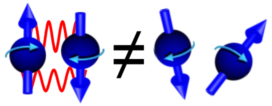
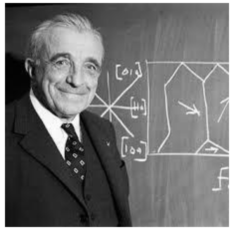
THz Magnetism of Antiferromagnets

Alexey V. Kimel

Ultrafast Spectroscopy of Correlated Materials,
Radboud University, Nijmegen, The Netherlands

Magnetism – *the strongest quantum mechanical phenomenon*





L. Néel:
“Antiferromagnets – interesting, but useless.”

$S_z = \pm\hbar/2$

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

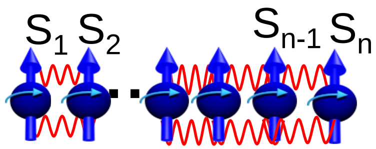
L. Néel

Spin
(1922)

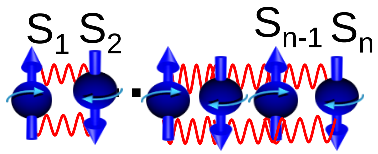
Exchange interaction
(1926)

Antiferromagnetism
(1930)

Ferromagnet (J>0)



Antiferromagnet (J<0)



Macrospin approximation

$$\mathbf{M} = -\gamma\frac{\sum S_i}{V} \neq 0 \qquad \mathbf{MH} \neq 0$$

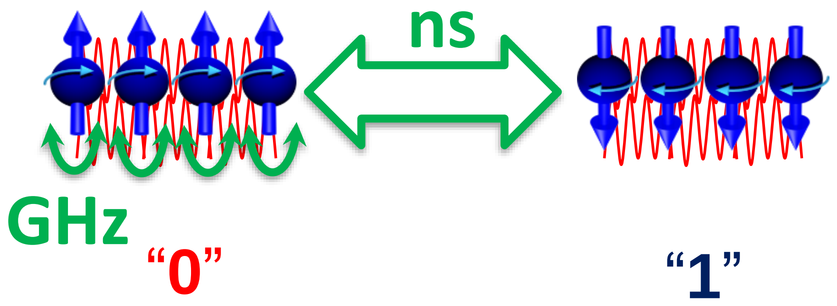
$$\mathbf{M} = 0$$

$$\mathbf{L} = -\gamma\frac{\sum(\mathbf{S}_{2i-1}-\mathbf{S}_{2i})}{V} \qquad \mathbf{LH} = 0$$

State-of-the-art

Ferromagnets

(well understood and used in data storage)

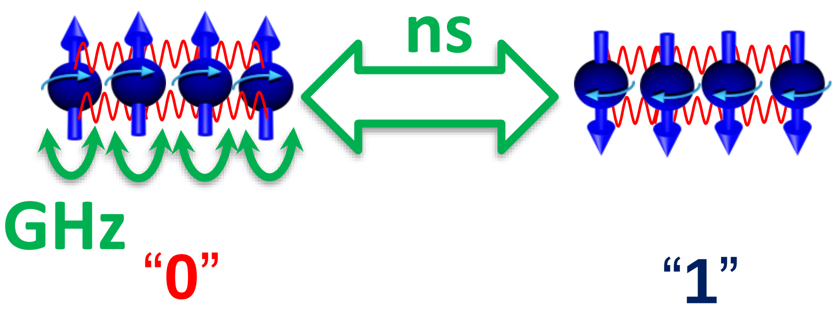


Non-zero magnetization M≠0
Controlled by magnetic fields

State-of-the-art

Ferromagnets

(well understood and used in data storage)



ns

GHz

"0"

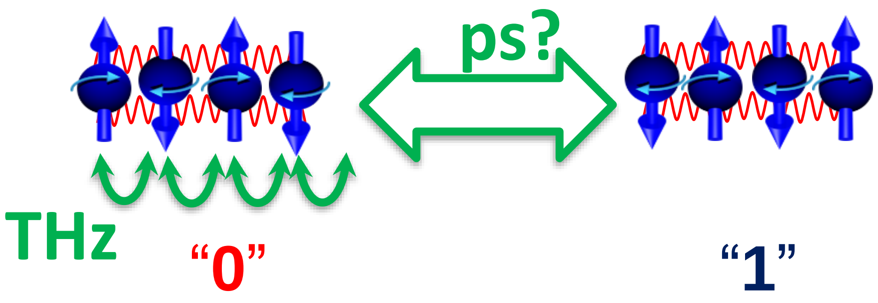
"1"

Non-zero magnetization M≠0

Controlled by magnetic fields

Antiferromagnets

(far less explored, 1000 times faster)



ps?

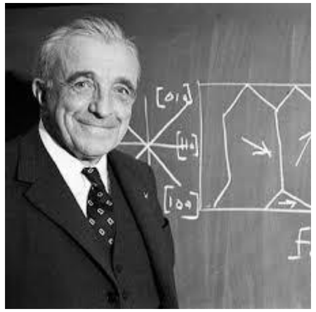
THz

"0"

"1"

No net magnetization M=0

Insensitive to magnetic fields



L. Néel:
"Antiferromagnets – interesting, but useless."

State-of-the-art

- How to detect?
- How to control?
- How to control ultrafast?

Antiferromagnets

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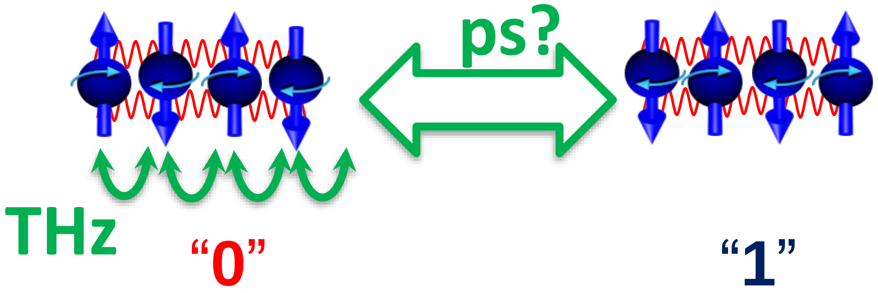
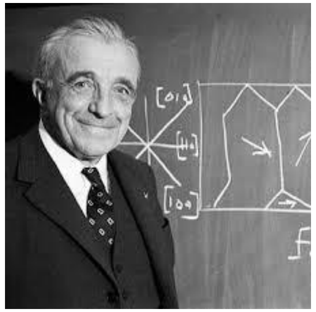


No net magnetization M=0
Insensitive to magnetic fields



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Symmetry in physics of antiferromagnets in thermodynamic equilibrium

I. A. Dzyaloshinskii

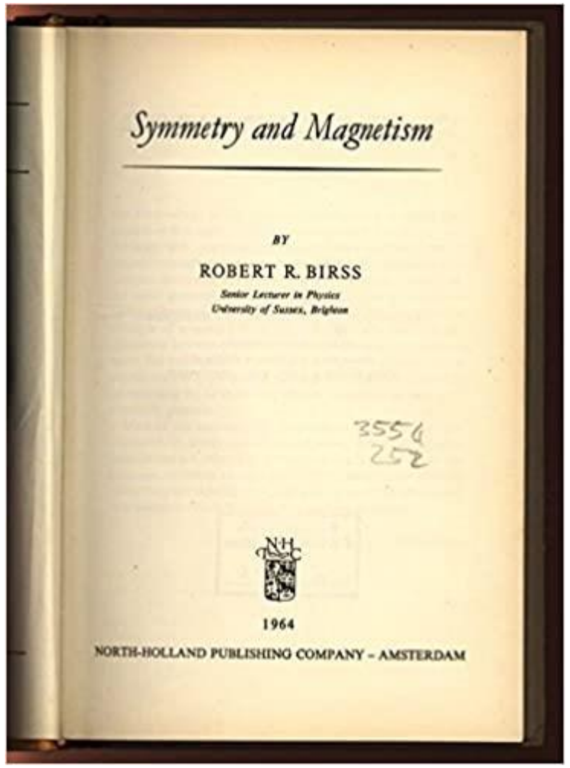
A. S. Borovik-Romanov

E. A. Turov

Е.А. ТУРОВ, А.В. КОЛЧАНОВ, В.В. МЕНЬШЕНИН,
И.Ф. МИРСАЕВ, В.В. НИКОЛАЕВ

СИММЕТРИЯ
И ФИЗИЧЕСКИЕ СВОЙСТВА
АНТИФЕРРОМАГНЕТИКОВ

МОСКВА
ФИЗМАТЛИТ
2001



Symmetry and Magnetism

BY

ROBERT R. BIRSS

Senior Lecturer in Physics
University of Sussex, Brighton

N.H.
P.C.

1964

NORTH-HOLLAND PUBLISHING COMPANY – AMSTERDAM

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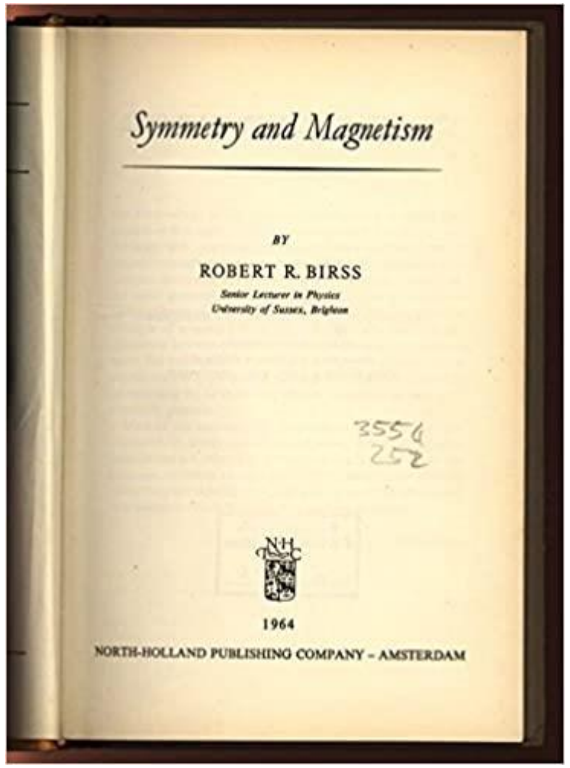

Symmetry and Magnetism

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Senior Lecturer in Physics
University of Sussex, Brighton

3556
252

1964

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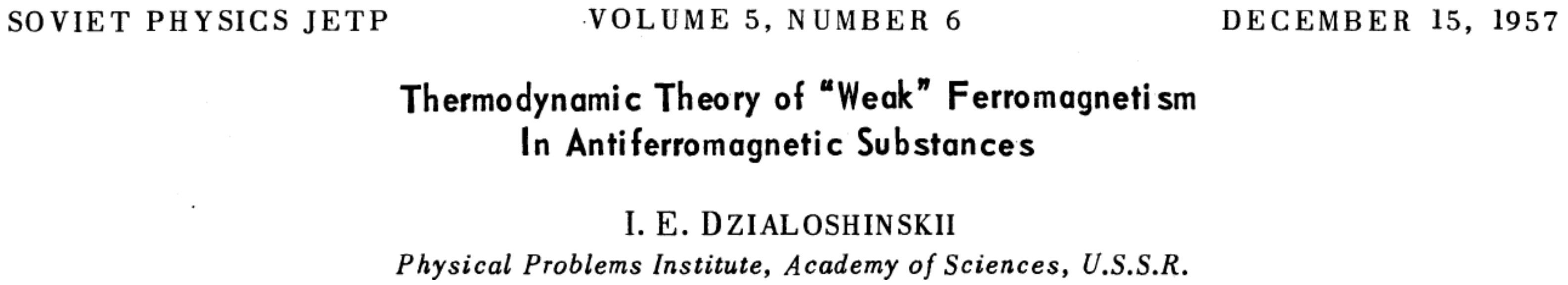
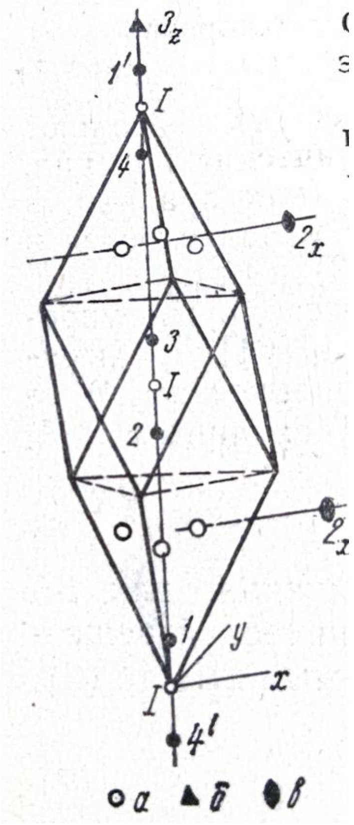
altermagnet!

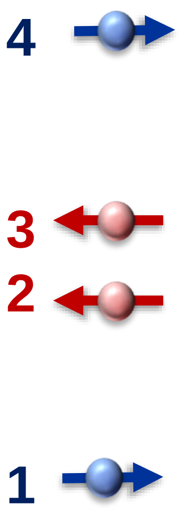


SOVIET PHYSICS JETP VOLUME 5, NUMBER 6 DECEMBER 15, 1957

Thermodynamic Theory of "Weak" Ferromagnetism In Antiferromagnetic Substances

I. E. DZIALOSHINSKII
Physical Problems Institute, Academy of Sciences, U.S.S.R.





$$\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2 + \mathbf{M}_3 + \mathbf{M}_4$$

$$\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2 - \mathbf{M}_3 + \mathbf{M}_4$$

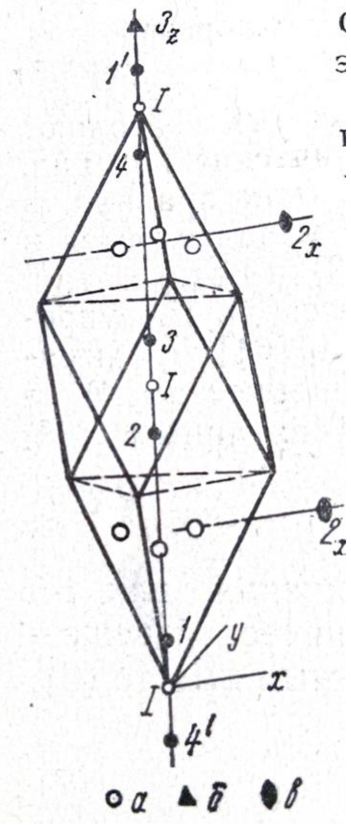
$$M_x = DL_y$$
$$M_y = -DL_x$$
$$M_z = 0$$

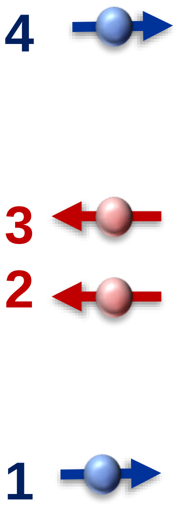
D shows the strength of the Dzyaloshinskii-Moriya interaction!

$$2_x M_x = M_x, 2_x L_x = -L_x$$
$$2_x M_y = -M_y, 2_x L_y = L_y$$

L_x behaves as M_y
L_y behaves as M_x

Antiferromagnetic Hall effect in hematite α-Fe_2O_3





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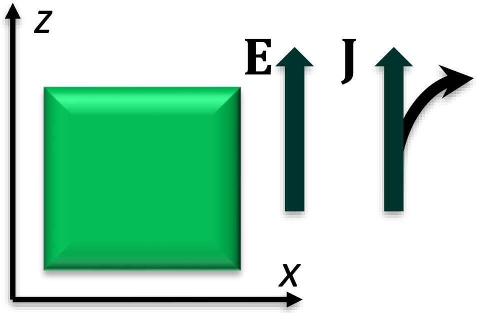
$$\mathbf{L} = \mathbf{M_1} - \mathbf{M_2} - \mathbf{M_3} + \mathbf{M_4}$$

$$M_x = DL_y$$
$$M_y = -DL_x$$
$$M_z = 0$$

$$\boxed{\sigma_{xz}^{(a)} = \chi^{(H)} H_y + \chi^{(M)} M_y - \chi^{(L)} L_x}$$



$$J_z = \sigma_{zz} E_z$$

$$J_x = \sigma_{xz} E_z$$

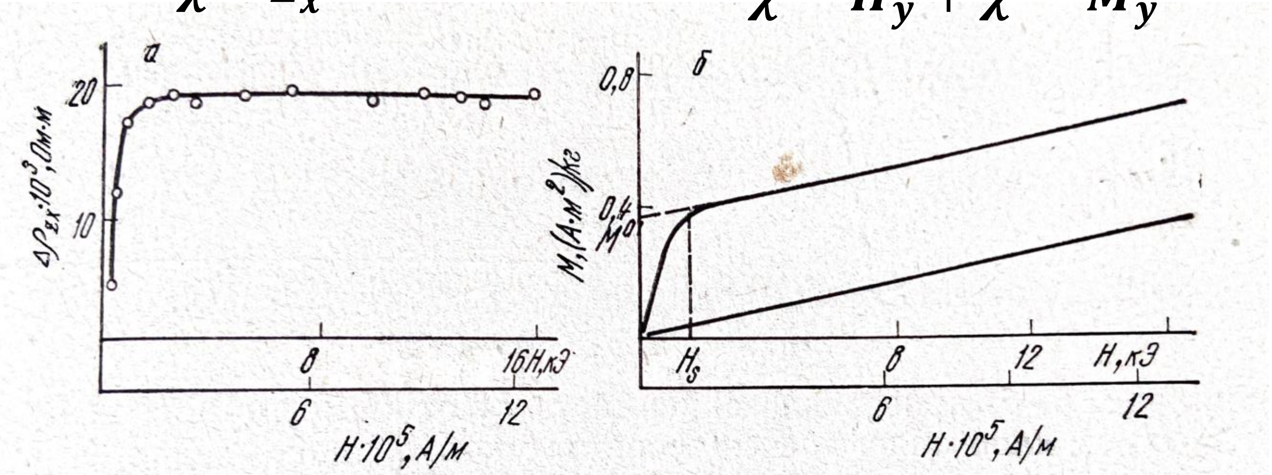
$$\sigma_{xz} = -\sigma_{zx} = \sigma_{xz}^{(a)}$$

Measured

$\boldsymbol{\chi^{(L)} L_x}$

Expected

$\boldsymbol{\chi^{(H)} H_y + \chi^{(M)} M_y}$

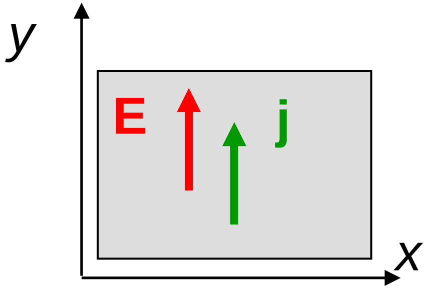


K. B. Vlasov et al., *Sov. Phys. Solid State* **22**, 967 (1980).
K. B. Vlasov et al., *Физика металлов и металловедение* **42**,513-517 (1976).

Hall *vs* Faraday effect

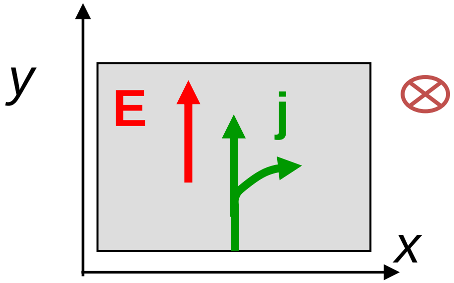
$$j_k = \sigma_{kl} E_l$$

j_k – current density
E_l – electric field
σ_{kl} – conductivity



H=0
M=0

$$\sigma_{kl} = \begin{bmatrix} \sigma_{xx} & 0 \\ 0 & \sigma_{yy} \end{bmatrix}$$



⊗ **H≠0**
M ≠ 0

$$\sigma_{kl} = \begin{bmatrix} \sigma_{xx} & \sigma_{xy} \\ \sigma_{yx} & \sigma_{yy} \end{bmatrix}$$

$$\sigma_{kl}^{(a)} = -\sigma_{lk}^{(a)} \neq 0$$

$$\sigma_{kl}^{(a)} = \underbrace{\chi_{klm}^{(H)} H_m}_{\text{dia- and para-magnetic (normal)}} + \underbrace{\chi_{klm}^{(M)} M_m}_{\text{ferro-magnetic (anomalous)}} + \underbrace{\chi_{klm}^{(L)} L_m}_{\text{antiferro-magnetic (alter)}}$$

Contributions to the Hall effect

Hall *vs* Faraday effect

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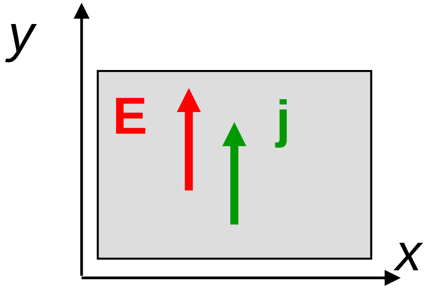

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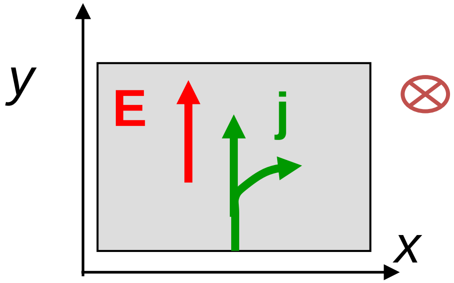

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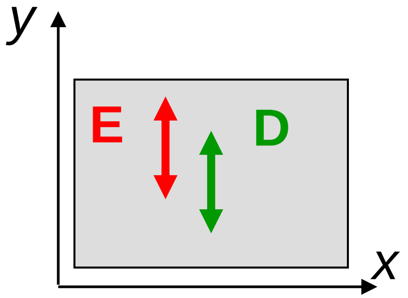
Contributions to the Hall effect

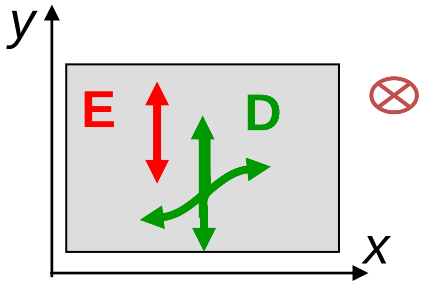
$$D_k = \varepsilon_{kl} E_l$$

D_k –electric displacement
E_l – electric field
ε_{kl} – permittivity



H=0
M=0

$$\varepsilon_{kl} = \begin{bmatrix} \varepsilon_{xx} & 0 \\ 0 & \varepsilon_{yy} \end{bmatrix}$$



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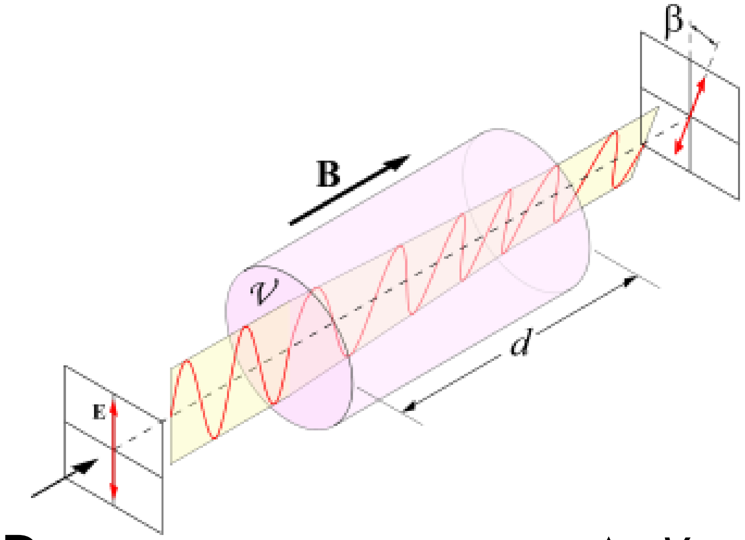
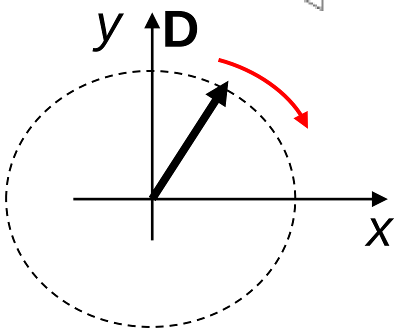
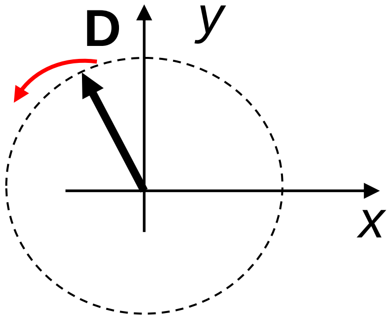
Contributions to the Faraday effect

The magneto-optical Faraday effect





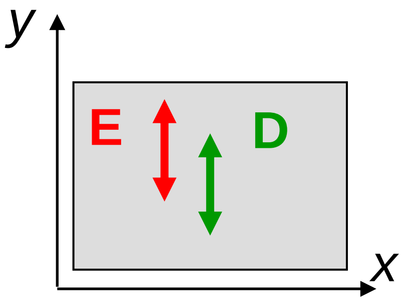
$$n_+ = \sqrt{\varepsilon_{xx} + \varepsilon_{xy}}$$



$$n_- = \sqrt{\varepsilon_{xx} - \varepsilon_{xy}}$$

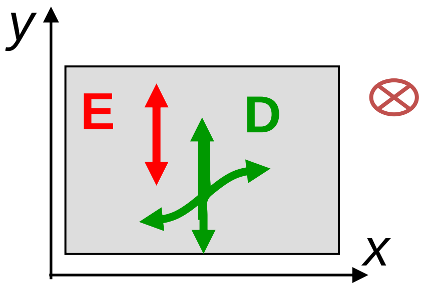
$$D_k = \varepsilon_{kl}E_l$$

D_k – electric displacement
E_l – electric field
ε_{kl} – permittivity



H=0
M=0

$$\varepsilon_{kl} = \begin{bmatrix} \varepsilon_{xx} & 0 \\ 0 & \varepsilon_{yy} \end{bmatrix}$$



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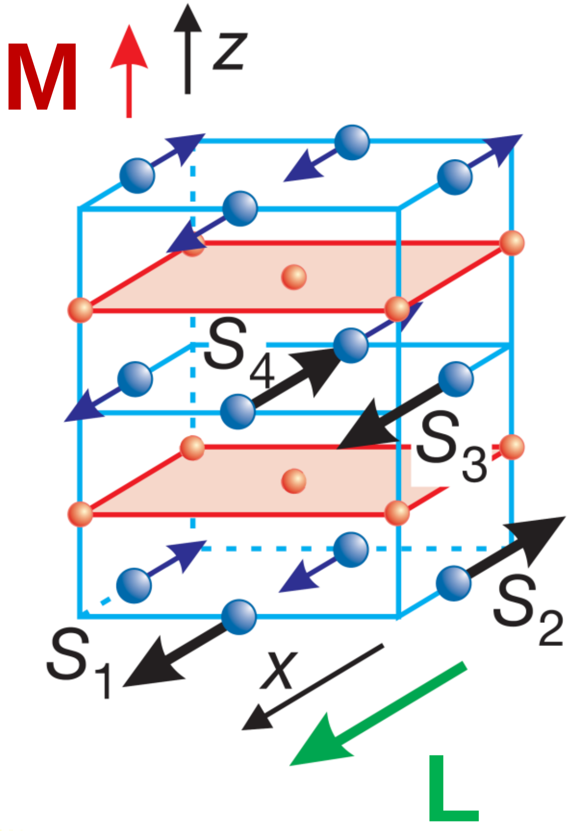
Contributions to the Faraday effect

Antiferromagnetic and ferromagnetic Faraday effect in yttrium orthoferrite $YFeO_3$

B. B. Krichevtsov, K. M. Mukimov, R. V. Pisarev, and M. M. Ruvinshteĭn

(Submitted 25 August 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **34**, No. 7, 399–402 (5 October 1981)



Dzyaloshinskii-Moriya interaction and weak-ferromagnetism

$$M_z = D L_x$$

$$\varepsilon_{xy}^{(a)} = -\varepsilon_{yx}^{(a)} = \underbrace{\chi^{(H)} H_z}_{\text{dia- and para-magnetic}} + \underbrace{\chi^{(M)} M_z}_{\text{ferro-magnetic}} - \underbrace{\chi^{(L)} L_x}_{\text{antiferro-magnetic}}$$

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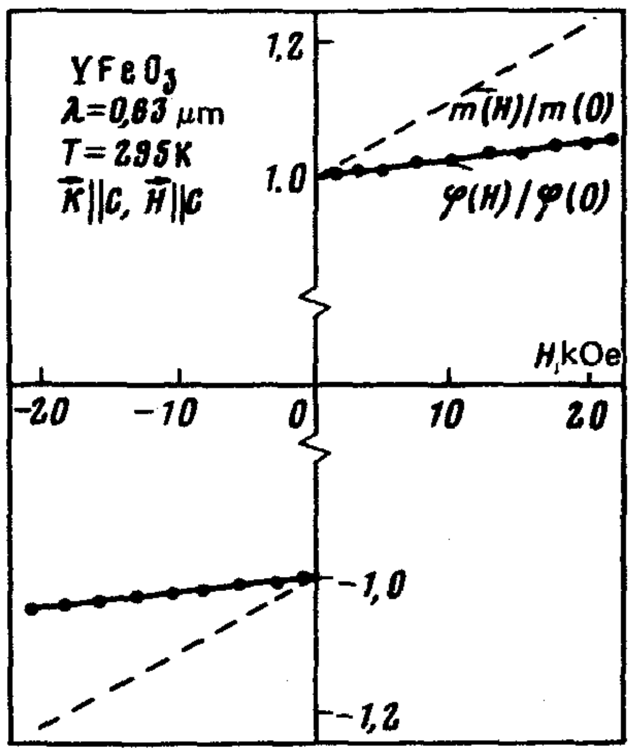

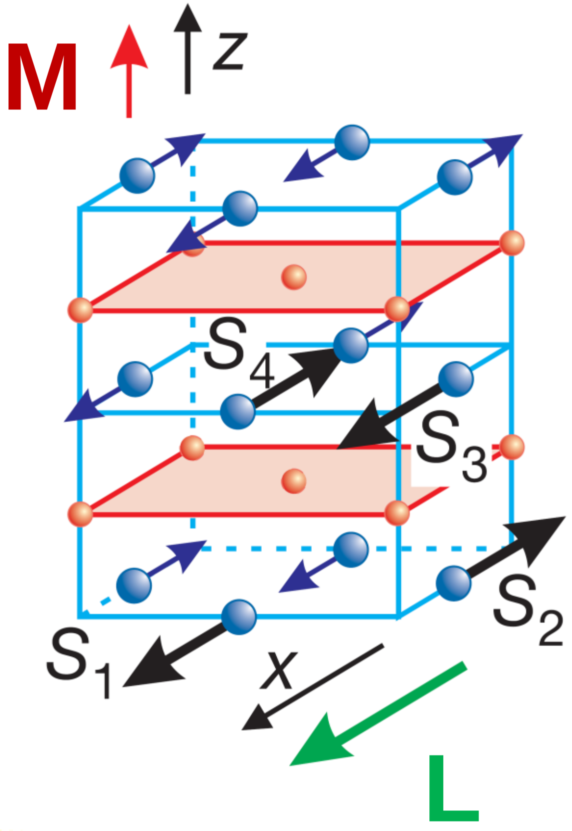

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$$M_z = DL_x$$

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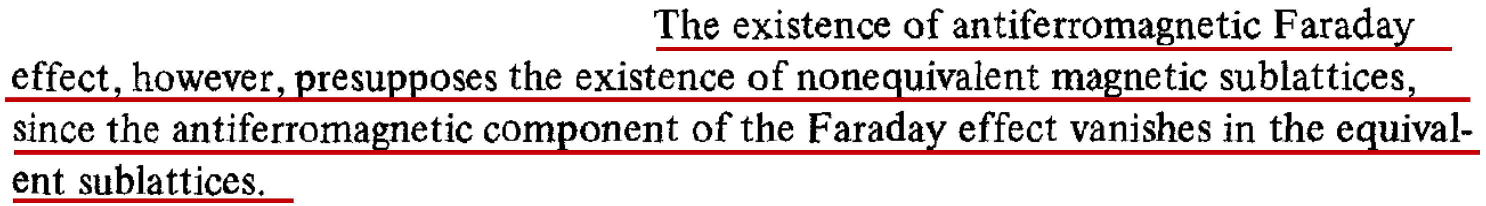
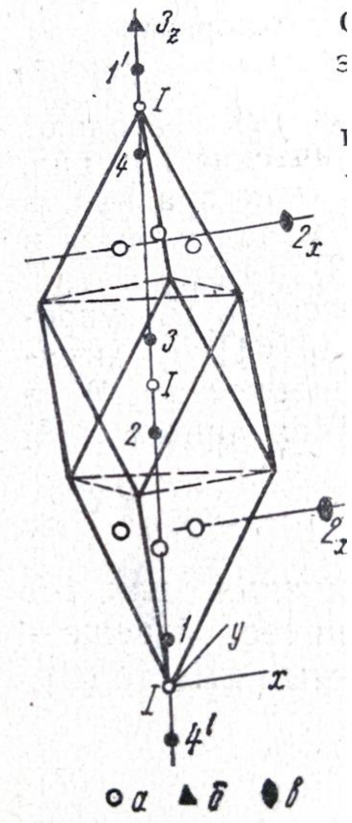
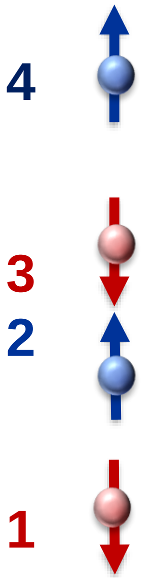


The existence of antiferromagnetic Faraday effect, however, presupposes the existence of nonequivalent magnetic sublattices, since the antiferromagnetic component of the Faraday effect vanishes in the equivalent sublattices.

Faraday effect in antiferromagnetic Cr_2O_3





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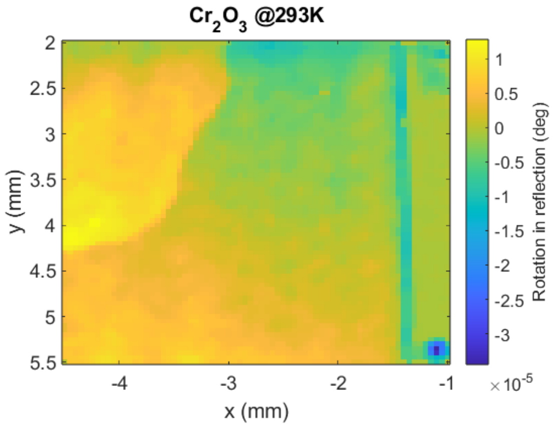
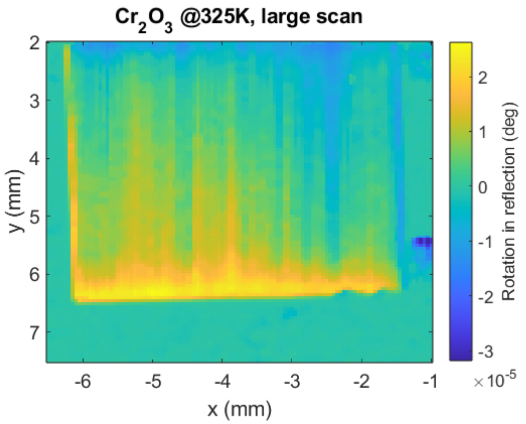
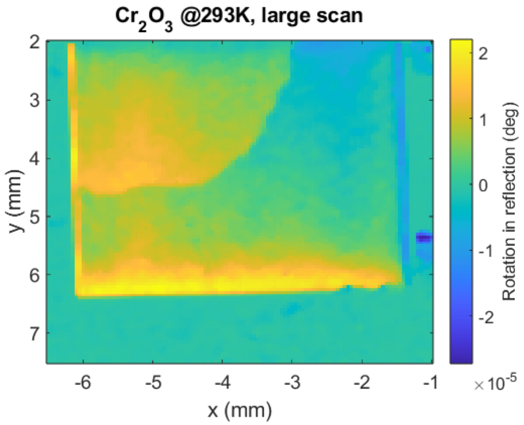
$$\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2 + \mathbf{M}_3 - \mathbf{M}_4$$

$$\boxed{\varepsilon_{xy}^{(a)} = \chi^{(H)} H_z + \chi_{xyzz}^{LE} E_z L_z}$$

Measurements are done by N. Khokhlov (Radboud University)







See also:

B B Krichevtsov et al, *Spontaneous non-reciprocal reflection of light from antiferromagnetic Cr2O3*, J. Phys.: Condens. Matter 5, 8233 (1993).

J. Wang, Ch. Binek, *Dispersion of Electric-Field-Induced Faraday Effect in Magnetoelectric Cr2O3*, Phys. Rev. Appl. 5, 031001 (2016).

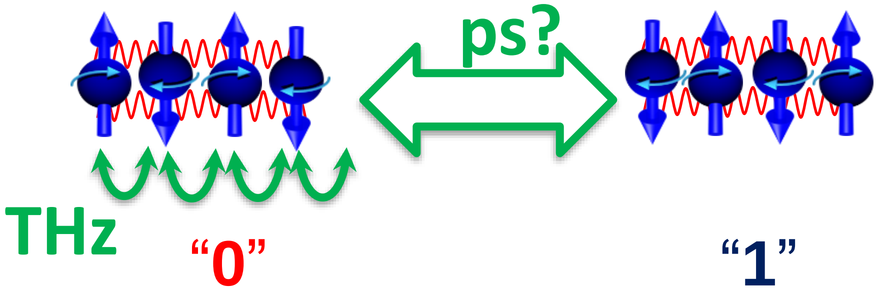
The talk of T. Jungwirth

State-of-the-art

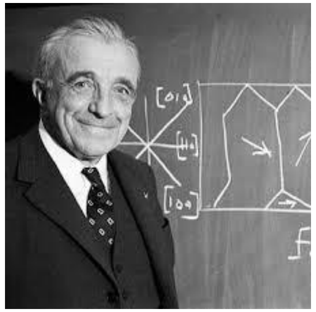
- How to detect?
- How to control?
- How to control ultrafast?

Antiferromagnets

(far less explored, 1000 times faster)



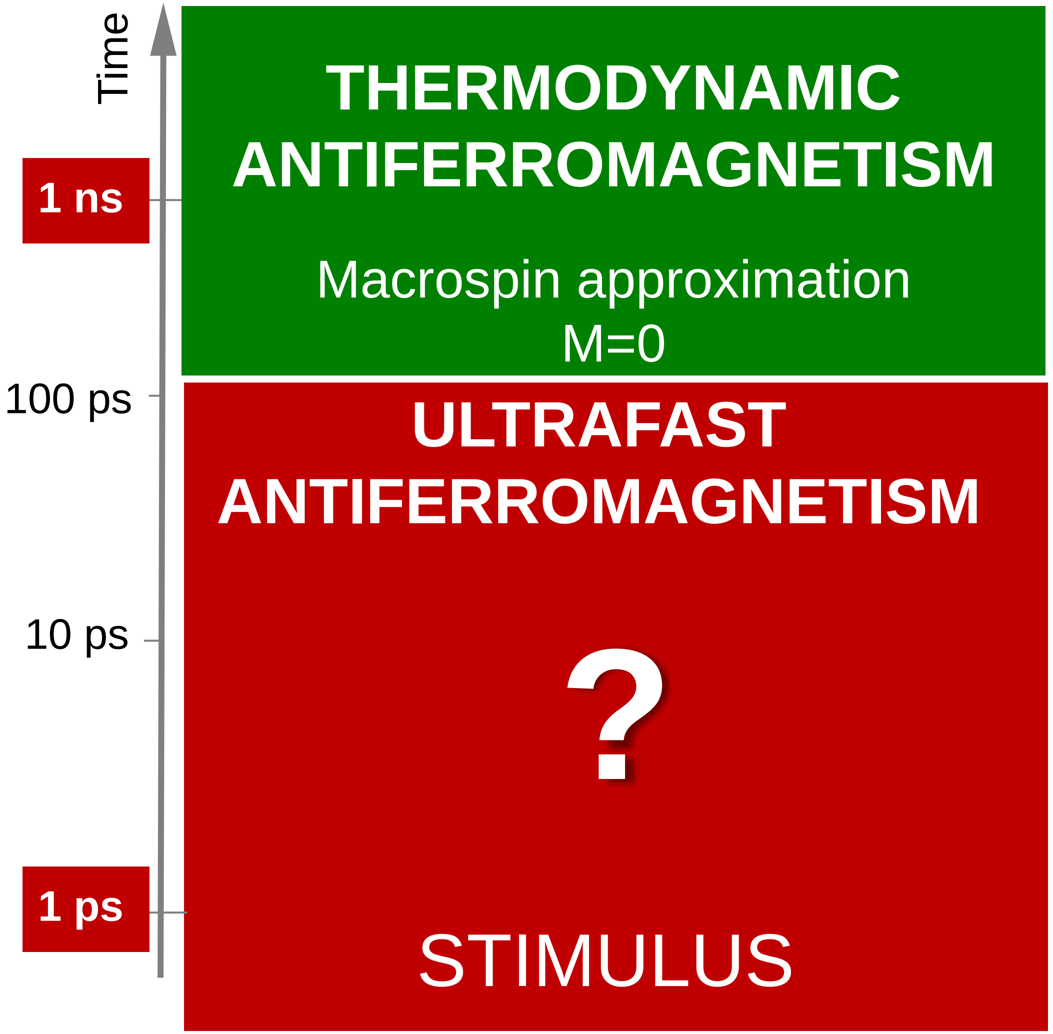
No net magnetization M=0
Insensitive to magnetic fields



L. Néel:
"Antiferromagnets – interesting, but useless."

THz antiferromagnetism

terra incognita of modern science



THERMODYNAMIC ANTIFERROMAGNETISM

Macrospin approximation
M=0

ULTRAFAST ANTIFERROMAGNETISM

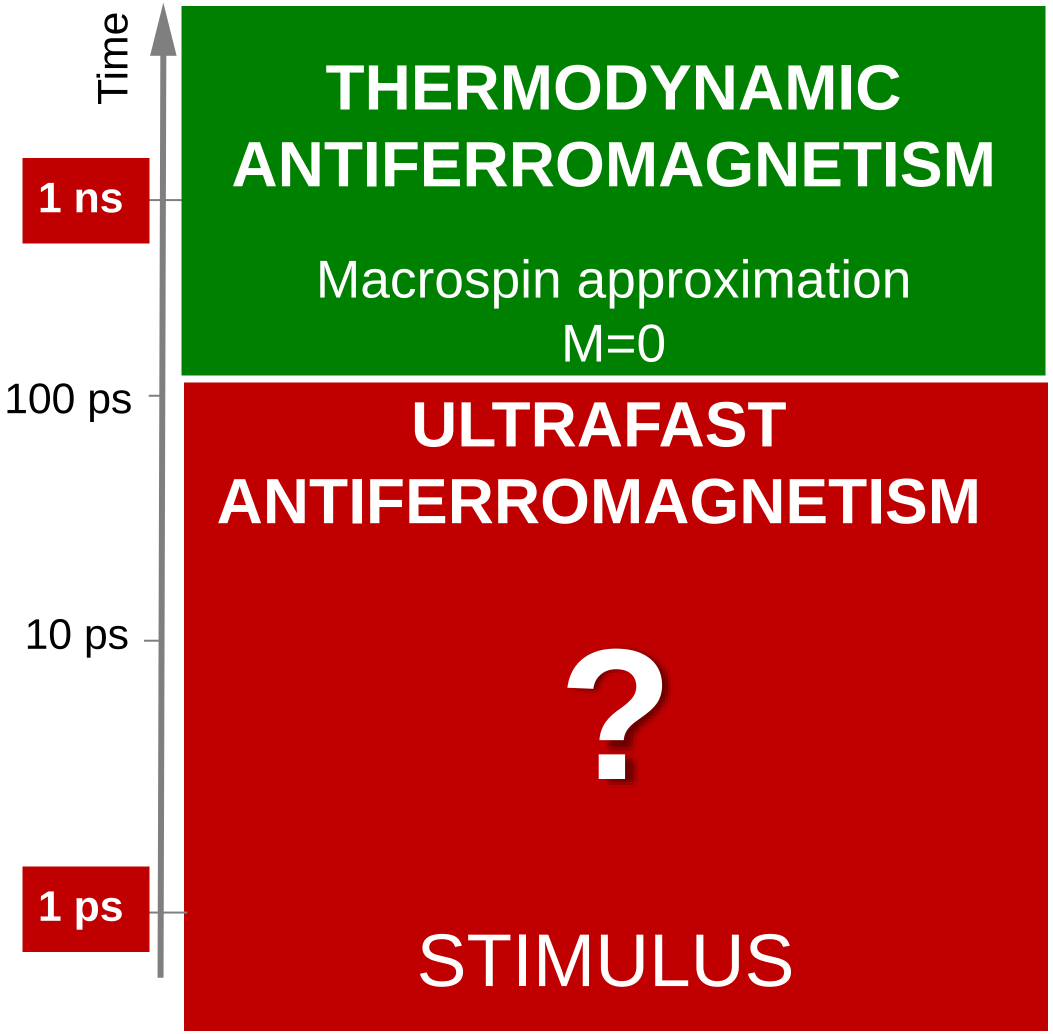
?

STIMULUS

- **M≠0**
- **Nonlinear spin dynamics** (*1+1>2*)
- **New channels of spin-lattice interaction**
- **Macrospin approximation fails**

THz antiferromagnetism

terra incognita of modern science



Time

1 ns

100 ps

10 ps

1 ps

THERMODYNAMIC ANTIFERROMAGNETISM

Macrospin approximation
M=0

ULTRAFAST ANTIFERROMAGNETISM

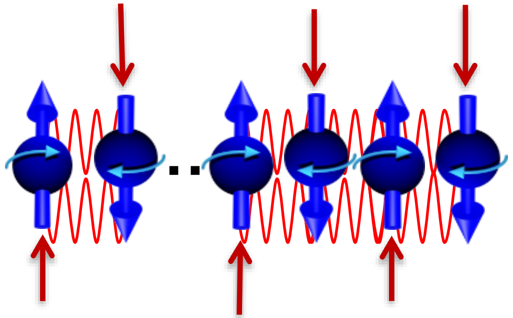
?

STIMULUS

- **M≠0**
- **Nonlinear spin dynamics** (*1+1>2*)
- **New channels of spin-lattice interaction**
- **Macrospin approximation fails**

Magnetic field as a stimulus for spins in antiferromagnets

$$\mathbf{M}_1 = \frac{\sum_i S_{2i}}{V}$$



$$\mathbf{M}_1 = \frac{\sum_i S_{2i-1}}{V}$$

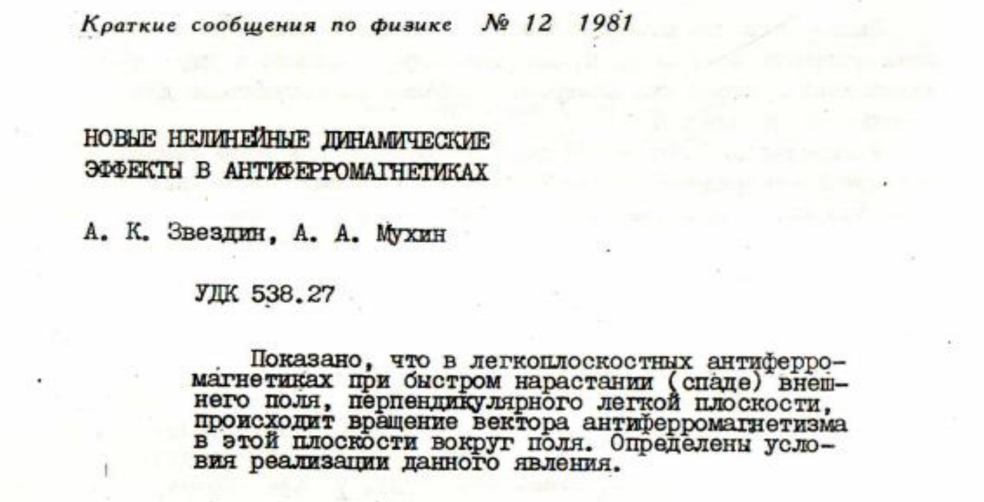
$$\mathbf{m}_1 = \frac{\mathbf{M}_1}{|\mathbf{M}_1|} \qquad \mathbf{m}_2 = \frac{\mathbf{M}_2}{|\mathbf{M}_2|}$$

$$\mathbf{l} = \frac{\mathbf{m_1} - \mathbf{m_2}}{2} \qquad \mathbf{m} = \frac{\mathbf{m_1} + \mathbf{m_2}}{2}$$

Even if $\mathbf{H}(t)$=0, $\boxed{\mathbf{m} = -\frac{1}{\gamma H_E}\frac{\partial \mathbf{l}}{\partial t} \times \mathbf{l}}$

If $\mathbf{H}(t) \neq \mathbf{0}$, $\boxed{\frac{\partial}{\partial t}\left[\frac{\partial \mathbf{l}}{\partial t} \times \mathbf{l}\right] = \gamma \frac{\partial H(t)}{\partial t} + \gamma^2 H_E [\mathbf{m} \times \mathbf{H}(\mathrm{t})]}$

- While spins in antiferromagnets are not sensitive to magnetic field $\mathbf{H}$, they can be controlled by $\frac{\partial \mathbf{H}(t)}{\partial t}$.
- Coherent spin oscillations in antiferromagnets induce **m**.



Краткие сообщения по физике № 12 1981

НОВЫЕ НЕЛИНЕЙНЫЕ ДИНАМИЧЕСКИЕ
ЭФФЕКТЫ В АНТИФЕРРОМАГНЕТИКАХ

А. К. Звездин, А. А. Мухин

УДК 538.27

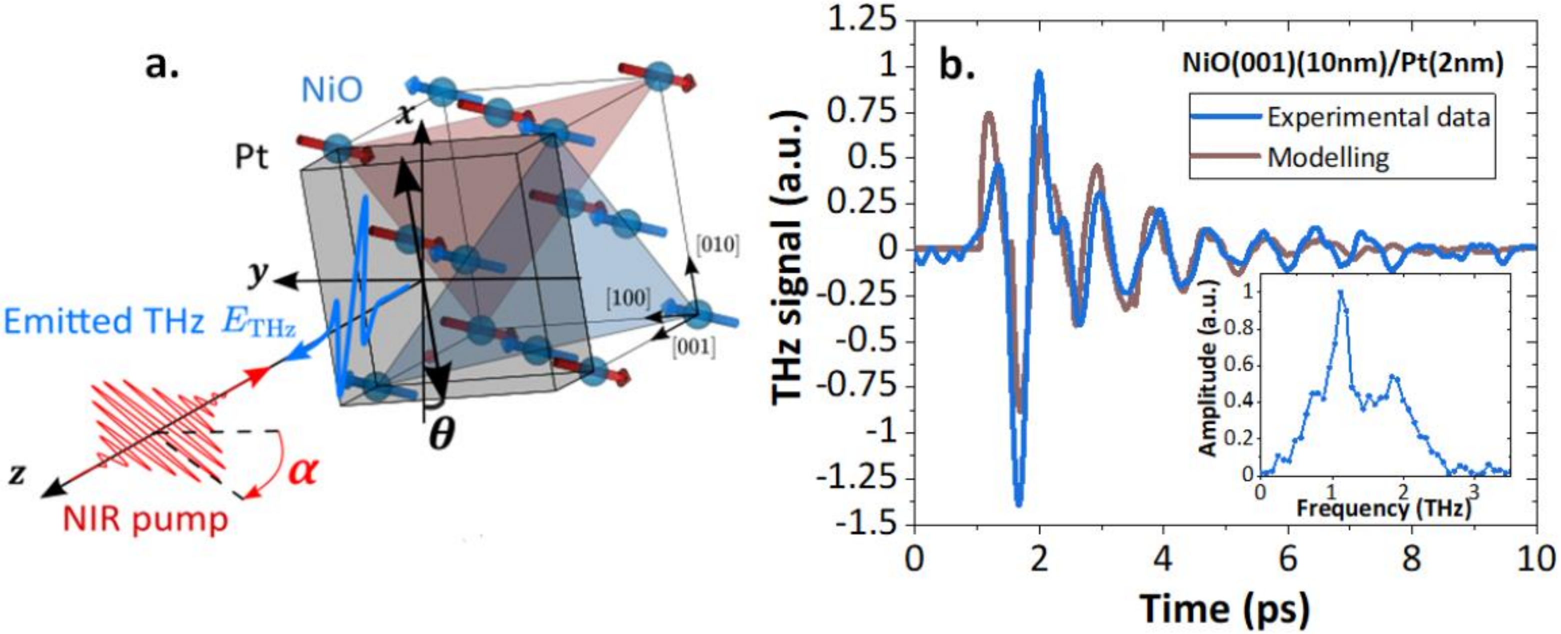
Показано, что в легкоплоскостных антиферромагнетиках при быстром нарастании (спаде) внешнего поля, перпендикулярного легкой плоскости, происходит вращение вектора антиферромагнетизма в этой плоскости вокруг поля. Определены условия реализации данного явления.

A. K. Zvezdin, *JETP* **29**, 553 (1979).
A. F. Andreev, V. I. Marchenko, *Sov. Phys. Uspekhi* **23** 21 (1980).
V.G. Baryakhtar, B. A. Ivanov, M. V. Chetkin, *Sov. Phys. Usp.* **28** 563 (1985).

T. Satoh et al, *Phys. Rev. Lett.* **105**, 077402 (2010).
T. Kampfrath et al, *Nature Photonics* **5**, 31 (2011).

A. V. Kimel et at, *Physics Reports* **852**, 1 (2020).

See the talks of E. Rongione and J. Bakker



THz antiferromagnetism

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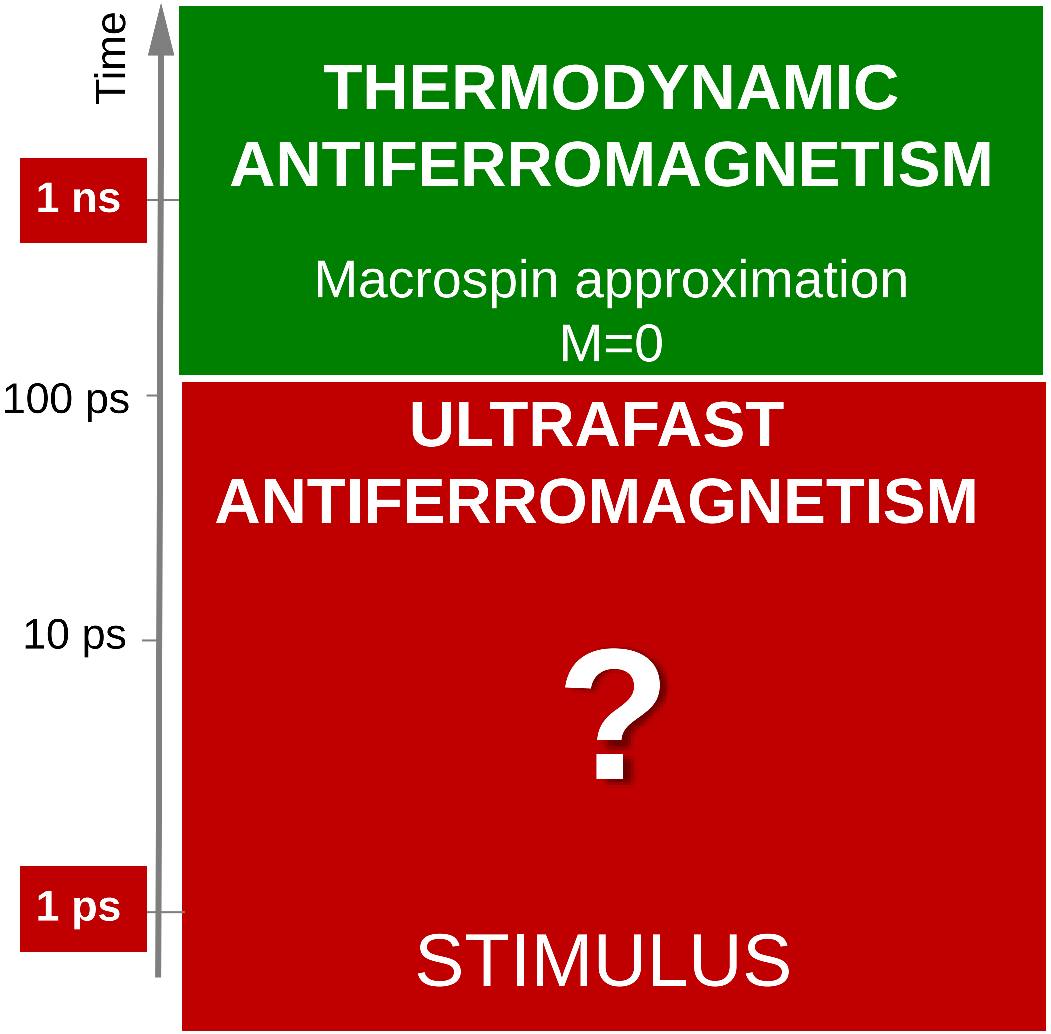


THERMODYNAMIC ANTIFERROMAGNETISM

Macrospin approximation
M=0

ULTRAFAST ANTIFERROMAGNETISM

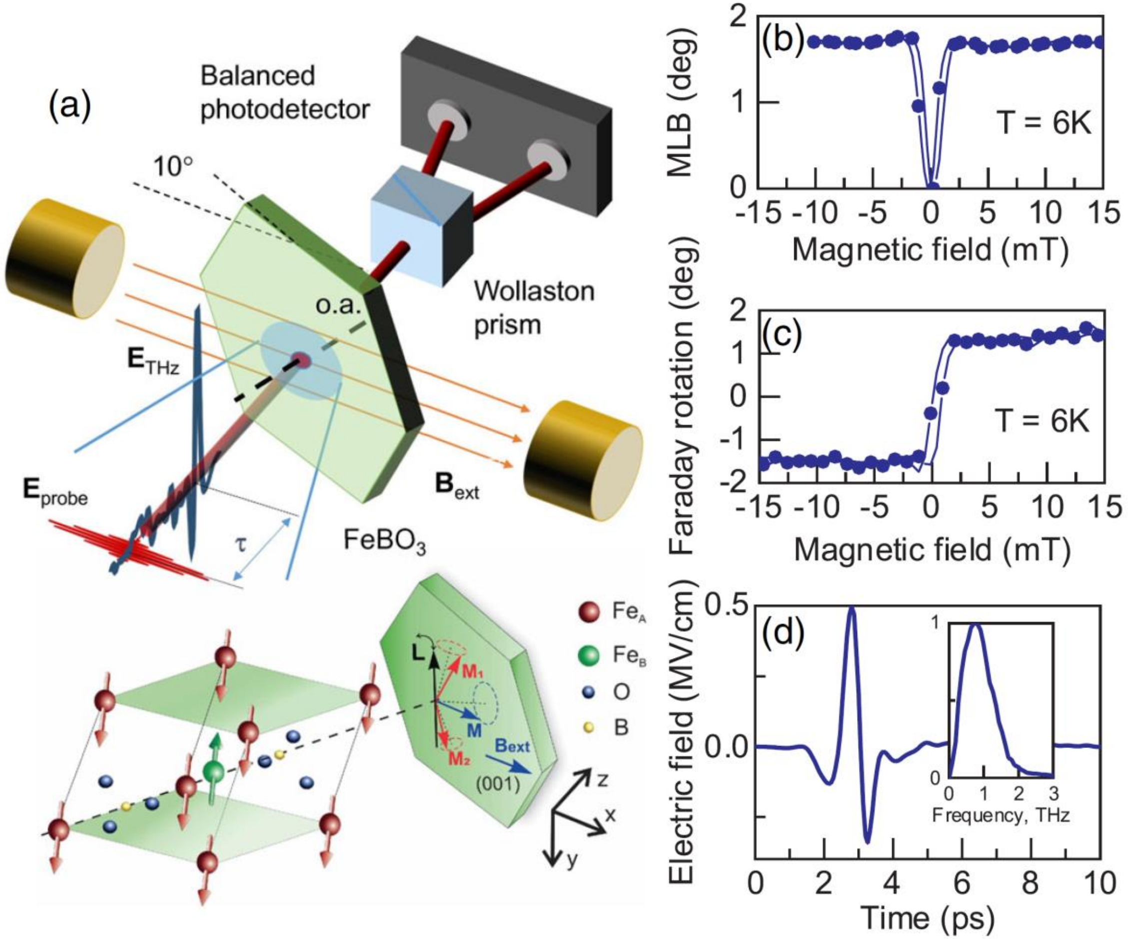
?

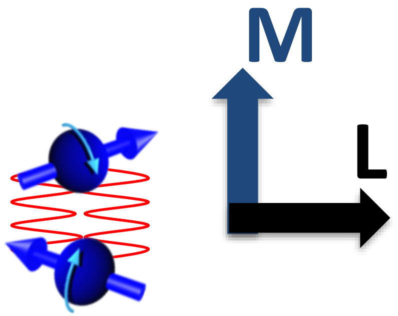
STIMULUS

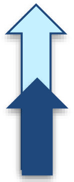
- M≠0
- **Nonlinear spin dynamics** (1+1>2)
- **New channels of spin-lattice interaction**
- **Macrospin approximation fails**

Iron borate $FeBO_3$

- *antiferromagnets with weak ferromagnetism*





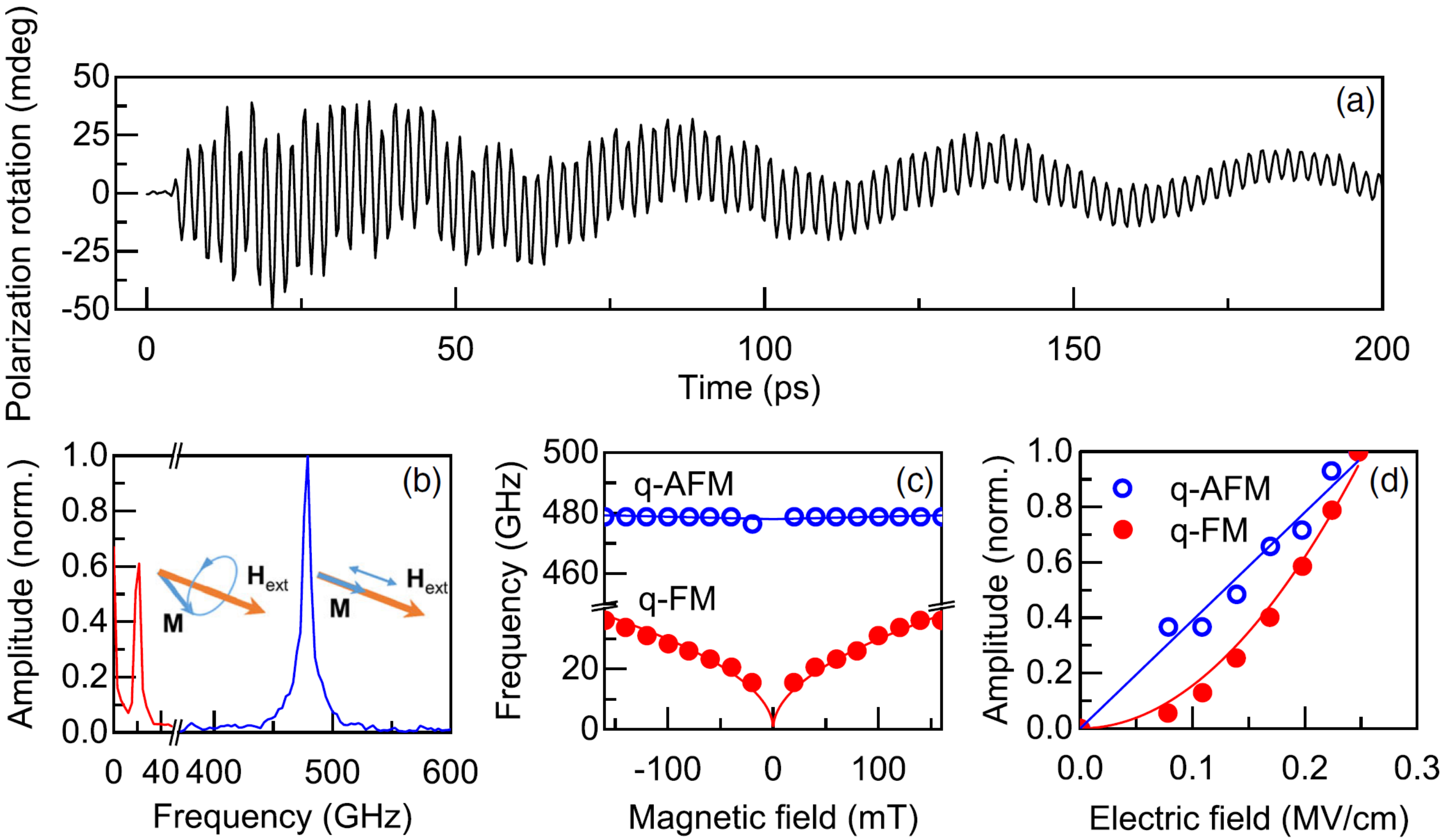


quasi-antiferromagnetic mode

quasi-ferromagnetic mode

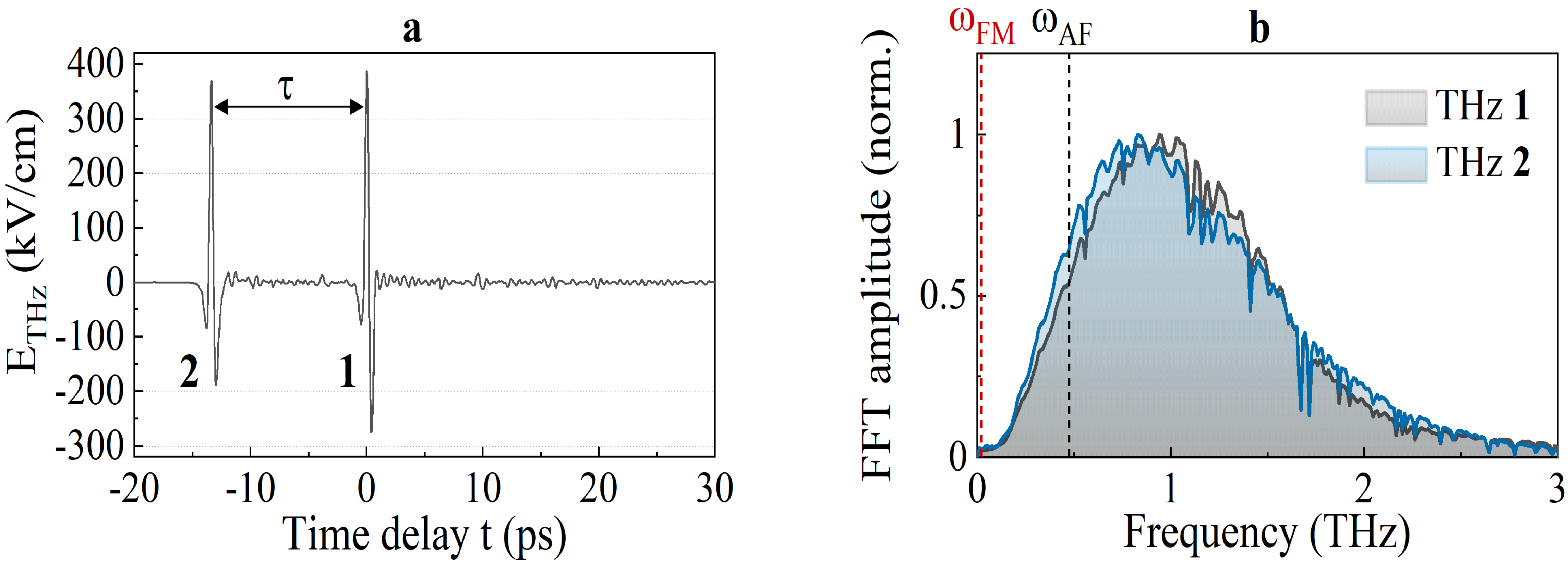
E. A. Mashkovich et al,
Phys. Rev. Lett. **123**, 157202 (2019).

THz excitation of spin resonances in $FeBO_3$

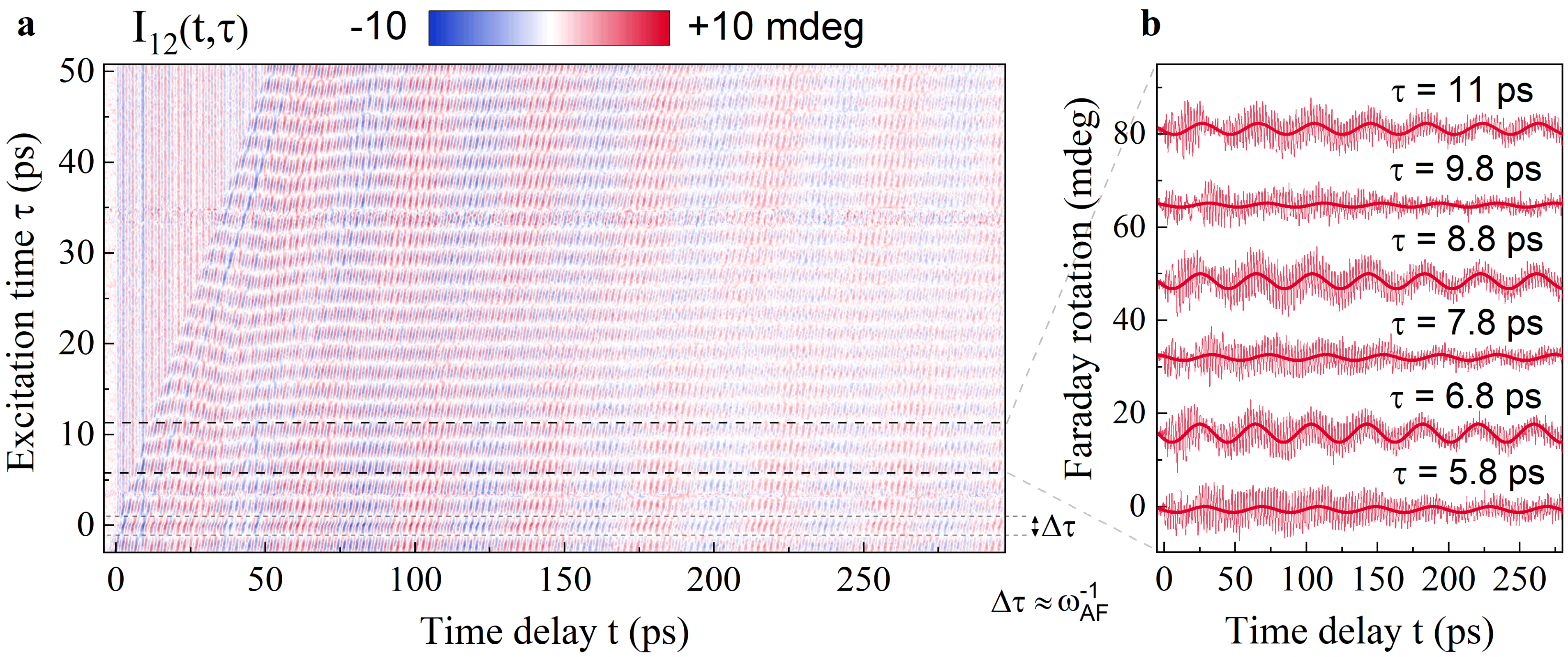


E. A. Mashkovich et al,
Phys. Rev. Lett. **123**, 157202 (2019).

Double pulse excitation of $FeBO_3$



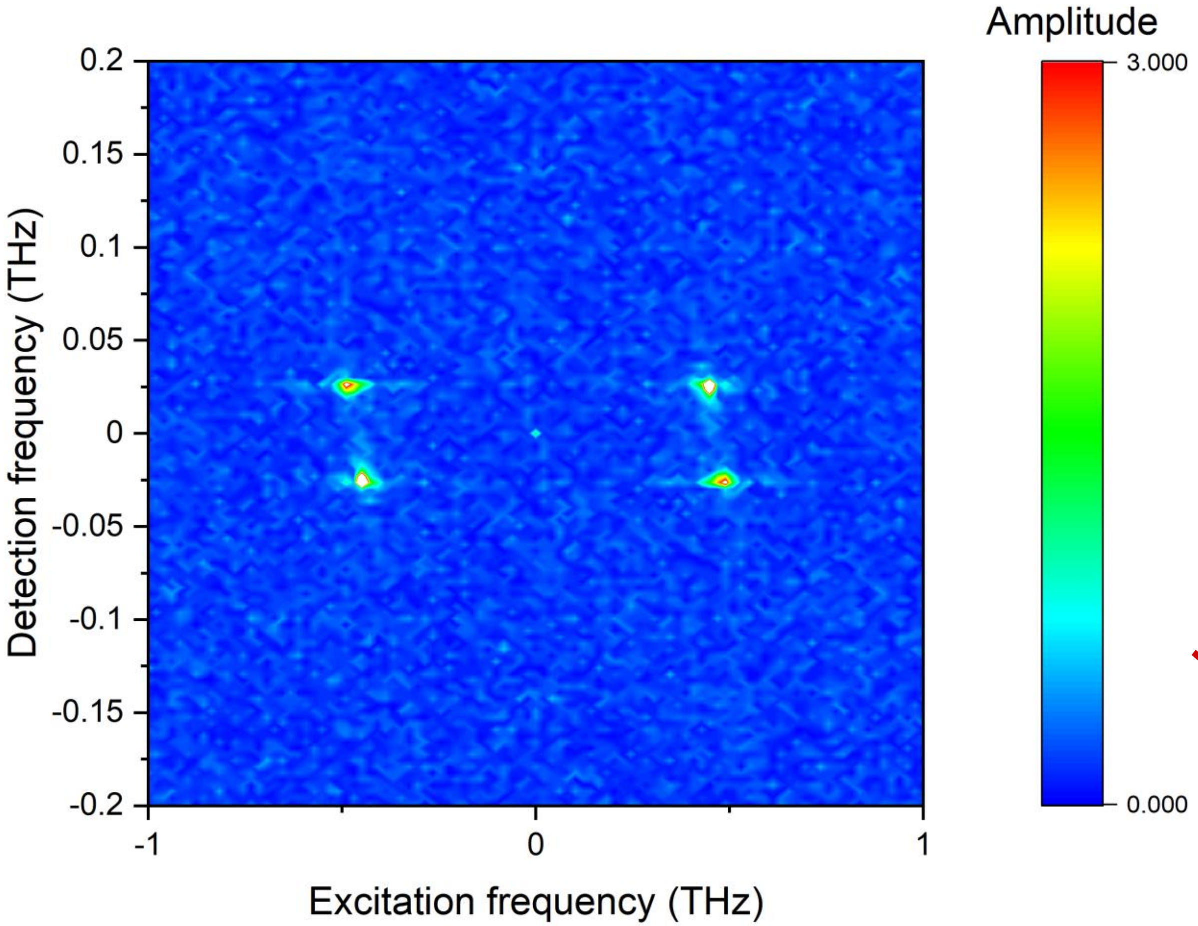
Double pulse excitation of $FeBO_3$

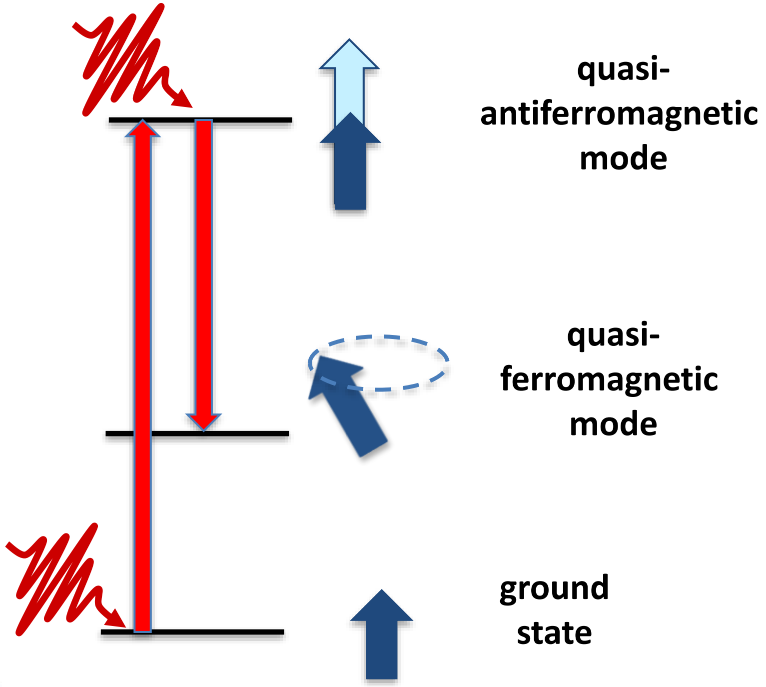


Nonlinear signal = **Full signal** – **Dynamics after the 1st pulse only** – **Dynamics after the 2nd pulse only**

T. Blank et al
https://arxiv.org/abs/2212.09532.

Nonlinear 2D THz spectroscopy of $FeBO_3$



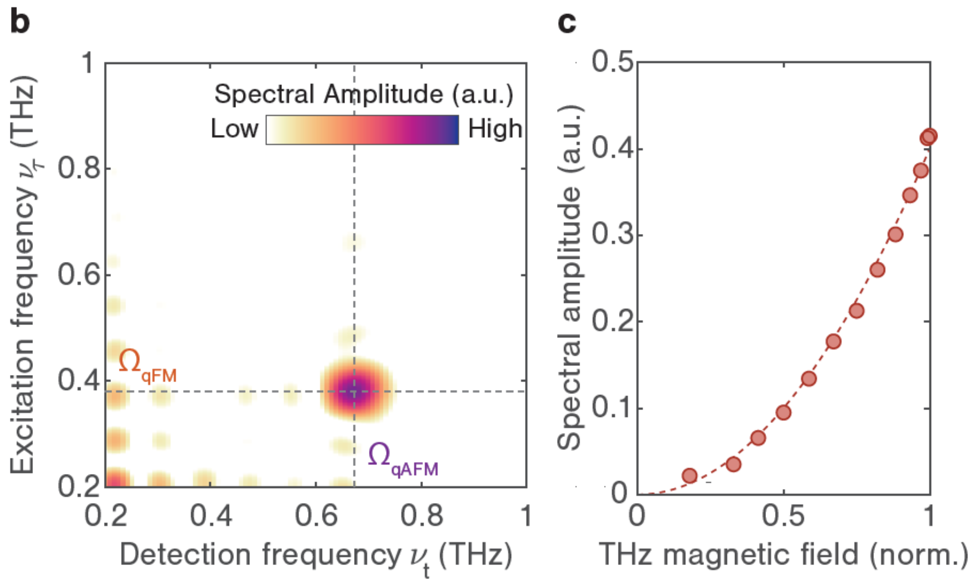


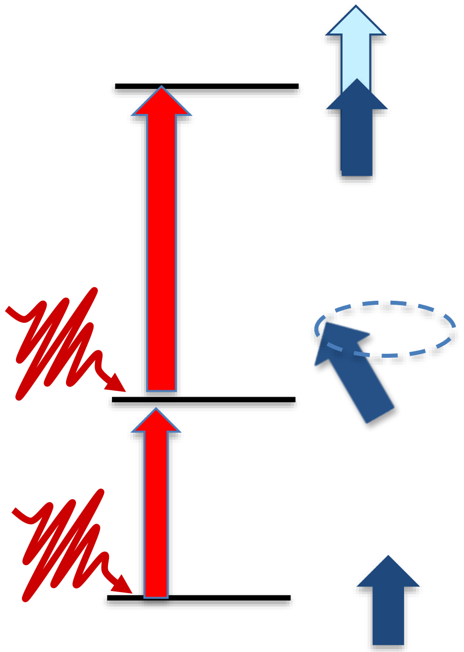
T. Blank et al, https://arxiv.org/abs/2212.09532.
T. Blank et al, PRL **131**, 096701 (2023).

Terahertz field-driven magnon upconversion in an antiferromagnet

Zhuquan Zhang[1†], Frank Y. Gao[2†], Yu-Che Chien[1], Zi-Jie Liu[1], Jonathan B. Curtis[3], Eric R. Sung[1], Xiaoxuan Ma[4], Wei Ren[4], Shixun Cao[4*], Prineha Narang[3], Alexander von Hoegen[5], Edoardo Baldini[2*], and Keith A. Nelson[1*]

arXiv:2207.07103





quasi-antiferromagnetic mode

quasi-ferromagnetic mode

ground state

THz antiferromagnetism

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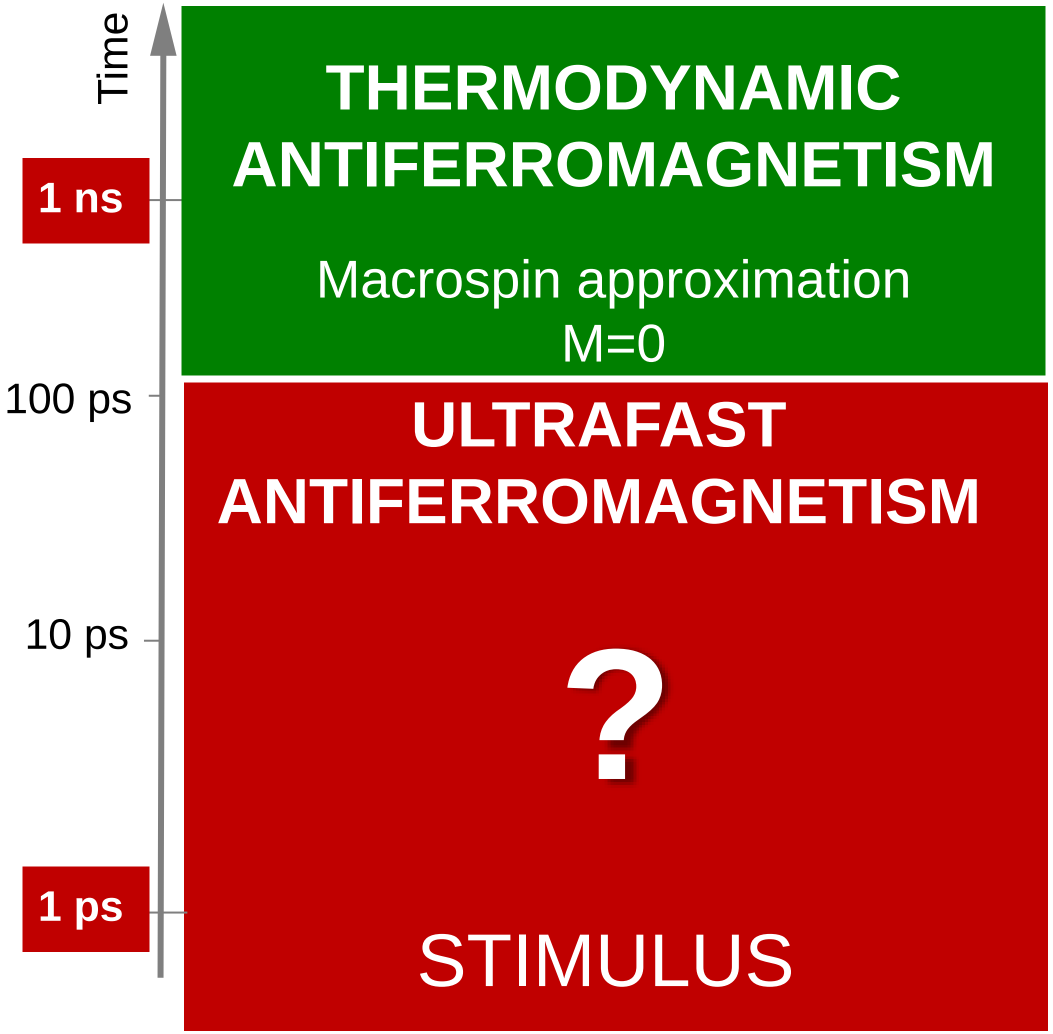
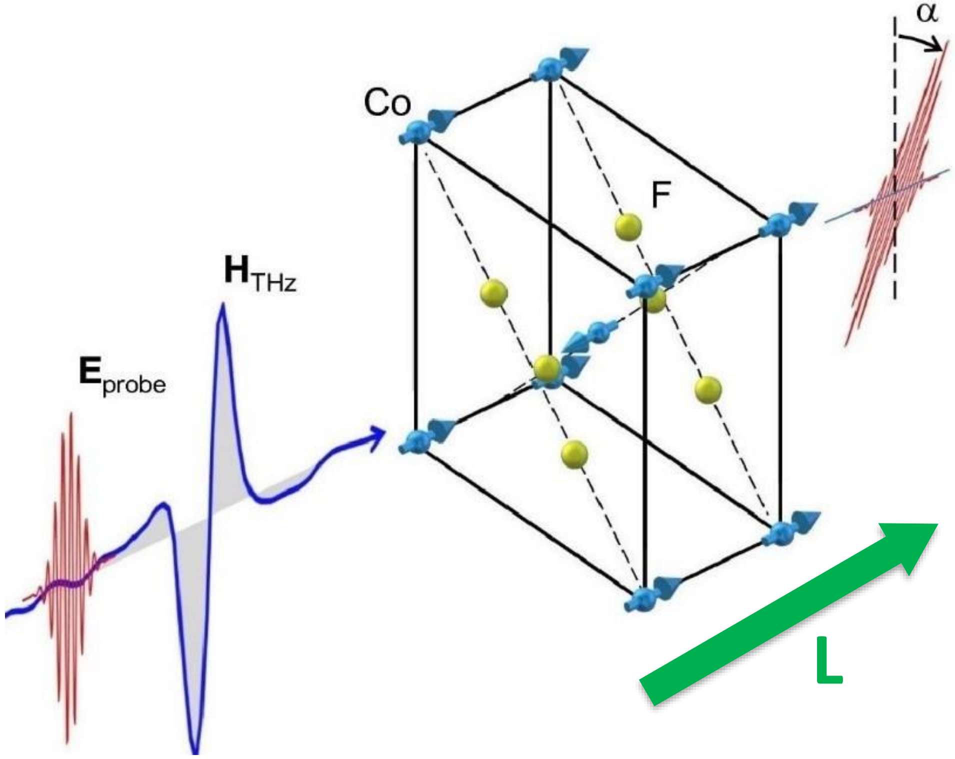


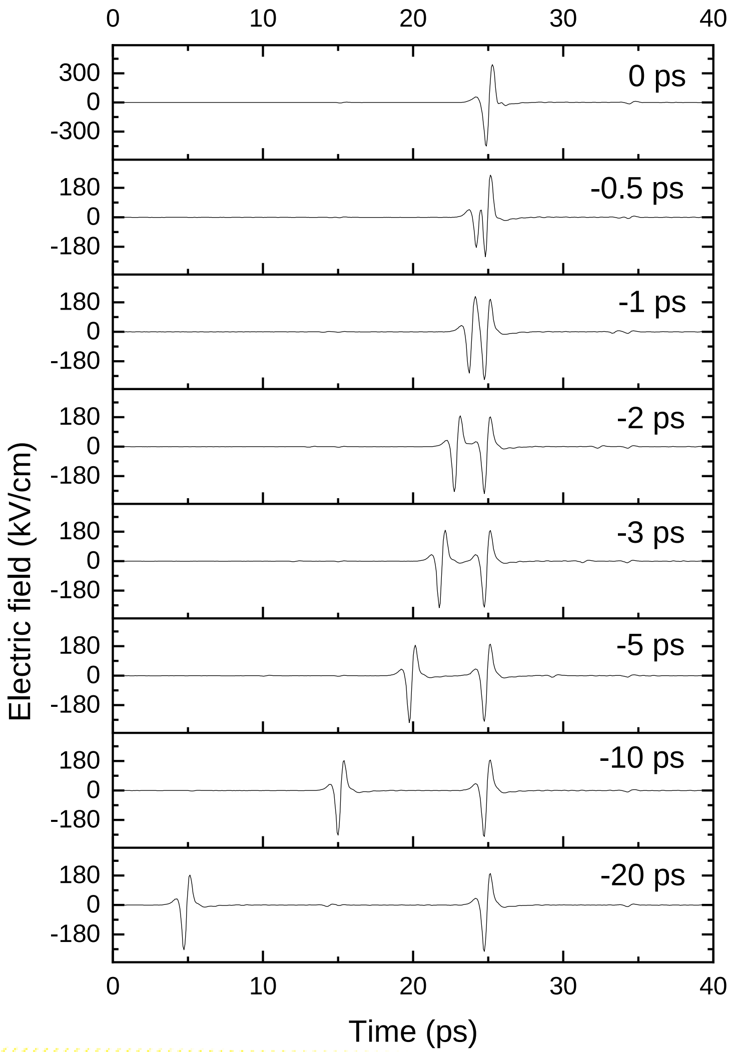
- M≠0
- Nonlinear spin dynamics (1+1>2)
- **New channels of spin-lattice interaction**
- Macrospin approximation fails

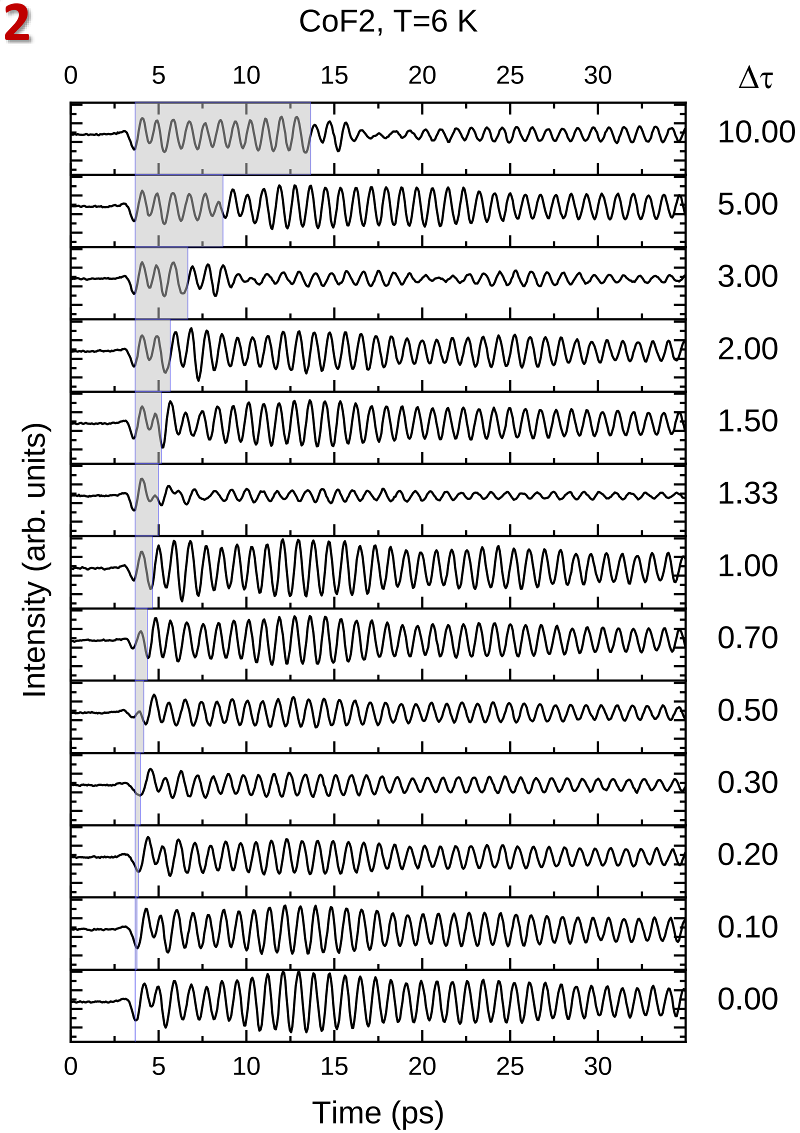
Response of antiferromagnetic spins in CoF_2 to THz magnetic fields

CoF_2
T_N=39 K



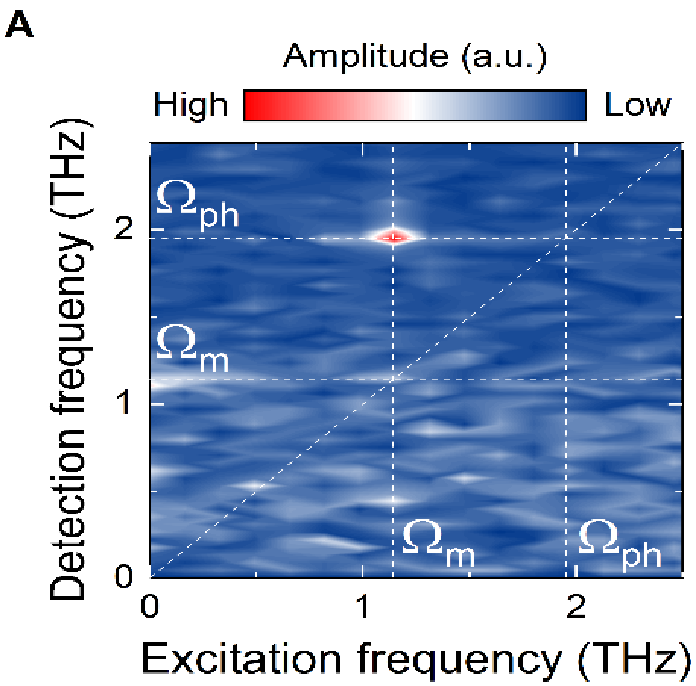
Double-pulse THz excitation of CoF_2





E. Mashkovich et al Science **374** ,1608 (2021).

2D THz spectroscopy of CoF_2

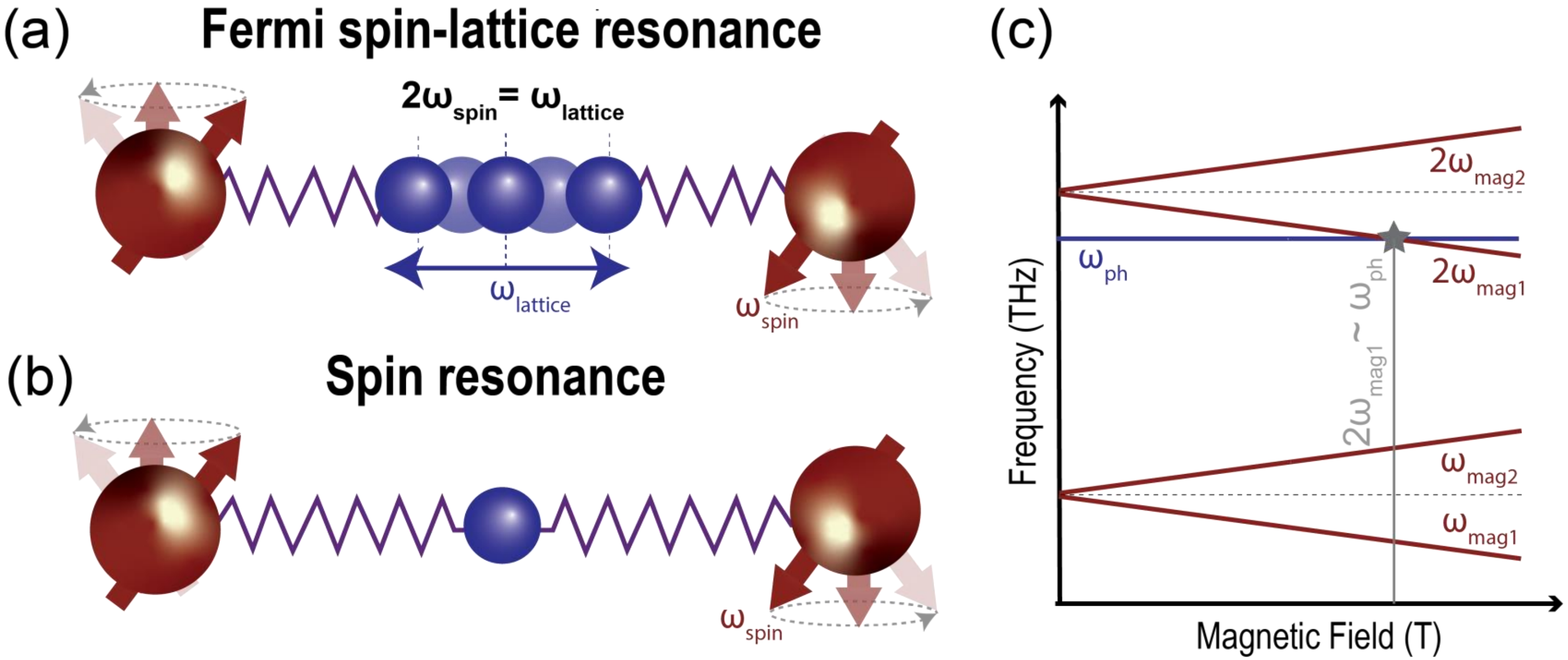


$$\alpha_{NL}(t_{pump1-probe}; t_{pump1-pump2}) = \alpha(t_{pump1-probe}; t_{pump1-pump2}) - \alpha1(t_{pump1-probe}) - \alpha2(t_{pump2-probe})$$

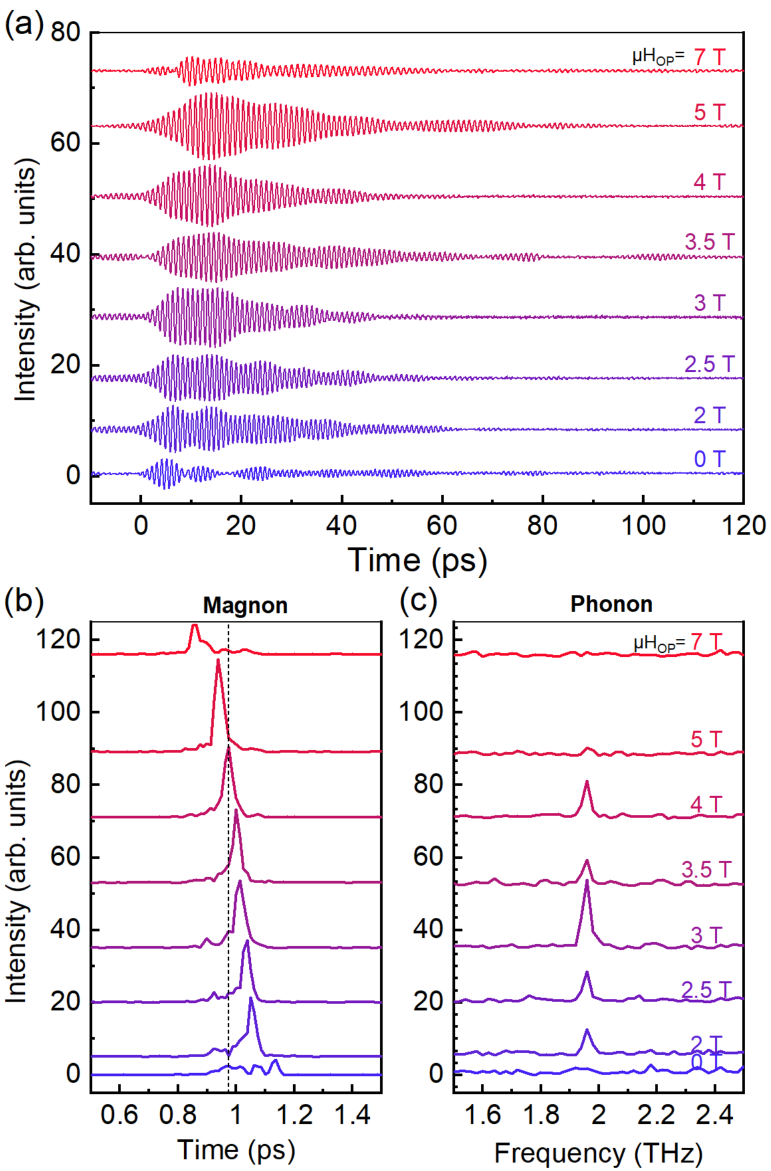
Nonlinear signal = **Full signal** – **Dynamics after the 1st pulse only** – **Dynamics after the 2nd pulse only**

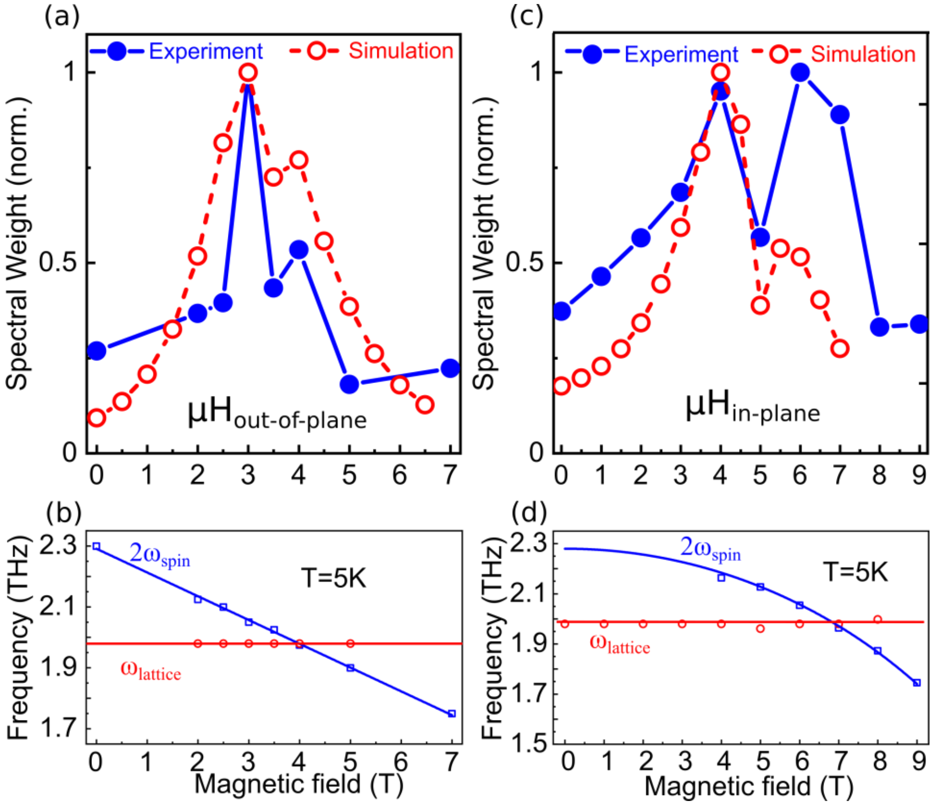
E. Mashkovich et al Science **374** ,1608 (2021).

Spin-lattice Fermi resonance in CoF$_2$



Spin-lattice Fermi resonance in CoF_2





T. Metzger et al, arXiv:2308.01052.

THz antiferromagnetism

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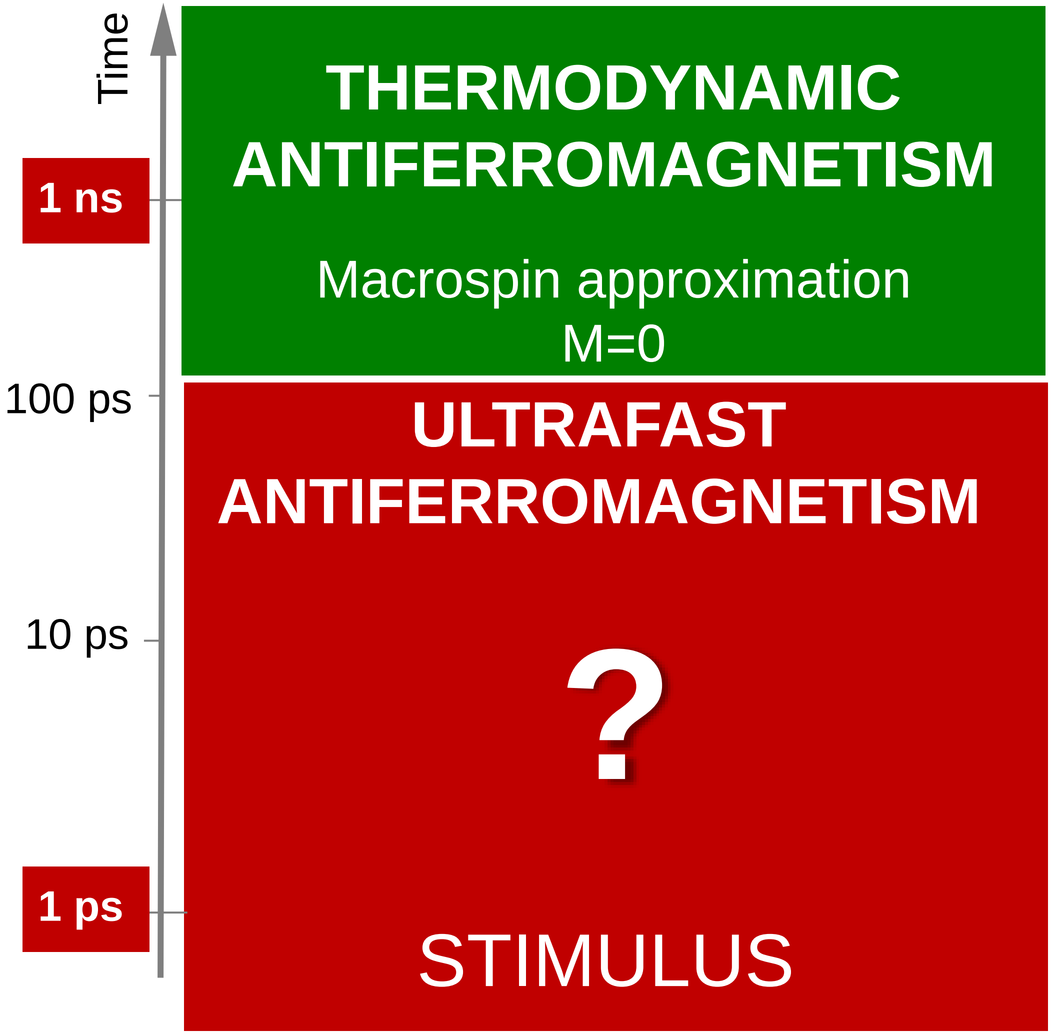

THERMODYNAMIC ANTIFERROMAGNETISM

Macrospin approximation
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ULTRAFAST ANTIFERROMAGNETISM

?

STIMULUS

- M≠0
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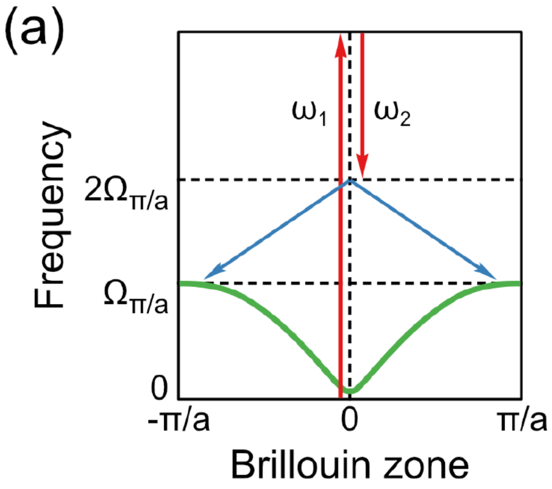
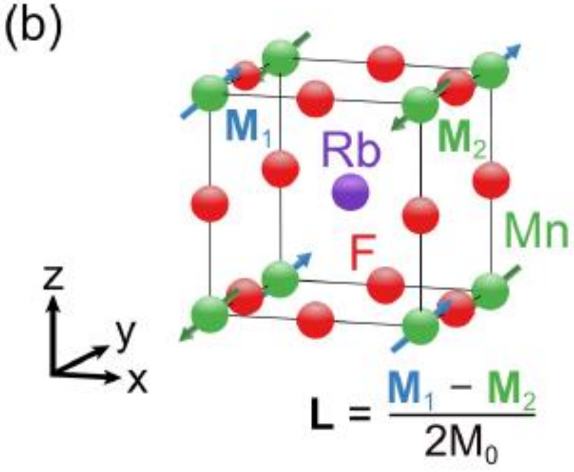
- **New channels of spin-lattice interaction**
- **Macrospin approximation fails**

Two-magnon excitation in antiferromagnet



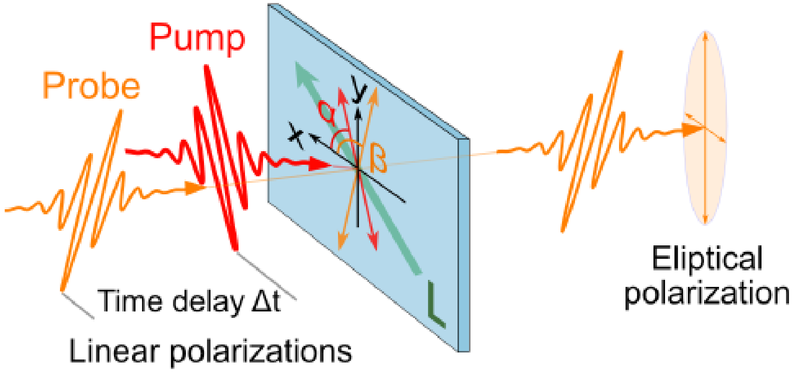


Thermodynamic potential

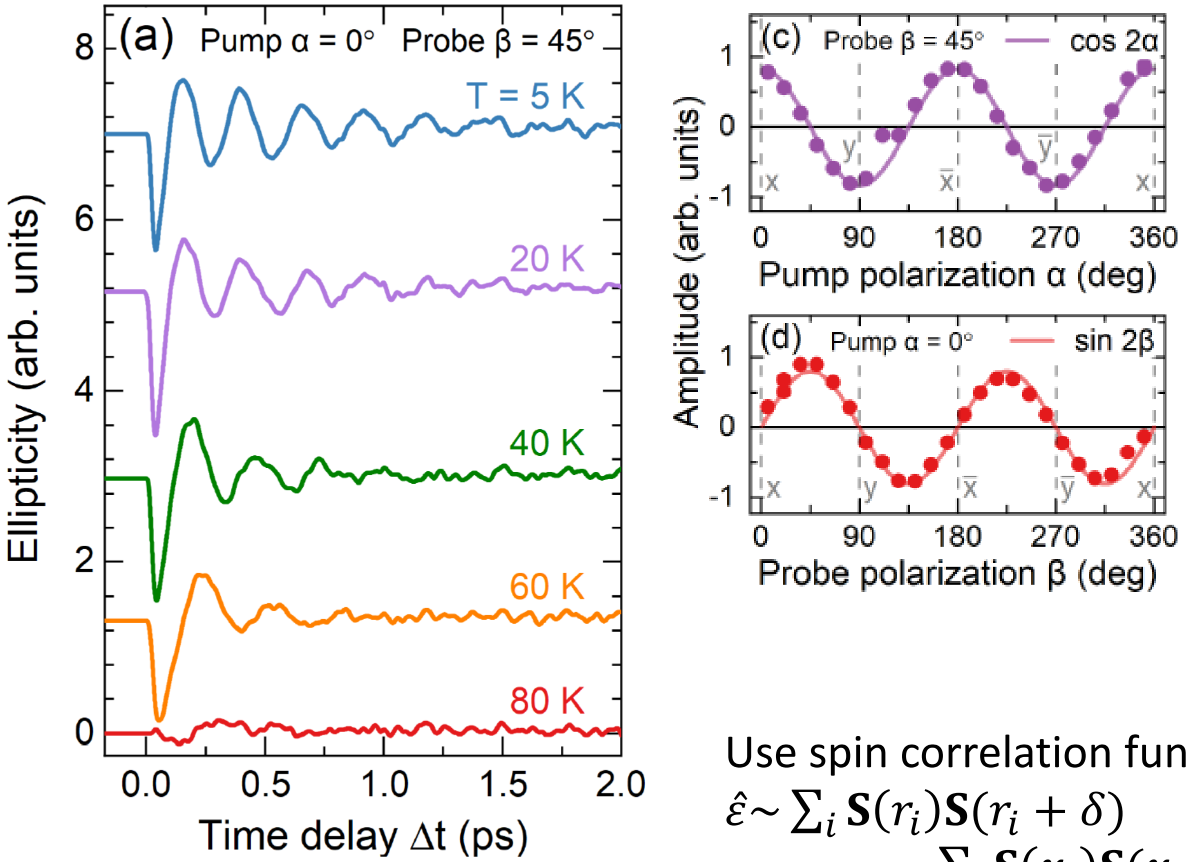
$$\Phi = \Phi_0 + \chi_{ijkl} E_i(\omega_1) E_j^*(\omega_2) L_k(\Omega;\mathbf{k}) L_l^*(\Omega;\mathbf{k})$$

Probe

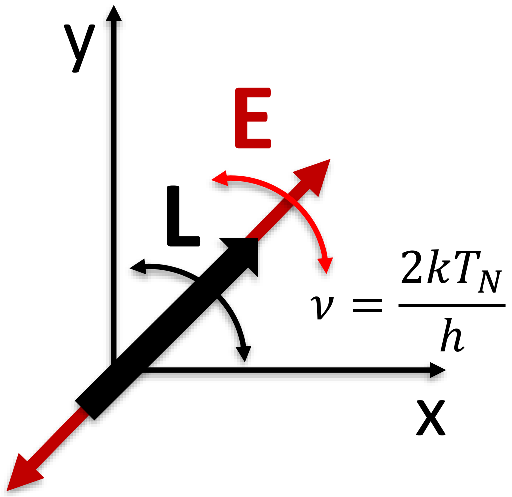
$$\varepsilon_{ij} = \frac{\partial^2 \Phi}{\partial E_i(\omega) \partial E_j^*(\omega)} = \chi_{ijkl} L_k(\Omega;\mathbf{k}) L_l^*(\Omega;\mathbf{k})$$



Two-magnon excitation in antiferromagnetic RbMnF$_3$



F. Formisano et al,
https://arxiv.org/abs/2303.06996.



$$\nu = \frac{2kT_N}{h}$$

Such fast oscillations of **L** are impossible!

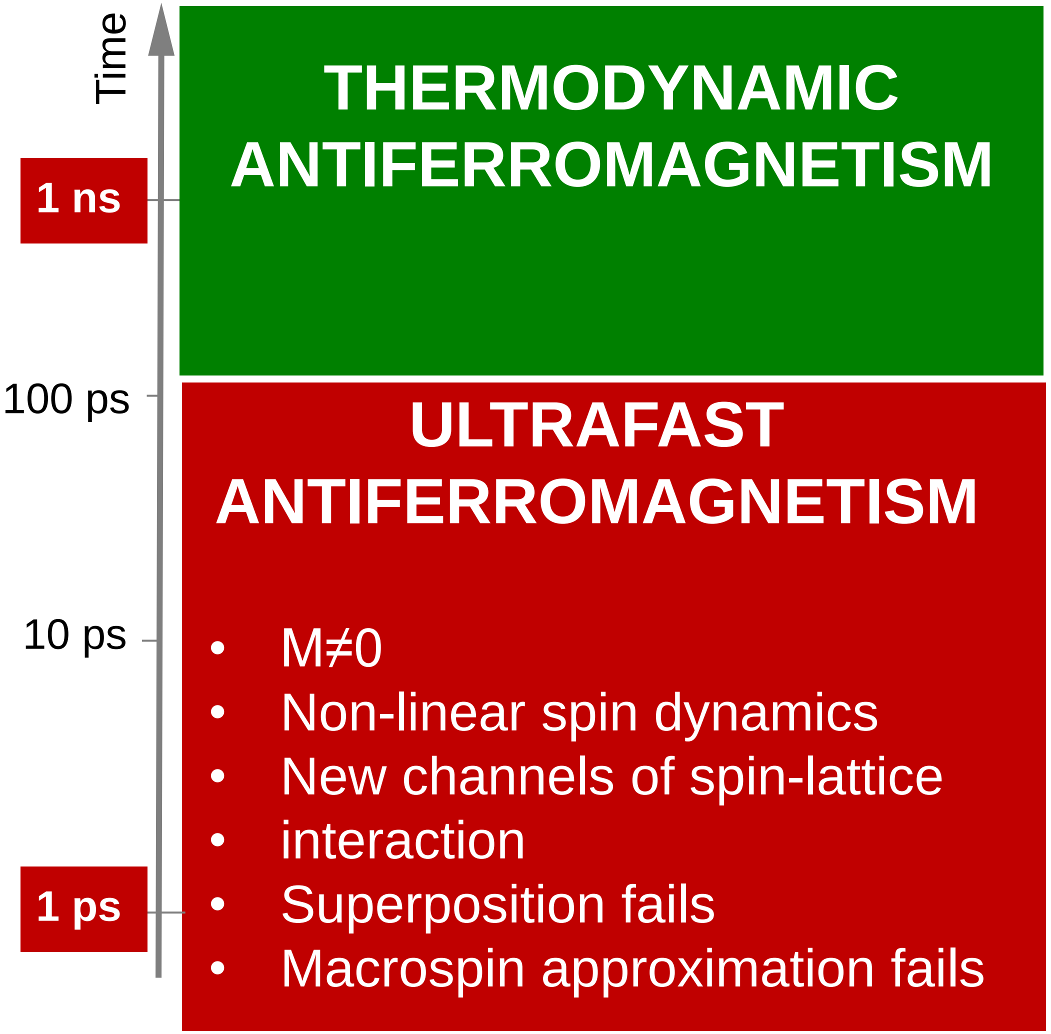
Use spin correlation function $\sum_i \mathbf{S}(r_i)\mathbf{S}(r_i+\delta)$ instead of $\mathbf{L}$.

$\hat{\varepsilon} \sim \sum_i \mathbf{S}(r_i)\mathbf{S}(r_i+\delta)$

$$\sum_i \mathbf{S}(x_i)\mathbf{S}(x_i+\delta) \neq \sum_i \mathbf{S}(y_i)\mathbf{S}(y_i+\delta)$$

$$\varepsilon_{xx} \neq \varepsilon_{yy}$$

Conclusions and Outlook



Time

1 ns

THERMODYNAMIC ANTIFERROMAGNETISM

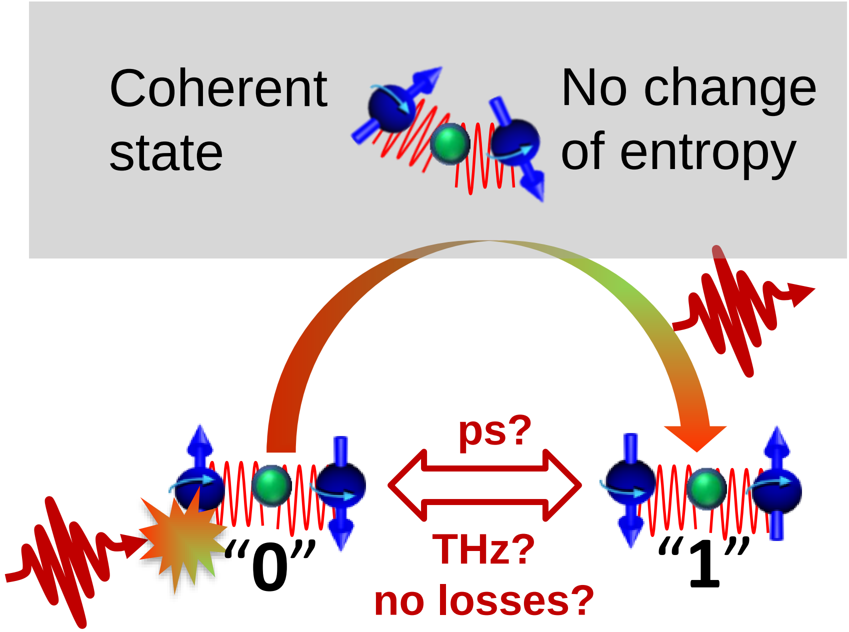
100 ps

ULTRAFAST ANTIFERROMAGNETISM

10 ps

- M≠0
- Non-linear spin dynamics
- New channels of spin-lattice
- interaction
- Superposition fails
- Macrospin approximation fails

1 ps



Coherent state

No change of entropy

“0”

ps?

THz?

no losses?

“1”

Acknowledgements

T. Blank, T. Metzger, T. Gareev, D. Khusyainov,
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B. A. Ivanov, M. I. Katsnelson

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A. Kalashnikova		*J.-Ch. Deinert*

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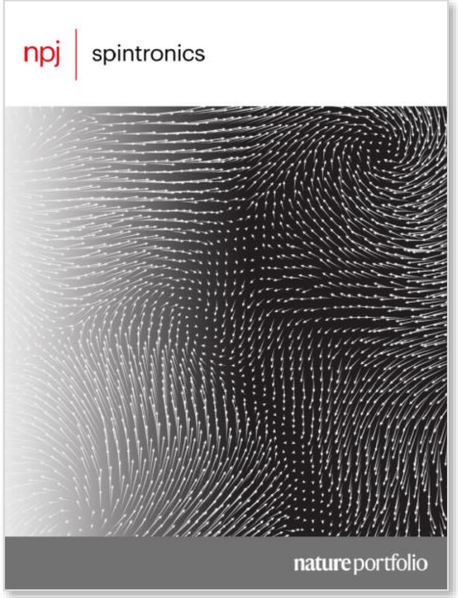

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