CRC / TRR 227
Ultrafast Spin Dynamics

Towards a "complete" picture of ultrafast dynamics in Fe_2GeTe_3 (FGT)

Dr. Y. Will Windsor

Technische Universität Berlin & Fritz Haber Institute

FRITZ-HABER-INSTITUT
MAX-PLANCK-GESELLSCHAFT

SPICE-SPIN+X Seminar 16 Nov. 2022

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SPIN PHENOMENA
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SFB/TRR 173
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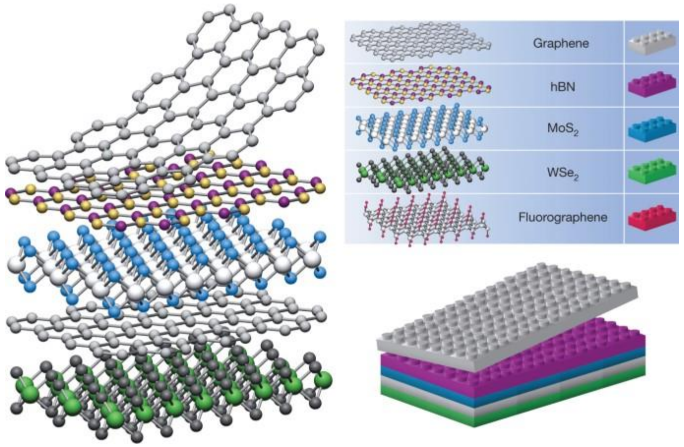
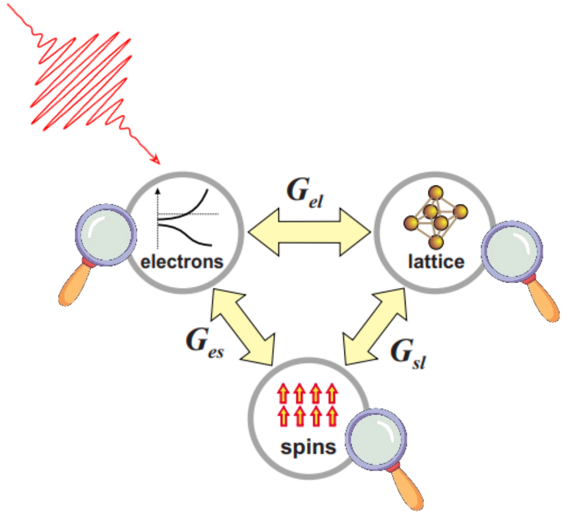
Overview

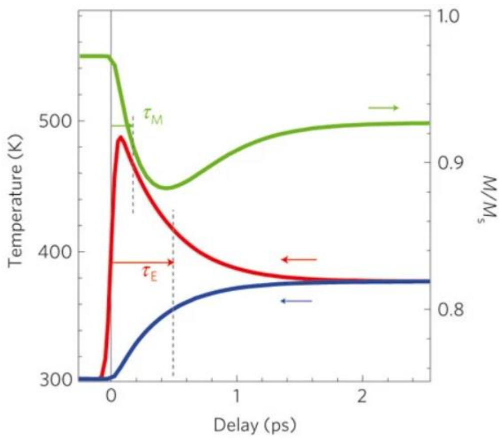
2D materials



Geim & Grigorieva, Nature 499, 419–425 (2013)

Ultrafast dynamics



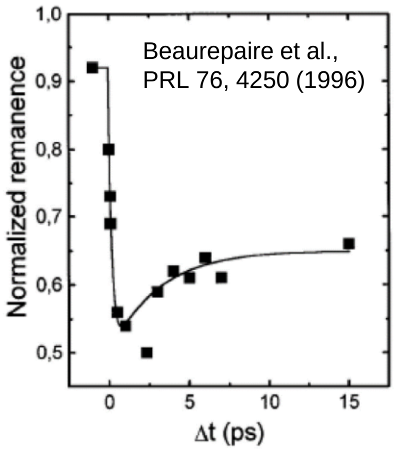


Kirilyuk, Rev. Mod. Phys. 82, 2731 (2010)

Koopmans Nat Mater 9, 259–265 (2010)

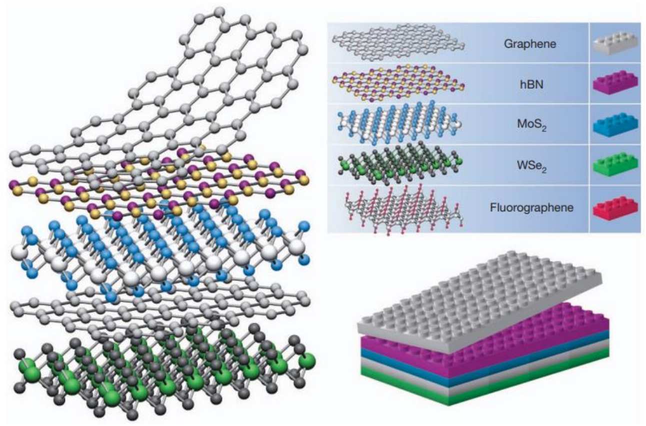
Why 2D magnets?

FM spin dynamics: very fast



~1 ns (1000 times slower)

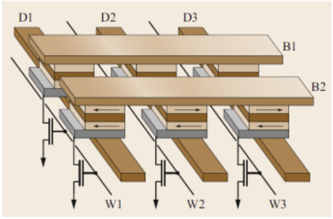
Van der Waal Heterostructures

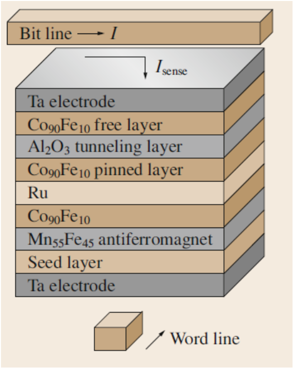


A. K. Geim and I. V Grigorieva, Nature **499**, 419 (2013).

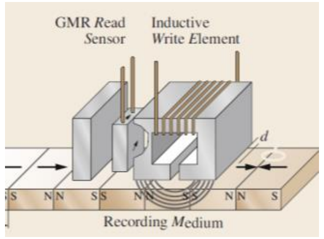
Applications

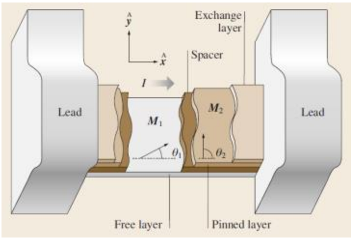
MRAM





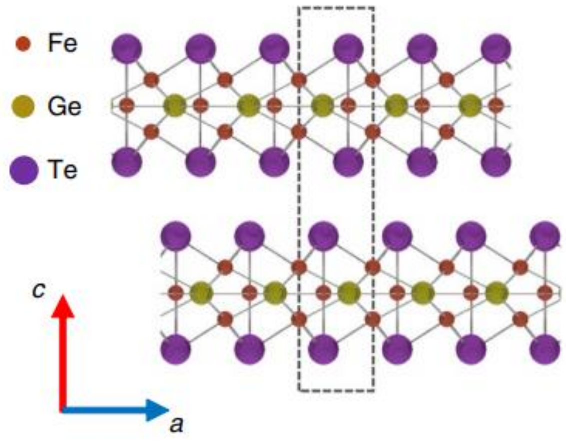
Hard disk Read/write head





Why Fe_2GeTe_3 (FGT)?

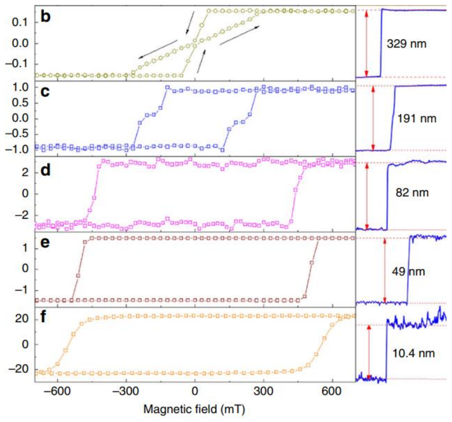
Van der Waals layered material



Fei, Nat. Mater. 17, 778–782 (2018)

Thickness dependent properties

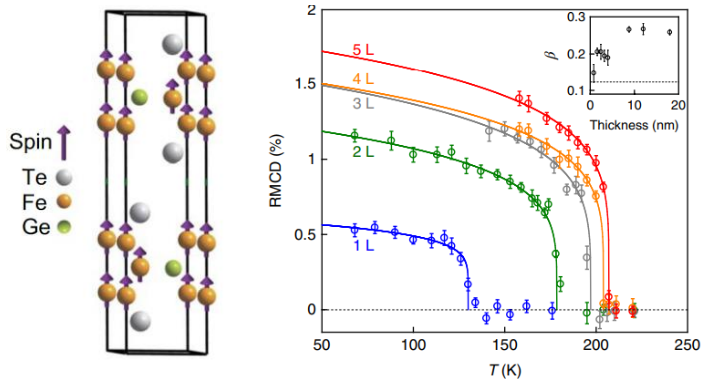
Control crystalline layer number



Tan, Nat Comm 9, 1554 (2018)

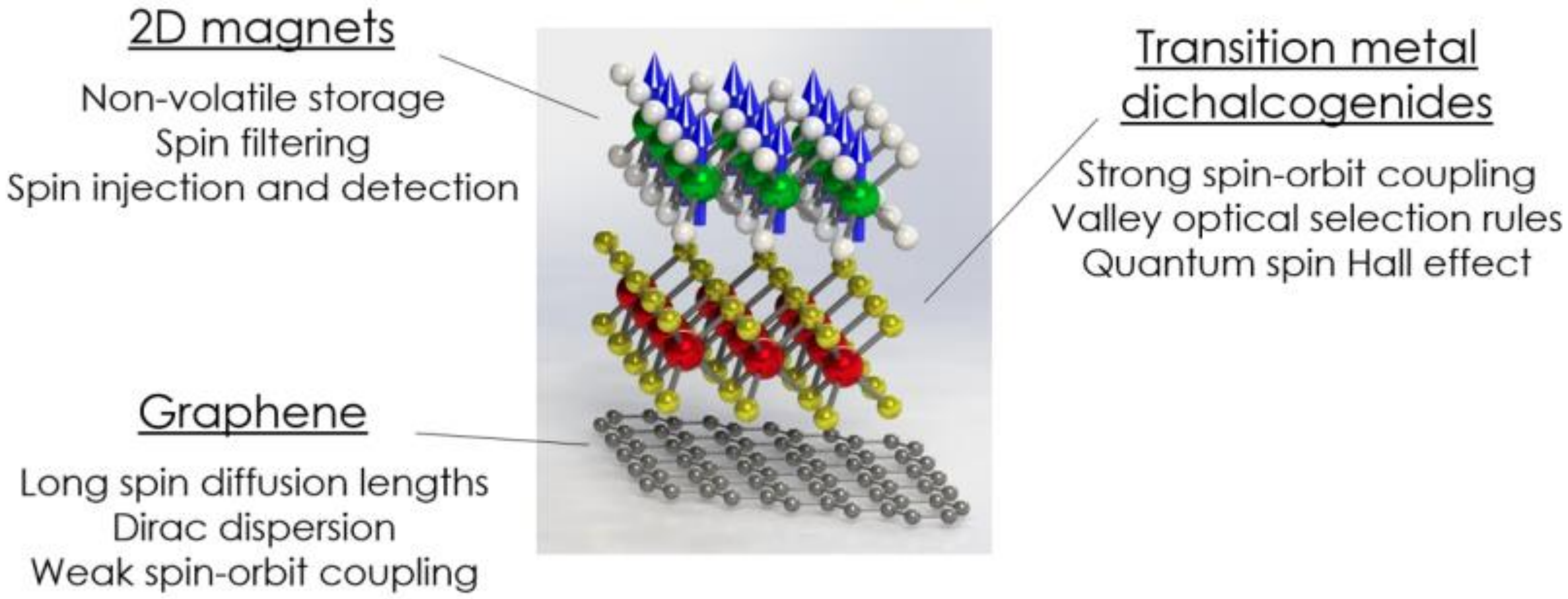
Metallic ferromagnet

Perpendicular anisotropy



Liu, PRL 125, 267205 (2020)

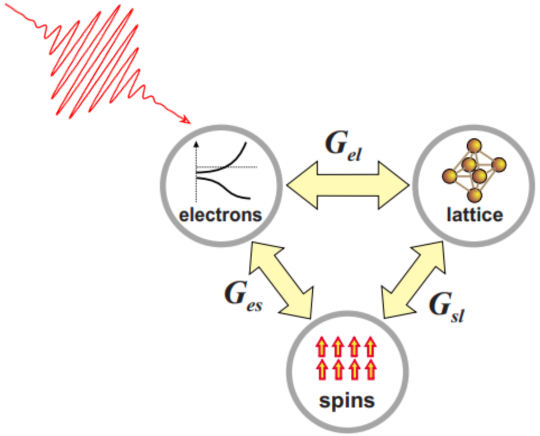
Vision: atomically flat, crystalline Heterostructures



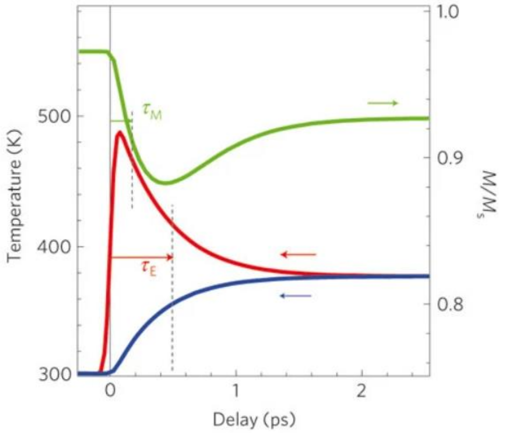
J. Phys. D: Appl. Phys. 53 (2020) 453001

Why ultrafast science?

1. Study nonequilibrium behavior
2. Study process at their intrinsic time scales
3. Understand **coupling** between intrinsic properties through **energy flow**



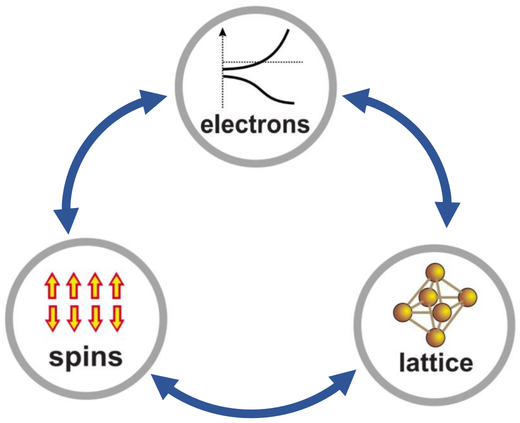
Kirilyuk, Rev. Mod. Phys. 82, 2731 (2010)



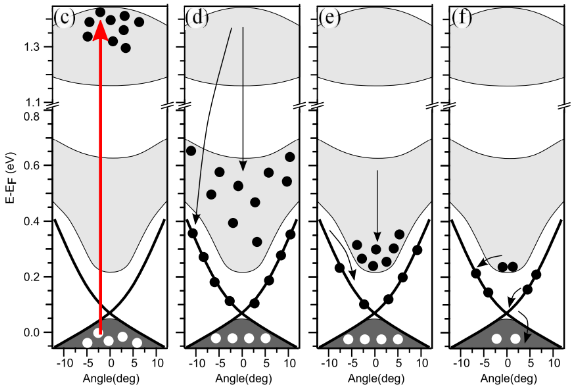
Koopmans Nat Mater 9, 259–265 (2010)

(my) three levels of understanding coupling

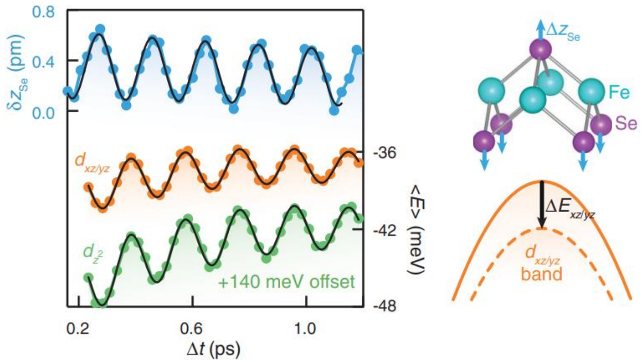
1. Coupled thermalized baths



2. Internal thermalization



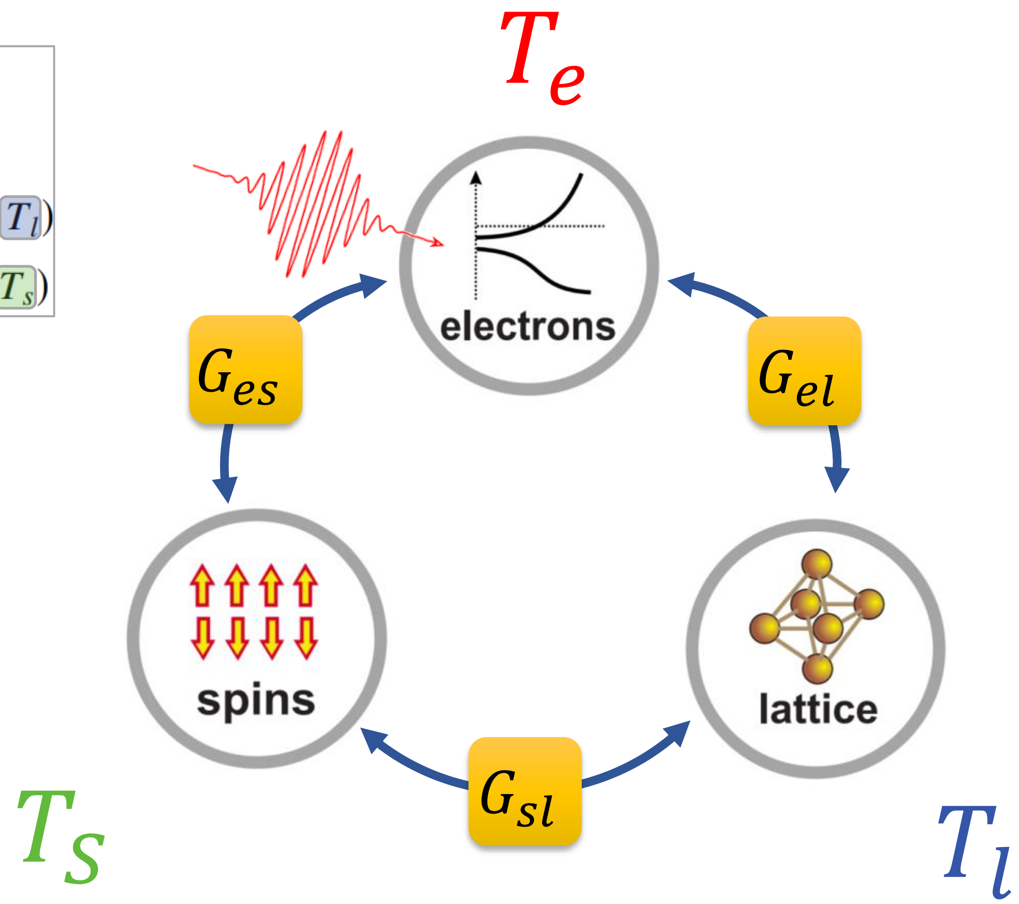
3. intra-DOF coupling

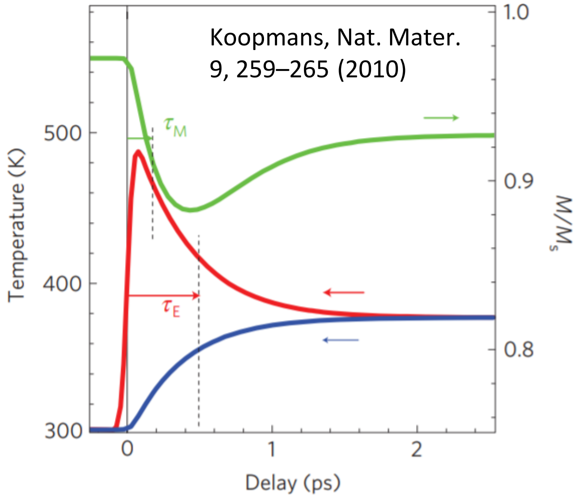


1. Coupled thermalized baths

Energy flow model

$$
\begin{aligned}
C_e(T_e)dT_e/dt &= -\,G_{el}(T_e - T_l) \\
&\quad -\,G_{es}(T_e - T_s) + P(t)\,, \\
C_s(T_s)dT_s/dt &= -\,G_{es}(T_s - T_e) - G_{sl}(T_s - T_l) \\
C_l(T_l)dT_l/dt &= -\,G_{el}(T_l - T_e) - G_{sl}(T_l - T_s)
\end{aligned}
$$





1. Coupled thermalized baths

Can we assume these baths are in thermal equilibrium?

Can we describe them with a temperature?

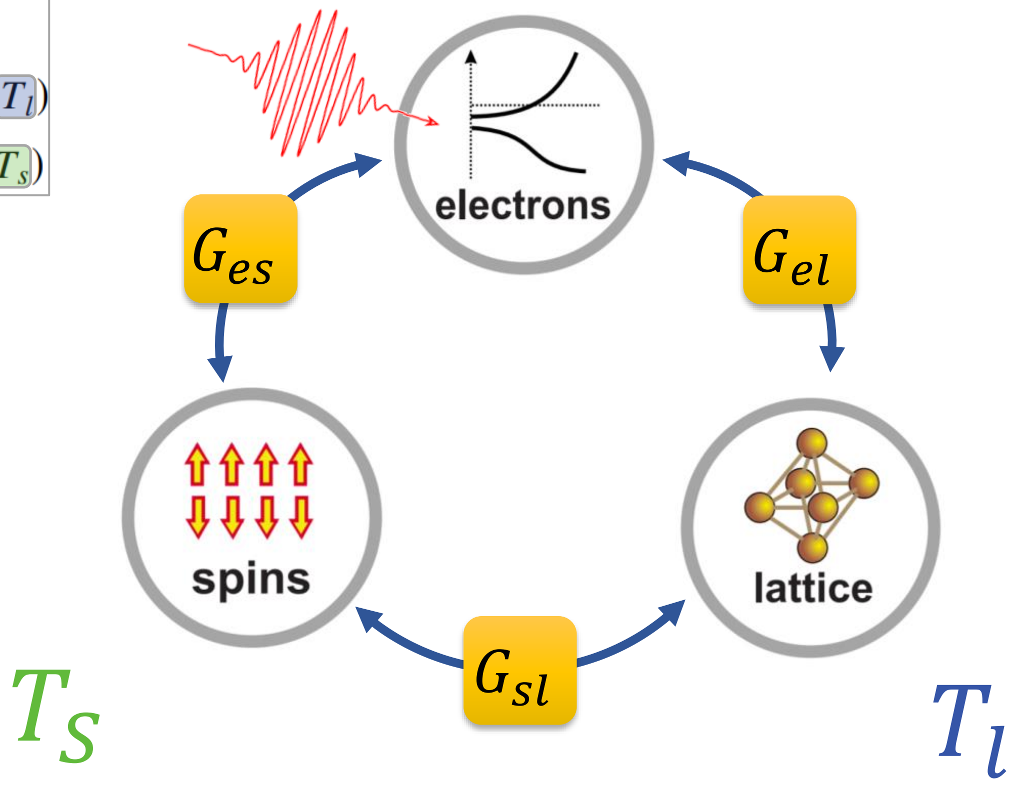
Energy flow

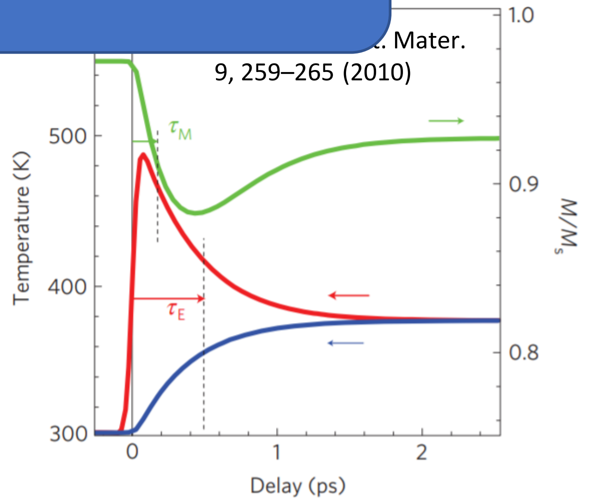
$$C_e(T_e)d\boxed{T_e}/dt \ldots$$

$$- G_{es}(\boxed{T_e} - \boxed{T_s}) + P(t)\,,$$

$$C_s(T_s)d\boxed{T_s}/dt = - G_{es}(\boxed{T_s} - \boxed{T_e}) - G_{sl}(\boxed{T_s} - \boxed{T_l})$$

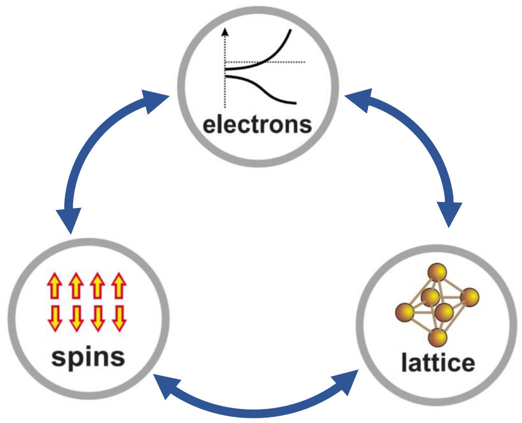
$$C_l(T_l)d\boxed{T_l}/dt = - G_{el}(\boxed{T_l} - \boxed{T_e}) - G_{sl}(\boxed{T_l} - \boxed{T_s})$$



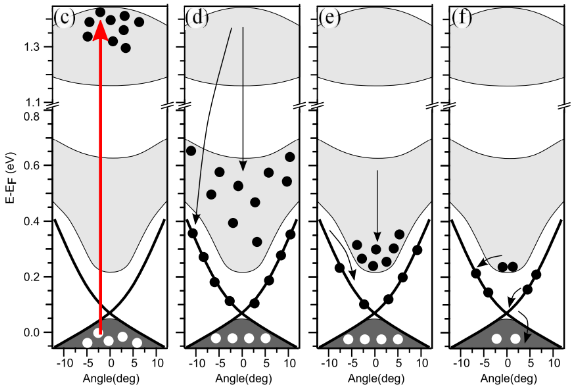


Three levels of understanding coupling

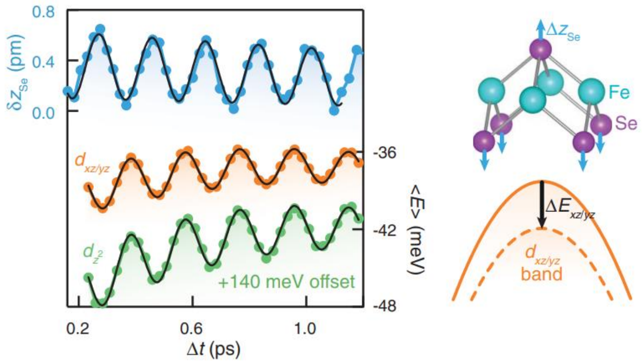
1. Coupled thermalized baths



2. Internal thermalization



3. intra-DOF coupling



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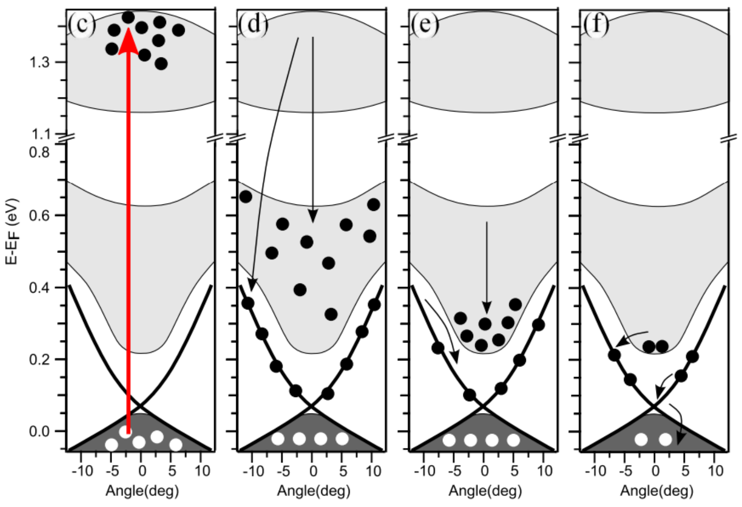
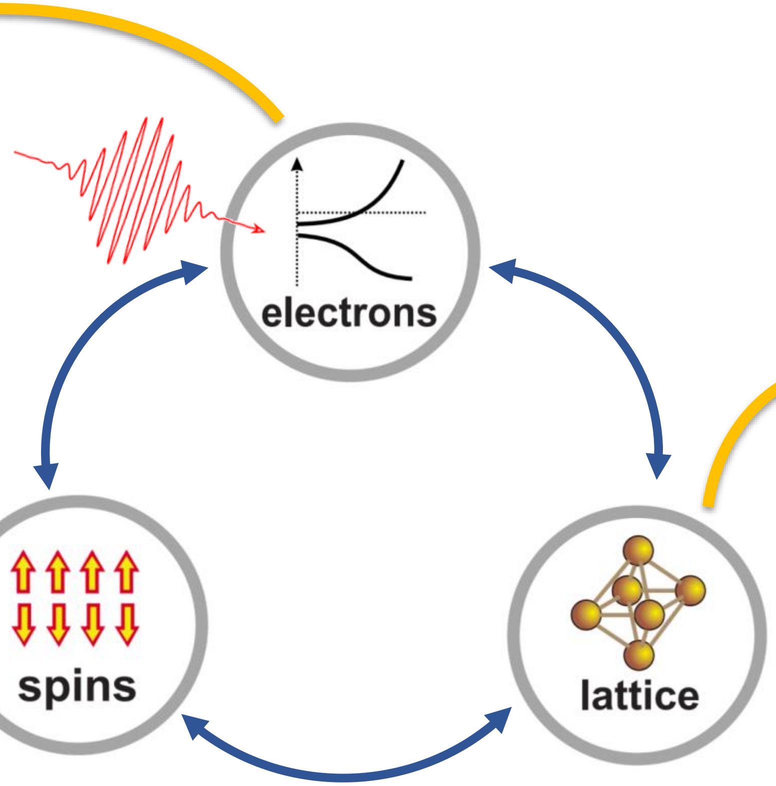
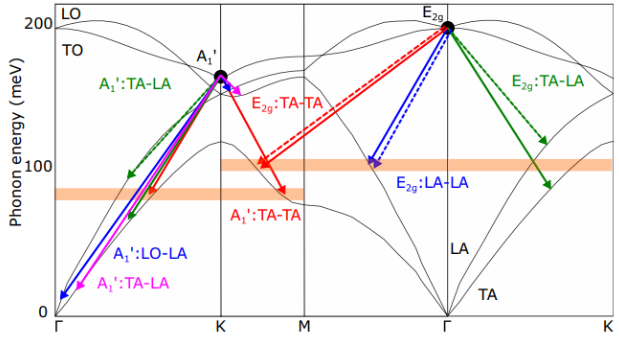


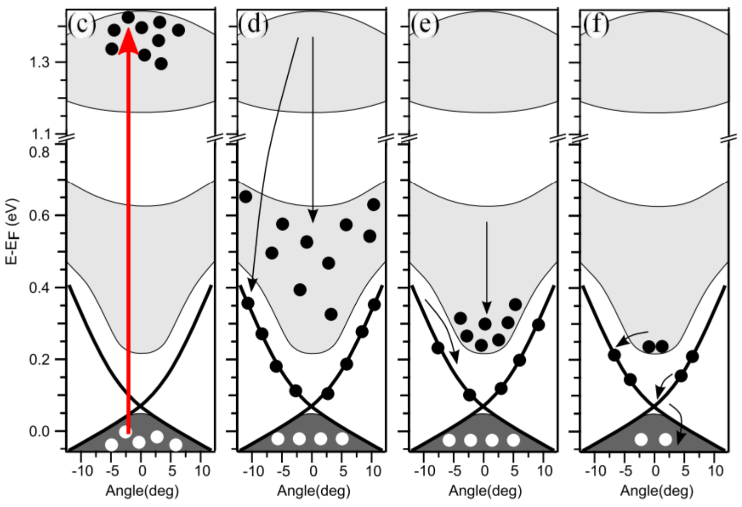
Sabota, PRL 108, 117403 (2012)



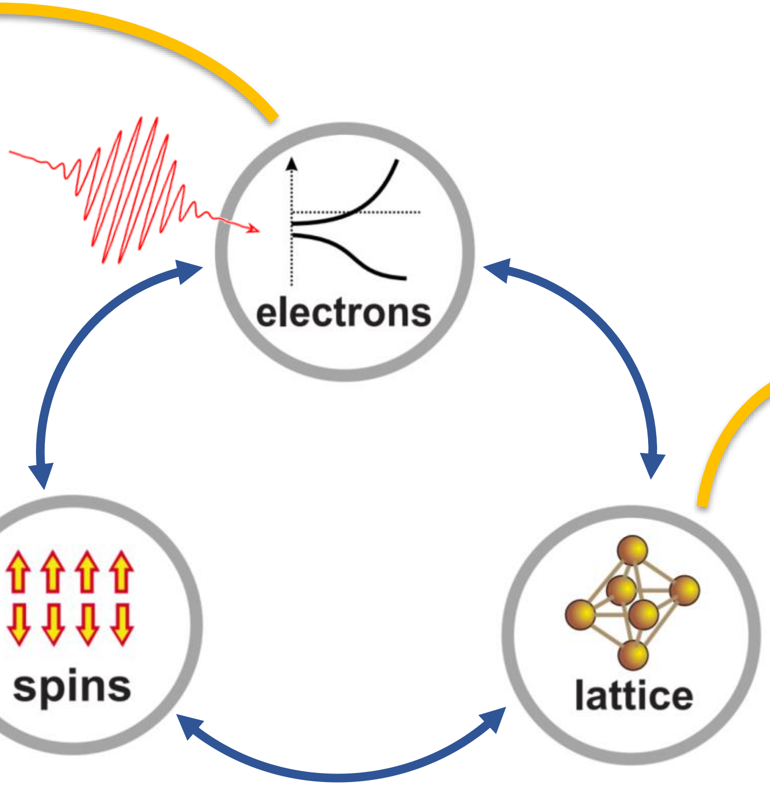


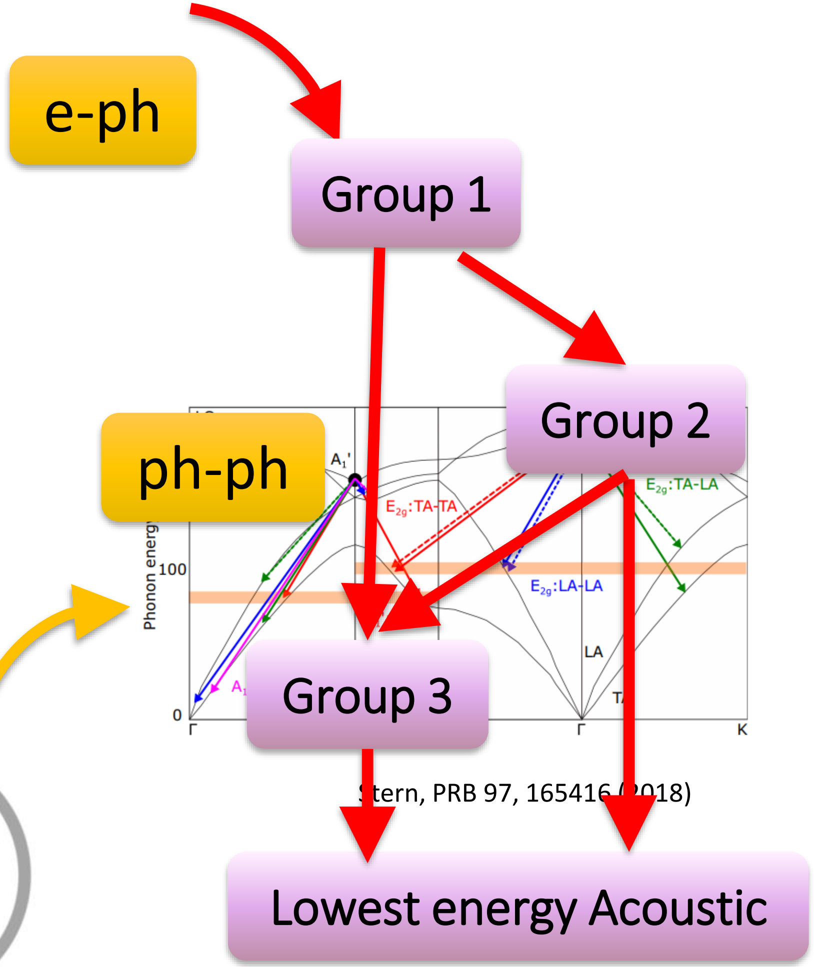
Stern, PRB 97, 165416 (2018)

2. Internal thermalization



Sabota, PRL 108, 117403 (2012)

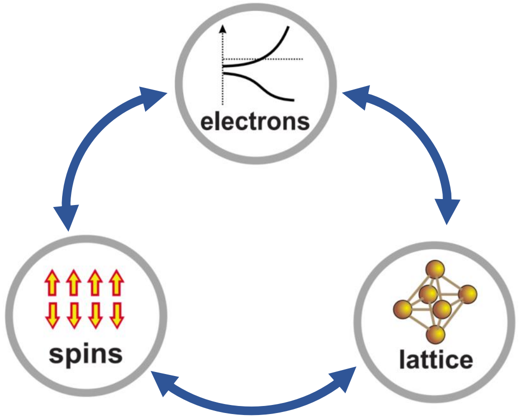




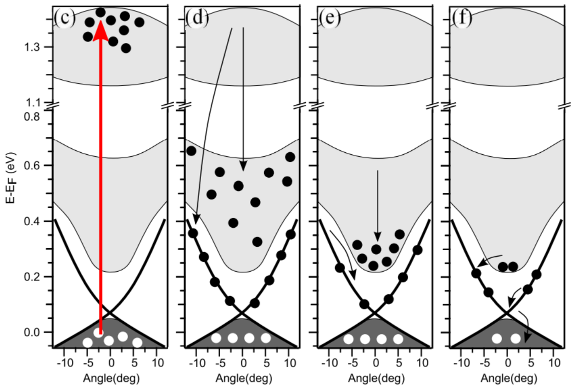
Stern, PRB 97, 165416 (2018)

Three levels of understanding coupling

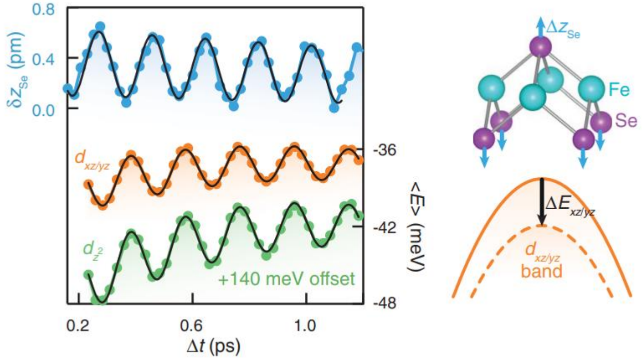
1. Coupled thermalized baths



2. Internal thermalization



3. intra-DOF coupling



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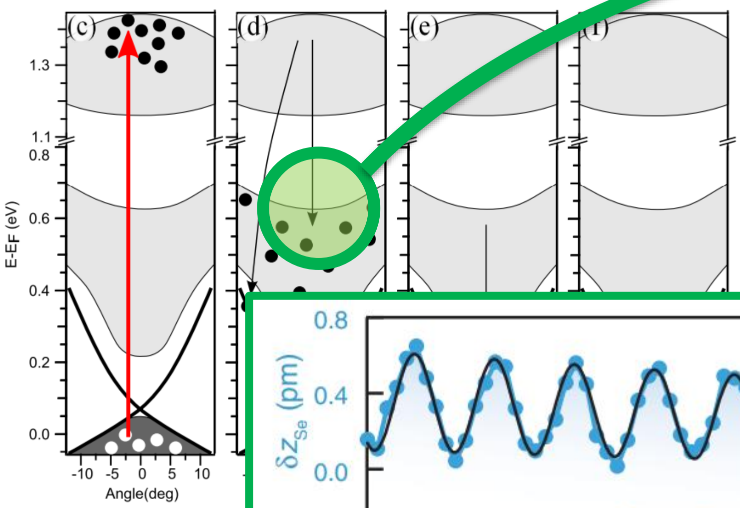
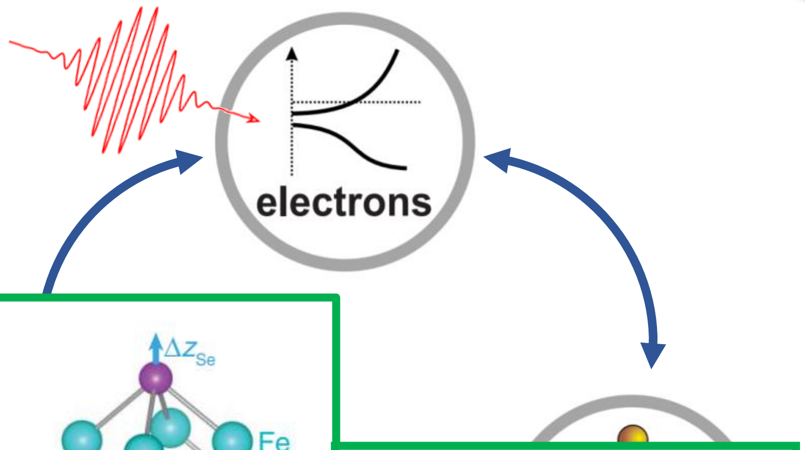
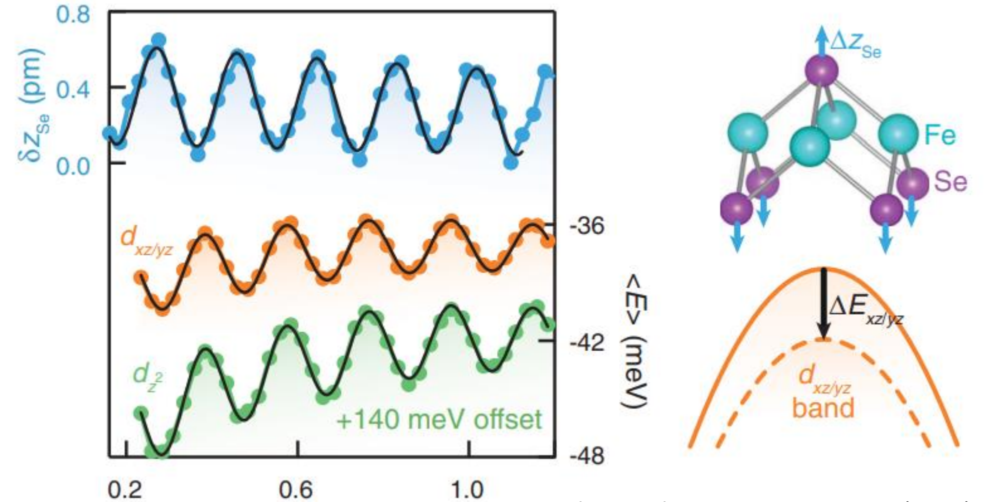


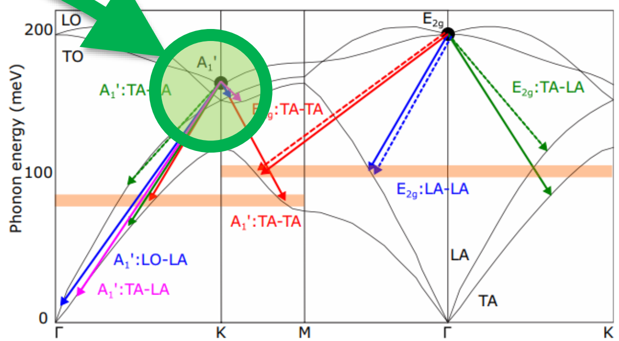
Sabota, PRL 108, 1





Gerber et al., Science 357, 71–75 (2017)

$$\frac{\Delta E}{\Delta Z}$$ “coupling potential”

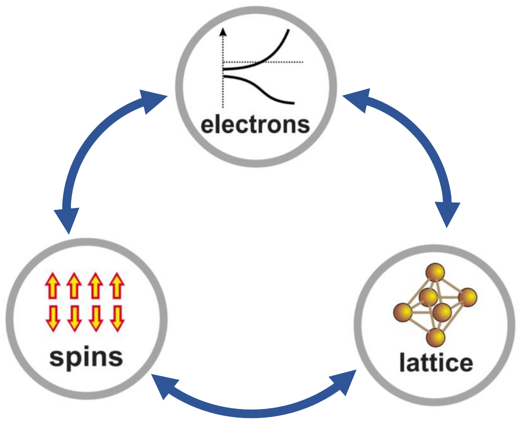


Stern, PRB 97, 165416 (2018)

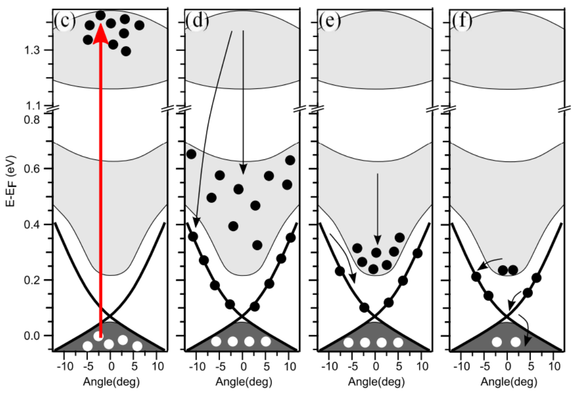
$BaFe_2As_2$
Yang, PRL 112, 207001 (2014)
Rettig PRL 114, 067402 (2015)

Understanding dynamics in FGT

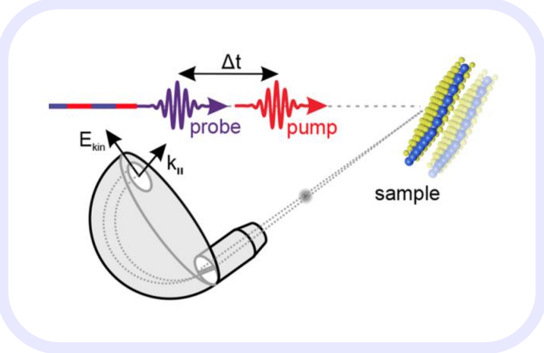
1. Coupled thermalized baths



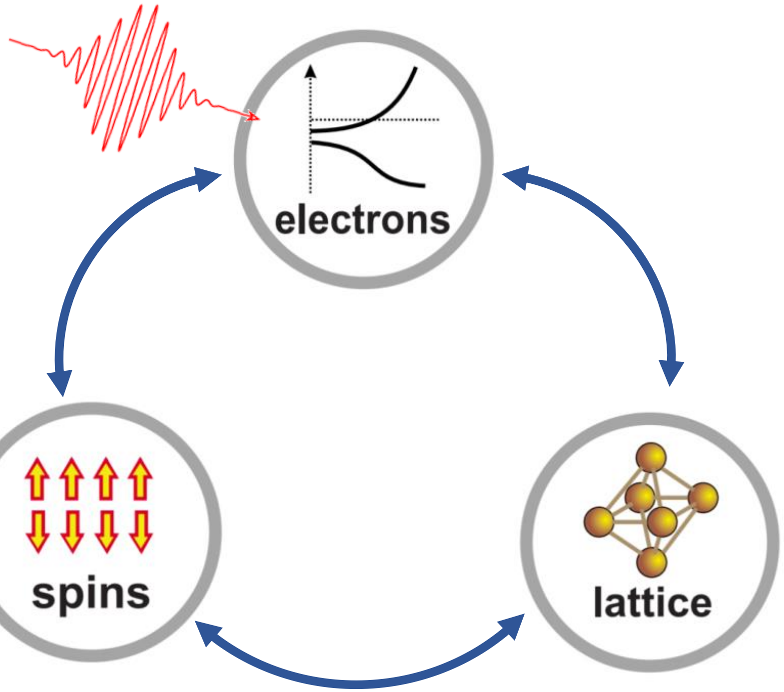
2. Internal thermalization



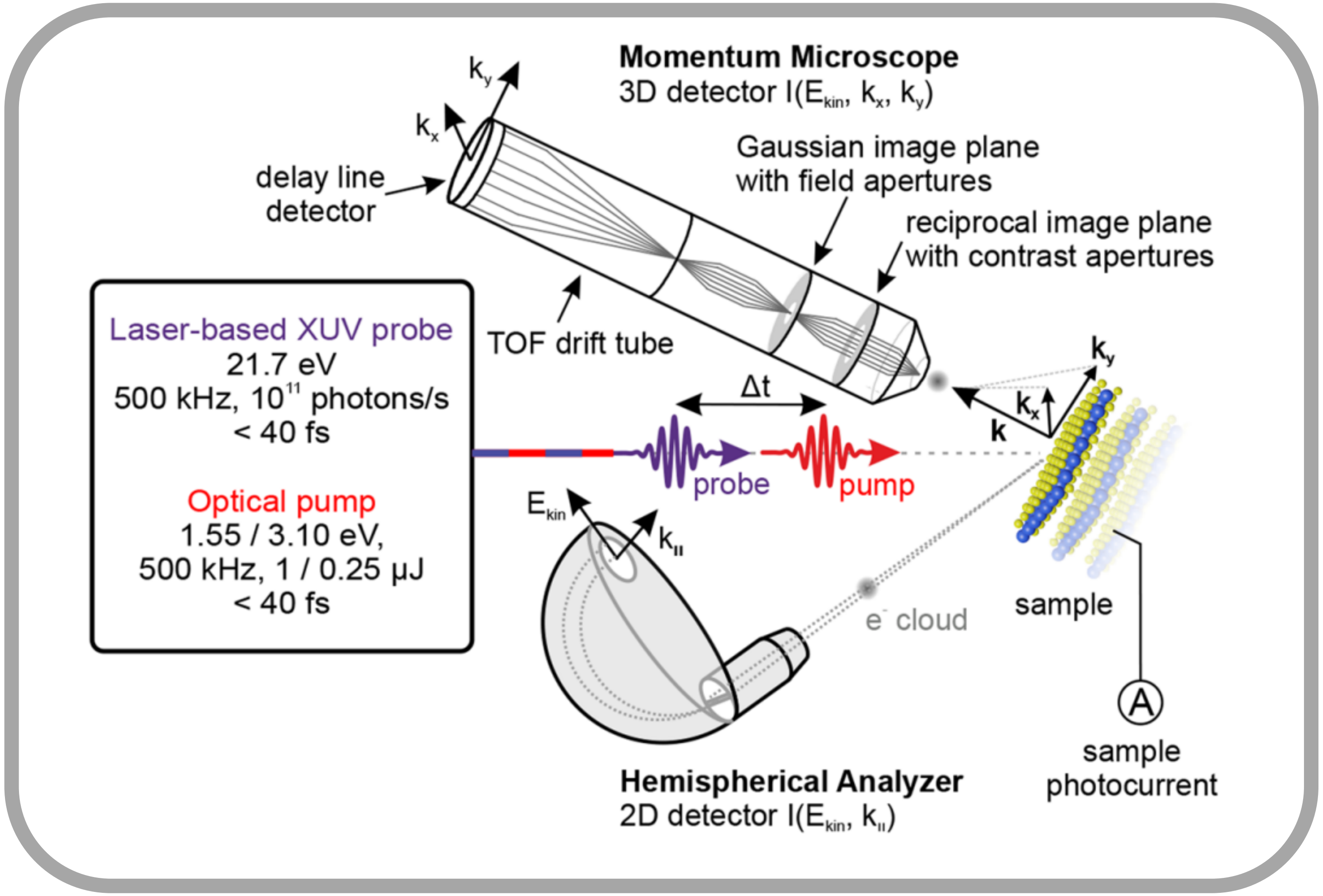
3. intra-DOF coupling



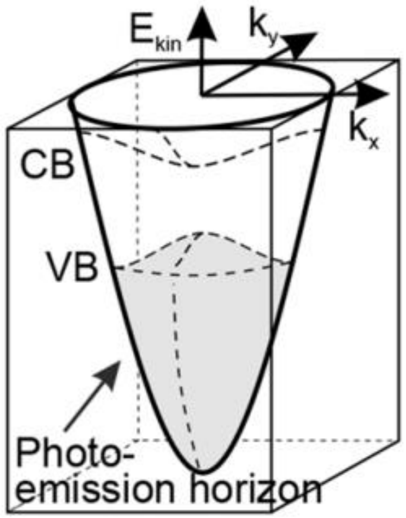
time resolved
ARPES

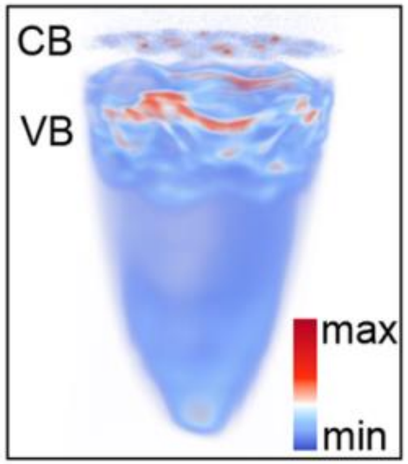


Time resolved ARPES at FHI



4-dimentional data (k_x, k_y,E, Δt)

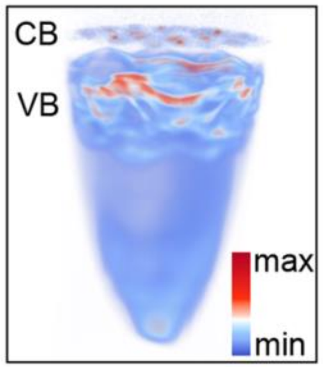


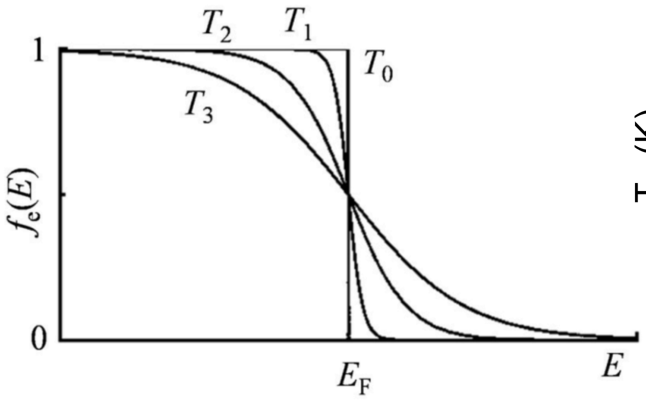


Maklar et al, Rev. Sci. Instr. 91, 123112 (2020)

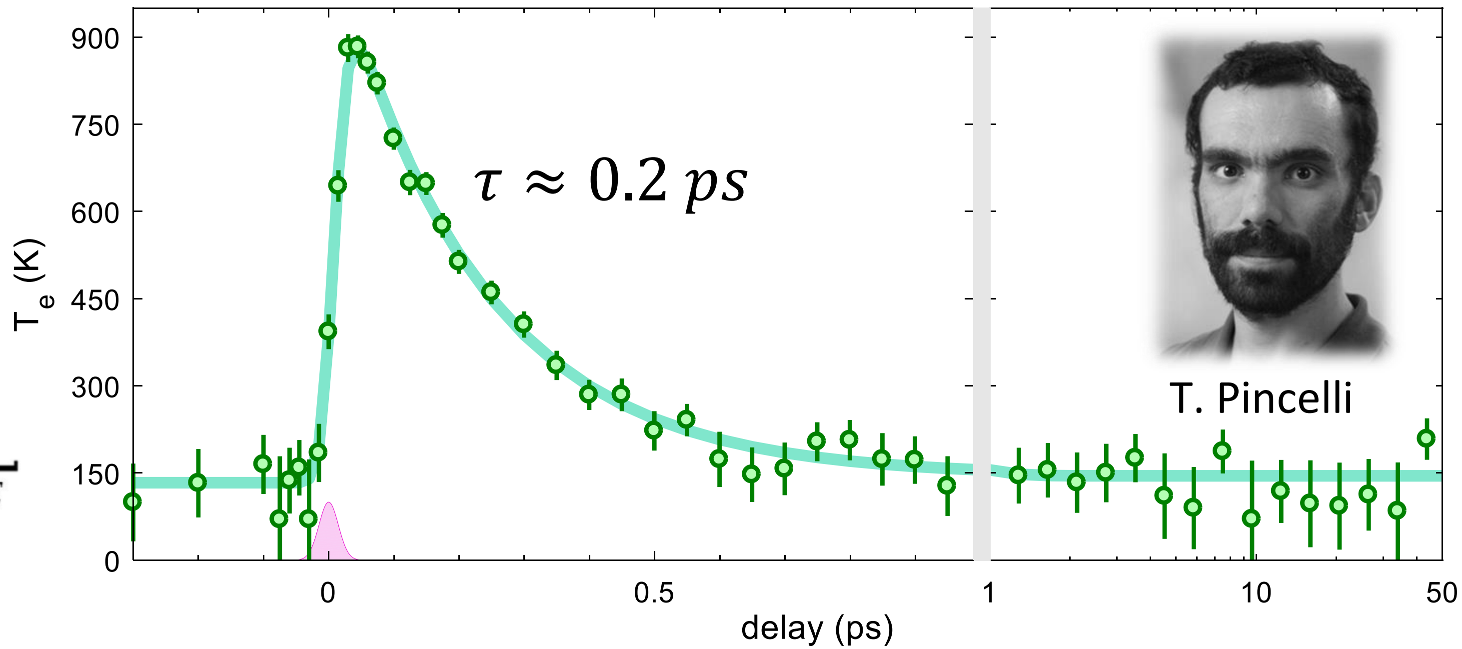
Electron temperature dynamics

$$I(\boldsymbol{k},\omega) = |M(\boldsymbol{k},\omega)|^2 f_{FD}(\omega) A(\boldsymbol{k},\omega)$$

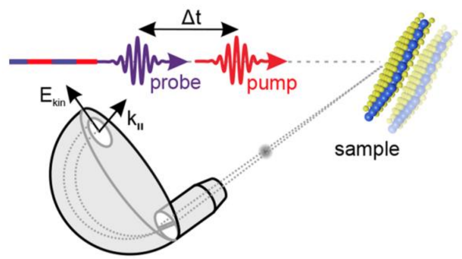




Electronic temperature

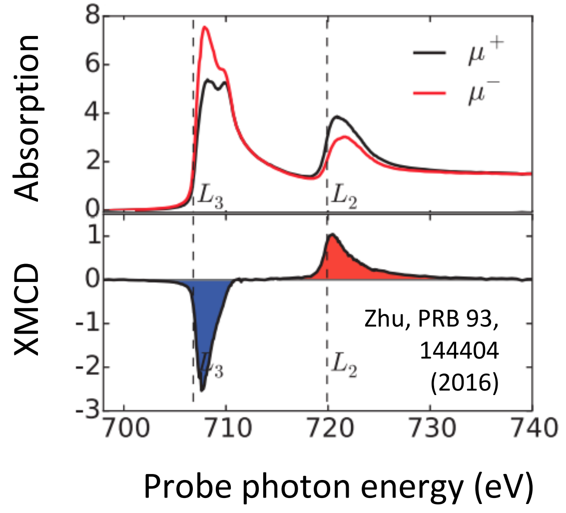


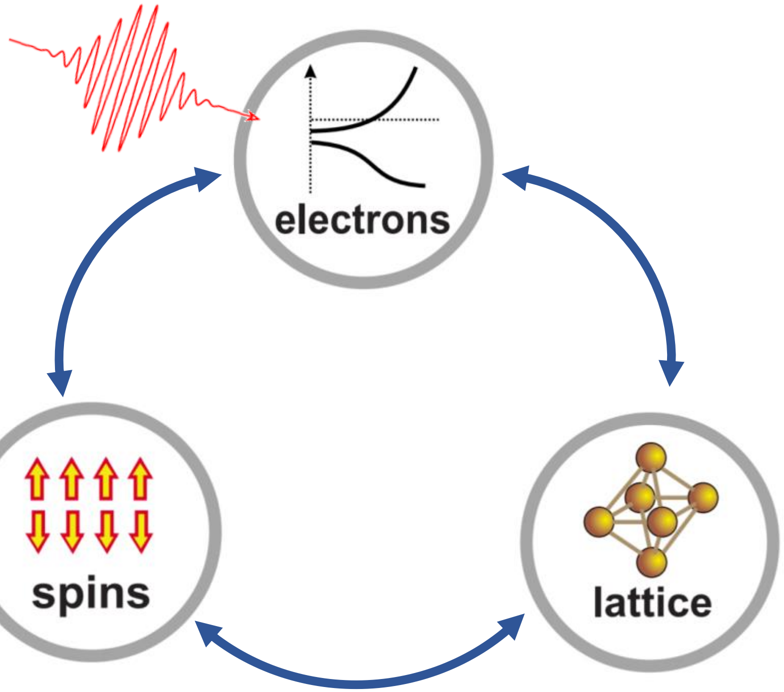
Pump: 800 nm
Probe: 21 eV (HHG XUV)
Resolution: ~35 fs



time resolved ARPES

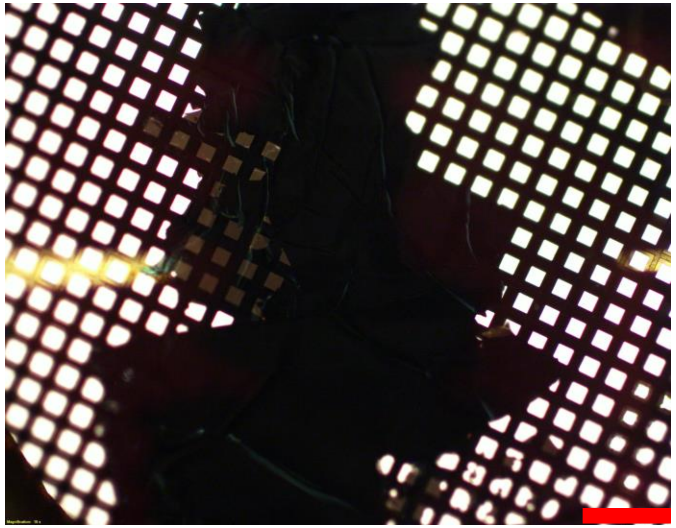
Ultrafast XMCD





XMCD from FGT in transmission

free standing FGT crystal (50 nm)



200 μm

FEMTOSPE

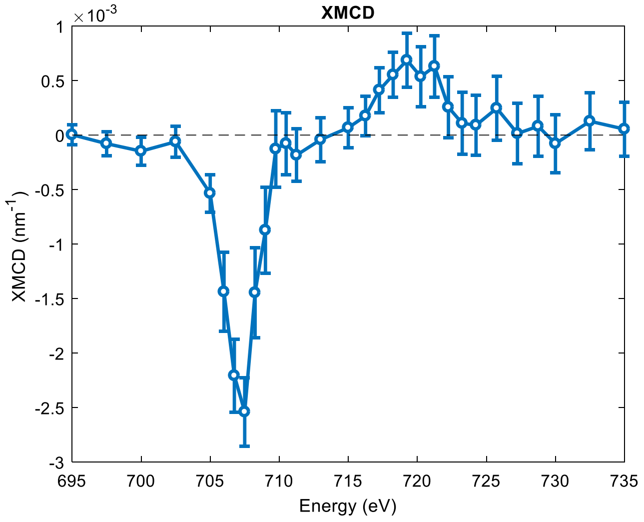
Pump: 800 nm
Probe: 707 eV (Fe L_3)
Resolution: ~120 fs

Ch. Schüßler-Langenheine

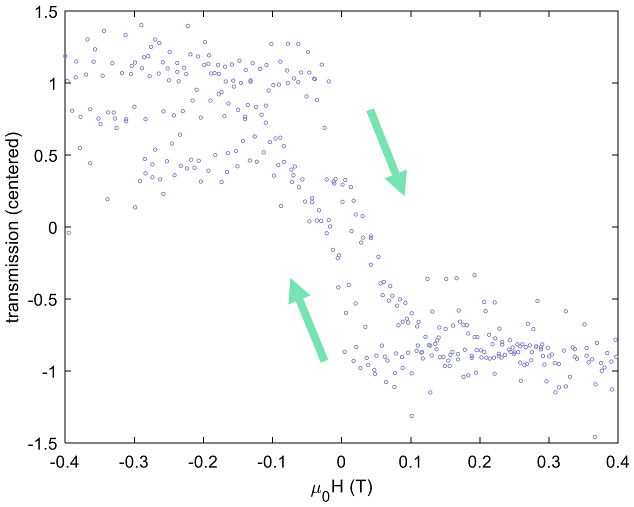
N. Pontius

XMCD results

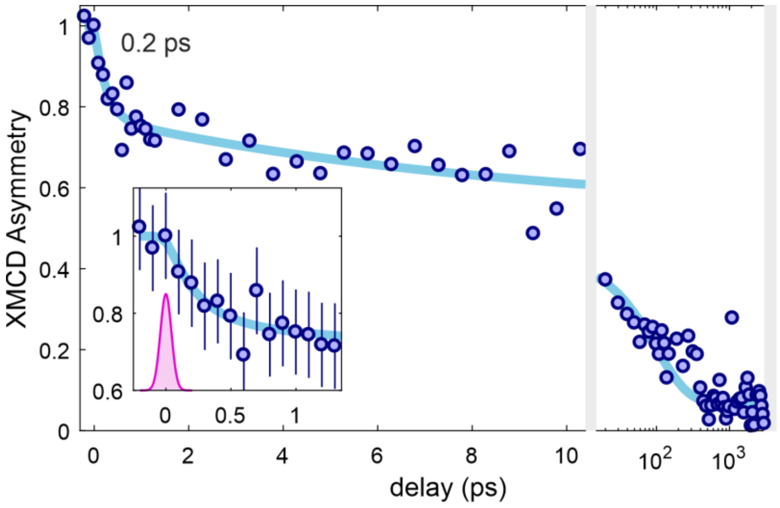
XMCD
(real units)



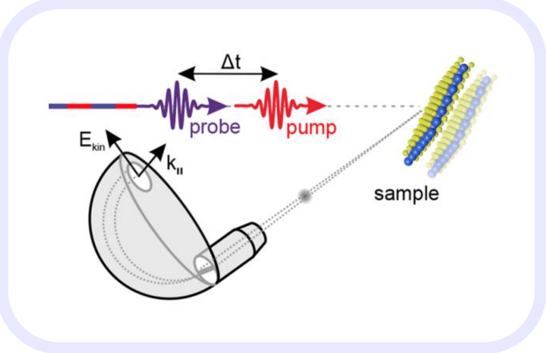
Hysteresis
(magnetic as expected)



Pump effect
(jump at axis break: artifact)

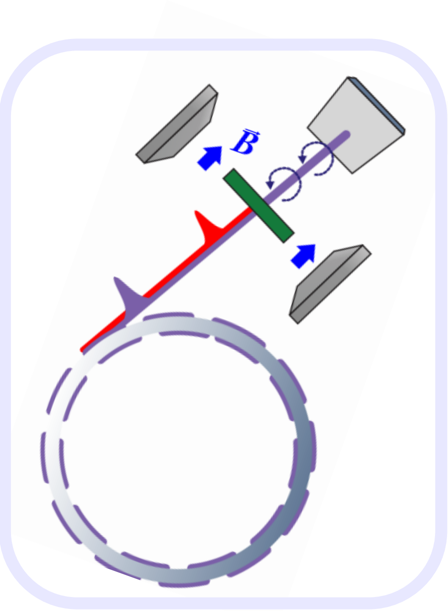


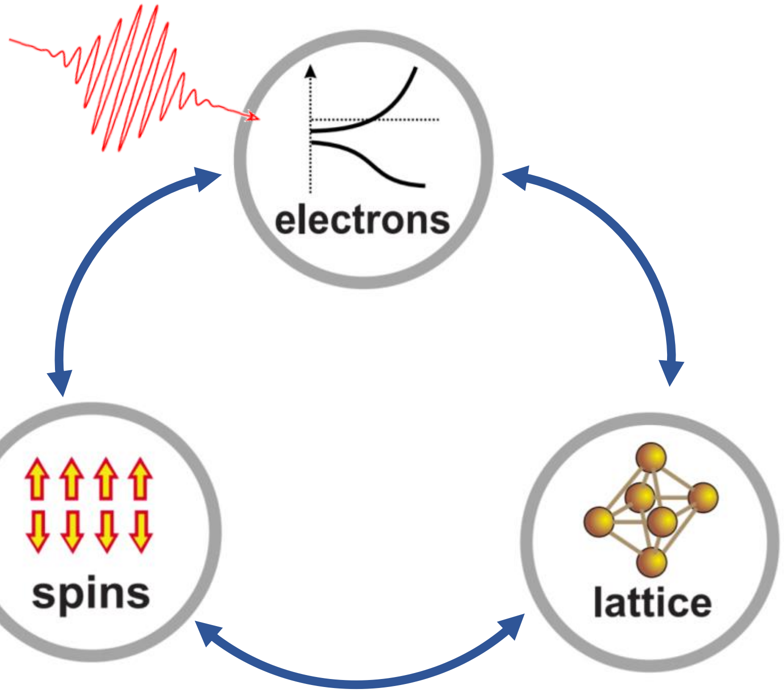
Spin dynamics time scale very similar to electron dynamics



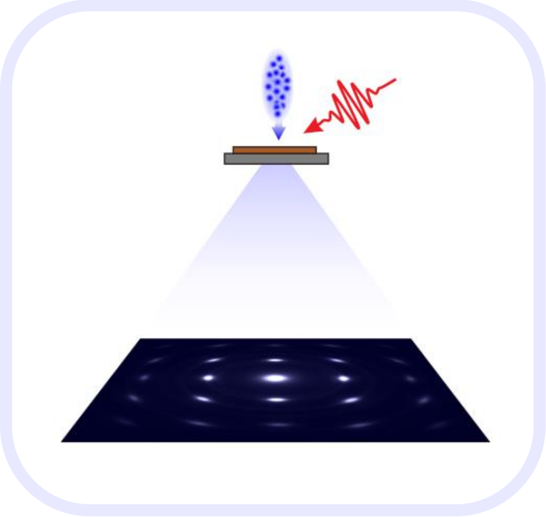
time resolved ARPES

Ultrafast XMCD

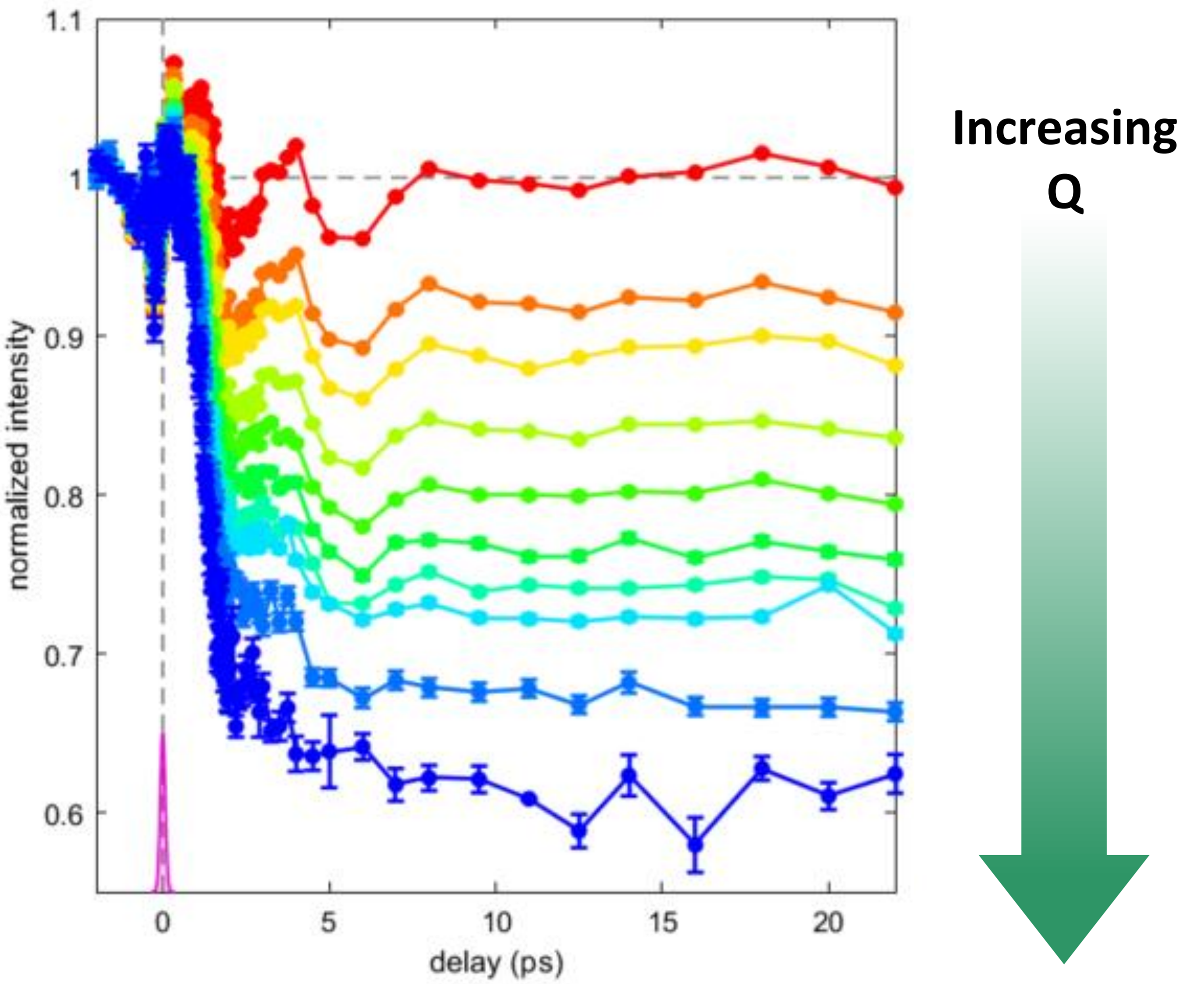


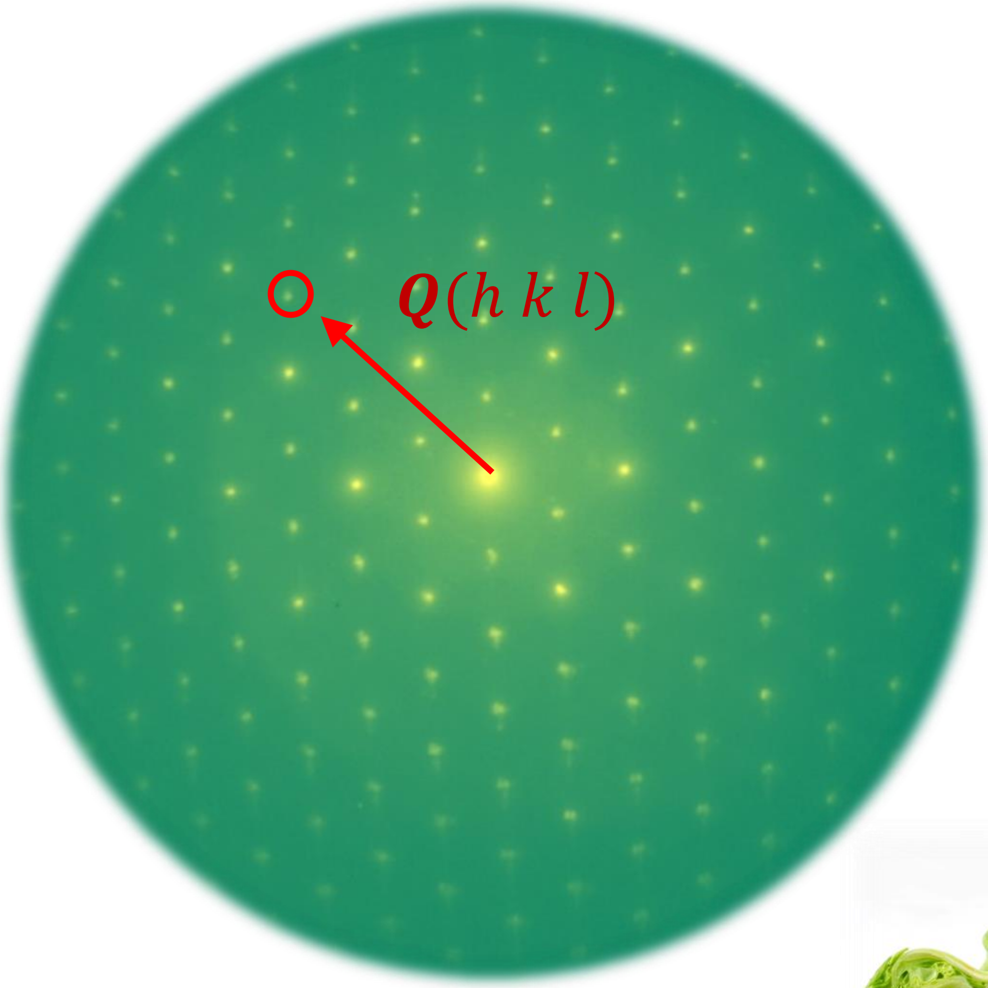


Femtosecond Electron Diffraction



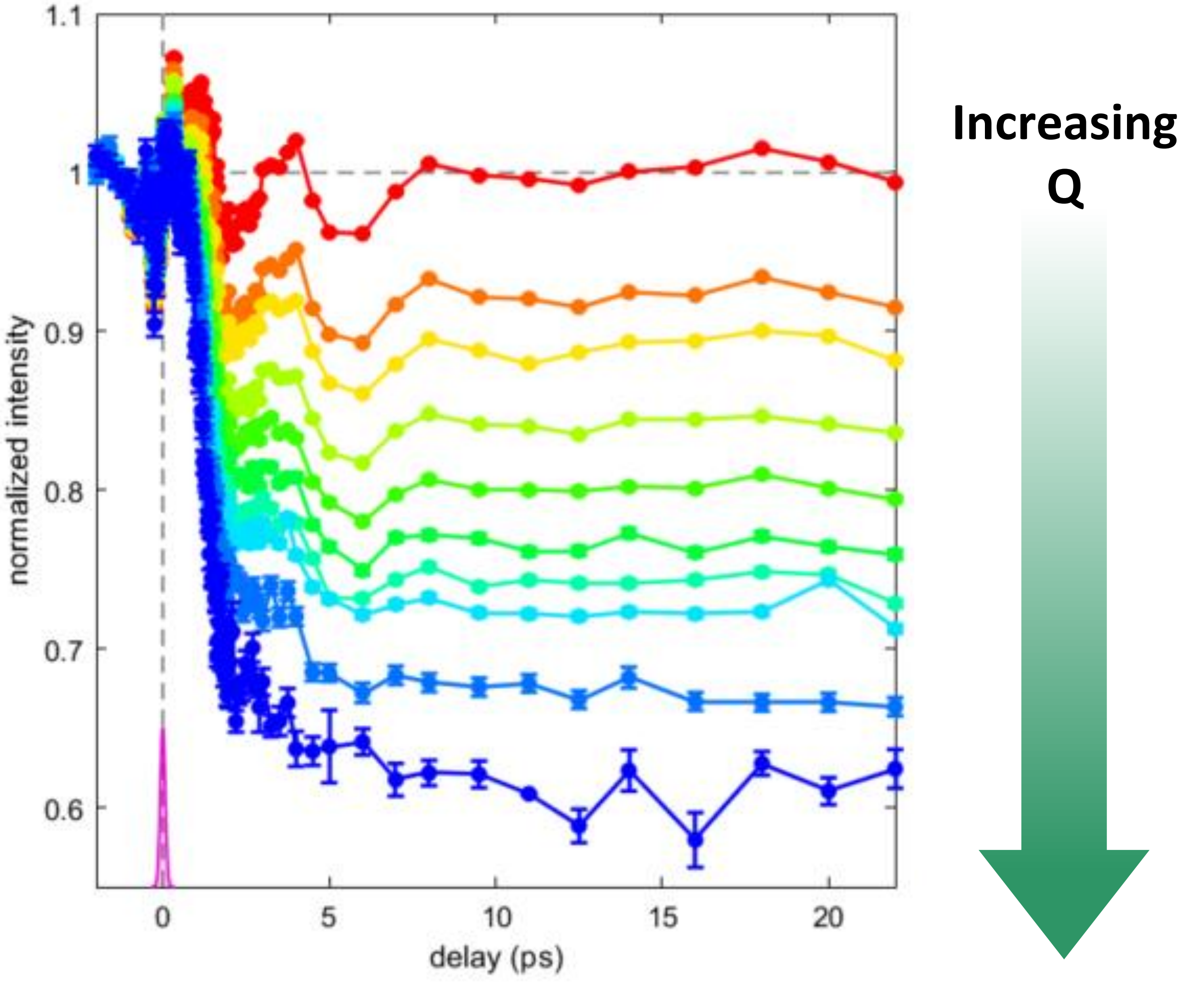
Lattice dynamics

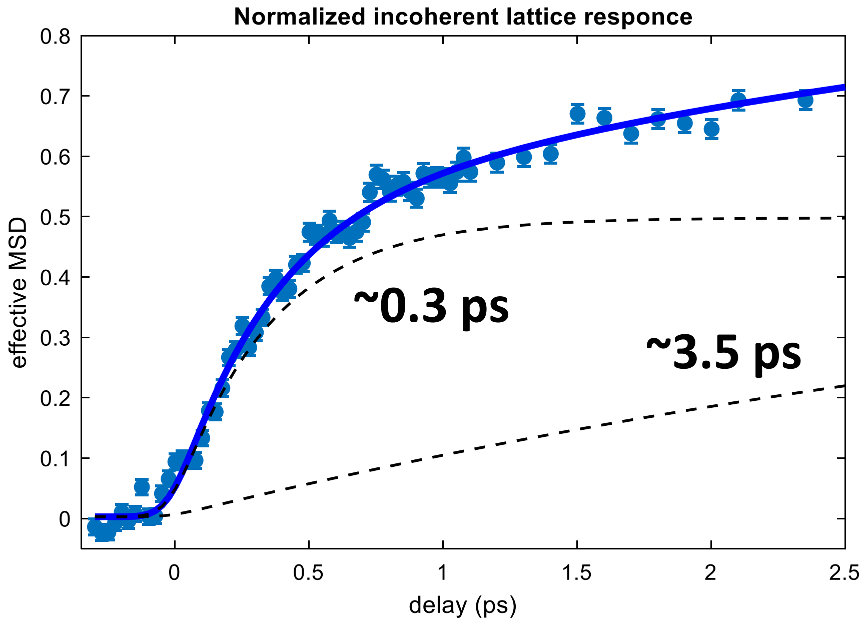


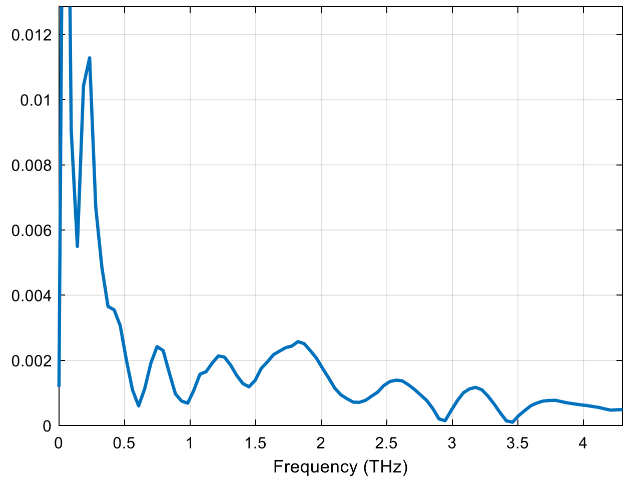


470 peaks, 56 unique $|Q|$

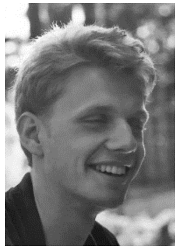
Lattice dynamics



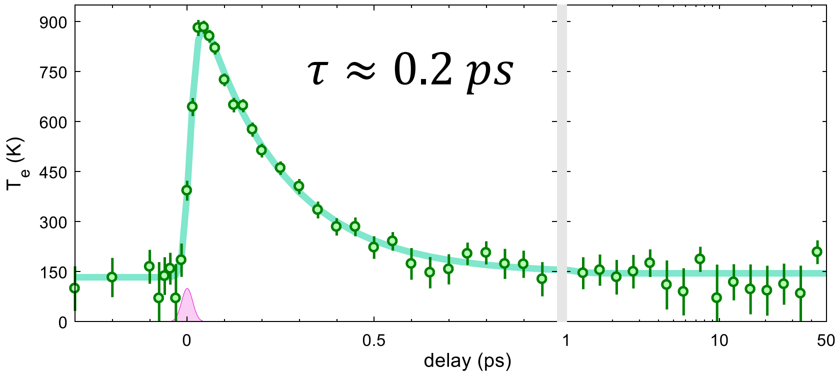




Theoretical modeling

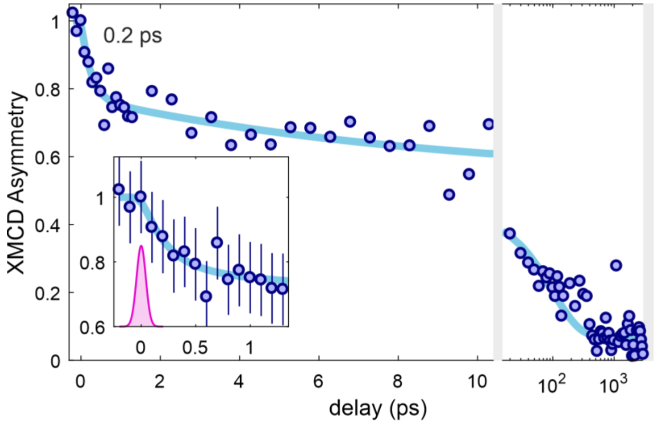



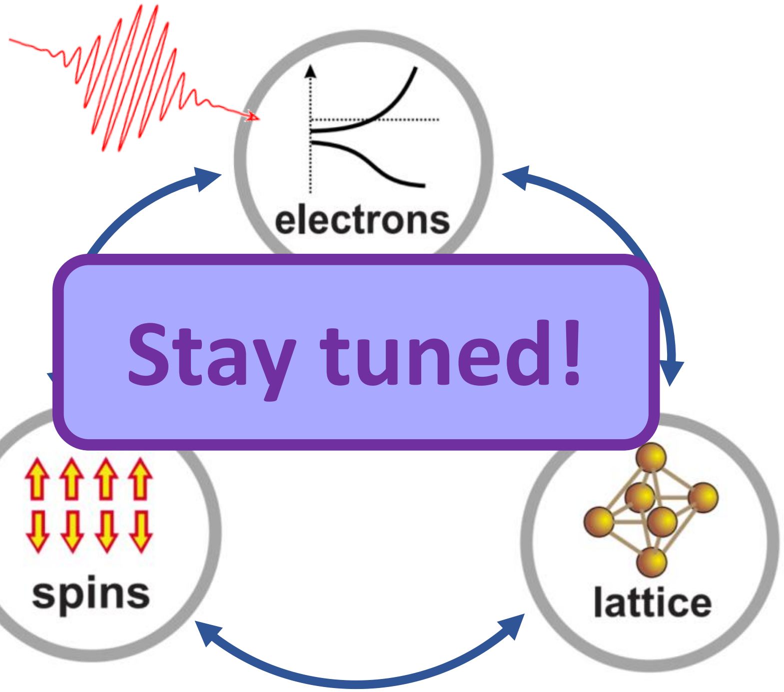
U. Atxitia **T. Griepe**



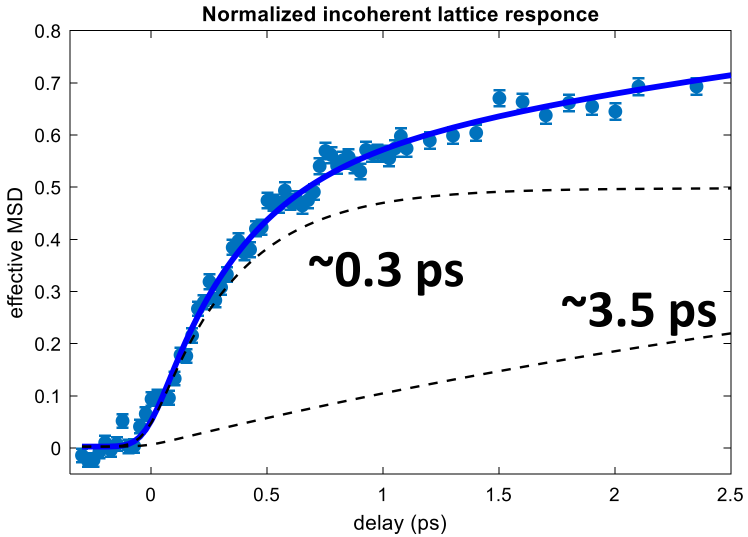
time resolved ARPES

Ultrafast XMCD



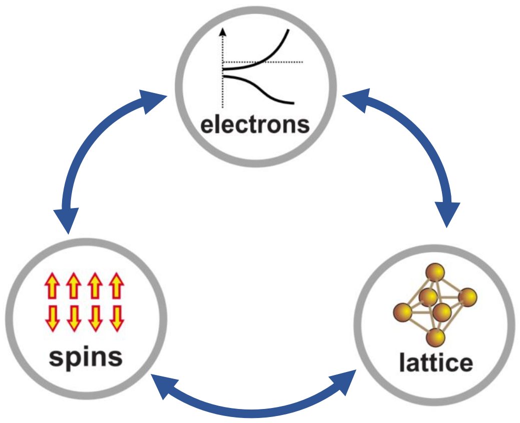


Femtosecond Electron Diffraction

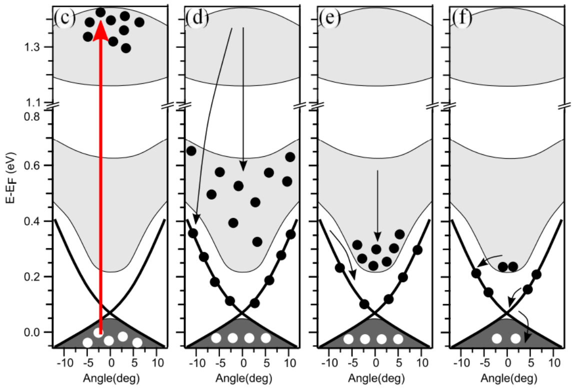


Understanding dynamics in FGT

1. Coupled thermalized baths

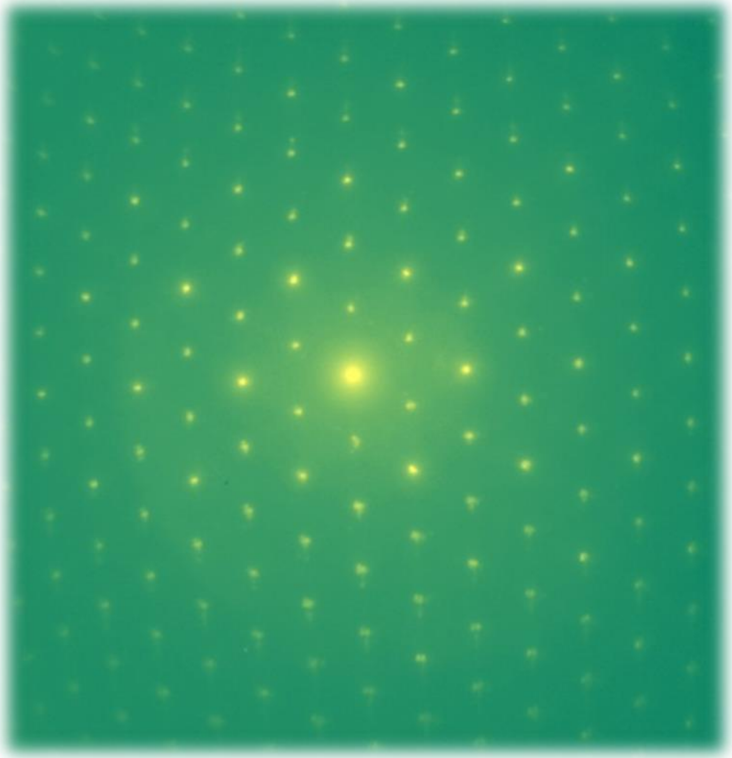


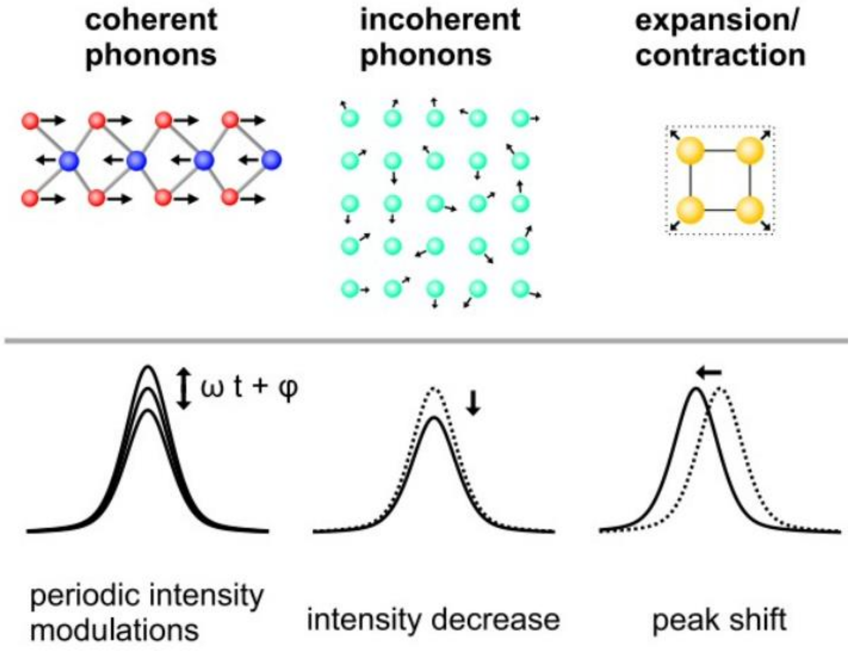
2. Internal thermalization



3. intra-DOF coupling

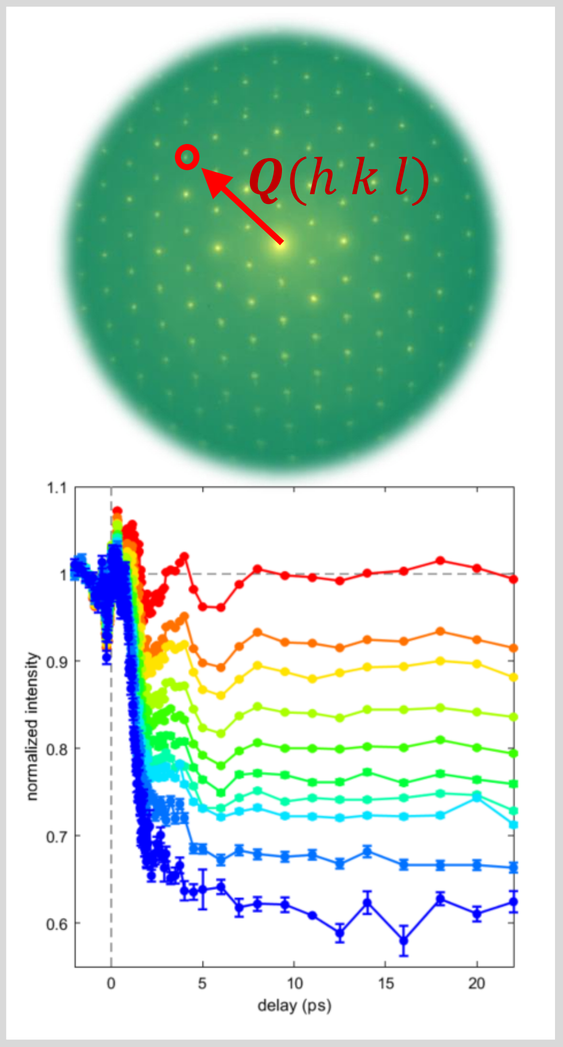
Diffraction



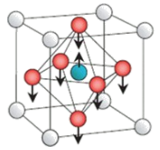


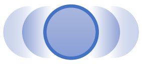
D. Zahn
Phd thesis

Evolution of the phonon system



$$I(Q) \propto \left| \sum_i \overbrace{f_i(Q)}^{\text{Scattering factors (tabulated)}} \overbrace{e^{2\pi i \mathbf{Q}\cdot \mathbf{r}_i}}^{\text{phase factors}} \overbrace{e^{-\frac{4\pi^2}{3}\langle u_i^2 \rangle Q^2}}^{\text{Temperature factor (Debye Waller effect)}} \right|^2$$

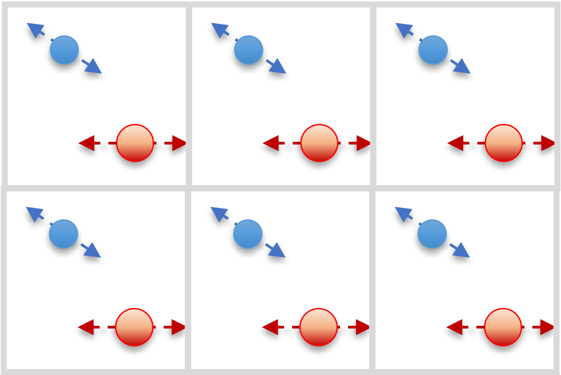




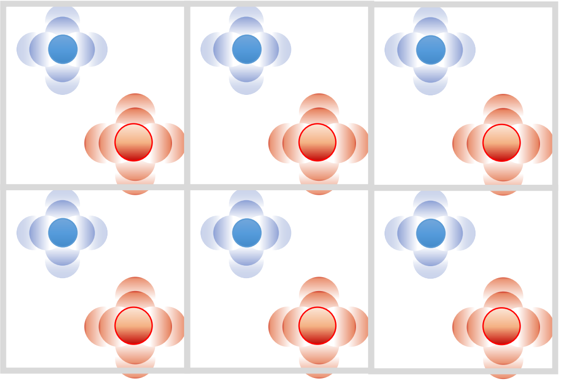
Changes intensity

Reduces intensity

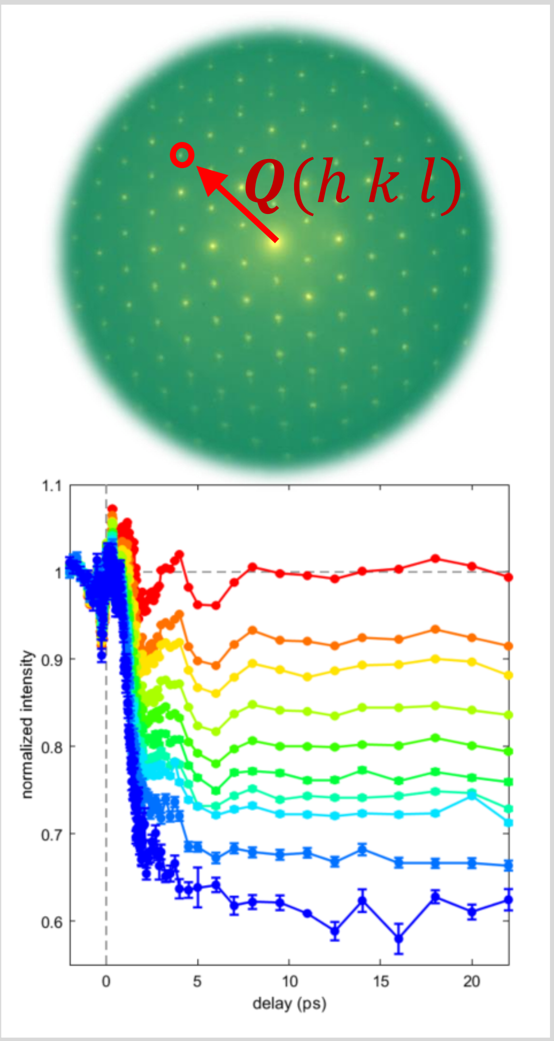
Coherent phonons



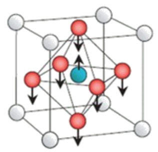
Incoherent phonons



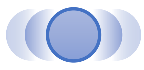
Evolution of the phonon system



$$I(Q) \propto \left| \sum_i \overbrace{f_i(Q)}^{\text{Scattering factors (tabulated)}} \overbrace{e^{2\pi i \boldsymbol{Q}\cdot\boldsymbol{r}_i}}^{\text{phase factors}} \overbrace{e^{-\frac{4\pi^2}{3}\langle u_i^2\rangle Q^2}}^{\text{Temperature factor (Debye Waller effect)}} \right|^2$$



Changes intensity

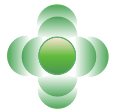


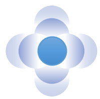
Reduces intensity

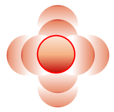
Fe: $\boldsymbol{r}_i(t) = \boldsymbol{r}_i^0 + \boldsymbol{\Delta}(t)$

Ge: $\boldsymbol{r}_i(t) = \boldsymbol{r}_i^0 + \boldsymbol{\Delta}(t)$

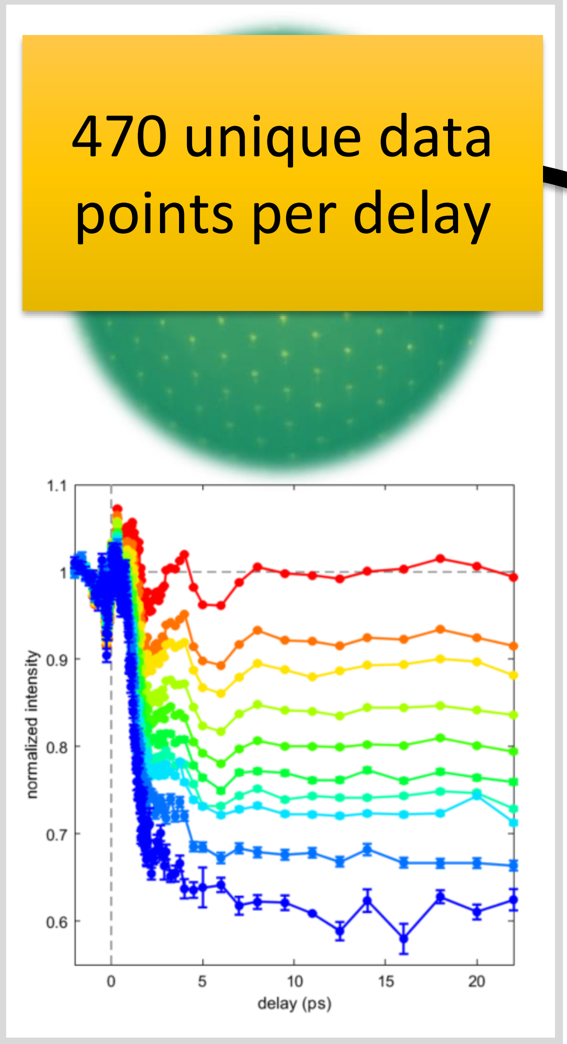
Te: $\boldsymbol{r}_i(t) = \boldsymbol{r}_i^0 + \boldsymbol{\Delta}(t)$

Fe:  $\begin{pmatrix} U_{Fe} & 2U_{Fe} & 0 \\ 2U_{Fe} & U_{Fe} & 0 \\ 0 & 0 & \cancel{U'_{Fe}} \end{pmatrix}$

Ge:  $\begin{pmatrix} U_{Ge} & 2U_{Ge} & 0 \\ 2U_{Ge} & U_{Ge} & 0 \\ 0 & 0 & \cancel{U'_{Ge}} \end{pmatrix}$

Te:  $\begin{pmatrix} U_{Te} & 2U_{Te} & 0 \\ 2U_{Te} & U_{Te} & 0 \\ 0 & 0 & \cancel{U'_{Te}} \end{pmatrix}$

Evolution of the phonon system

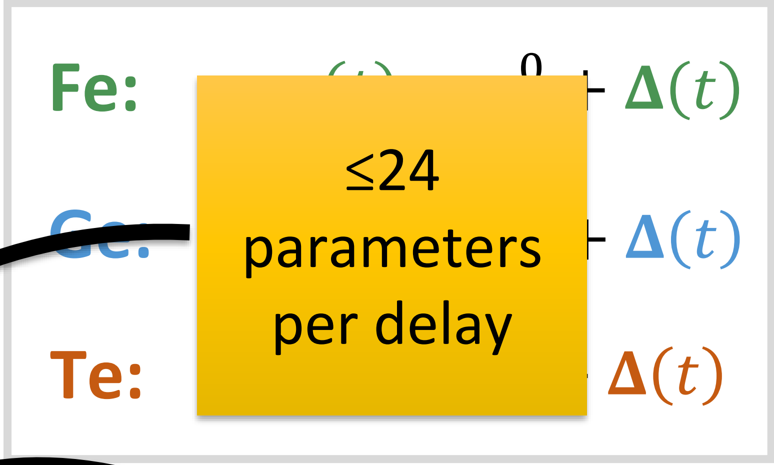


470 unique data points per delay

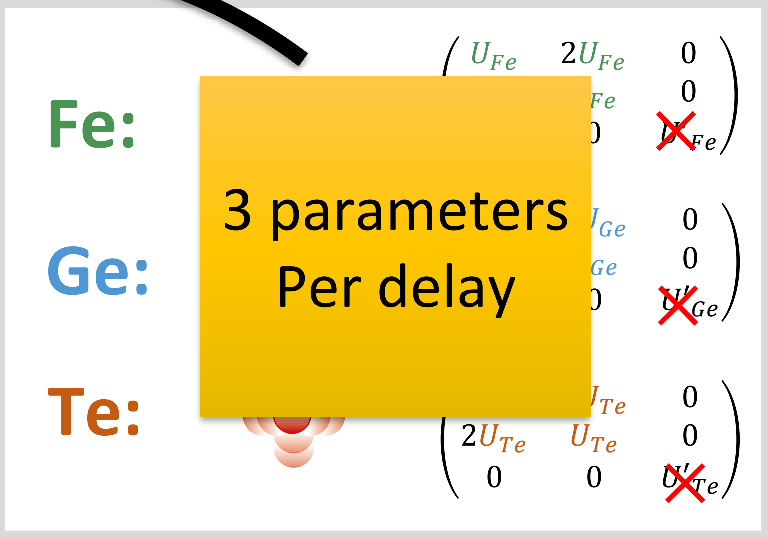
$$I(Q) \propto \left| \sum_i \overbrace{f_i(Q)}^{\text{Scattering factors (tabulated)}} \overbrace{e^{2\pi i \boldsymbol{Q}\cdot\boldsymbol{r}_i}}^{\text{phase factors}} \overbrace{e^{-\frac{4\pi^2}{3}\langle u_i^2\rangle Q^2}}^{\text{Temperature factor (Debye Waller effect)}} \right|^2$$

Can we systematically extract these quantities?

…Towards capability to solve crystal structures in the time domain!



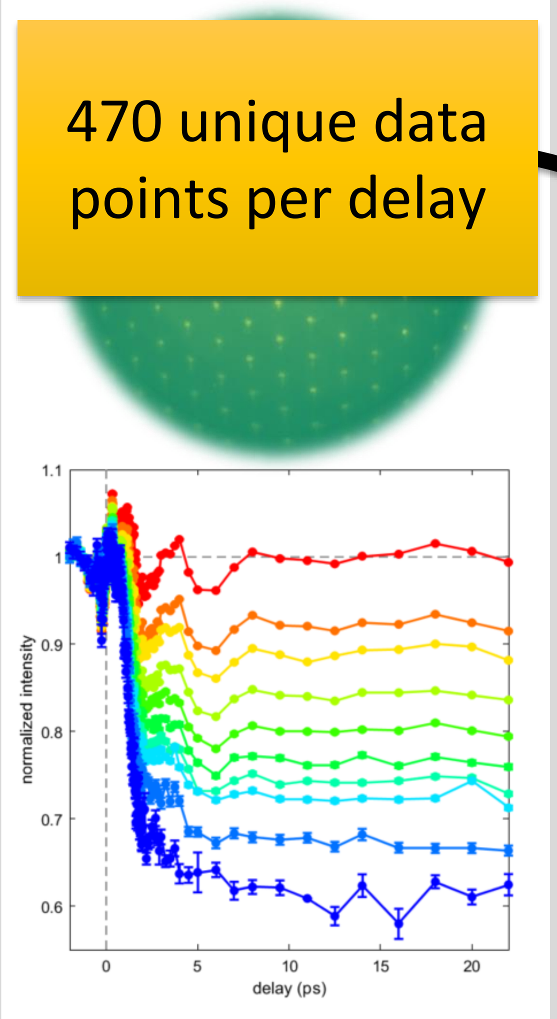
≤24 parameters per delay



3 parameters Per delay

Evolution of the phonon system

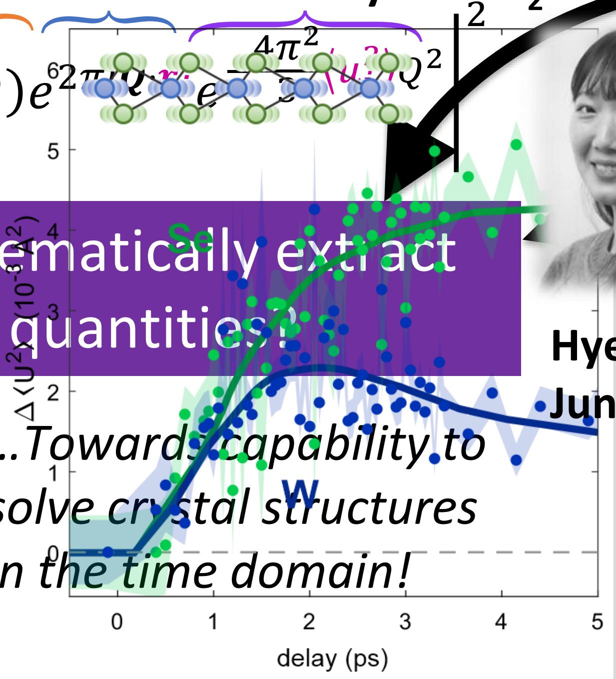
470 unique data points per delay



Scattering factors (tabulated) | phase factors | Temperature factor (Debye Waller effect)

$$I(Q) \propto \left| \sum_i f_i(Q) e^{2\pi i Q \cdot r_i} e^{-\frac{4\pi^2}{3}\langle u_i^2 \rangle Q^2} \right|^2$$

Can we systematically extract these quantities?



...Towards capability to solve crystal structures in the time domain!

Hyein Jung

AG Bratschitsch

WWU MÜNSTER

Fe: $+ \Delta(t)$

Ge: $+ \Delta(t)$

Te: $\Delta(t)$

≤24 parameters per delay

e-ph

Group 1

Group 2

ph-ph

Group 3

Fe:

Ge:

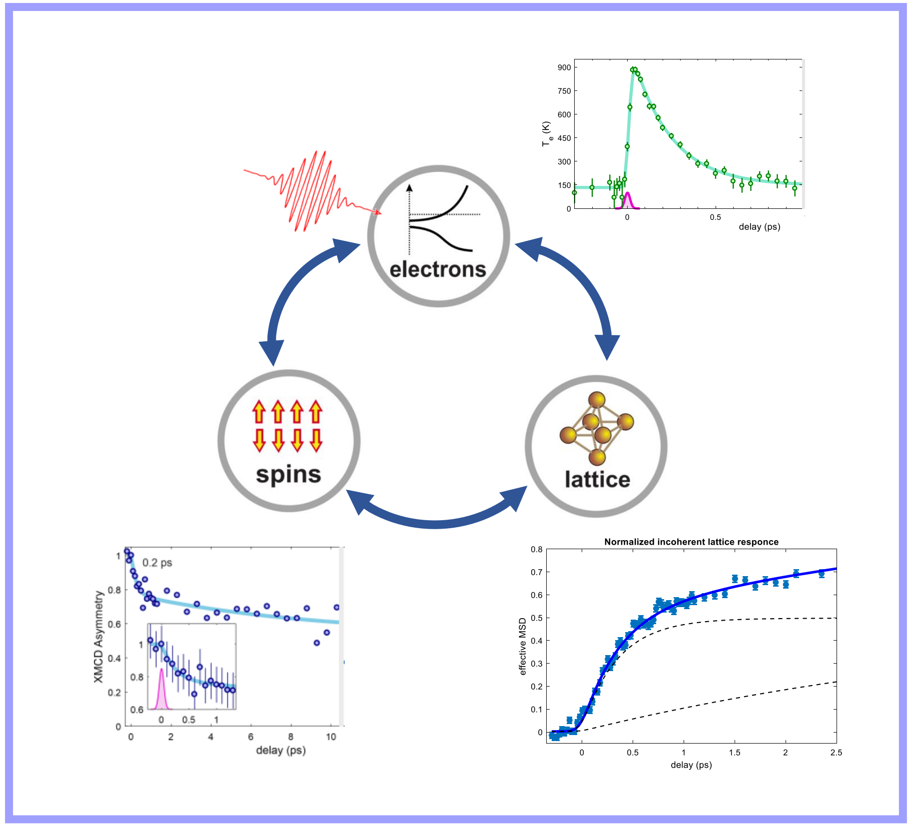
Te:

3 parameters Per delay

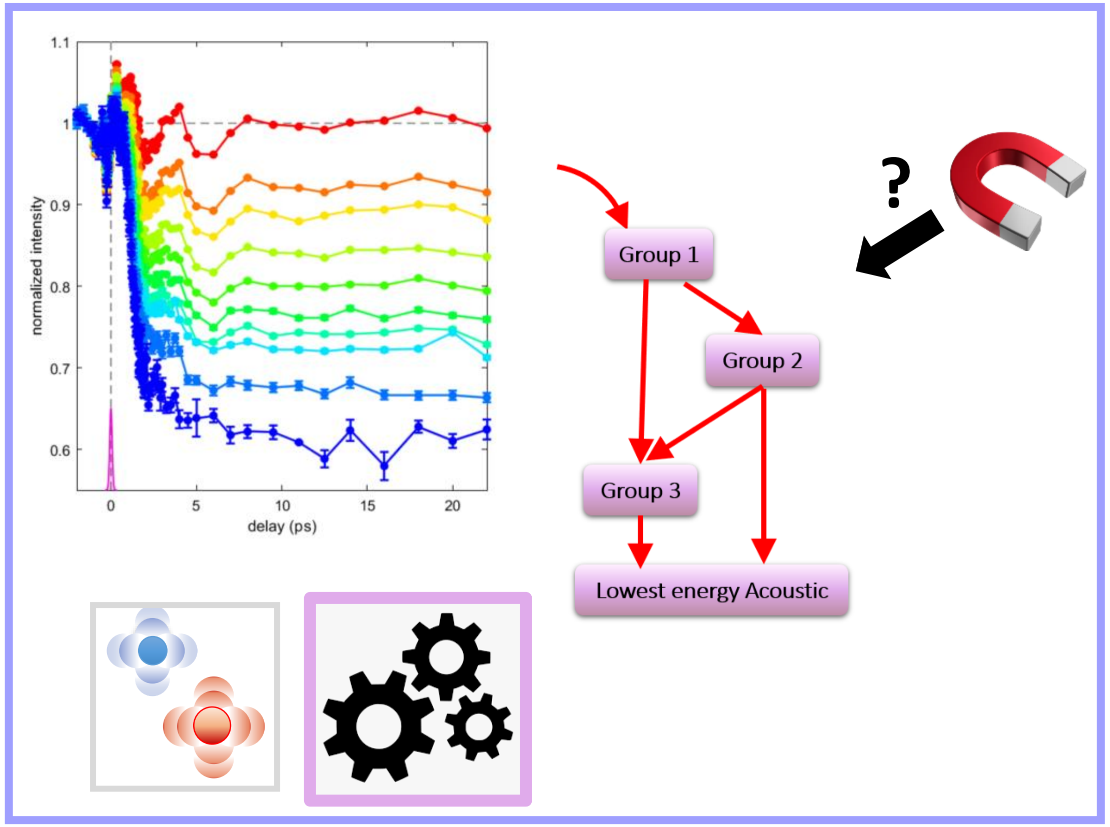
Lowest energy Acoustic

Stay tuned!

Towards "Full" energy flow picture



Towards detailed ph-ph picture



Acknowledgements

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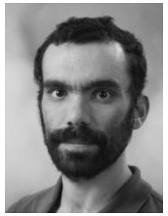
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U. Atxitia


T. Griepe

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Thank you.