# Resolving chicken-or-egg causality dilemma for magneto-structural phase transition in FeRh


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https://www.ru.nl/science/uscm/

# Workshop 'Ultrafast Antiferromagnetic Writing'

- May 9th – 10th 2022, Ingelheim (Germany)

**Categories**

- Pathways to ultrafast switching of antiferromagnets
- Optically-driven spin excitations in antiferromagnets
- Switching of antiferromagnetic metals
- Switching of antiferromagnetic insulators
- Phononic control of antiferromagnets
- THz excitations in heterostructures and complex spin textures

https://www.spice.uni-mainz.de/ultrafast-antiferromagnetic-writing-2022-program/

# Webinar Series

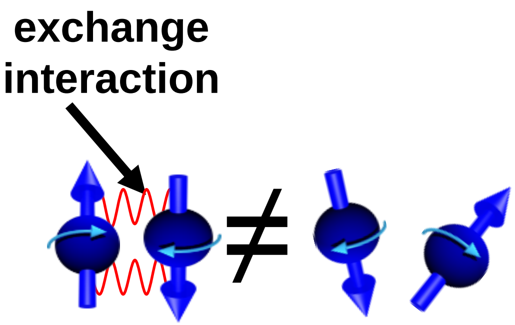
Prof. B. Ivanov
Theory of Antiferromagnets
May 2, $15^{00}$

Prof. A. Kimel
Ultrafast Magnetism
May 3, $15^{00}$

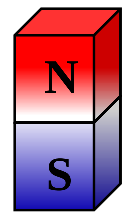
Dr. B. Heinrich
Meet the Editor
May 5, $11^{00}$

Prof. T. Jungwirth
Antiferromagnetic Spintronics
May 6, $15^{00}$

# How fast can we make "useless" antiferromagnet "useful"?

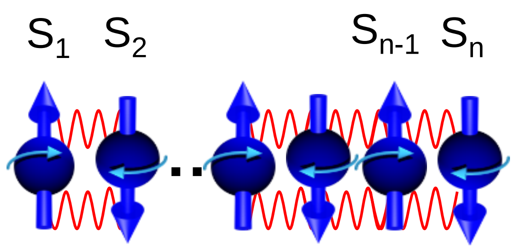



$E_{ex}=-J\mathbf{S}_i\mathbf{S}_j$




$\mathbf{M} = 0$

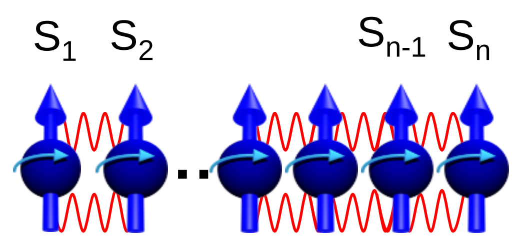
***Antiferromagnet (J<0)***



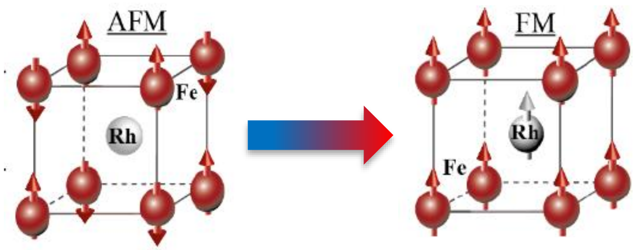


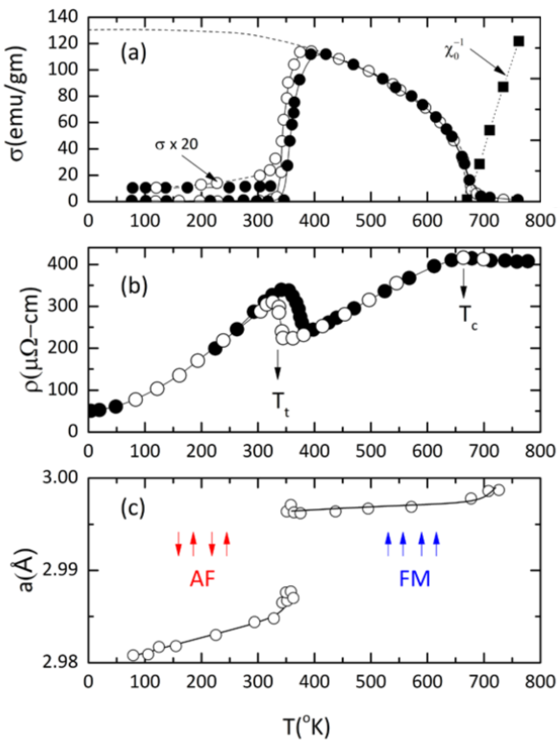
$\mathbf{M} = -\gamma \frac{\sum s_i}{V}$

***Ferromagnet (J>0)***



# Magneto-structural phase transition and chicken-or-egg causality dilemma in FeRh

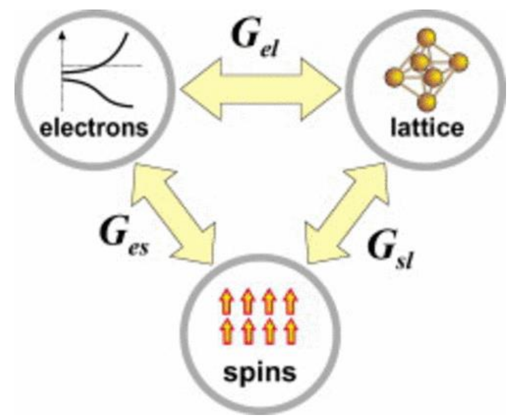



Magnetization

Electronic conductivity

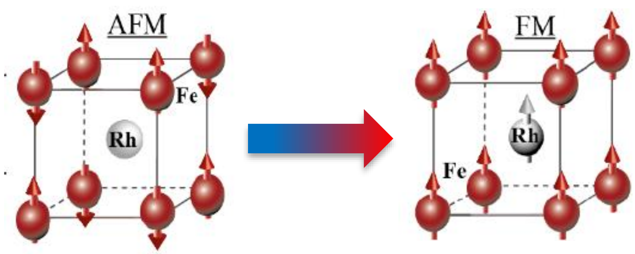
Lattice expansion




**Does magnetism drive lattice?**
**Does lattice drive magnetism?**
**Chicken-or-egg problem!**

L. H. Lewis, C. H. Marrows and S. Langbridge
*J. Phys. D: App. Phys.* **49** 323002 (2016).

# Magneto-structural phase transition and chicken-or-egg causality dilemma in FeRh




**1938**
**Discovery**
M. Fallot, Ann. Phys., Paris*,* **10**,
291-332 (1938).

**1960 (Lattice)**
**Model of exchange inversion**
C. Kittel, Phys. Rev B*,* **120,**
335-342 (1960).

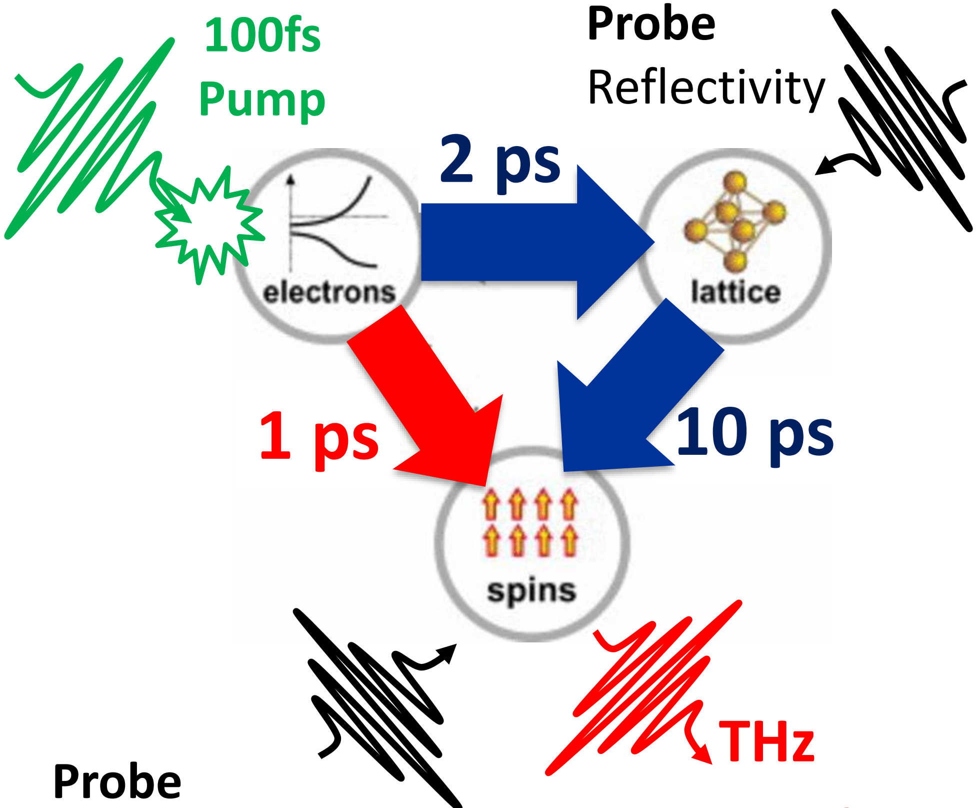
**1966 (Spins)**
**Entropy analysis**
J. C. Kouvel, J. Appl. Phys.,**137**,
1257-1258 (1966).

…

**2005 (Spins)**
**Bi-quadratic exchange**
O.N. Mryasov, Phase transitions **78**,
197-208 (2005).

**2020 (Lattice)**
**Phonon spectrum**
M. P. Belov et al,
Phys. Rev. B **101**, 13430 (2020).

# Simple idea to resolve chicken-or-egg problem



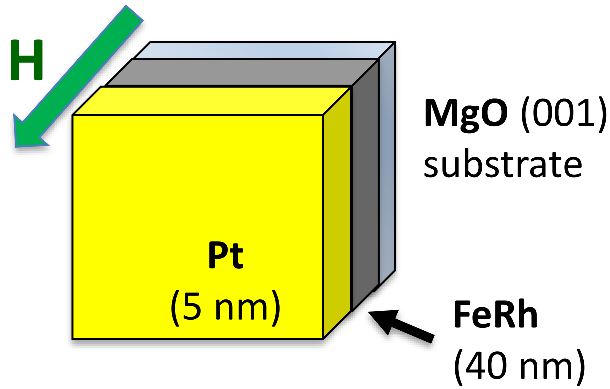

**Probe**
Magneto-optical
Kerr effect

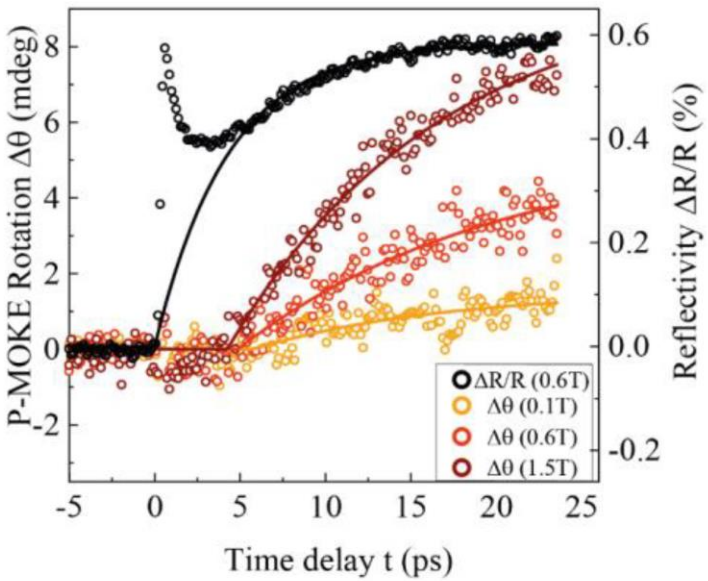
**magnetism changes first and drives the lattice**

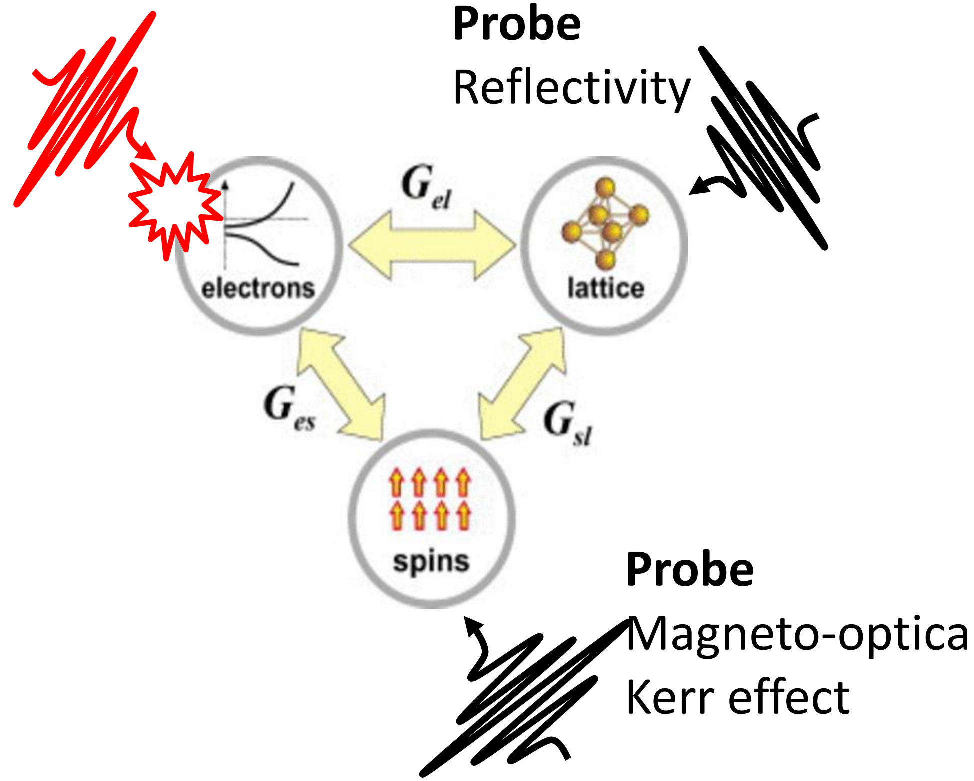
$$E(t) \sim \frac{\partial^2 M}{\partial t^2}$$

E. Beaurepaire et al, APL **84**, 3465 (2004).

# Dynamics of spins and lattice in low magnetic fields



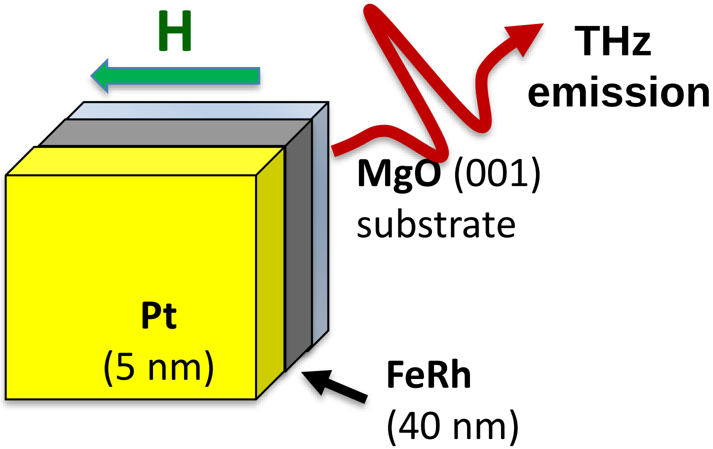



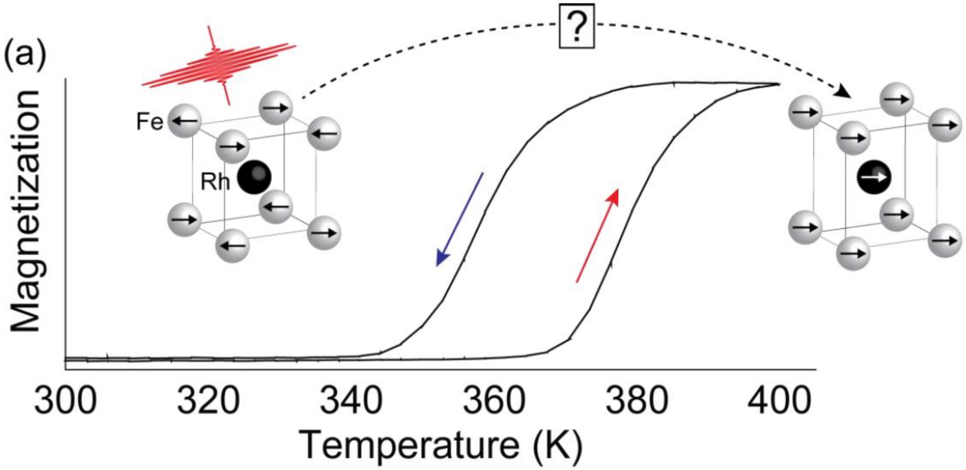



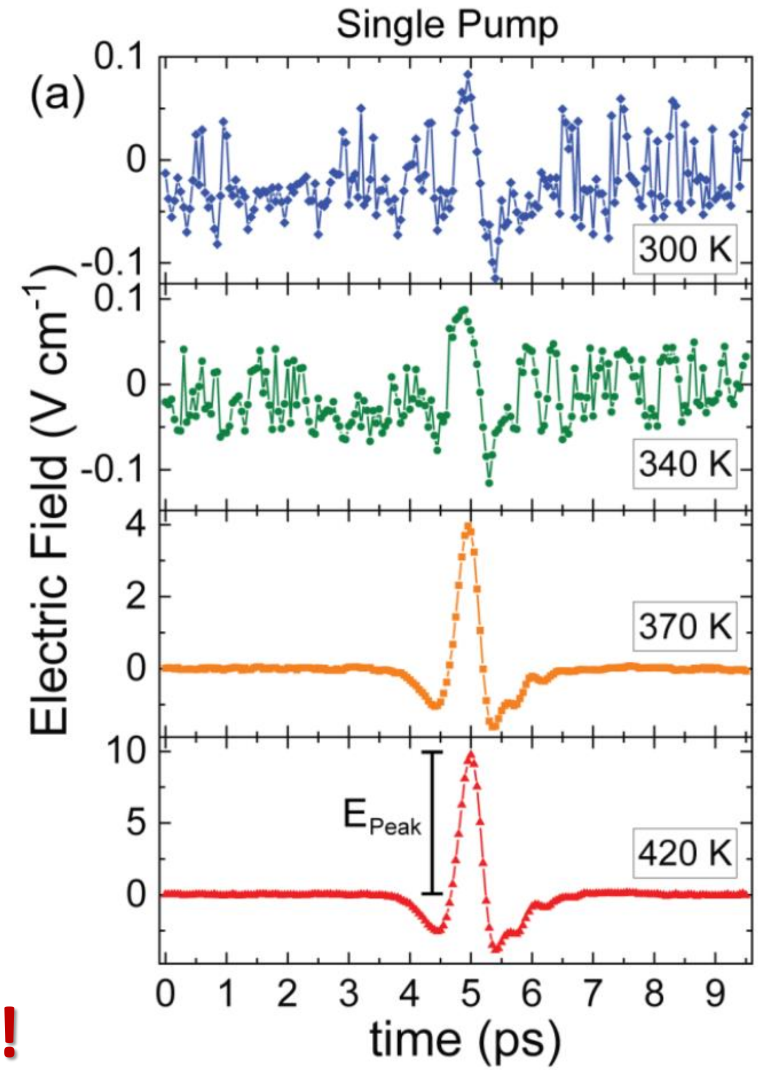

**Lattice is faster!**

I. Dolgiih et al, (submitted).
https://arxiv.org/abs/2202.03931

# Dynamics of spins as seen in THz spectroscopy



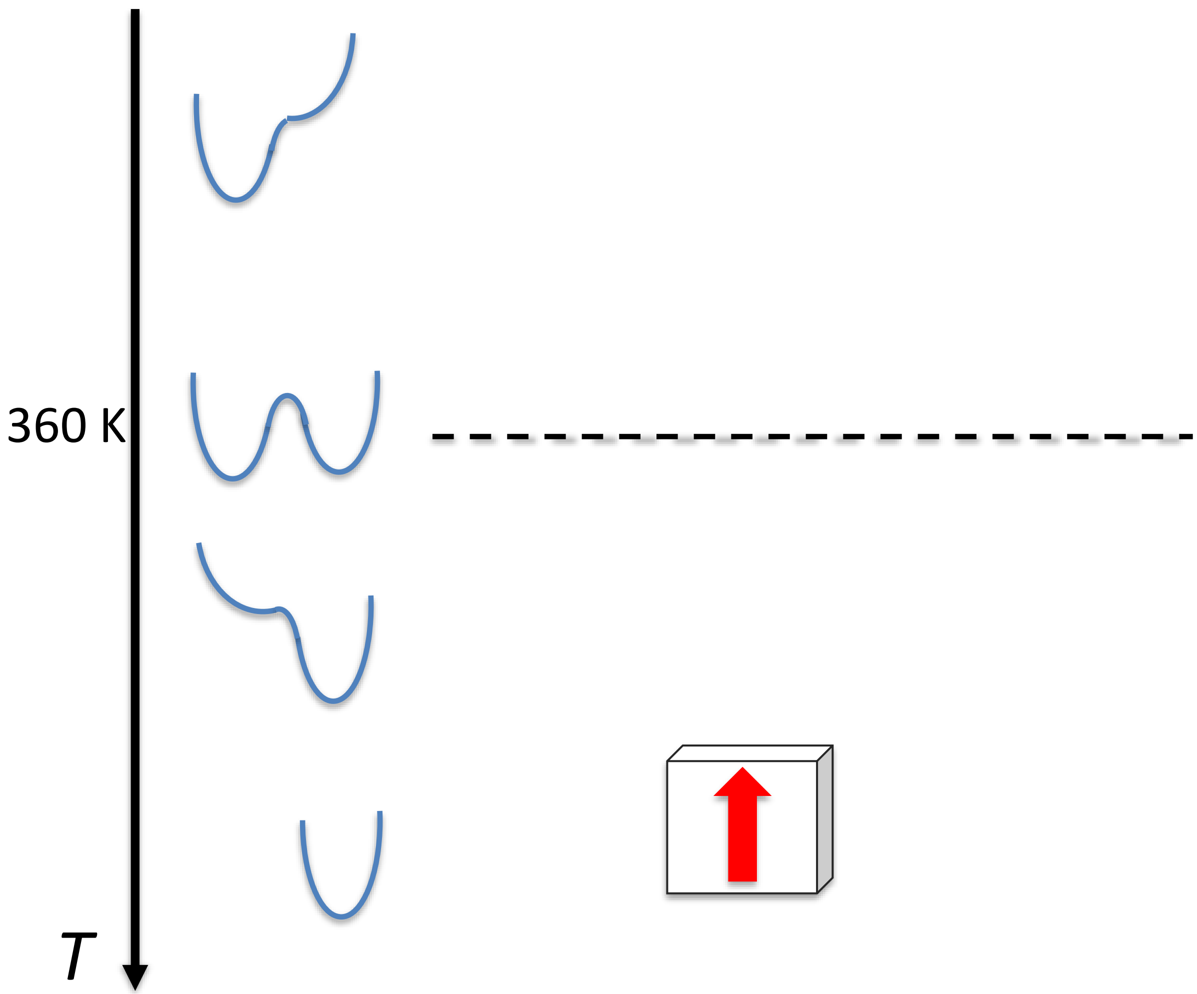



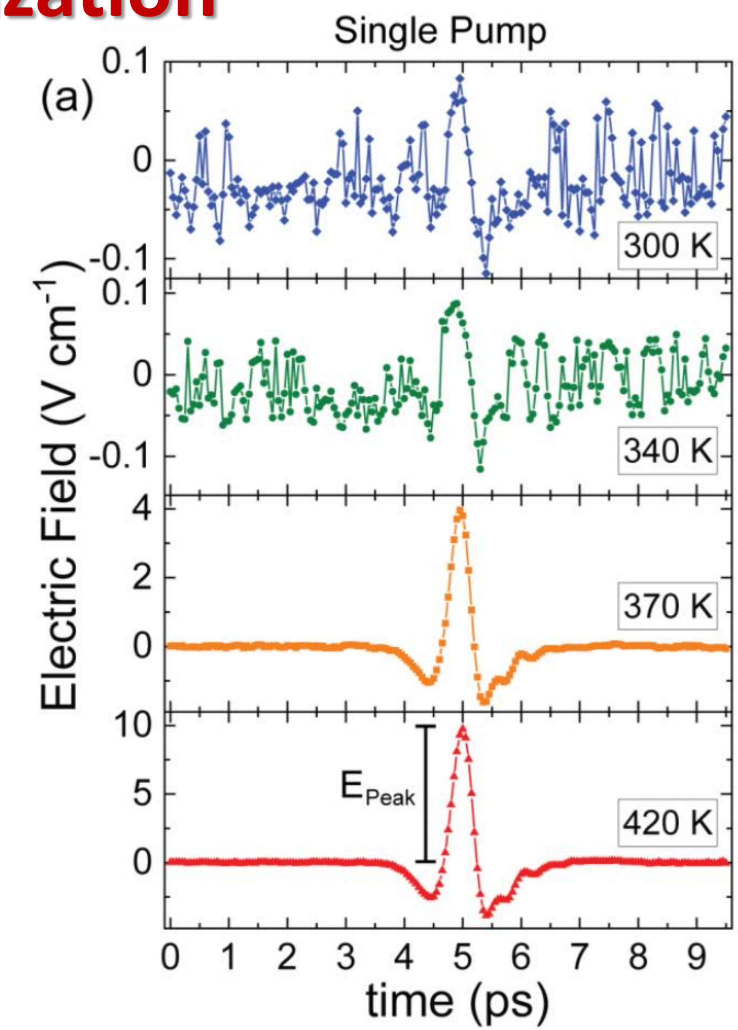




R. Medapalli et al,
Appl. Phys. Lett. **117**, 142406 (2020).

**Spins are faster!**

# THz emission from FeRh/Pt as a result of ultrafast demagnetization




G. Li et al, Nature Comm (accepted). arXiv:2001.06799v3.

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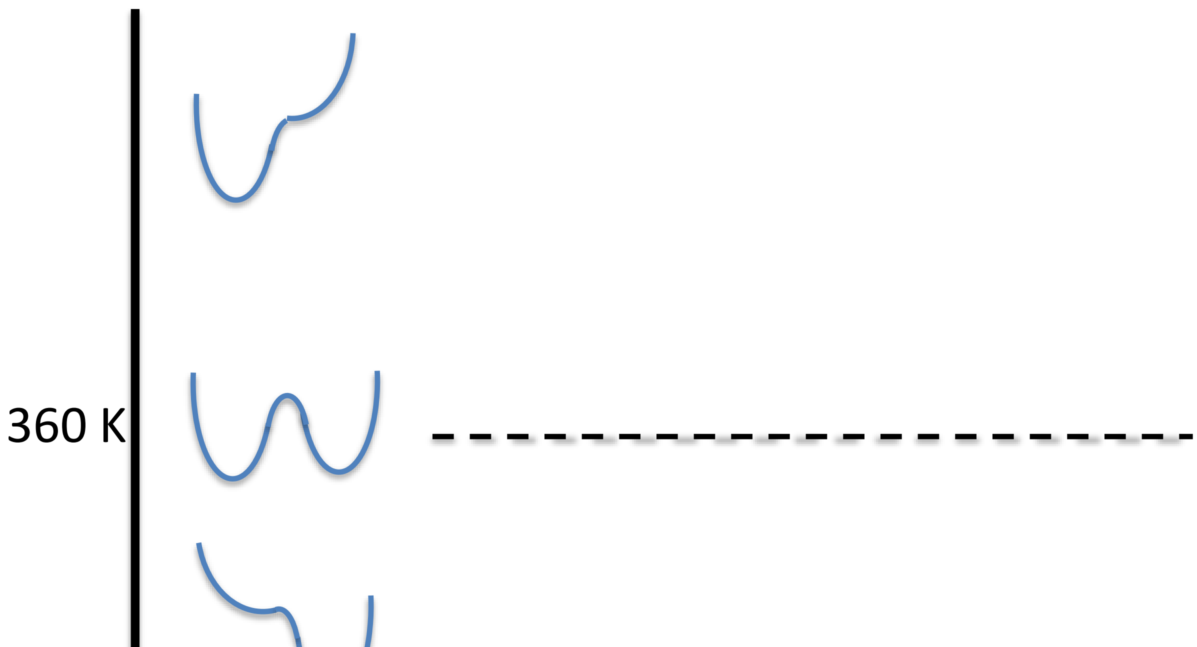


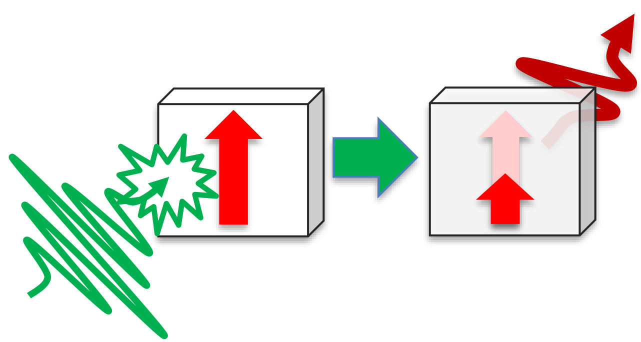

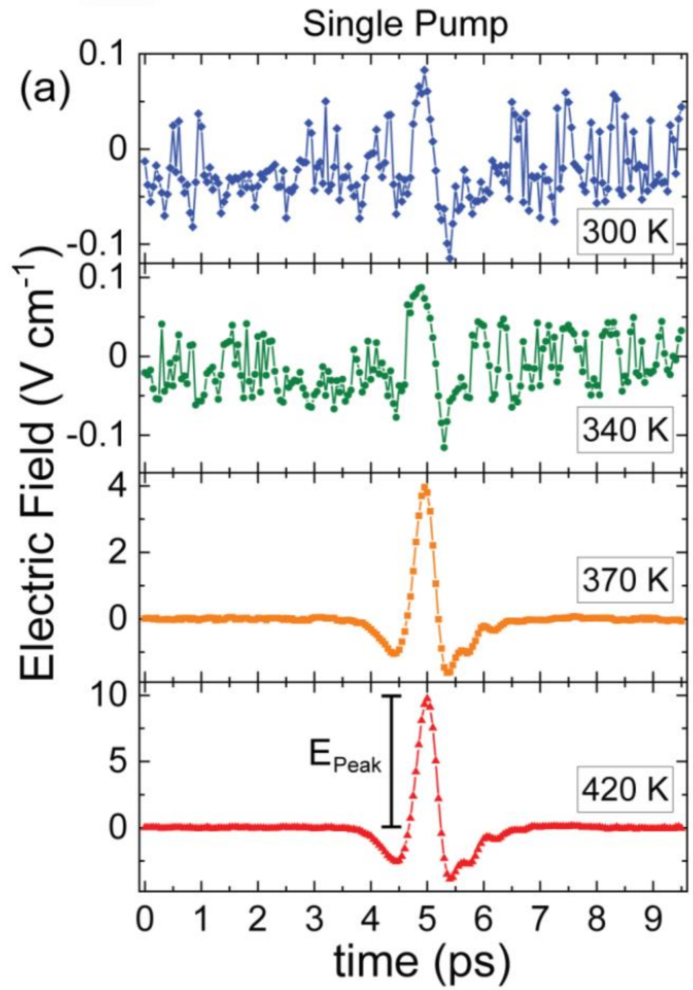


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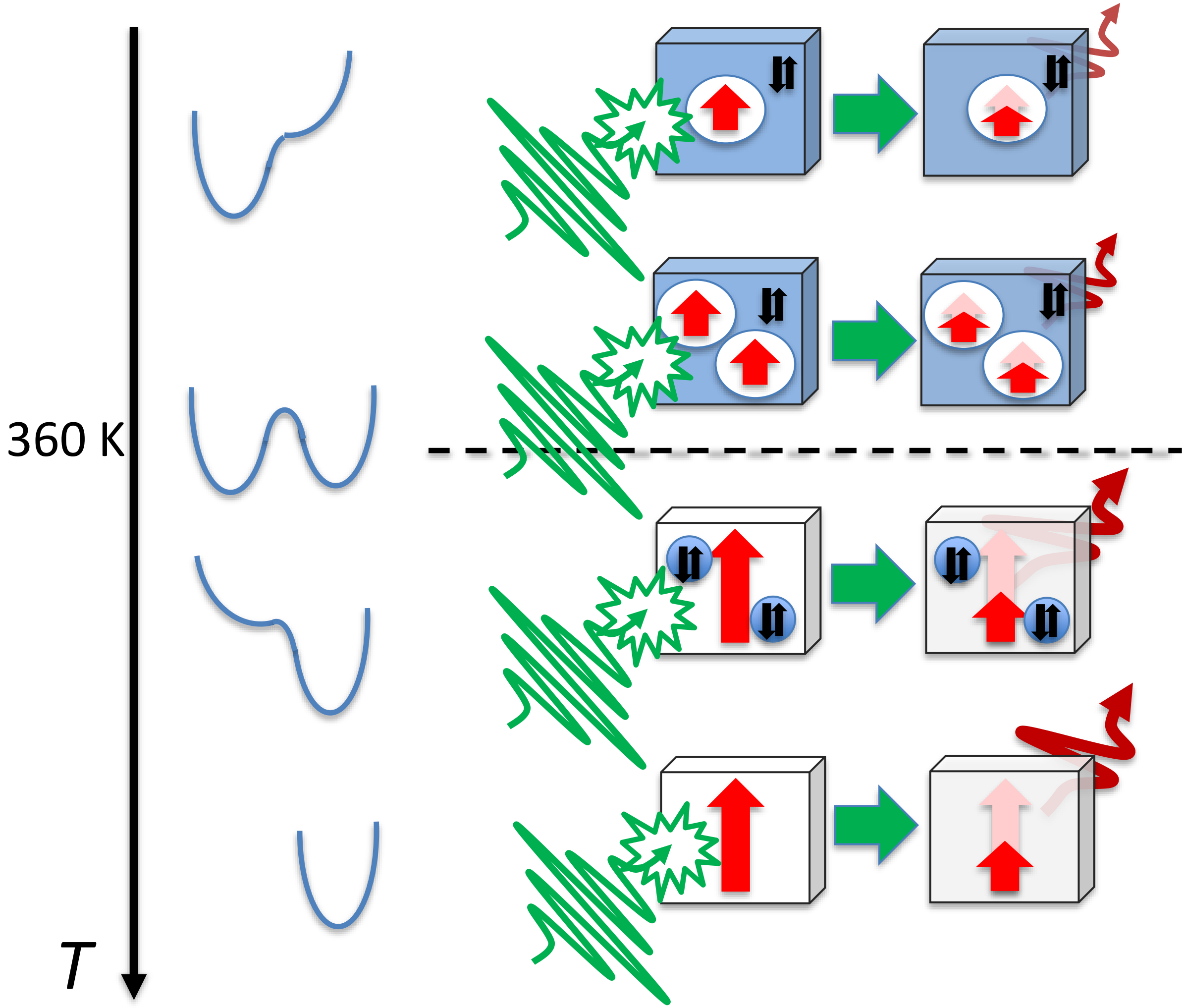


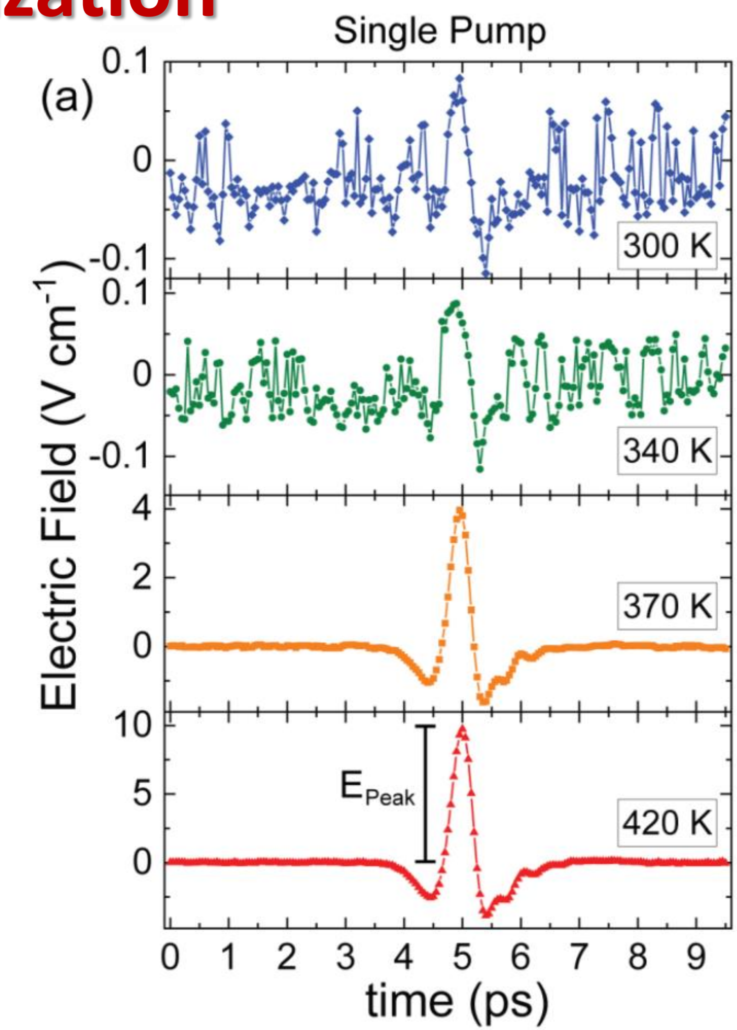


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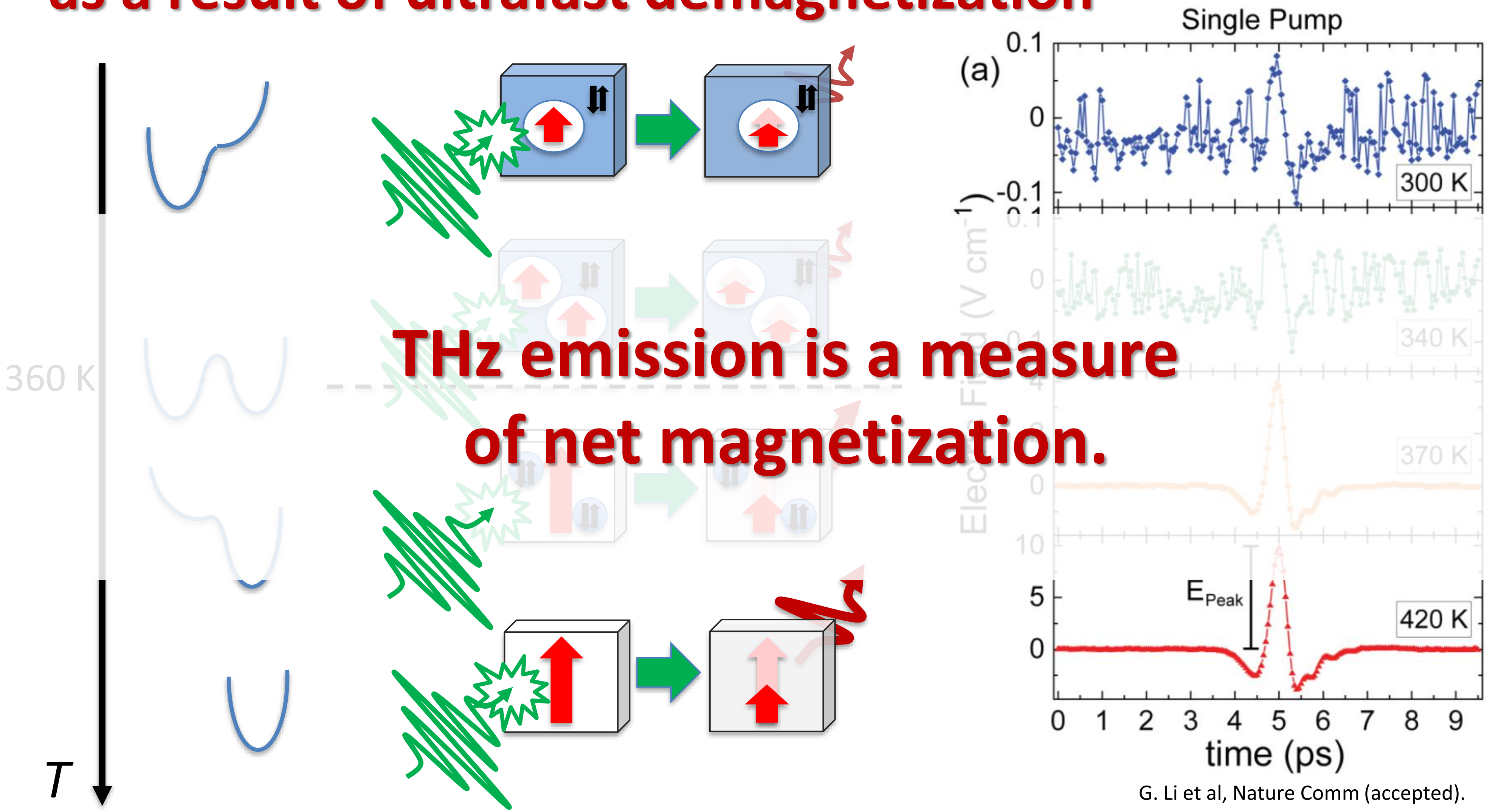
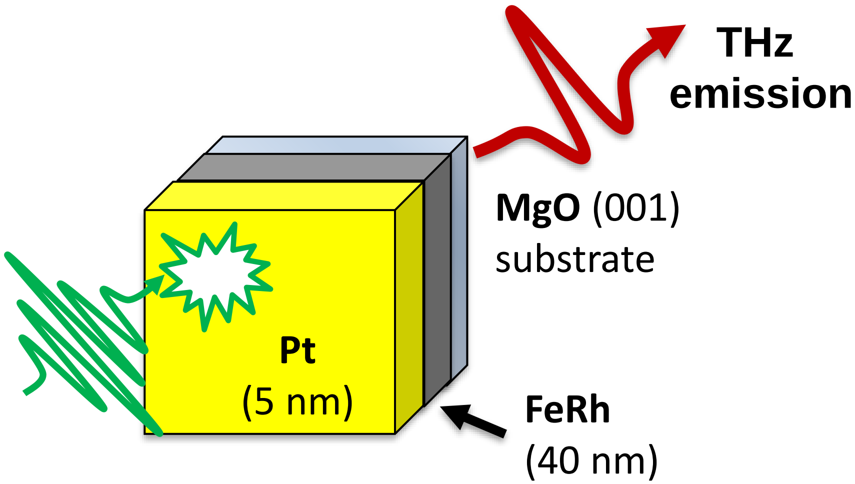
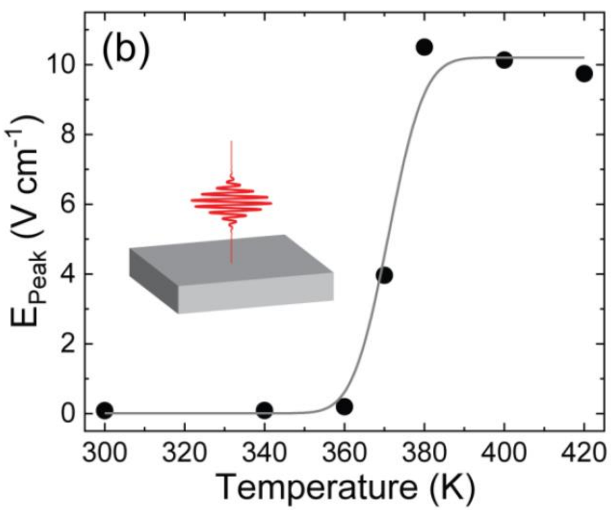


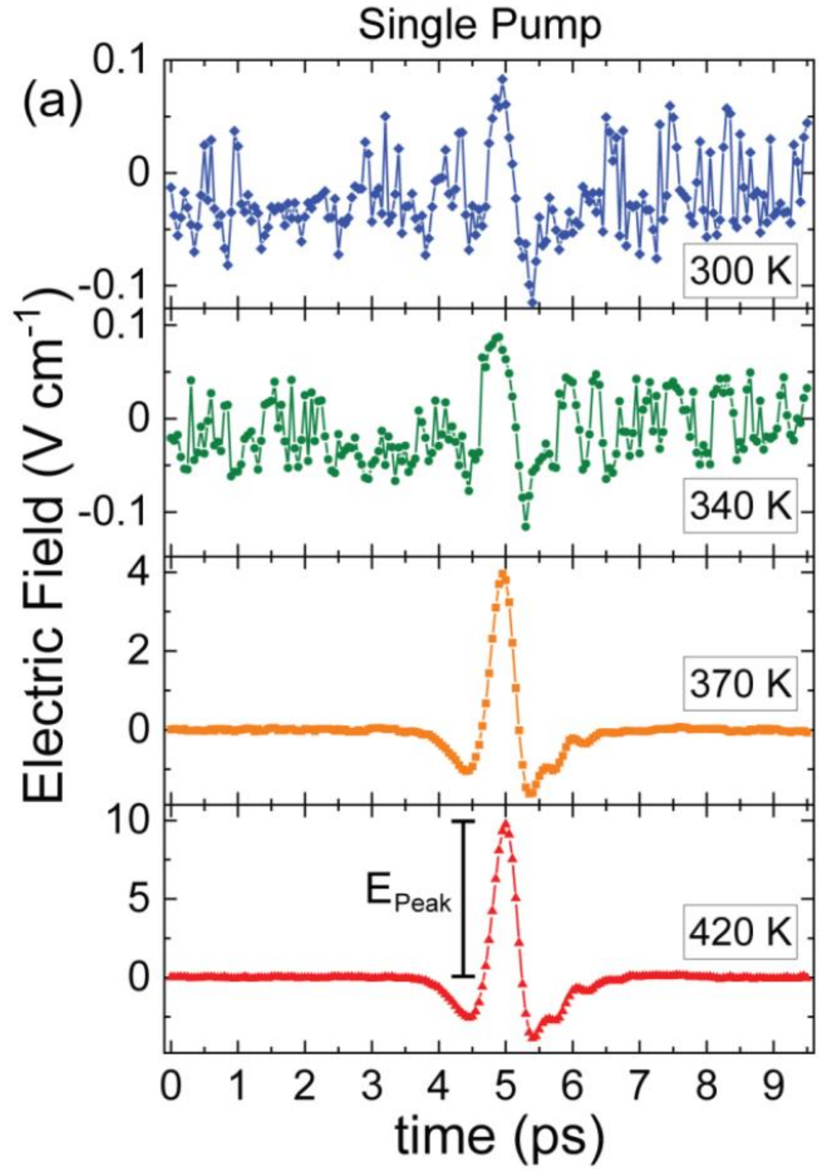

**THz emission is a measure of net magnetization.**

G. Li et al, Nature Comm (accepted). arXiv:2001.06799v3.

# Studied FeRh/Pt bilayer

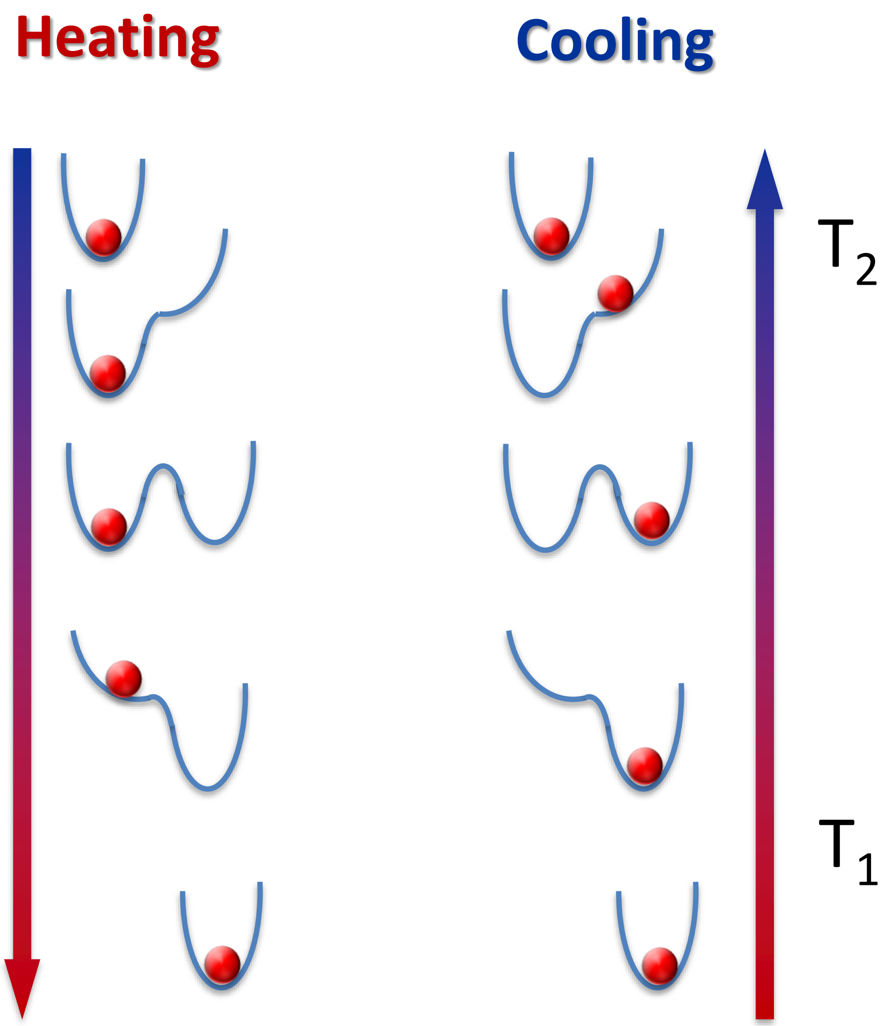


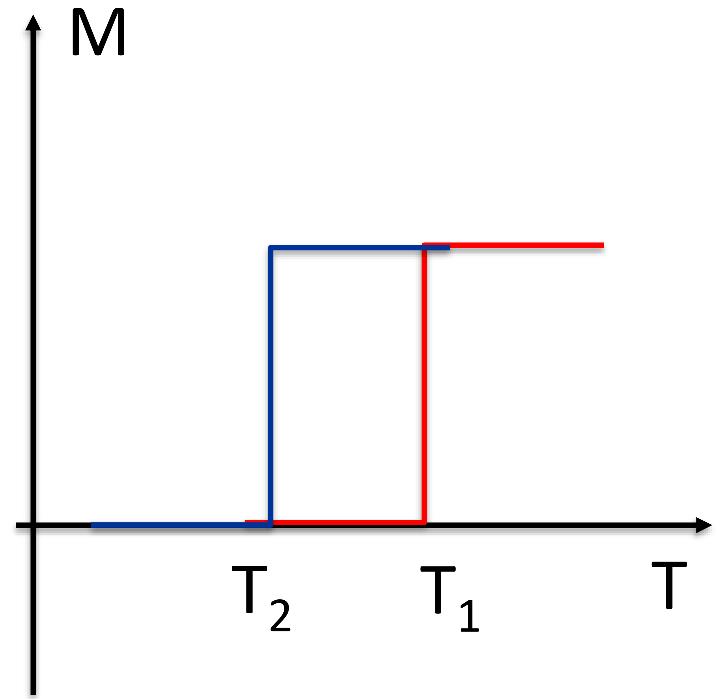




G. Li et al, arXiv:2001.06799v3.

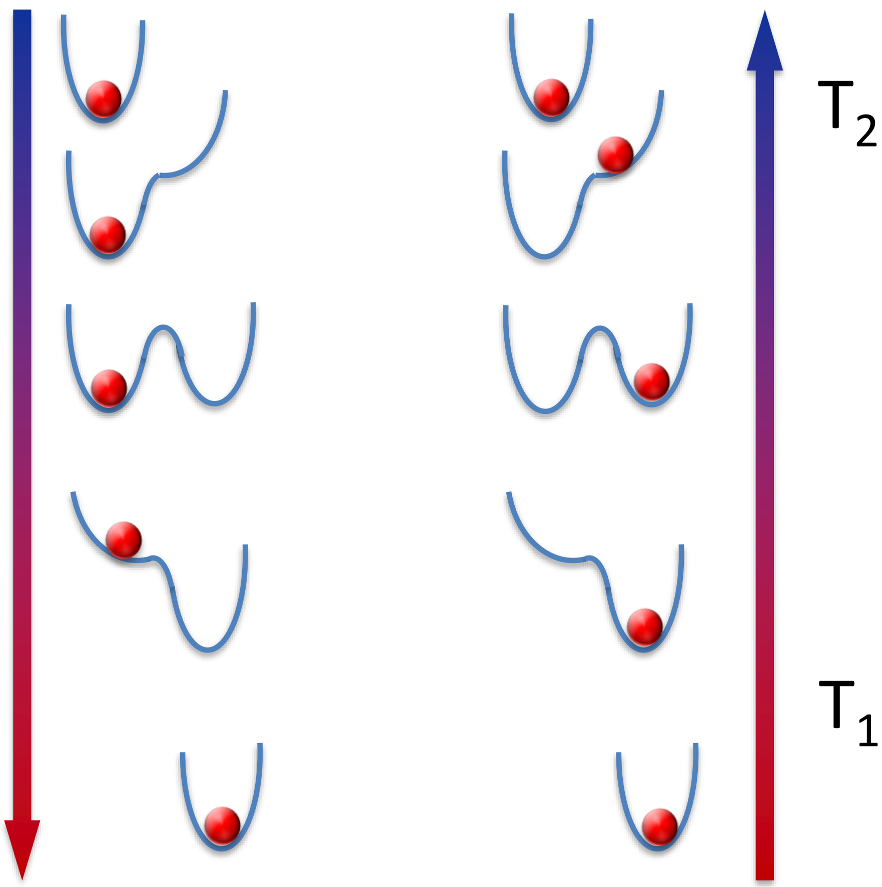
# Origin of hysteresis

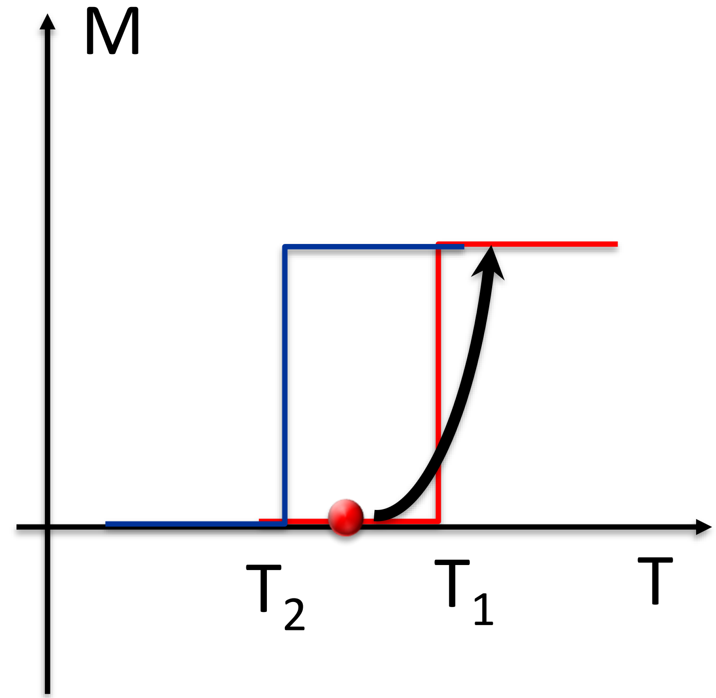


# Disappearance of the hysteresis as a result of uncontrolled relaxation

**Heating** **Cooling**



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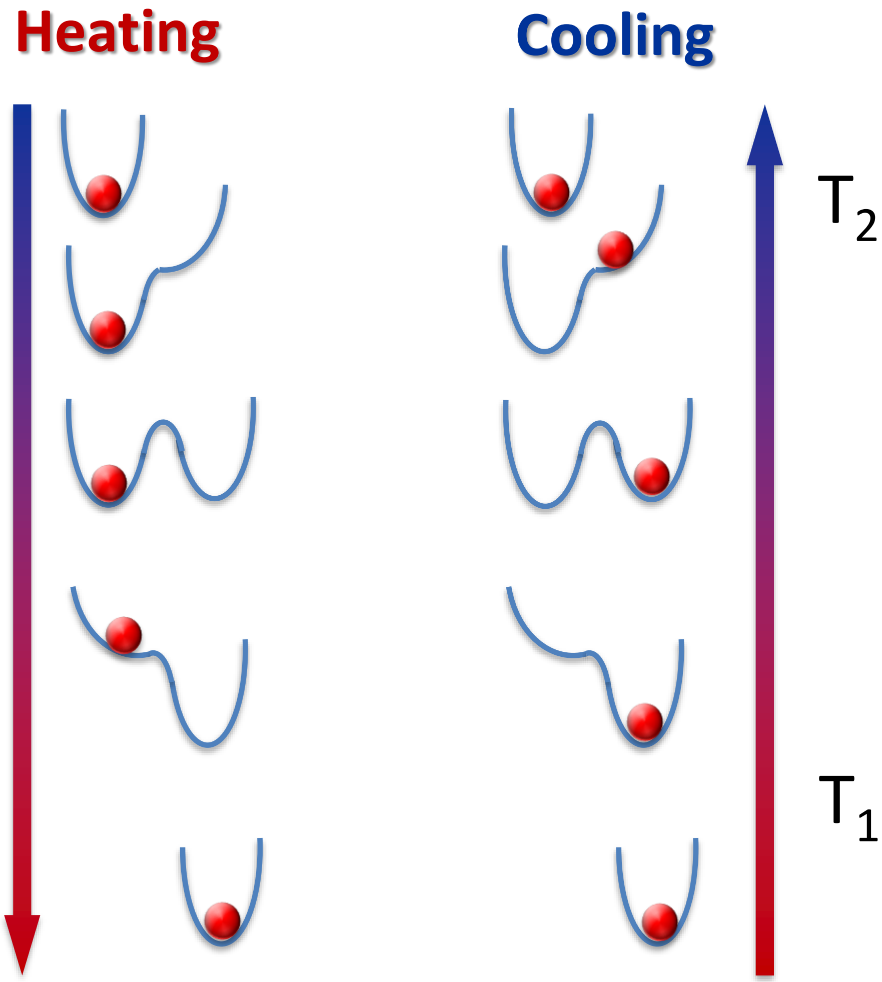


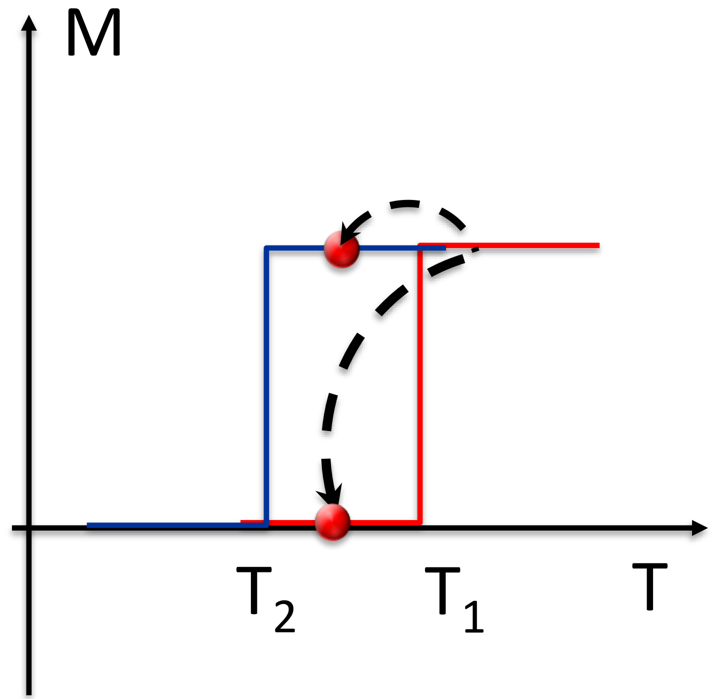

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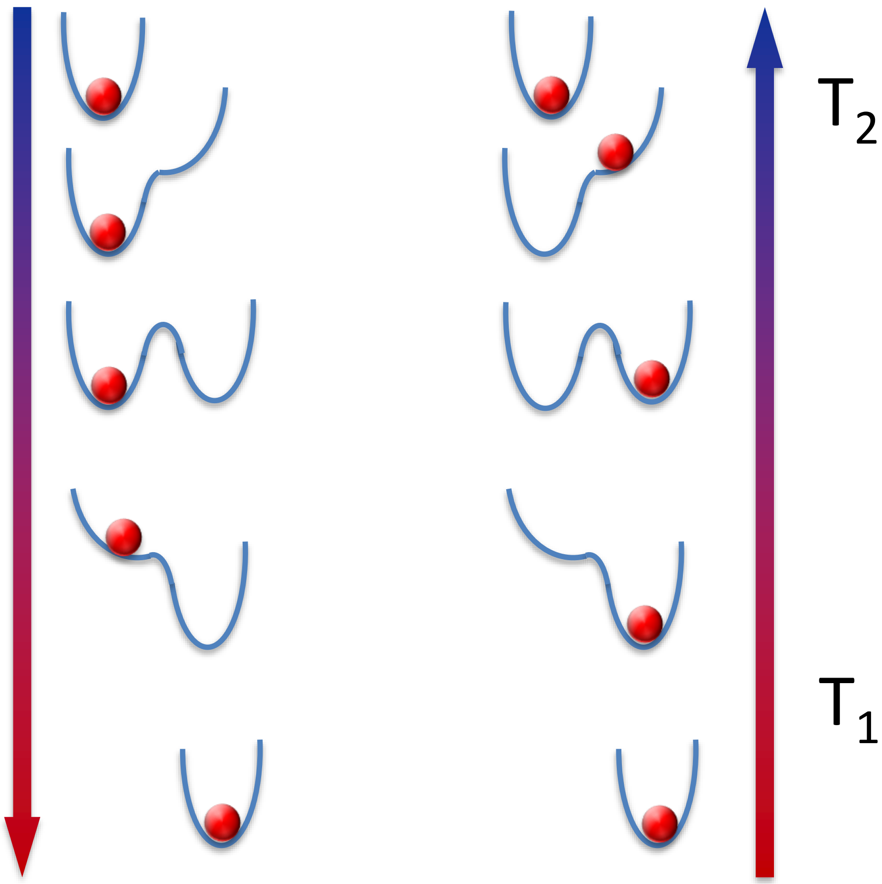


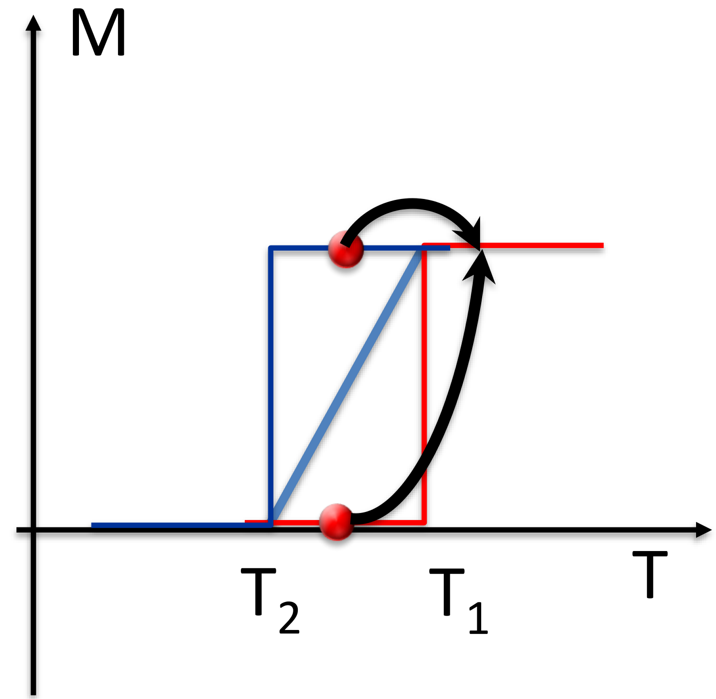

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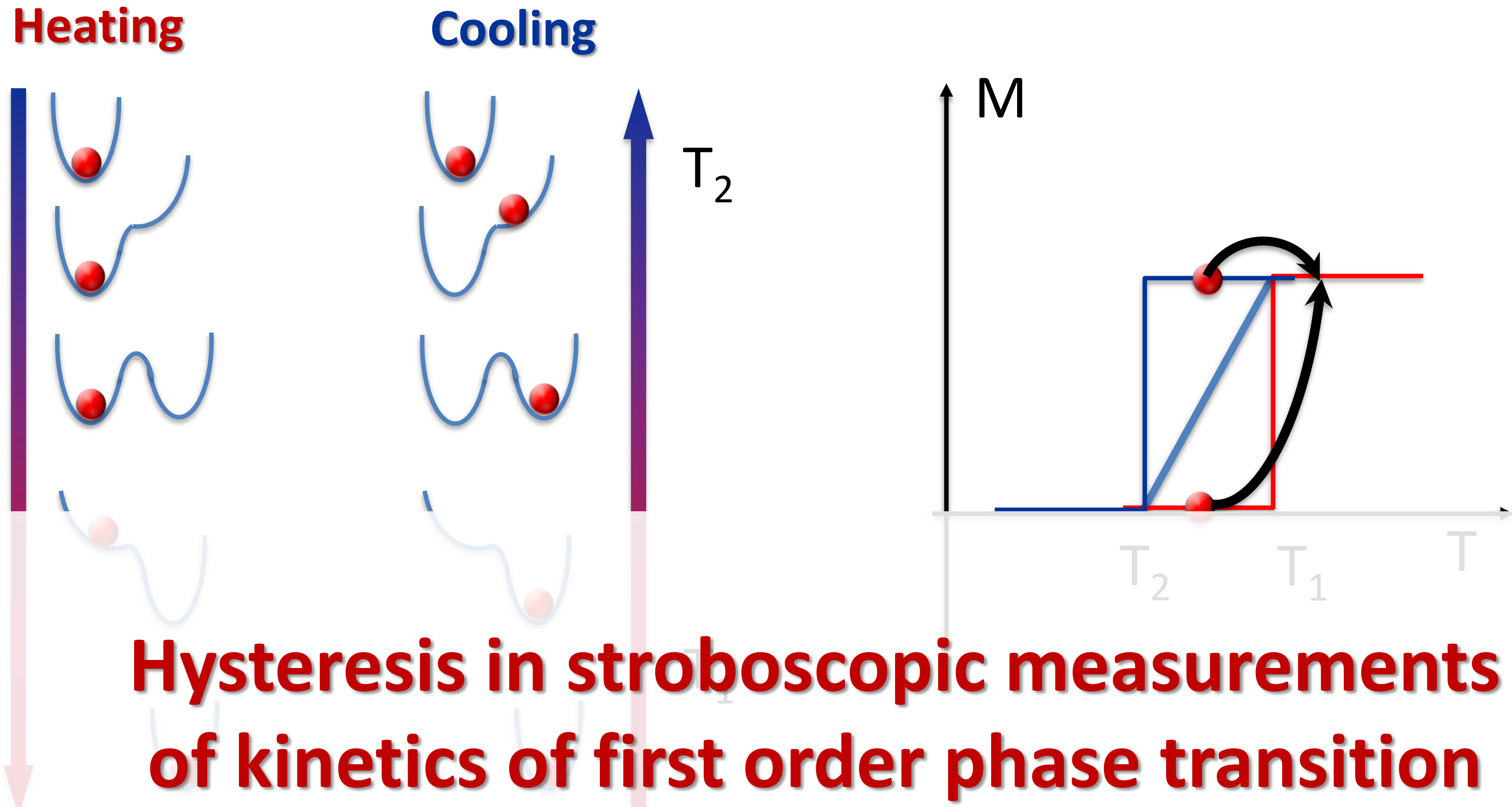
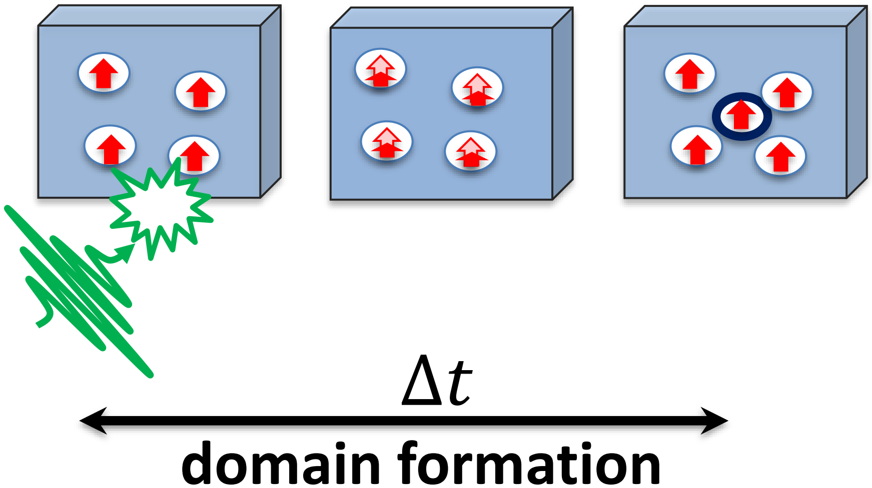



**Hysteresis in stroboscopic measurements of kinetics of first order phase transition is rather anomaly than law.**

# How can we see the formation and growth of ferromagnetic domains?



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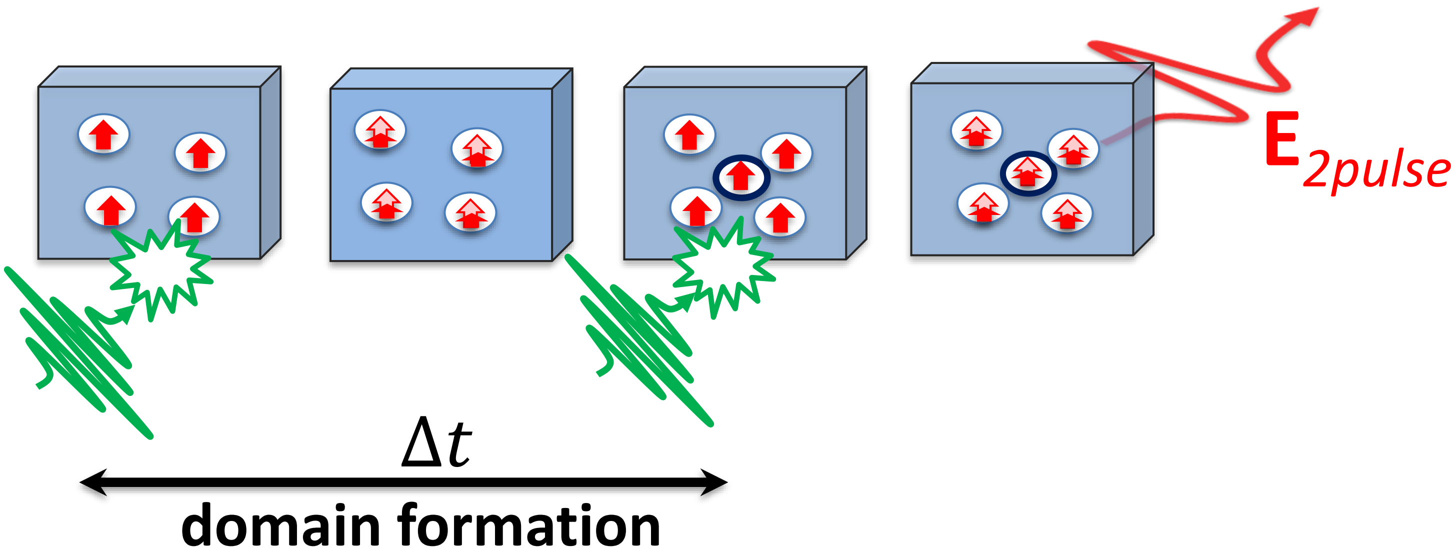

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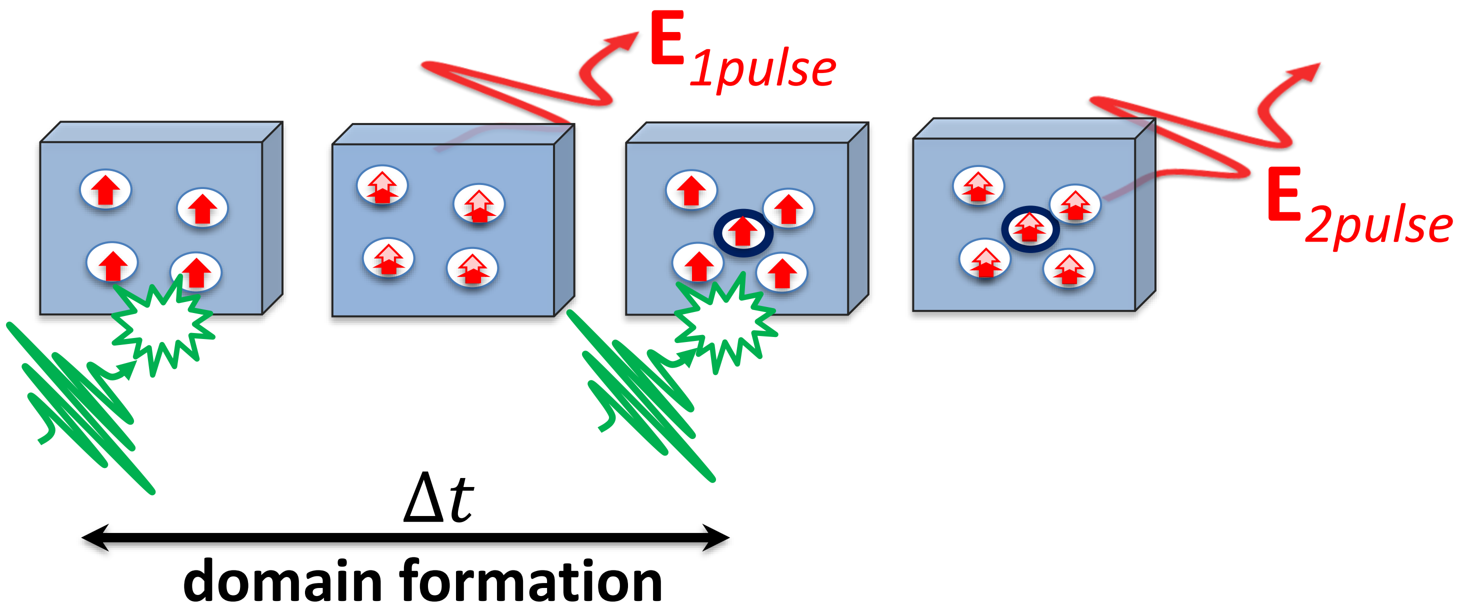
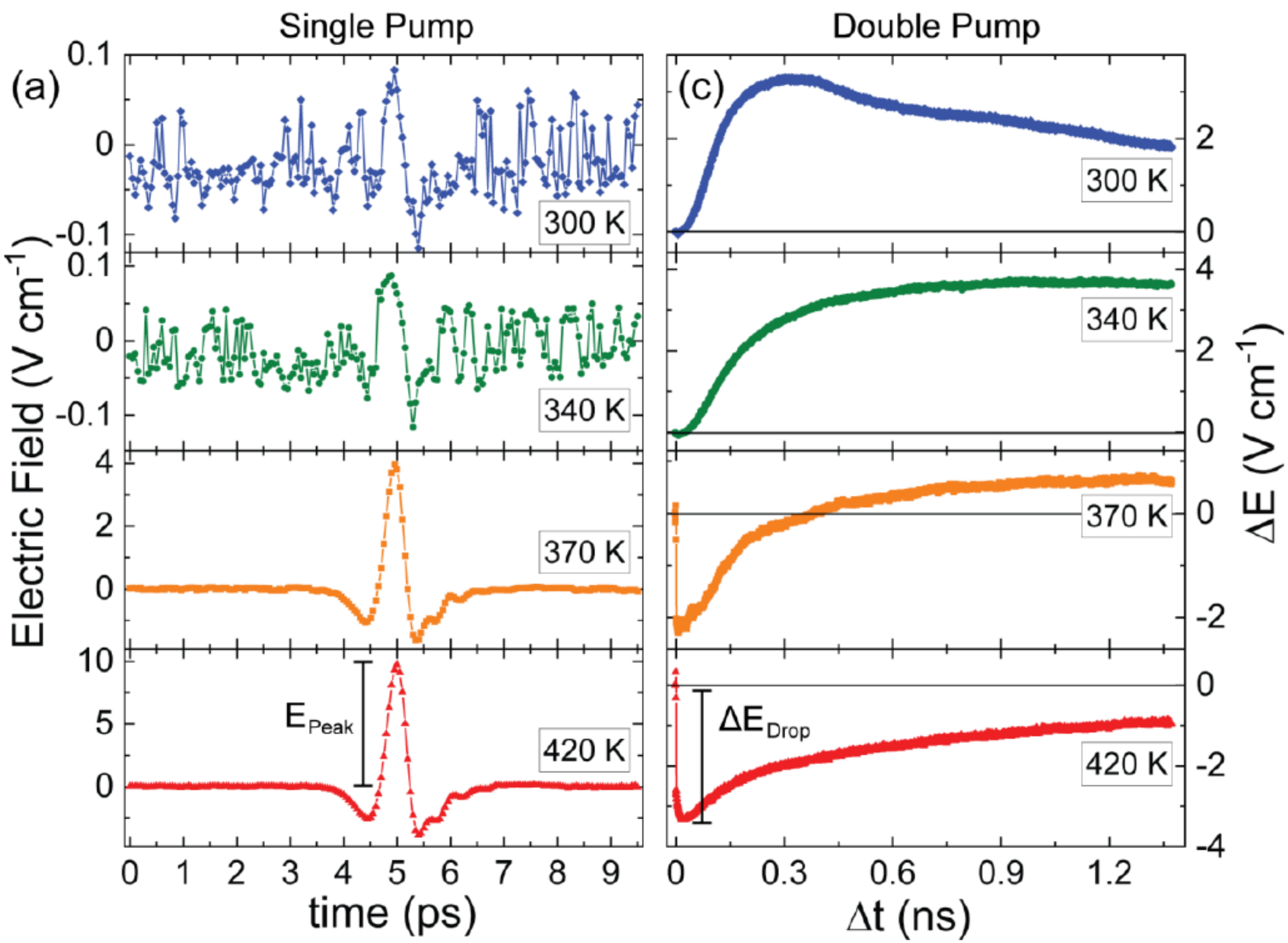


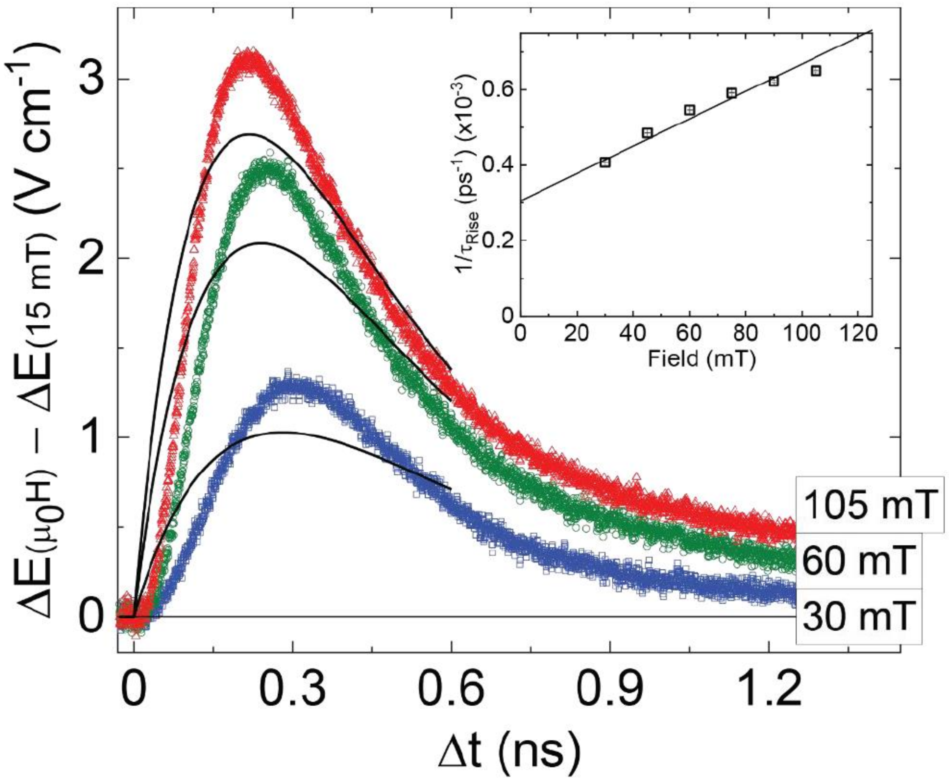

**$E_{2pulse}$-$E_{1pulse}$ contains information about domains formed between the pump pulses.**

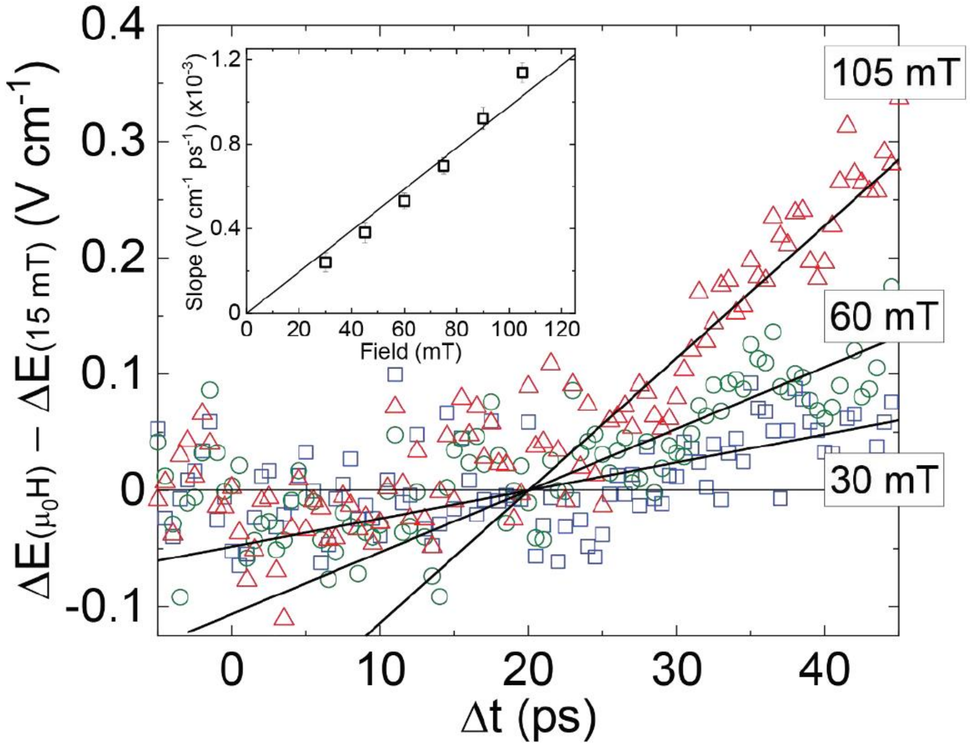
# Single *vs* Double pump THz emission spectroscopy




G. Li et al, Nature Comm (accepted).
arXiv:2001.06799v3.

# THz emissivity of ferromagnetic domains in FeRh/Pt




**It takes 10-20 ps to nucleate a ferromagnetic domain in FeRh**

G. Li et al, Nature Comm (accepted).
arXiv:2001.06799v3.

# Mean field theory

$\boldsymbol{M_1}, \boldsymbol{M_2}$ Iron sublattice magnetizations, $\boldsymbol{M_1}, \boldsymbol{M_2} \sim 3\mu_B$ at transition temperature ($\sim$ 350 K)

$$F = \underbrace{f(M_1^2) + f(M_2)^2}_{\text{Non-equilibrium exchange energy density}} - \underbrace{J_{FF}^{(2)}(\mathbf{M}_1 \cdot \mathbf{M}_2)^2}_{\text{4-spin exchange (bi-quadratic exchange)}} - \underbrace{(J_{FF}^{(1)} + \rho_J u)\mathbf{M}_1 \cdot \mathbf{M}_2}_{\text{Lattice expansion (u) dependent Heisenberg exchange}} - \underbrace{\mathbf{H_0} \cdot (\mathbf{M}_1 + \mathbf{M}_2)}_{\text{Zeeman energy}} - \underbrace{\beta^{-1} \ln 2 \cosh(\beta\Delta)}_{\text{Ising-like paramagnetic rhodium ion}} + \underbrace{\frac{1}{2}\epsilon u^2}_{\text{Elastic stiffness (Hooke's law)}}$$

$$\left.\begin{array}{ll} \Delta = \mu_{Rh}|J_{FR}(\boldsymbol{M_1} + \boldsymbol{M_2}) + \boldsymbol{H_0}| & \mu_{Rh} \sim 1\,\mu_B \\ \rho_J \equiv \partial J_{FF}^1/\partial u & \langle u \rangle = \rho_J^2 \boldsymbol{M_1} \cdot \boldsymbol{M_2}/\epsilon \end{array}\right\} \text{in equilibrium}$$

$$F_{eq} = -\left(J_{FF}^{(2)} + \frac{\rho_J^2}{2\epsilon}\right)(\mathbf{M}_1 \cdot \mathbf{M}_2)^2 - J_{FF}^{(1)}\mathbf{M}_1 \cdot \mathbf{M}_2 - \mathbf{H_0} \cdot (\mathbf{M}_1 + \mathbf{M}_2) - \beta^{-1} \ln 2 \cosh(\beta\Delta) + \underbrace{K\left(\frac{(\mathbf{M}_1 \cdot \hat{\mathbf{z}})^2}{\mathbf{M}_1^2} + \frac{(\mathbf{M}_2 \cdot \hat{\mathbf{z}})^2}{\mathbf{M}_2^2}\right)}_{\text{Added uniaxial anisotropy}}$$

$$\frac{1}{\gamma}\frac{d}{dt}\mathbf{M}_{1,2} = \mu\lambda_{so}\mathbf{H}_{1,2} + \mu\lambda_{ex}(\mathbf{H}_{1,2} - \mathbf{H}_{2,1}) + \mathbf{M}_{1,2} \times \mathbf{H}_{1,2}. \qquad \mathbf{H}_i \equiv -\frac{\delta F}{\delta \mathbf{M}_i}$$

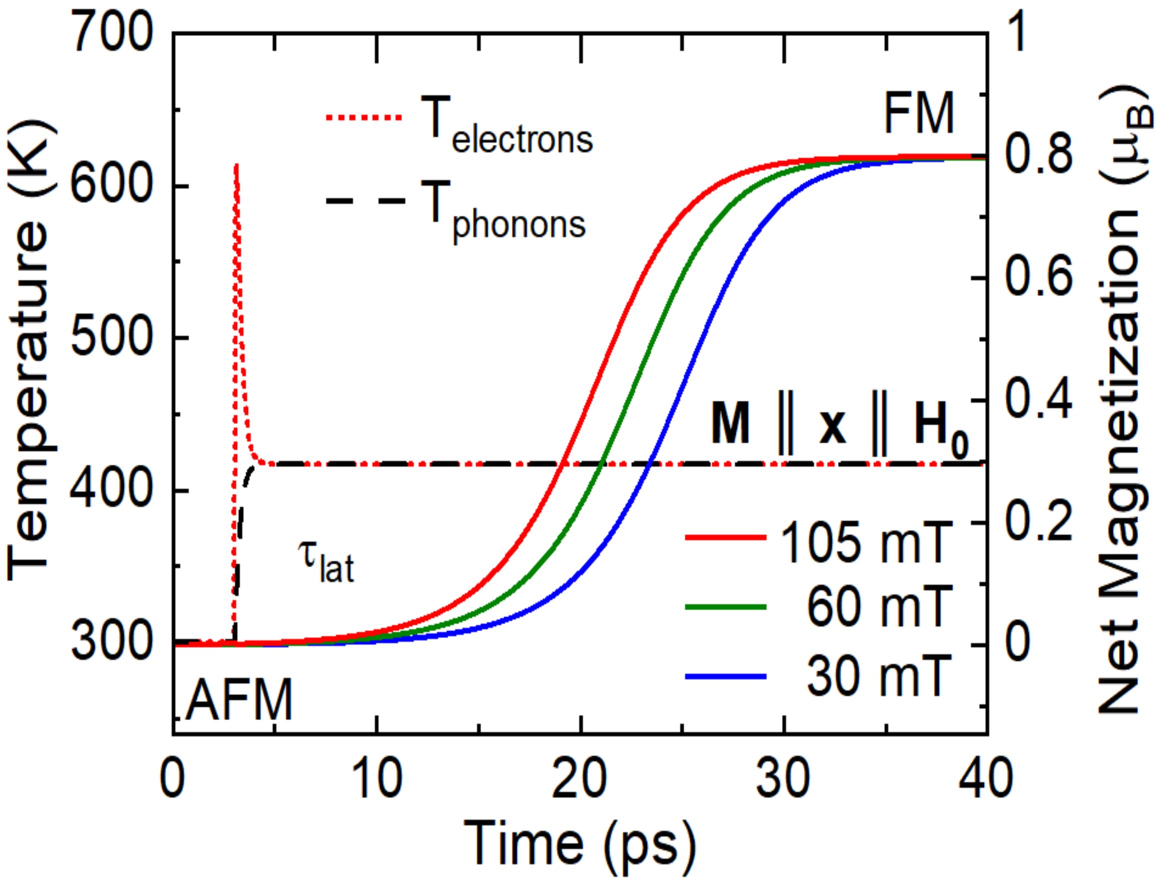
$\gamma$ – gyromagnetic ratio

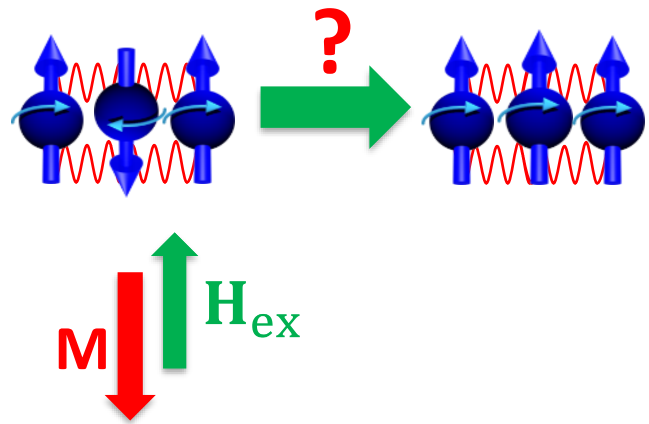
$\lambda_{ex}$ – exchange mediated angular momentum transfer between the Fe-sublattices.

$\lambda_{so}$ – angular momentum transfer to the lattice.

# Picosecond dynamics

**Assumption:** Laser-induced heating causes ultrafast expansion of the lattice $u(T)$ and thus changes the exchange interaction.




$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{\mathrm{ex}} = \mathbf{0}$$

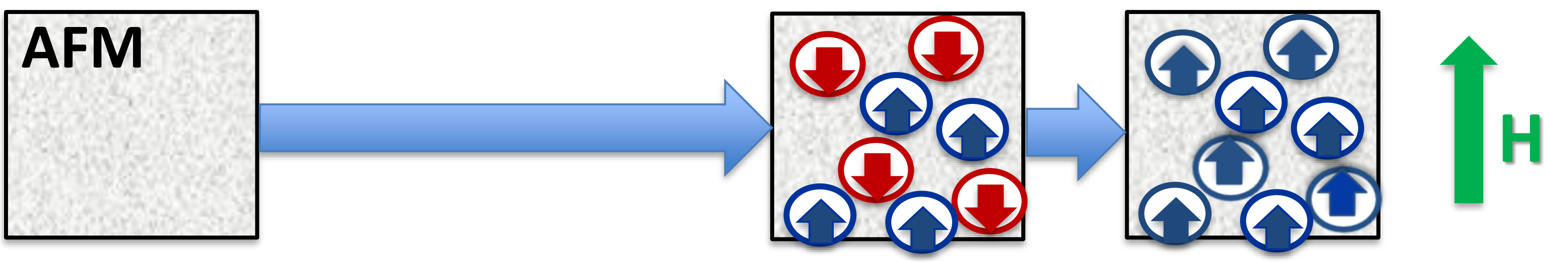
# Emergence of magnetization in FeRh

| fs excitation | Electron-Phonon interaction | | Spin Lattice interaction | Cooling down |
|---|---|---|---|---|

1 ps — 10 ps — 100 ps — 1 ns — time

Hot electrons

Spin precession period in fields 0.01-0.1 T




**Induction of M**

$\Sigma M \neq 0$

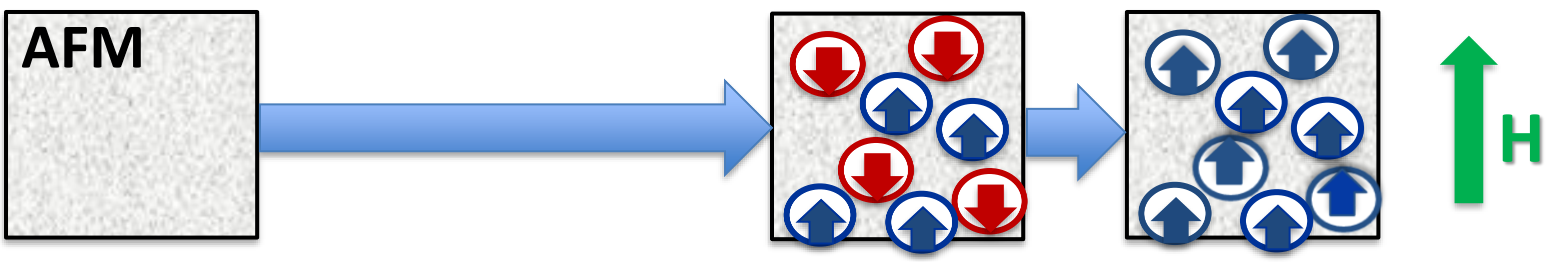
# Emergence of magnetization in FeRh

fs excitation | Electron-Phonon interaction | Spin Lattice interaction | Cooling down

1 ps | 10 ps | 100 ps | 1 ns | time

Hot electrons

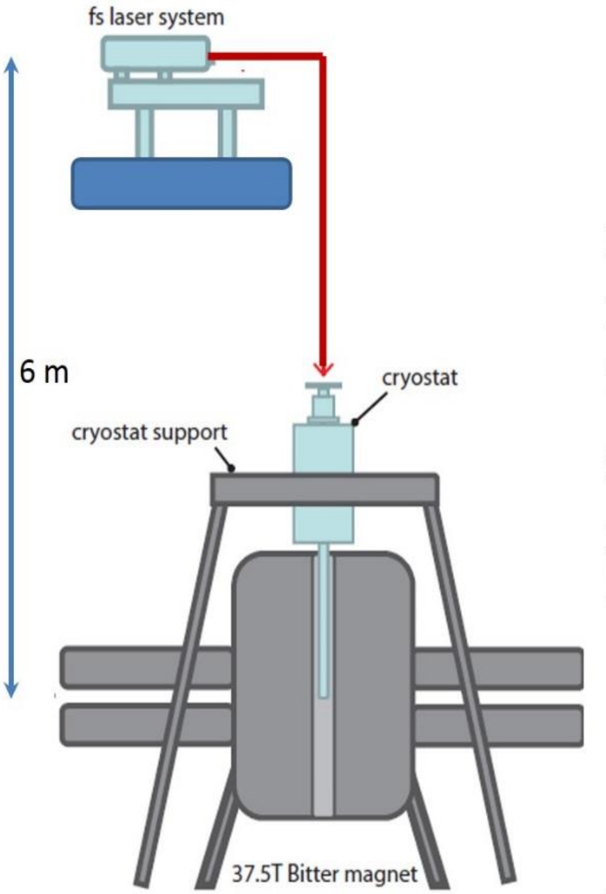
Spin precession period in fields ~~0.01-0.1 T~~ 1-30T



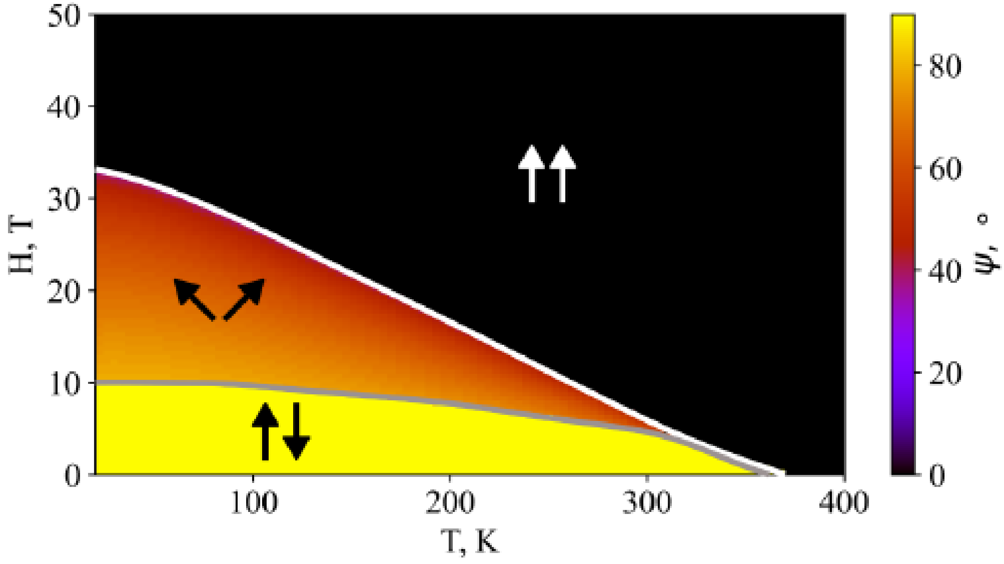

Induction of M

ΣM≠0

# Time-resolved measurements in 38 Tesla

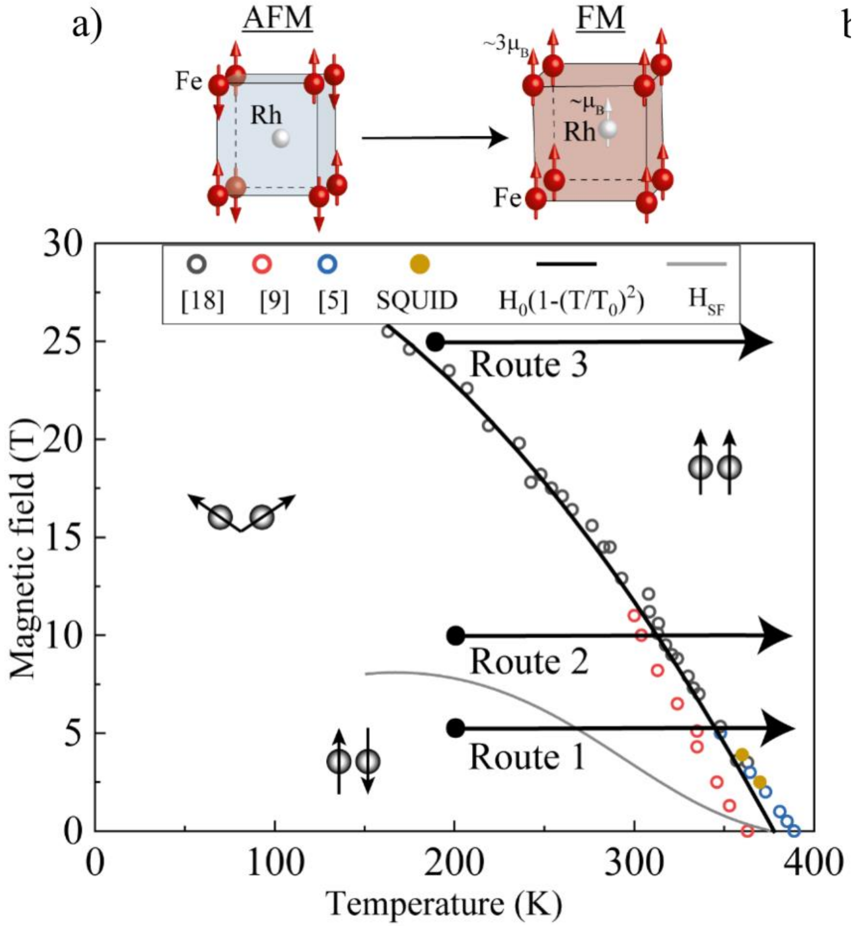


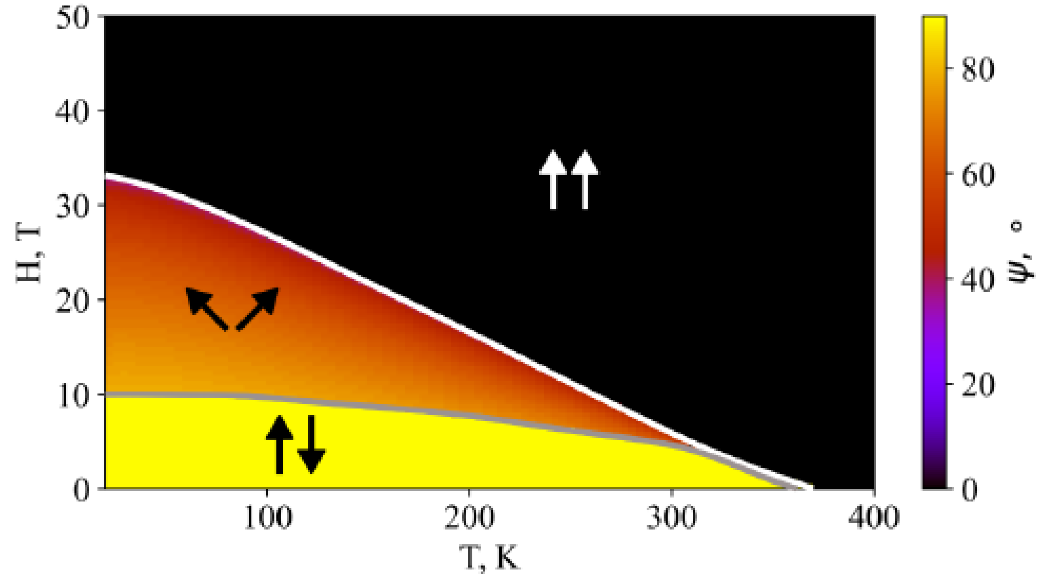

PhD thesis of J. Becker (Nijmegen).



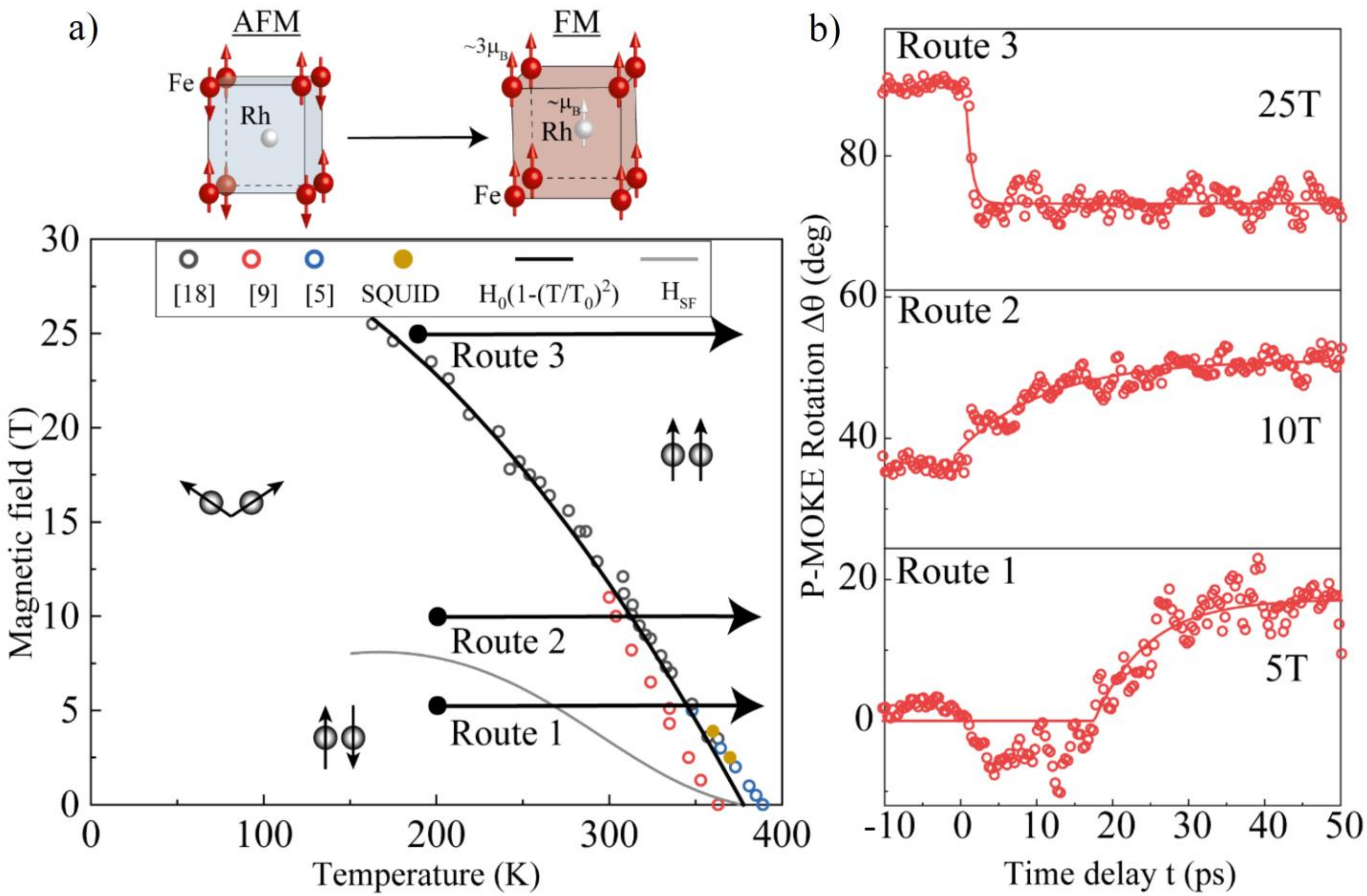

J. B. McKinnon et al,
J. Phys. C: Solid State Phys. **3**, S46 (1970).

# Magnetism of FeRh in high magnetic fields



# Ultrafast magnetism of FeRh in high magnetic fields



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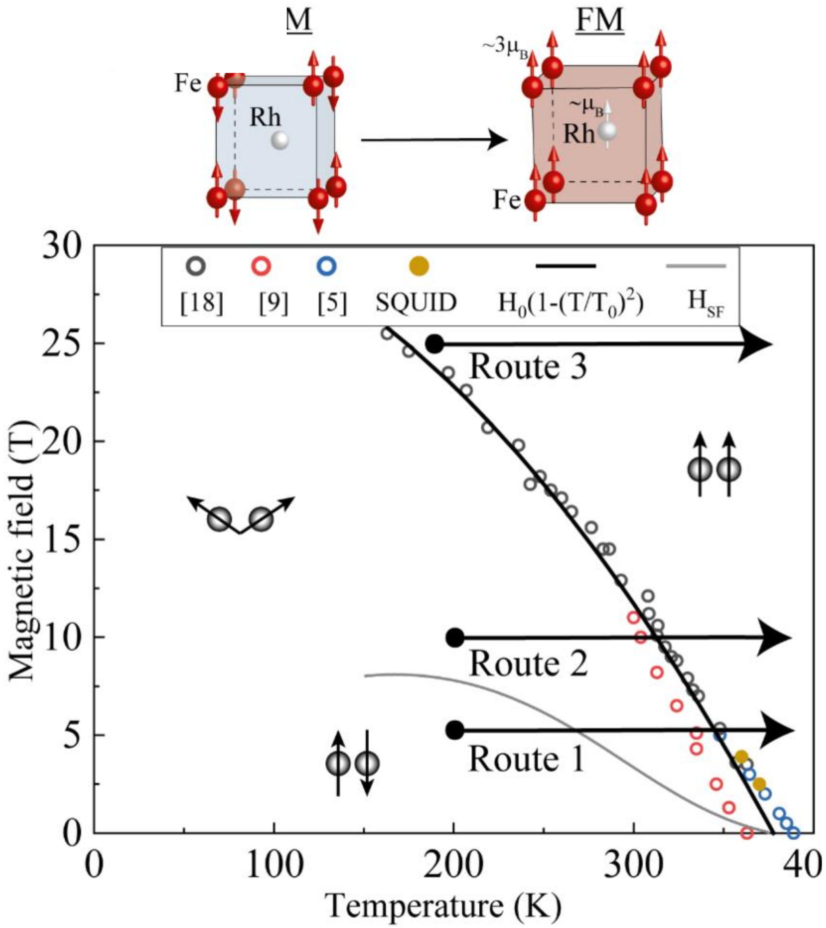


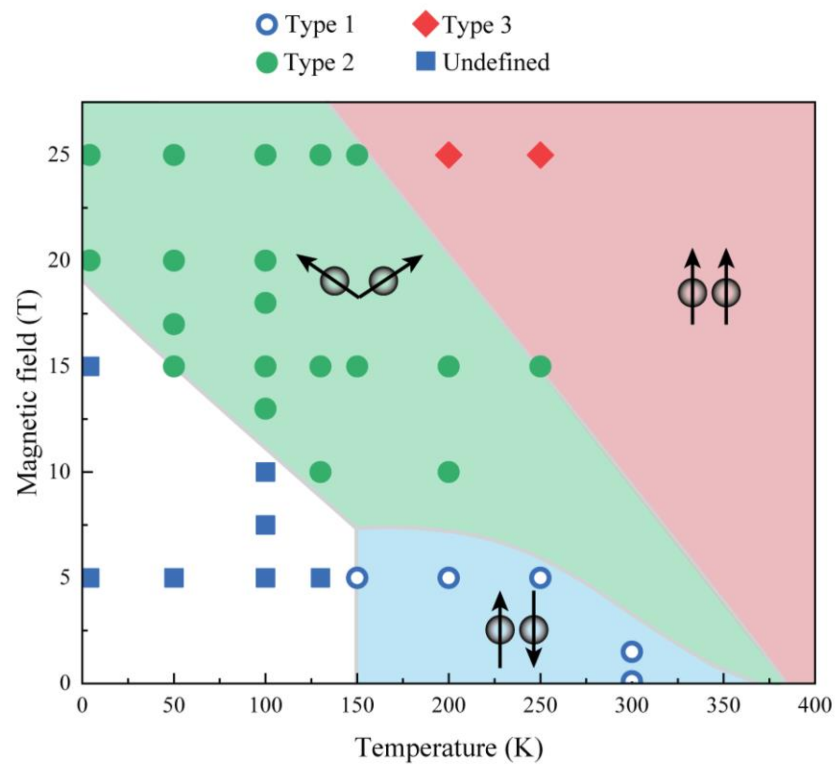

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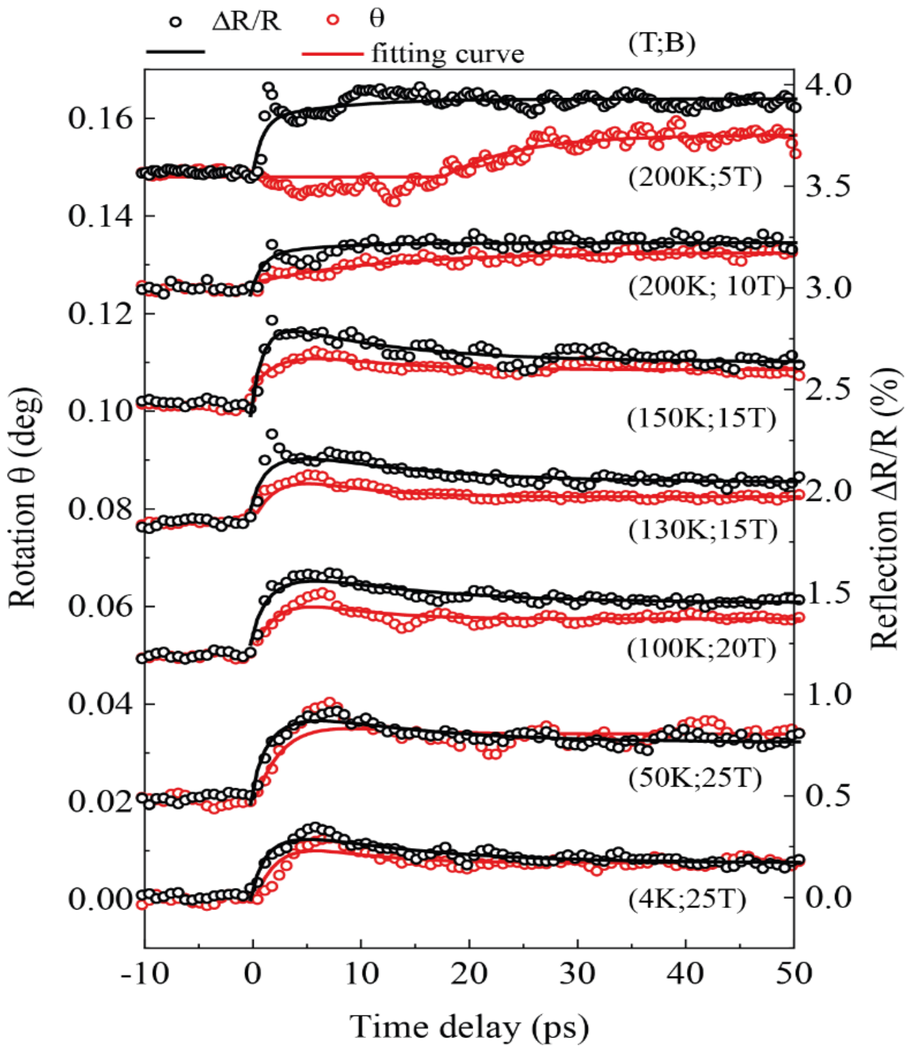


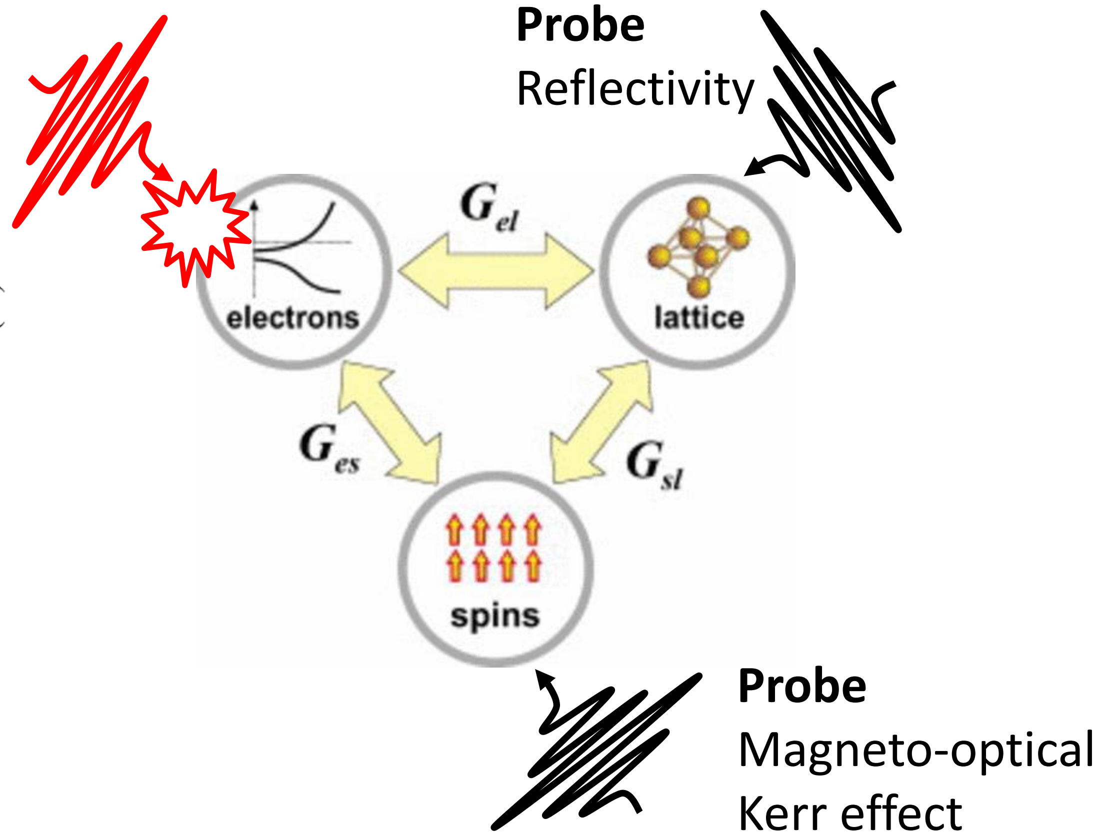


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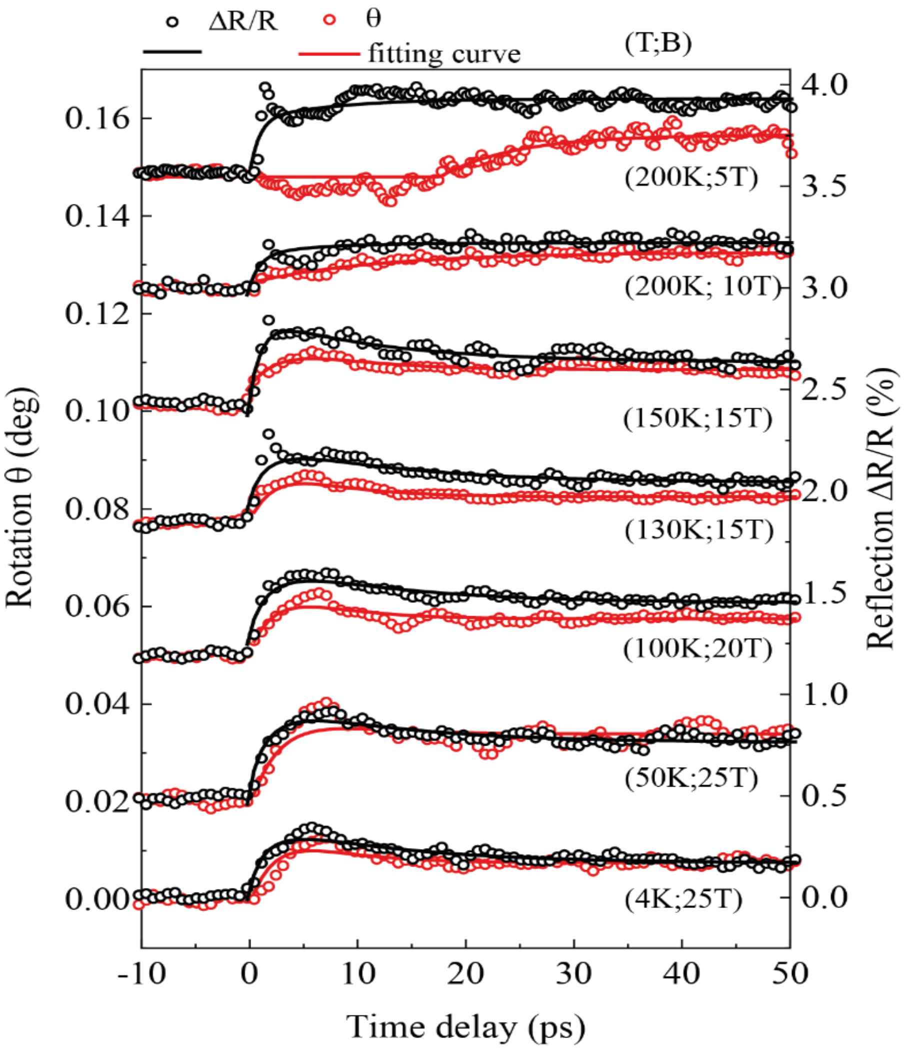
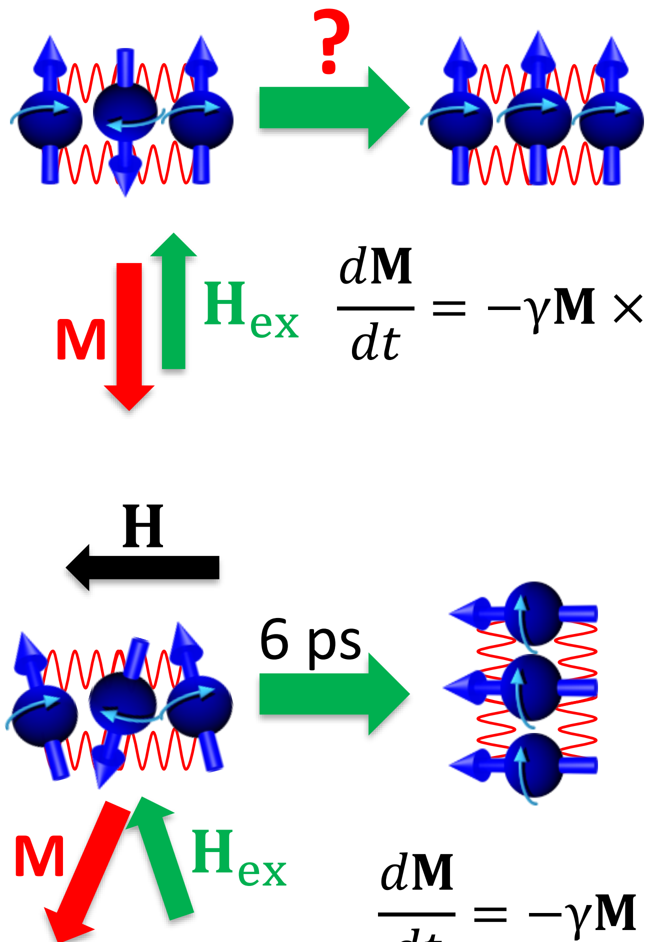
https://arxiv.org/abs/2202.03931

# Ultrafast magnetism of FeRh in high magnetic fields




$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{ex} = \mathbf{0}$$

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{ex} \neq \mathbf{0}$$

I. Dolgiih et al, (submitted).
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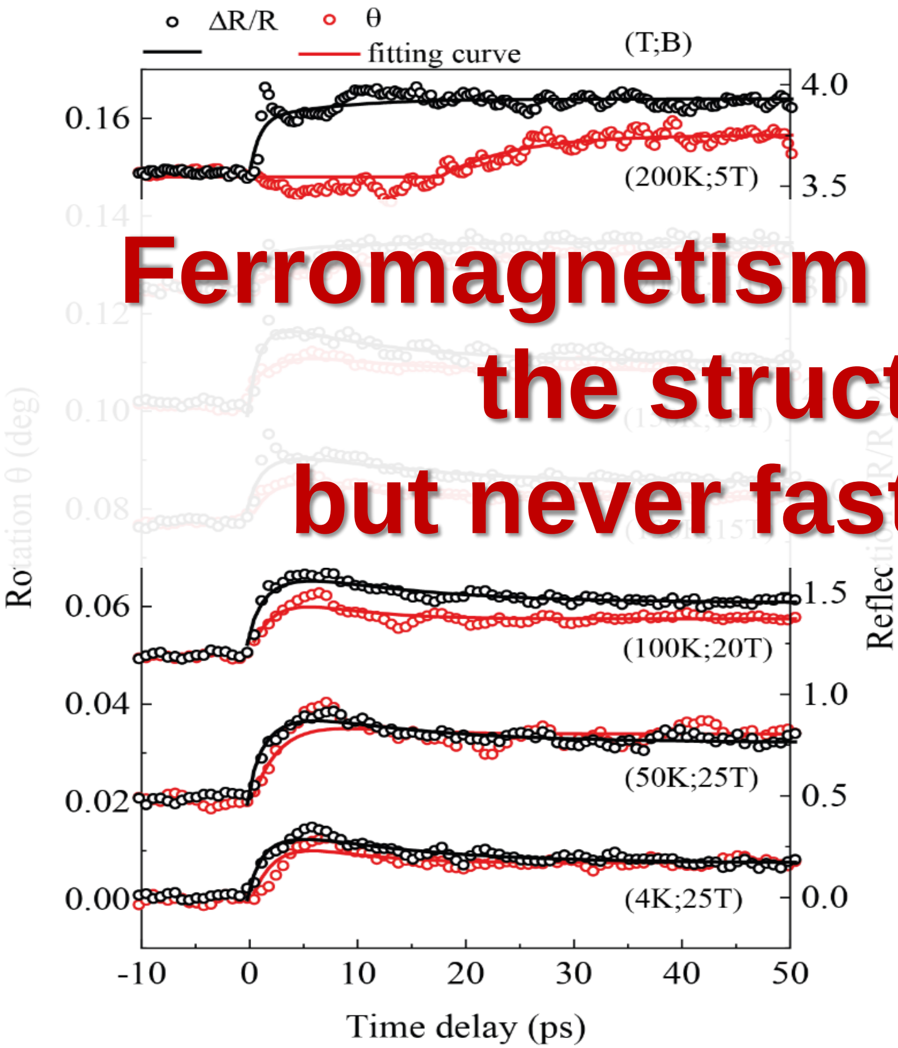


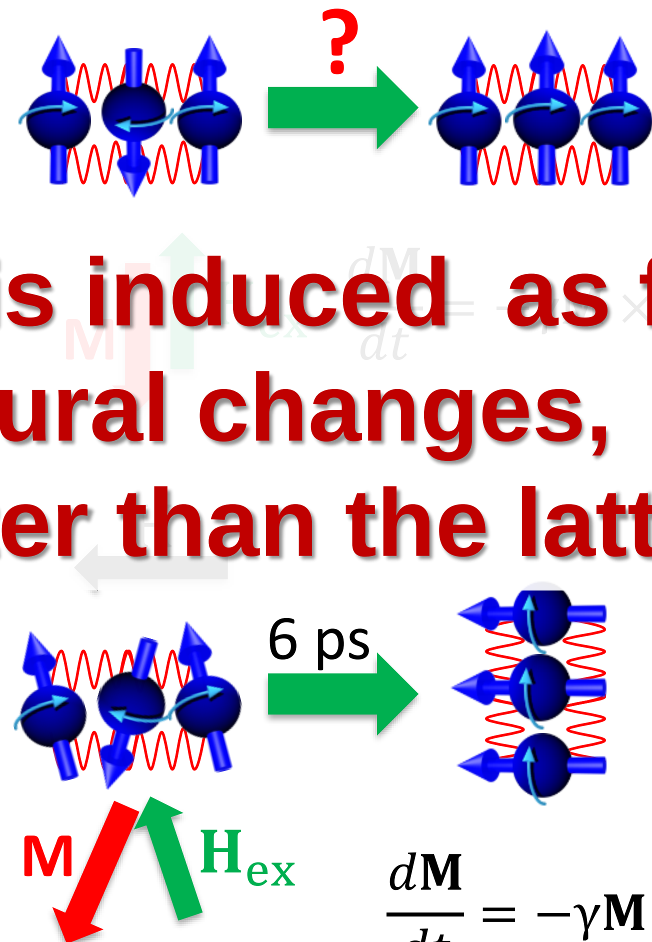


**Ferromagnetism is induced as fast as the structural changes, but never faster than the latter.**

$$\frac{d\mathbf{M}}{dt} = -\gamma \mathbf{M} \times \mathbf{H}_{ex} \neq \mathbf{0}$$

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# Conclusions

- Femtosecond laser pulses trigger magneto-structural phase transition from antiferromagnetic to ferromagnetic phase in FeRh
- Excitation of FeRh with femtosecond laser pulses generates nuclei's of ferromagnetic phase. The ferromagnetic order in the nuclei's is established only in 10-20 ps after the excitation.
- Structural changes estimated from reflectivity also occur on a time-scale of 10 ps (characteristic time of 3 ps).
- At 5 T the characteristic time of the phase transition reaches the limit of 3 ps (i.e. the characteristic time of the structural changes). Further increase of the field does not accelerate the phase transition any further.

## Chicken and egg evolve together!

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