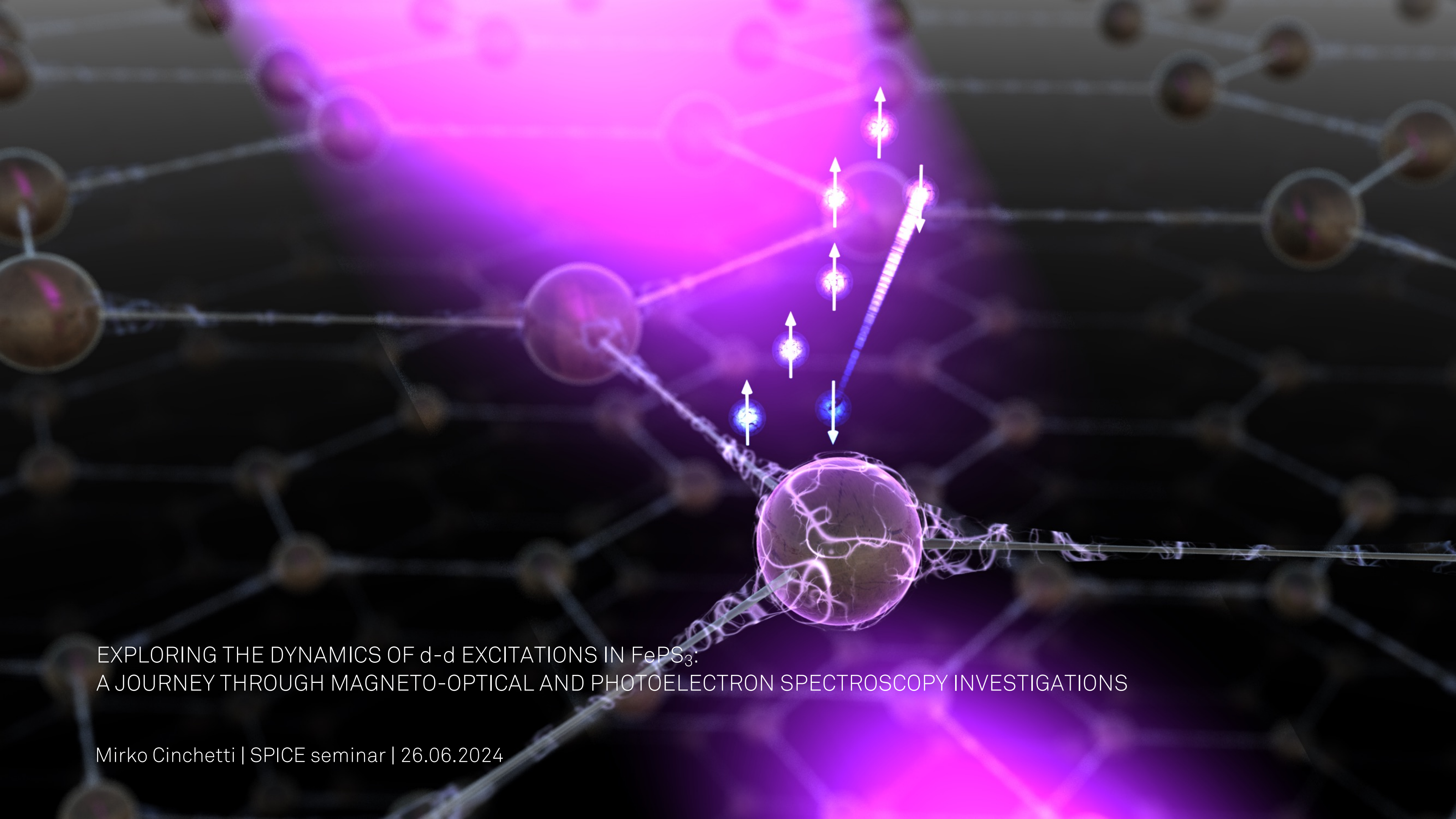
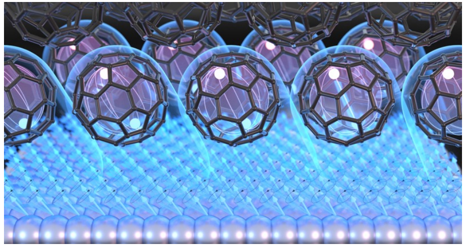


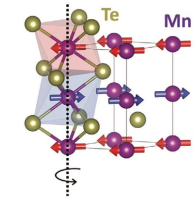
EXPLORING THE DYNAMICS OF d-d EXCITATIONS IN $FePS_3$: A JOURNEY THROUGH MAGNETO-OPTICAL AND PHOTOELECTRON SPECTROSCOPY INVESTIGATIONS

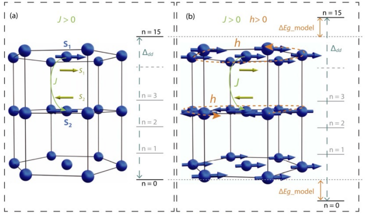
Mirko Cinchetti | SPICE seminar | 26.06.2024

Optical control of quantum materials using elementary excitations

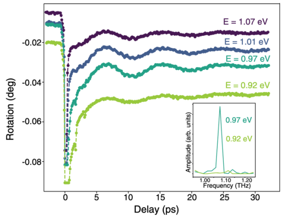


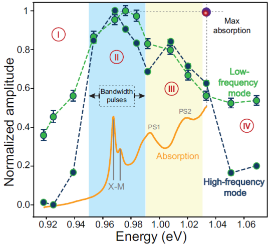
Molecular compounds, proximity effects
Exciton-mediated optical control of GHz spin dynamics
*Advanced Materials Interfaces **10**, 2300236 (2023)*
*Advanced Materials **35**, e2205698 (2023)*





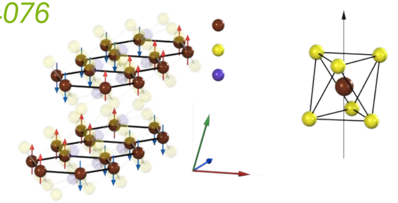
Spin-charge correlations in the altermagnet MnTe generate THz lattice and spin dynamics
*New Journal of Physics **2020**, 22, 083029*
*Physical Review B **2021**, 104, 224424*
*Physical Review Materials **2023**, 7, 054601*
*Advanced Materials **2024**, 2314076*





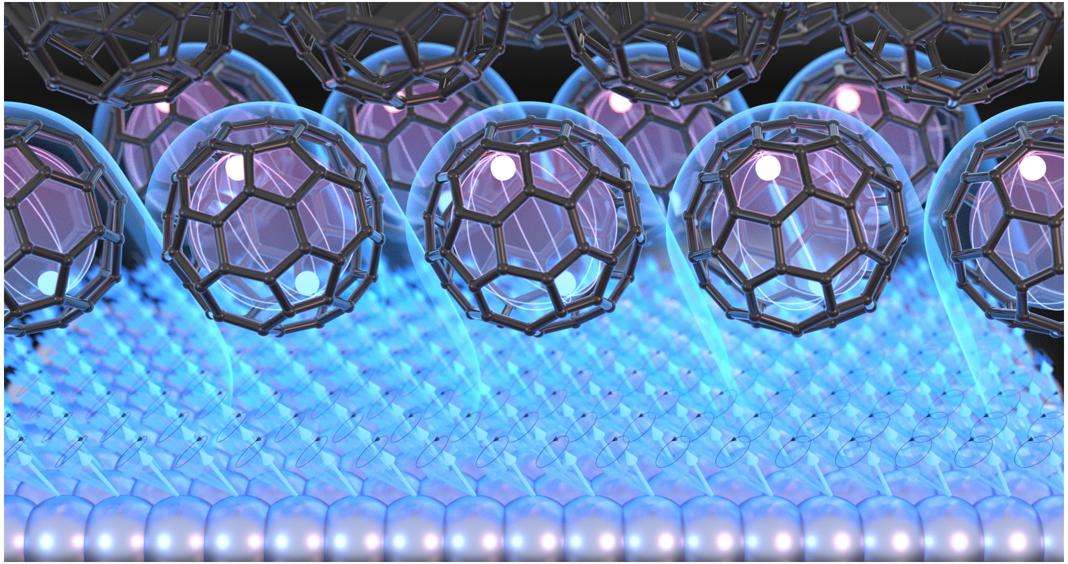
Exciton-magnon coupling in NiO generate THz spin dynamics
Physical Review Letters 127, 077202 (2021)

$TMPS_3$ **CrSBr**



Localized d-d excitations generate THz spin dynamics in $FePS_3$
*Advanced Materials **2023**, 35, 2208355*
*Materials Today Electronics **2023**, 6, 100061*
*Physical Review Materials **2024**, 014408*
*arXiv:2402.03018 **2024***

Molecular compounds, proximity effects



Exciton-mediated optical control of GHz spin dynamics in Co/C60

On arXiv soon!

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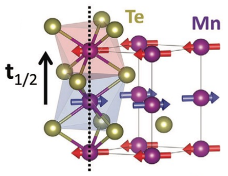
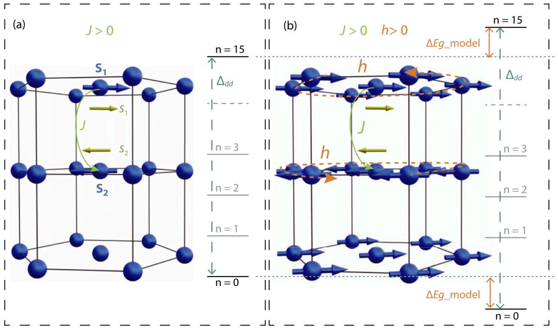
Bulk antiferromagnetic dielectrics: THz spin dynamics

▪ MnTe

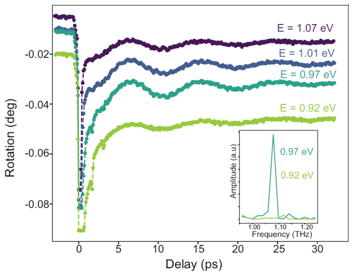


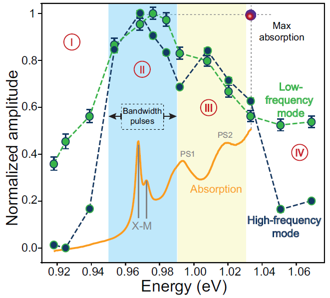


Spin-charge correlations in the altermagnet MnTe generate THz spin dynamics

New Journal of Physics ***2020****, 22, 083029*
Physical Review B ***2021****, 104, 224424*
Physical Review Materials ***2023****, 7, 054601*
Advanced Materials ***2024****, 2314076*

▪ NiO

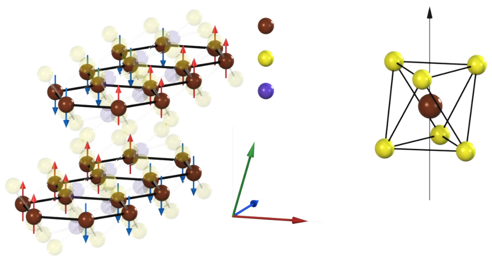


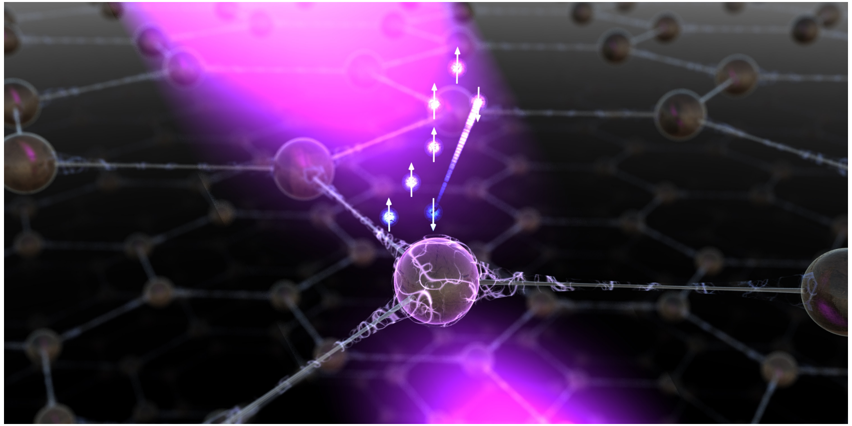


Exciton-magnon coupling in NiO enhance THz and GHz magnon amplitude

Physical Review Letters 127, 077202 (2021)

2D antiferromagnetic semiconductors: $TMPS_3$, CrSBr

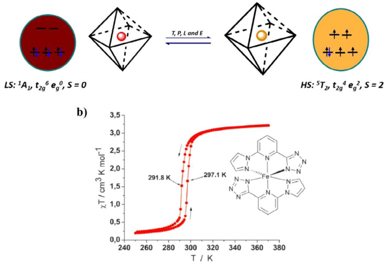




This talk: Localized d-d excitations generate THz spin dynamics in $FePS_3$

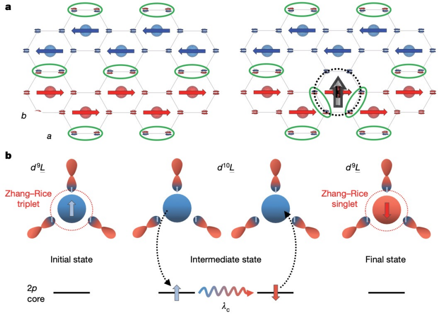
Advanced Materials ***2023****, 35, 2208355*
Materials Today Electronics ***2023****, 6, 100061*
Physical Review Materials ***2024****, 014408*
arXiv:2402.03018 ***2024***

d-d excitations are ubiquitous



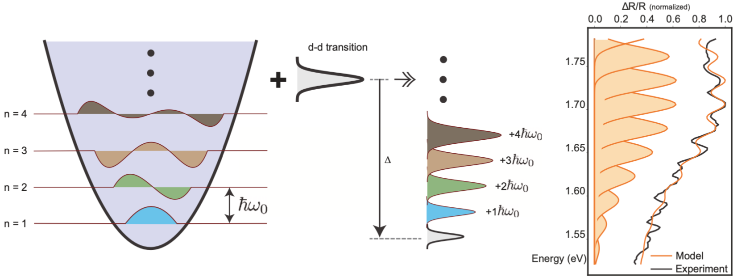
Color of TM-oxides, cathalysis
Spin-crossover complexes

Nature **1960**, *187*, 493
Coordination Chemistry Reviews **2017**, 176, 346



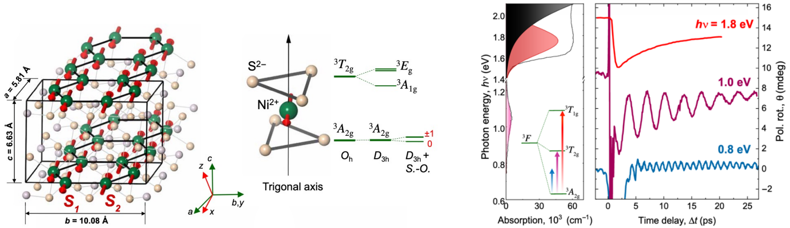
Zhang-Rice singlet/triplet
Superconductivity in Cu-O-based perovskites

Nature ***2020***, *583*, *785*



Electron-phonon bound states in $NiPS_3$

Nat. Commun. ***2022***, *13*, *98*



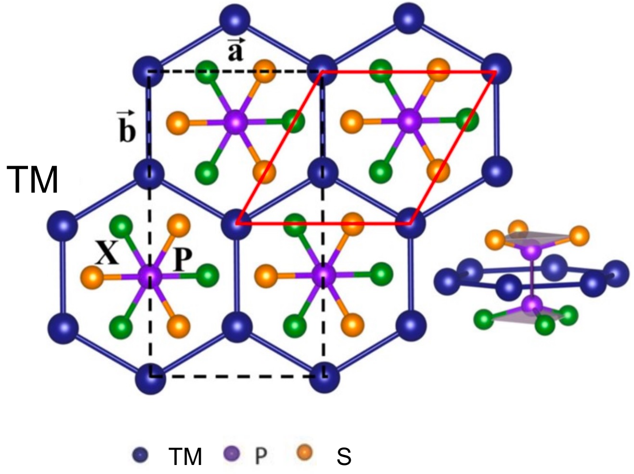
THz magnon generation in $NiPS_3$

Sci. Adv. ***2021***, *7*, *eabf3096.*

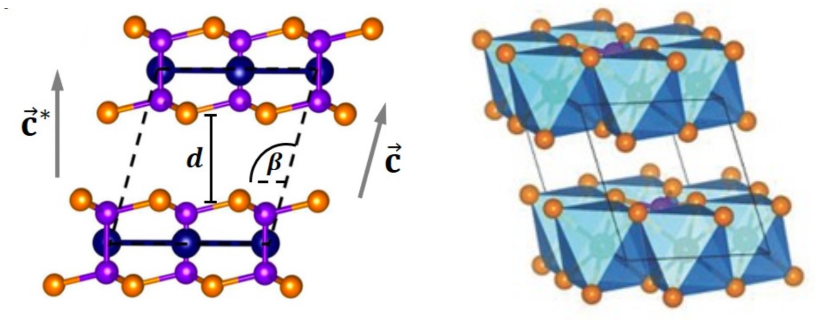
The $TMPS_3$ family: 2D antiferromagnetic semiconductors

Material	$FePS_3$	$NiPS_3$	$CoPS_3$	$MnPS_3$
Bandgap	1.5 eV	1.6 eV	1.4 eV	3.5 eV
d-electrons (TM^{2+})	d^6	d^8	d^7	d^5
T_N	118 K	155 K	132 K	78 K
Intralayer coupling	zz-AFM	zz-AFM	zz-AFM	Neél-AFM
Interlayer coupling	AFM	FM	FM	FM
2D magnetism model	2D Ising	XY-model	XY-model	Heisenberg

The $TMPS_3$ family: crystal structure



Top view



Side view

Materials Research Bulletin 20, 1181 (1985)
Physical Review B 94, 184428 (2016)
Advanced Functional Materials (2018): *https://doi.org/10.1002/adfm.201802151*

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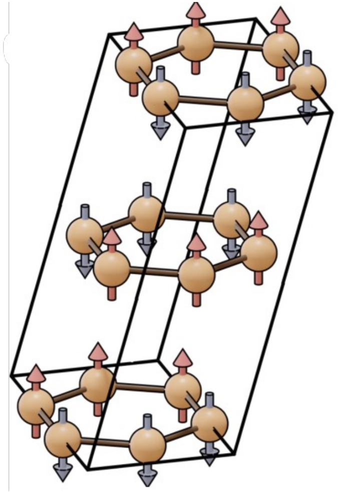
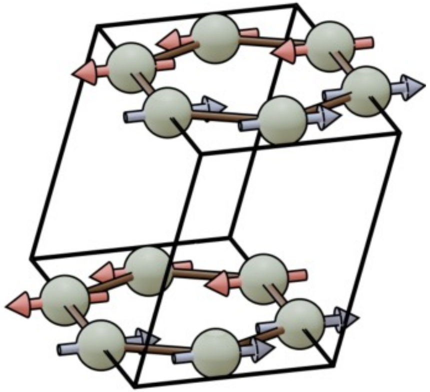
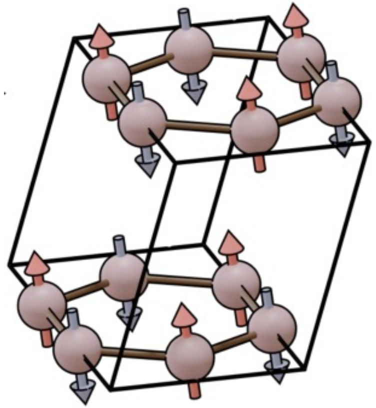
The TMPS$_3$ family: magnetic structure



$FePS_3$
2D Ising

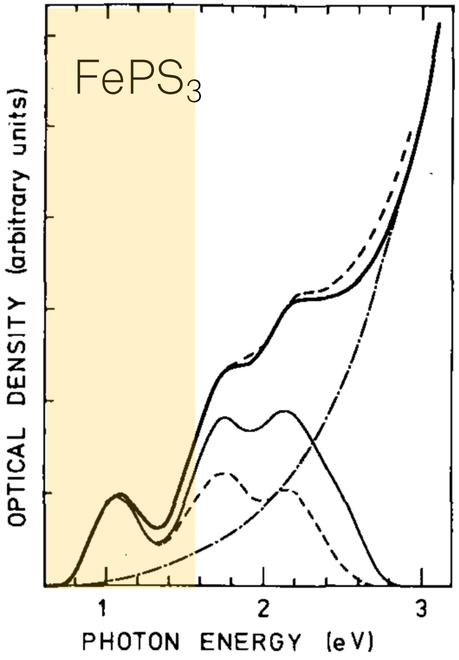


$CoPS_3$/$NiPS_3$
XY



$MnPS_3$
Heisenberg

d-d excitations in the $TMPS_3$ family: optical absorption spectroscopy



Transition	Energy (eV)
$^5T_{2g} \rightarrow {}^5E_g$ (D)	1.08
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	1.78
$^5T_{2g} \rightarrow {}^3T_{2g}$ (H)	2.14
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	2.46
$^5T_{2g} \rightarrow {}^3T_{2g}$ (F)	2.58
$^5T_{2g} \rightarrow {}^3T_{1g}$ (P)	2.84

Piacentini, et al., *Chemical Physics* **72**, 61-71 (1982)

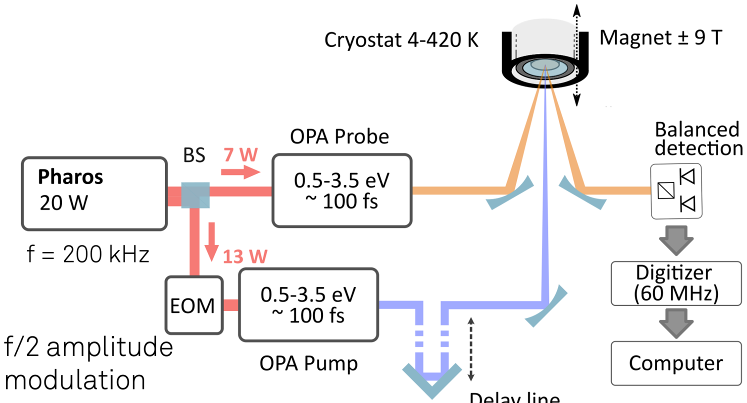
Open questions: Nature? Excitation? Lifetime?
Decay mechanisms?
Relaxation pathways?

Part 1

THE METHODS

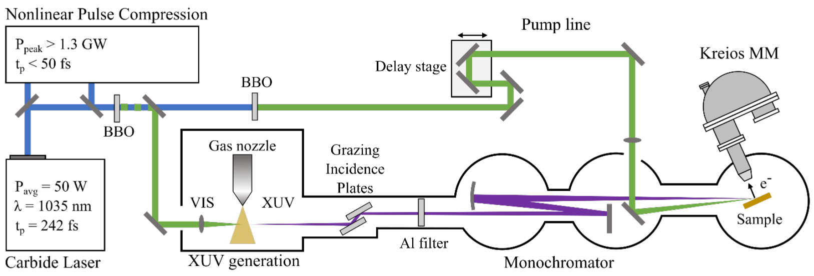
Complementary experimental methods

- Time-resolved magneto-optical spectroscopy



Review of Scientific Instruments **2020**, 2, 113001

- Time-resolved ARPES



*Advanced Physics Research **2023**, 2, 2200016*
https://doi.org/10.1364/opticaopen.26124829.v1

Complementary experimental methods

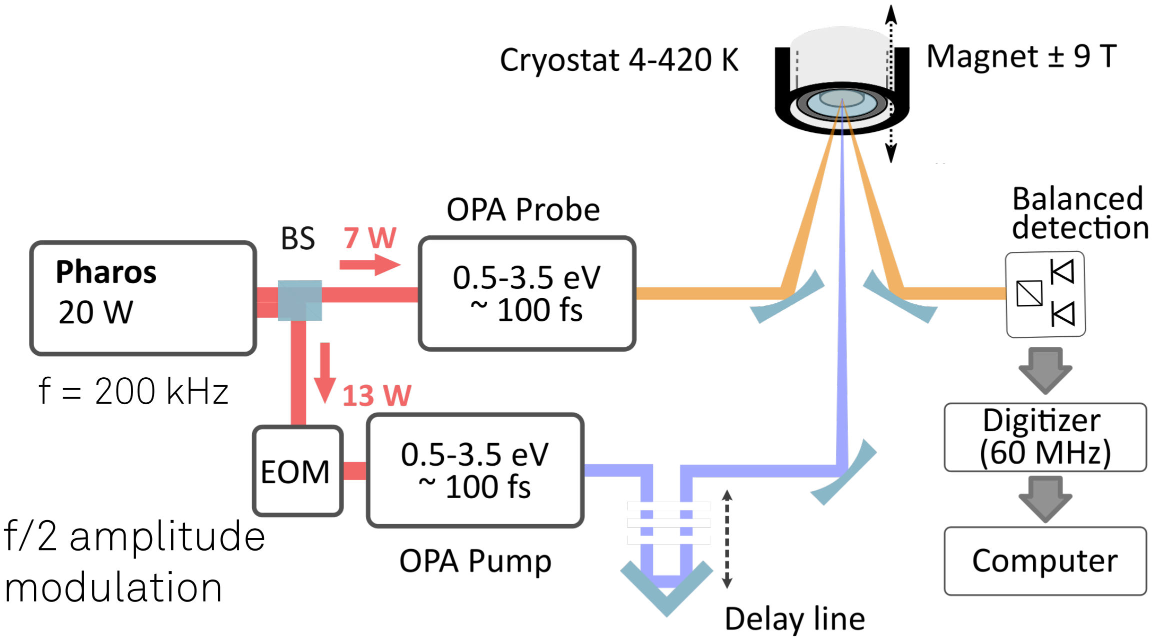
- Time-resolved magneto-optical spectroscopy
- Time-resolved ARPES

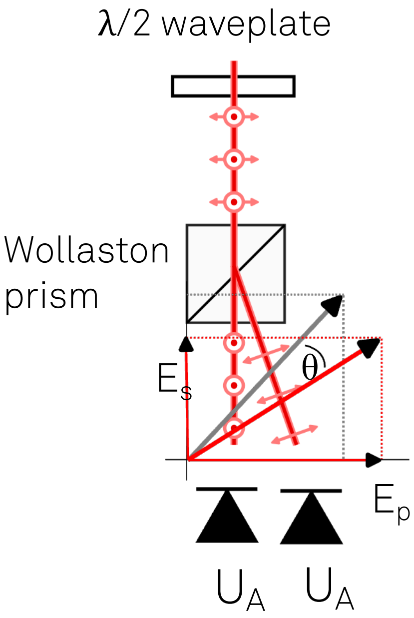
Degrees of freedom

Electronic
Lattice
Magnetic

Spectral function
Quasiparticles
(Excitons, d-d transitions)

Time-resolved magneto-optical spectroscopy



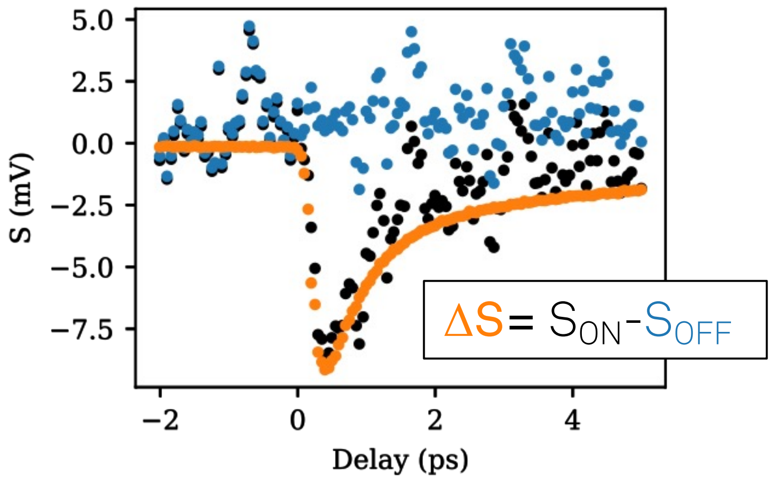


Reflectivity R ~ $U_A + U_B$

Rotation $\theta \sim U_A - U_B$

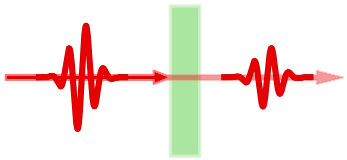
*Review of Scientific Instruments **2020**, 2, 113001*

Time-resolved magneto-optical spectroscopy

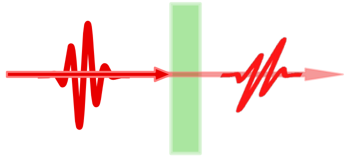


Signal

Intensity $(\Delta T/T)$ / $(\Delta R/R)$



Polarization rotation $\Delta\theta$
(sensitivity ~ 70 µdeg)



Degrees of freedom

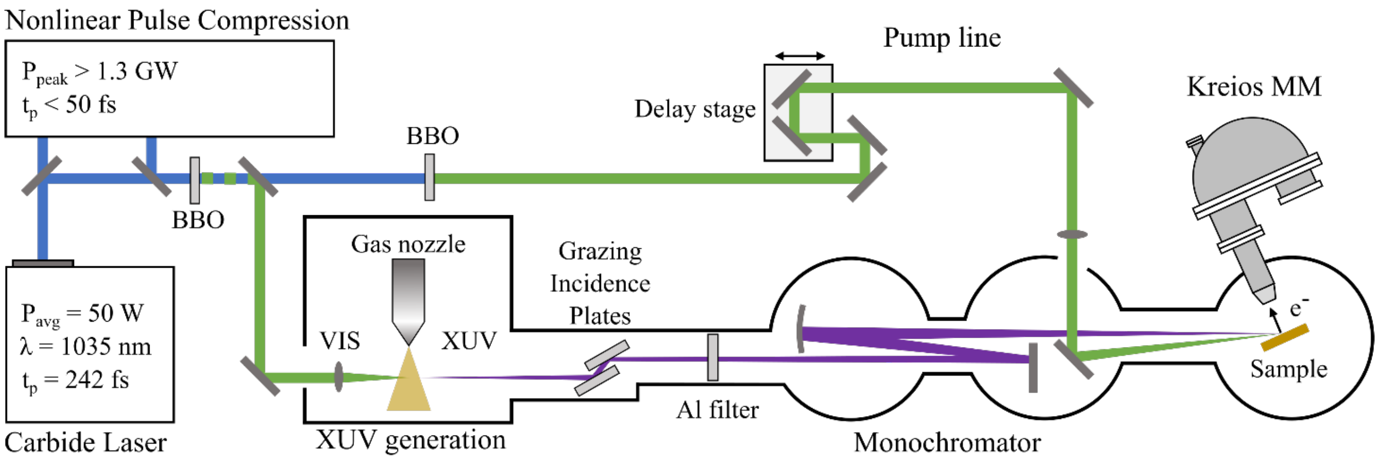
Electronic
Lattice
(Magnetic)

Magnetic

~ M (Kerr, Faraday)
~ L^2

Review of Scientific Instruments ***2020****, 2, 113001*

Time-resolved ARPES: momentum microscope coupled to a fs-XUV source



f_{rep} 540 – 600 kHz

9th harmonic @ 21.6 eV

Advanced Physics Research ***2023***, *2, 2200016*
https://doi.org/10.1364/opticaopen.26124829.v1

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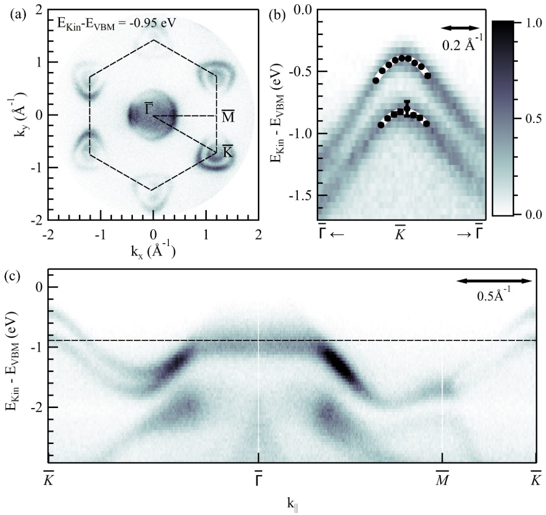
tr-ARPES setup performance: measurements on a WS_2 crystal

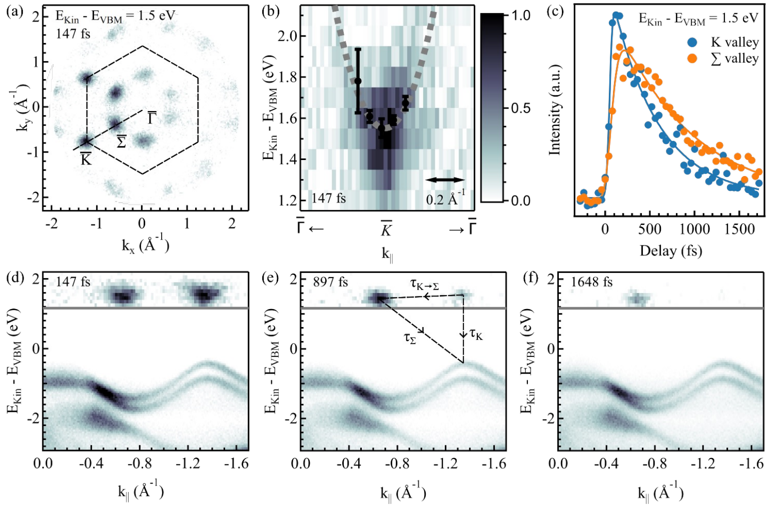
Ground state



Energy resolution < 49 meV;
momentum resolution <0.005 Å^{-1}
Lateral resolution <40 nm

Advanced Physics Research 2023, 2, 2200016

Excited state after pump at 1.2eV



Energy resolution < (107 ± 2) meV
Time-resolution < (49 ± 17) fs

https://doi.org/10.1364/opticaopen.26124829.v1

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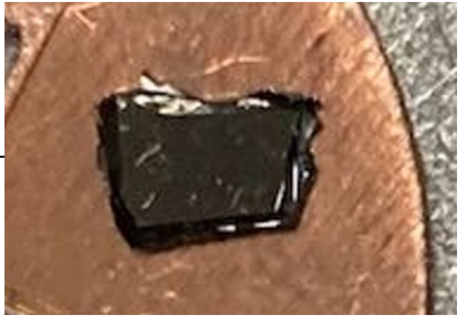
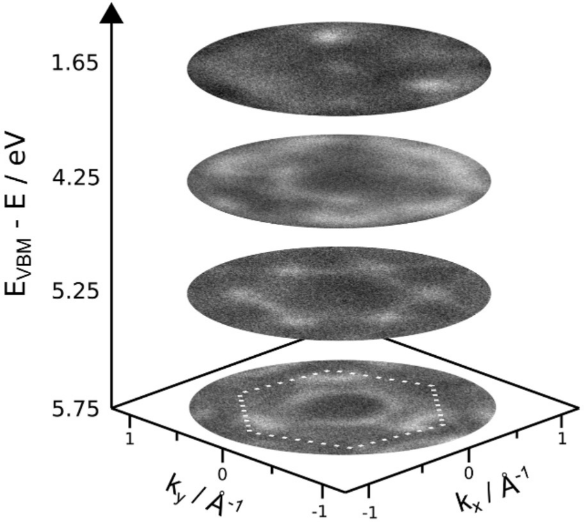
Part 2

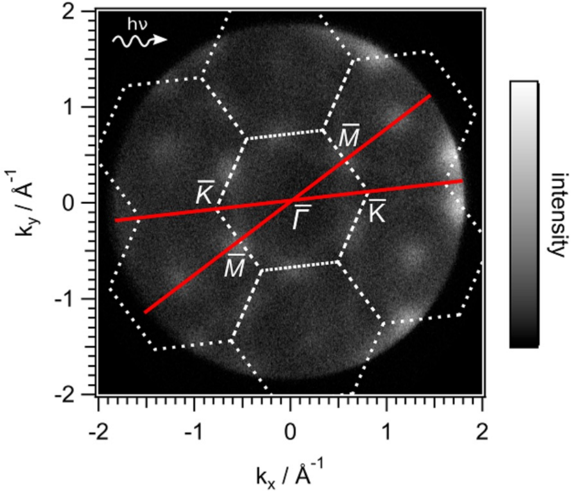
ELECTRONIC STRUCTURE (ground state of the $TMPS_3$ family)



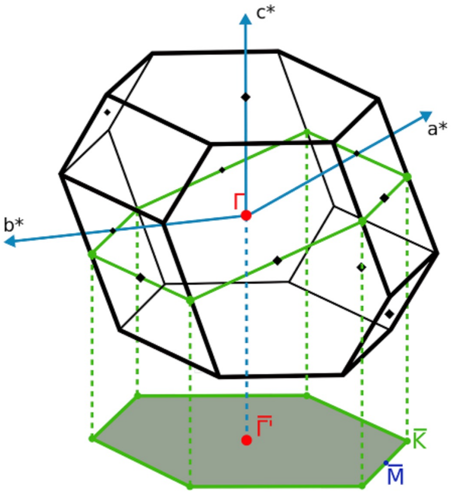
μ- ARPES : $FePS_3$ valence band characterization



Momentum maps

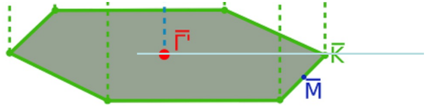


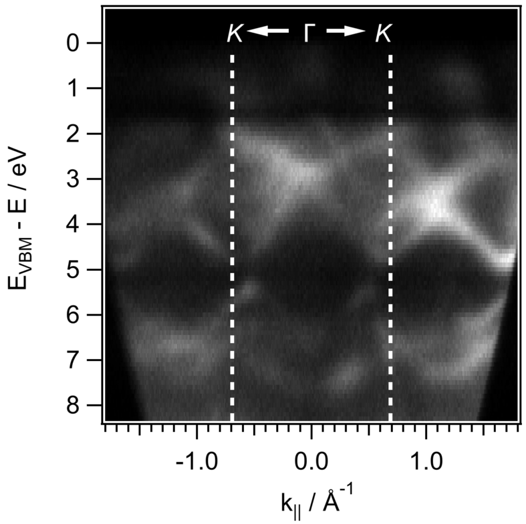
Momentum map
@ 5.15 eV below the VBM



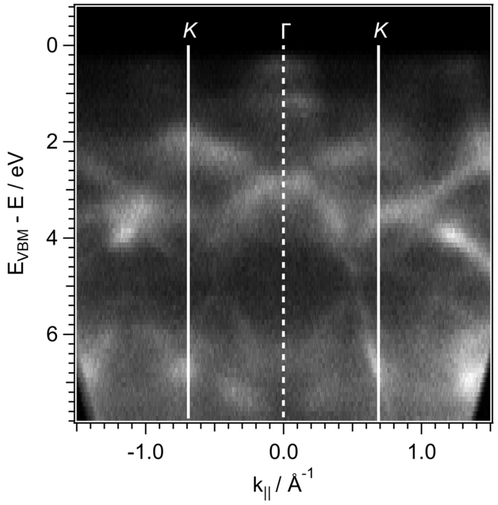
Bulk and surface
Brillouin Zone

μ- ARPES : valence band characterization

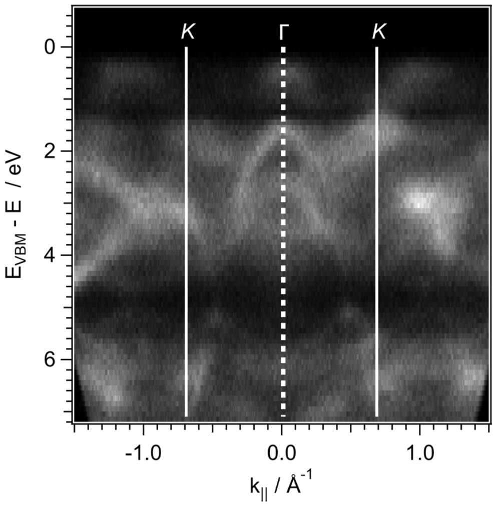




FePS$_3$

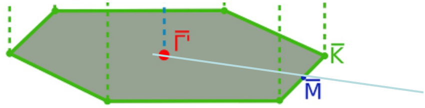


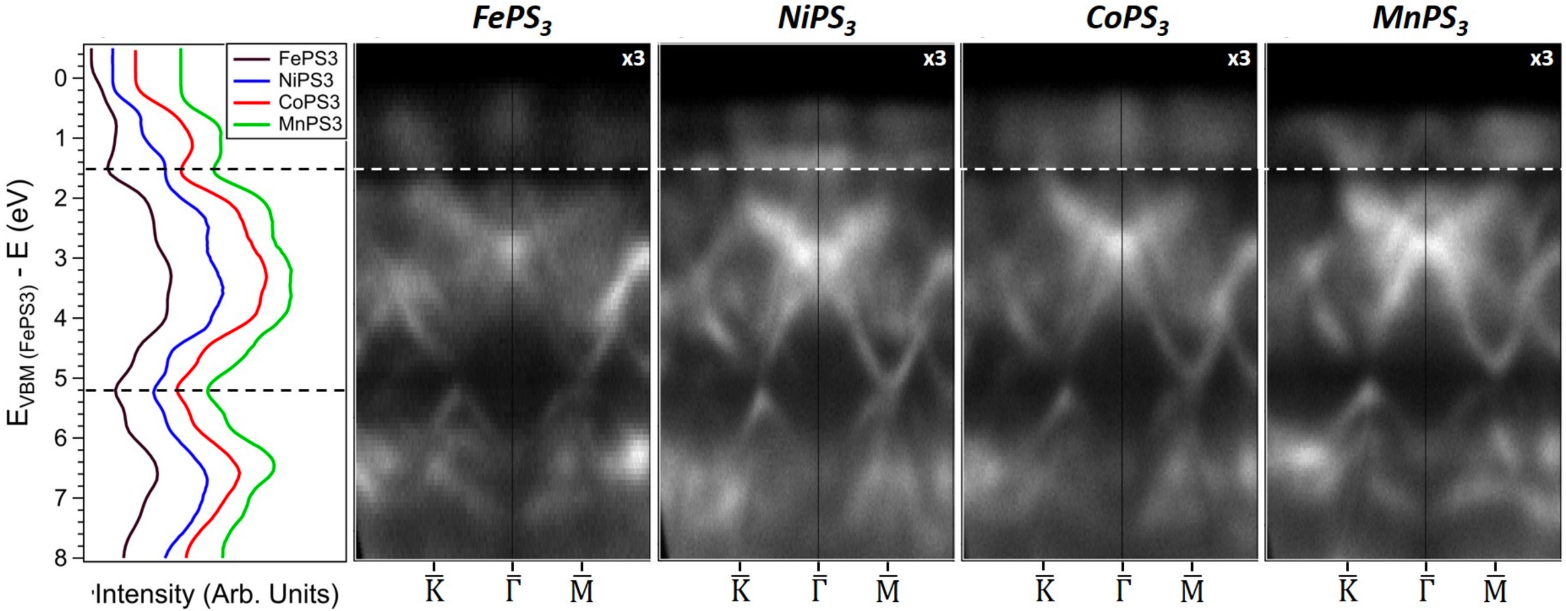
NiPS$_3$



MnPS$_3$

μ- ARPES : valence band characterization





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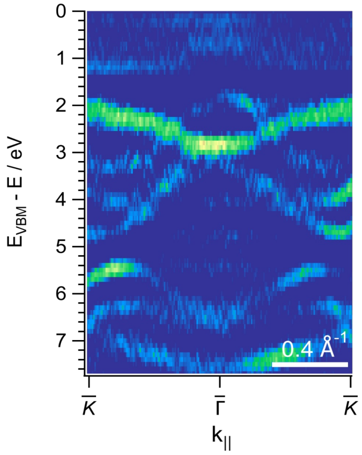
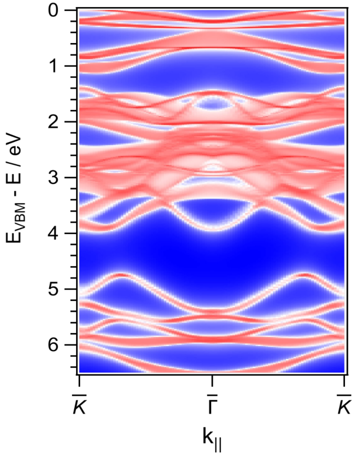
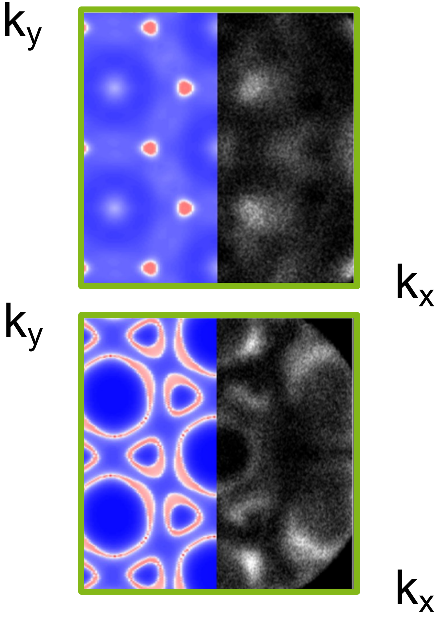
$FePS_3$ valence band characterization



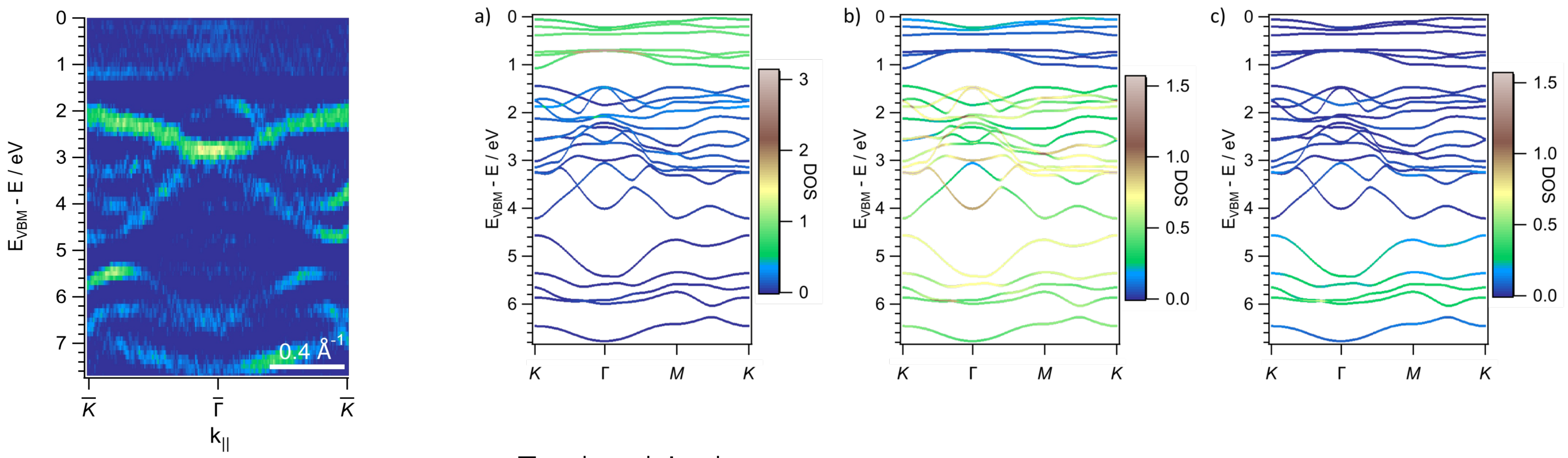




Comparison between experimental data (cruvature alogorithm) and Hubbard-corrected density functional theory (DFT+U) calculations

*Materials Today Electronics **2023**, 6, 100061*

$FePS_3$ as a Mott-Hubbard insulator



Fe d-orbitals

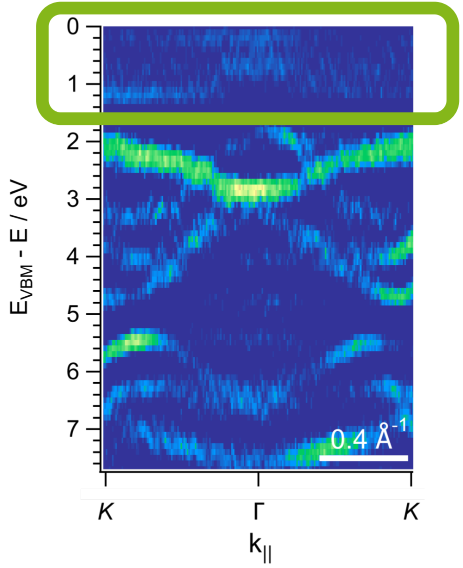
P p-orbitals

S p-orbitals

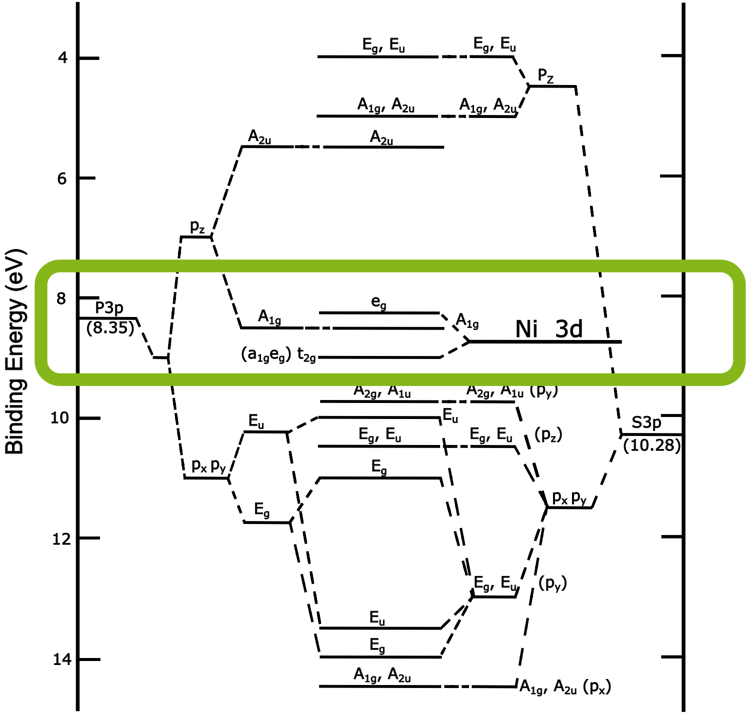
$FePS_3$ can be understood as Mott-Hubbard insulator with the top of the valence band formed by Fe 3d states

Simplified model for understanding d-d excitations in $FePS_3$

$FePS_3$ as ionic crystal: $Fe^{2+}(PS_3)^{2-}$



Local description
of the Fe 3d states: Fe^{2+} multiplet ($3d^6$)



M. Piacentini, et al. *Chemical Physics* 61–71 (1982).

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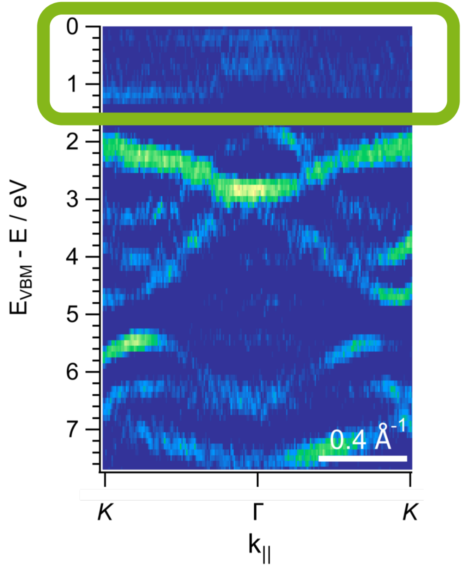
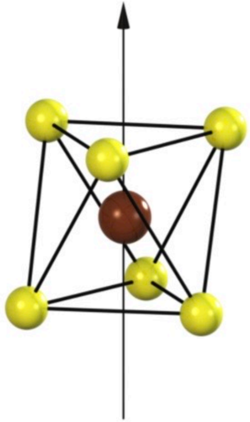
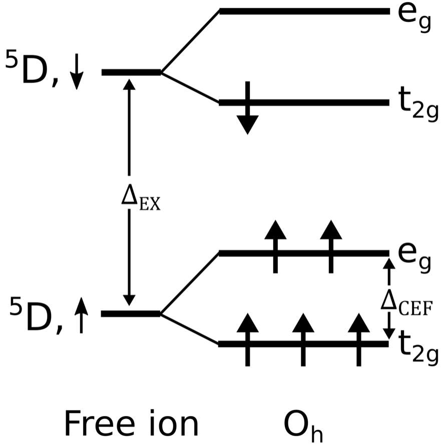
Ground state of the Fe^{2+} multiplet ($3d^6$)



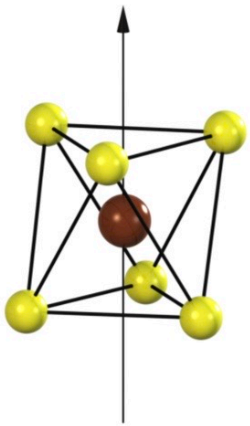


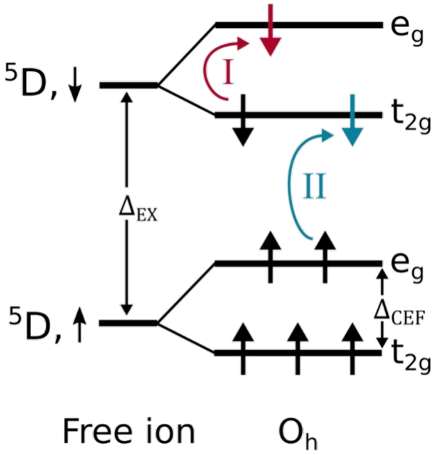


Fe^{2+} $3d^6$ multiplet in an octraedral ligand field
Involved energies: crystal field and exchange
Ground state: $^5T_{2g}$

d-d excitations: phonon and magnon generation

Transition	Energy (eV)
$^5T_{2g} \rightarrow {}^5E_g$ (D)	1.08
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	1.78
$^5T_{2g} \rightarrow {}^3T_{2g}$ (H)	2.14
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	2.46
$^5T_{2g} \rightarrow {}^3T_{2g}$ (F)	2.58
$^5T_{2g} \rightarrow {}^3T_{1g}$ (P)	2.84





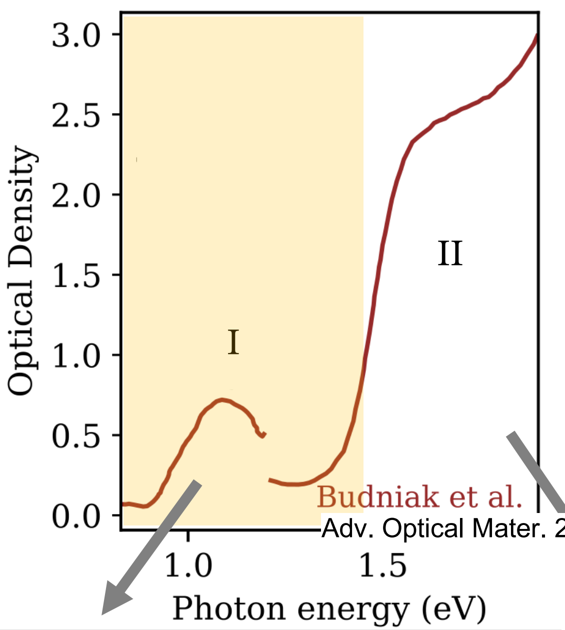
I : spin conserving transition → modifies the cristal field: dynamic Jahn-Teller
II : not spin conserving → phonon and magnon generation


0D excitations generate lattice
and spin dynamics in a 2D material

Part 3

OPTICALLY INDUCED DYNAMICS

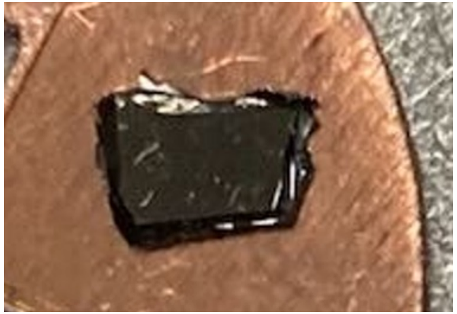
Time-resolved magneto-optical measurements on $FePS_3$



Adv. Optical Mater. 2022, 10, 2102489

Below band gap excitation
*Advanced Materials **2023**, 35, 2208355*

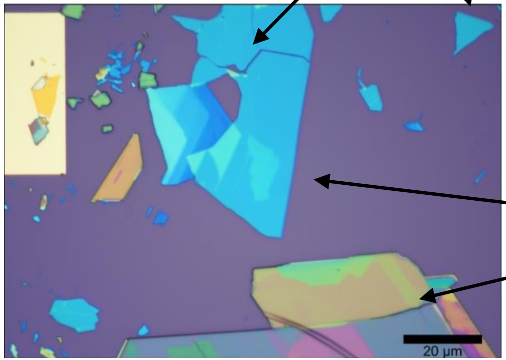
Above band gap excitation
*Physical Review Materials **2024**, 014408*



$FePS_3$ crystal

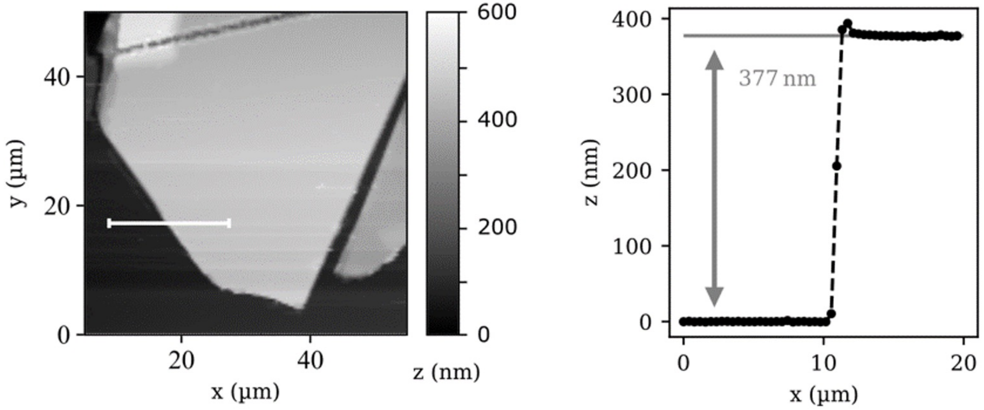
$FePS_3$ flakes

h-BN



$FePS_3$

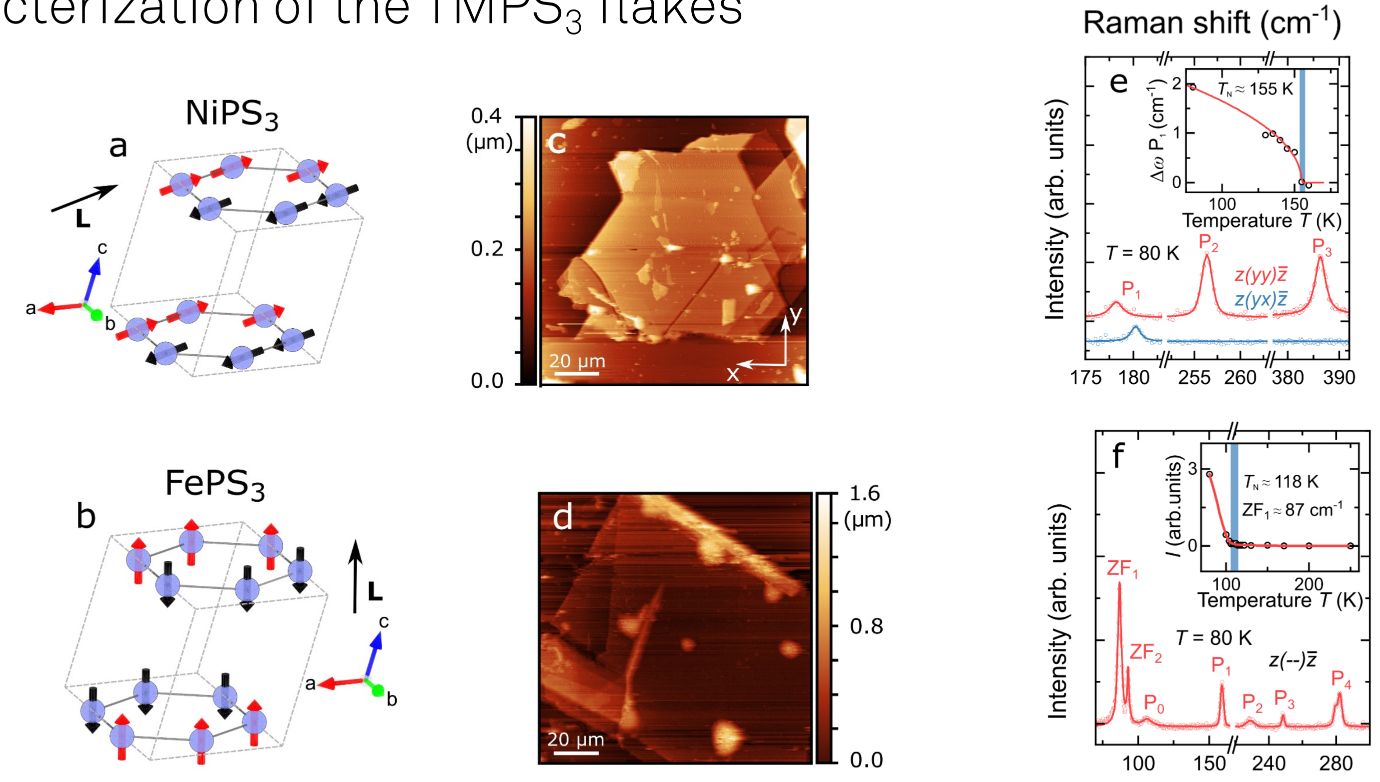
Characterization of the $FePS_3$ flakes



$FePS_3$ exfoliated
on SiO|Si substrate
h = 380 nm
d ≈ 50 µm

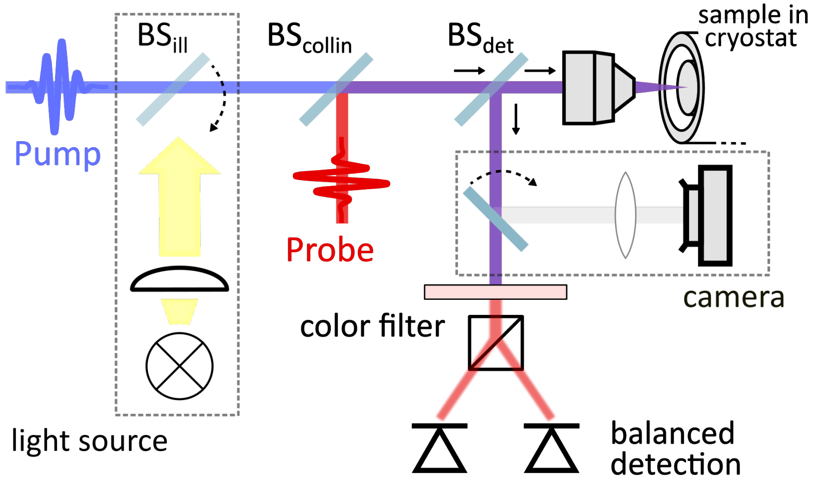
Atomic force microscopy

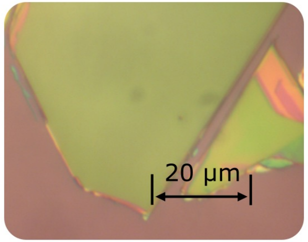
Characterization of the $TMPS_3$ flakes



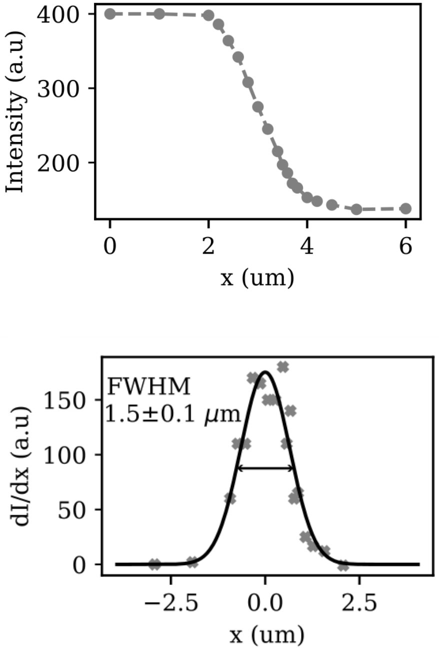
Physical Review Materials ***2024***, 014408

Microscope pump-probe setup

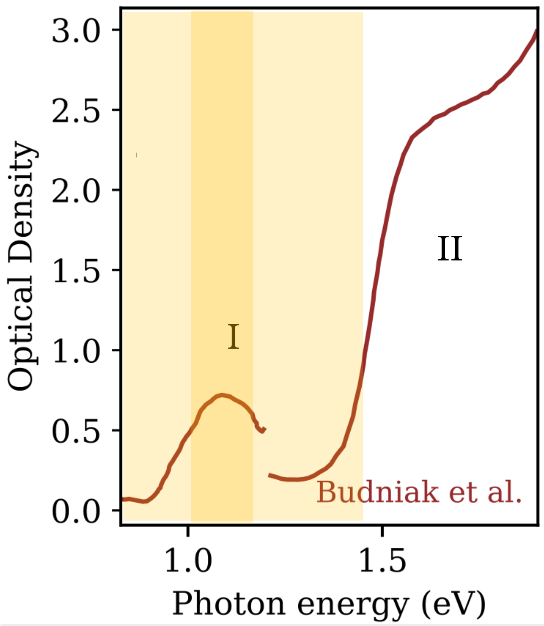




Knife edge



$FePS_3$ flake– below band gap excitation

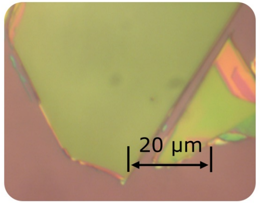


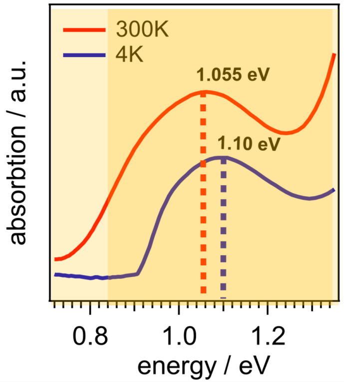
$FePS_3$

- E_{gap} = 1.5 eV
- T_N= 118 K

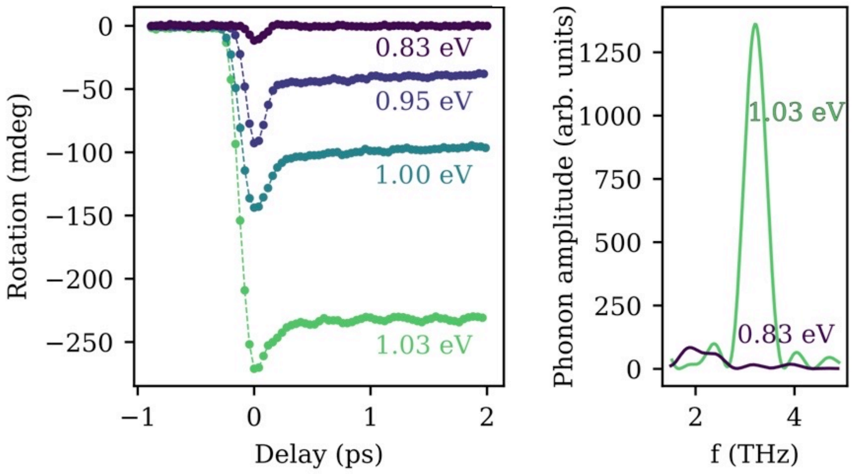
Experiment

- E_{probe}= 1.45 eV
- E_{pump}= (0.83 – 1.08) eV
  resonant with $^5T_{2g} \rightarrow {}^5E_g$ transition
- T= 10K



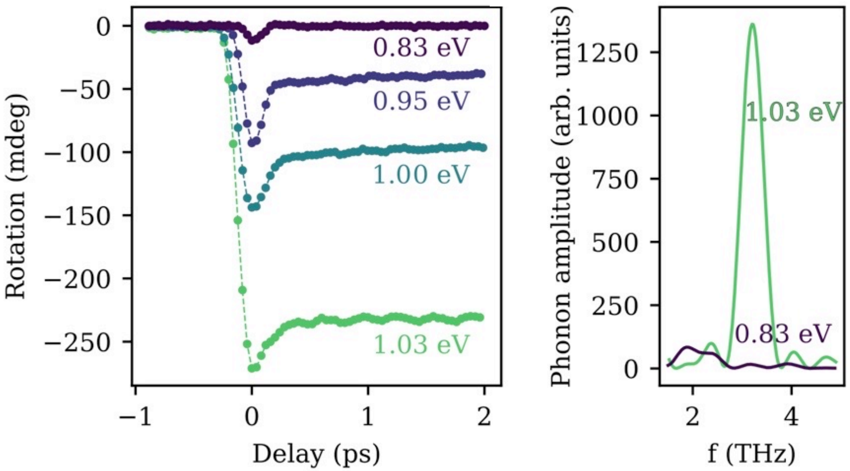


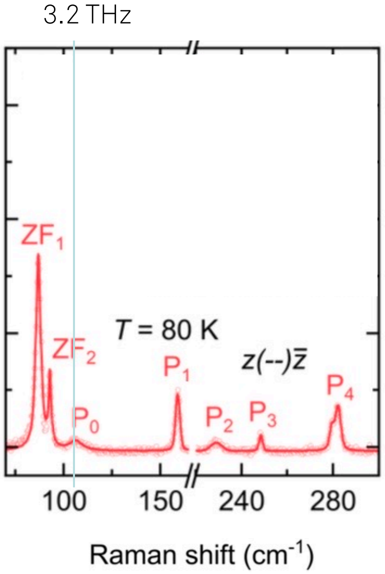
$FePS_3$ flake: results

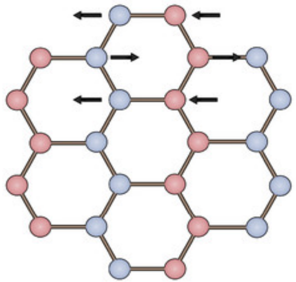


Coherent contribution to the signal: oscillations at 3.2 THz

$FePS_3$ flake: results







Coherent contribution to the signal: oscillations at 3.2 THz
Excitation of optical phonon modes, displacement of the Fe^{2+} atoms and of the ligands

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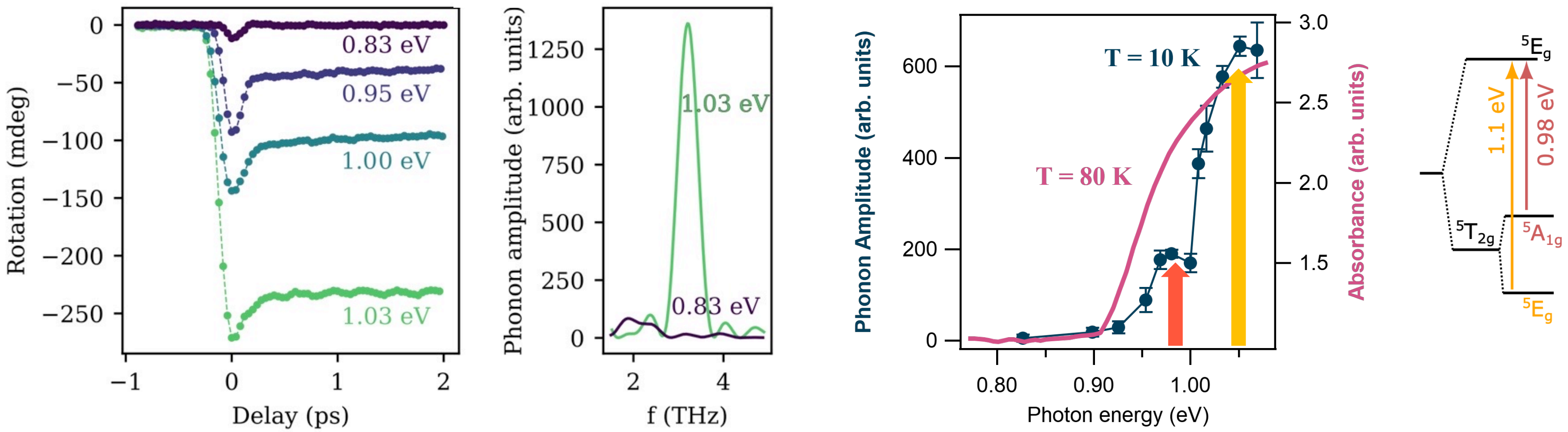
$FePS_3$ flake: results



Phonon amplitude follows trend of optical absorption, including the degeneracy of the $^5T_{2g}$→^5E_g transition due to distortion of the ligands → d-d transition plays a crucial role

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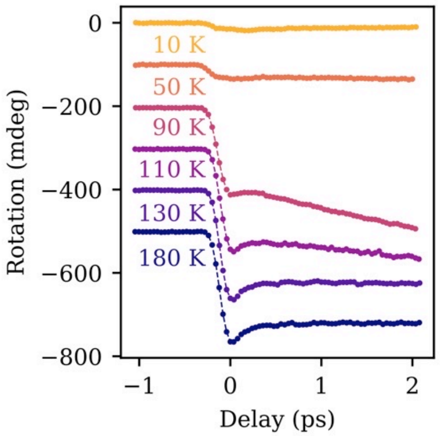
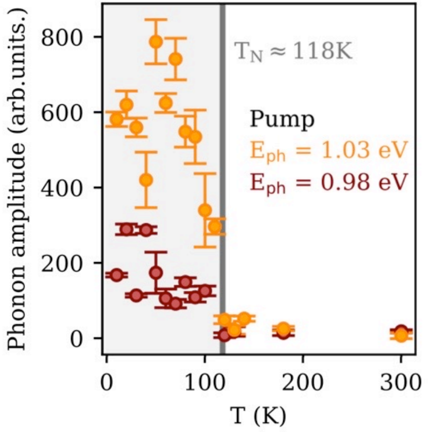
Temperature dependence

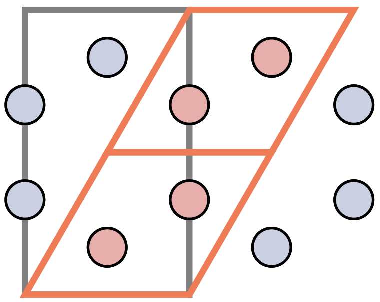


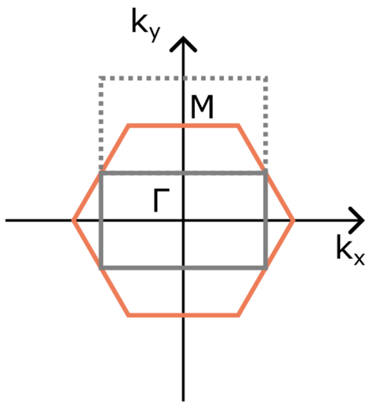


Phonon amplitude vanishes for $T>T_N$

Temperature dependence and zone-folding

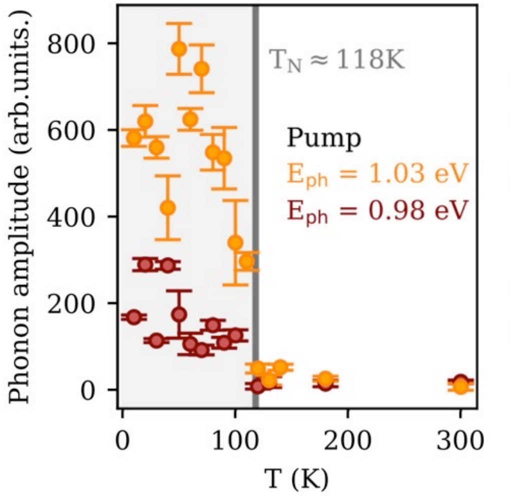
$T \lessgtr T_N$





Paramagnetic

Antiferromagnetic

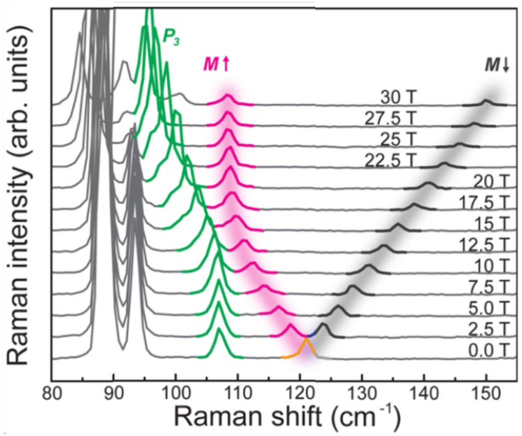


Zone-folded phonon mode

Generation of zone-folded optical phonons @3.2 THz

WHAT ABOUT SPIN DYNAMICS?

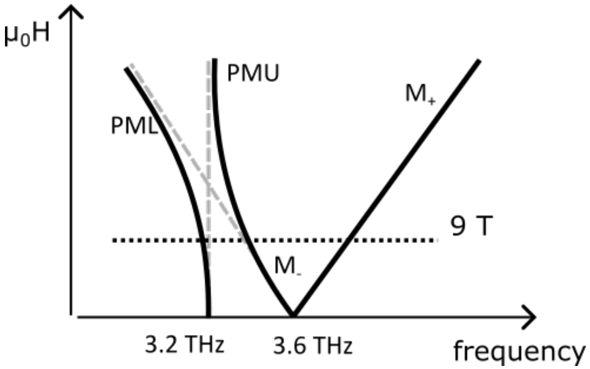
Phonon-magnon hybridization



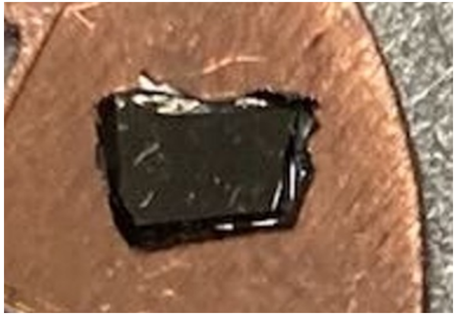
Liu et al. *Phys. Rev. Lett.* **127** (2021)

0 T
P: 3.2 THz
M: 3.6 THz

Phonon-magnon hybridization: experiments on a $FePS_3$ crystal



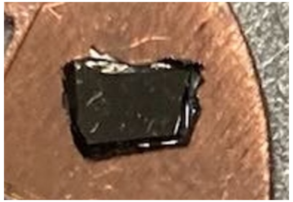
Liu et al. *Phys. Rev. Lett.* **127** (2021)

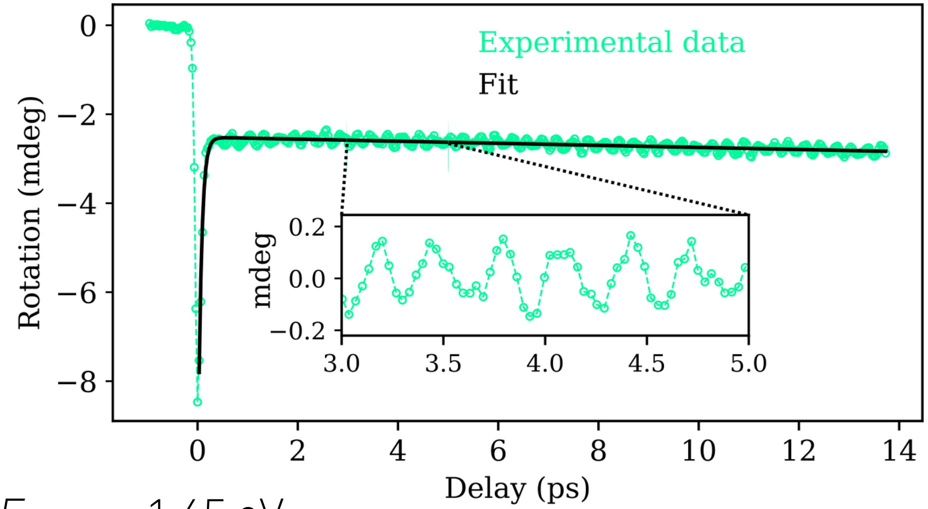


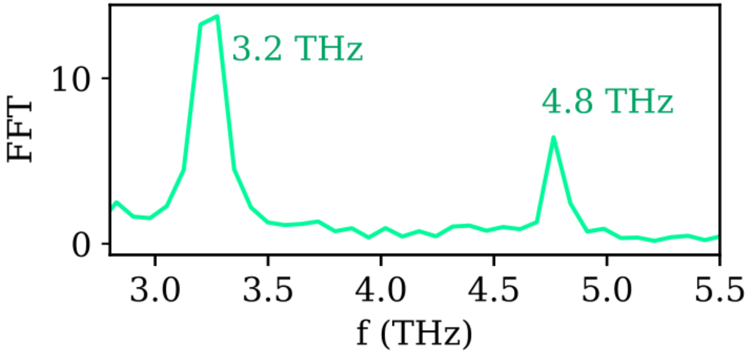
$FePS_3$ crystal

Lower branch phonon-magnon (PML)
Upper branch phonon-magnon (PMU)

Bulk $FePS_3$: results







E_{Probe} = 1.45 eV
E_{Pump} = 1.03 eV
T = 10 K

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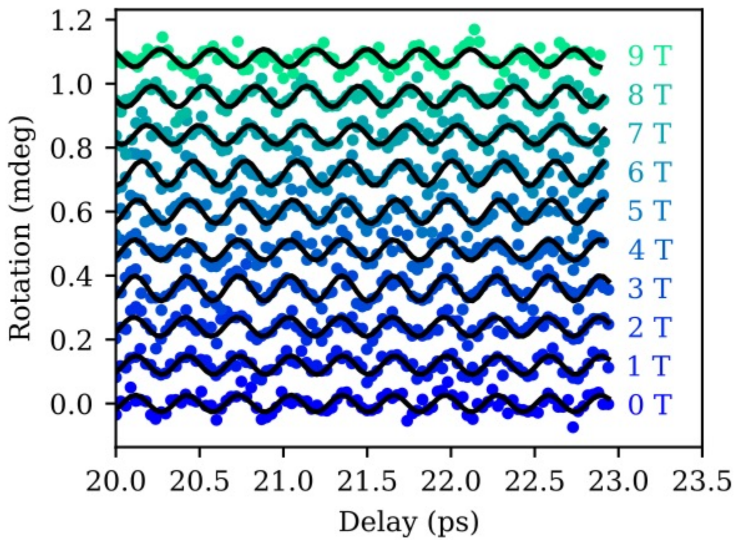
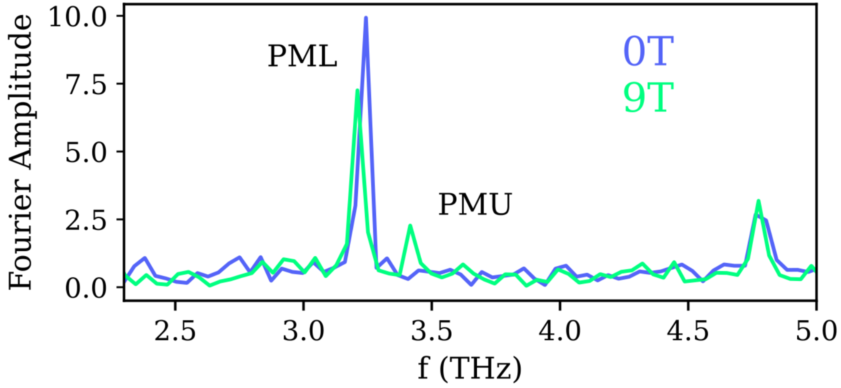
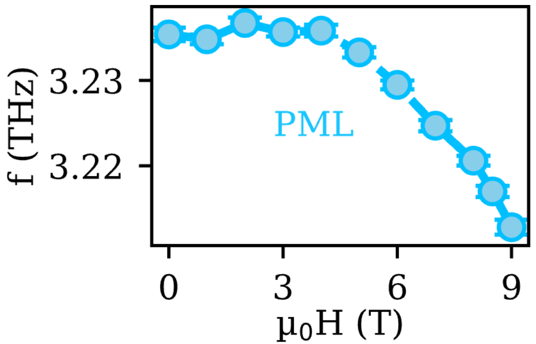
Phonon-magnon hybridization



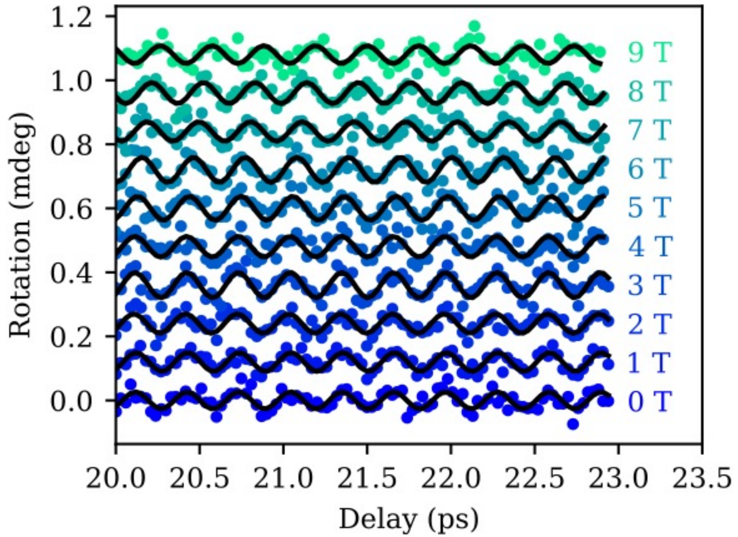


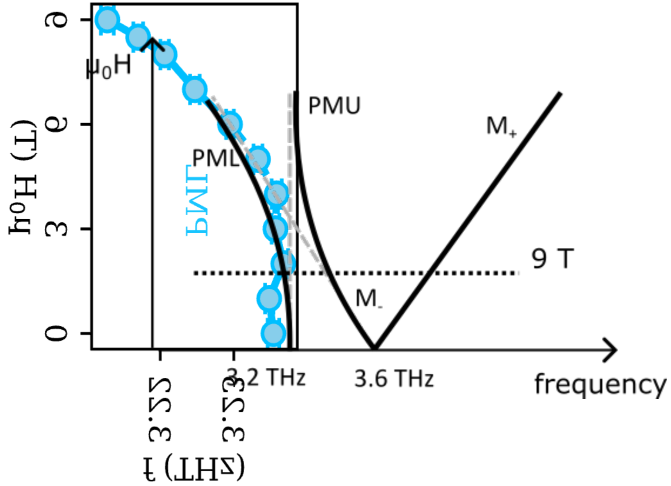


E_{Probe} = 1.45 eV
E_{Pump} = 1.03 eV
T = 10 K

Emergence of upper and lower branch of the phonon-magnon mode

Phonon-magnon hybridization



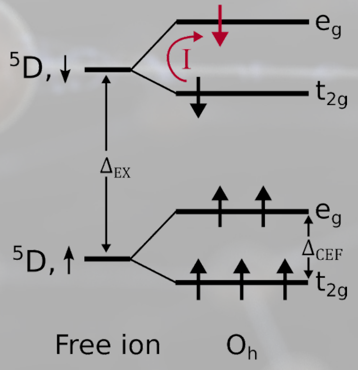


E_{Probe} = 1.45 eV
E_{Pump} = 1.03 eV
T = 10 K

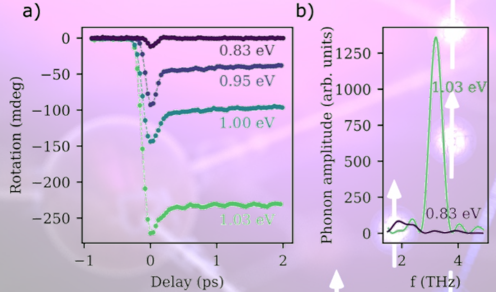
Emergence of upper and lower branch of the phonon-magnon mode

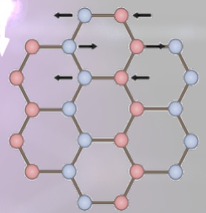
We generated THz spin dynamics using resonant optical excitation

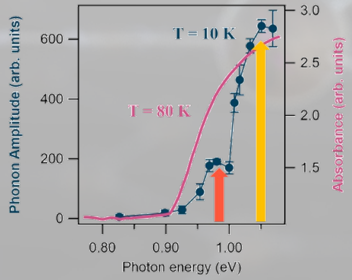
The story so far



Exciting the
$^5T_{2g} \rightarrow {}^5E_g$ d-d transition

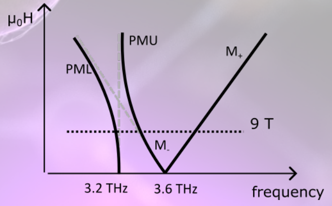


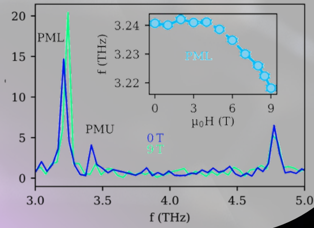




Generation of (zone folded) optical phonons @3.2 THz

Hybrid phonon-magnon mode generated @9T



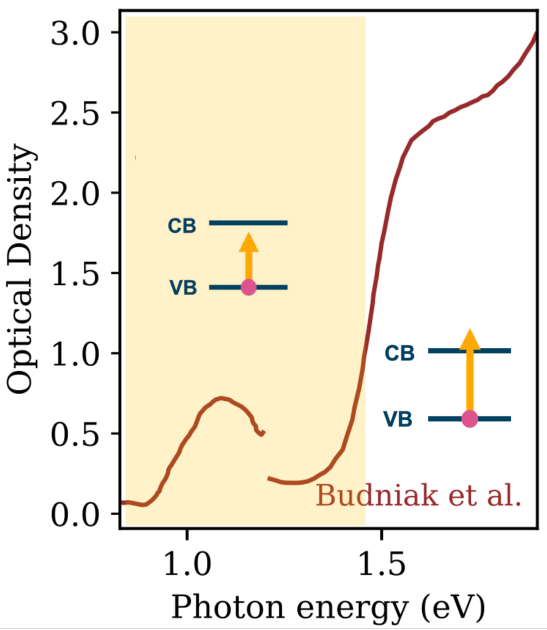


Part 4

TRACING d-d TRANSITIONS ON ULTRAFAST TIME-SCALES BY trARPES

Uncovering d-d transitions dynamics with trARPES

Transition	Energy (eV)	
$^5T_{2g} \rightarrow ^5E_g$ (D)	1.08	Below band gap
$^5T_{2g} \rightarrow ^3T_{1g}$ (H)	1.78	Above band gap
$^5T_{2g} \rightarrow ^3T_{2g}$ (H)	2.14	
$^5T_{2g} \rightarrow ^3T_{1g}$ (H)	2.46	
$^5T_{2g} \rightarrow ^3T_{2g}$ (F)	2.58	
$^5T_{2g} \rightarrow ^3T_{1g}$ (P)	2.84	



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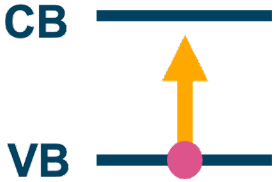
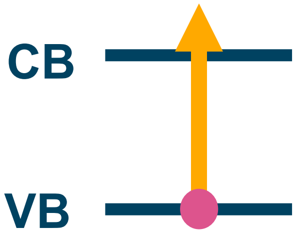
Uncovering d-d transitions dynamics with trARPES

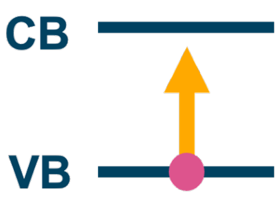


Below band gap excitation (E_{pump} = 1.2 eV)

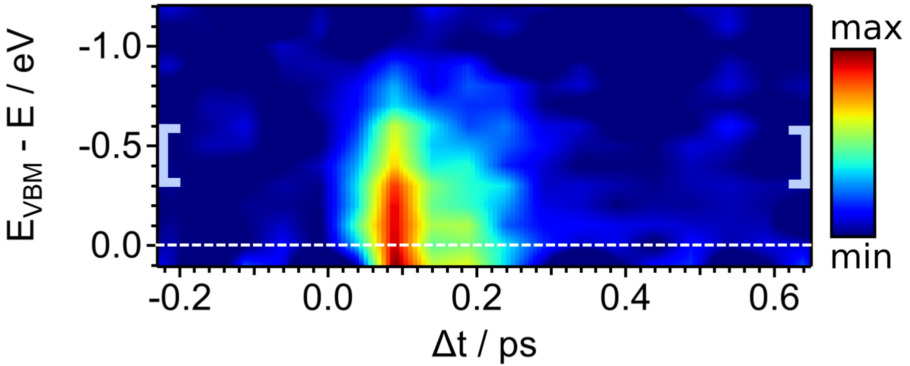


Above band gap excitation (E_{pump} = 2.4 eV)

Uncovering d-d transitions with trARPES: below band gap excitation



Below band gap excitation (E_{pump} = 1.2 eV)



Momentum integrated transient ARPES signal (DOS) above the valence band maximum (VBM)

→ **excited state dynamics**

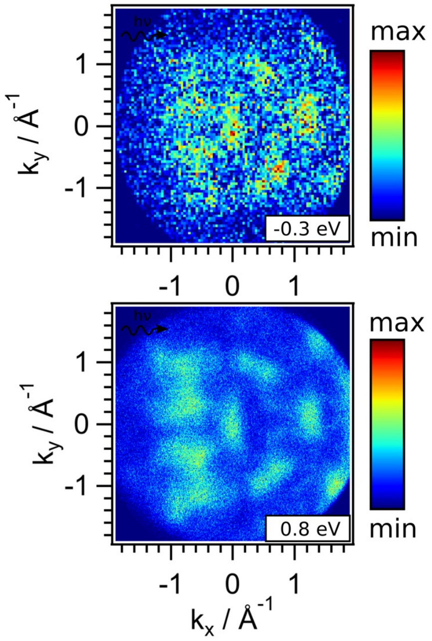
Origin of the trARPES signal: momentum maps

Unoccupied states → excited state

- 0.3 eV

+ 0.8 eV

Occupied states → ground state



Origin of the trARPES signal: momentum maps

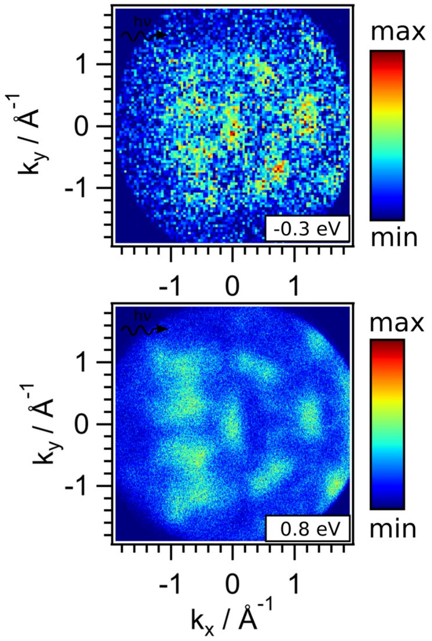
Unoccupied states → excited state

- 0.3 eV

Energy difference 1.1eV

+ 0.8 eV

Occupied states → ground state



Origin of the trARPES signal: fingerprint of the $^5T_{2g} \rightarrow {}^5E_g$ transition

Unoccupied states → excited state

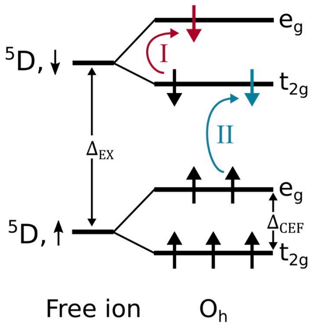
- 0.3 eV

Energy difference 1.1eV

+ 0.8 eV

Occupied states → ground state

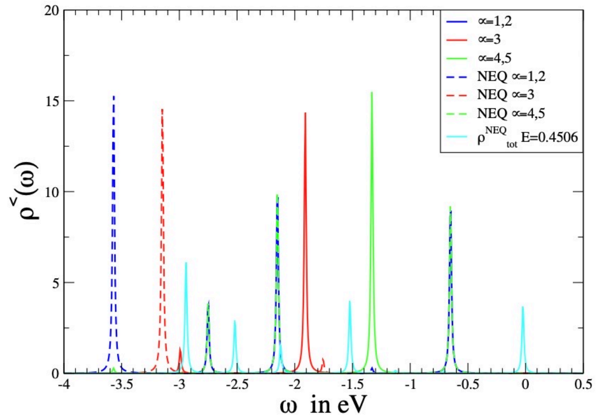
Transition	Energy (eV)
$^5T_{2g} \rightarrow {}^5E_g$ (D)	1.08 I
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	1.78
$^5T_{2g} \rightarrow {}^3T_{2g}$ (H)	2.14
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	2.46
$^5T_{2g} \rightarrow {}^3T_{2g}$ (F)	2.58
$^5T_{2g} \rightarrow {}^3T_{1g}$ (P)	2.84



$^5T_{2g} \rightarrow {}^5E_g$ transition is at ~1.1 eV

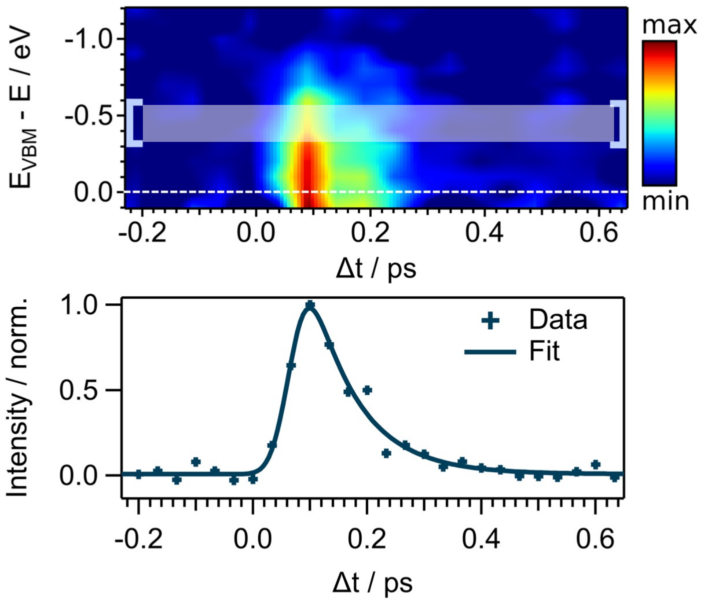
Fingerprint of the $^5T_{2g}$ → ^5E_g d-d transition in momentum space!

Transition	Energy (eV)
$^5T_{2g} \rightarrow {}^5E_g$ (D)	1.08
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	1.78
$^5T_{2g} \rightarrow {}^3T_{2g}$ (H)	2.14
$^5T_{2g} \rightarrow {}^3T_{1g}$ (H)	2.46
$^5T_{2g} \rightarrow {}^3T_{2g}$ (F)	2.58
$^5T_{2g} \rightarrow {}^3T_{1g}$ (P)	2.84



Equilibrium spectrum of the lesser Green function and spectrum with the excited state $|E_{ex} = 1.1\text{eV}, S = 2\rangle$

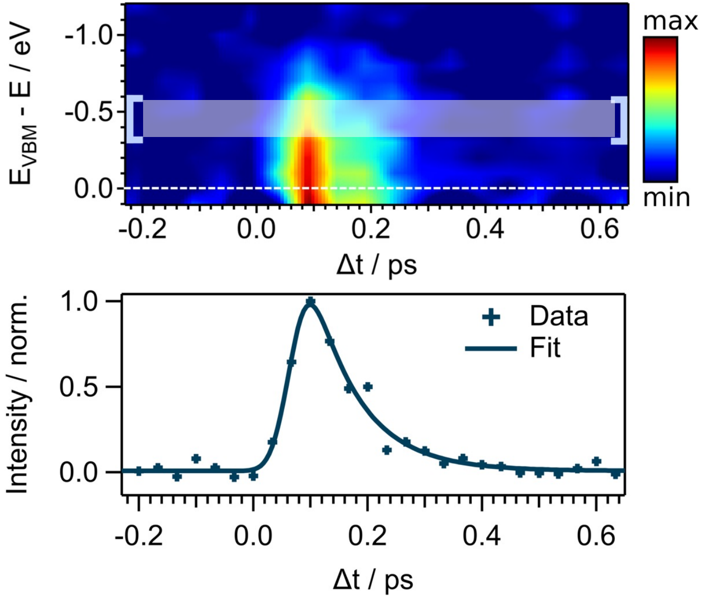
Intrinsic dynamics of the $^5T_{2g} \rightarrow {}^5E_g$ transition

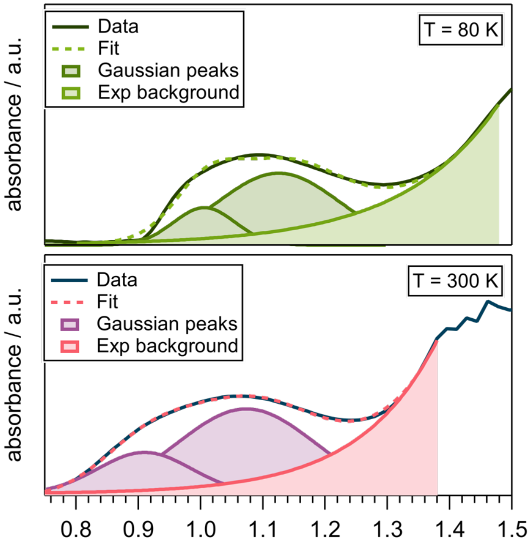


Instantaneous excitation

within experimental resolution of (49 ± 17) fs

Lifetimes vs. Linewidths $\Gamma_{hom} = \frac{\hbar}{\tau}$





Position / eV	FWHM / eV
1.006	117
1.1257	229

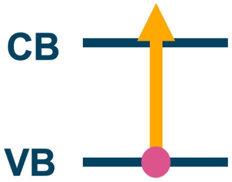
Position / eV	FWHM / eV
0.91	177
1.073	257

$\tau = 83 \pm 10$ fs → $\Gamma_{hom} = 48$ meV

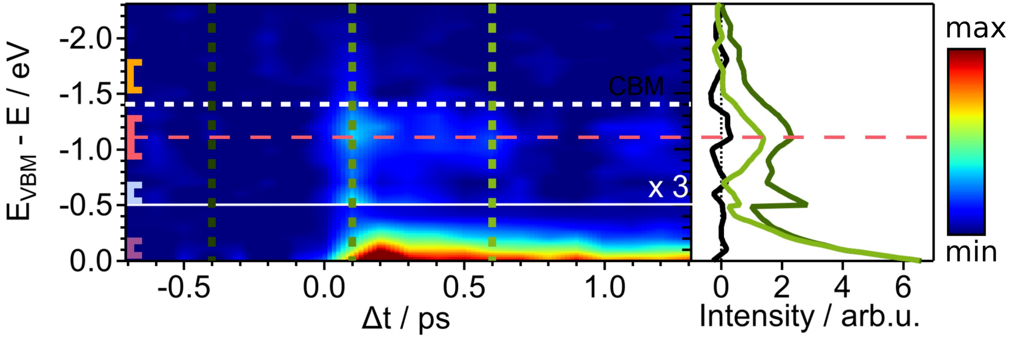
$\Gamma = 177$ meV → $\tau = 23$ fs

$\Gamma = 258$ meV → $\tau = 15$ fs

Uncovering d-d transitions with trARPES: above band gap excitation



Above band gap excitation (E_{pump} = 2.4 eV)



Momentum integrated transient ARPES signal (DOS) above the valence band maximum (VBM)
→ excited state dynamics

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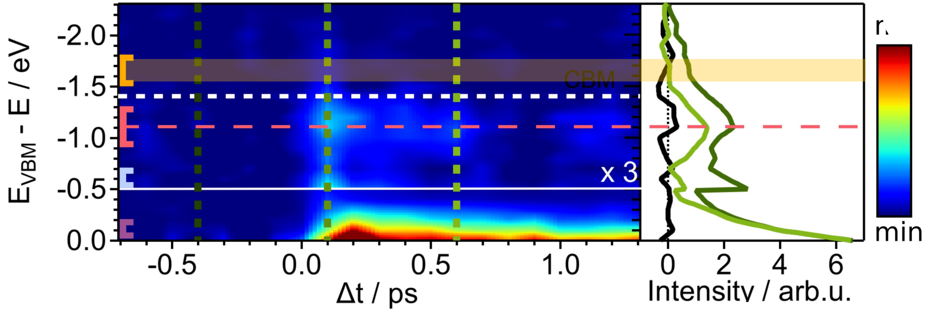
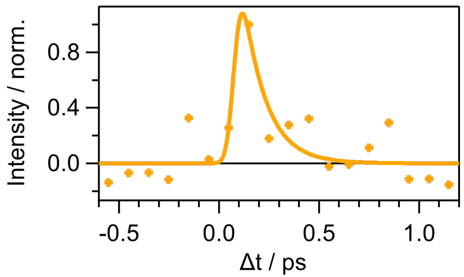
Above band gap excitation: excitations and decay-times





τ_{yellow} = 115 ± 100 fs

Conduction band

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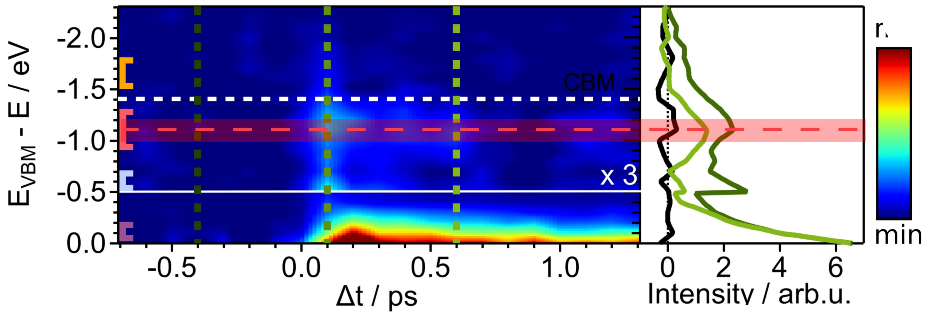
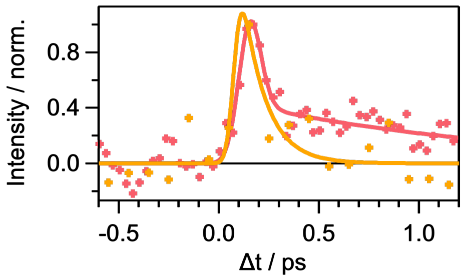
Above band gap excitation: excitations and decay-times





τ_{yellow} = 115 ± 100 fs

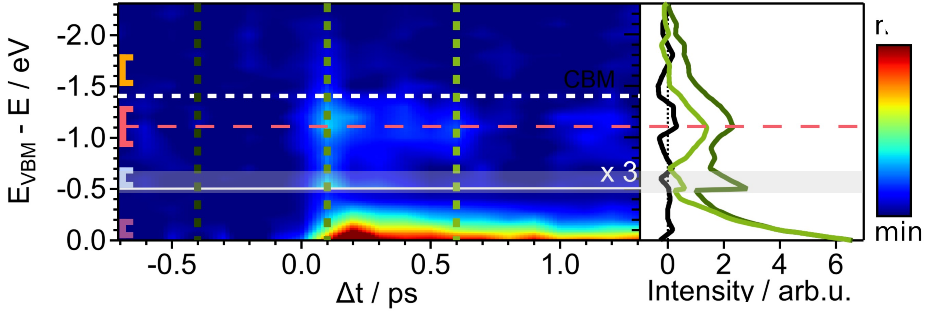
$\tau_{red,1}$ = 46 ± 34 fs

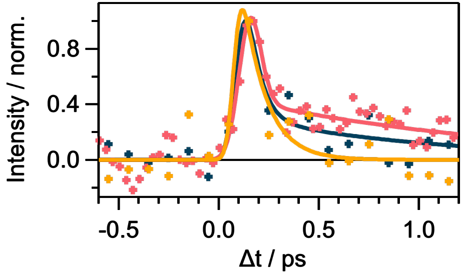
$\tau_{red,2}$ = 1273 ± 268 fs

Conduction band

$^5T_{2g} \rightarrow {}^3T_{1g}$ transition

Above band gap excitation: excitations and decay-times





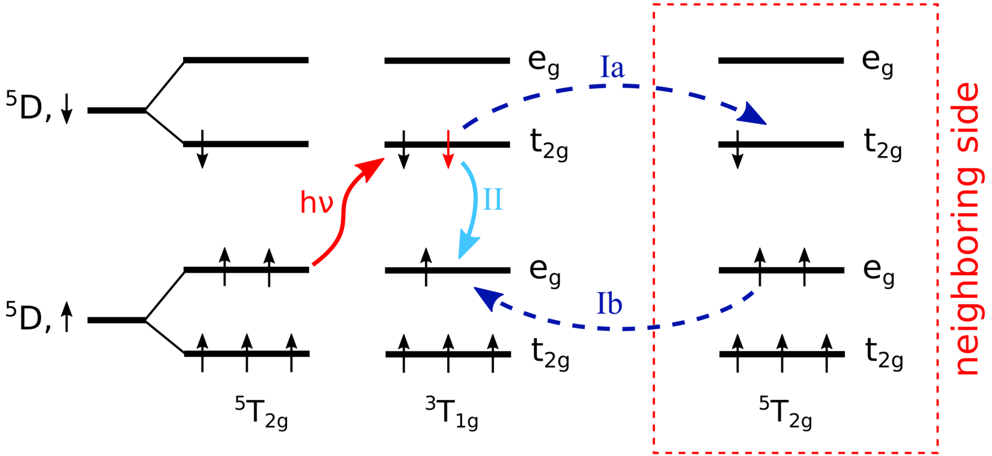
τ_{yellow} = 115 ± 100 fs — Conduction band

$\tau_{red,1}$ = 46 ± 34 fs — $^5T_{2g} \rightarrow {^3T_{1g}}$ transition

$\tau_{red,2}$ = 1273 ± 268 fs

$\tau_{gray} = 66 \pm 128$ fs — $^5T_{2g} \rightarrow {^5E_g}$ transition

Decay mechanisms of the $^5T_{2g} \rightarrow {}^3T_{1g}$ transition



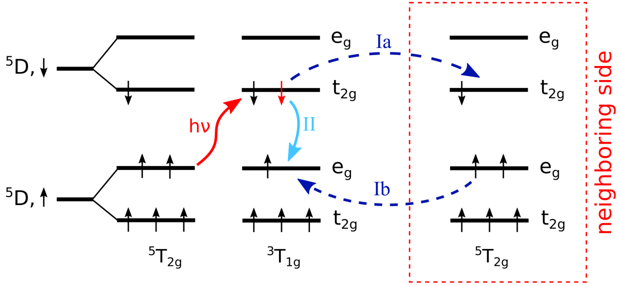
$\tau_{red,1} = 46 \pm 34$ fs

Hopping between neighboring sites

$\tau_{red,2} = 1273 \pm 268$ fs

Spin-orbit-mediated on-site spin-flip

Decay mechanisms of the $^5T_{2g} \rightarrow {}^3T_{1g}$ transition



$\tau_{red,2} = 1273 \pm 268$ fs

Spin-orbit-mediated on-site spin-flip

DFT+ U+ SOC

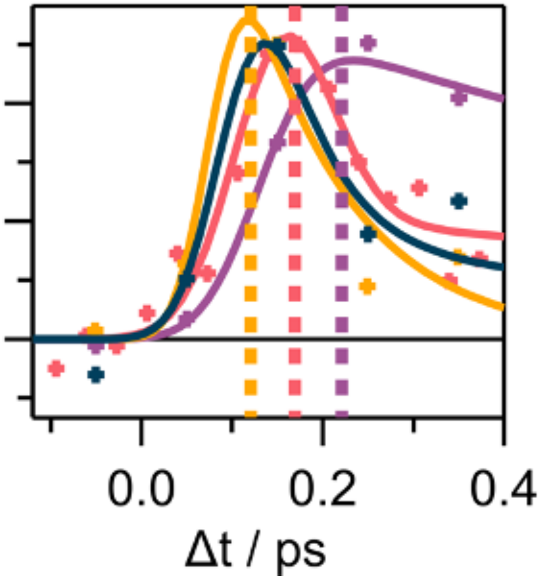
λ_{SOC} (Fe^{2+})= 14.04 meV → τ_{SOC} (Fe^{2+})= h/ $\lambda_{SOC} \cong 300$ fs

Mixing of the ^5E_g and $^3T_{1g}$ states (energy separation 0.72 eV)

Similar to $NiPS_3$ in PRB 46, 5134 (1992)

λ_{SOC} =34.7 meV, $\Delta E(^1E_g - {}^3T_{2g})$=0.98eV

Ultrafast dynamics for off-resonant excitation: build-up time



Conduction band: instantaneous

$^5T_{2g} \rightarrow {}^5E_g$ transition
exhibits a quicker buildup (22 fs)
compared to the spin-forbidden
$^5T_{2g} \rightarrow {}^3T_{1g}$ transition (48 fs)

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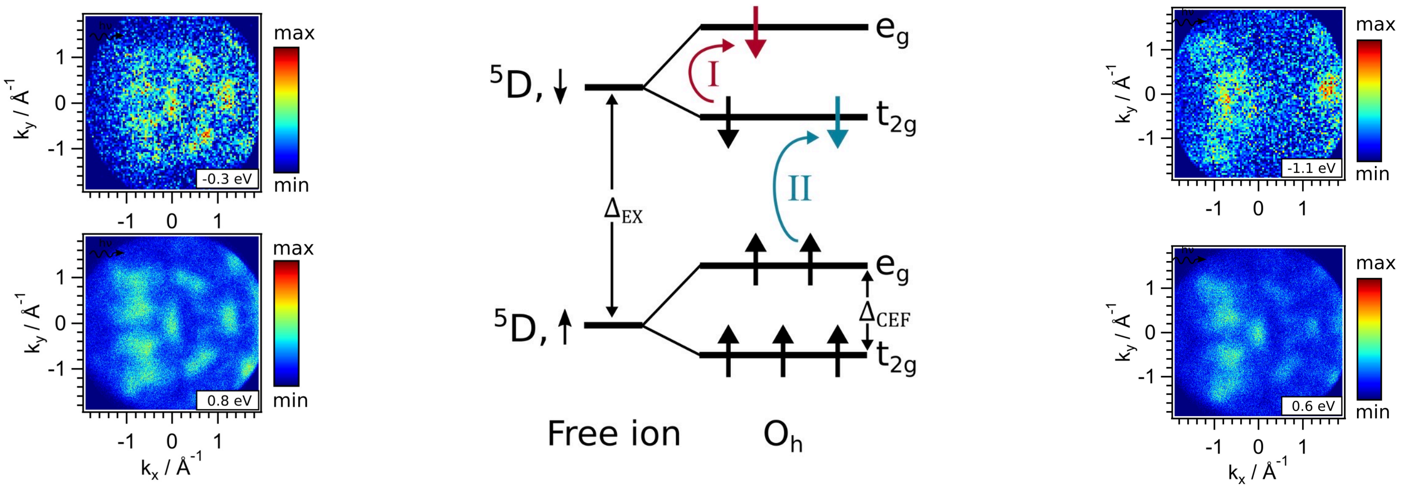
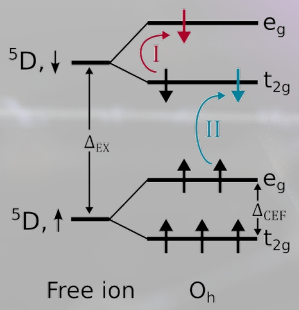
The last part of the story: summary trARPES experiments

arXiv:2402.03018 2024



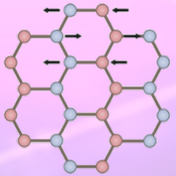
Fingerprint in momentum space of d-d transitions
Transitions with energy above band gap can be also detected
Ultrafast dynamics (buildup and decay) accessible!

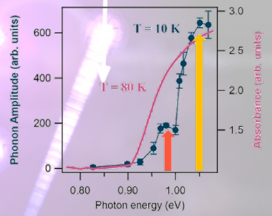
Conclusions

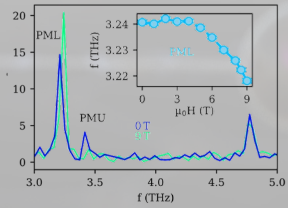


d-d transitions in $FePS_3$ explored by trMO and trARPES

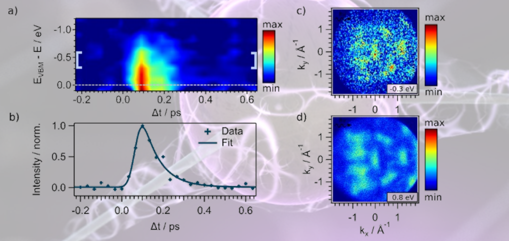
***Advanced Materials* 2023, 35, 2208355**
***Materials Today Electronics* 2023, 6, 100061**
***Physical Review Materials* 2024, 014408**
***arXiv:2402.03018* 2024**
https://doi.org/10.1364/opticaopen.26124829.v1

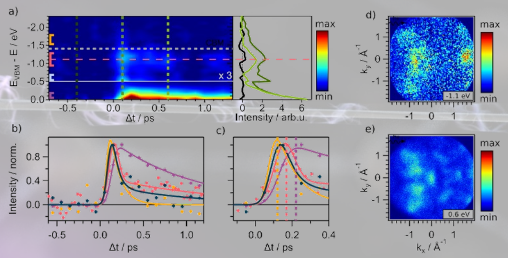






Generation of (zone folded) optical phonons @3.2 THz
Hybrid phonon-magnon mode generated @9T





Fingerprint in momentum space of d-d transitions
Ultrafast dynamics accessible!

Collaborations

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Matjia Stupar

Jonah Nitschke

Umut Parlak

Fabian Mertens

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MERCUR
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MERCUR Kooperation
Towards an UA Ruhr Ultrafast Laser Science Center

DFG-TRR160
Coherent Manipulation of Interacting Spin Excitations in Tailored Semiconductors

TRR 160

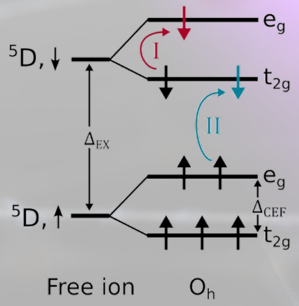
ERC Consolidator
Coherent optical control of multi-functional nano-scale hybrid units

erc

SINFONIA FET
Selectively activated INFOrmation techNology by hybrId organic interfAces

SINFONIA

Conclusions



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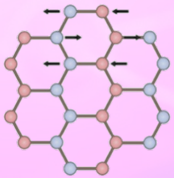

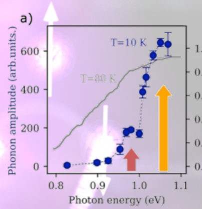

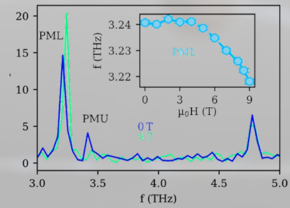

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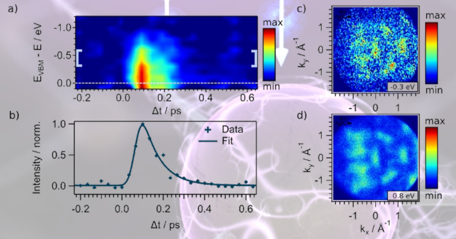

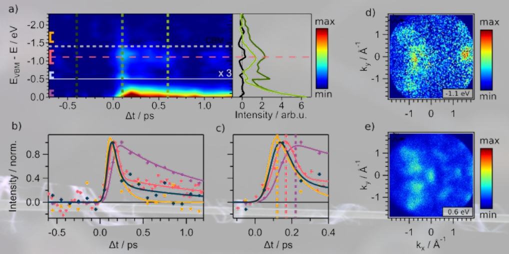

Fingerprint in momentum space of d-d transitions
Ultrafast dynamics accessible!

Open PhD and postdoc positions!