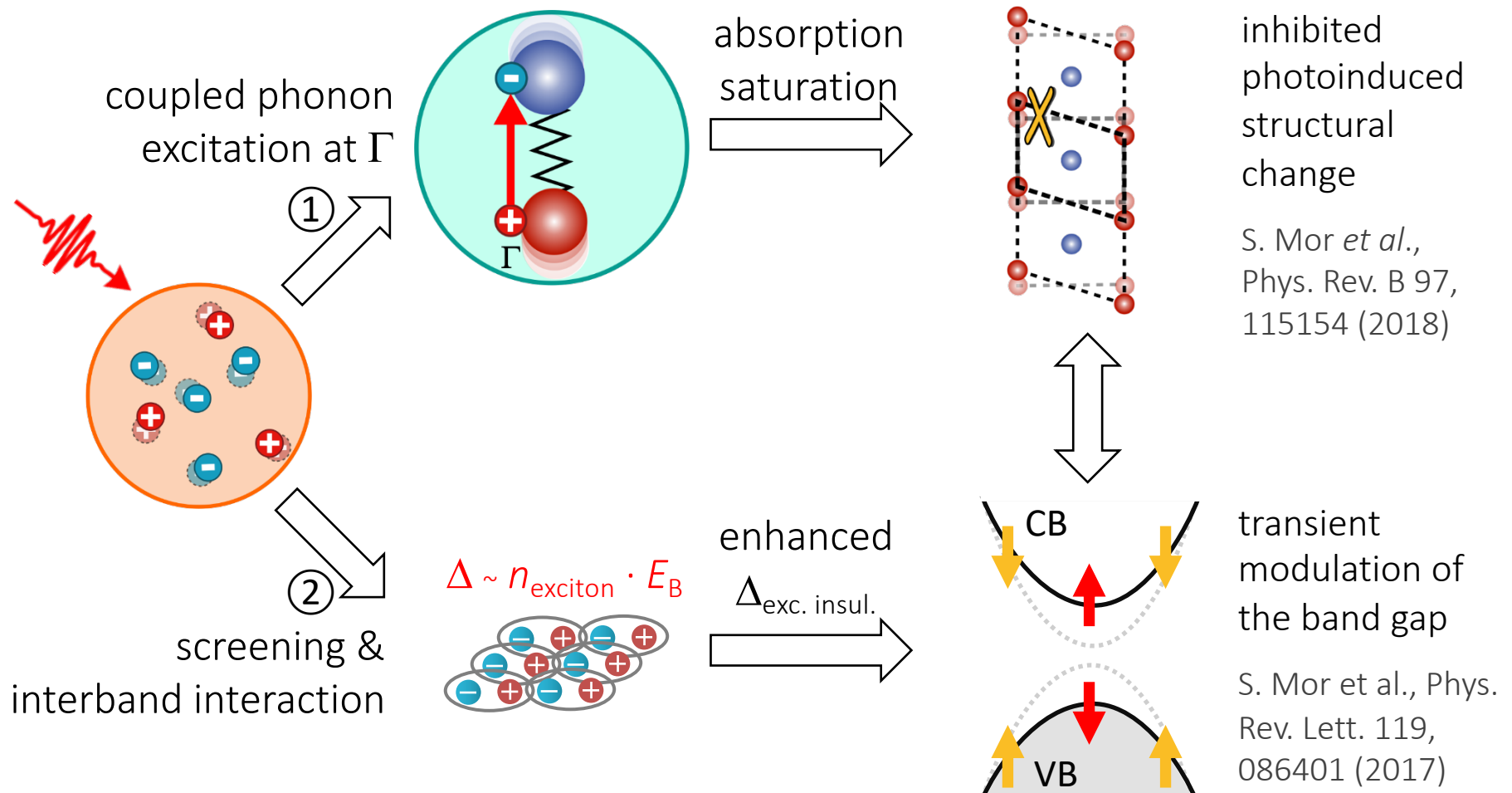
Summary: Ultrafast electronic and structural dynamics of Ta₂NiSe₅



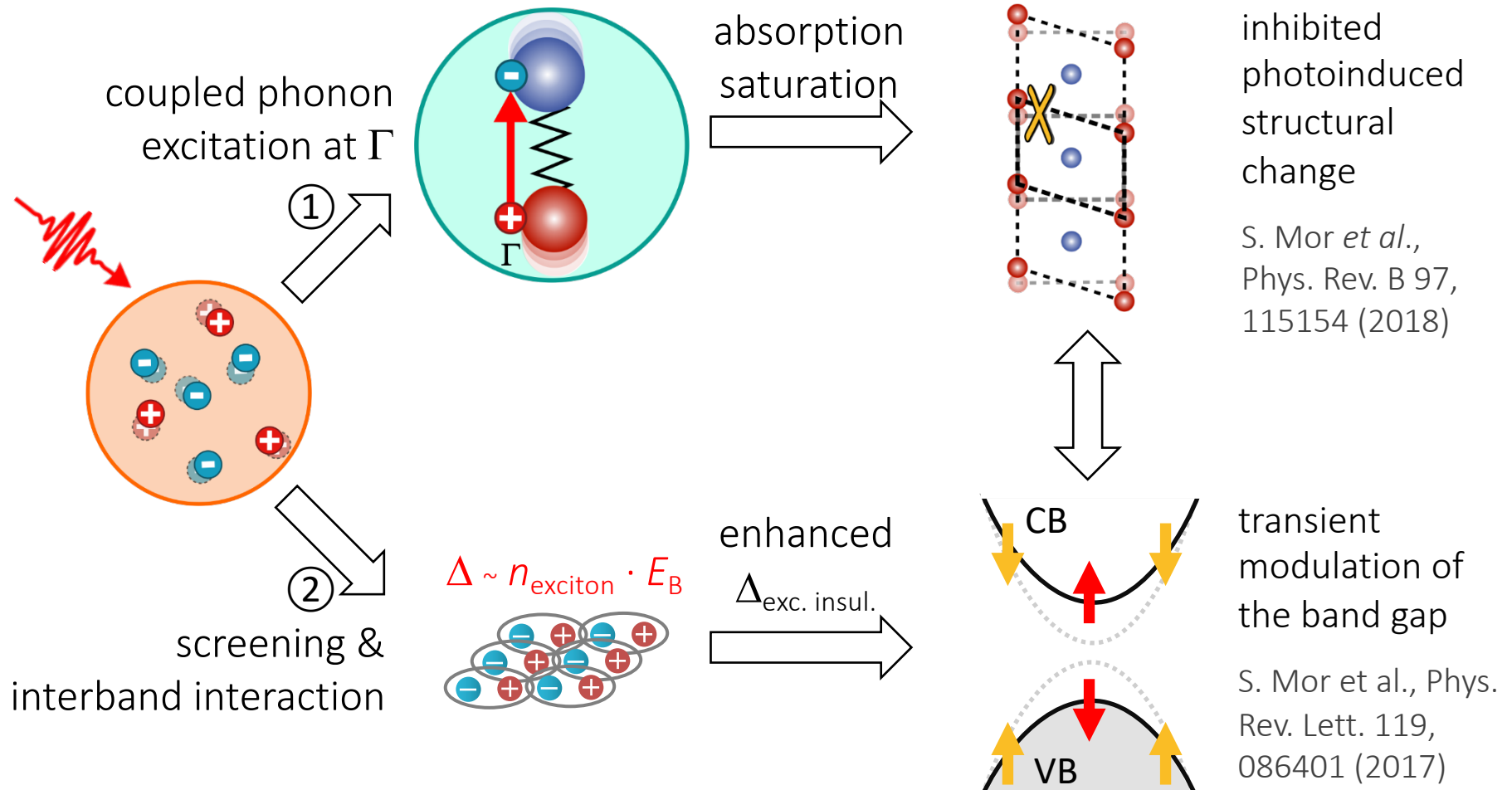
Summary: Ultrafast electronic and structural dynamics of Ta₂NiSe₅



Summary: Ultrafast electronic and structural dynamics of Ta₂NiSe₅



Summary: Ultrafast electronic and structural dynamics of Ta₂NiSe₅



Thank you!