Combining ferroelectricity and magnetism: the low energy electrodynamics

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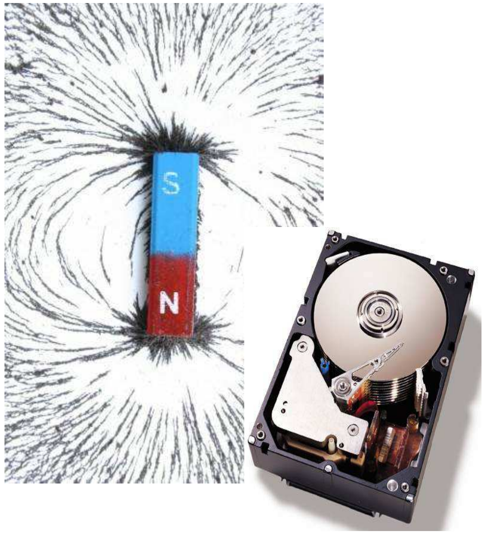
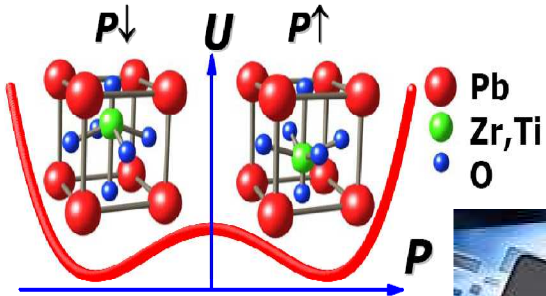
Ferroic properties





FERROMAGNETISM

High-power current-driven write operation

FERROELECTRICITY

Low-power voltage-driven write

But…

Issues of fatigue need to be overcome

… enter **MULTIFERROICITY!!!**

Magnetoelectric multiferroics

MULTIFERROICS – materials with coexisting magnetism and ferroelectricity.

Magnetic order ⟷ Electric polarization

magnetoelectric interaction

Possible applications for magnetoelectric materials

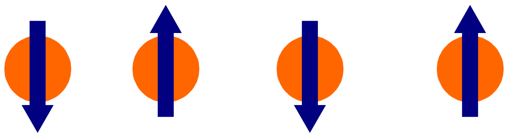
Magnetoelectric memory
Magnetically switched electro-optic device
Electric-field-modulated visible Faraday rotator
Magnetically (electrically)-modulated piezoelectric (piezomagnetic) devices, etc.

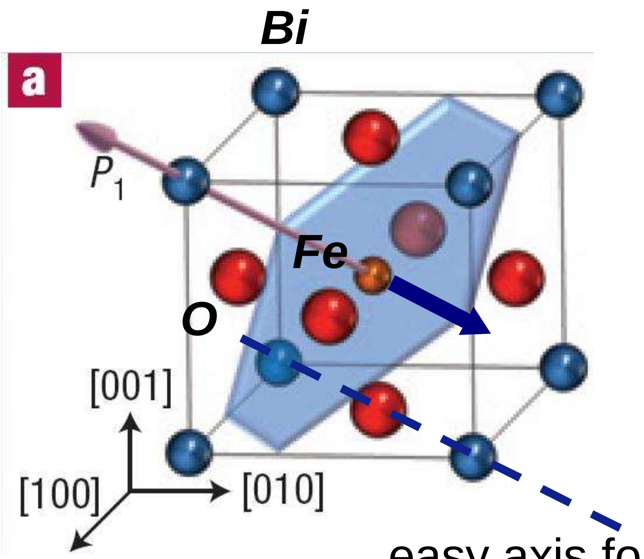
V. E. Wood, A. F. Austin, Int. J. Magnetism 5, 303-315 (1974)

Magnetoelectric effect

$BiFeO_3$: ferroelectric antiferromagnet

Fe ions:





easy axis for magnetization

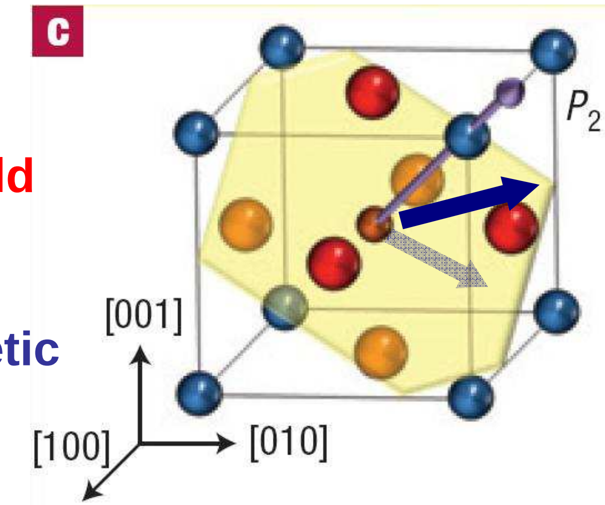
Y.-H. Chu et al.,

Nat. Materials **7** 478 (2008)

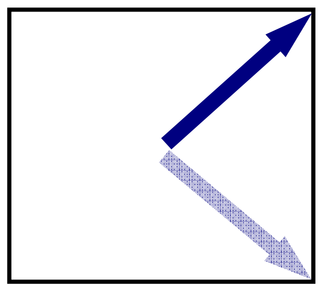
Apply electric field

E

Turn the magnetic moments!

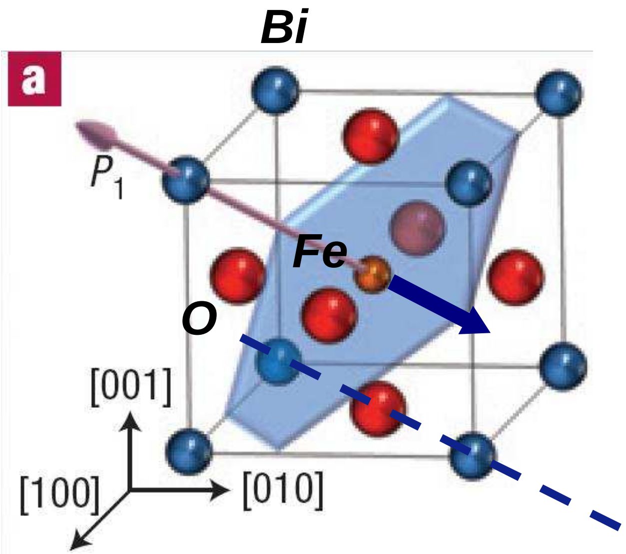


Top view

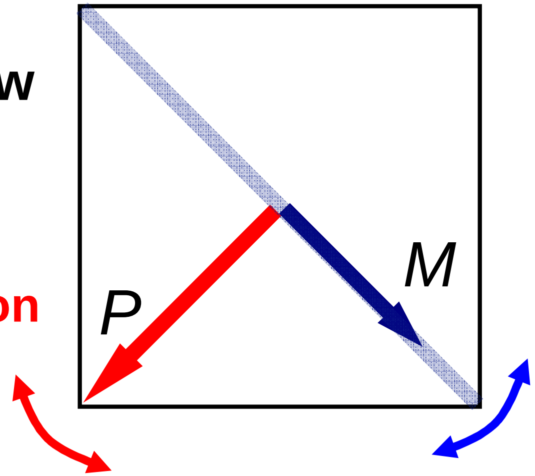


Dynamic magnetoelectric effect

$BiFeO_3$



Top view



Rock the polarization

excite phonons

Magnetic moments respond!

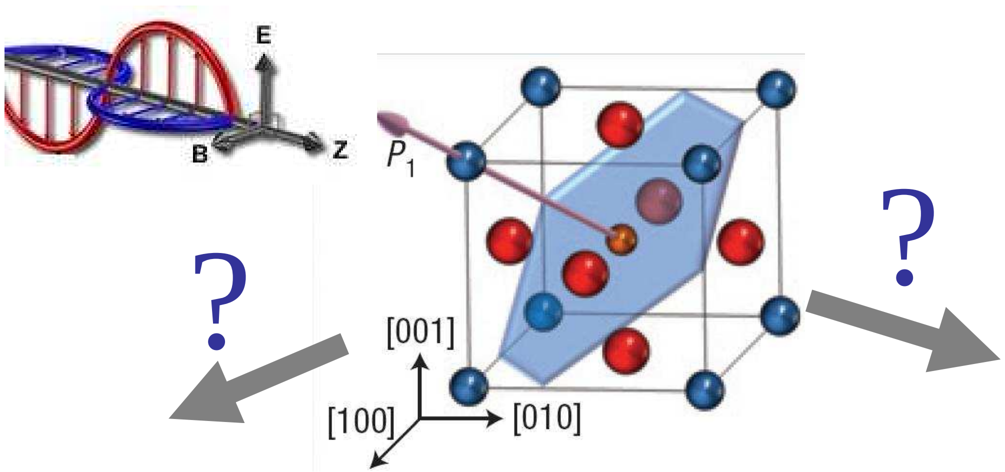
Mixing of magnetic and lattice vibrations –
DYNAMIC MAGNETOELECTRIC EFFECT

Y.-H. Chu et al.,

Nat. Materials **7** 478 (2008)

Magnetic and lattice motion interacts with light

Electromagnetic wave incident on the crystal



Phonons ~ oscillating electric dipole

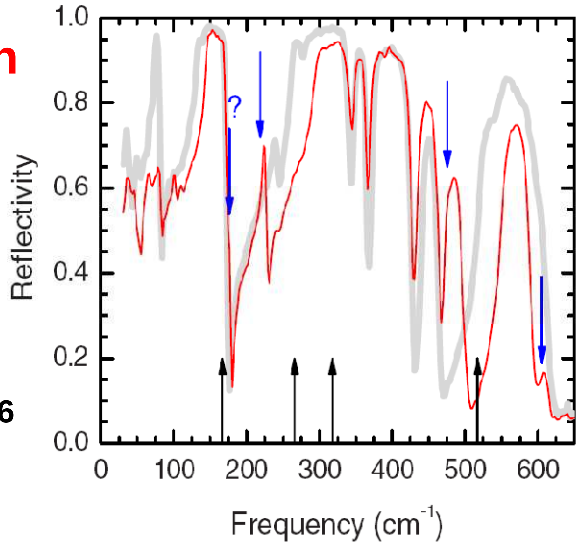
→ Coupling to the electric field of the light wave

→ Resonance in the dielectric function, a.k.a. optical conductivity:

$$\varepsilon(\omega) \leftrightarrow \sigma(\omega)$$

33 cm^{-1} ~ 1 THz ~ 4 meV

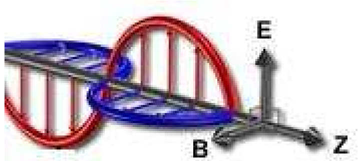
Phonons in $BiFeO_3$



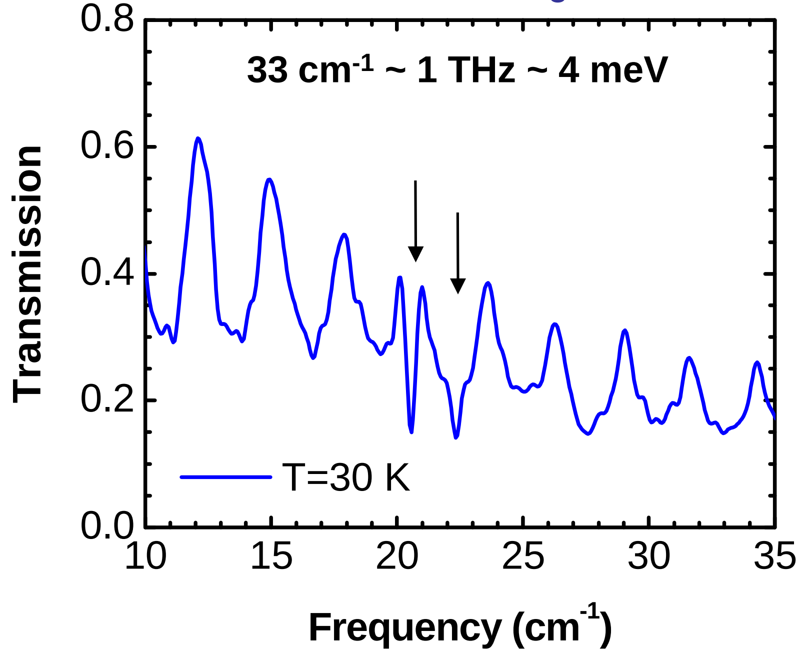
Lobo et al.,
Phys. Rev. B **76** 172105 (2007)

Magnetic and lattice motion interacts with light

Electromagnetic wave incident on the crystal



Magnetic excitations in $BiFeO_3$



D. Talbayev et al., unpublished

Magnons ~ oscillating magnetic dipole

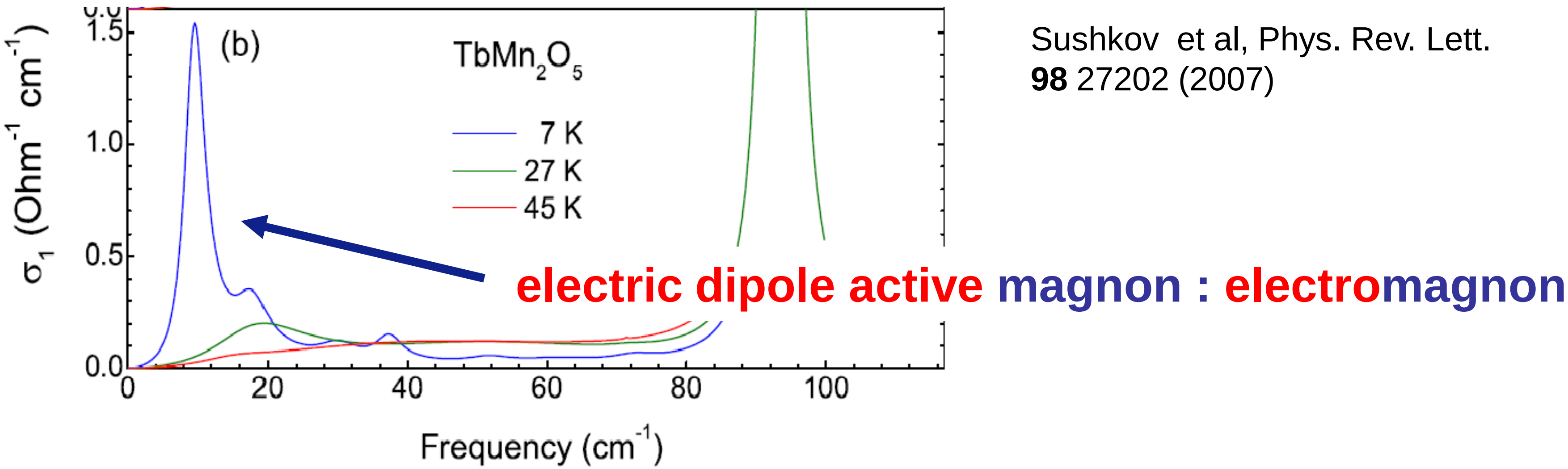
→ Coupling to the magnetic field of the light wave

→ Resonance in the magnetic susceptibility:

$$\chi(\omega) \leftrightarrow \mu(\omega)$$

Mix it up: electromagnons

$TbMn_2O_5$: antiferromagnet and ferroelectric



Sushkov et al, Phys. Rev. Lett. **98** 27202 (2007)

Similar observations : multiferroics $TbMnO_3$, $Eu_{0.75}Y_{0.25}MnO_3$

Pimenov et al., Nature Phys. **2** 97 (2006)

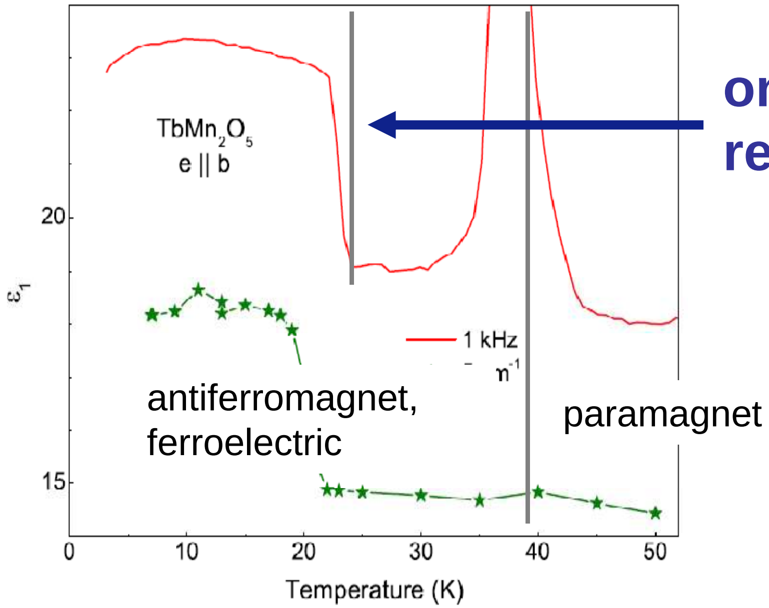
Valdes Aguilar et al., Phys. Rev. B 76 060404R (2007)

Pimenov et al., Phys. Rev. B 77 014438 (2008)

Electromagnons determine magnetoelectric functionality

$TbMn_2O_5$: antiferromagnet and ferroelectric

Sushkov et al, Phys. Rev. Lett. **98** 27202 (2007)



onset of electromagnon response

(i) **Electromagnon contributes to static dielectric constant:**

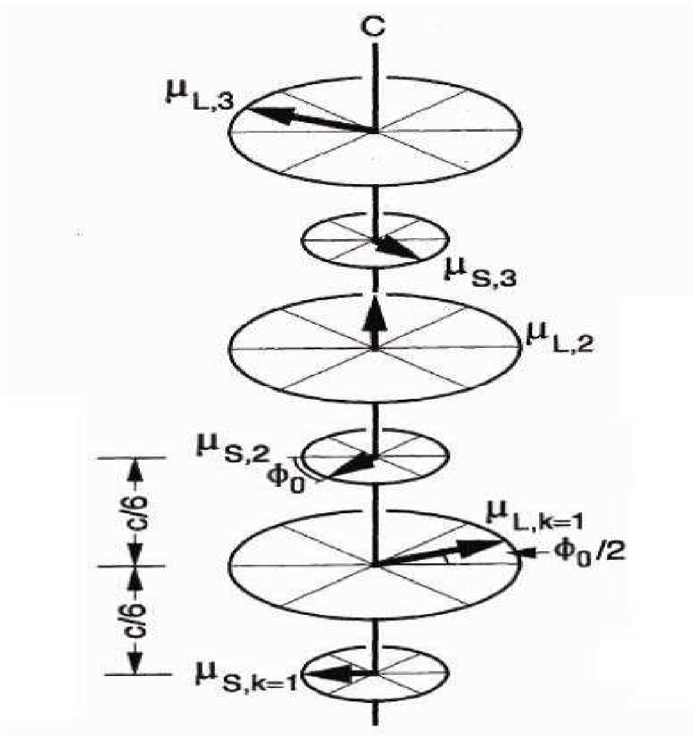
$$\varepsilon_1(0) = 1 + 8\int_0^\infty \frac{\sigma_1(\omega)}{\omega^2} d\omega$$

(ii) **Electromagnon properties depend on the microscopic magnetoelectric coupling**

i) Properties of electromagnons

Dynamic magnetoelectric effect in hexagonal $Ba_{0.6}Sr_{1.4}Zn_2Fe_{12}O_{22}$

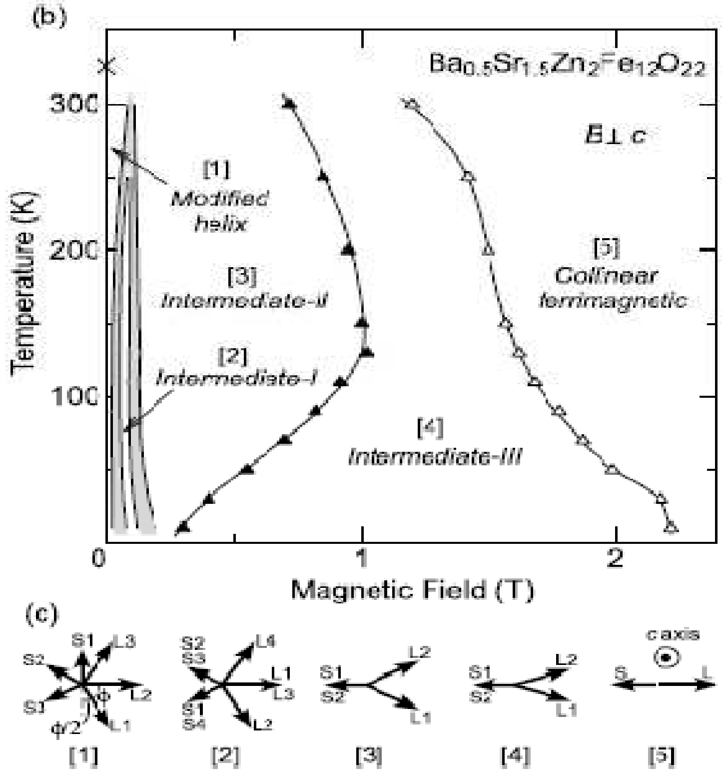
Layered magnetic structure



N. Momozawa and Y. Yamaguchi,

J. Phys. Soc. Jpn. **62** 12992 (1993)

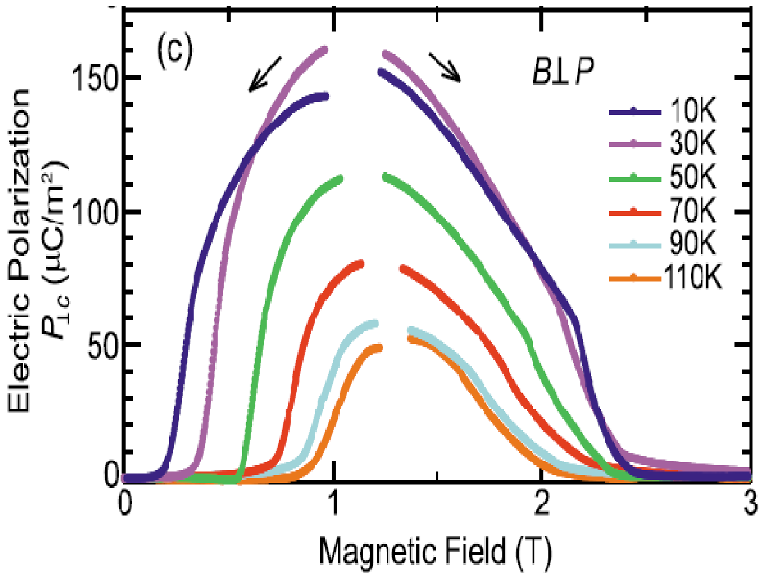
Several magnetic phases

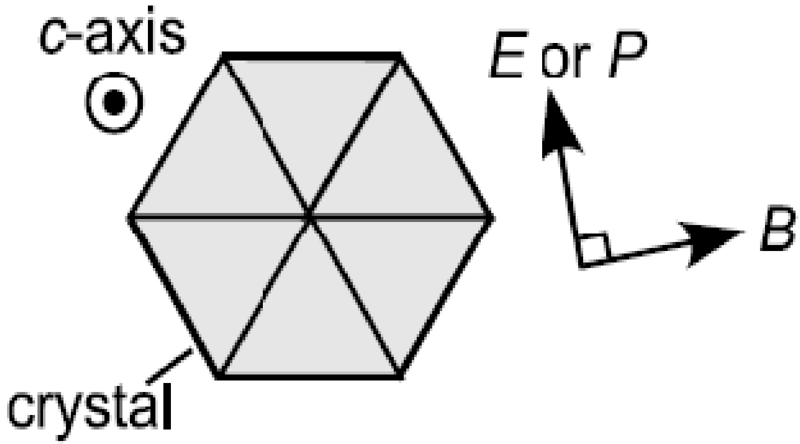


T. Kimura, G. Lawes, and A.P. Ramirez,
PRL **94** 137201 (2005)

Dynamic magnetoelectric effect in hexagonal $Ba_{0.6}Sr_{1.4}Zn_2Fe_{12}O_{22}$

Control of electric polarization by magnetic field:



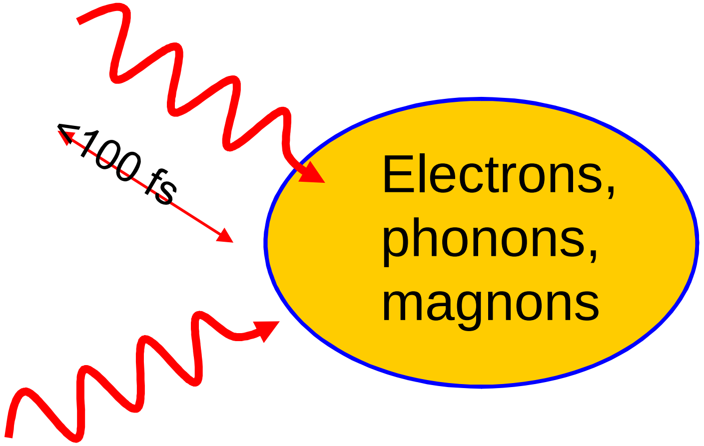


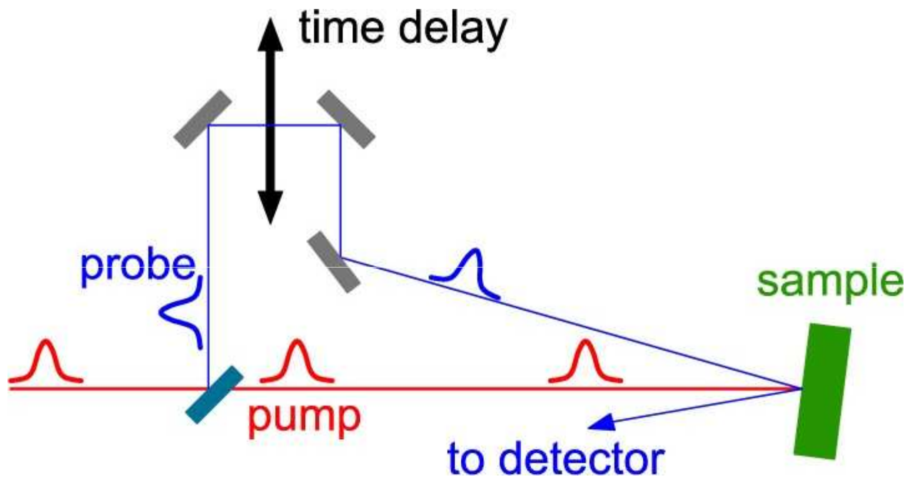
T. Kimura, G. Lawes, and A.P. Ramirez, PRL **94** 137201 (2005)

Time-domain studies of elementary excitations

visible optical pulse excitation, a.k.a. pump

Pump and probe wavelength: 800 nm (~1.5 eV)



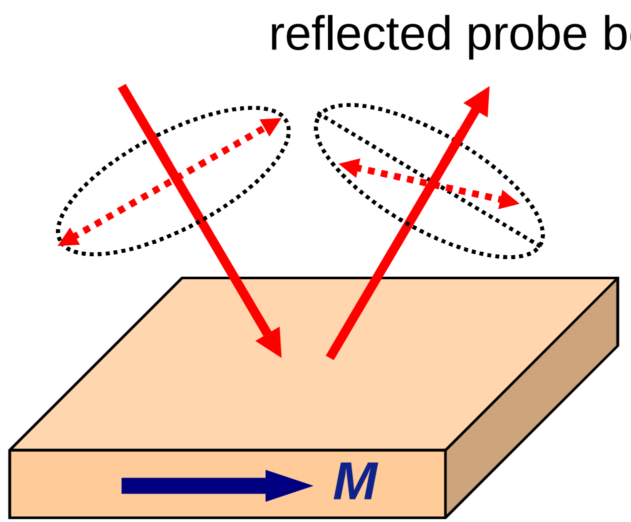


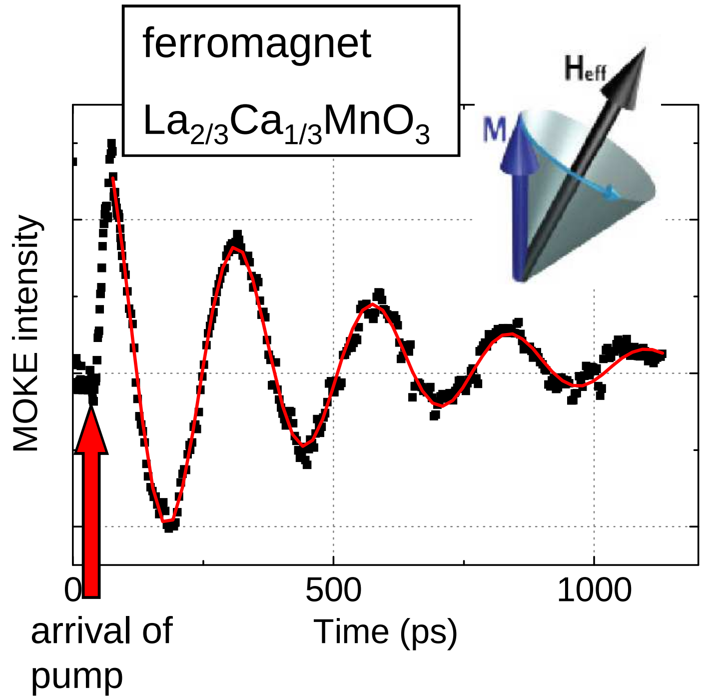
optical pulse interrogation (probe) of material's response

Dynamics of photo-excited quasiparticles often exposes properties not detected by conventional probes – transport, magnetization, or optical conductivity

Time-domain detection of magnetic motion

Magneto-optical Kerr effect (MOKE) – rotation of polarization of light upon reflection by a magnetized medium



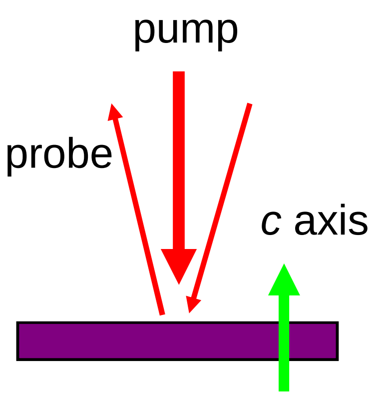


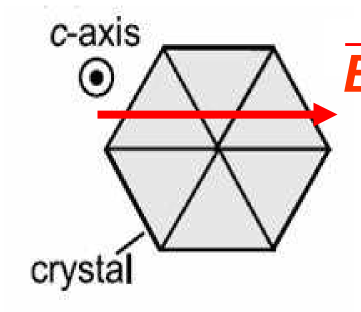
$$\dot{M} = -\gamma M \times H_{eff} \qquad H_{eff} = H_0 + H_{anisotropy} + H_{demag}$$

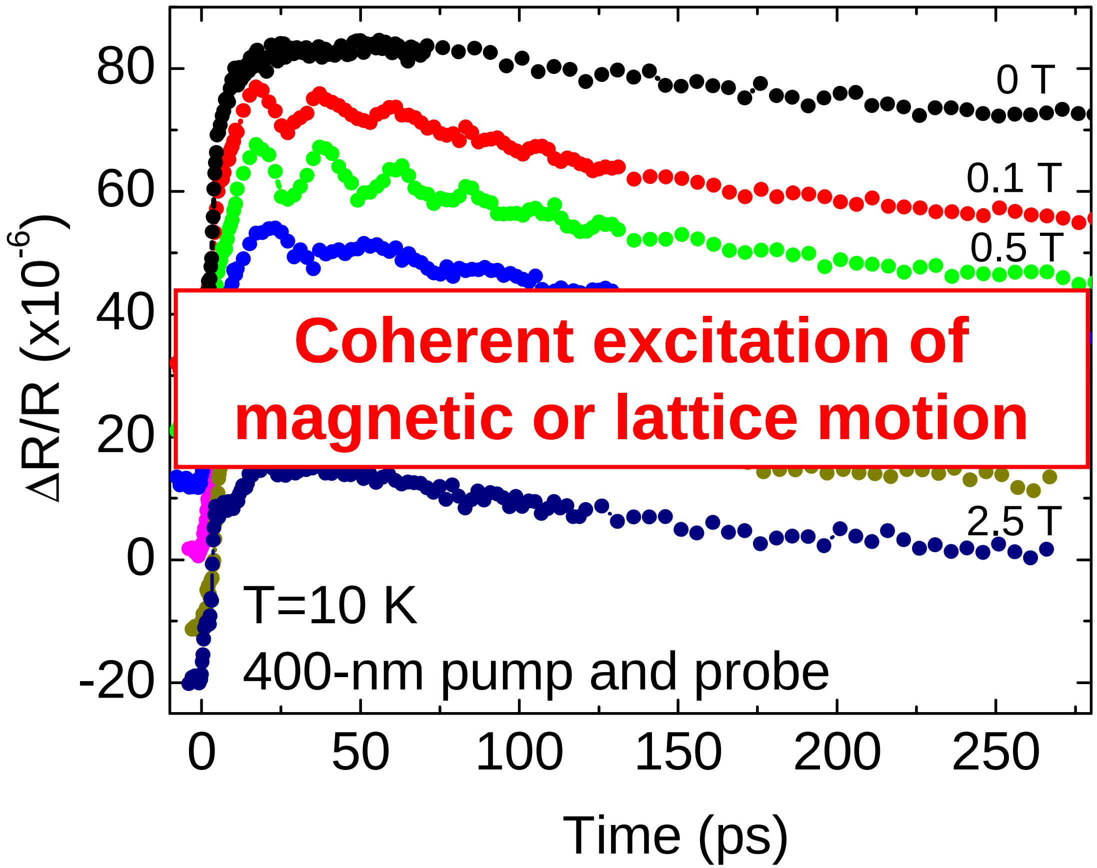
D. Talbayev et al, Phys. Rev. B **73** 14417 (2006); Appl. Phys. Lett. **86** 182501 (2005)

Time-resolved reflectance : coherent response

$Ba_{0.6}Sr_{1.4}Zn_2Fe_{12}O_{22}$: photoinduced change in reflectance

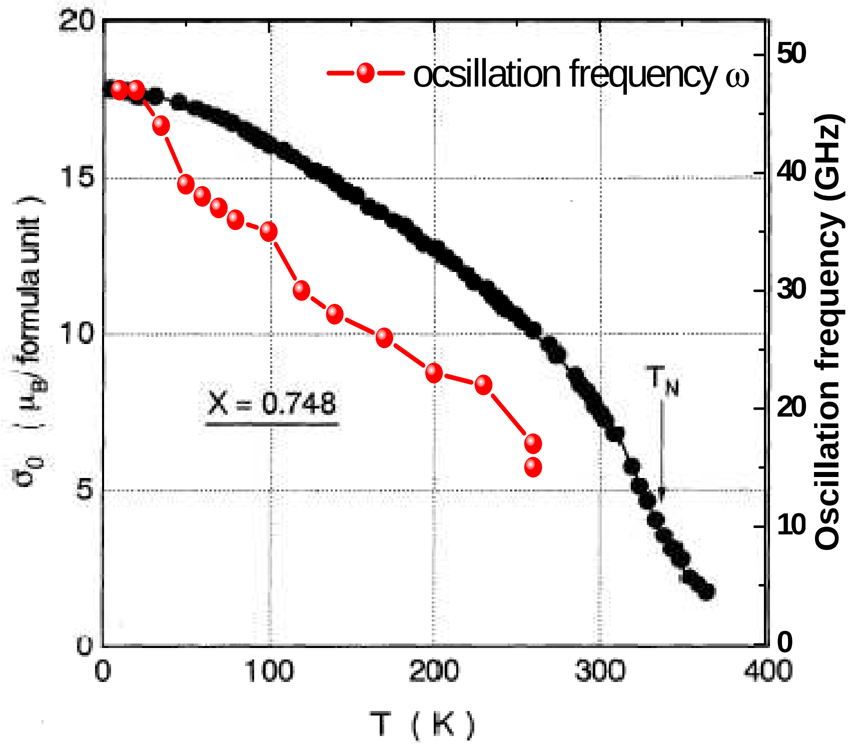




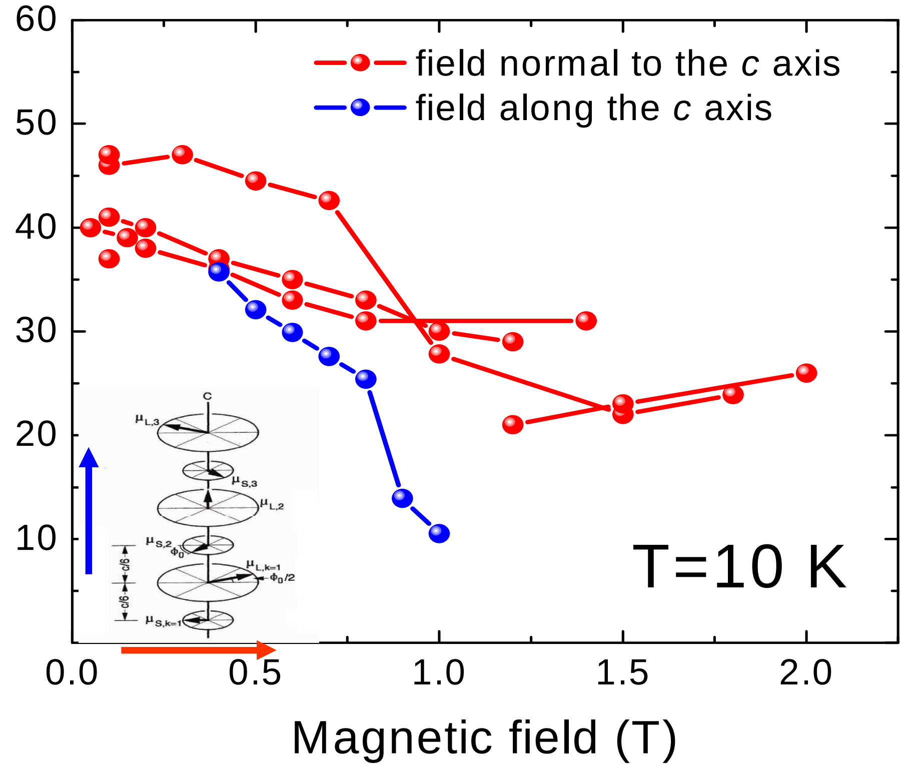


D. Talbayev et al., Phys. Rev. Lett. **101** 097603 (2008)

Magnetic or lattice?



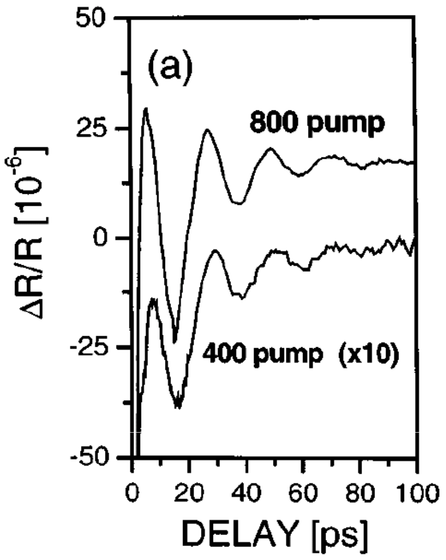
N. Momozawa and Y. Yamaguchi, J. Phys. Soc. Jpn. **62** 12992 (1993)



Strong evidence in support of magnetic origin of the excitation – electron spin resonance, i.e., *k*=0 magnon

D. Talbayev et al., Phys. Rev. Lett. **101** 097603 (2008)

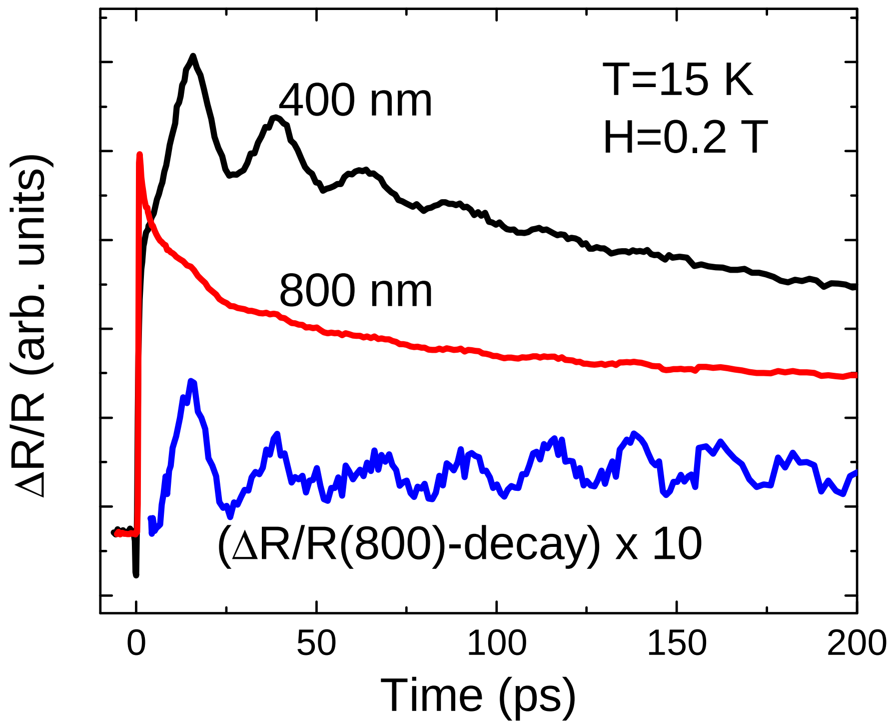
More evidence against the lattice



Oscillation frequency:

$$f_{PHONONS} \propto n / \lambda$$

D. Lim et al., Appl. Phys. Lett. **83** 4800 (2003)



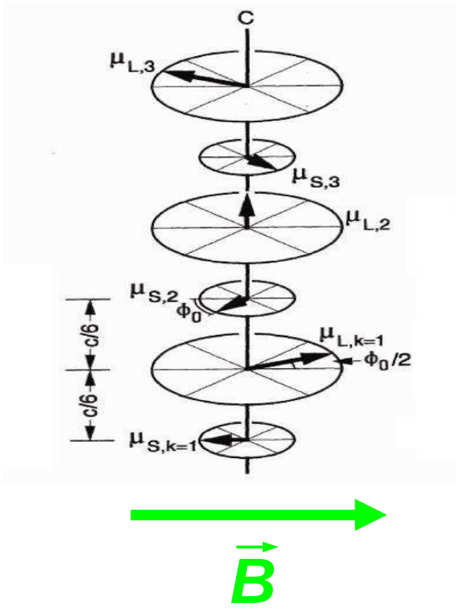
Identical oscillation frequency at different probe wavelengths

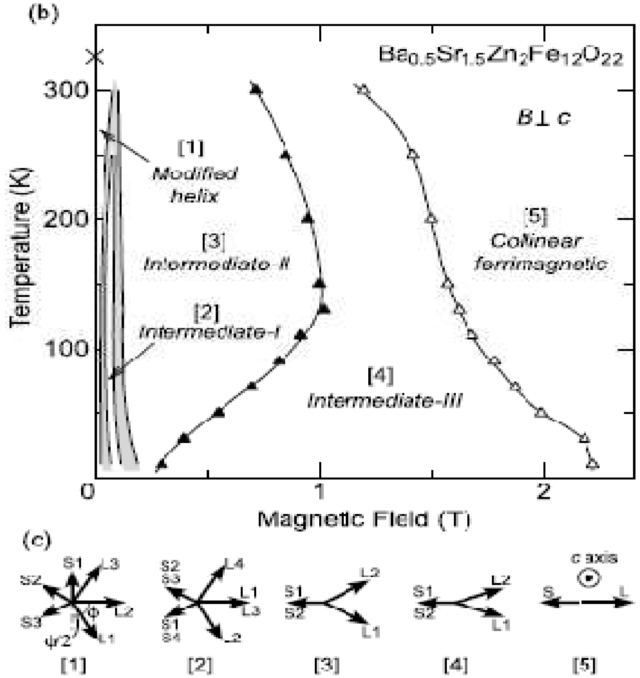
D. Talbayev et al., Phys. Rev. Lett. **101** 097603 (2008)

Los Alamos NATIONAL LABORATORY EST.1943

NNSA

Calculation of magnon frequencies





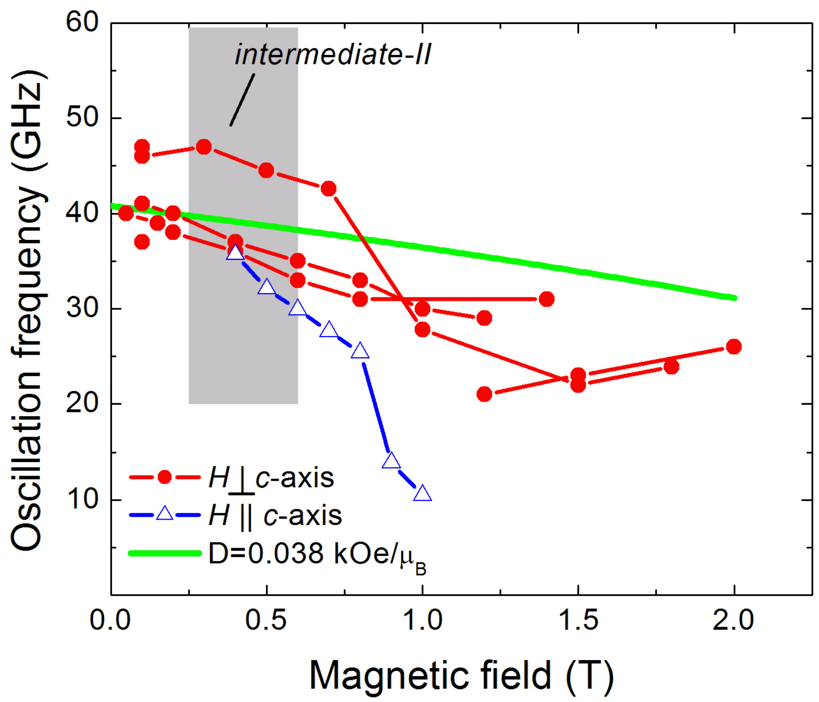
$$H = H_{ex}(M_{Si} \cdot M_{Lj}; M_{L1} \cdot M_{L2}; M_{S1} \cdot M_{S2}) + D\sum(M_{Liz}^2 + M_{Siz}^2)$$

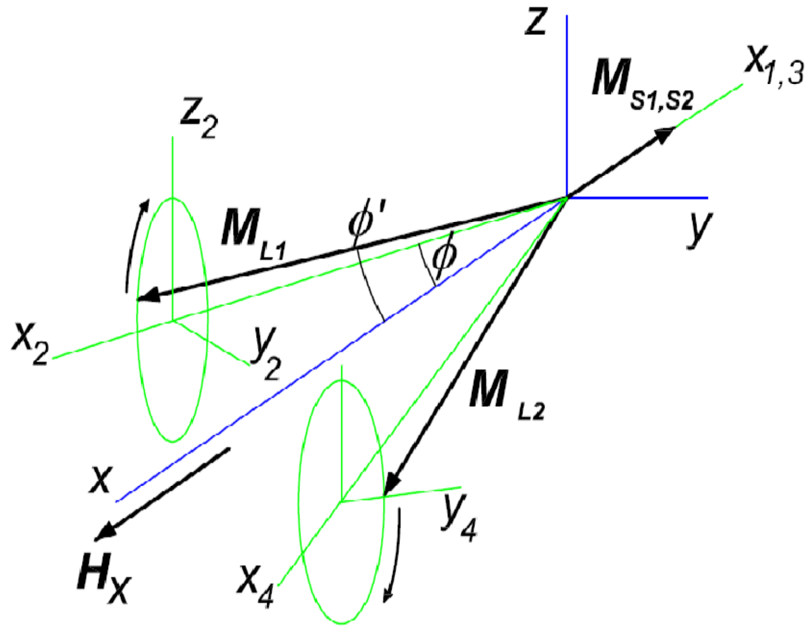
D – the only free parameter

T. Kimura, G. Lawes, and A.P. Ramirez, PRL **94** 137201 (2005)

N. Momozawa and Y. Yamaguchi,J. Phys. Soc. Jpn. **62** 12992 (1993)

Calculation of magnon frequencies

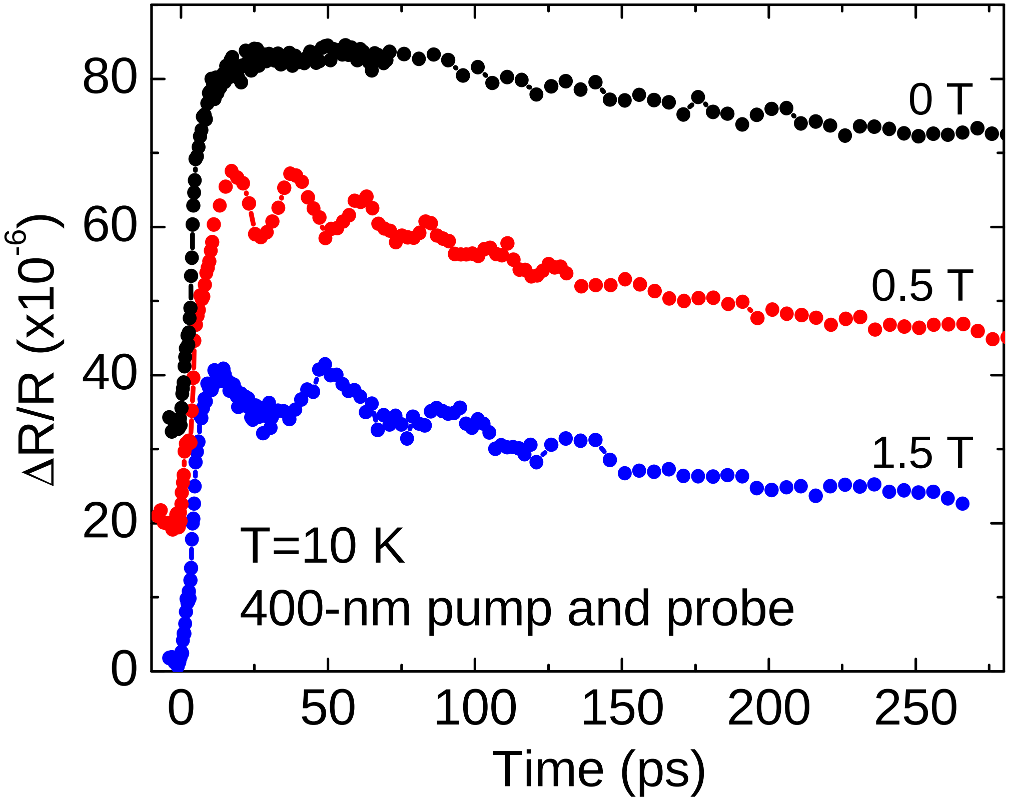




D. Talbayev et al., Phys. Rev. Lett. **101** 097603 (2008)

Dynamic magnetoelectric effect

$Ba_{0.6}Sr_{1.4}Zn_2Fe_{12}O_{22}$



Modulation of reflectance

$$R = \left| \frac{n-1}{n+1} \right|^2$$

$$n = \sqrt{\varepsilon}$$

Modulation of *n* and ε by magnon motion:

dynamic magnetoelectric effect

Optical detection of magnetic state using reflection – implications for data storage and spintronics.

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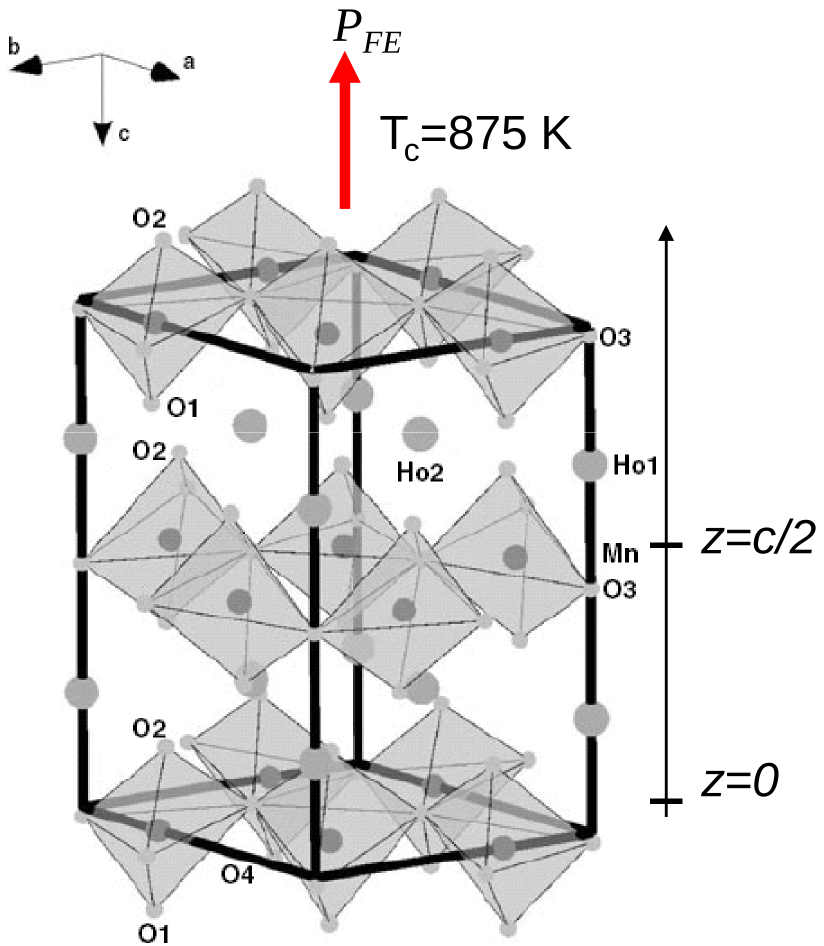
Magnetoelectricity in hexagonal $HoMnO_3$

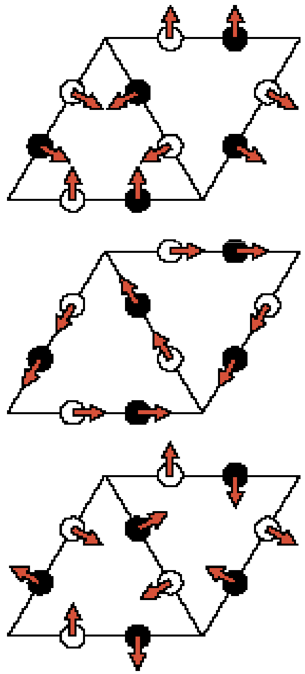
Ferroelectric



Litvinchuk et al., JPCM **16** 809 (2004)

Triangular antiferromagnet, T_N=72 K

Mn ions: ● *z=c/2* ○ *z=0*



40 K < T < 72 K

T_R T_N

5 K < T < 40 K

T_{Ho} T_R

T < 5 K

T_{Ho}

Vajk et al., PRL **94** 87601 (2005)

Magnetoelectricity in hexagonal $HoMnO_3$

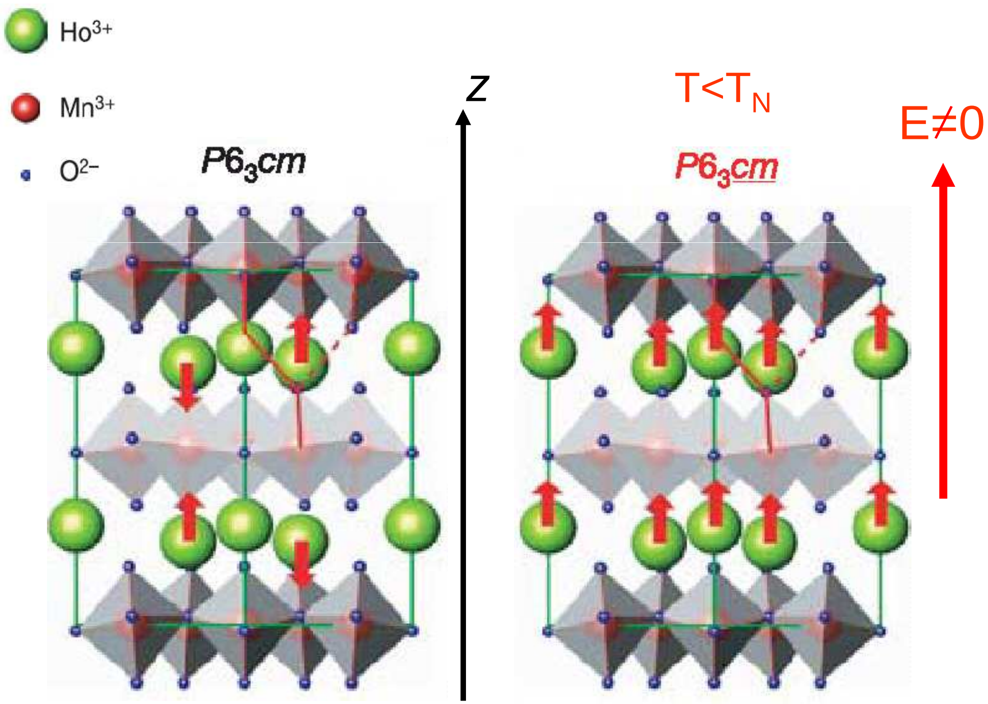
Magnetism of Ho^{3+} (S=2, L=6, J=8) ions:

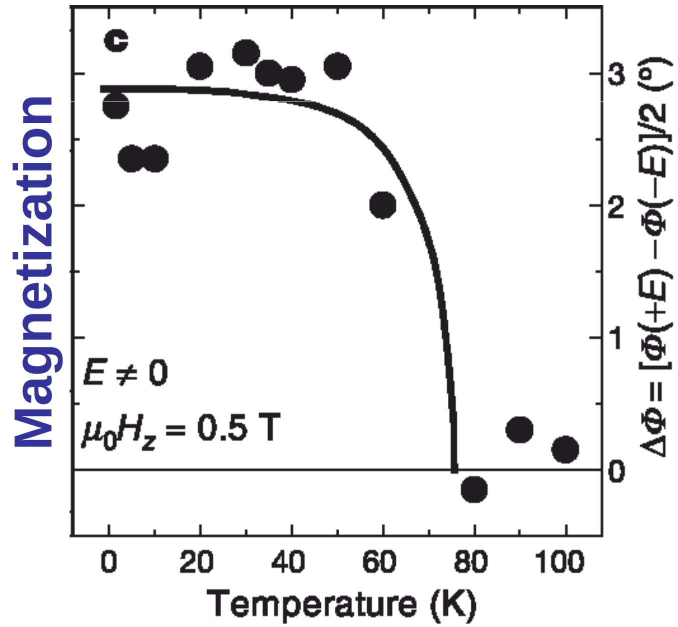
Two sites: Ho(1) C_{3v}

Ho(2) C_3 – ordered for $T<T_{Ho}$=5 K

Applied electric field induces magnetization of Ho ions:

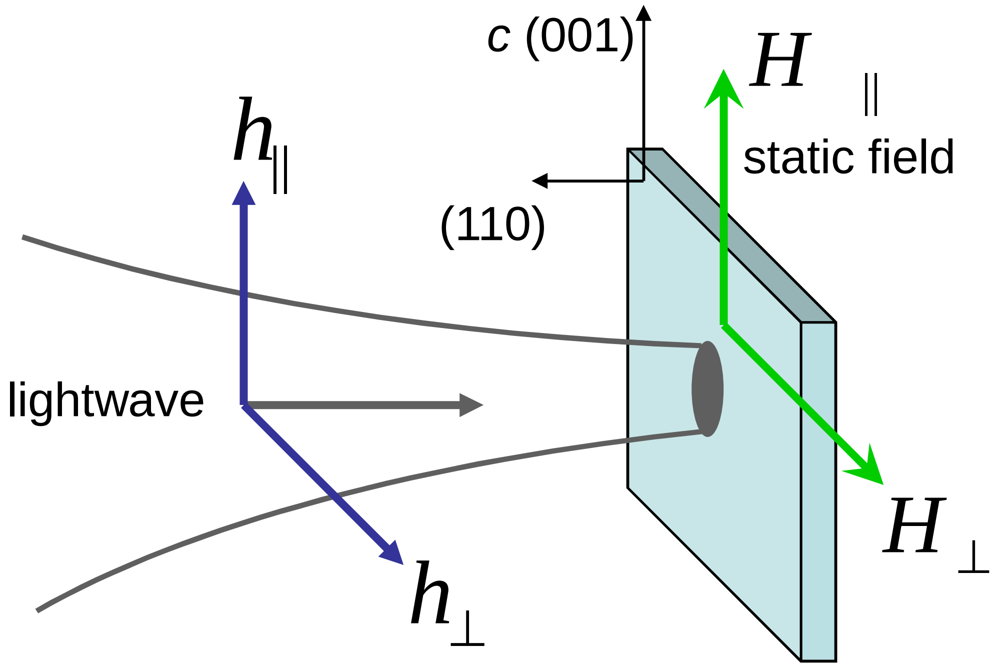
$$H_{int}=\Sigma S^{Ho} \Lambda S^{Mn}$$



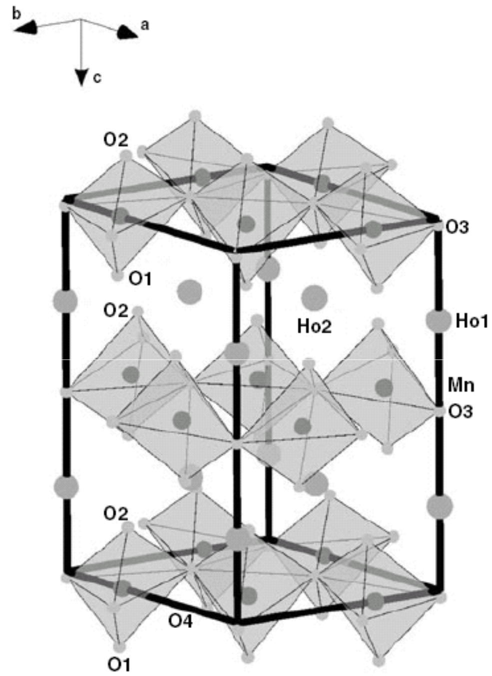


Lottermoser et al., Nature **430** 541 (2004)

Far-infrared study of magnetic excitations

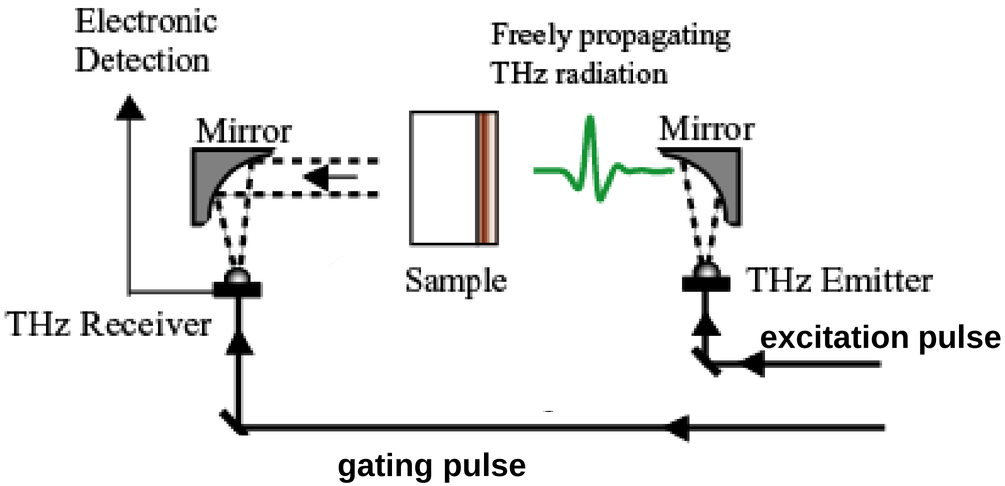


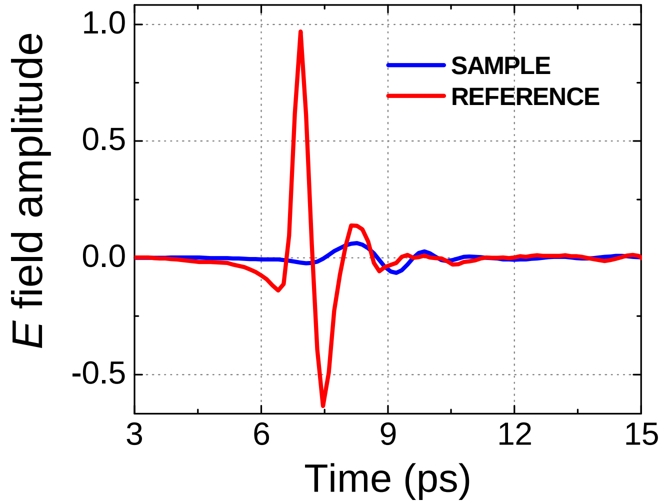
$HoMnO_3$



Far-infrared study of magnetic excitations

1 THz → 300 μm → 0.004 eV → 33cm^{-1} → 47 K

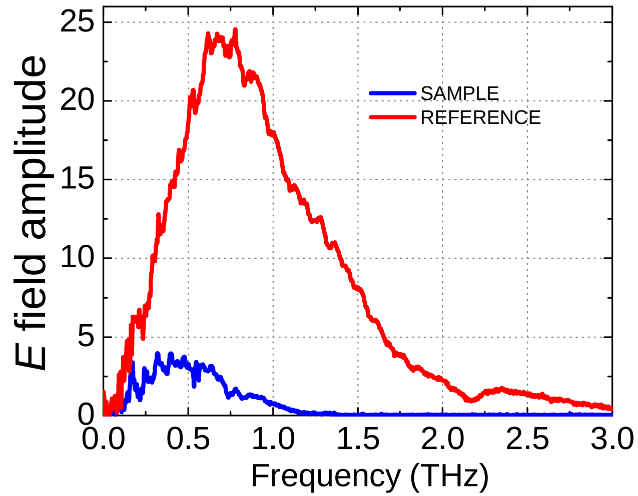




- Broadband
- Measurement of electric field *E*

(1) Photoconductive antennas

(2) Electro-optic crystals



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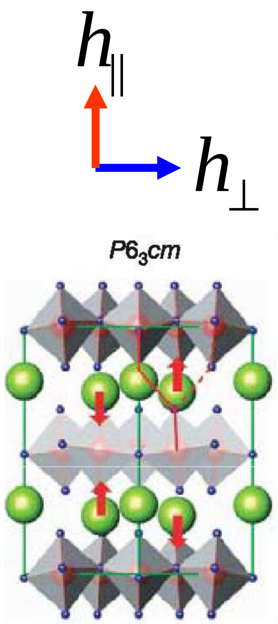
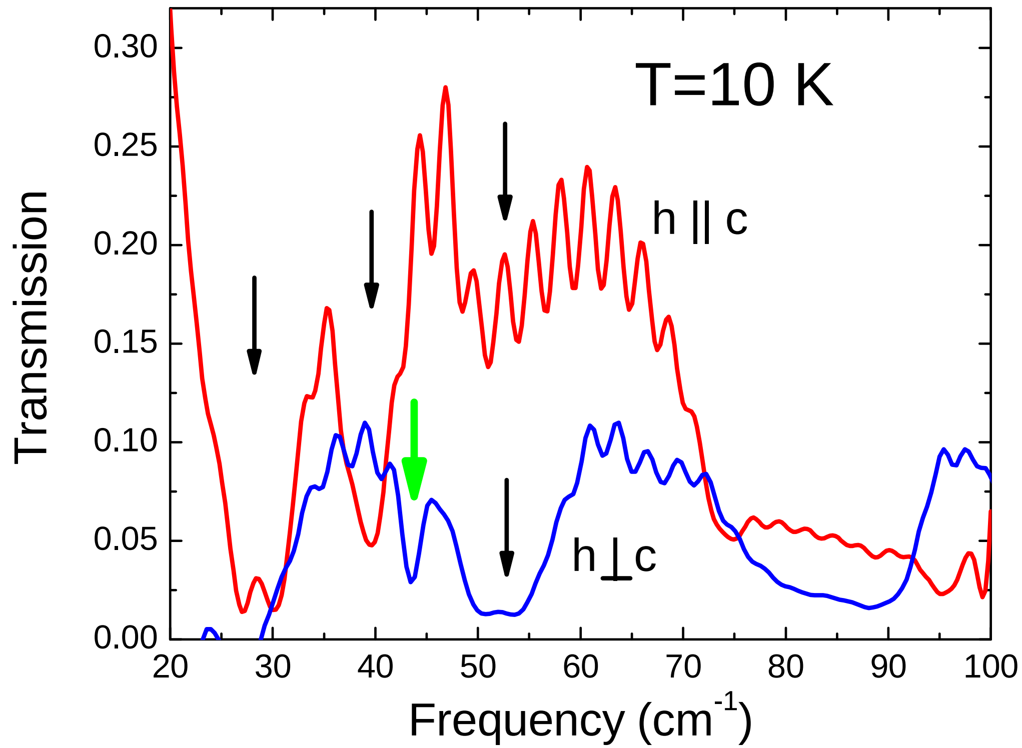
Magnetic excitations in $HoMnO_3$





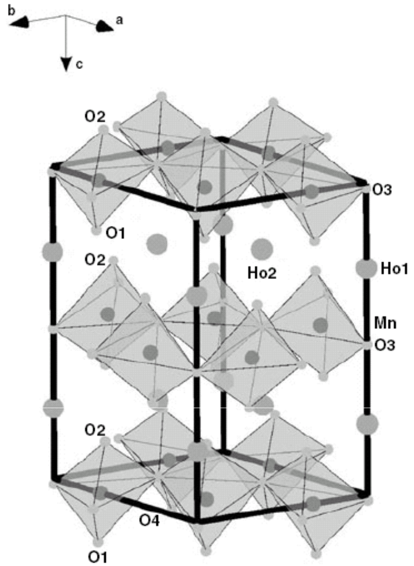
→ Crystal field excitations of Ho ions

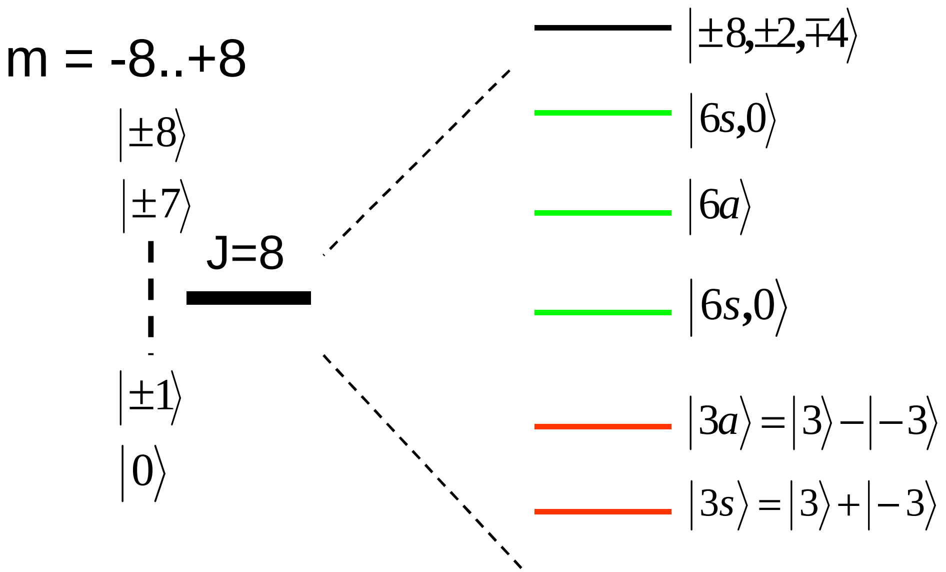
→ Antiferromagnetic resonance (AFMR) of Mn ions

Neutron scattering: Vajk et al., Phys. Rev. Lett. **94** 87601 (2005)

Crystal field splitting of Ho^{3+} ground state

Two Ho^{3+} (S=2, L=6, J=8) cites with C_3 and C_{3v} point symmetries

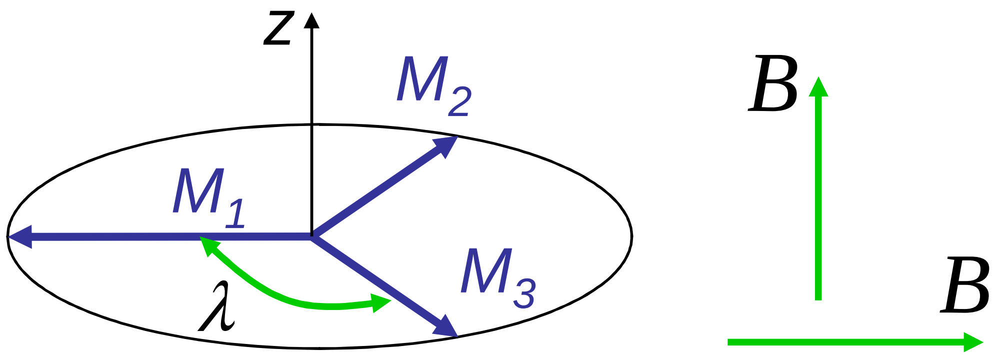




The electrostatic environment of Ho ions determines the crystal field splitting

Abragam and Bleaney, Electron paramagnetic resonance of transition ions, Clarendon press, 1970

Magnetic resonance of Mn ions

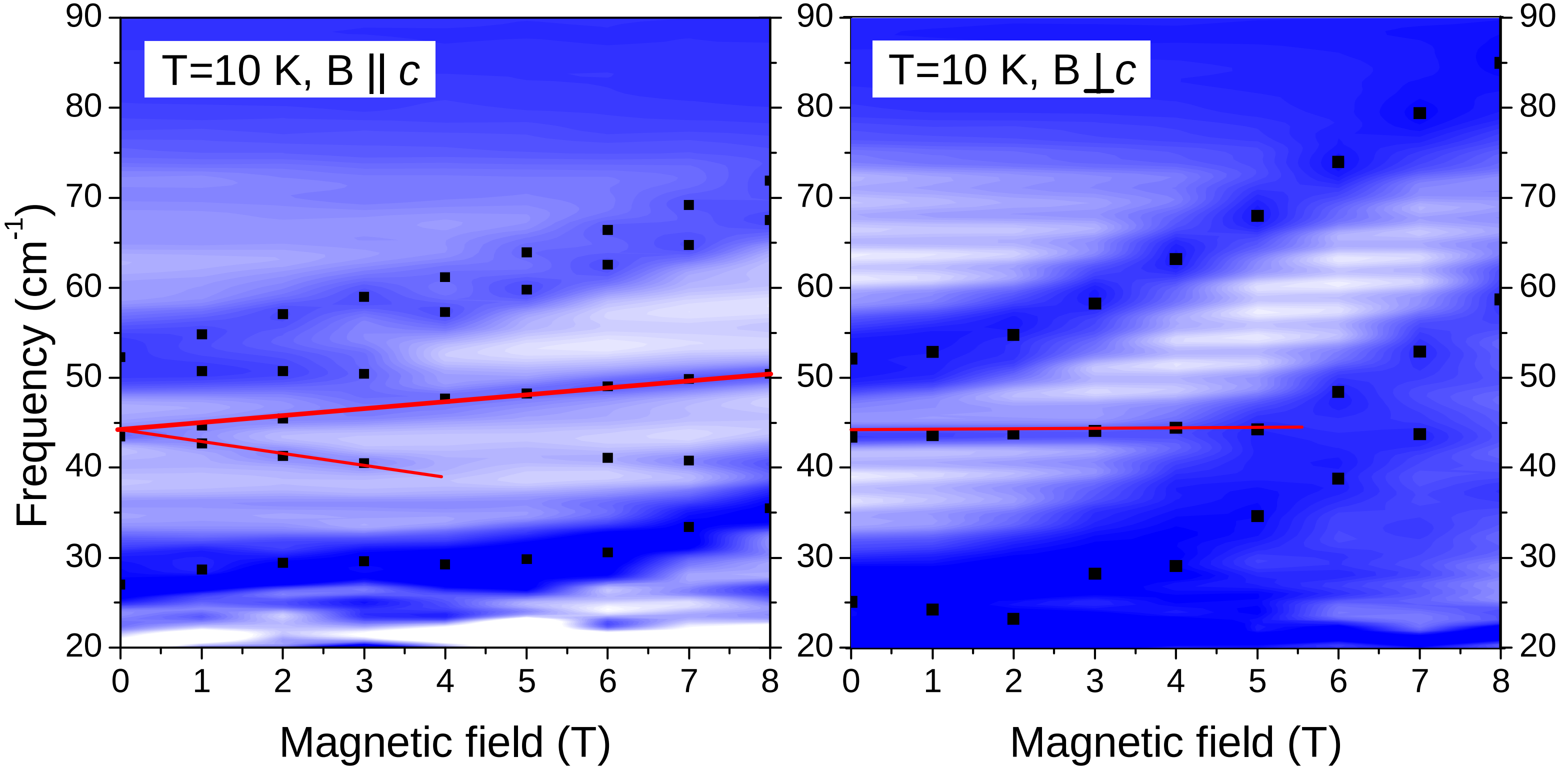


Classical treatment

$$F = \lambda \Sigma M_i \cdot M_j + K \sum (M_i^z)^2 - B \Sigma M_i^z$$

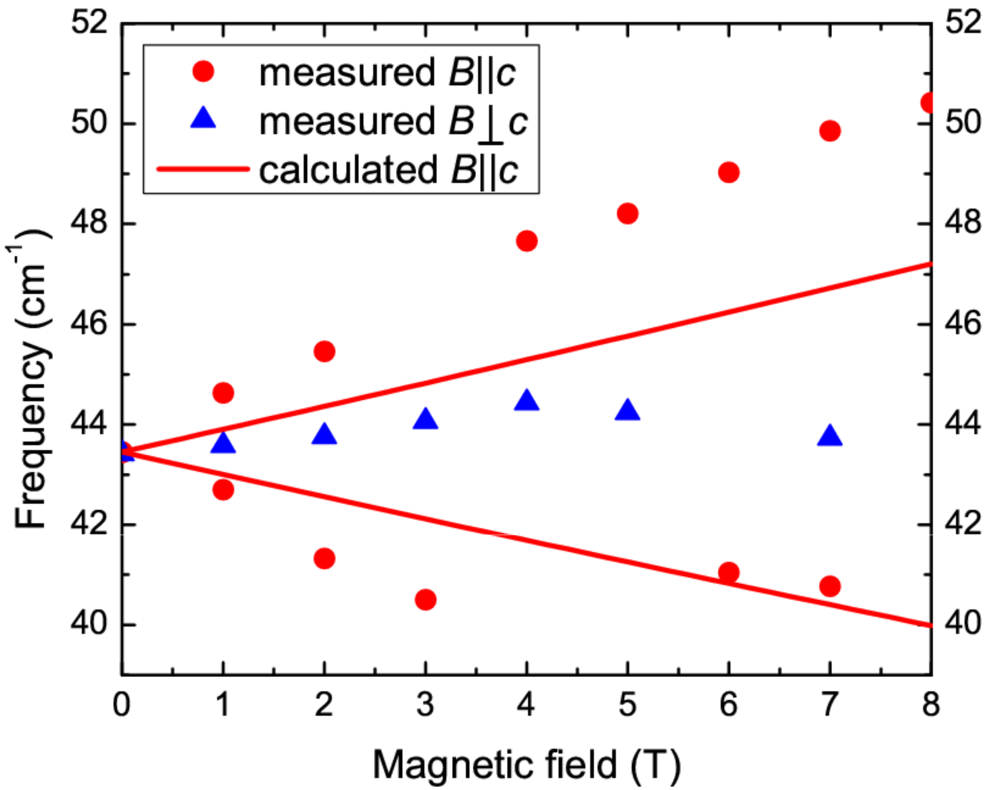
Interaction parameters measured by neutron scattering in zero field by Vajk et al., Phys. Rev. Lett. 94 87601 (2005)

Magnetic resonance of Mn ions



D. Talbayev et al., Phys. Rev. Lett. **101** 247601 (2008)

Magnetic resonance of Mn ions



$$\omega_{\pm}^2 = \frac{ab}{2} - \frac{b(a-b)}{2(a+b)^2}B^2 \pm \frac{bB}{2(a+b)^2} \times \sqrt{B^2 b(b-2a) + 2ab(a+b)^2}$$

$$a = 2H_d, \quad b = 3H_{\text{ex}}$$

$$H_{ex} = \lambda M_0$$

$$H_d = KM_0$$

Palme et al., Solid State Comm. **76** 873 (1990)

Why the discrepancy?

D. Talbayev et al., Phys. Rev. Lett. **101** 247601 (2008)

Exchange coupling between Ho and Mn ions

$YMnO_3$: similar material – hexagonal lattice, ferroelectric, triangular antiferromagnet … but Y ions are not magnetic

…and the calculation works!!

Penney et al., J. Appl. Phys. **40** 1234 (1969)   Sato et al., Phys. Rev. B **68** 014432 (2003)

Discrepancy in $HoMnO_3$ is due to Ho-Mn (HM) exchange interaction:

$$H_{ex}^{HM} = \tilde{J}_{ij} S_{iz}^{Ho} S_{jz}^{Mn} = J_z \frac{\chi B}{g_z \mu_B} S_z^{Mn}$$

$$F_{HM} = \frac{6 J_z \chi}{g_z g \mu_B^2} B \sum M_{iz}^{Mn} = \lambda_{HM} B \sum M_{iz}^{Mn} \qquad \boxed{\lambda_{HM} = -1}$$

Effective magnetic field acting on Mn ions!

D. Talbayev et al., Phys. Rev. Lett. **101** 247601 (2008)

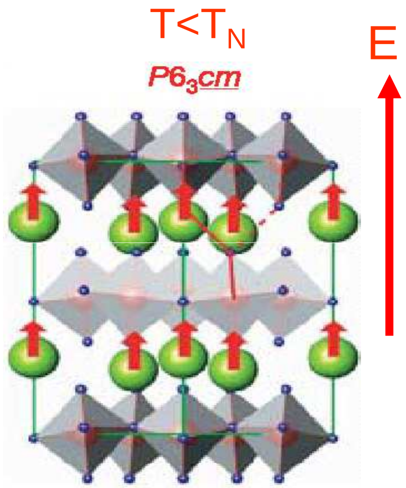
Magnetoelectricity in $HoMnO_3$

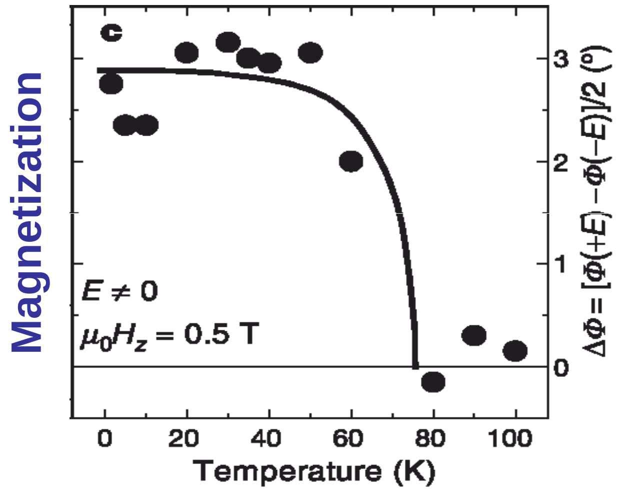
Ferromagnetic exchange between Ho and Mn:

$$H_{ex}^{HM} = \tilde{J}_{ij}^{z} S_{iz}^{Ho} S_{jz}^{Mn} \qquad \tilde{J}_{ij}^{z} < 0$$

Same interaction responsible for the magnetoelectricity:

Lottermoser et al.,
Nature **430** 541 (2004)





Magnons allow the determination of microscopic details of magnetoelectric interaction.

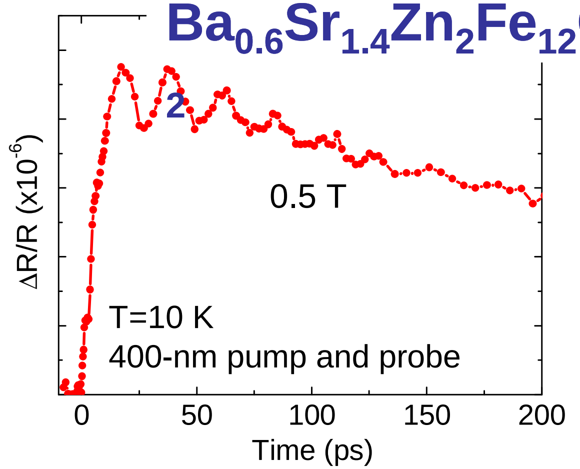
In $HoMnO_3$, it is the Ho-Mn magnetic exchange coupling.

Summary

(i) **Magnetic and lattice excitations (magnons and phonons) play a fundamental role in the quest for understanding and exploiting magnetoelectric materials**

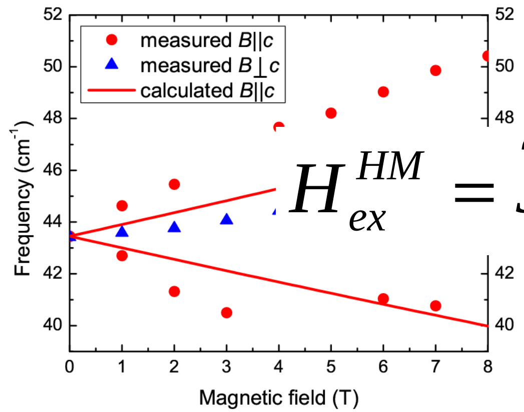
(ii) **Detection of magnetic motion and magnetic state via the modulation of reflectance**

$Ba_{0.6}Sr_{1.4}Zn_2Fe_{12}O_{22}$



(iii) **Properties of magnons and phonons help reveal the microscopic details of magnetoelectric interaction**

$HoMnO_3$



$$H_{ex}^{HM} = \tilde{J}_{ij}^{z} S_{iz}^{Ho} S_{jz}^{Mn}$$