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Title: Ultrafast dynamics of excitons and spin carriers in 2D semiconductors and 3D Rashba materials

Author(s): Sim, Sangwan

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Ultrafast dynamics of excitons and spin carriers in 2D semiconductors and 3D Rashba materials

Sangwan Sim

Center for Integrated Nanotechnologies
Los Alamos National Laboratory

Contents

- Personal introduction
- Ultrafast pump-probe spectroscopy
- Ultrafast exciton dynamics in ReS_2
  - Coherent exciton dynamics
  - Incoherent exciton dynamics
- Ultrafast spin carrier dynamics in BiTeBr
- Summary

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Personal Introduction

Sangwan Sim

- Director's postdoctoral fellow, Center for Integrated Nanotechnologies,
  Los Alamos National Laboratory (2018.01 – present)
  mentor: Dr. Rohit P. Prasankumar

- Ph.D., Electrical and Electronic Engineering, Yonsei Univ., South Korea (2017)
  advisor: Prof. Hyunyong Choi

Research interests

Ultrafast optical phenomena in 2D semiconductors and topological materials.

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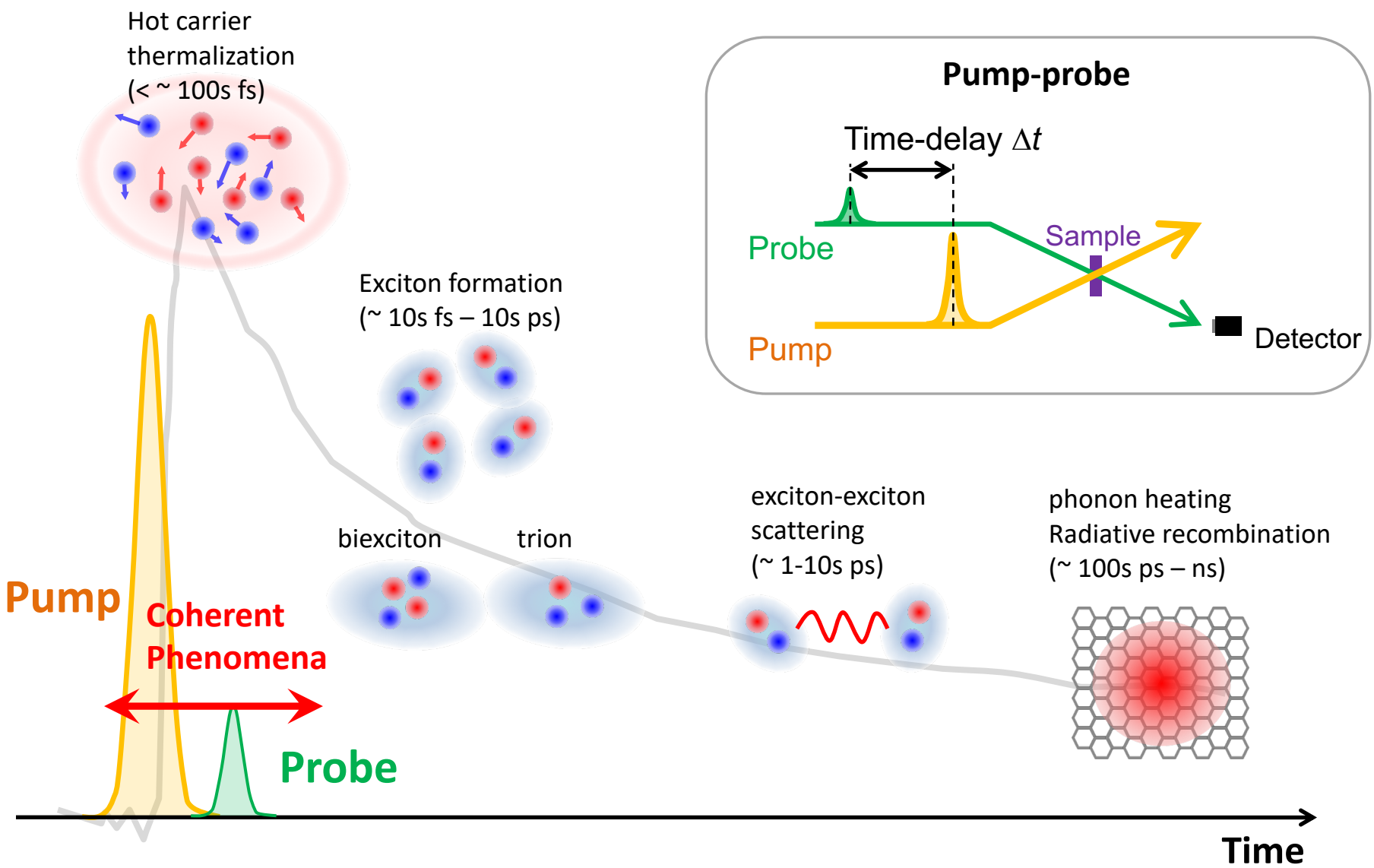
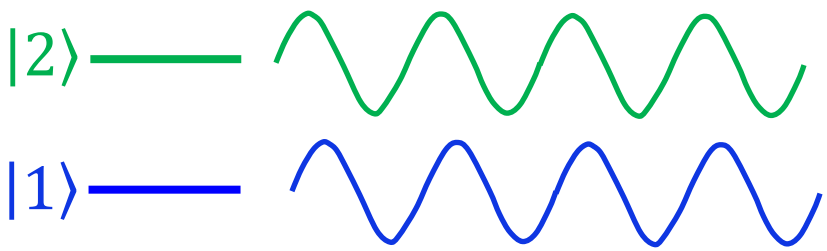
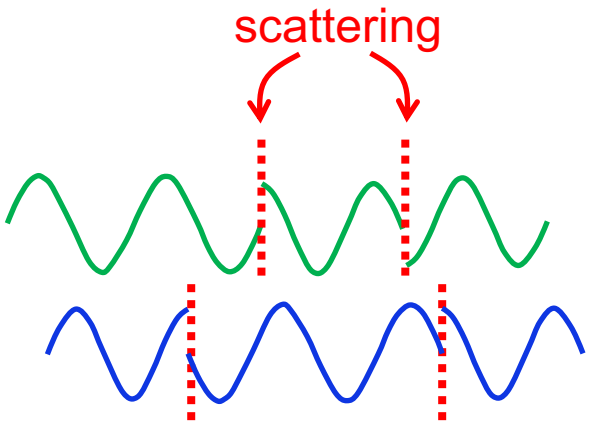
Ultrafast response in semiconductors



Ultrafast response in semiconductors





Coherence: constant phase difference

dephasing

Quantum beats, optical Stark effect, etc.

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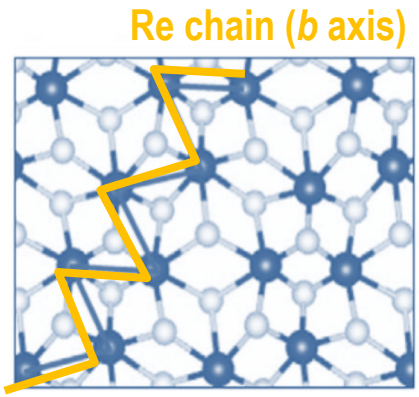
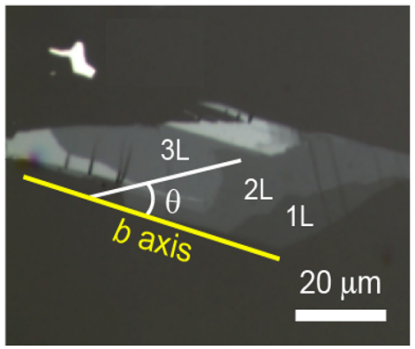
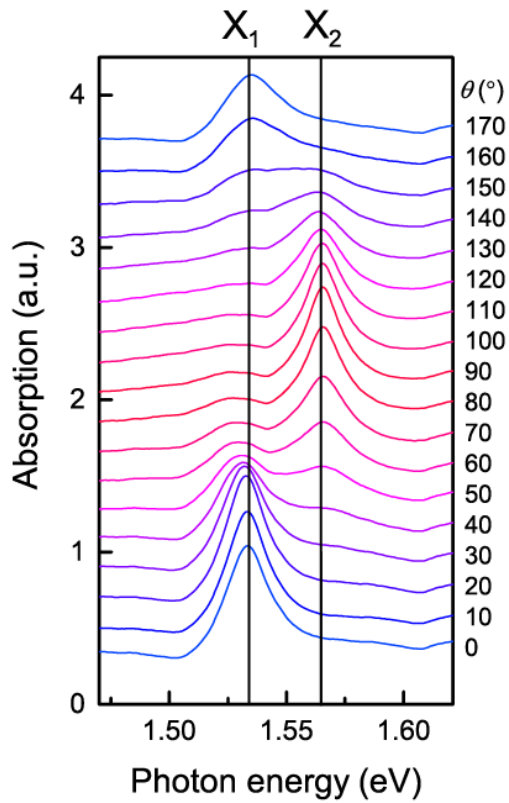
Rhenium disulfide (ReS_2)

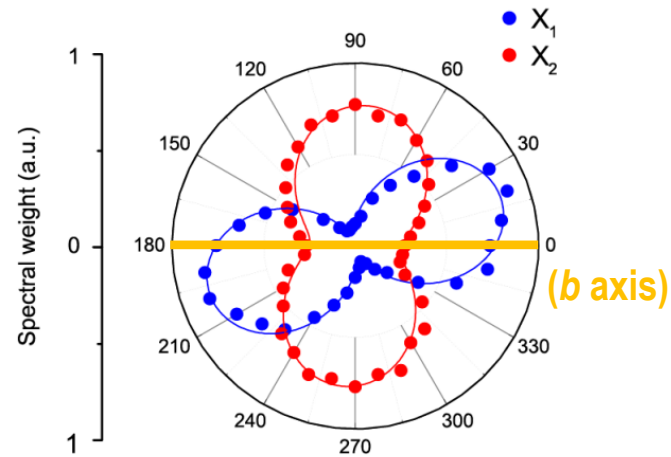




- Layered TMD
- Anisotropic structure
- Direct or indirect?

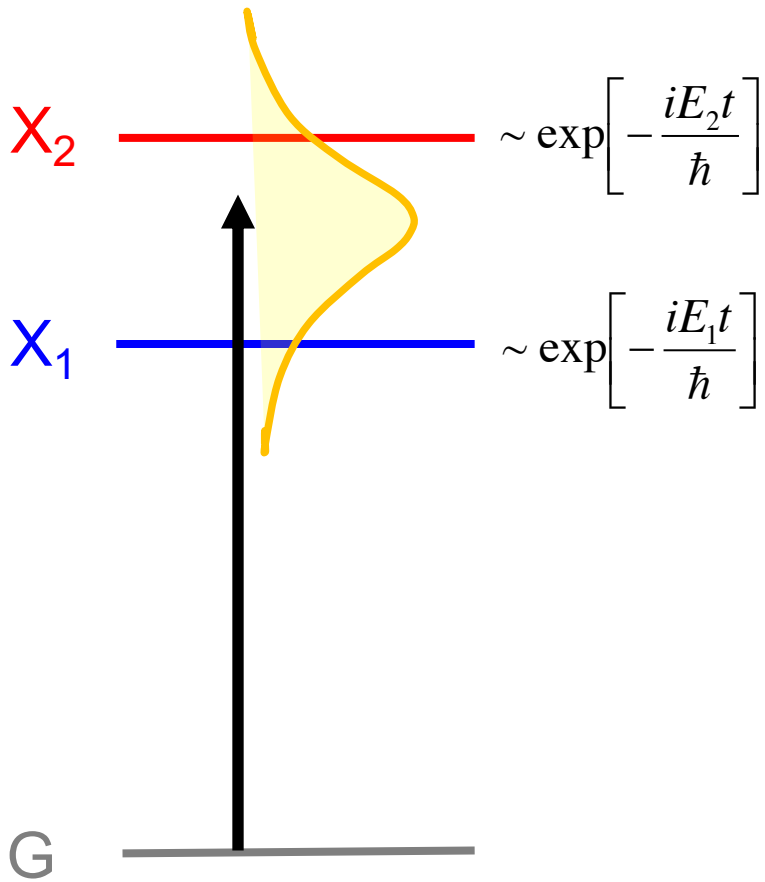


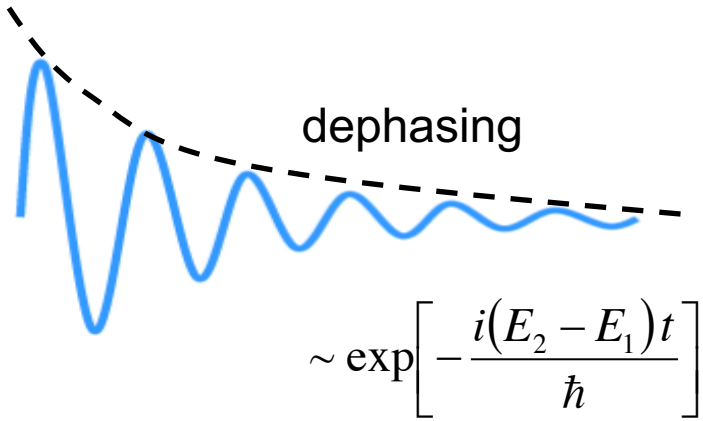
- ***Anisotropic excitons***



S. Sim *et al.* Nature Commun. **7**, 13569 (2016)
S. Tongay *et al.* Nature Commun. **5**, 3252 (2014)
O. Aslan *et al.* ACS Photonics **3**, 96 (2015)

Quantum beats



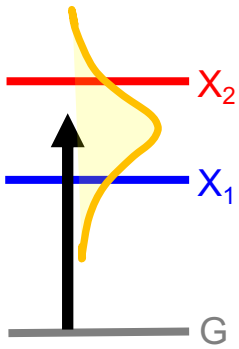


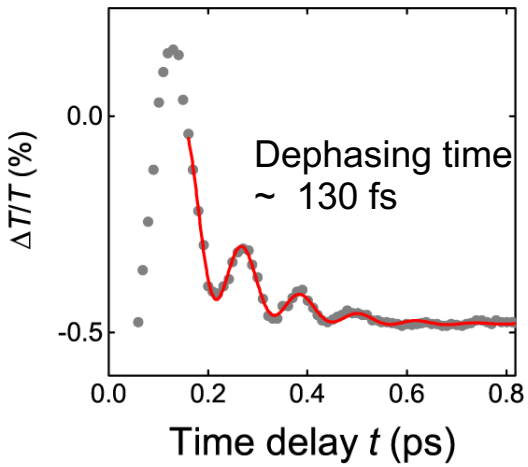
Quantum beats

- THz pulse generation
- Ultrafast optical modulator
- Lasing without inversion
- Quantum information processing
- etc..

J. Shah. Ultrafast spectroscopy of semiconductors and semiconductor nanostructures, Springer (1999)

Quantum beats in ReS_2



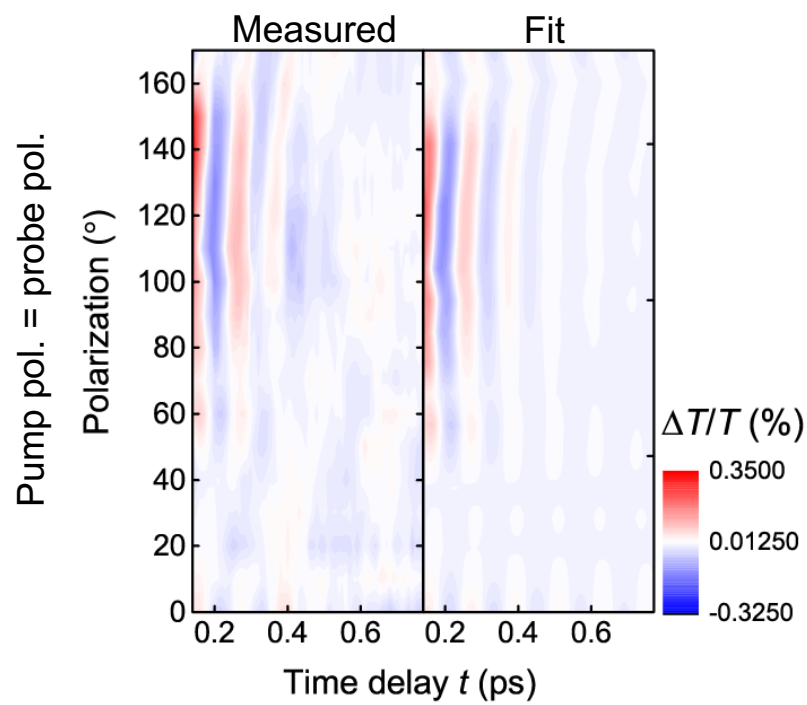


Oscillation period: $T_p = 116$ fs

$$T_p = 2\pi\hbar / \Delta E$$

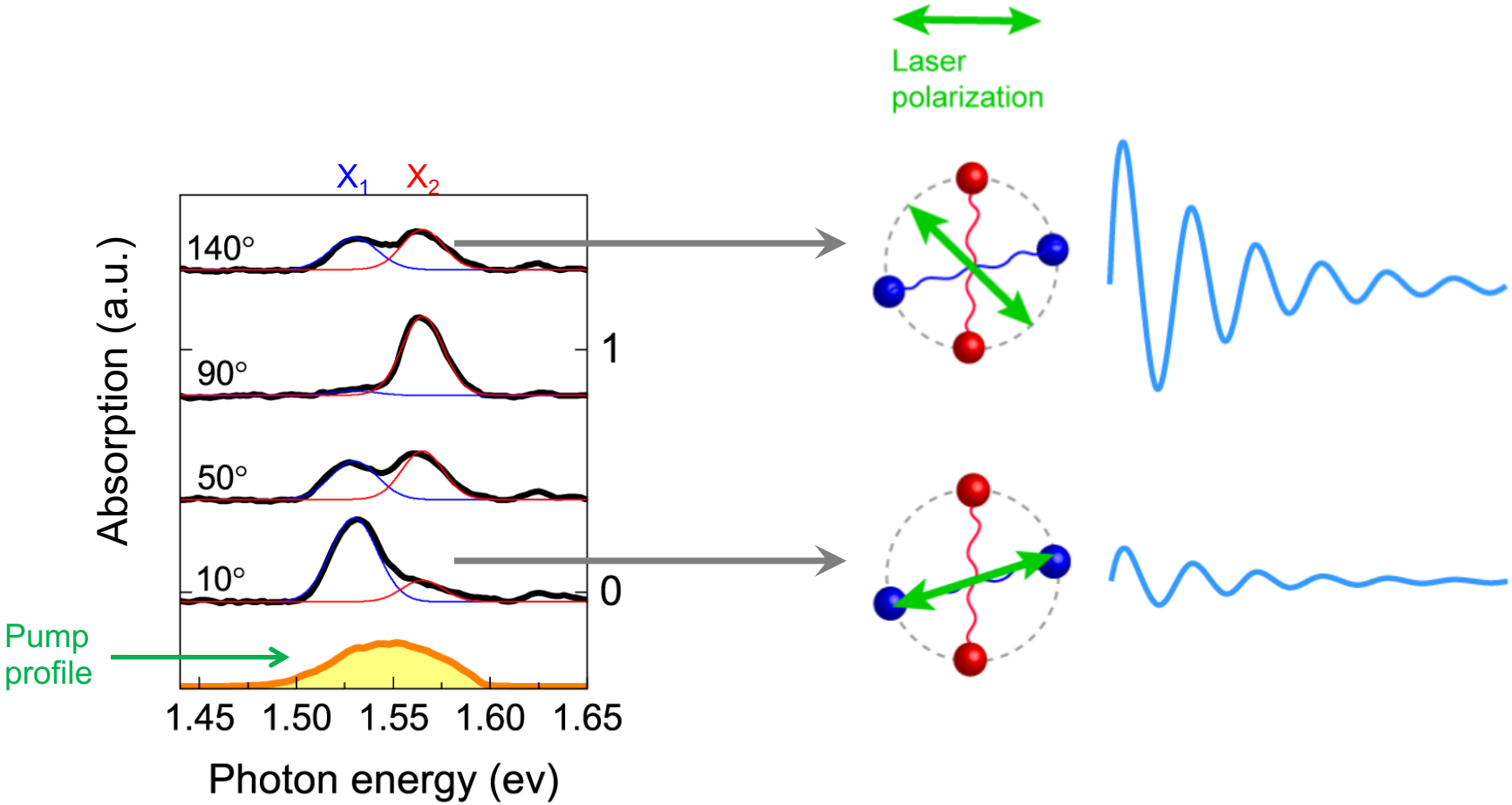
X_1-X_2 energy difference: $\Delta E \sim 35$ meV

Polarization-dependent quantum beats



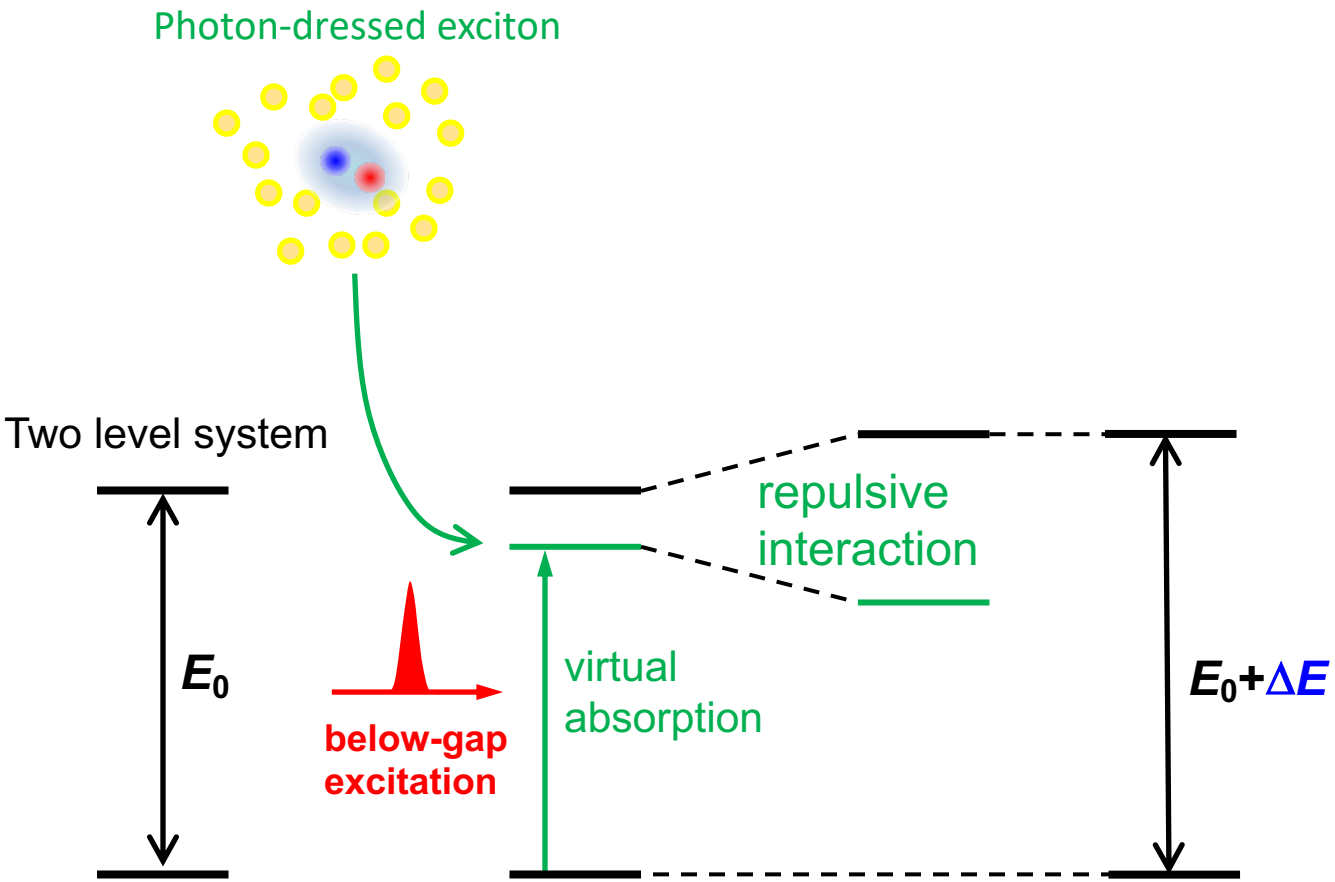
S. Sim *et al. Nature Commun.* (2018)

Polarization dependent quantum beats



S. Sim *et al. Nature Commun.* (2018)

Optical Stark effect



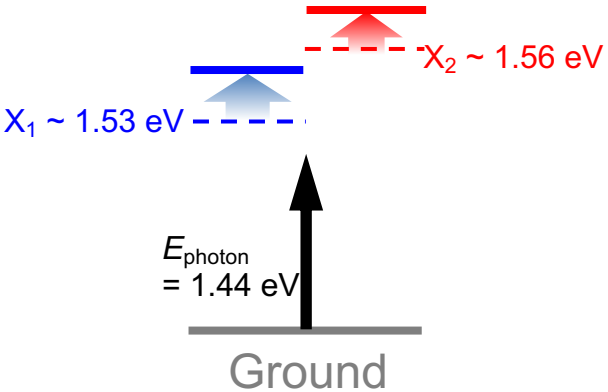
Energy blue-shift

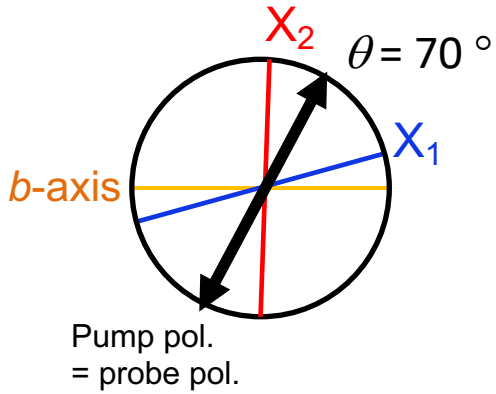
$$\Delta E = \frac{\mu^2 \varepsilon^2}{\delta}$$

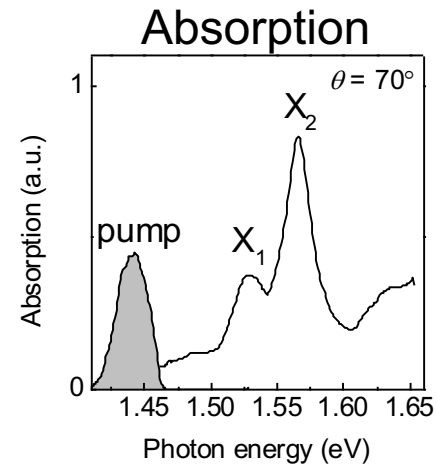
$\mu =$ transition dipole moment
$\varepsilon =$ electric field
$\delta =$ pump energy detuning

Mysyrowicz *et al,* Phys. Rev. Lett. **56**, 2748 (1986).
Sie *et al.* Nature Mat. **14**, 290. (2015).

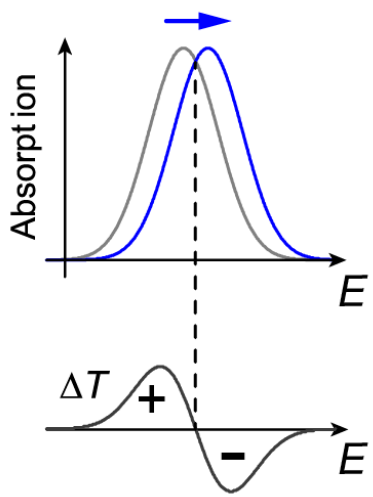
Optical Stark effect in ReS_2

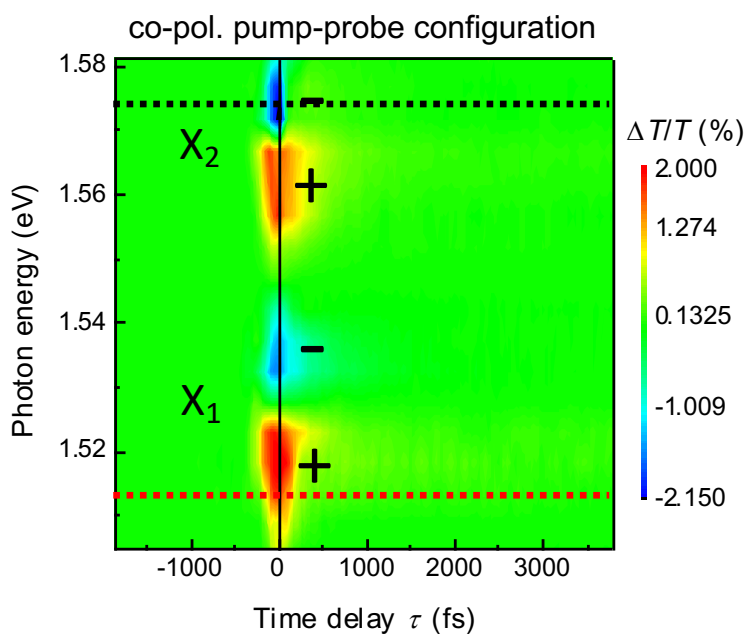




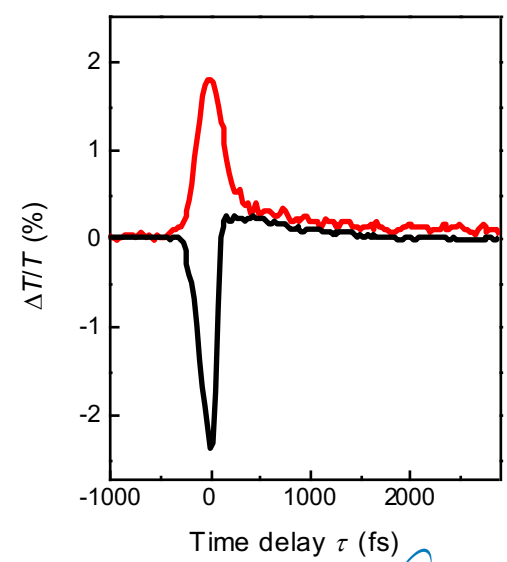


Blue-shift in exciton resonances





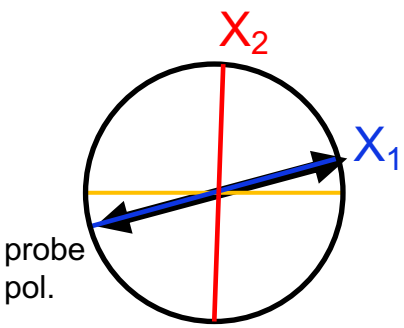
Transient response at zero time-delay

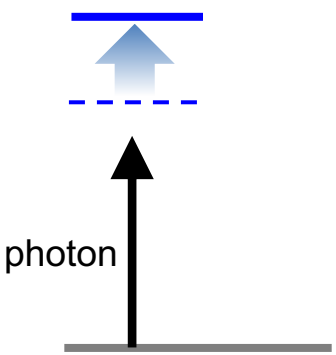


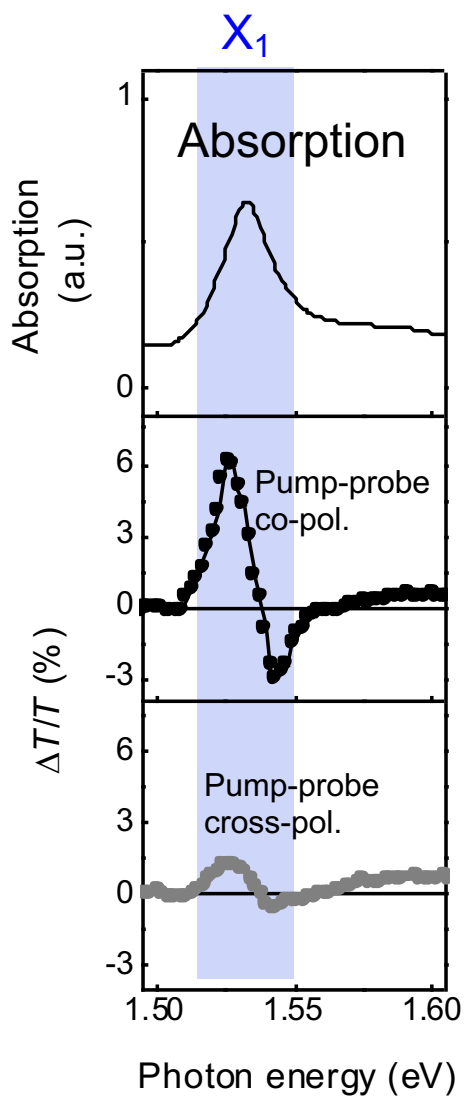
S. Sim *et al. Nature Commun.* (2016)

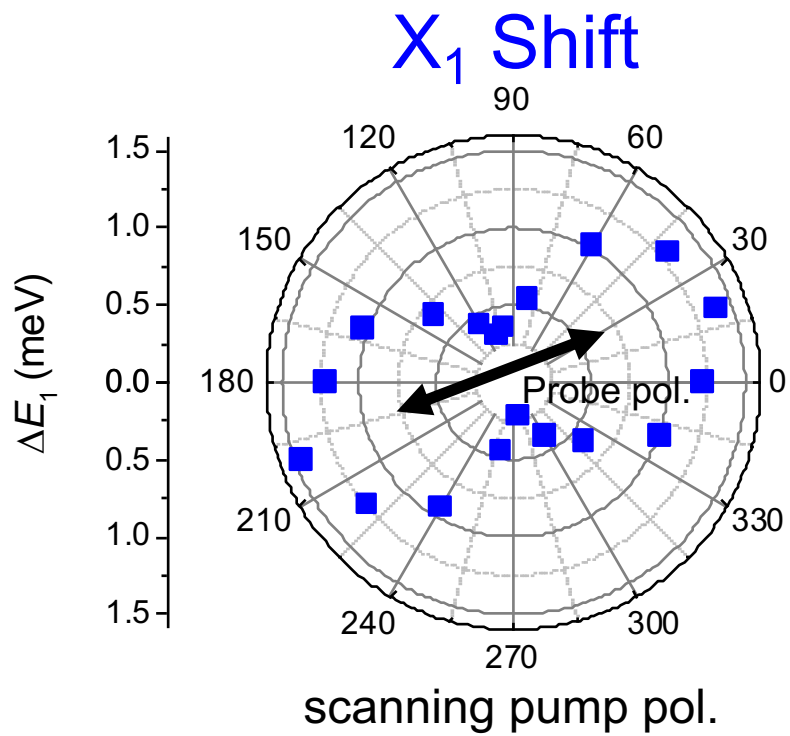
Selective observation of optical Stark shift in X_1

Probe pol. = 20°





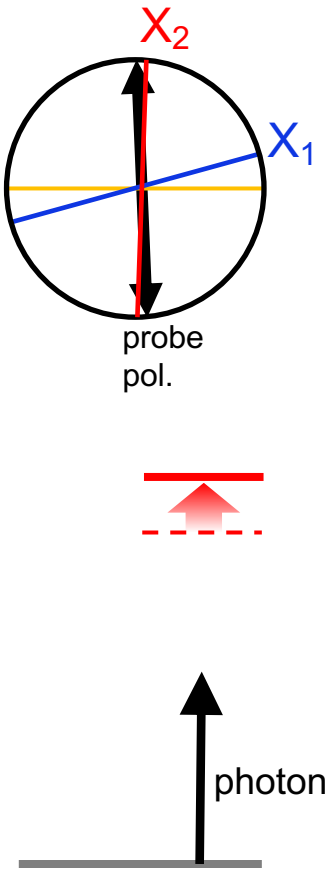


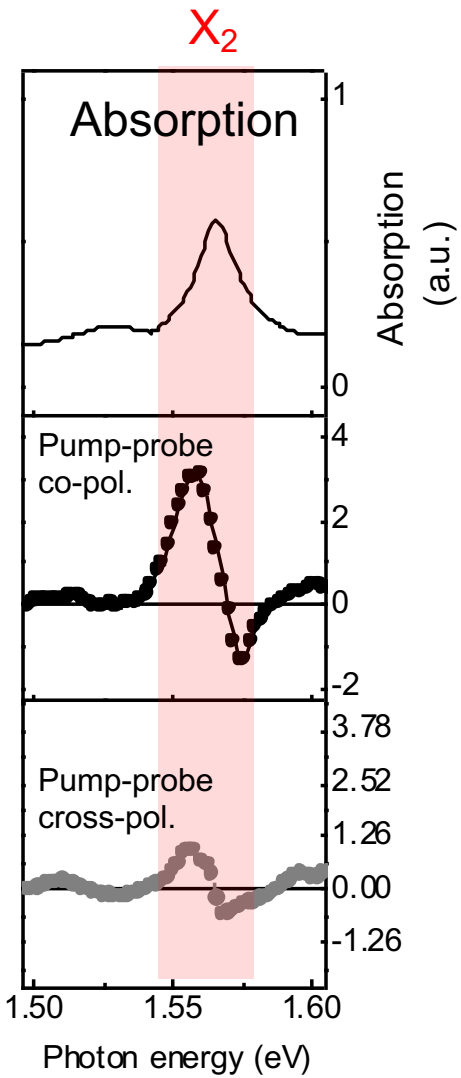


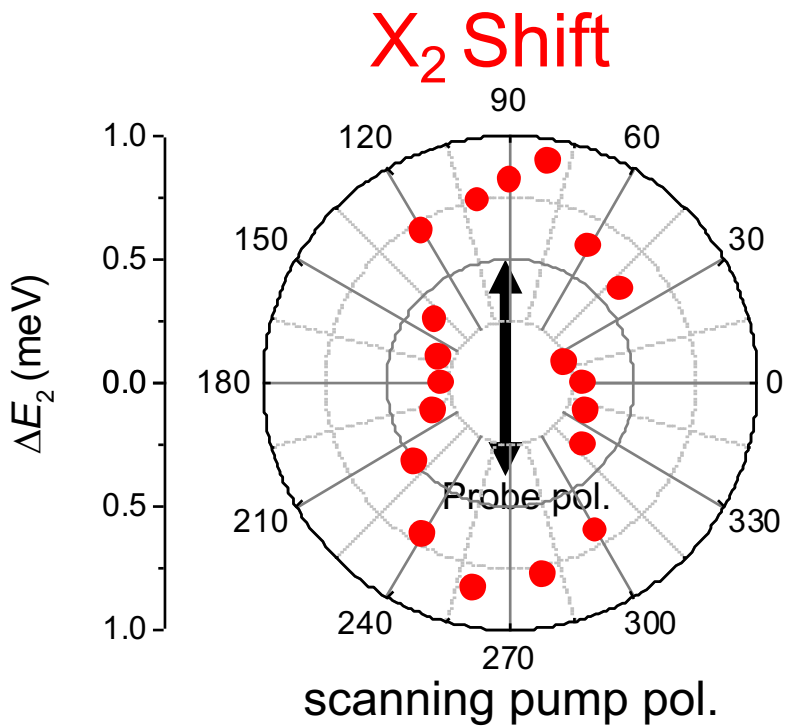
S. Sim *et al. Nature Commun.* (2016)

Selective observation of optical Stark shift in X_2

Probe pol. = 90°

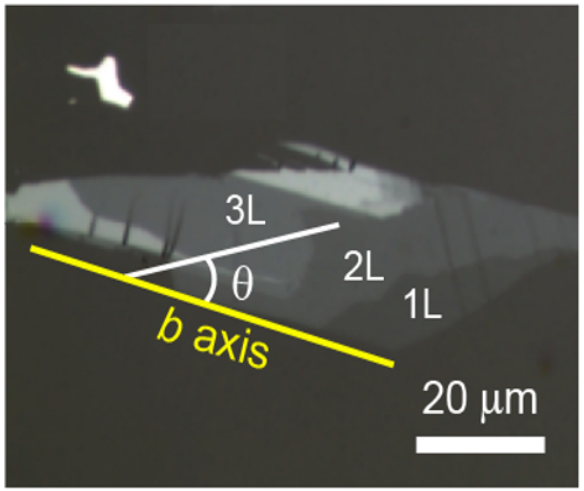


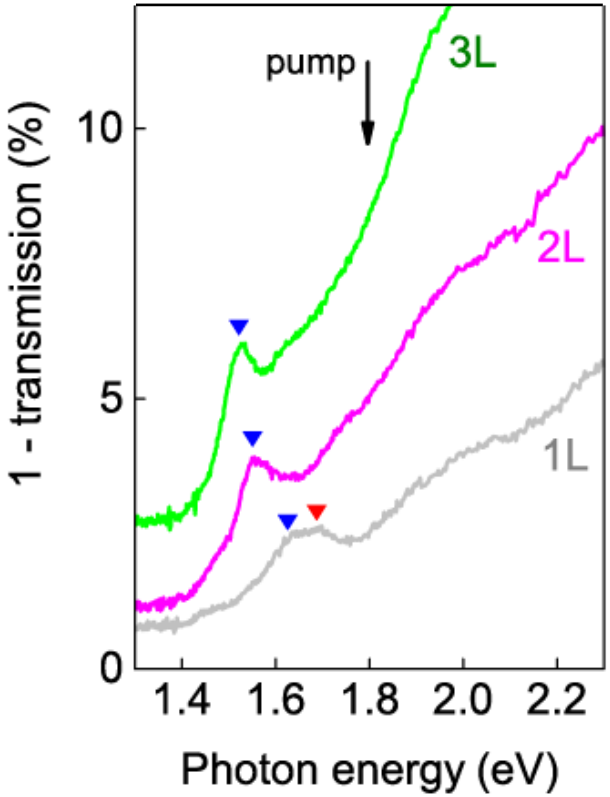




S. Sim *et al. Nature Commun.* (2016)

Thickness dependent exciton dynamics





S. Sim *et al. in preparation* (2018)

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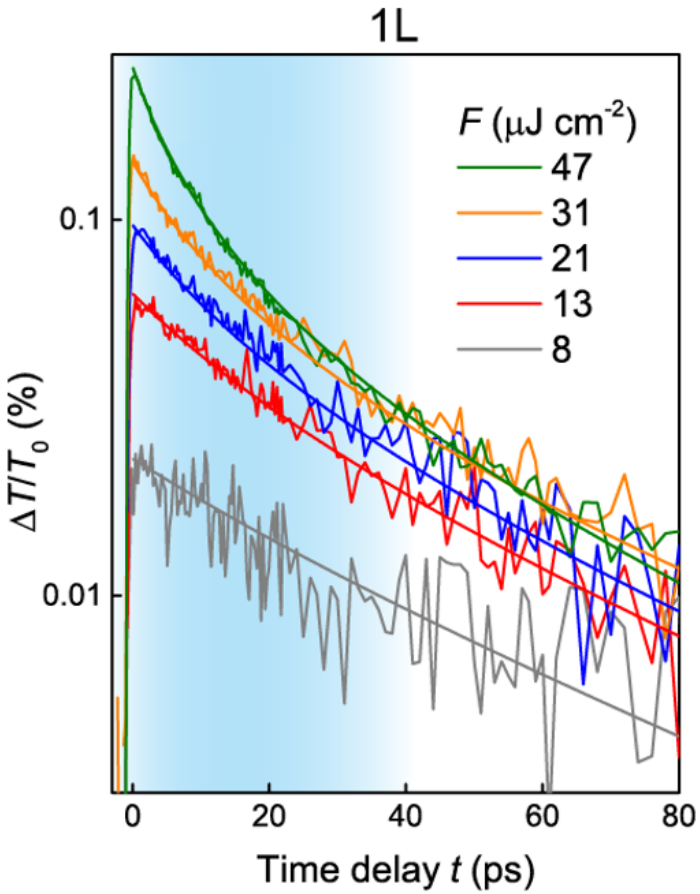
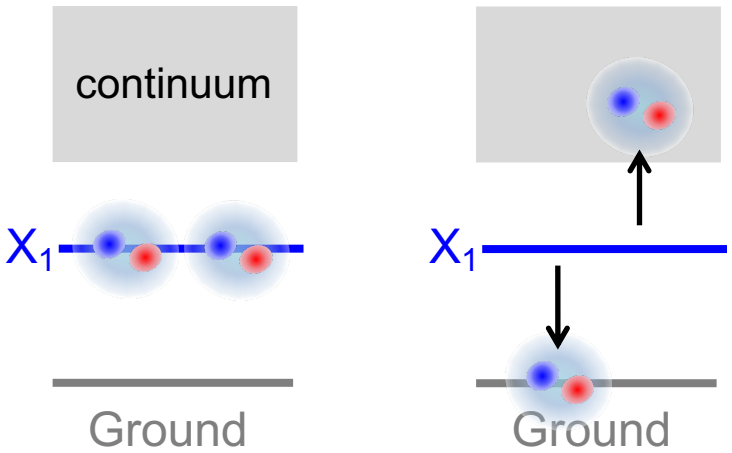
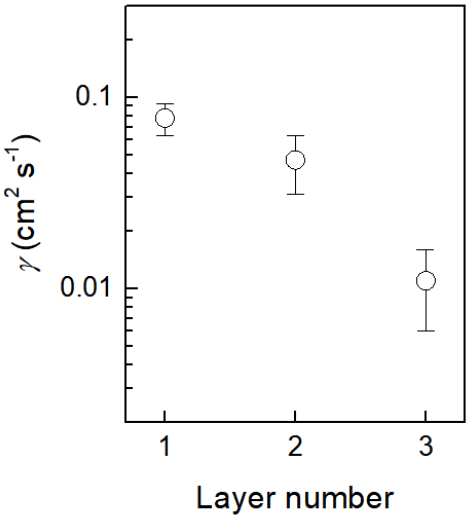
Thickness dependent exciton dynamics



Exciton-exciton annihilation (EEA)



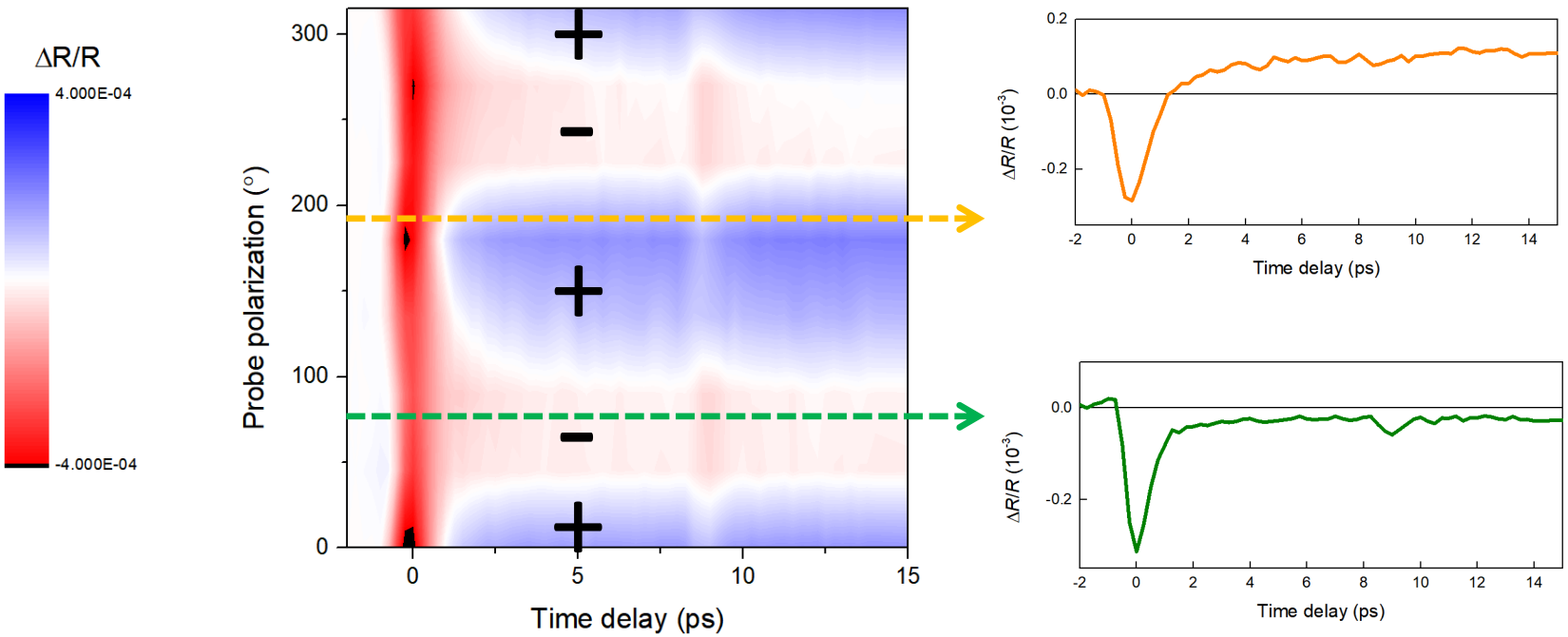


EEA rate↓
as thickness↑

S. Sim *et al. in preparation* (2018)

Polarization dependent exciton dynamics

Pump: 3.1 eV
Probe: 1.5 eV ~ exciton2



G. Bordia *et al. in preparation* (2018)

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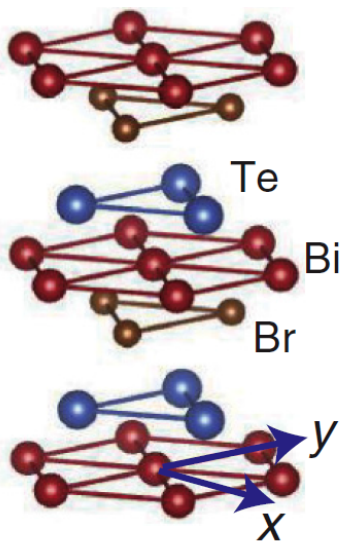
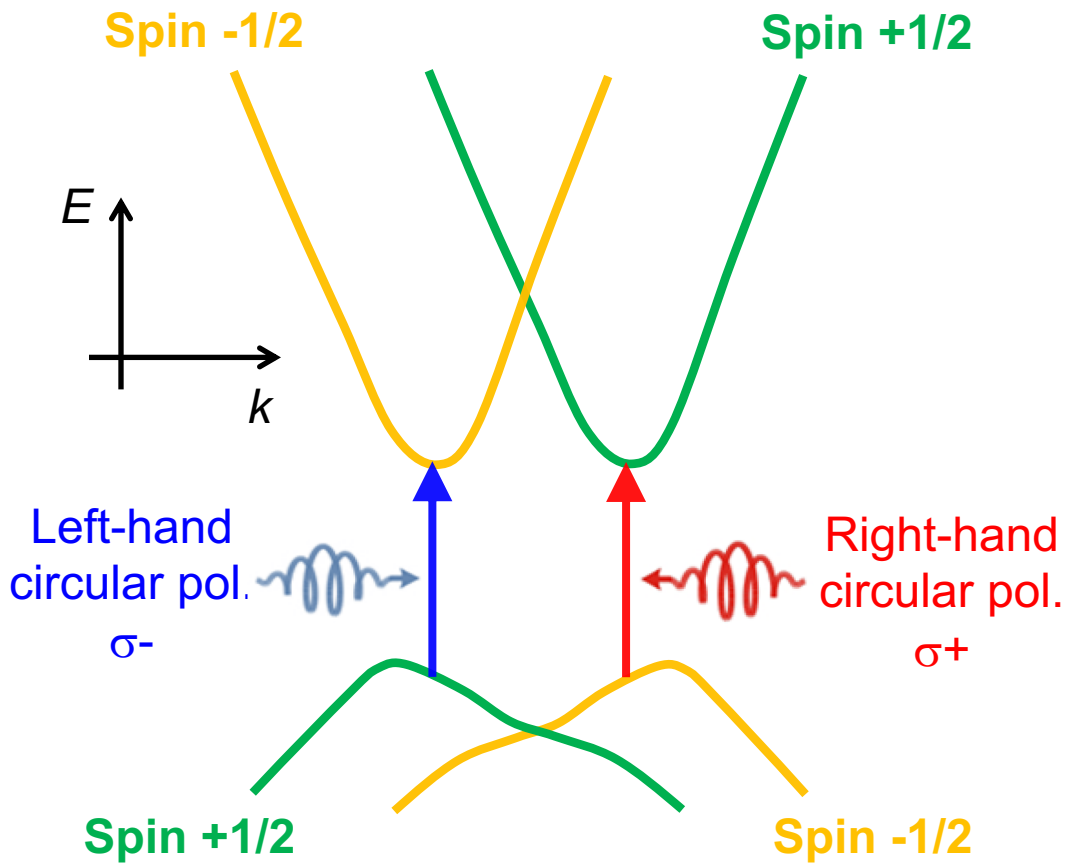
3D Rashba spin splitting material

BiTeBr

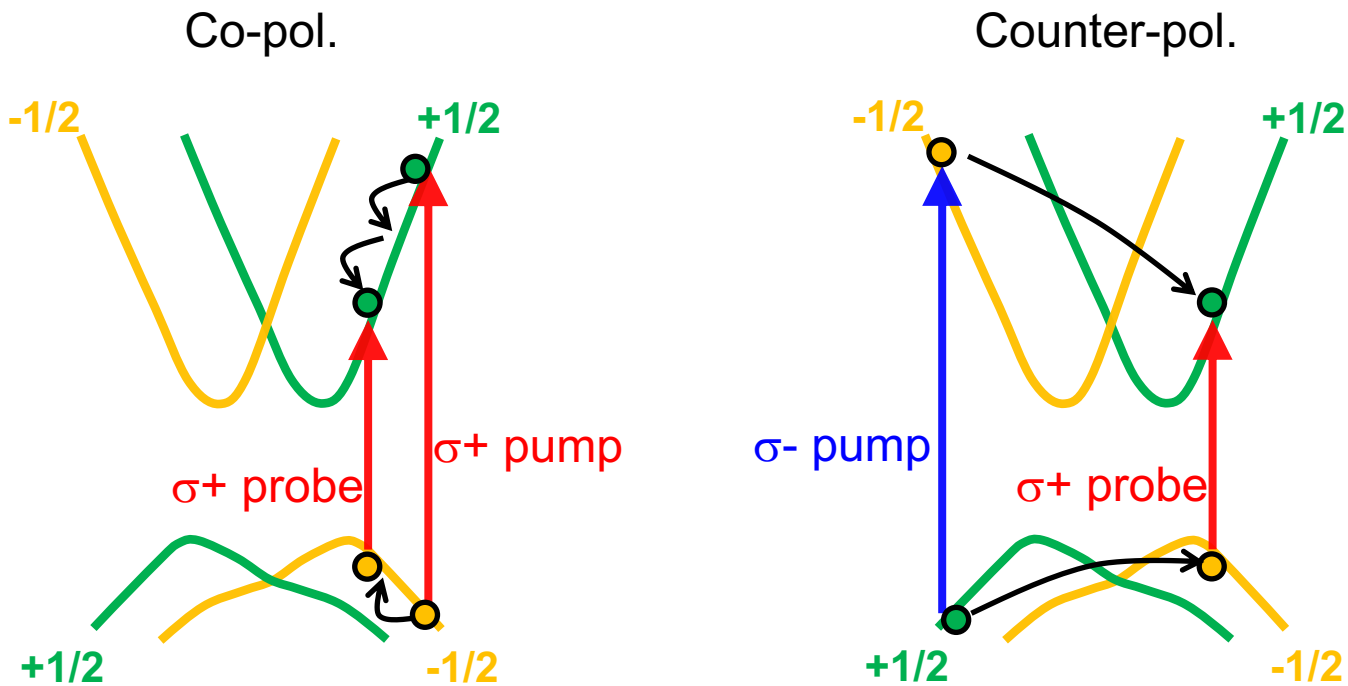


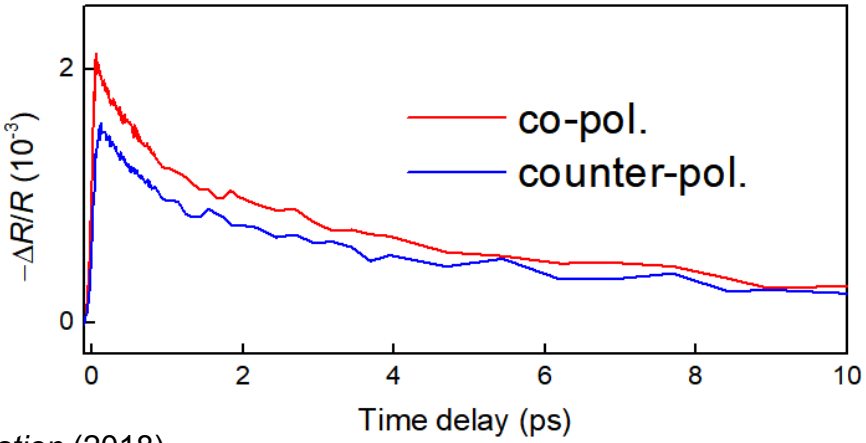


Band gap ~ 0.5 eV

N. Ogawa et al., PRB 90, 125121 (2014)
S. Sim *et al. in preparation* (2018)

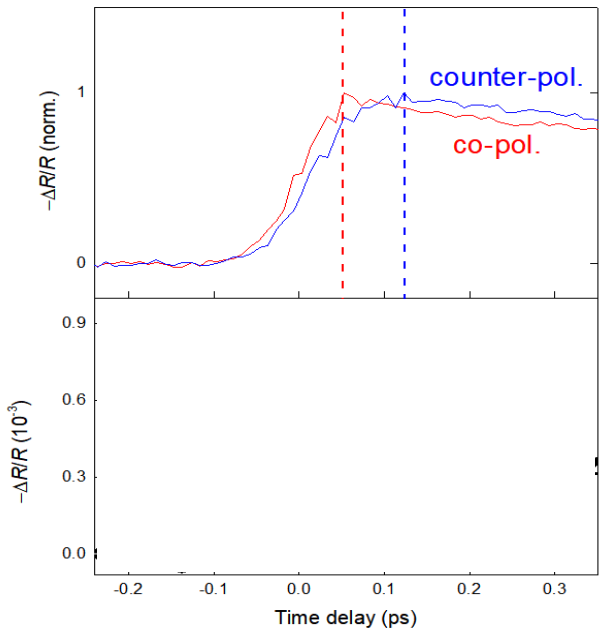
Spin carrier dynamics in BiTeBr

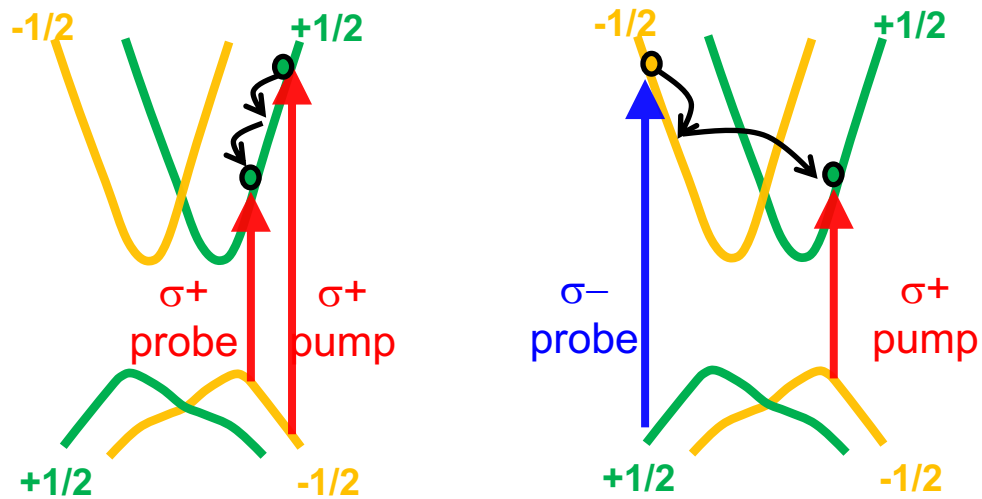




S. Sim *et al. in preparation* (2018)

Spin carrier dynamics in BiTeBr





S. Sim *et al. in preparation* (2018)

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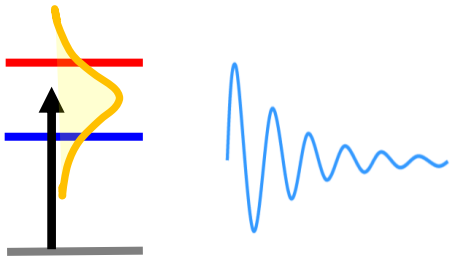
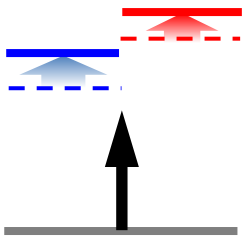
Exciton dynamics in ReS_2

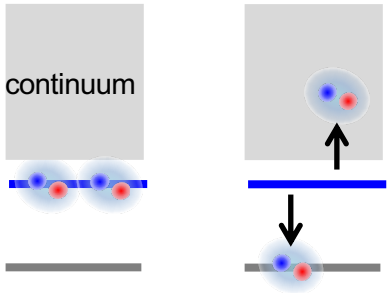
Quantum beats



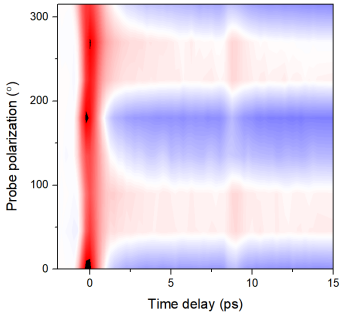
Optical Stark effect



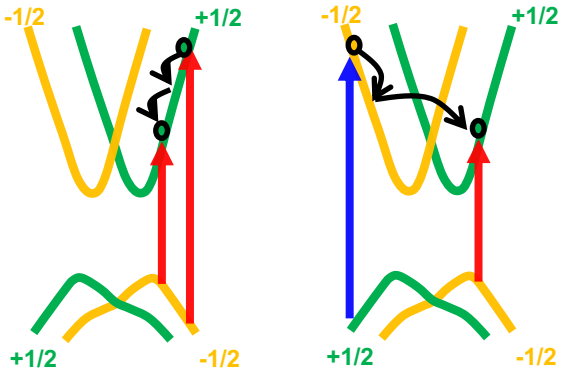
Exciton annihilation

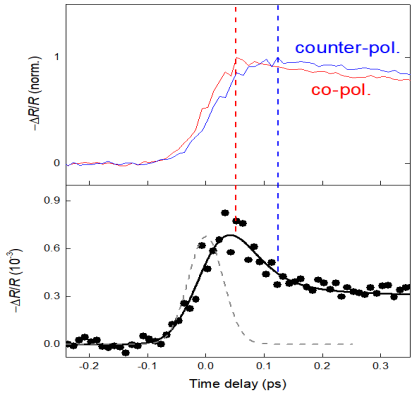


Pol.-dependent decay



Spin carrier dynamics in BiTeBr





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CINT
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Mr. Doeon Lee
Dr. Soonyoung Cha

Cheong's group at Rutgers Univ.
Prof. Sang-Wook Cheong
Mr. Jaewook Kim

Thank you for your attention