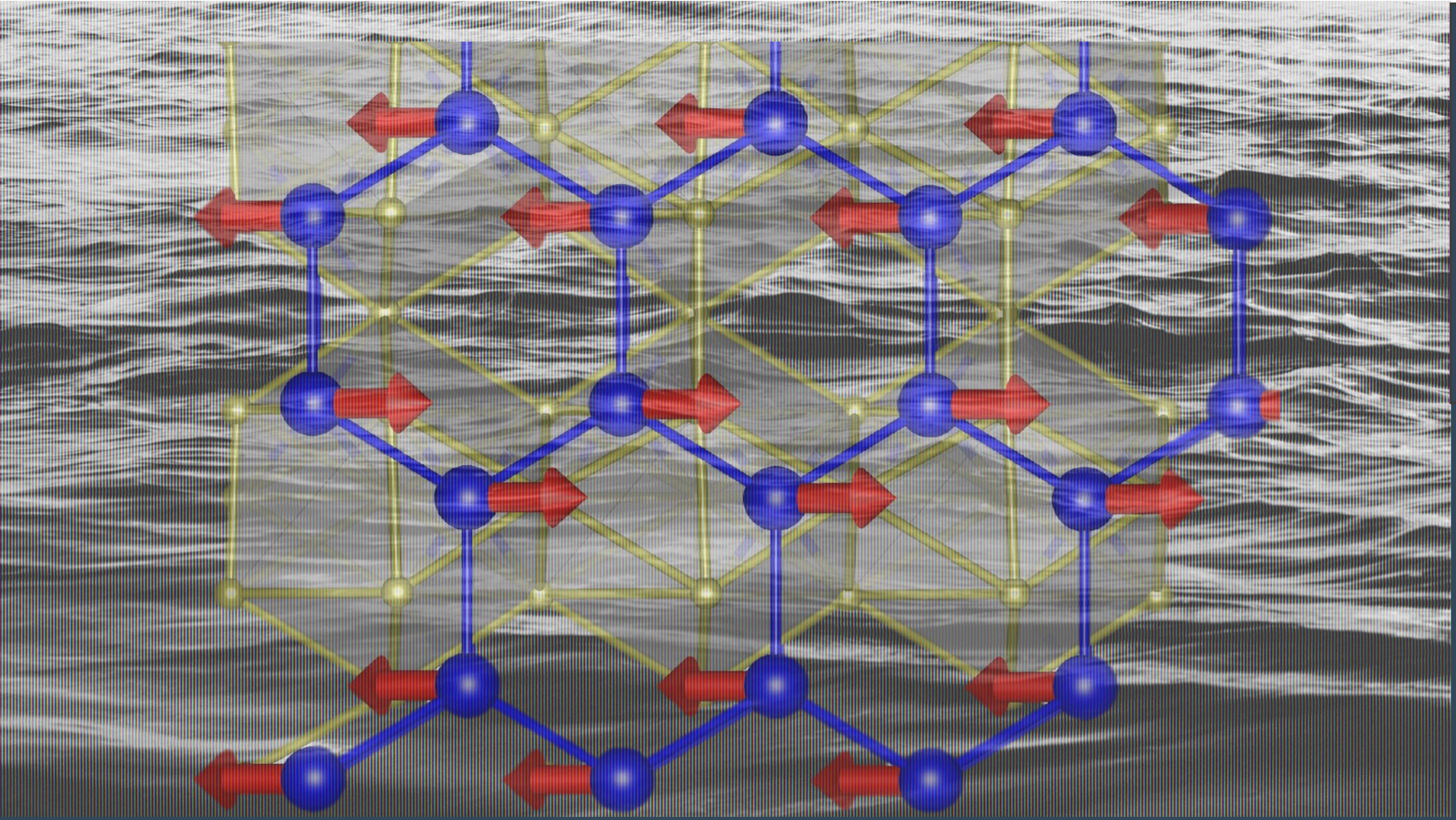


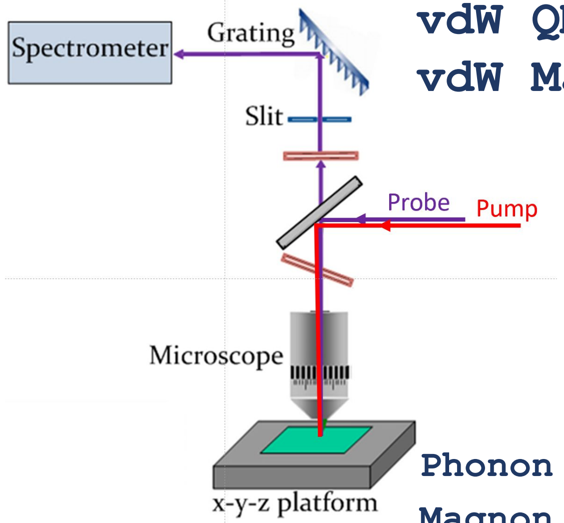
University of Cologne

Hamoon Hedayat
II. Physics Institute,
Department of Physics
University of Cologne

Light-Induced Metastable Disordered Spins in the Kitaev Candidate α-$RuCl_3$

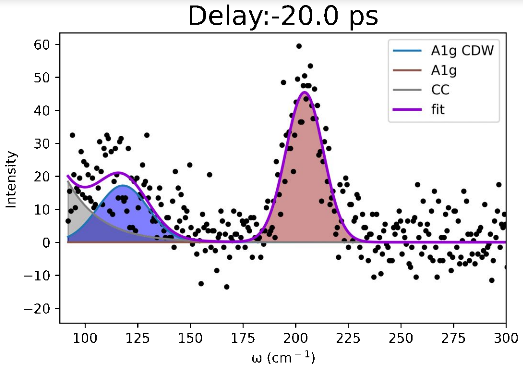
Ultrafast Optical Spectroscopy of Quantum Materials

TR Raman



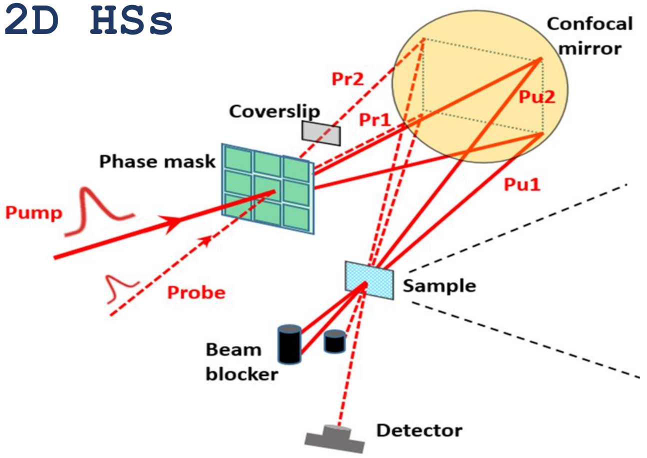
vdW QM, CDWs
vdW Magnets

Phonon dynamics.
Magnon dyanmics.

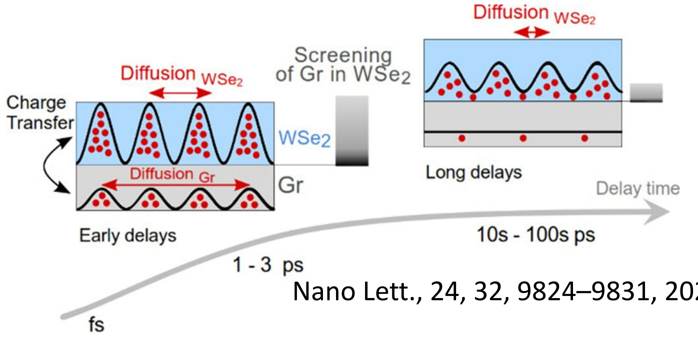


TR grating

2D HSs

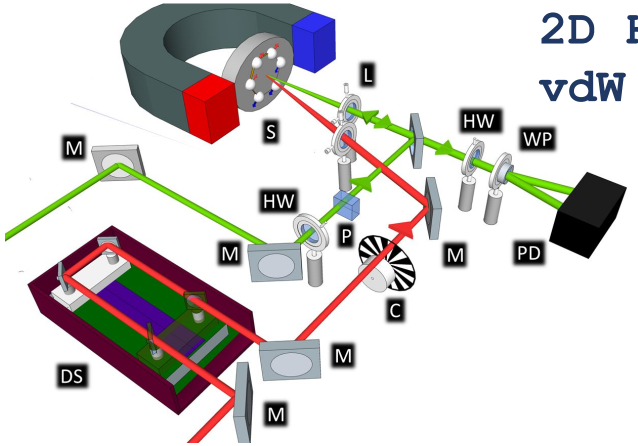


Dynamical diffusion.



Nano Lett., 24, 32, 9824–9831, 2024

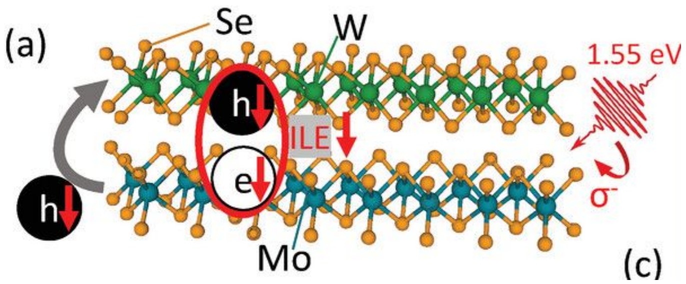
TR Magneto-optical Spec.



2D HSs
vdW Magnets

Charge transfer.
Spin dynamics.

8T	2K



Adv. Optical Mater, 13, 2402703, 2025

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In equilibrium
measurements: low field, more details on metamagnetic transitions

Out-of equilibrium

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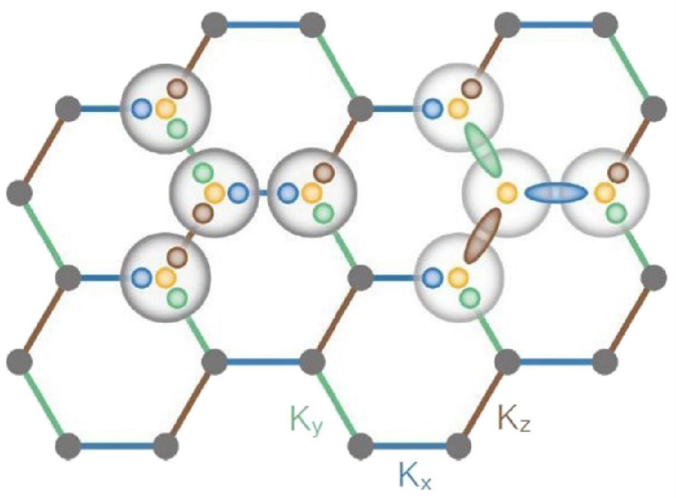
α-$RuCl_3$ – promising SL material ?!

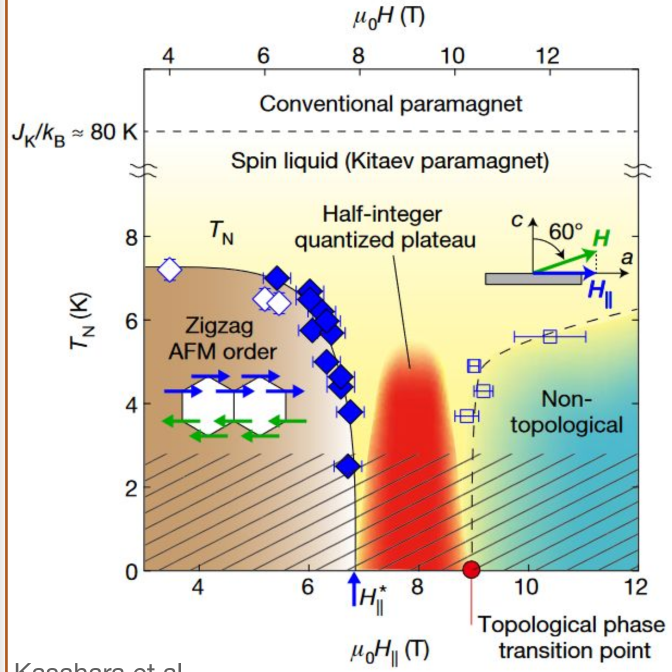
α-RuCl3 as promising material for Kitaev model describing spins *S*=1/2 on a honeycomb lattice



Simon Trebst
Kitaev Materials
arXiv:1701.07056

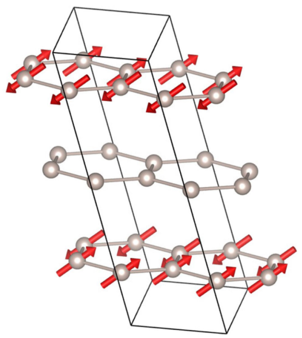
$$\mathcal{H}_{ij}^{(\gamma)} = K S_i^\gamma S_j^\gamma$$

Searching for Experimental fingerprints of KSLQ



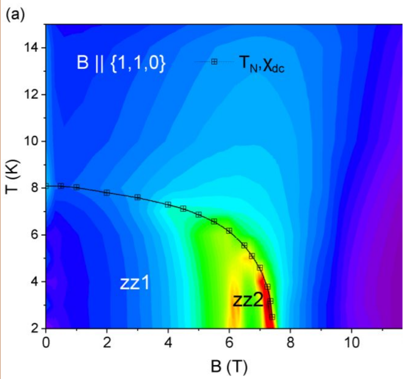
Kasahara et al.
Nature volume 559, pages227–231(2018)

ZZ ground state

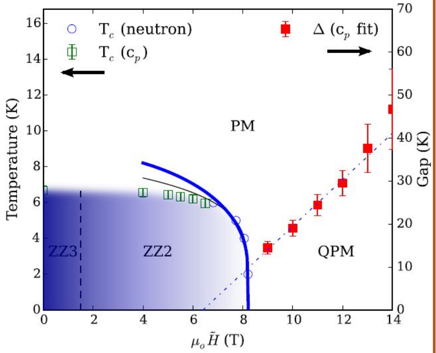


Phys. Rev. B
93 134423

Characterizing ZZ spin configuration



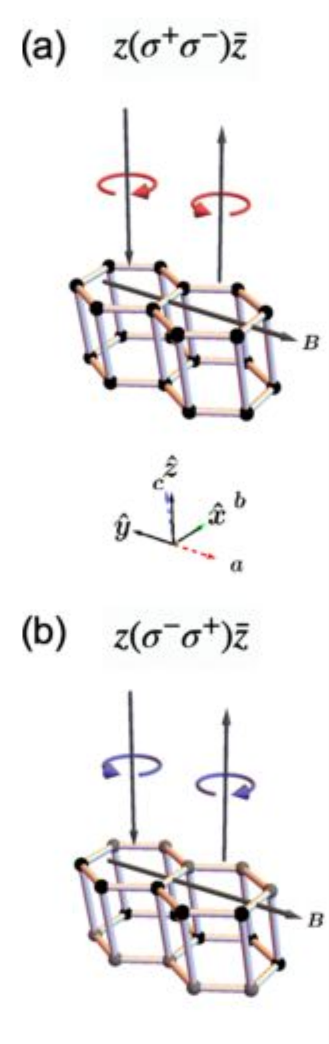
PHYSICAL REVIEW B
103, 174417 (2021)

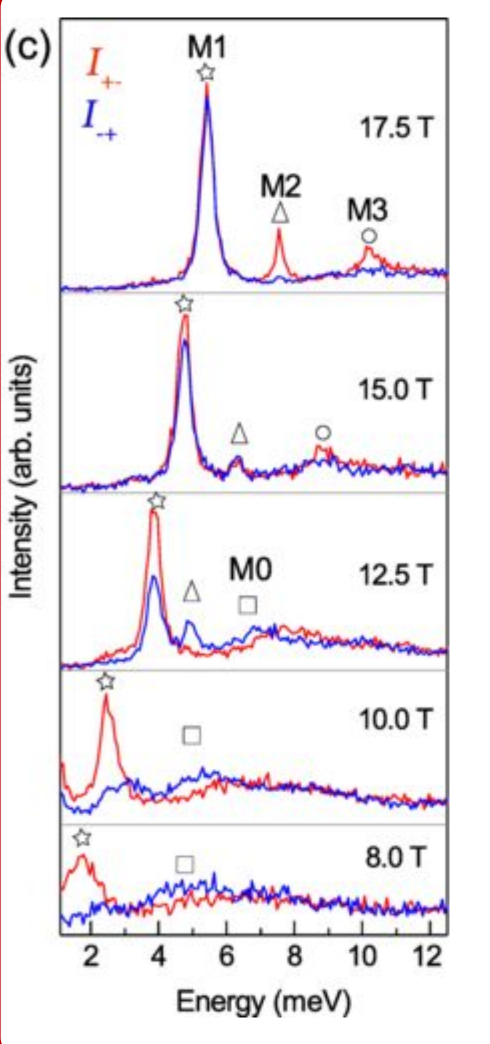


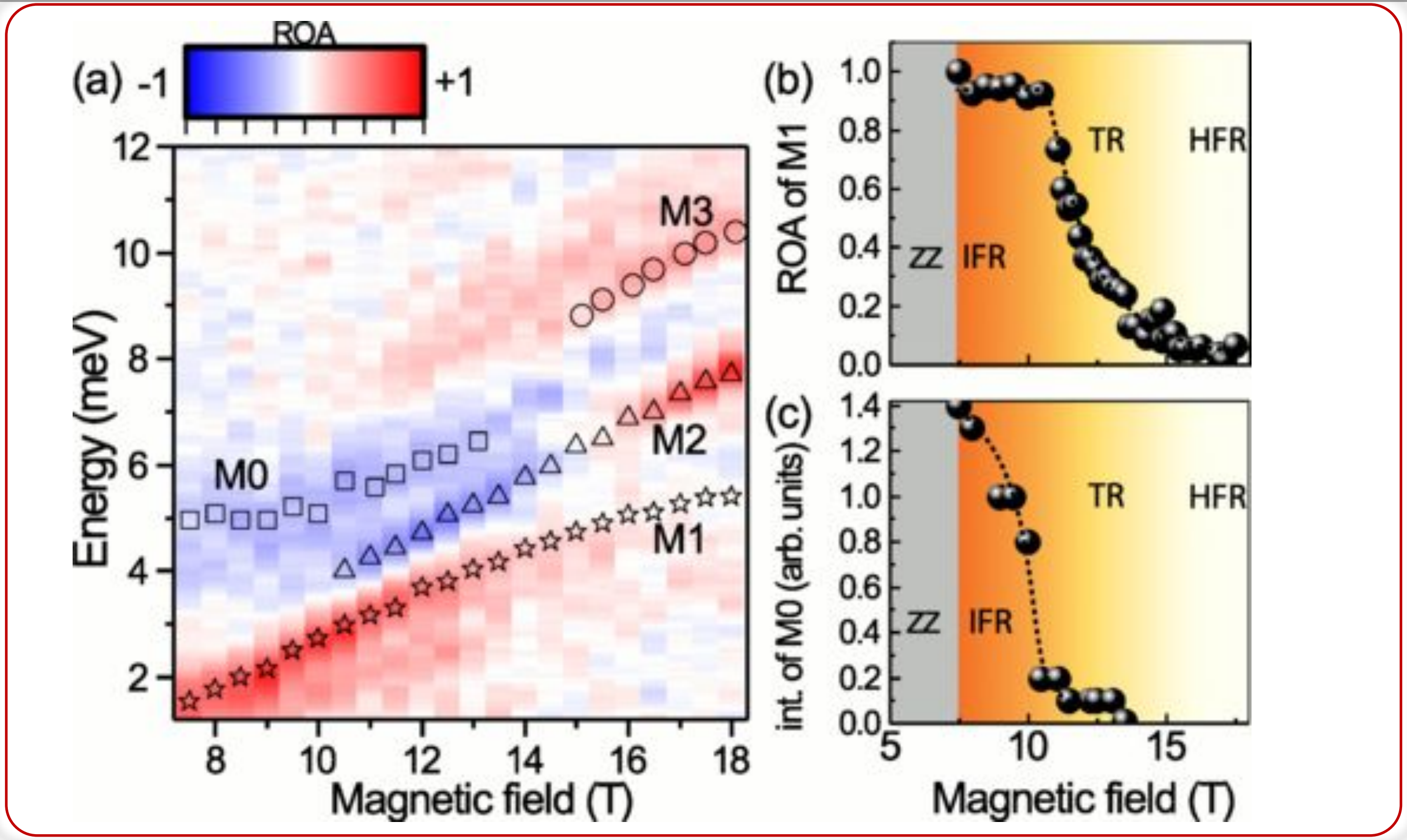
Phys. Rev. B 95, 180411(R) (2017)

$$\mathcal{H}_{ij}^{(\gamma)} = J S_i \cdot S_j + K S_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right)$$

α-$RuCl_3$ – ground sate and T-B diagram





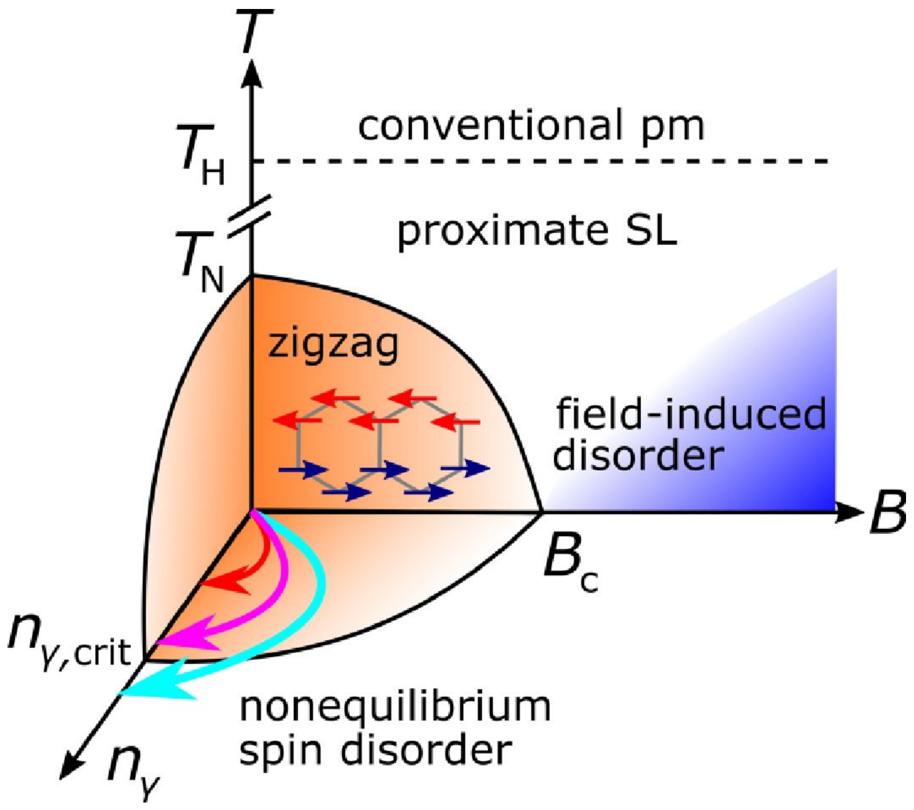


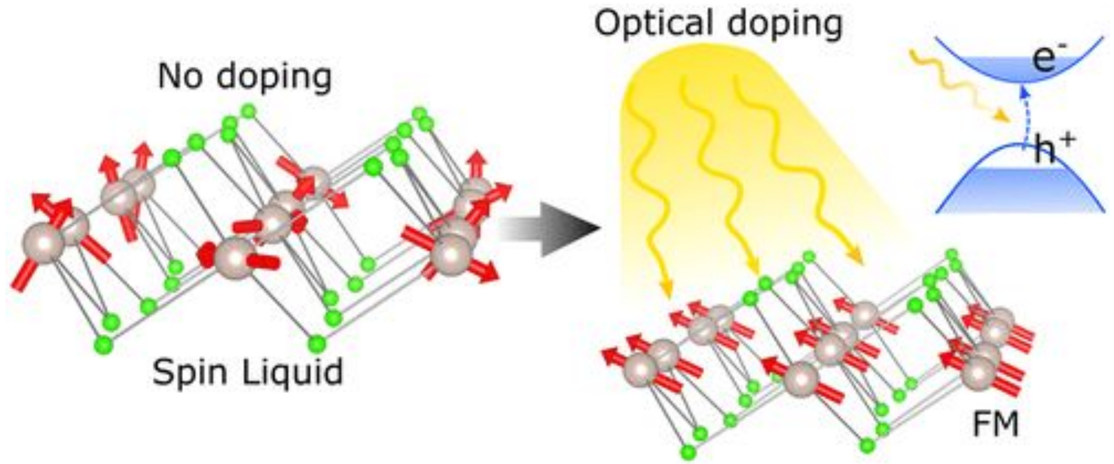
$$\mathrm{ROA} = \frac{I_{+-} - I_{-+}}{I_{+-} + I_{-+}}$$

Between 7.5 and 10.5 T, a clear plateau in chiral spin-flip signals points to a unique intermediate phase with chiral excitations.

PHYSICAL REVIEW RESEARCH 6, L022005 (2024)

α-$RuCl_3$ – ground sate and T-B diagram, OPTICALLY





Nano Lett. 2019, 19, 11, 7673–7680

Photoexcitation

Magneto optical spect.

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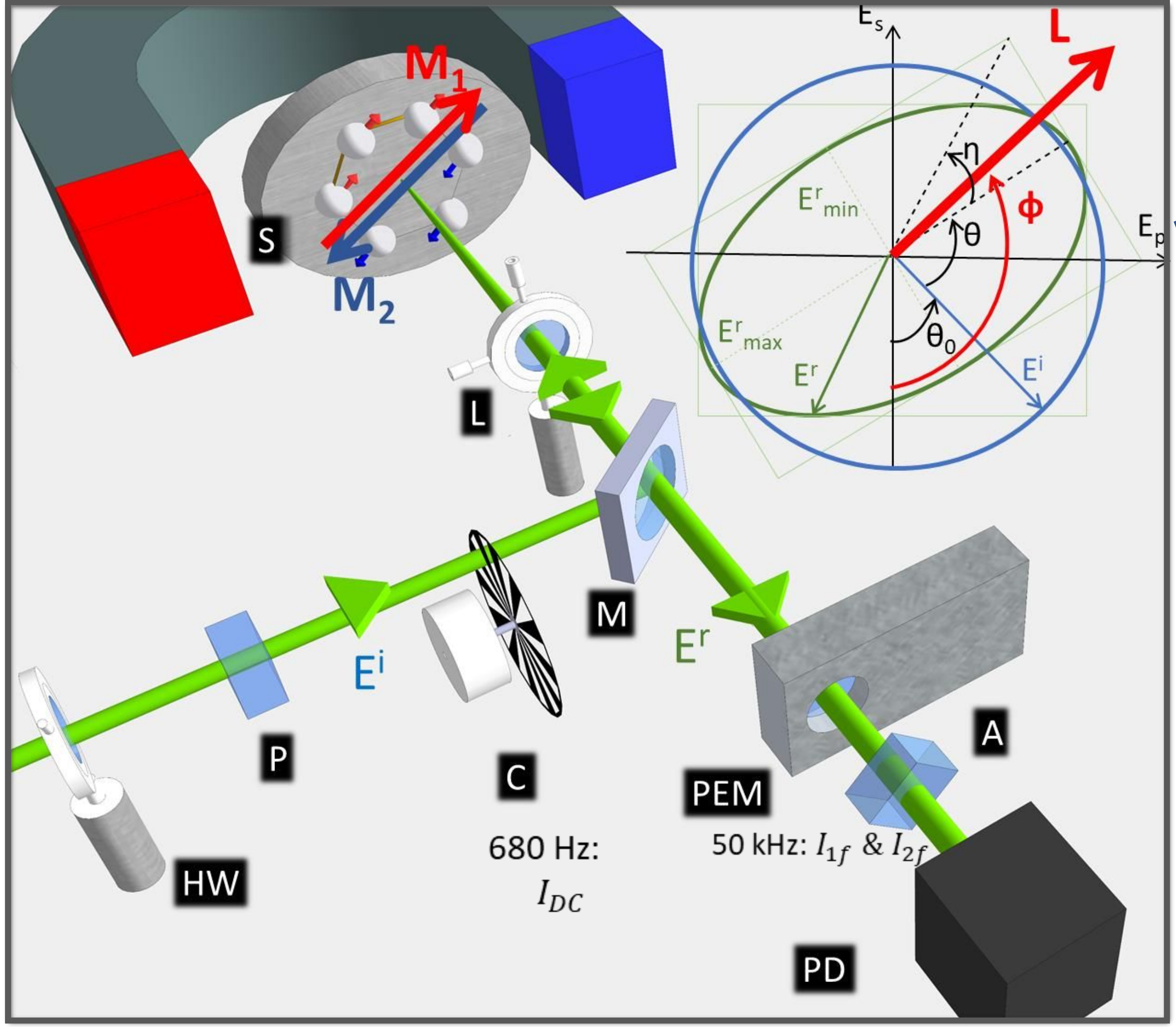
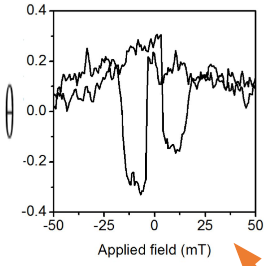
Magneto-optical (MO) Spec.

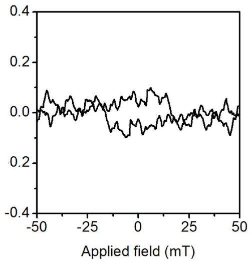
Experimental setup



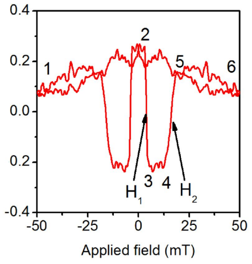


$$\Theta_{\mathcal{MLD}} = \frac{1}{2}(\Theta_{tot}(+B) + \Theta_{tot}(-B))$$

$$\Theta_{\mathcal{Lin}} = \frac{1}{2}(\Theta_{tot}(+B) - \Theta_{tot}(-B))$$



+



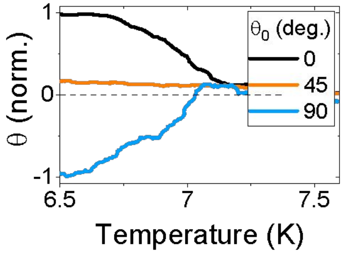
$$\theta = \mathcal{A}^{\mathcal{Lin}} + (\mathcal{A}^{\mathcal{MLD}} \sin[2(\phi - \theta_0)])$$

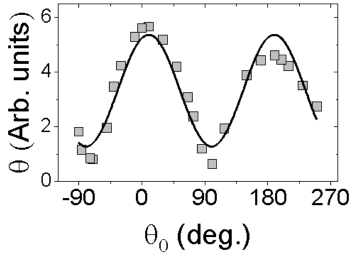
Linear MOKE

$\propto L^2$

Direction of **L**

Experiment on α-RuCl3





MLD: $\frac{I_{2f}}{I_{DC}} \propto \theta$

MLB: $\frac{I_{1f}}{I_{DC}} \propto \eta$

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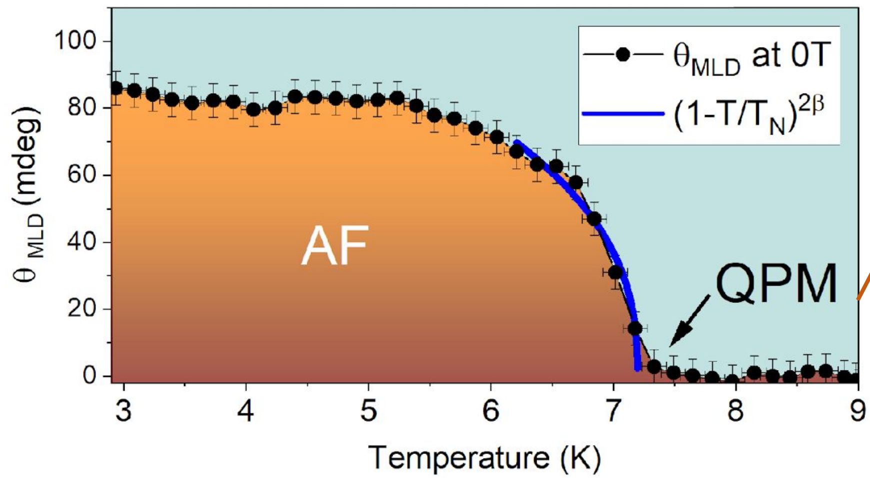
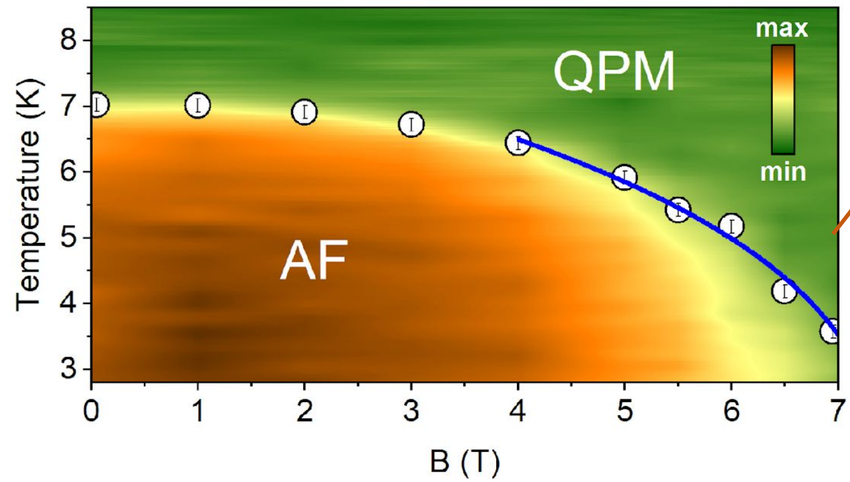
In equilibrium MO results on α-$RuCl_3$

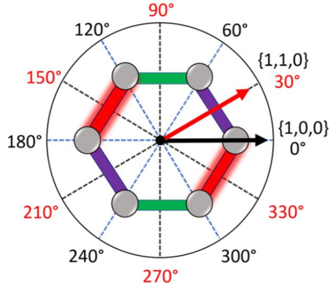


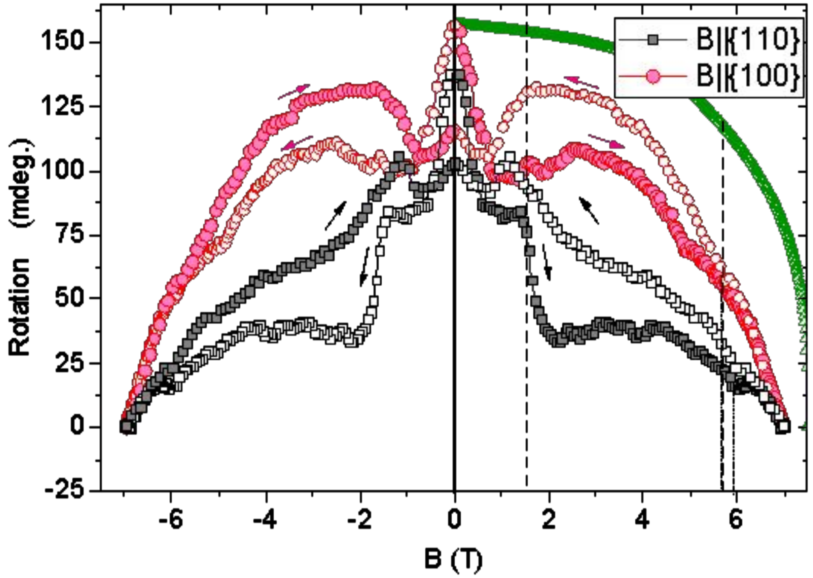


$(1-T/T_N)^{2\beta}$
$\beta = 0.19 \pm 0.07$
2D Ising universality class

$(H-H_c)^{\gamma}$
$H_c = 7.480$ T and
$\gamma = (0{:}31 \pm 0.07)$,
->
0.32, 2D Ising symmetry class

Direction and field dependent isothermal MO spectroscopy





Highly anisotropic MLD response

$$\theta_{\mathcal{MLD}} = \mathcal{A}^{\mathcal{MLD}} \sin[2(\phi - \theta_0)]$$

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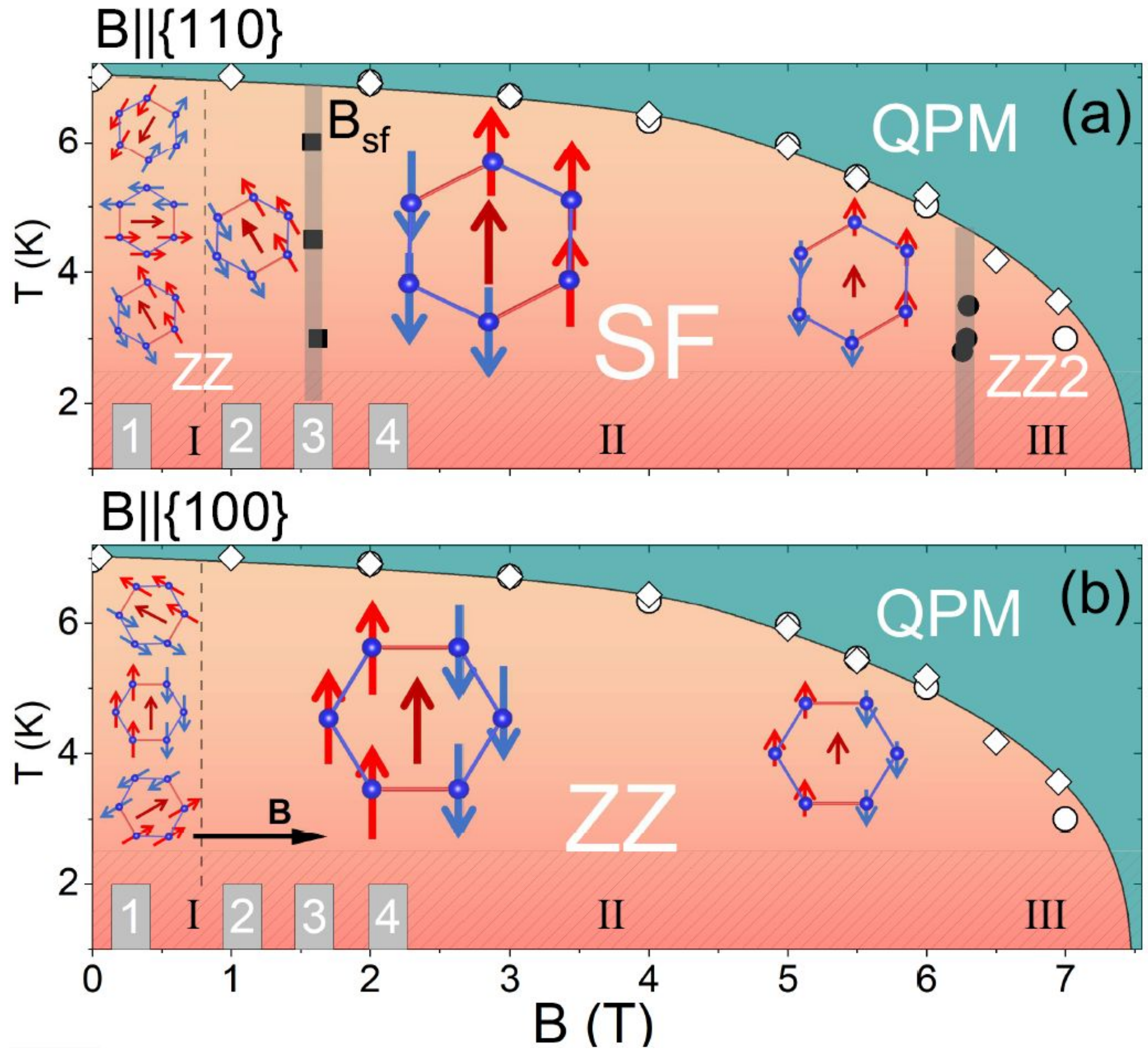
Results on α-RuCl3



Temperature-field phase diagram of α-$RuCl_3$ is shown for two in-plane field directions:

- ❑ {1,1,0}: perpendicular to the Ru–Ru bond.
- ❑ {1,0,0}: along the Ru–Ru bond.

Three magnetic regimes are identified:

- ❑ Regime I (ZZ): Zigzag order with three initial domains.
- ❑ Regime II (SF): Spin-flop phase.
- ❑ Regime III (ZZ2): Modified zigzag order.

npj Quantum Materials (2022) 7:28

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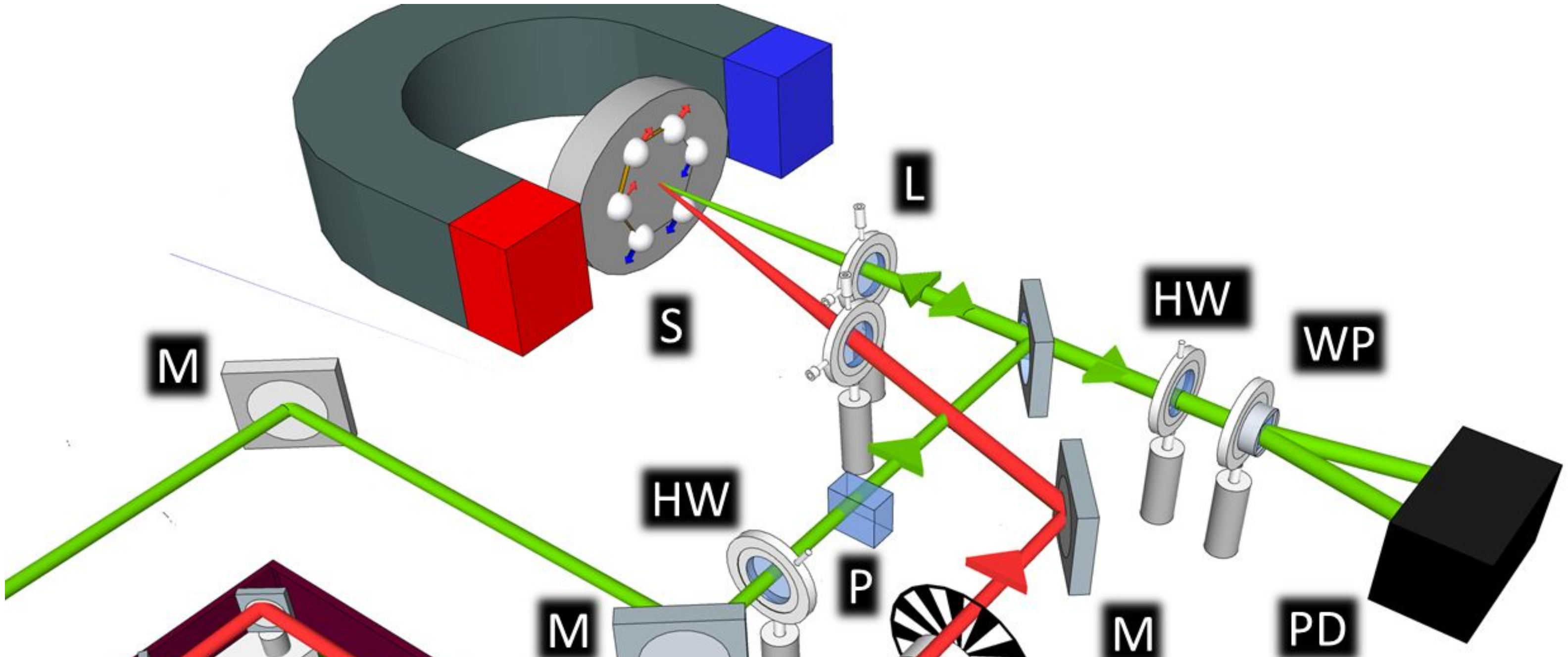
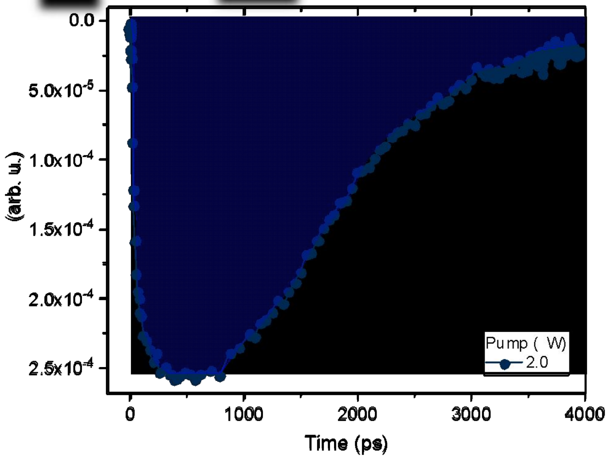
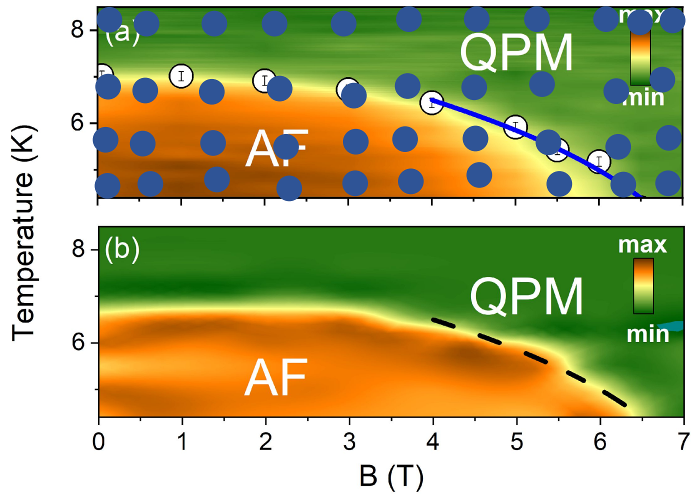




Static phase diagram



Dynamics phase diagram

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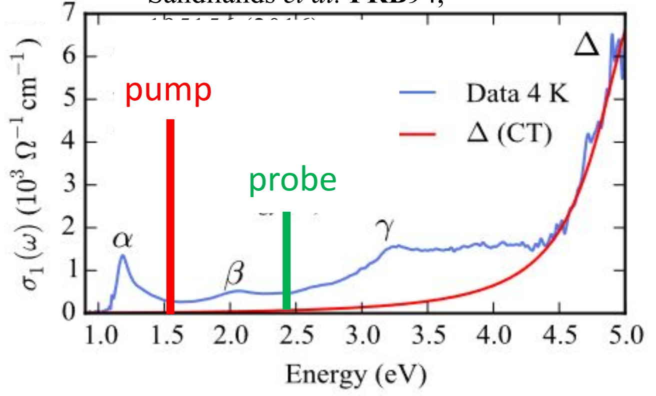
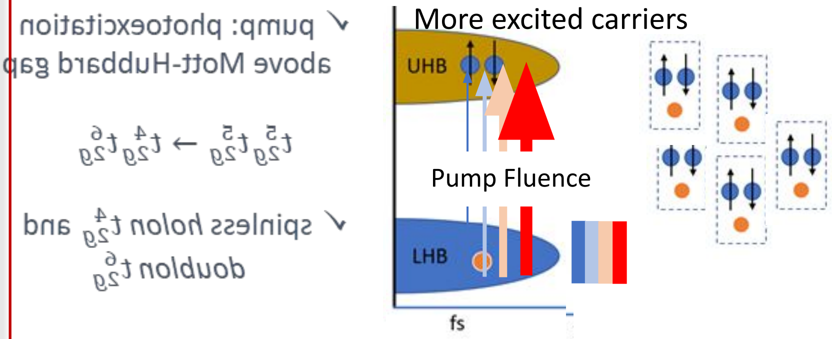
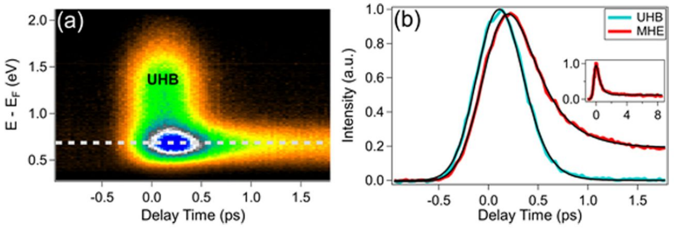
photoexcited α-RuCl3

Sandilands *et al.* **PRB**94,

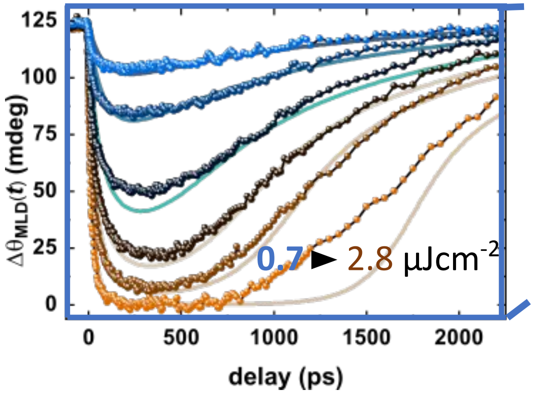




Physical Review B 103, 245105 (2021)

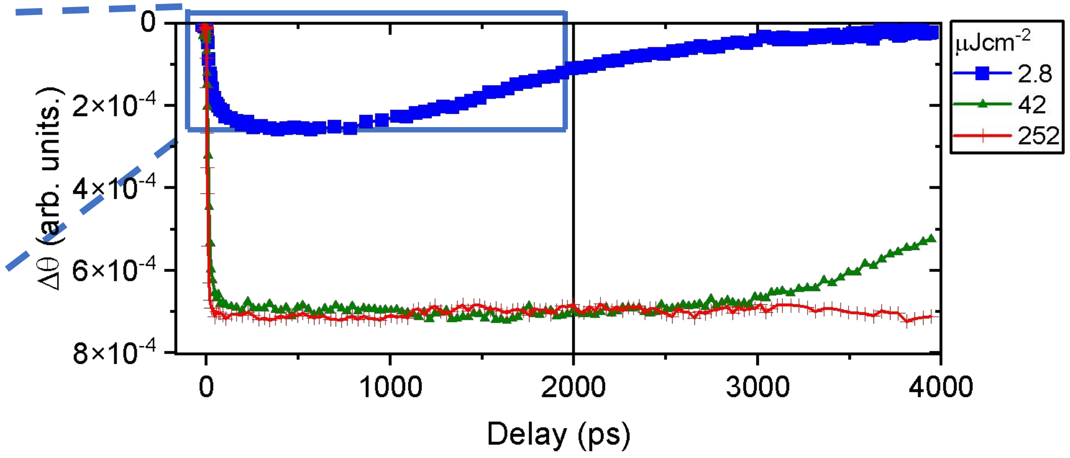


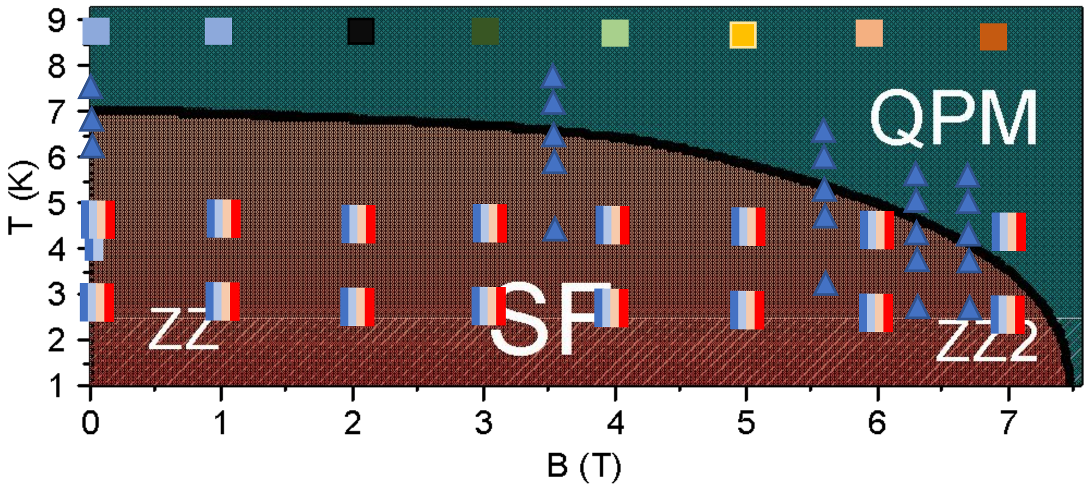
Low Fluence- Zero Magnetic field



PHYSICAL REVIEW B 105, 224428 (2022)

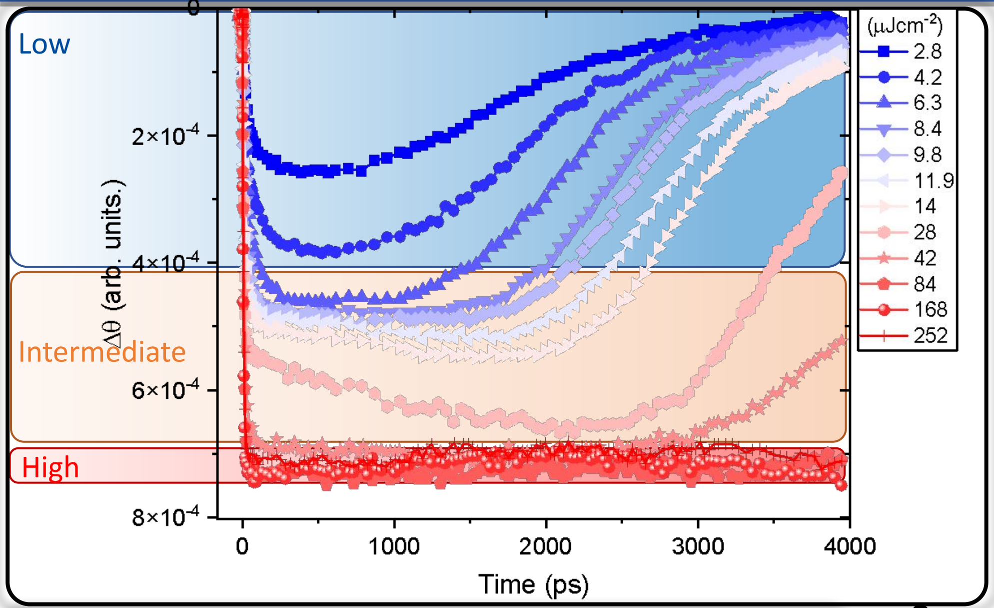
We extended the study





Extended
Not only in time and
Excitation density
But
To all
ZZ-SF-ZZ2 and PM
phases

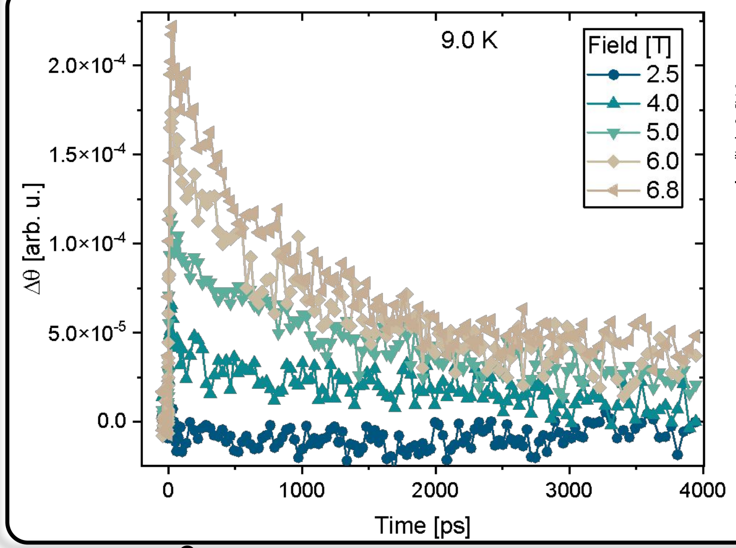
photoexcited α-$RuCl_3$

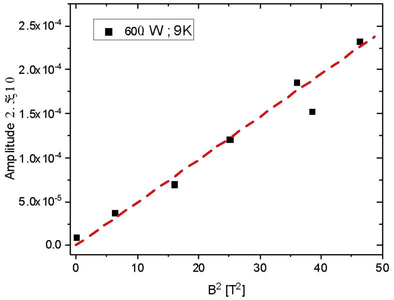


ZZ

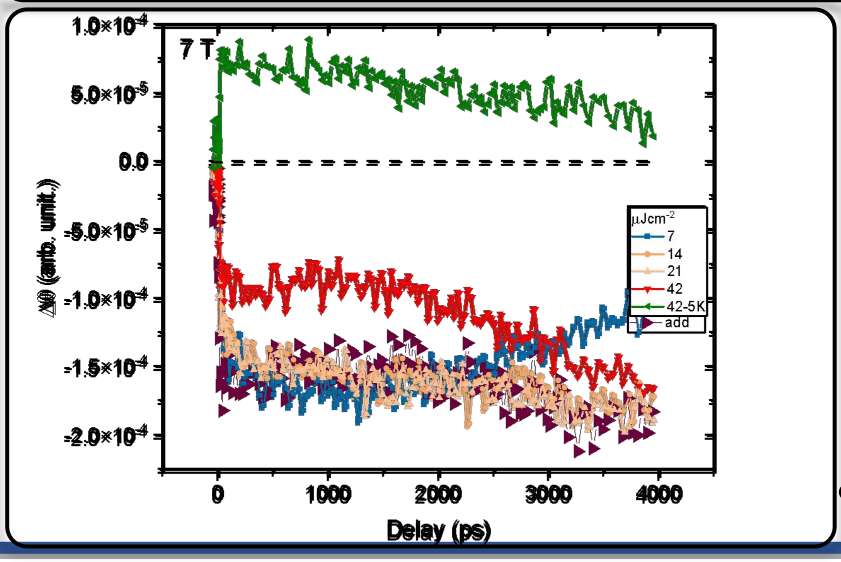
Opposite sign

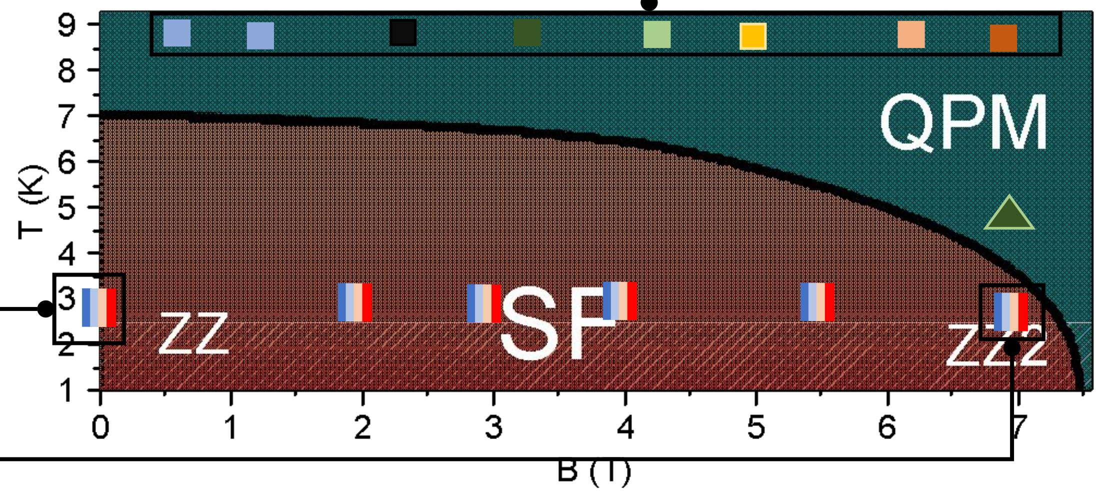
QPM





Magnetization dynamics in QPM phase





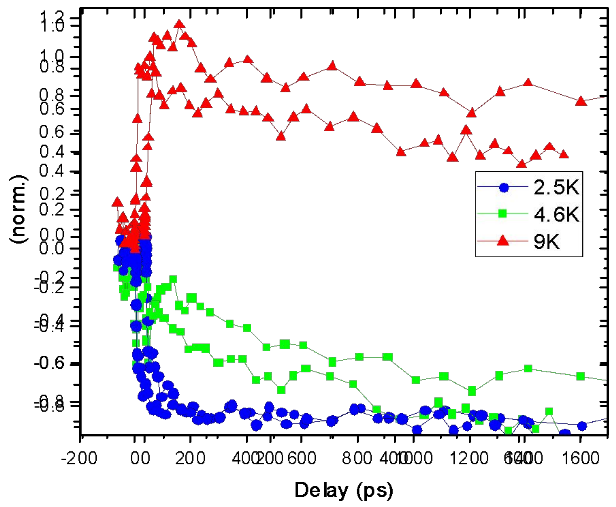
Fluence dependence

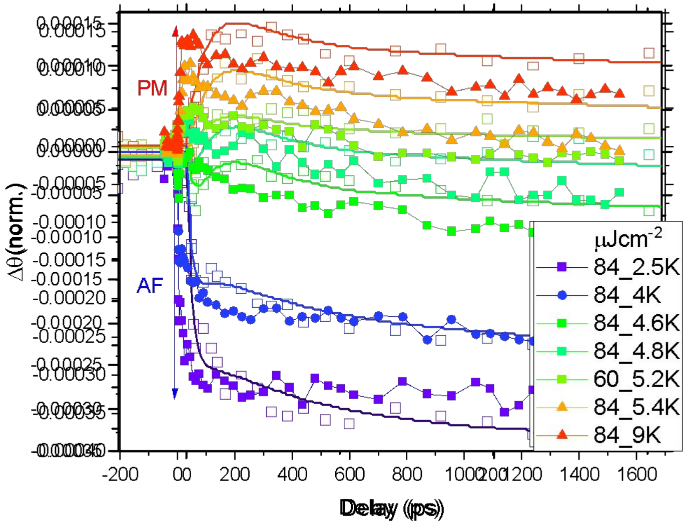
Fast recovery

Long lived disorder

Signature of QPM

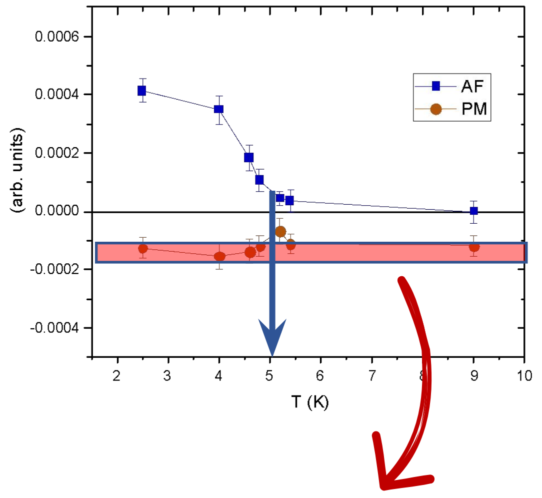
photoexcited α-RuCl3



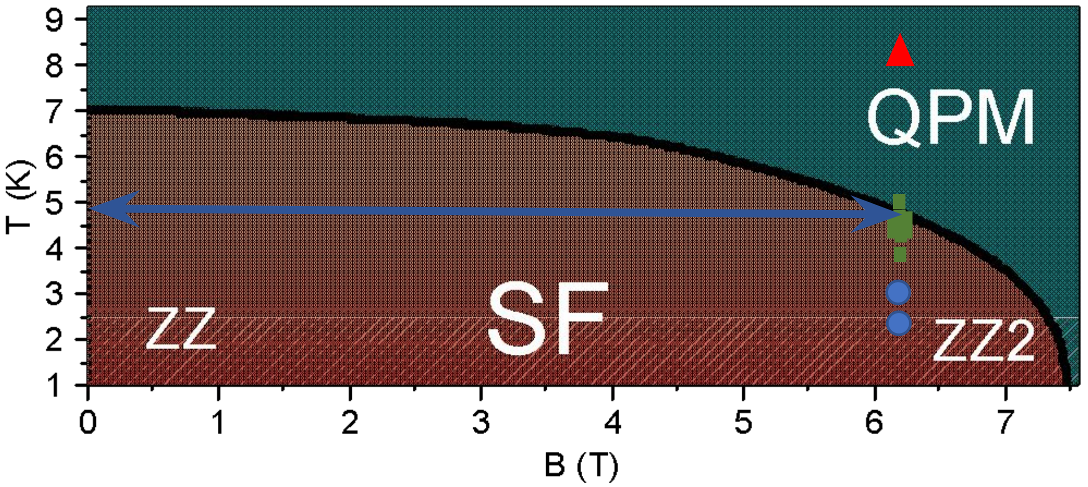


Phenomenological model

$$\Delta\theta = \{AF\,[1 - e^{-t/\tau_{AF}}]e^{-t/\tau_{D1}} + PM\,[1 - e^{-t/\tau_{PM}}]e^{-t/\tau_{D2}}\} * g(\sigma)$$



Signature of QPM
For high excitation

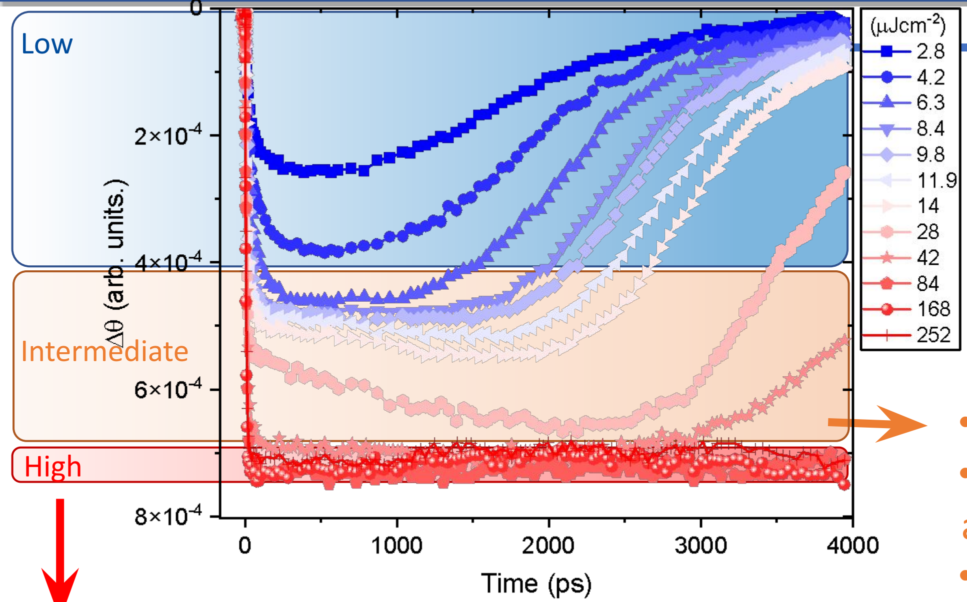


$\tau_{PM} \approx 13$ ps
$\tau_{AF} \approx 5$ ps

Different non-thermal
Dynamics/mechanisms

Faraday Discuss., 2022, 237, 237

photoexcited α-RuCl3



- Low holon-doublon population
- Less HBE creation
- Still Strong antiferromagnetic background
- No plateau- long lived AF disorder

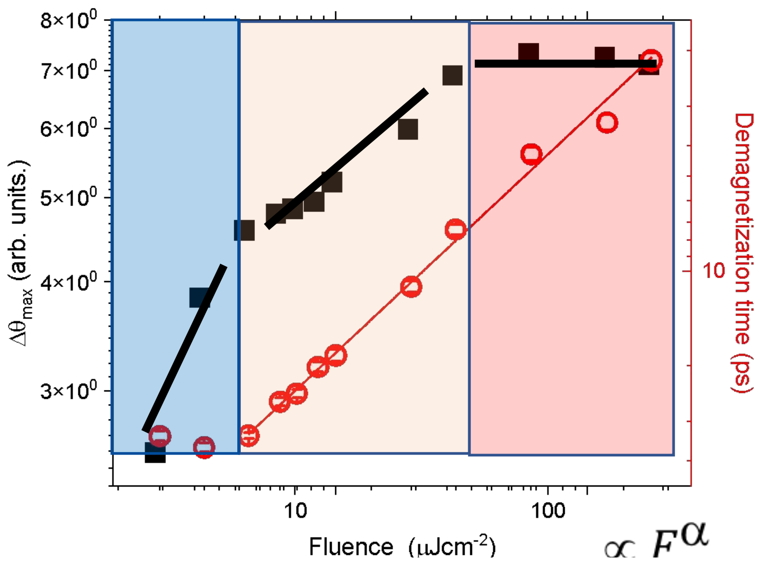
- Weaker antiferromagnetic background
- High holon-doublon population -> faster and more MHE pop
- Fluence dependent plateau- long lived AF disorder

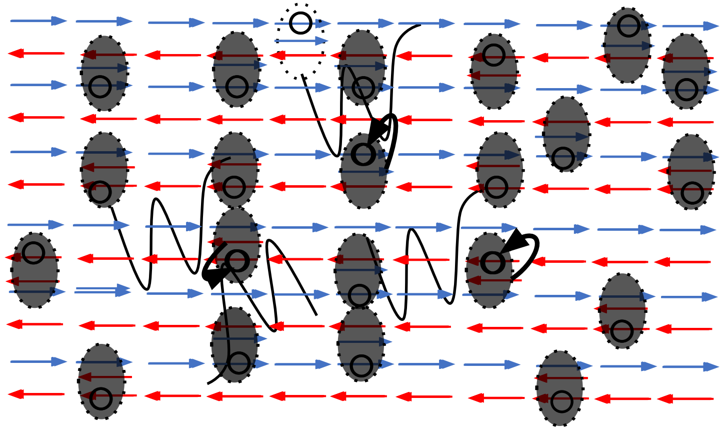
- Saturation of Δθ -> suppression of AF
- Signatures of PM phase in high field
- Non-thermal phase transition
- No recovery in 4 ns

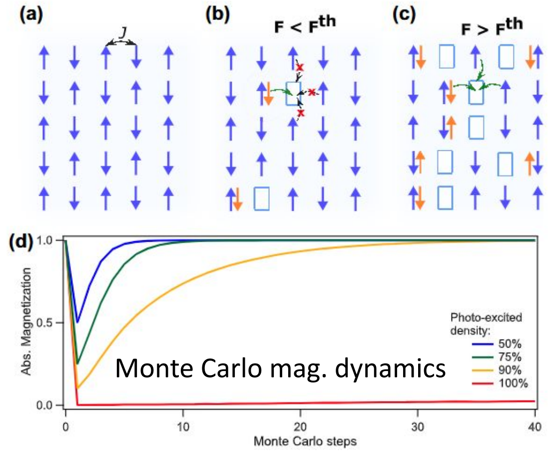
LOW

Magnon emission/disturbing AF BG

Suppression of AF phase vs PED







PRB 105 064409 (2022)

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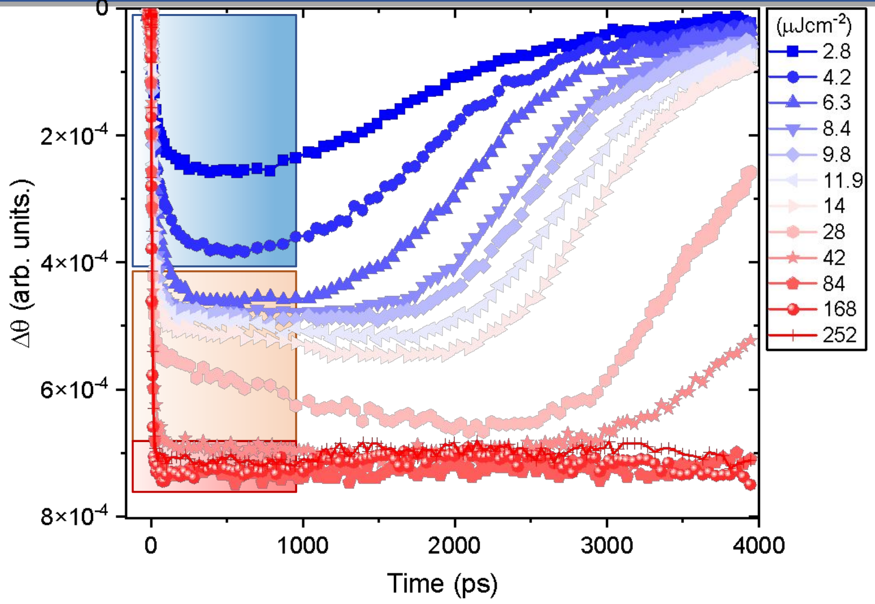
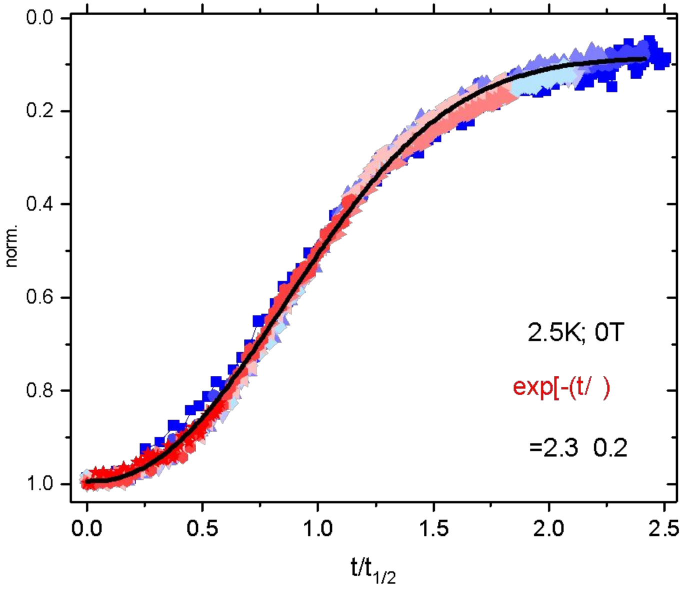
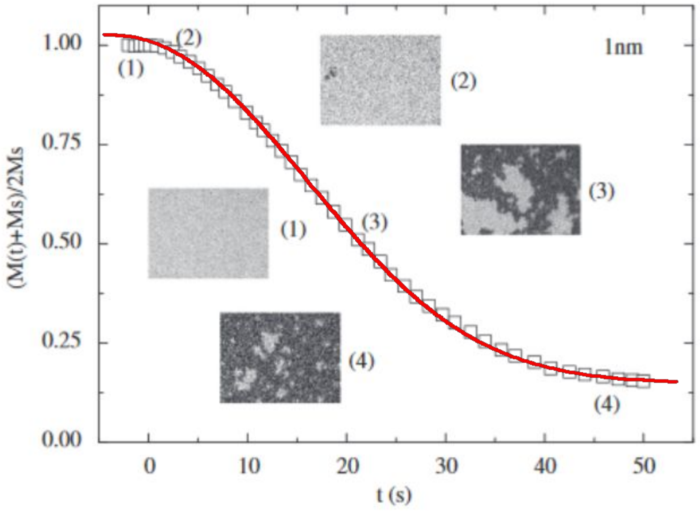
photoexcited α-RuCl3





- Adapted Fattuzo model
- $\propto \exp[-(\frac{t}{\tau})^{\beta}]$
- $k = \frac{v}{Rr_c} = \frac{\text{domain propagation}}{\text{nucleation}}$
  > ns domain growth
- β > 1: domain growth

Faraday Discuss., 2022, 237, 237

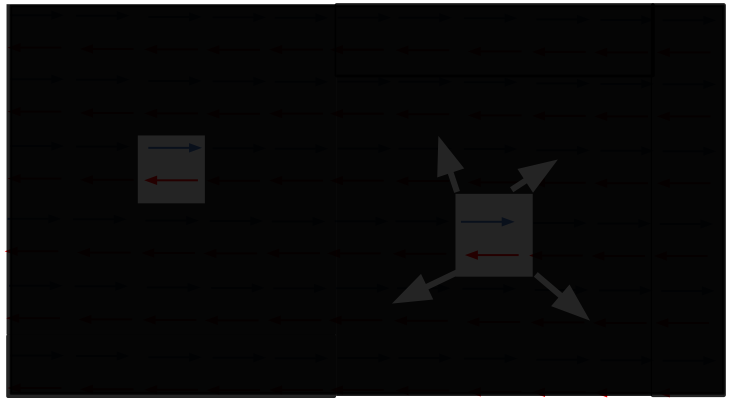


Fattuzo model

$$B(t') = \exp\left(-(t'/\tau(k))^{\beta(k)}\right),$$

$$k = \frac{v}{Rr_c} = \frac{\text{domain propagation}}{\text{nucleation}}$$

nucleation domain growth



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Conclusions

- More details on metamagnetic transitions by static MO spectroscopy

- Long-lived disorder AF phase in out-of-equilibrium
- Signatures of non-thermal phase transition induced by strong optical excitation
- The mechanisms lead to spin disorder are discussed but the true magnetic nature of this non-thermal phase needs more investigation
- Recovery dynamics is fluence independent and follows Fattuzo-like behaviour

Outlook

Characterize the non-thermal phase using different methods (such as Raman)

Looking at non-thermal phase transition at 2D limit

Extend this kind of studies to other correlated materials and spin liquid candidates

Acknowledgments

University of Cologne

- OCMS group of

Paul van Loosdrecht

- Julian Wagner
- Anuja Sahasrabudhe
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In memory of the late Daniel I. Khomskii
whose contributions continue to inspire!!!

Contact:

Hamoon Hedayat

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Website

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PhD students:

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- Danilo Comini
- A fully funded PhD position is available

CRC 1238 Control and Dynamics of Quantum Materials

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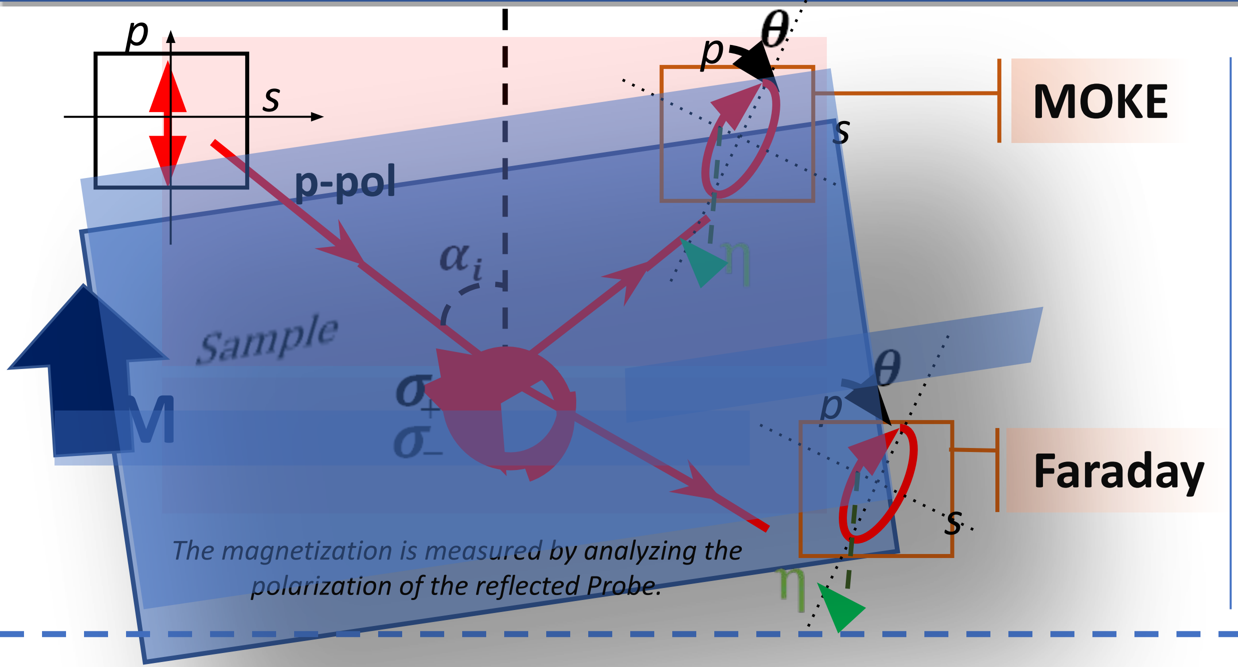
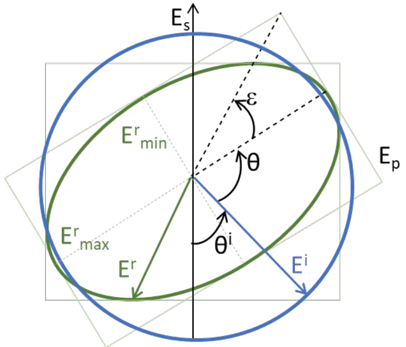
Magneto-optical (MO) Spec. and Antiferromagnetism



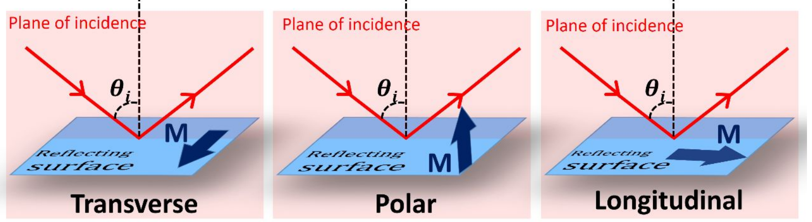


Rotation

$$\Theta = \theta + i\eta$$

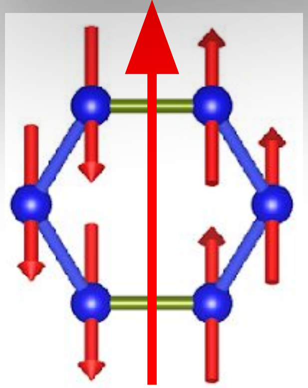
Ellipticity

MOKE



$$\theta(-M) = -\theta(M)$$

Antiferromagnet



$M\downarrow + M\uparrow = 0$

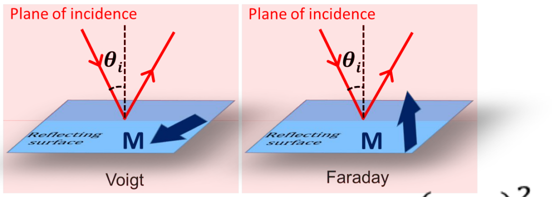
Expansion dielectric tensor for |M|<<|L|

$$\epsilon_{ij} = \epsilon_{ij}^{(0)} + \left[\frac{\partial \epsilon_{ij}}{\partial M_k}\right]_{M_0} \cdot L_i + \frac{1}{2}\left[\frac{\partial^2 \epsilon_{ij}}{\partial L_k \partial L_l}\right]_{L_0} \cdot L_k L_l$$

$$\theta(-M) = \theta(M)$$

$$\theta \propto L^2$$

QMOKE



$$\varepsilon, \theta \approx n_{||} - n_{\perp} \propto \epsilon_{||} - \epsilon_{\perp} - \frac{(\epsilon_{OD})^2}{\epsilon_{\perp}}$$

Magnetic linear dichroism (**MLD**) $\propto \theta$

Magnetic linear birefringence (**MLB**) $\propto \eta$