Coherence in semiconductor nanostructures
Part V: Coherent dynamics and imaging of excitons in transition metal dichalcogenides and their heterostructures

Jacek Kasprzak

UNIWERSYTET WARSZAWSKI

ZIP ZINTEGROWANY PROGRAM ROZWOJU

Equipe mixte CEA-CNRS "Nanophysique et semicondcuteurs"
Institut Néel - CNRS
Grenoble France
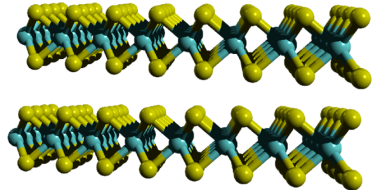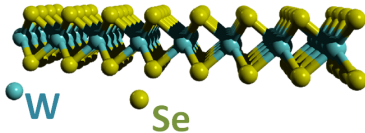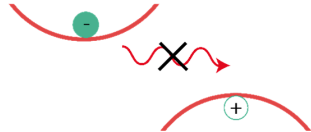
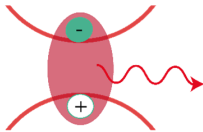Warsaw University, October-December 2020

Outline

Optically active 2D materials

K. F. Mak Phys. Rev. Lett. 105, 136805 (2010): "a crossover to a direct-gap material in the limit of the single monolayer (...) increase in luminescence by 10^4 compared with the bulk"
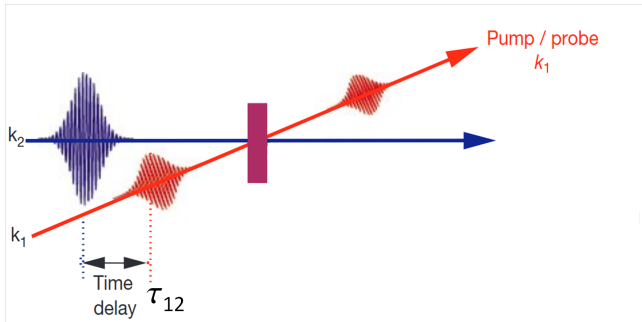
Four-wave mixing (FWM)
inferring coherence, correlations & kinetics in solids

📄 **Chemla & Shah Nature (2001), Axt & Kuhn Rep. Prog. Phys. (2004)**

FWM$\propto \mu^4 \mathcal{E}_1^* \mathcal{E}_2 \mathcal{E}_2 \Rightarrow$ Photon echo resolves σ and γ
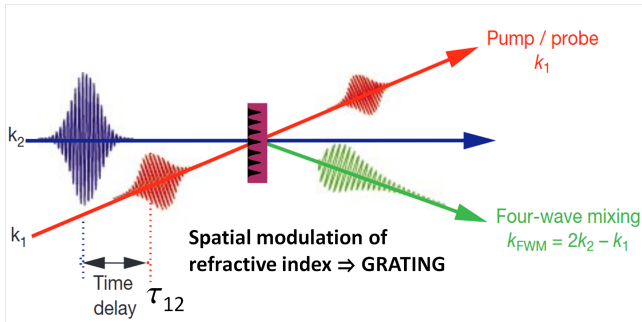
Major issue: spatial averaging $\Rightarrow$ Microscopy Required

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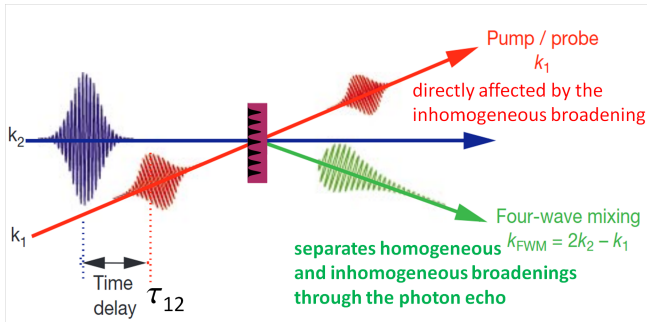
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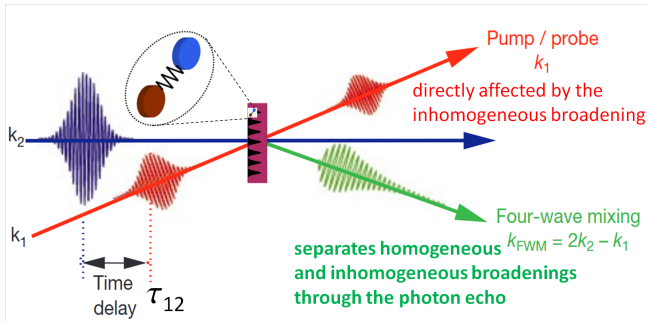
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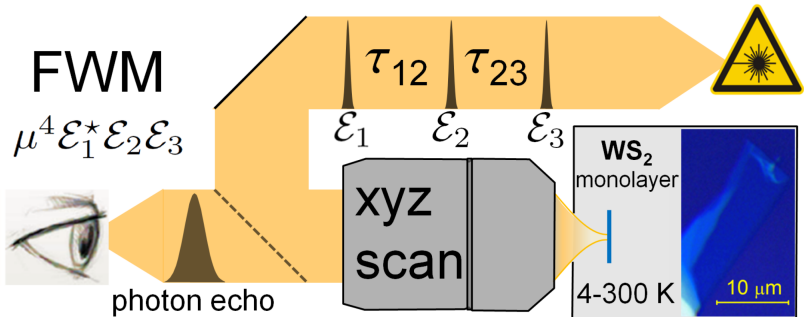FWM $\propto \mu^4 \mathcal{E}_1^* \mathcal{E}_2 \mathcal{E}_2 \Rightarrow$ **Photon echo resolves σ and γ**

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FWM micro-spectroscopy ⇒ optical lock-in

W. Langbein et al. Optics Letters 31, 1151 (2006)

intensly developed & exploited in Grenoble

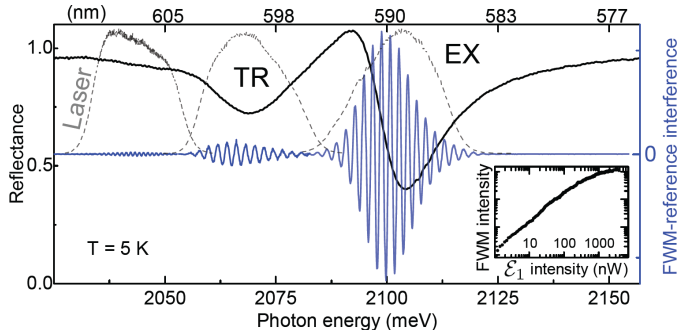## 3-beam heterodyne detection & spectral interferometry

Measurement of the exciton polarization and density dynamics
with an enhanced spatio-temporal resolution: (100 fs, 300 nm)

FWM micro-spectroscopy ⇒ optical lock-in

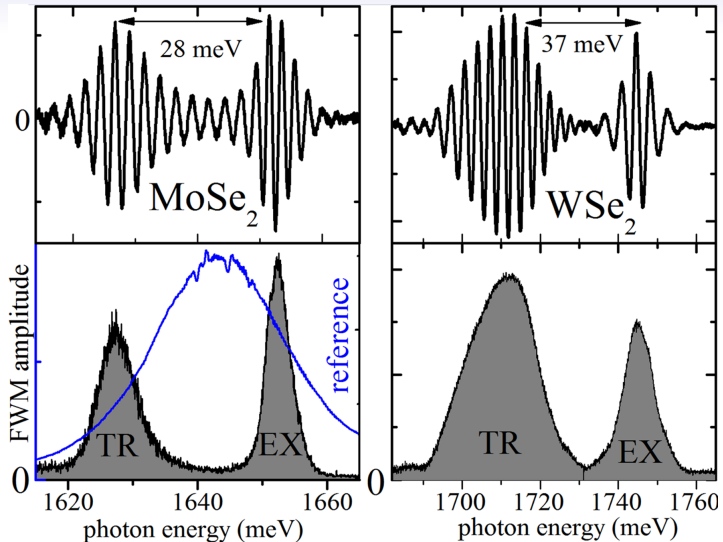W. Langbein et al.Optics Letters 31, 1151 (2006)

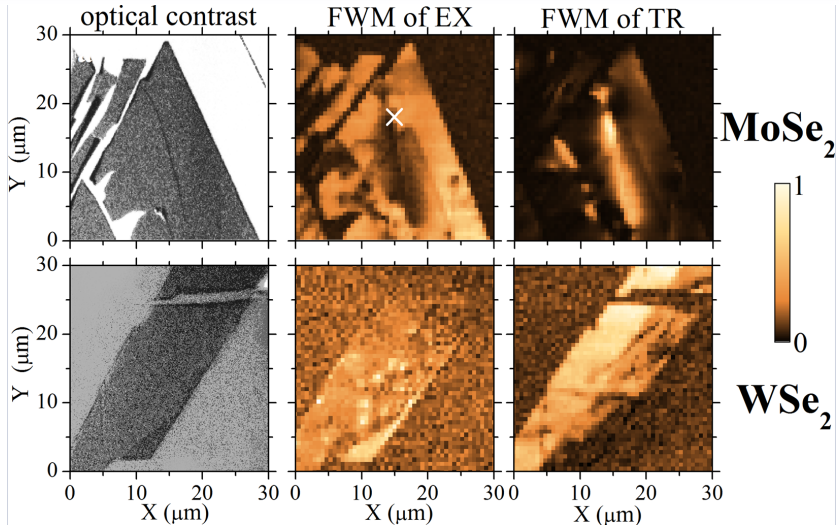intensly developed & exploited in Grenoble

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FWM response and imaging of MoSe₂ and WSe₂
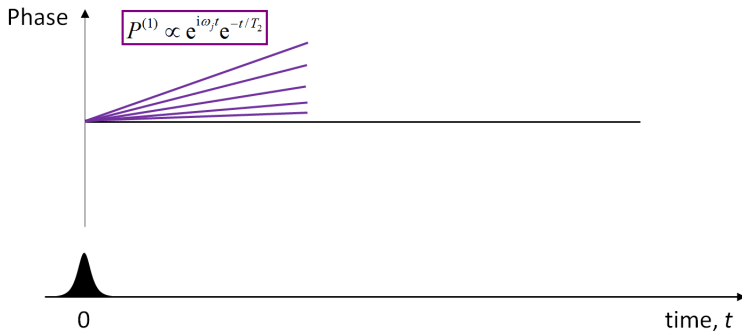
FWM response and imaging of MoSe$_2$ and WSe$_2$

Outline

1. Methodology

2. **Dephasing**

3. Density

4. Heterostructures

5. Outlook & Discussion

Why it is worth to look into FWM $\propto \mathcal{E}_1^* \mathcal{E}_2 \mathcal{E}_2$?

Because it offers an access to homogeneous dephasing time $T_2 = 2\hbar/\gamma$
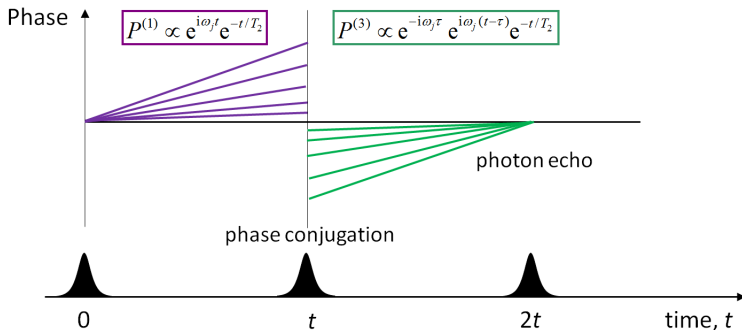in the presence of spectral inhomogeneous broadening σ via **photon echo**

$$P^{(1)} \propto e^{i\omega_j t} e^{-t/T_2}$$

Phase

0   time, t

Rephasing of all polarizations at $t = 2\tau \Rightarrow$
FWM is only sensitive to microscopic dephasing, independent of σ.
σ is inferred through the time-spread of the echo.

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$P^{(3)} \propto \mathrm{e}^{-\mathrm{i}\omega_j \tau} \, \mathrm{e}^{\mathrm{i}\omega_j (t-\tau)} \mathrm{e}^{-t/T_2}$

Phase

photon echo

phase conjugation

0   t $2t$ time, t

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Dephasing of exciton ensembles in 2D materials

Homogenous γ and inhomogeneous σ widths, correlations

Nano Letters (2016), 2D Materials (2017), ACS Nano (2019), Phys. Rev. Materials (2020), SPIE Proceed. (2020)
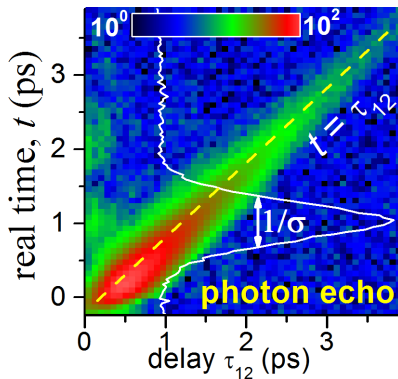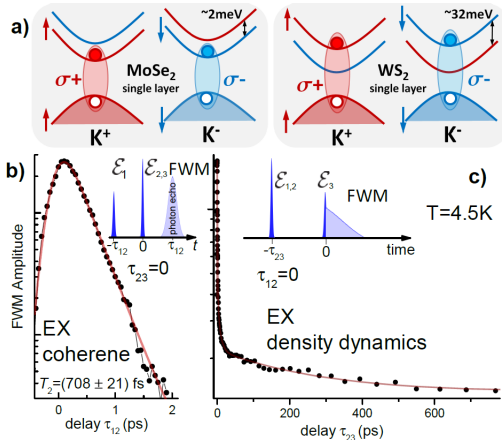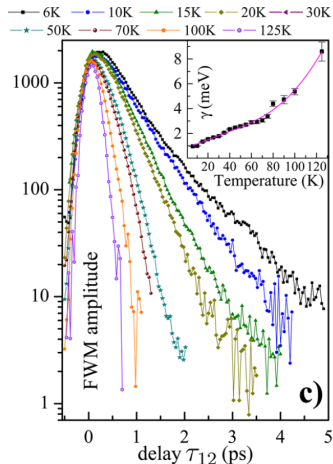

Photon echo versus τ_{12} $\Rightarrow$ measures and separates γ and σ

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Correlations: Localization vs. Lifetime

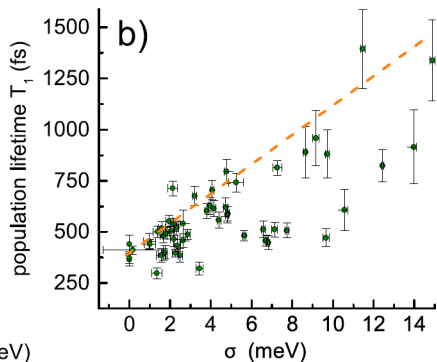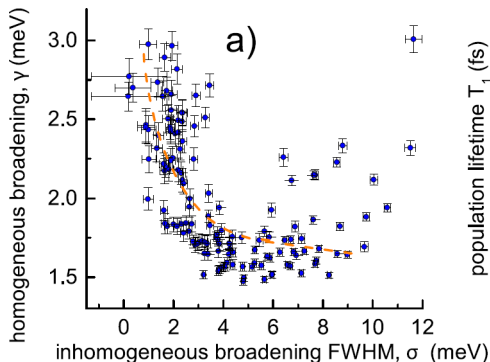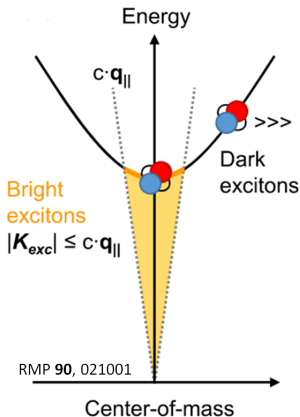Exploiting micro-spectroscopy: homogeneous (γ) versus inhomogeneous (σ) broadenings
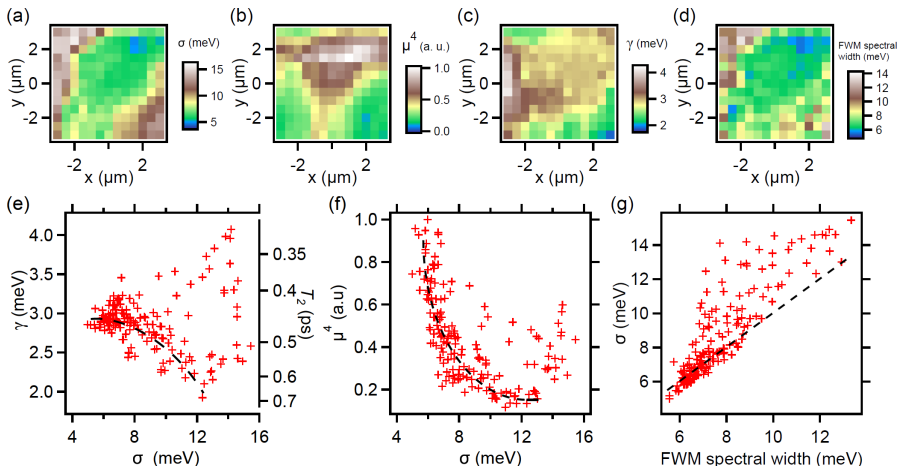
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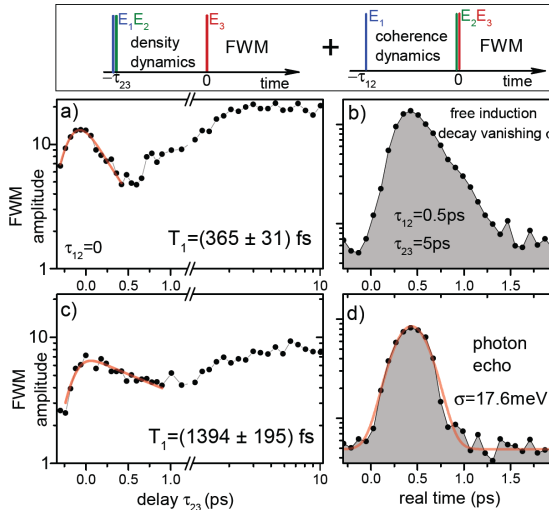
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1. Methodology

2. Dephasing

3. **Density**

4. Heterostructures

5. Outlook & Discussion

FWM used as "pump-probe": density dynamics

complex FWM field $\Rightarrow$ interferences vs. τ_{23}, access to the phase

Involved processes:
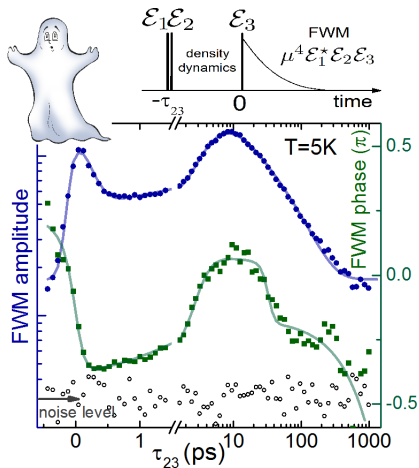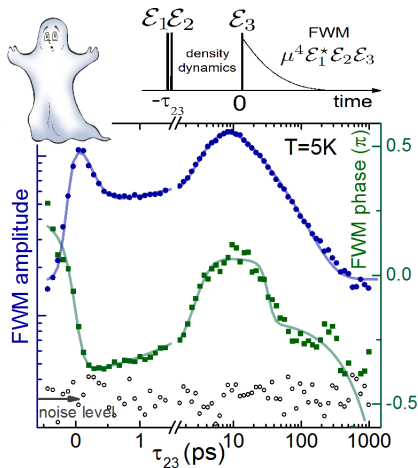
1. initial rise from negative delays (autocorrelation)

2. expected decay on 1 ps scale: radiative + scattering to dark states, valleys

3. unexpected rise owing to interaction enhancement with dark states (?)

4. final power law decay on a few ns scale

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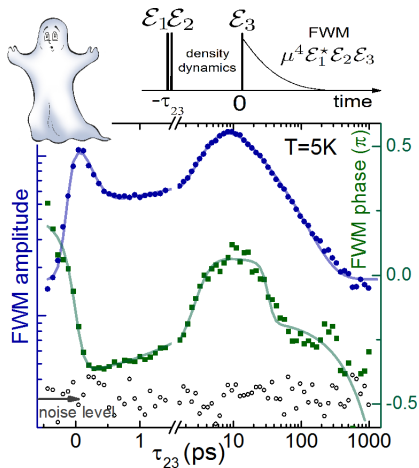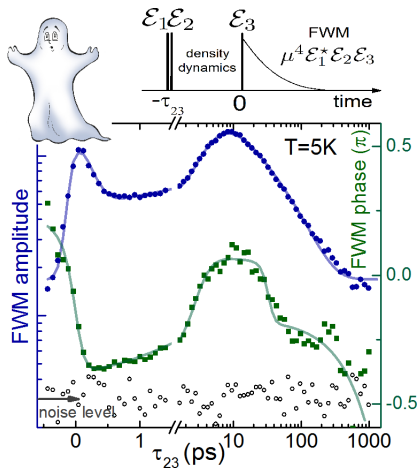

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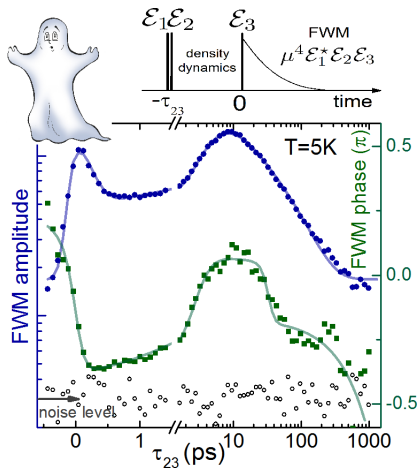complex FWM field $\Rightarrow$ interferences vs. τ_{23}, access to the phase

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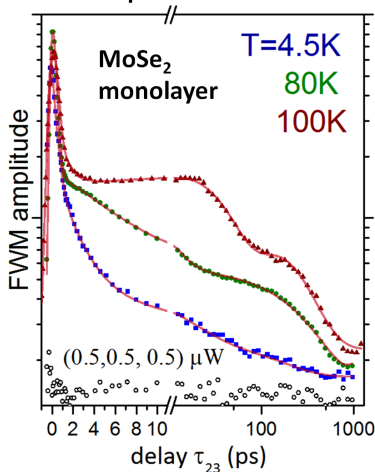complex FWM field $\Rightarrow$ interferences vs. τ_{23}, access to the phase

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ubiquitous in TMDs

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Manifestation of the valley degree of freedom

Raman beats between intra- and inter-valley trions in WS_2
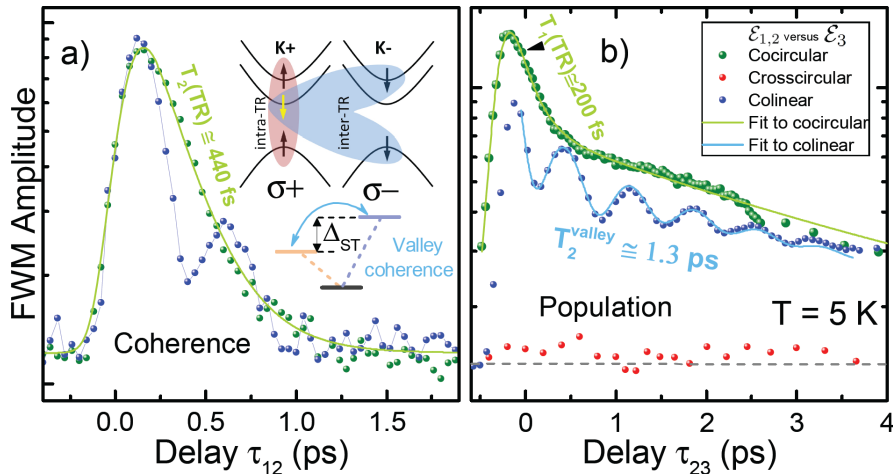
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4 **Heterostructures**

5 Outlook & Discussion

Improving optical quality with heterostructures
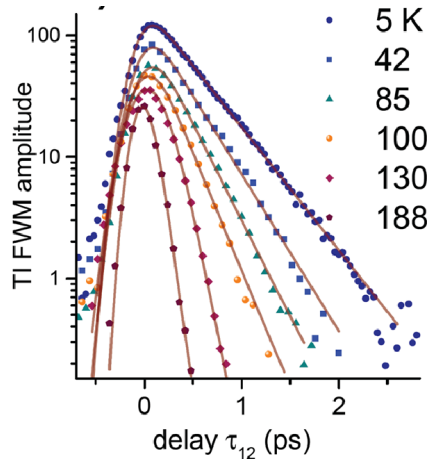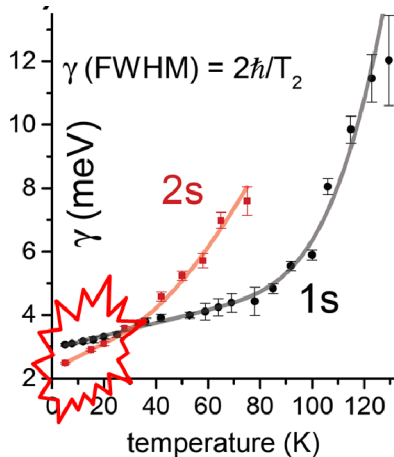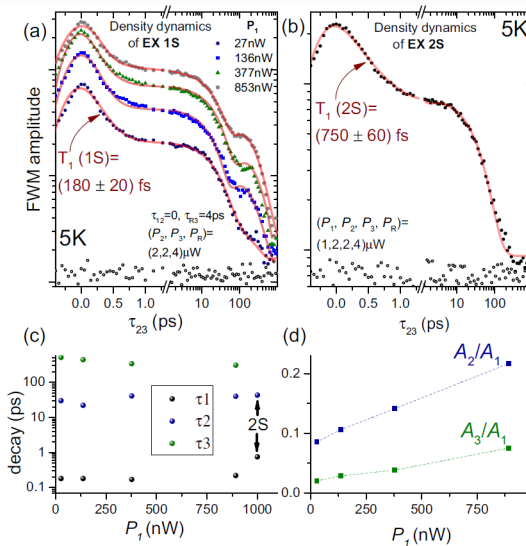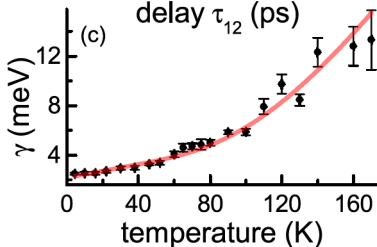Flattening, Shielding & Isolating from excess charges $\Rightarrow$ suppressing σ

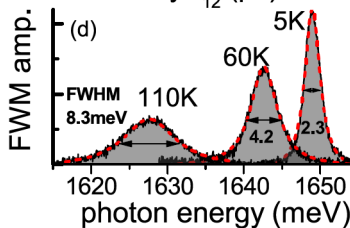# Improving optical quality with heterostructures
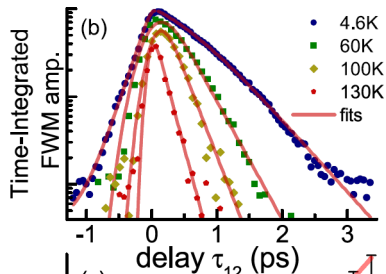Flattening, Shielding & Isolating from excess charges $\Rightarrow$ suppressing σ

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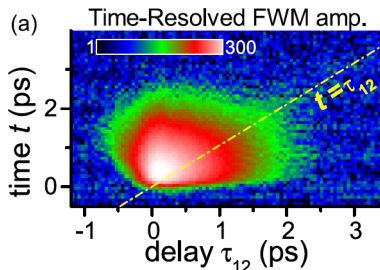Flattening, Shielding & Isolating from excess charges $\Rightarrow$ suppressing σ

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Conclusions & Outlook

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- Robust excitons in TMDs, generating enhanced linear absorption.

- Giant, nonlinear optical responses: SHG, THG, FWM.

- FWM $\Rightarrow$ insight into dynamics on a sub-micron scale: radiative lifetime influenced by localization.

- Step-like improvement of optical properties in TMD heterostructures, 2D Rydberg excitons.

Spectroscopy of TMDs: outlook

- $\Rightarrow$ Polaritonics

- $\Rightarrow$ Nanophotonics

- $\Rightarrow$ Optomechanics

- $\Rightarrow$ Single emitters

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Personal outlook
Polaritonics with 2D materials: quantum fluids of light
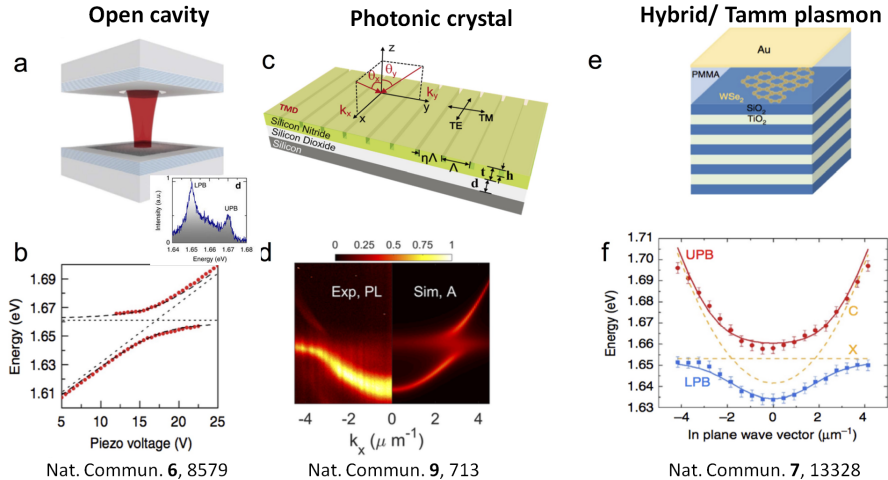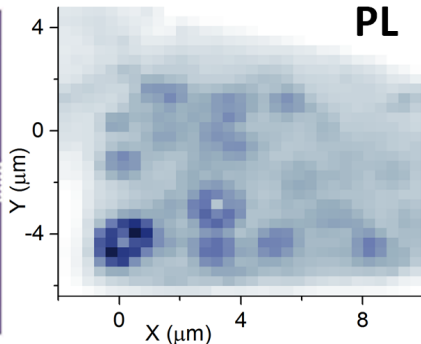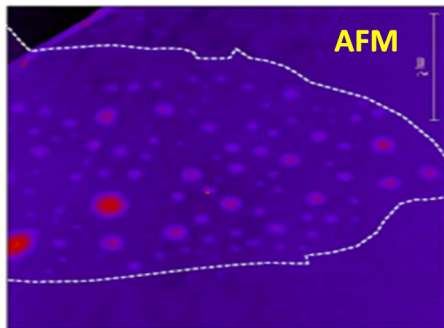
Open cavity

a

Photonic crystal

c

Hybrid/ Tamm plasmon

e

b

d

f

Nat. Commun. **6**, 8579

Nat. Commun. **9**, 713

Nat. Commun. **7**, 13328

Personal outlook
Nanophotonics with 2D materials

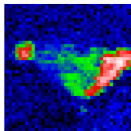Lensing by the micron-size defects in a **hBN/WS$_2$/hBN** heterostructure

Personal outlook
Nanophotonics with 2D materials
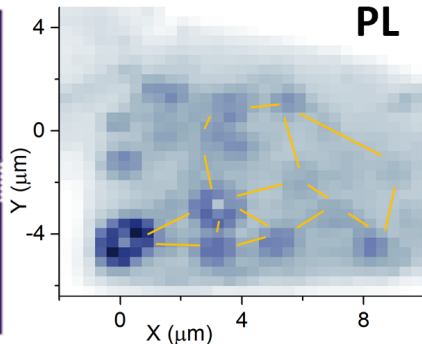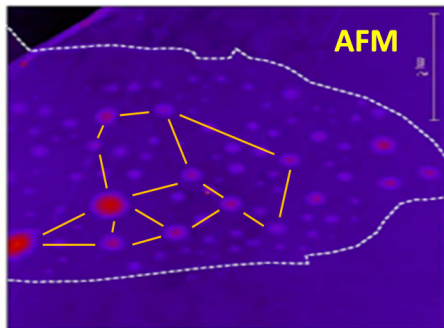
Lensing by the micron-size defects in a **hBN/WS$_2$/hBN** heterostructure

Personal outlook
Merging coherent spectroscopy and optomechanics
of suspended 2D materials

FWM image

20 µm

Y (µm)

0

X (µm)

20 µm

3 MLs

1 ML

opening for gas exchange

10.6 µm
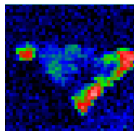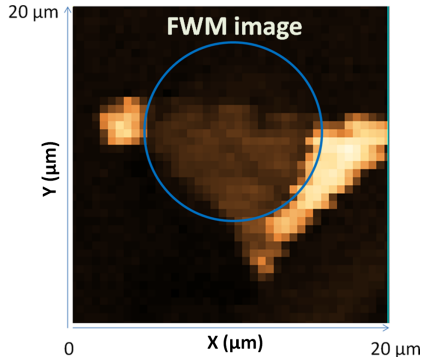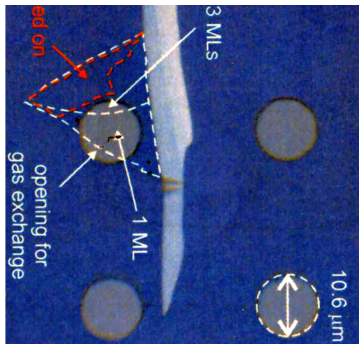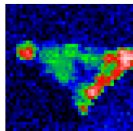
| 1655 meV | 1653 meV | 1650 meV | 1649 meV | 1648 meV |

Personal outlook

Toward coherent spectroscopy of single emitters in 2D Materials
driven by local strain/disorder, ...but on-demand arrays now at hand

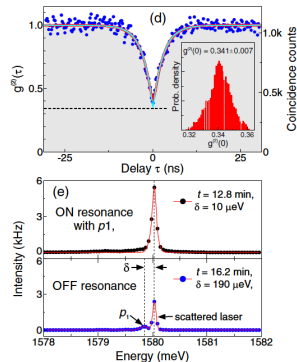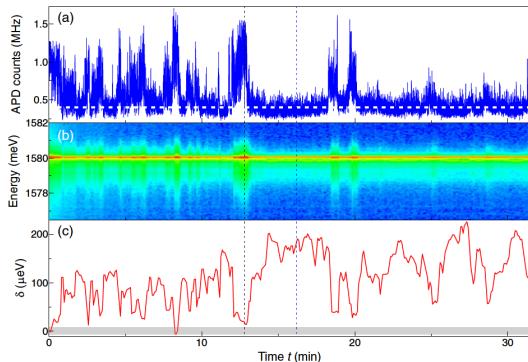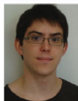# Personal outlook
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Nat. Commun. **8**, 15093
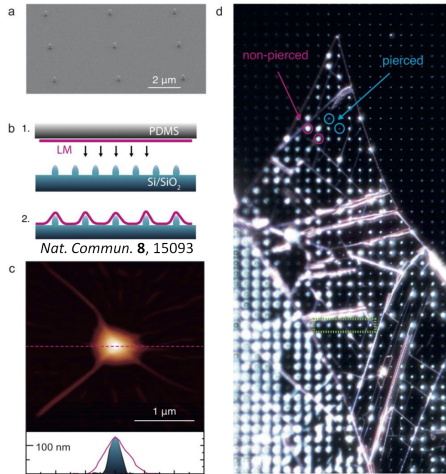
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Acknowledgement

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NÉEL institut

J-M. Gérard   J. Claudon   E. Peinke   cea INAC INSTITUT NANOSCIENCES ET CRYOGÉNIE   W. Langbein   CARDIFF UNIVERSITY PRIFYSGOL CAERDYDD   W. Pacuski

S. Höfling   M. Kamp   C. Schneider   Julius-Maximilians- UNIVERSITÄT WÜRZBURG   T. Kuhn   D.E. Reiter   D. Wigger   WESTFÄLISCHE WILHELMS-UNIVERSITÄT MÜNSTER

S. Reitzenstein   S. Fischbach   S. Rodt   M. Richter   TU berlin   M. Potemski   K. Nogajewski   M. Molas   LNCMI

Acknowledgement

Marek Potemski & his group
« **Magneto-optics of layered materials** »

Auxiliary results, WS₂
Photoluminescence hyperspectral imaging
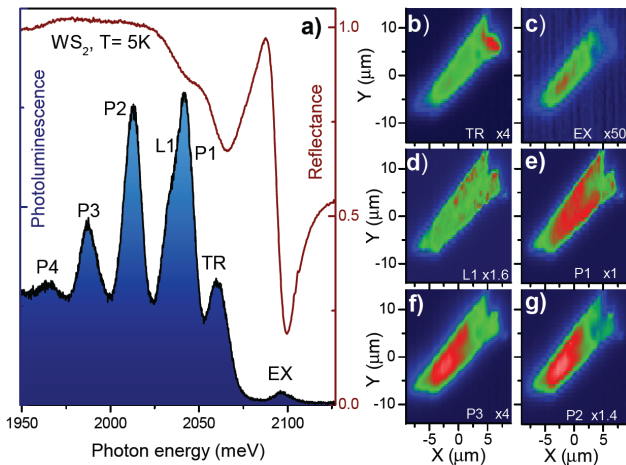
Auxiliary results, WS$_2$
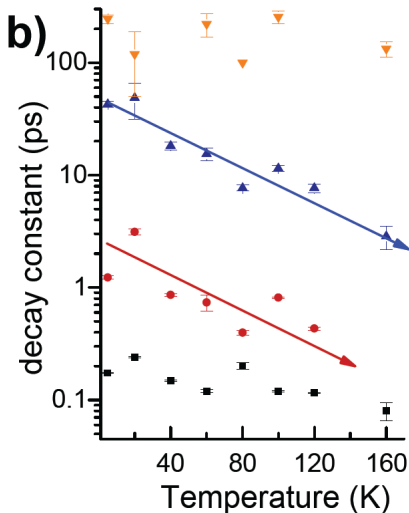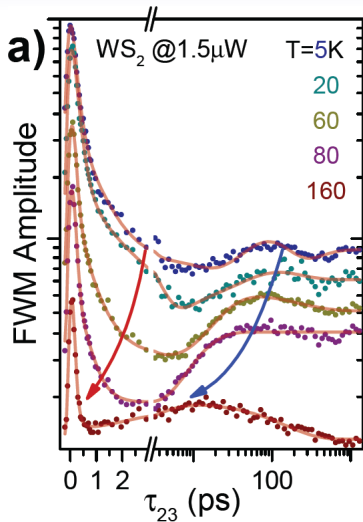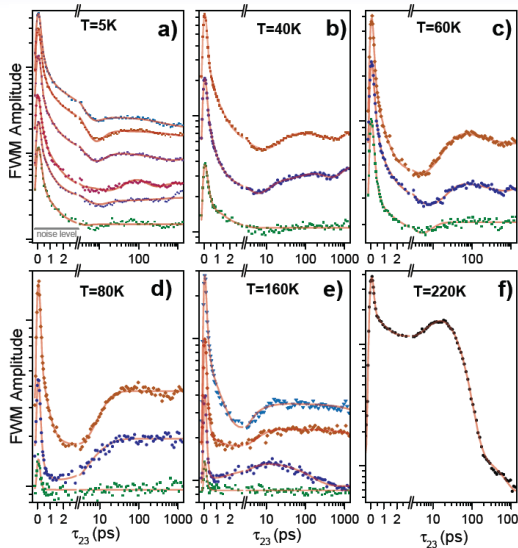## Raman beats of trions
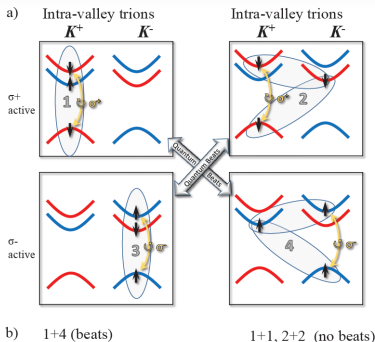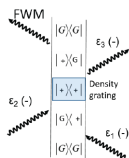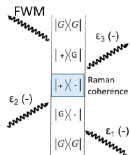
Auxiliary results, WS$_2$

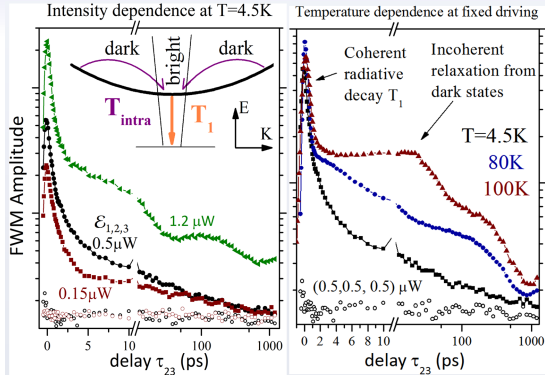## Density dynamics on a nano-second timescale

Auxiliary results, WS$_2$

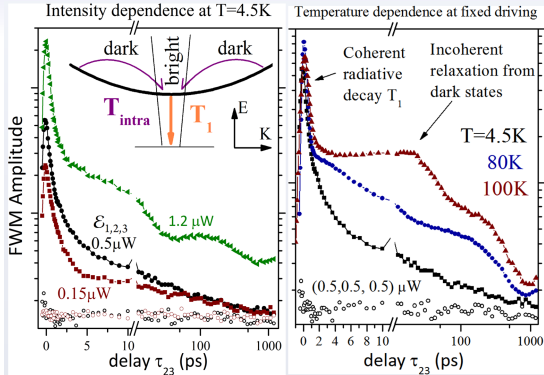## Density dynamics on a nano-second timescale

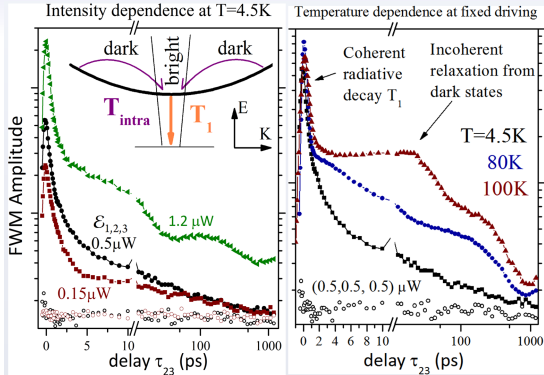# Exciton density dynamics in MoSe$_2$

- Initial decay $\Rightarrow$ radiative removal of coherent exciton density
- Later dynamics $\Rightarrow$ complex relaxation from dark states
- Enhanced dynamic range owing to the heterodyne detection
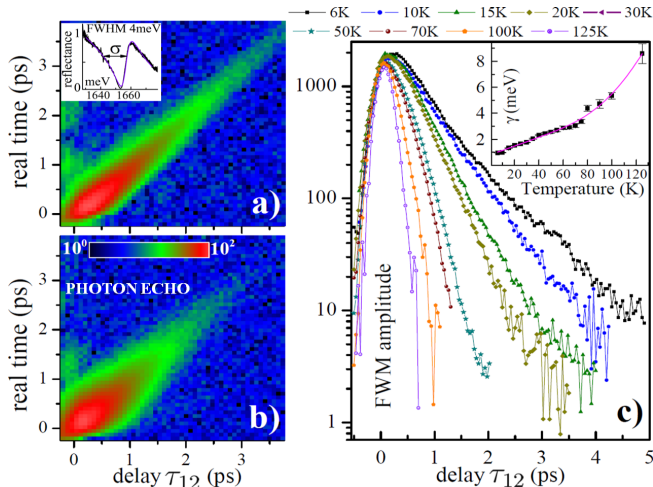
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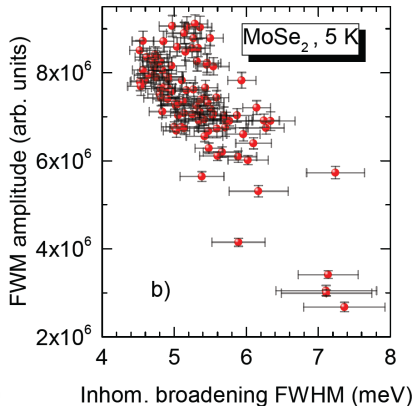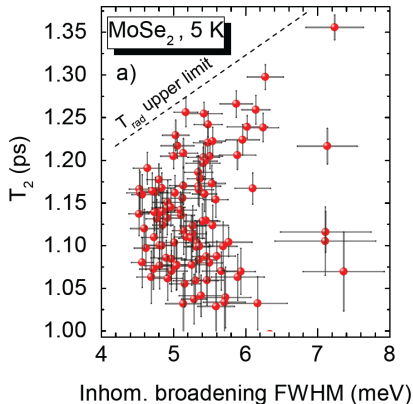
Exciton coherence dynamics in MoSe$_2$

Photon echo $\Rightarrow$ fingerprint of disorder (on a sub-μm scale)
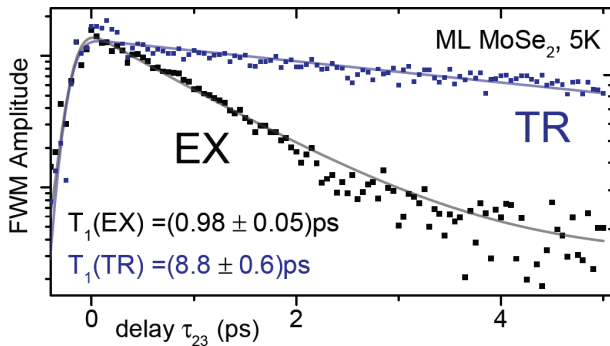
Phonon-induced dephasing $\Rightarrow$ as in semiconductor QWs

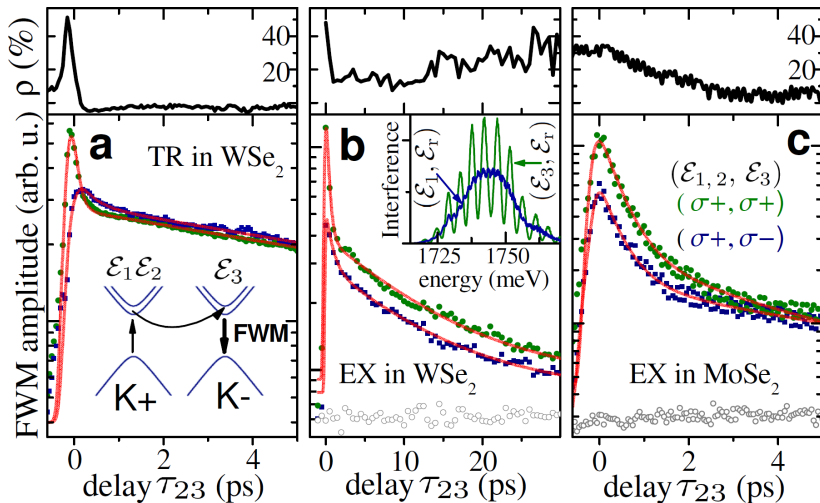# Exciton coherence dynamics in MoSe$_2$

correlations: T_2, σ, μ

Exciton coherence dynamics in MoSe$_2$
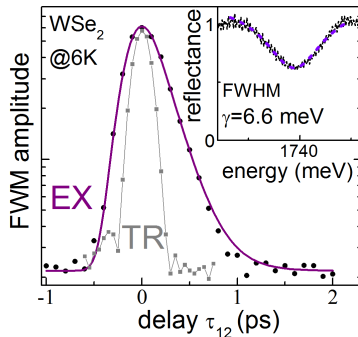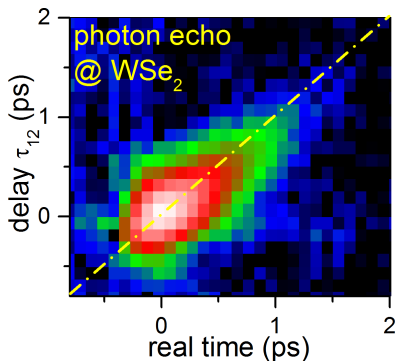## Exciton versus Trion lifetime

Inter-valley population dynamics in WSe$_2$ & MoSe$_2$
polarization-resolved driving of the FWM

Exciton & trion coherence in WSe$_2$ at 5K

Trion case: shorter dephasing, yet longer lifetime

Dynamics of secondary excitons at 5K
$MoSe_2$ versus WSe_2