NanoGe Spring Meeting 2022

Online Format

Ultrafast Continuum Infrared Spectroscopy of *n* - type HgSe Quantum Dots

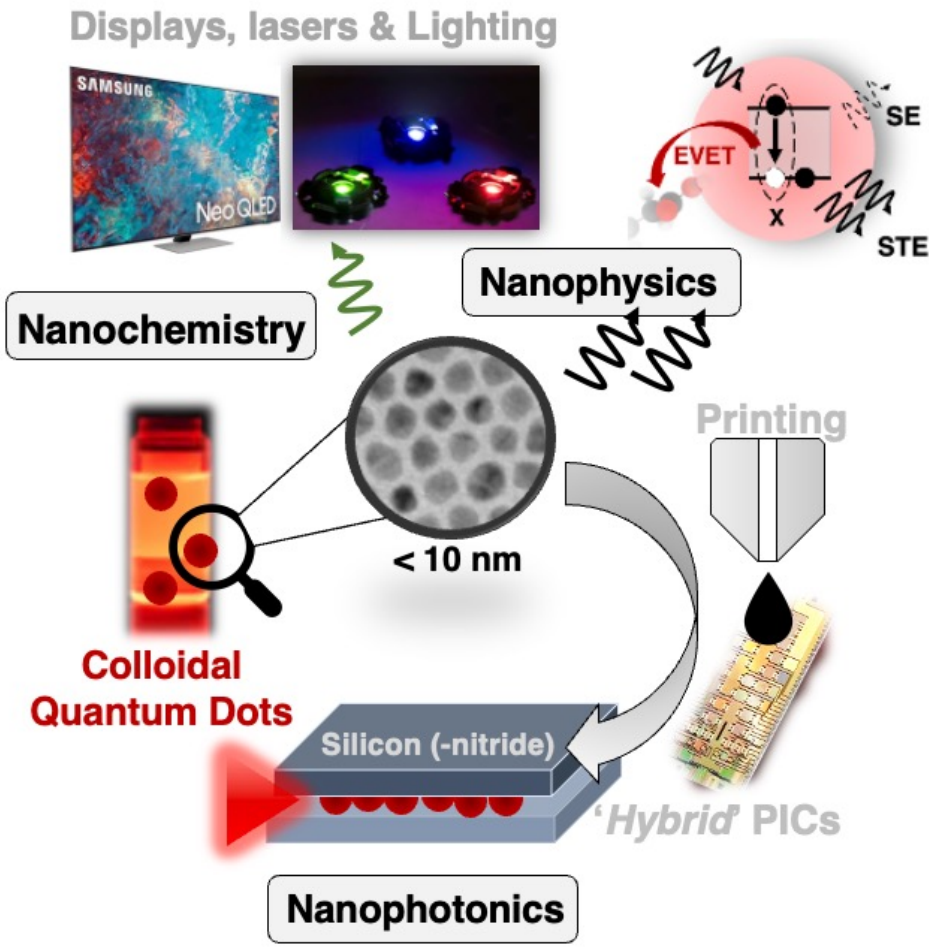
Jari Leemans, Zeger Hens, ***Pieter Geiregat****,
Department of Chemistry, UGent

Christophe Delerue
IEMN Lille, France

Ashley M. Stingel, Poul B. Petersen
Department of Chemistry, RU Bochum

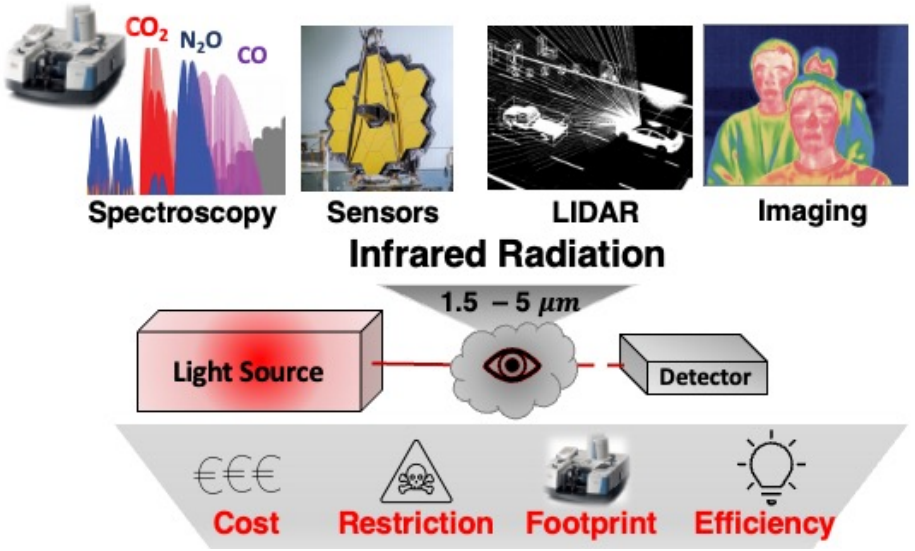
Pieter.Geiregat@ugent.be
@Pieter_Geiregat

Printable Infrared Opto-Electronics



Geiregat *et al*. "A Bright Future for Colloidal Quantum Dot Lasers." *NPG ASIA MATERIALS*, vol. 11, 2019

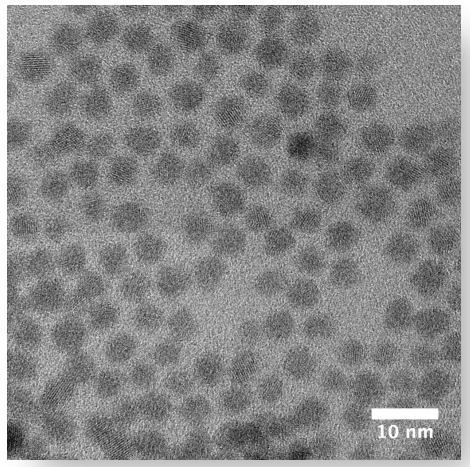
Can we extend the **optimization cycle** of VIS/NIR quantum dots (QDs) towards the **short and mid-wave infrared ?**

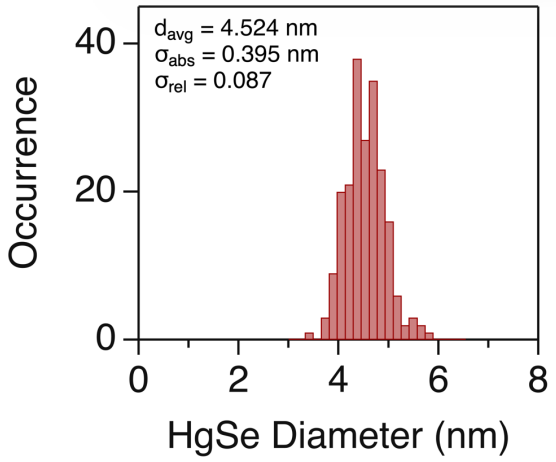


Step 1 ?

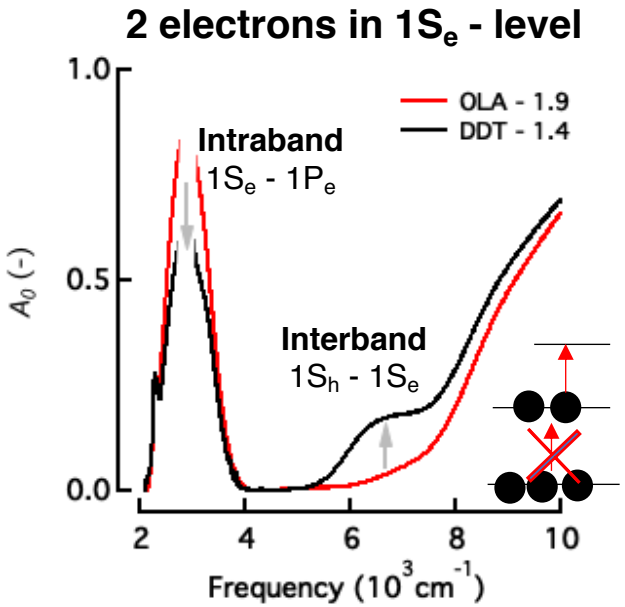
Study of fundamental light-matter interactions on relevant time-scales

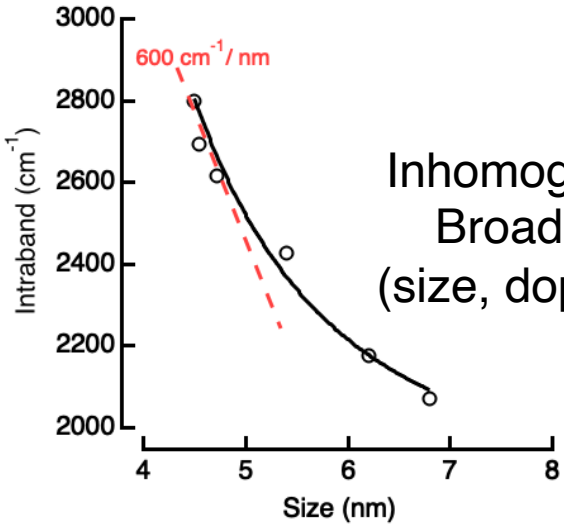
Intraband Transitions in *n*-doped HgSe Quantum Dots



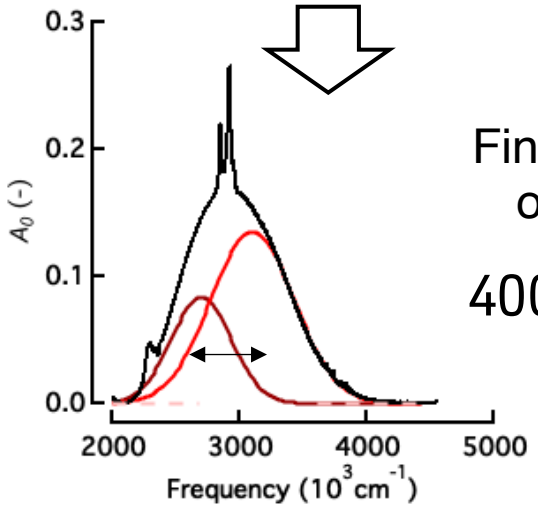


Chem. Mater. 2018, 30, 21, 7637–7647



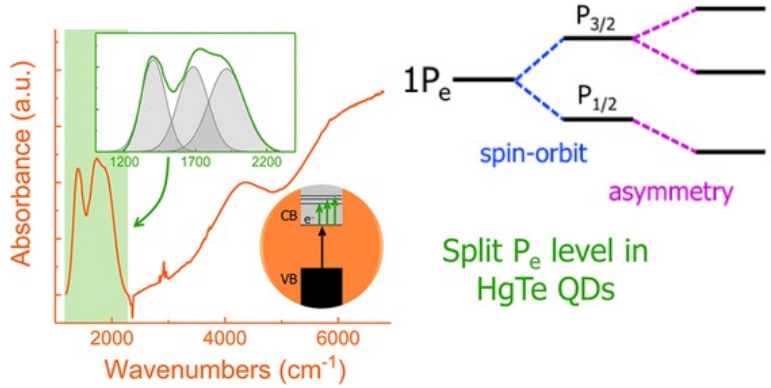


Inhomogeneous Broadening (size, doping, …)



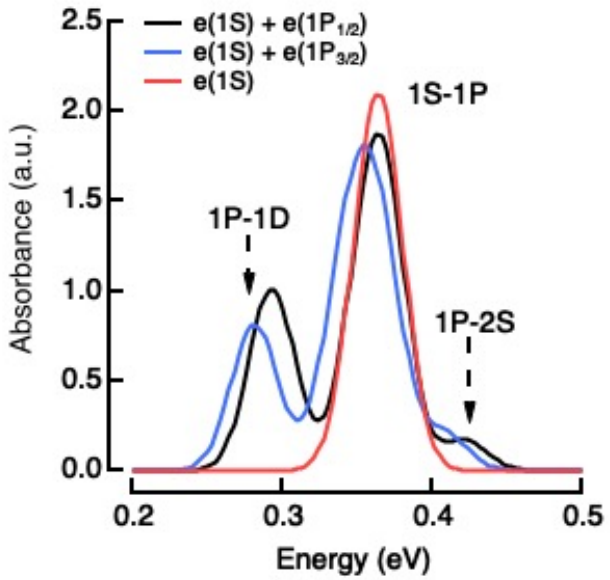
Fine Structure of 1S-1P !

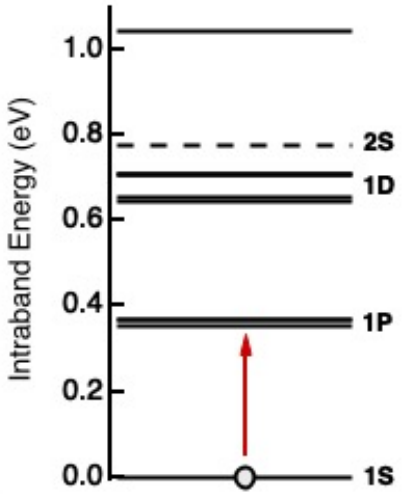
400 cm^{-1}=2k_BT

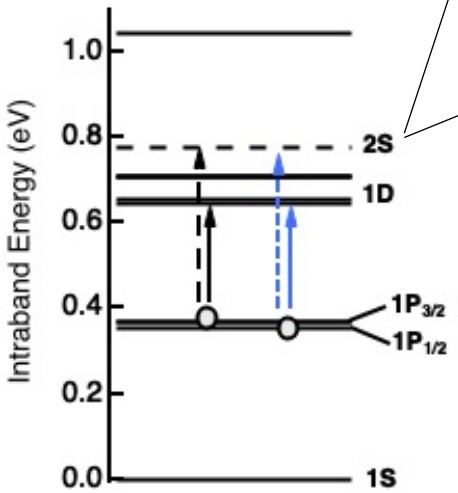


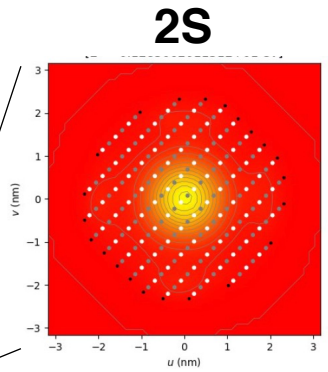
ACS Nano 2018, 12, 9, 9397–9404

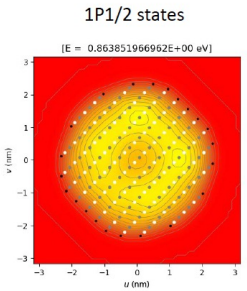
n-Doped HgSe Quantum Dots - Energy level structure



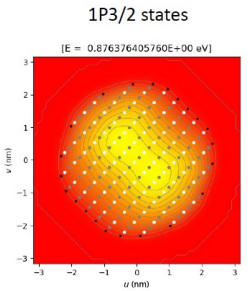




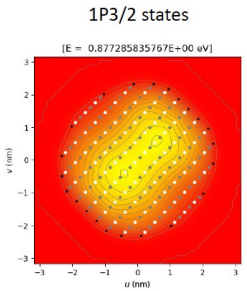




$1P_{1/2}$

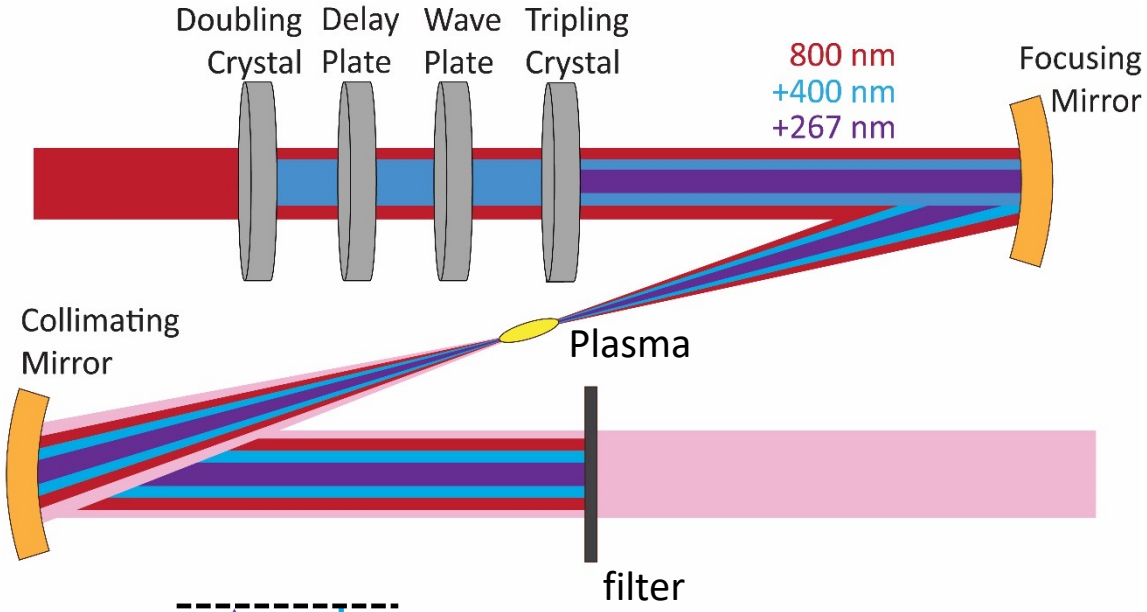


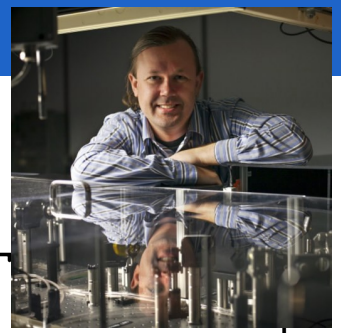
$1P_{3/2}$

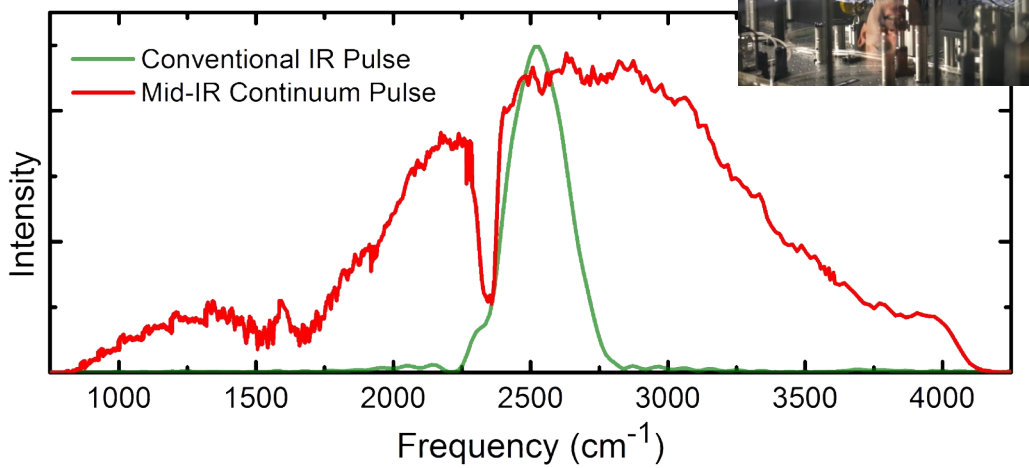


$1P_{3/2}$

Ultrafast Continuum Infrared Spectroscopy







- Sub-100 fs time resolution
- Spectral coverage 1000 - 4000 cm^{-1}
- Coverage of the entire spectrum in a single shot

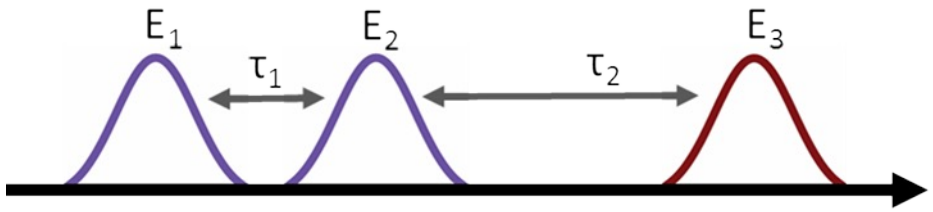
⇨ Very suited to study broad inter and intra-band transitions in QDs !

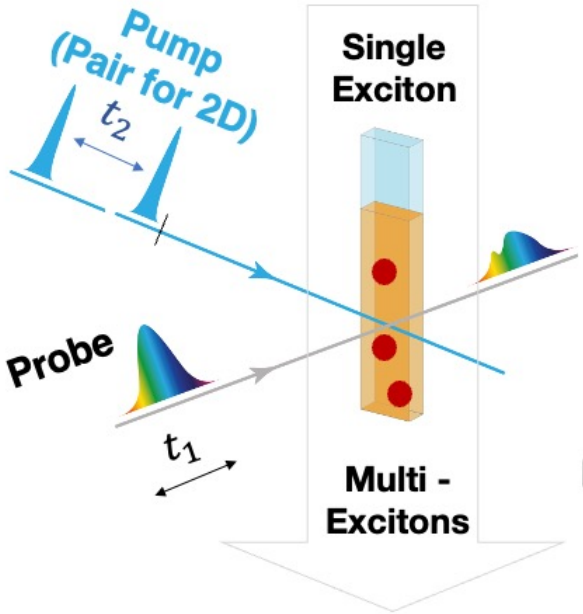
P. B. Petersen, A. Tokmakoff, *Opt. Lett*. 35, 1962-1963 (2010)
C. Calabrese, A. M. Stingel, L. Shen, P. B. Petersen, *Opt. Lett.* 37, 2265-2267 (2012)

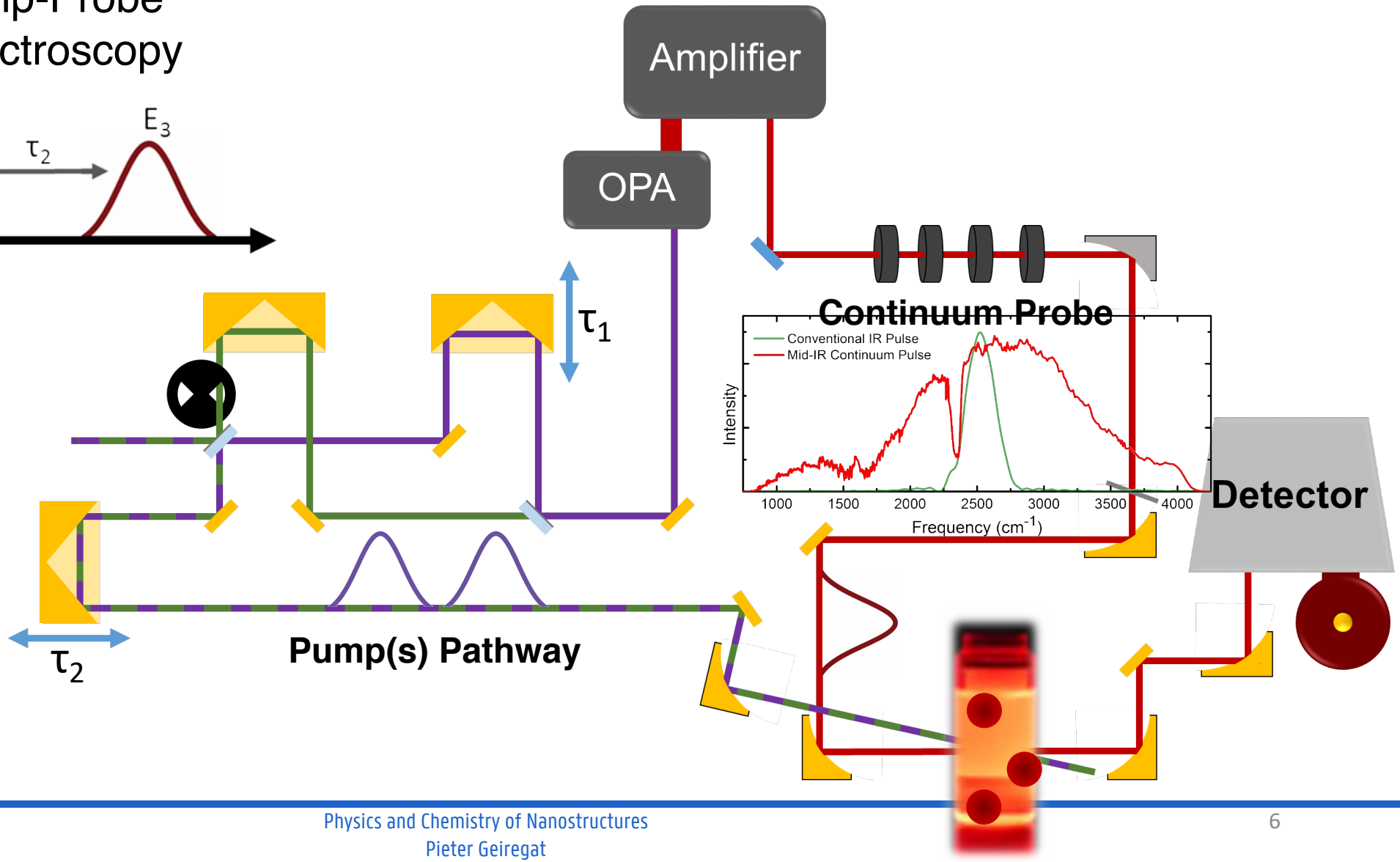
Ultrafast & Broadband Infrared Spectroscopy

$\tau_2 = 0 \rightarrow$ 1D Pump-Probe

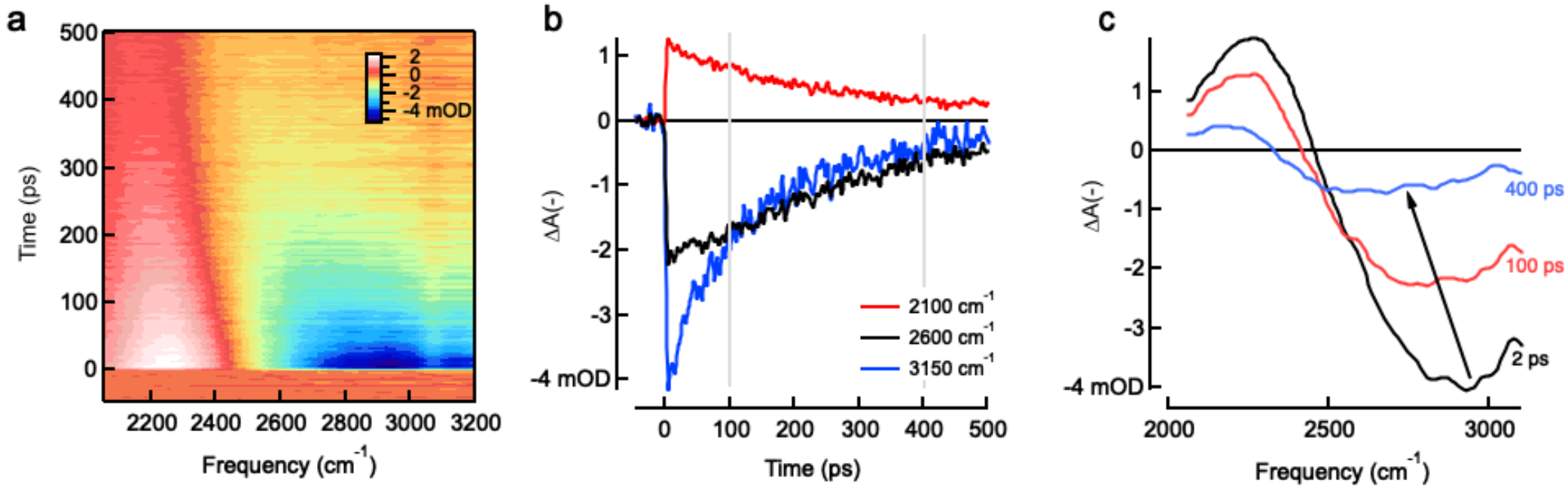
$\tau_2 \neq 0 \rightarrow$ 2D Spectroscopy



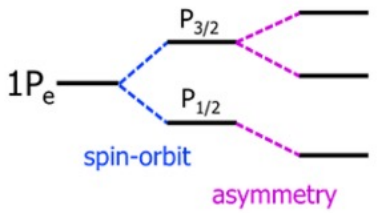




1D Pump-Probe Spectroscopy – Linear Regime ($\langle N \rangle = 0.2$)



Bleach kinetics indicative of P-P relaxation ?

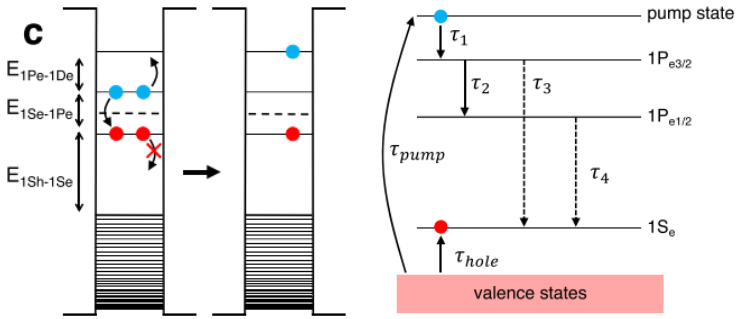


1D Pump-Probe Spectroscopy – Dynamics

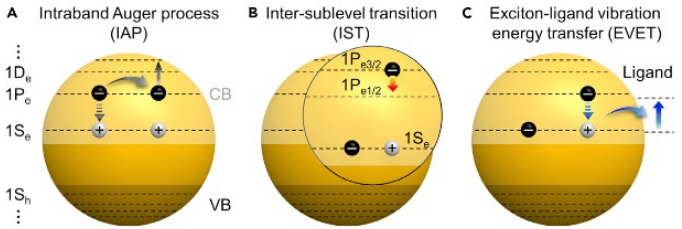
τ_{XX} Auger Recombination

τ_{PP} Intra-P state cooling

τ_X Exciton lifetime



ACS Nano 2019, 13, 9, 10512–10519



Matter, vol.4 (3) 2021, 1072-1086

$\tau_{XX} = \infty$

$\tau_{PP} = 5 - 10$ ps

$\tau_X = 300 - 600$ ps

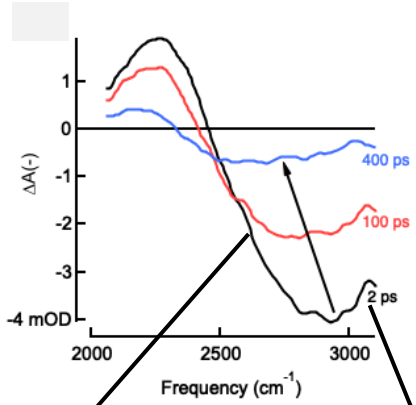
???

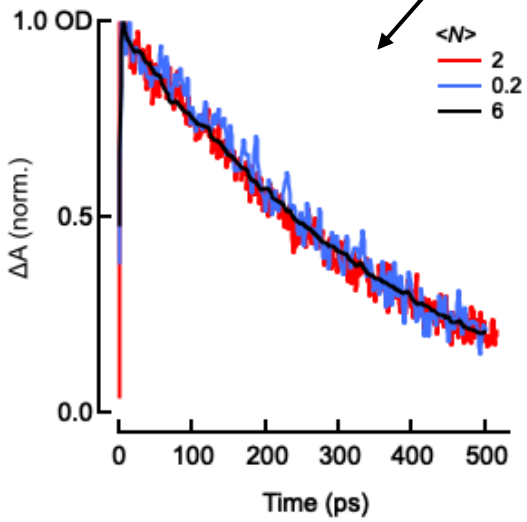
$\tau_{XX} = 1$ ps

$\tau_{PP} = 10 - 60$ ps

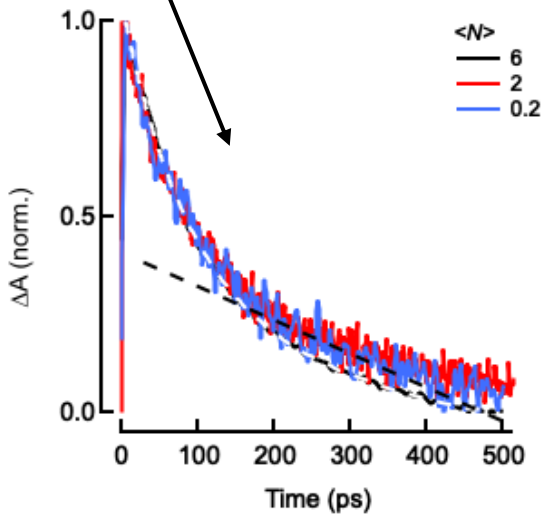
$\tau_X = 300 - 600$ ps

Narrow probing !





$\tau_X = 405$ ps

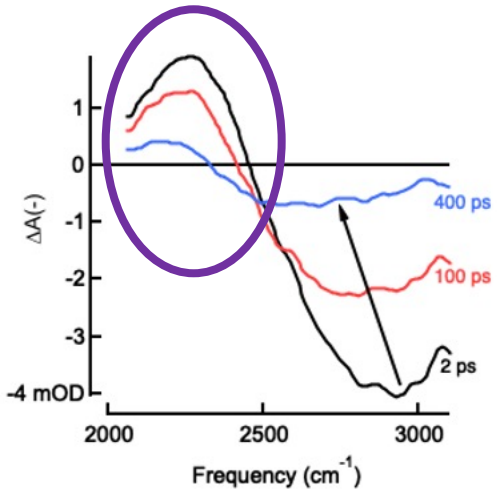


$\tau_X = 400$ ps

$\tau_{PP} = 83$ ps

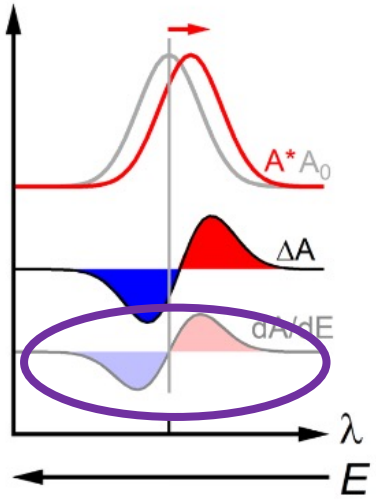
No Signs of Auger !

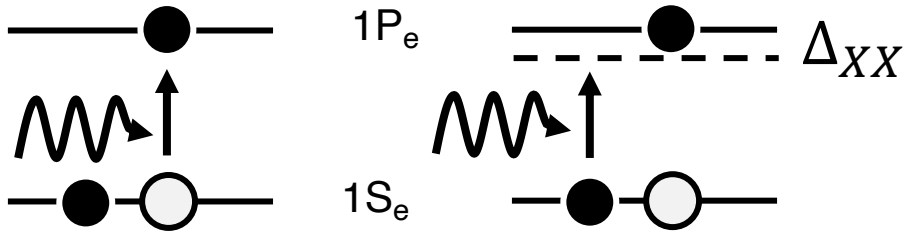
1D Pump-Probe Spectroscopy – Spectral Slices



Frequency Shifts

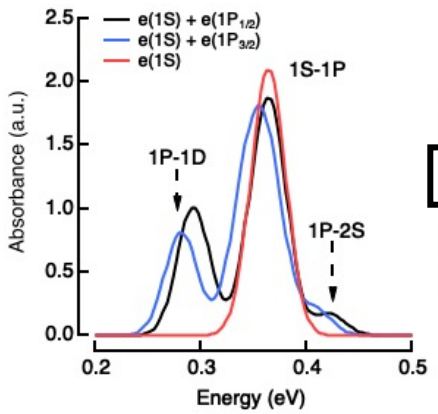
(X-X interactions)

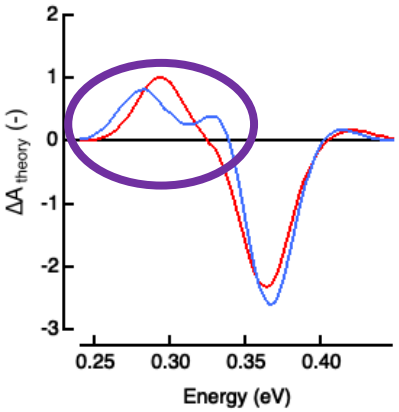




SP – Biexcitons show attractive interactions ?

Photo-Induced Absorption

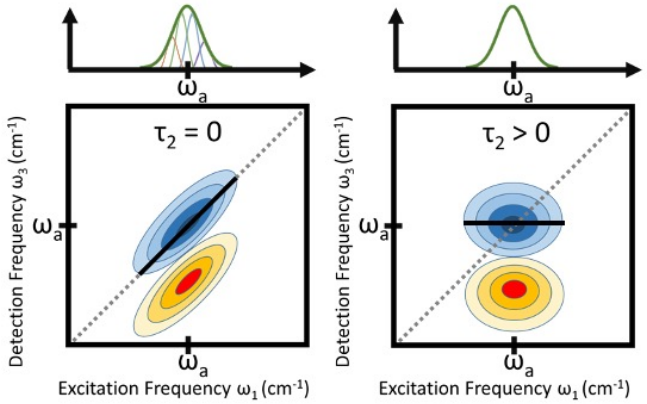




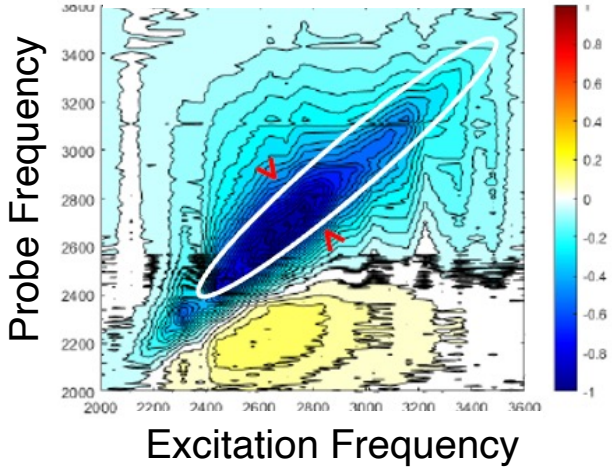
P-D transitions give a similar signature

Two - Dimensional (2D) Spectroscopy

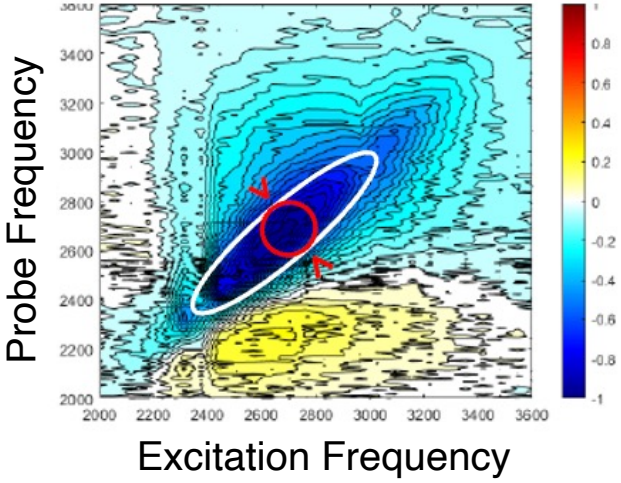
Spectral Diffusion & Broadening



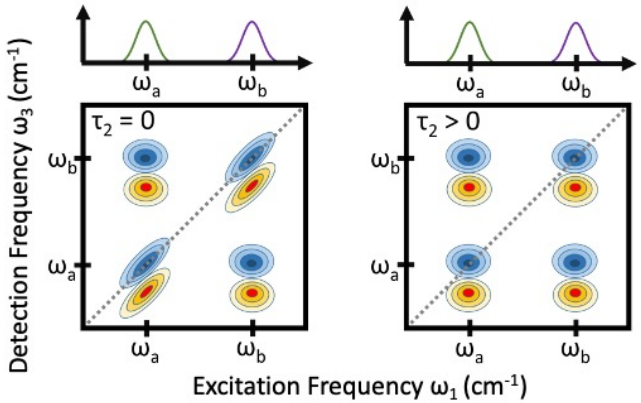
Waiting Time = 150 fs



Waiting Time = 10 ps



Mode Coupling



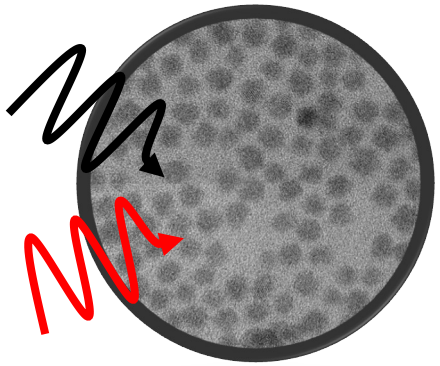
Anti-Diagonal = "**Homogeneous**" Linewidth
⇒ 200 cm^{-1}

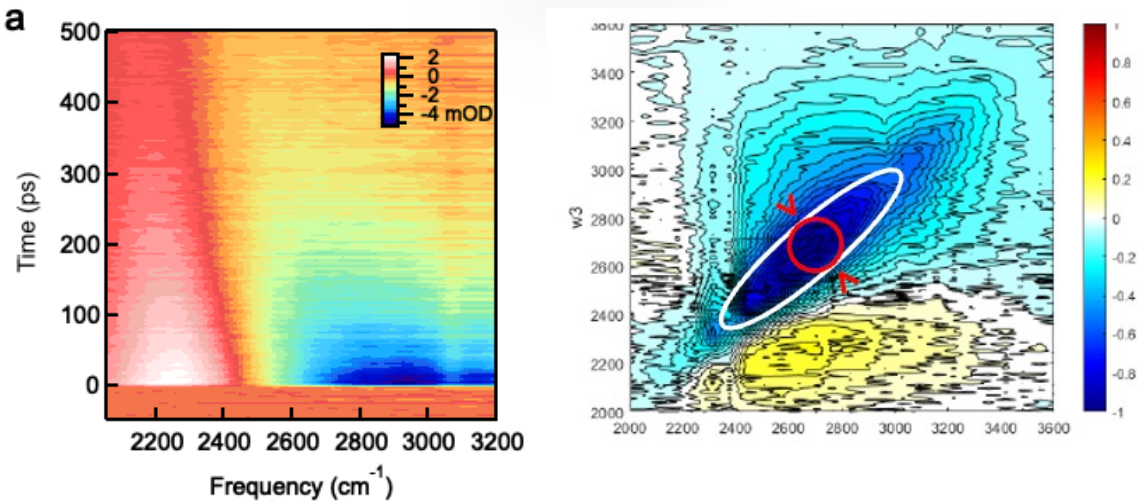
Diagonal = Full Linewidth (Hom. + Inhom.)
⇒ 600 cm^{-1}, dominated by heterogeneity (size, doping, …)
⇒ Static effect !

* Method => J. Chem. Phys. **155**, 104202 (2021)

Outlook

- ✓ Broadband probing reveals absence of Auger Recombination , in-line with fluorescence experiments.
- ✓ Intra-P state relaxation unabmiguosly determined at 80 ps.
- ✓ 2D full spectrum IR reveals linewidths are dominated by inhomogeneous broadening





Acknowledgements

Physics and Chemistry of Nanostructures Group

RUHR UNIVERSITÄT BOCHUM

@Pieter_Geiregat