UC SANTA BARBARA
Kavli Institute for
Theoretical Physics

Multipolar excitations and their magnetoelectric characters in åkermanites

Correlated Systems with Multicomponent Local Hilbert Spaces

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Nov 17 2020

Magnetoelectric effect

coupling between magnetic and electric degrees of freedom

when inversion symmetry is broken

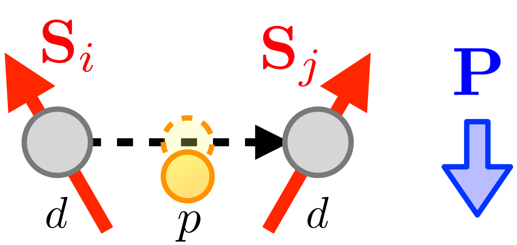
$\mathcal{H} \propto PS^2$ is allowed

inversion	time reversal
$\mathbf{P} \rightarrow -\mathbf{P}$	$\mathbf{P} \rightarrow \mathbf{P}$
$\mathbf{S} \rightarrow \mathbf{S}$	$\mathbf{S} \rightarrow -\mathbf{S}$

microscopic mechanisms for spin induced P

Katsura-Nagaosa-Balatsky **spin-current** mechanism for non collinear magnets:

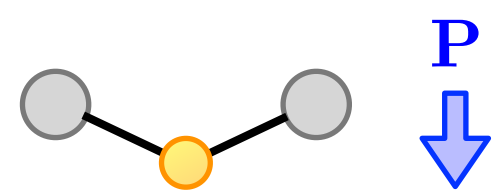
$$\mathbf{P} \propto \mathbf{e}_{ij} \times (\mathbf{S}_i \times \mathbf{S}_j)$$



magnetostrsiction in collinear magnets:

symmetric exchange interaction

$$J(e, \vartheta)\mathbf{S}_i \cdot \mathbf{S}_j$$



Magnetoelectric effect

coupling between magnetic and electric degrees of freedom

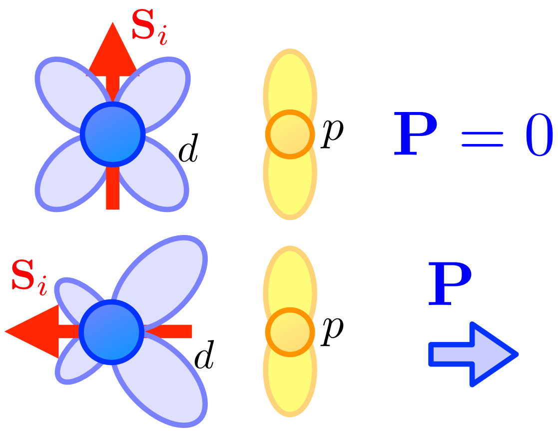
when inversion symmetry is broken

$\mathcal{H} \propto PS^2$ is allowed

	inversion	time reversal
	$\mathbf{P} \to -\mathbf{P}$	$\mathbf{P} \to \mathbf{P}$
	$\mathbf{S} \to \mathbf{S}$	$\mathbf{S} \to -\mathbf{S}$

microscopic mechanisms for spin induced P

spin-dependent metal-ligand **(p-d) hybridization**



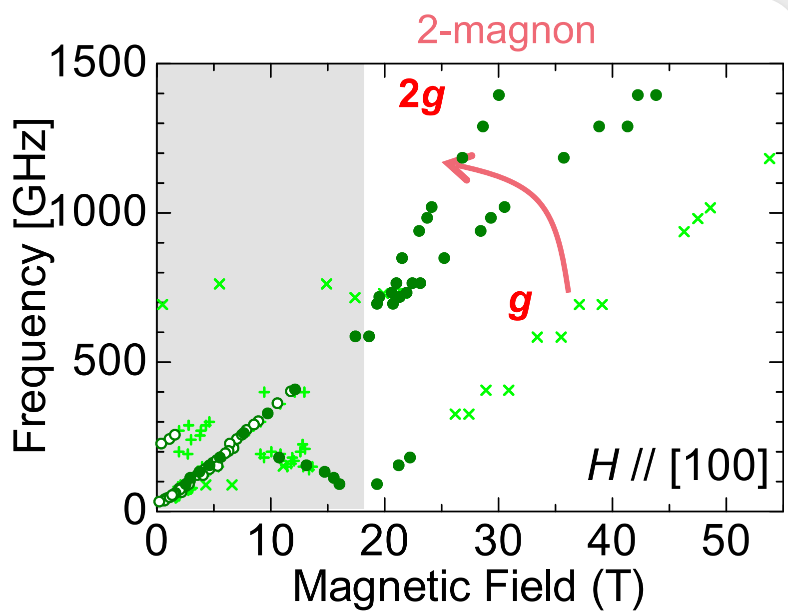
$$\begin{pmatrix} P^x \\ P^y \\ P^z \end{pmatrix} \propto \begin{pmatrix} S^y S^z \\ S^z S^x \\ S^x S^y \end{pmatrix}$$

Quadrupolar operators (rank 2)

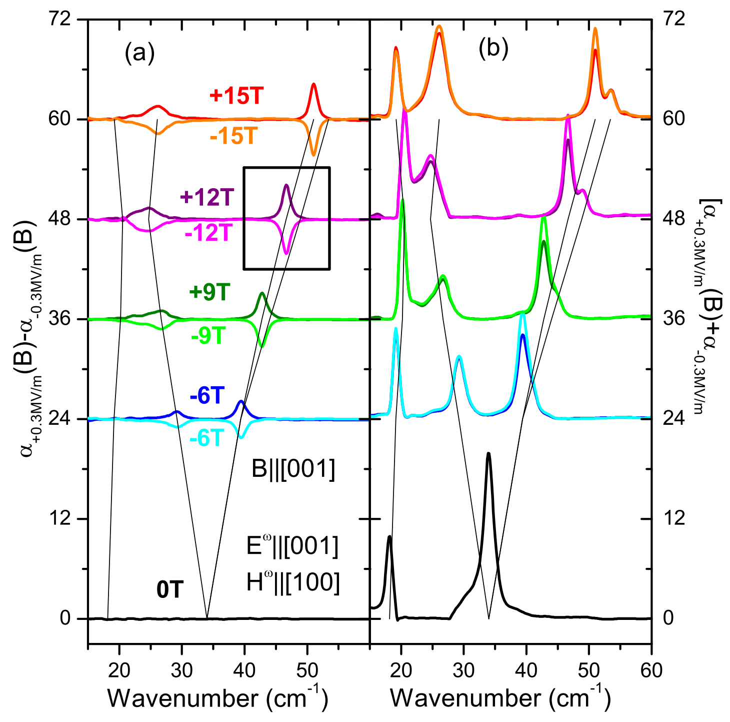
Jia et al PRB **74**, 224444 (2006)
Jia et al PRB **76**, 144424 (2007)
T. Arima, JPSJ **76**, 073702 (2007)

Detection of quadrupolar waves

Outline



Two magnon spin-quadrupolar excitations and their detection in the high-field excitations of $\mathbf{Sr_2CoGe_2O_7}$



Directional dichroism in the AFM phase of $\mathbf{Ba_2CoGe_2O_7}$

Special thanks to

Theory

Karlo Penc

Wigner RCP

$Ba_2CoGe_2O_7$

Sándor Bordács

Budapest University of Technology

István Kézsmárki

University of Augsburg

Jakub Vít

Thomas Rõõm

Urmas Nagel

Dávid Szaller

Titusz Fehér

Yoshinori Tokura

$Sr_2CoGe_2O_7$

Masayuki Hagiwara

Osaka University

Mitsuru Akaki

Kobe University

Daichi Yoshizawa

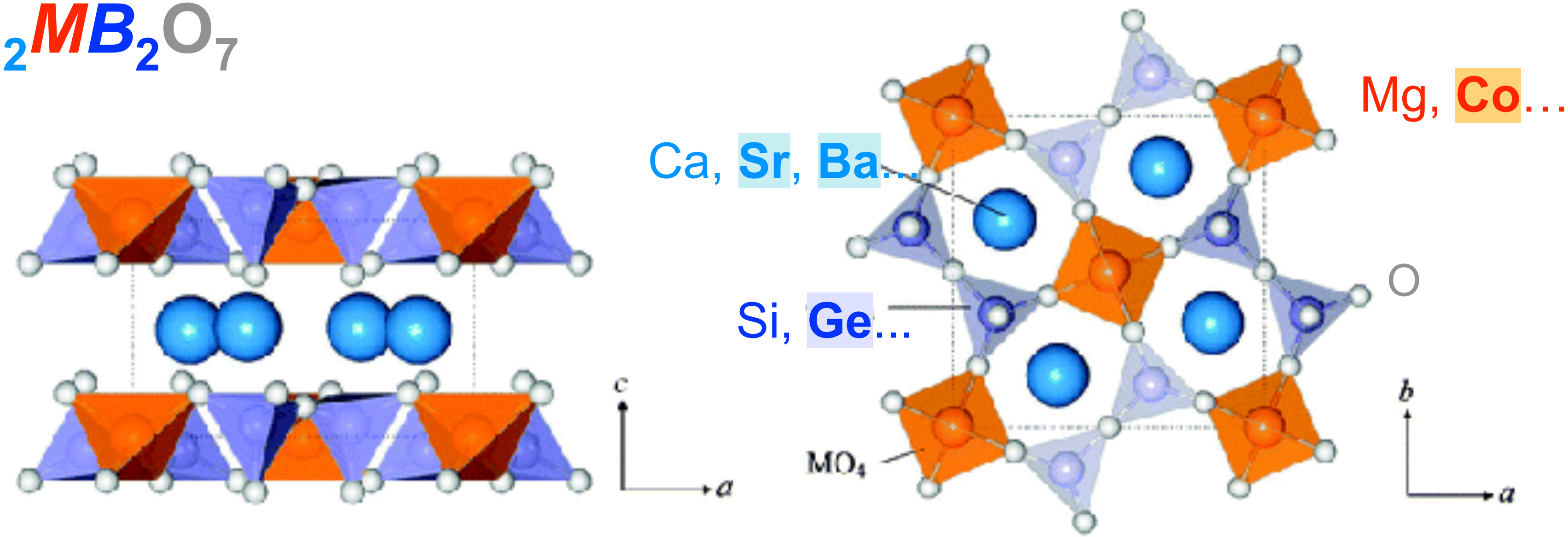
Akira Okutani

Takanori Kida

Yasuo Narumi

Åkermanite structure

$A_2MB_2O_7$



T Endo et al Inorg. Chem. **51**, 3572 (2012)

Tetragonal $P\bar{4}2_1m$

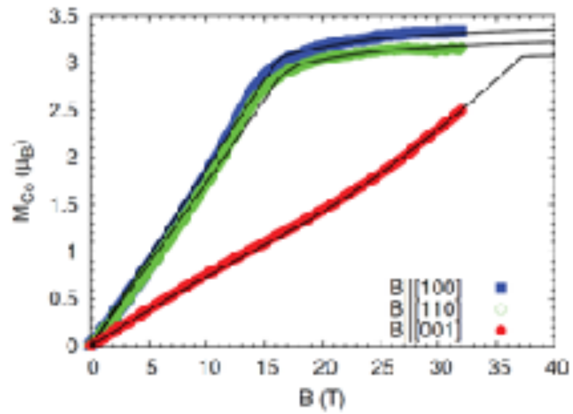
åkermanite ($Ca_2MgSi_2O_7$) structure + magnetic ion

p-d hybridization → spin-induced electric polarization

Magnetic properties

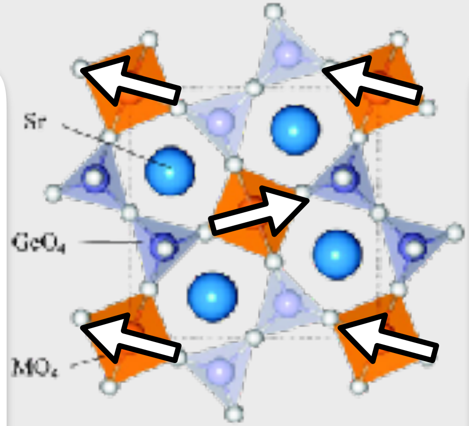
$Ba_2CoGe_2O_7$

AFM ordering below T_N=6.7 K



V. Hutanu et al., PRB **89**, 064403 (2014)

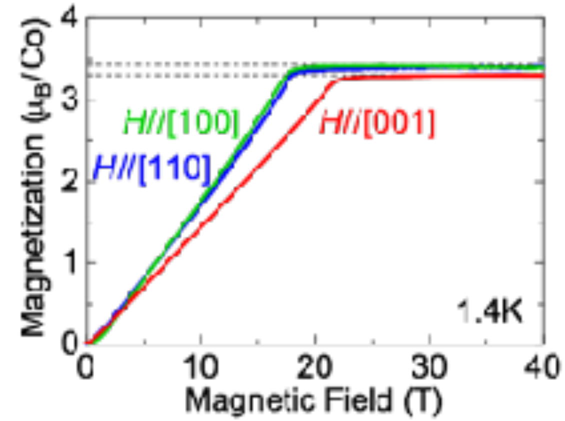
magnetic structure



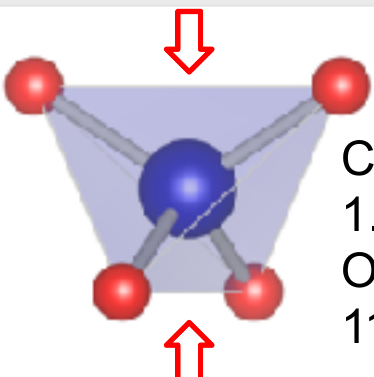
T. Endo et al., Inorg. Chem. **51**, 3572 (2012).

$Sr_2CoGe_2O_7$

AFM ordering below T_N=6.5 K



M. Akaki et al PRB **96**, 214406 (2017)

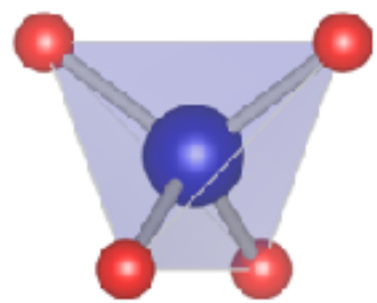


Co-O length 1.98Å
O-Co-O angle 116.7°

$Ba_2CoGe_2O_7$
single crystal neutron, @RT
A. Sazonov *et al.*, J. Appl. Cryst. **49**, 556 (2016).

the distortion of CoO_4

magnetic anisotropy

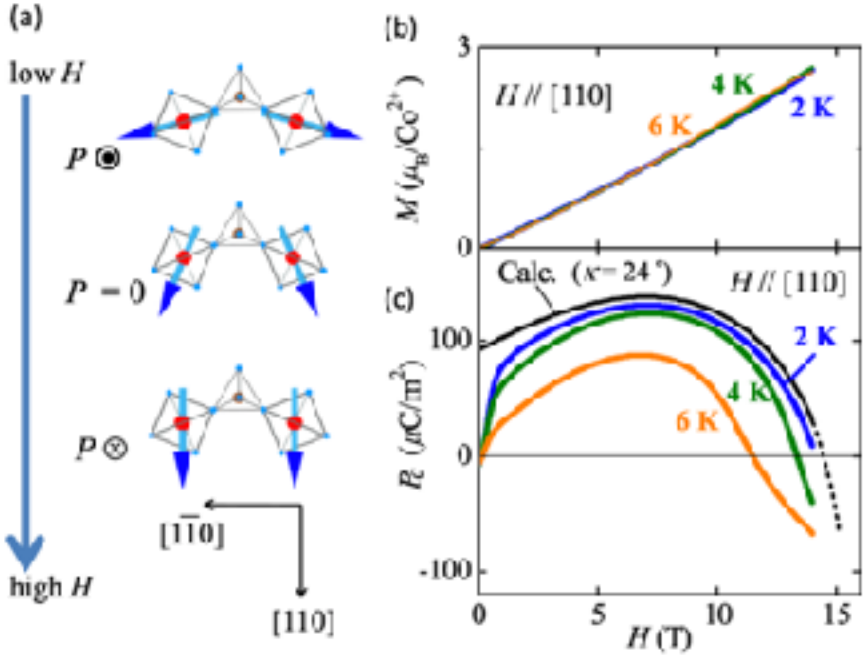


Co-O length 1.96Å
O-Co-O angle 110.7°

$Sr_2CoGe_2O_7$ powder neutron, 2.5K
T. Endo *et al.*, Inog Chem. **51**, 3572 (2012).

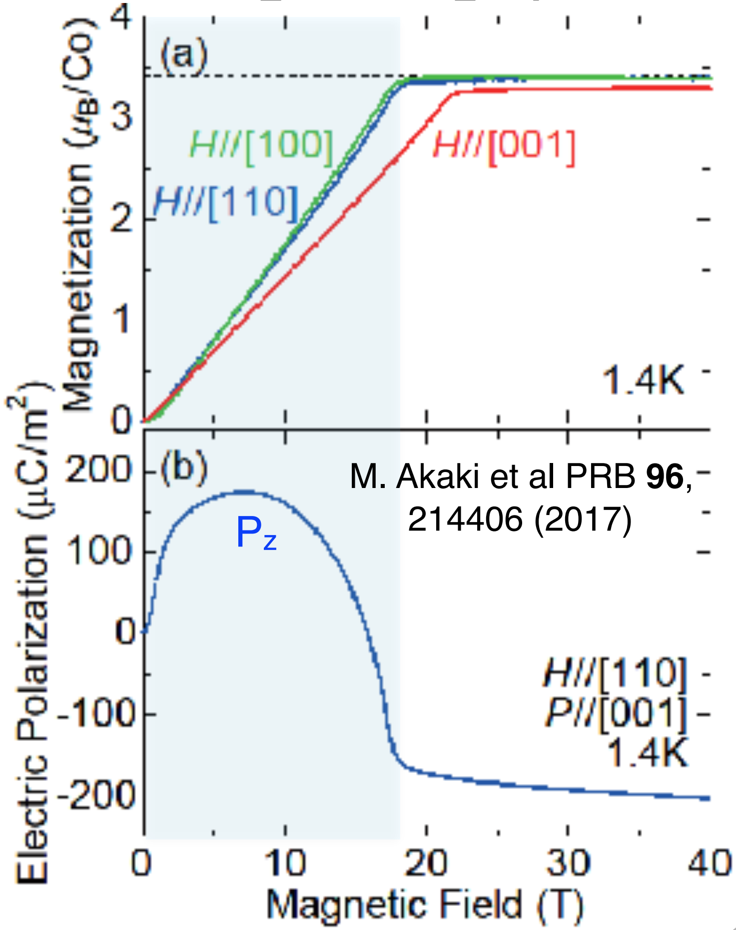
Spin induced polarization

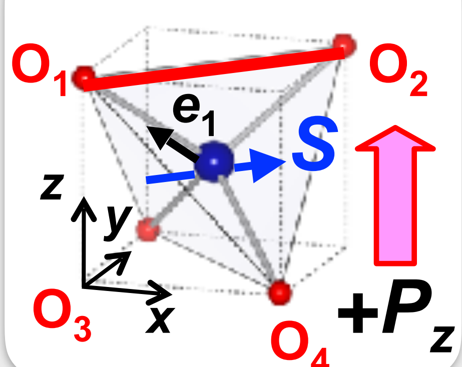
$Ba_2CoGe_2O_7$

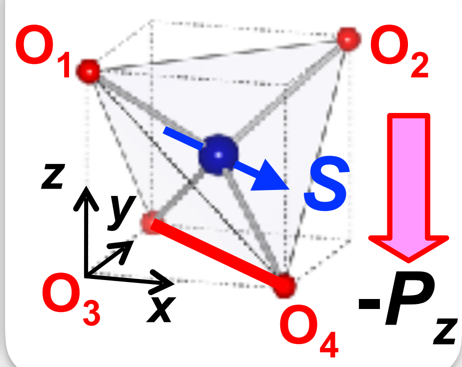


H. Murakawa et al PRL **105**, 137202 (2010)

$Sr_2CoGe_2O_7$







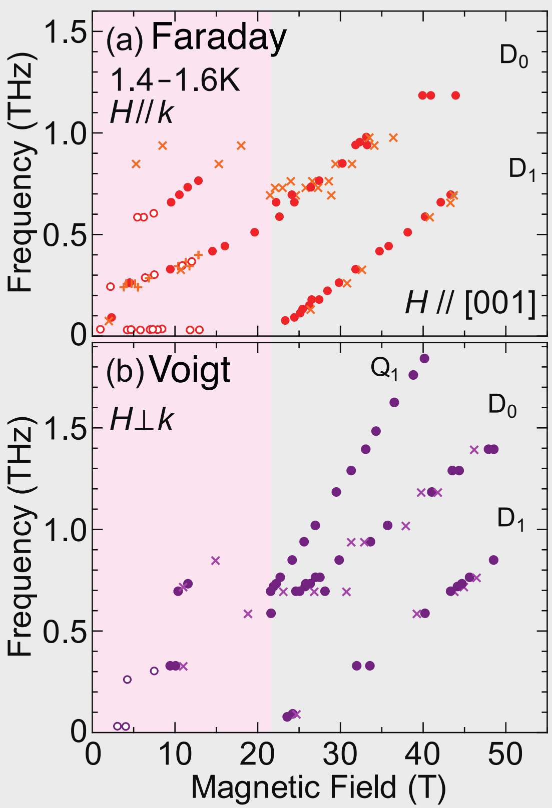
$$P \propto \sum_{i=1}^{4} (S \cdot e_i)^2 e_i$$

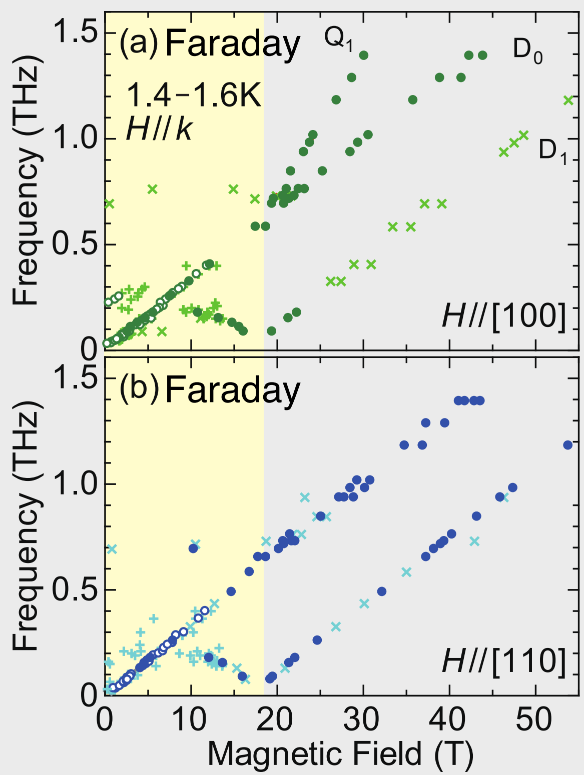
T. Arima, J. Phys. Soc. Jpn. **76**, 073702 (2007).

Dynamic properties from ESR

$Sr_2CoGe_2O_7$

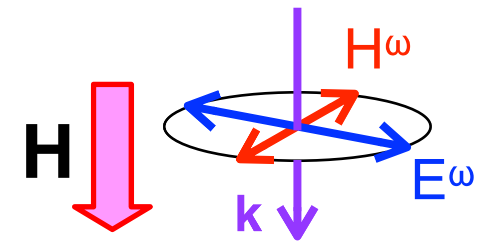
data collected in Faraday and Voigt geometries in fields along [001], [100], and [110]



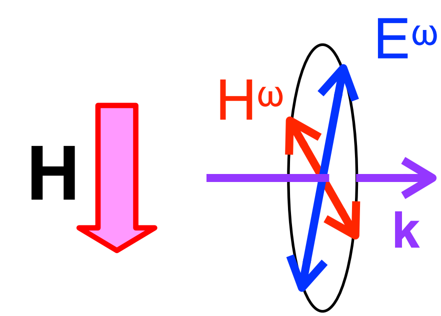


○+ : static field by SCM ●×: pulsed magnetic fields

Faraday H^ω and E^ω ⊥ to **H** can excite

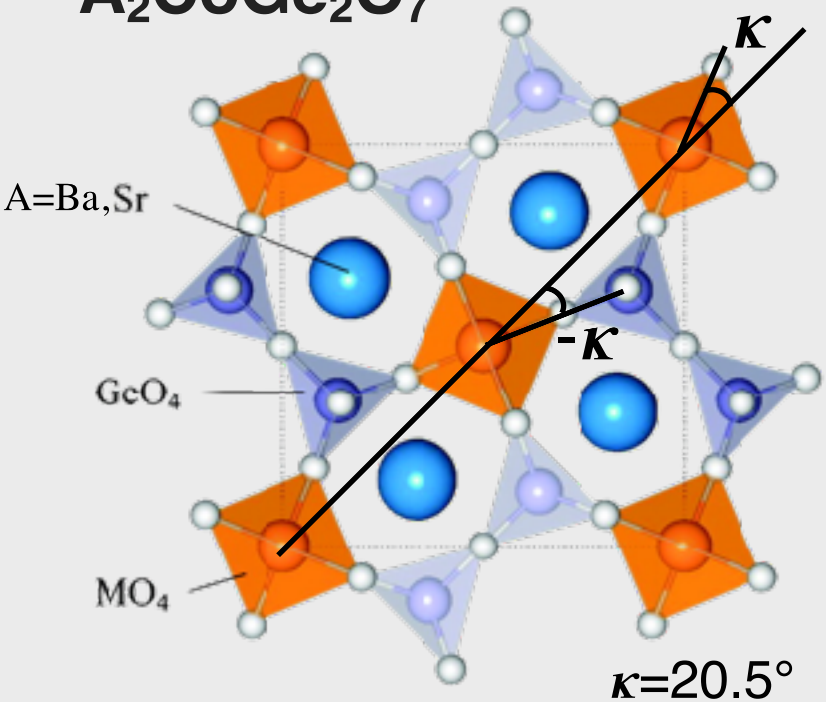


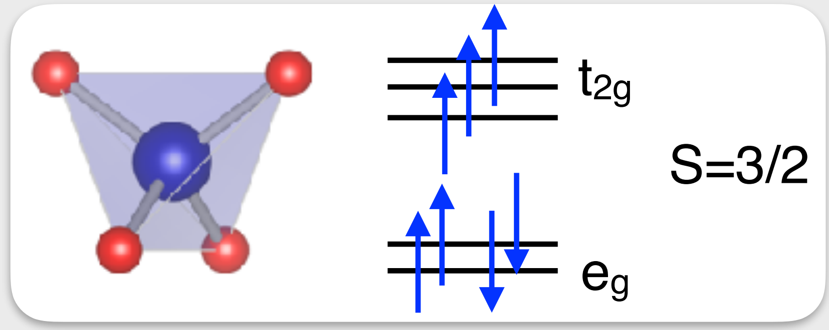
Voigt H^ω and E^ω ⊥ or ∥ to **H** can excite



Model Hamiltonian

$A_2CoGe_2O_7$





large local Hilbert space

$$\left|\tfrac{3}{2}\right\rangle = |\Uparrow\rangle$$

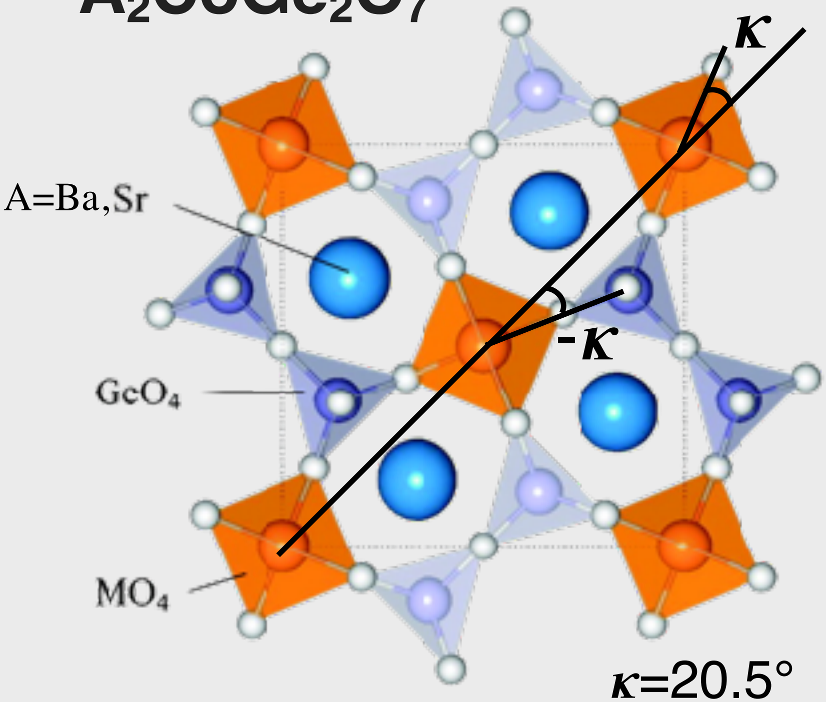
$$S^- \left|\tfrac{3}{2}\right\rangle = \left|\tfrac{1}{2}\right\rangle = |\uparrow\rangle \quad \text{dipole}$$

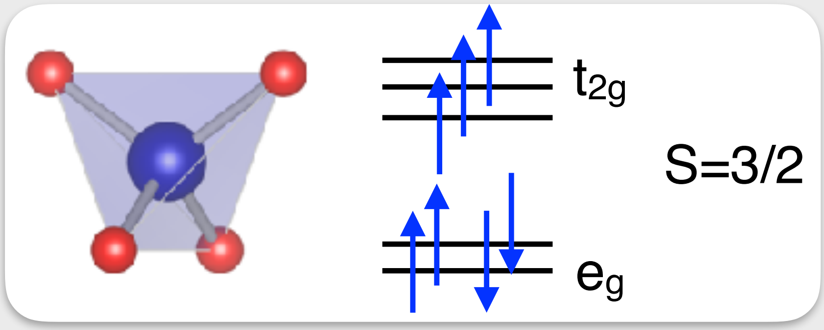
$$S^- S^- \left|\tfrac{3}{2}\right\rangle = \left|-\tfrac{1}{2}\right\rangle = |\downarrow\rangle \quad \text{quadrupole}$$

$$S^- S^- S^- \left|\tfrac{3}{2}\right\rangle = \left|-\tfrac{3}{2}\right\rangle = |\Downarrow\rangle \quad \text{octupole}$$

Model Hamiltonian

$A_2CoGe_2O_7$





multicomponent local Hilbert space

$$P_j^x \propto -\cos 2\kappa\, \overline{S_j^x S_j^z} - (-1)^j \sin 2\kappa\, \overline{S_j^y S_j^z}\ ,$$

$$P_j^y \propto \cos 2\kappa\, \overline{S_j^y S_j^z} - (-1)^j \sin 2\kappa\, \overline{S_j^x S_j^z}\ ,$$

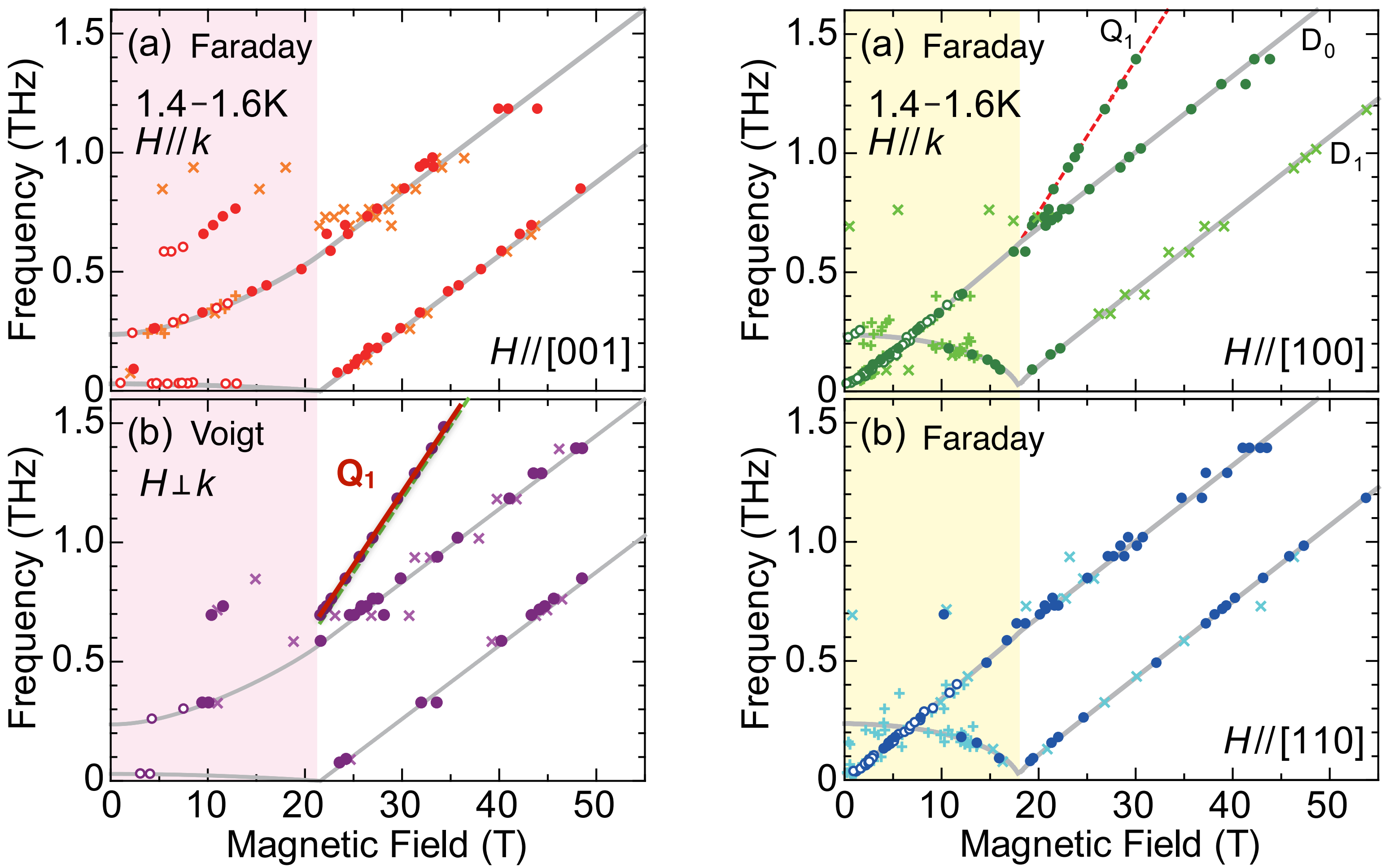
$$P_j^z \propto \cos 2\kappa \left[(\hat{S}_j^y)^2 - (\hat{S}_j^x)^2\right] - (-1)^j \sin 2\kappa\, \overline{\hat{S}_j^x \hat{S}_j^y}$$

T. Arima, J. Phys. Soc. Jpn. **76**, 073702 (2007).
H. Murakawa *et al.*, Phys. Rev. Lett. **105**, 172020 (2010).

$$\mathcal{H} = J \sum_{(i,j)} \left(\hat{S}_i^x \hat{S}_j^x + \hat{S}_i^y \hat{S}_j^y\right) + J_z \sum_{(i,j)} \hat{S}_i^z \hat{S}_j^z + \Lambda \sum_i (\hat{S}_i^z)^2 - \mu_B \sum_i \mathbf{h g_i S_i}$$

M. Akaki et al PRB **96**, 214406 (2017)

The full spectrum from multiboson calculation



Gray: multiboson spin wave

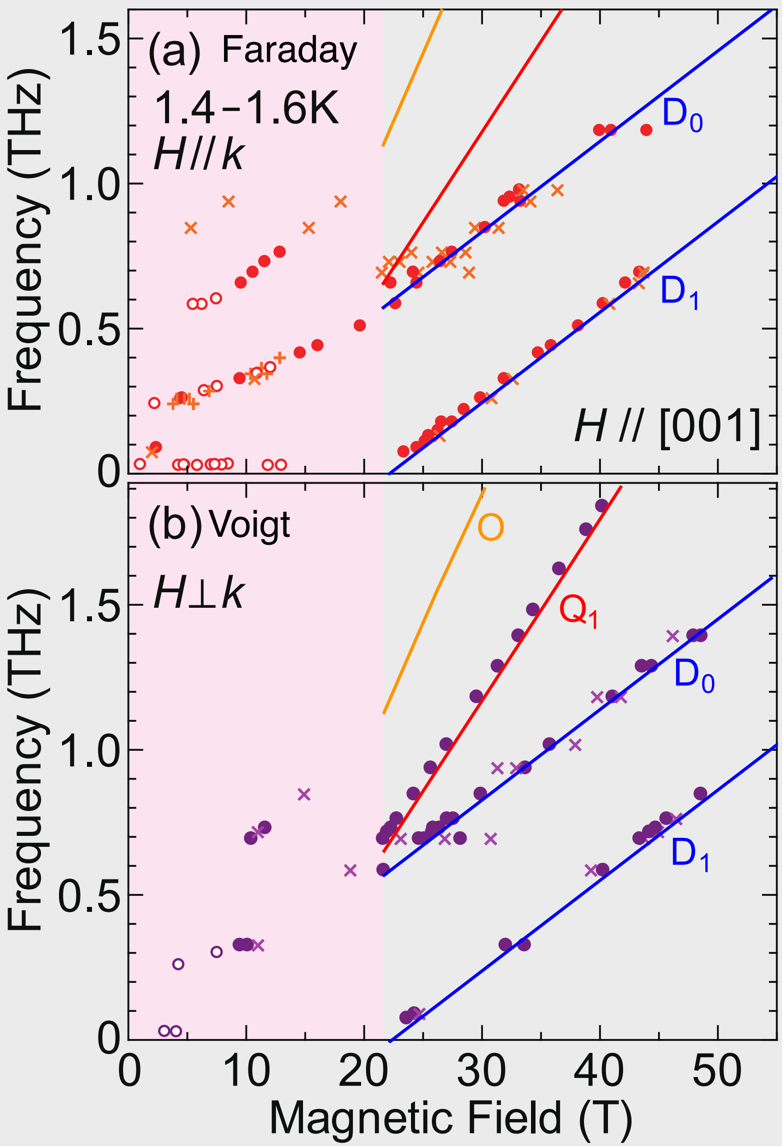
$$J = 49.1 \pm 0.8 \text{ GHz} = 2.36 \pm 0.04 \text{ K} ,$$

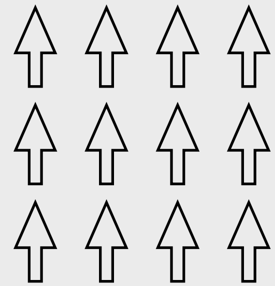
$$J_z = 45.0 \pm 1.1 \text{ GHz} = 2.16 \pm 0.05 \text{ K} ,$$

$$\Lambda = 49.7 \pm 4.2 \text{ GHz} = 2.39 \pm 0.20 \text{ K} .$$

M. Akaki et al PRB **96**, 214406 (2017)

Excitations in high field H ll [001]





Fully saturated state

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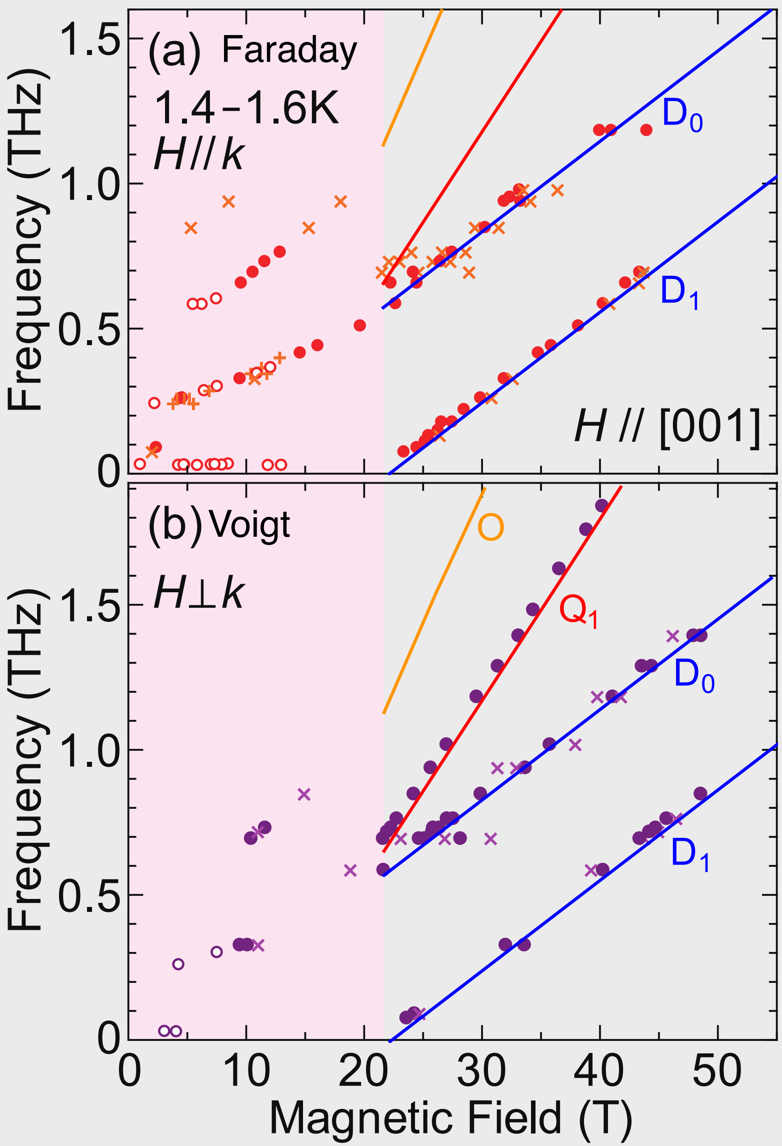
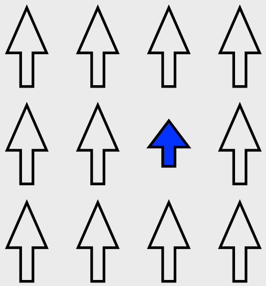
Excitations in high field H ll [001]





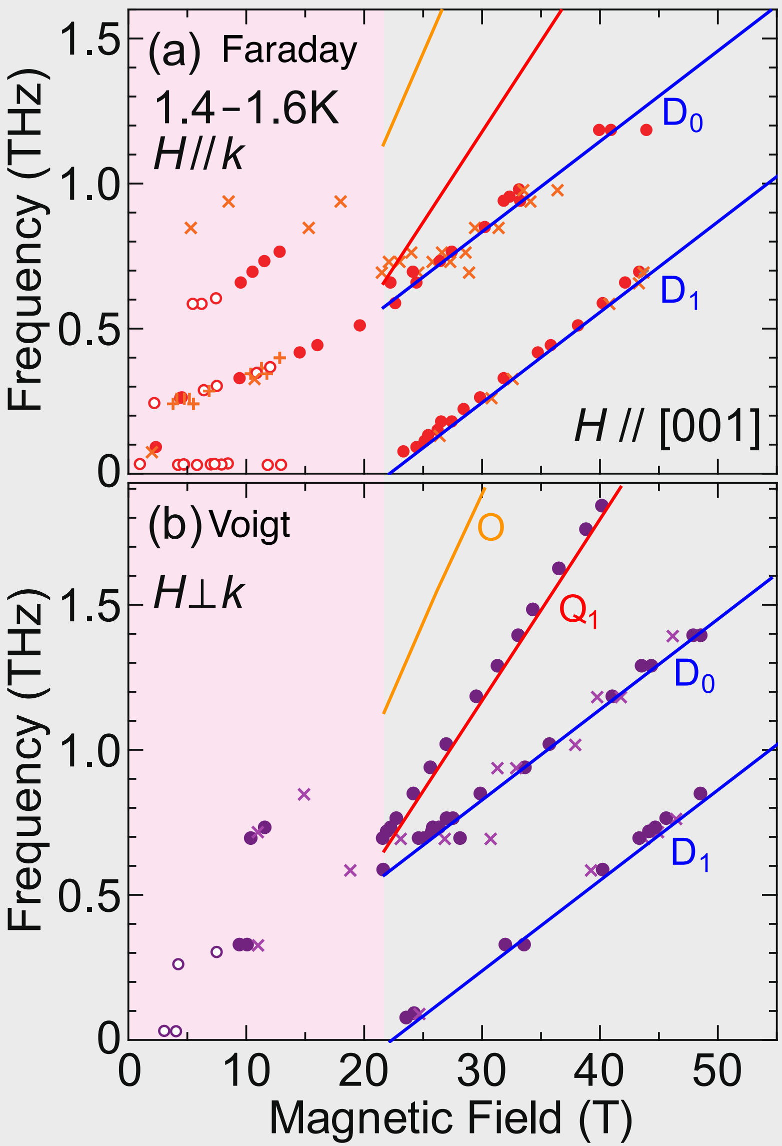
one-magnon dipolar transition

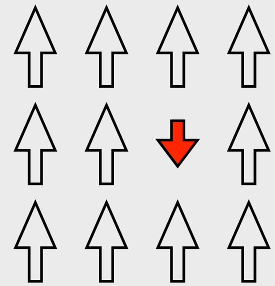
$$\omega_{D_0} = g_z h^z - 2\Lambda$$

$$\omega_{D_1} = g_z h^z - 2\Lambda - 12J$$

M. Akaki et al PRB **96**, 214406 (2017)

Excitations in high field H II [001]





one-magnon dipolar transition

$$\omega_{D_0} = g_z h^z - 2\Lambda$$

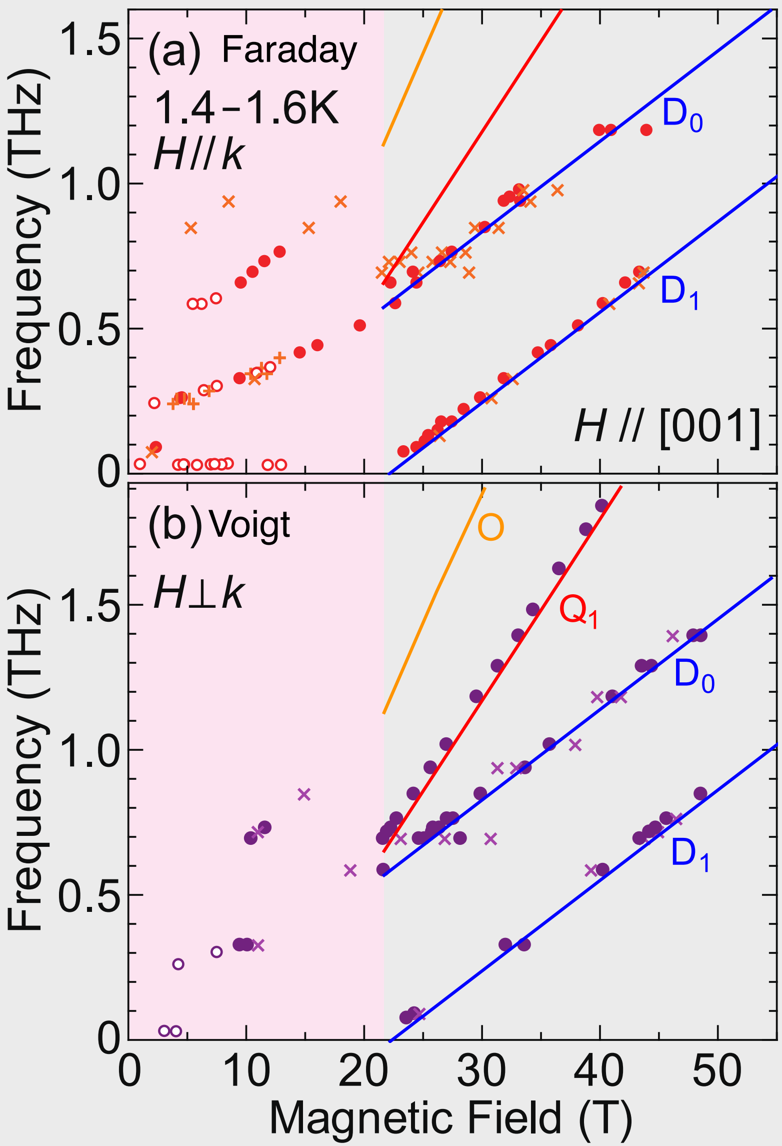
$$\omega_{D_1} = g_z h^z - 2\Lambda - 12J$$

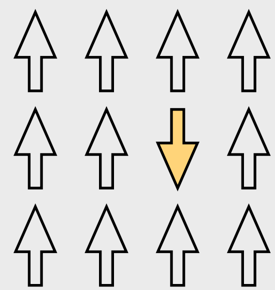
two-magnon quadrupolar transition

$$\omega_Q = 2g_z h^z - 12J - 2\Lambda$$

M. Akaki et al PRB **96**, 214406 (2017)

Excitations in high field H ll [001]





one-magnon dipolar transition

$$\omega_{D_0} = g_z h^z - 2\Lambda$$

$$\omega_{D_1} = g_z h^z - 2\Lambda - 12J$$

two-magnon quadrupolar transition

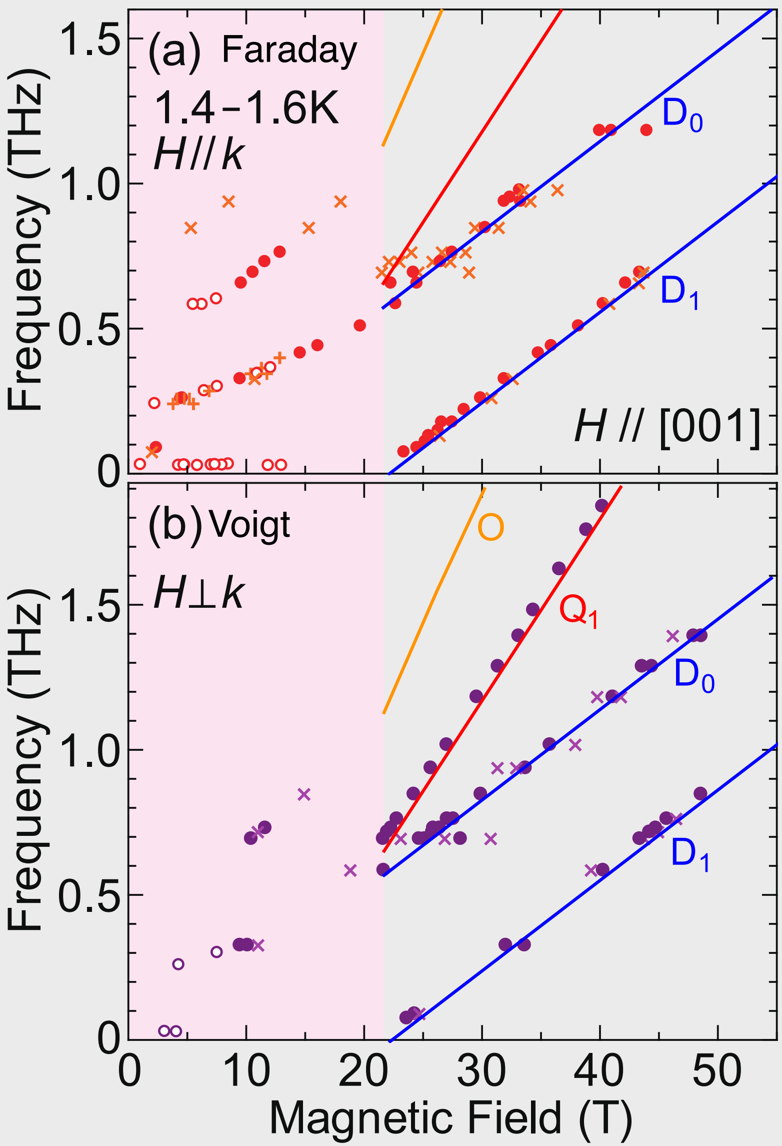
$$\omega_Q = 2g_z h^z - 12J - 2\Lambda$$

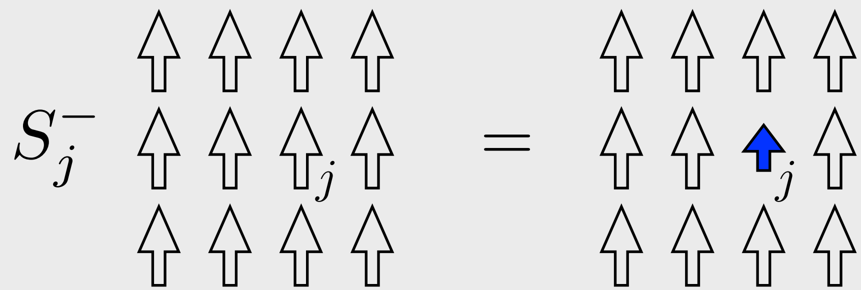
three-magnon octupolar transition

$$\omega_O = 3g_z h^z - 18J$$

M. Akaki et al PRB **96**, 214406 (2017)

One-magnon transition H ll [001]





the operators containing S_j^- are

S_j^x and S_j^y

P_j^x and $P_j^y \propto S_j^z S_j^x$ and $S_j^z S_j^y$

these are the perpendicular (H ll [001]) components, present in the Voigt & Faraday geometries

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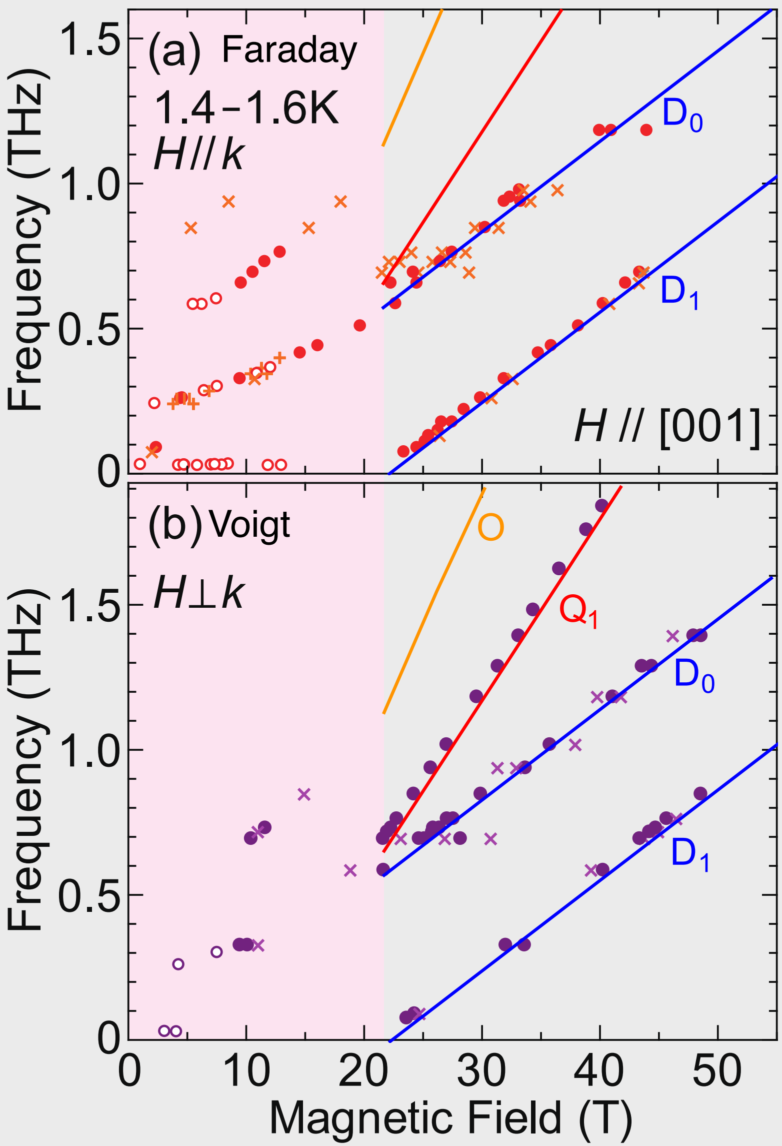
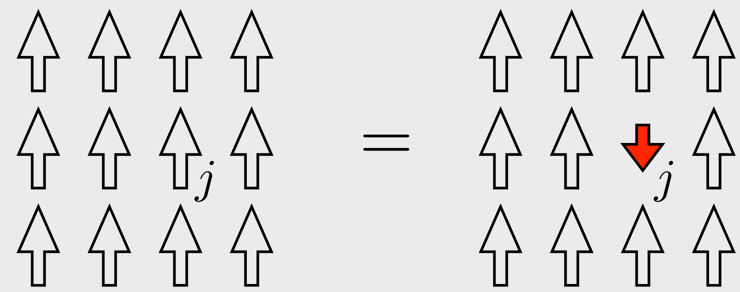
Two-magnon transition H ll [001]



$S_j^- S_j^-$ 

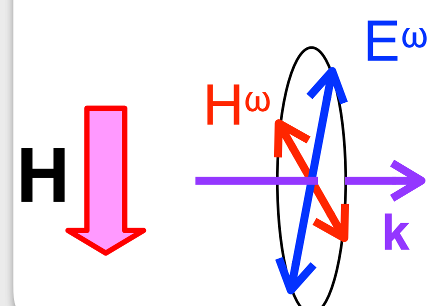
only operator containing $S_j^- S_j^-$ is

$$P_j^z \propto \cos 2\kappa \left[(\hat{S}_j^y)^2 - (\hat{S}_j^x)^2 \right] - (-1)^j \sin 2\kappa \, \overline{\hat{S}_j^x \hat{S}_j^y} \, ,$$

$$\mathrm{Im}\chi_{\alpha\alpha}^{ee}(\omega) \propto \sum_f |\langle f | P^\alpha | 0 \rangle|^2 \delta(\omega - \omega_f + \omega_0)$$

this is the parallel component, only present in the Voigt geometry

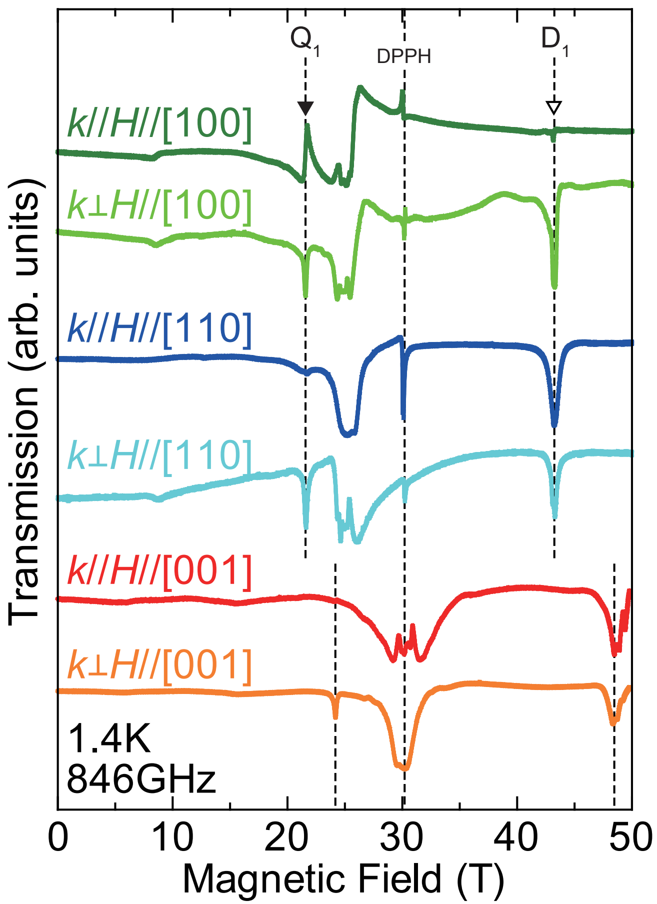
H^ω and E^ω ⊥ or ∥ to **H** can excite



Selection rules from magnetoelectric coupling

Experiment:

When a peak is **missing from the Faraday** geometry but **present in Voigt:**
it has been excited with
H^{ω} and E^{ω} ǁ to **H**

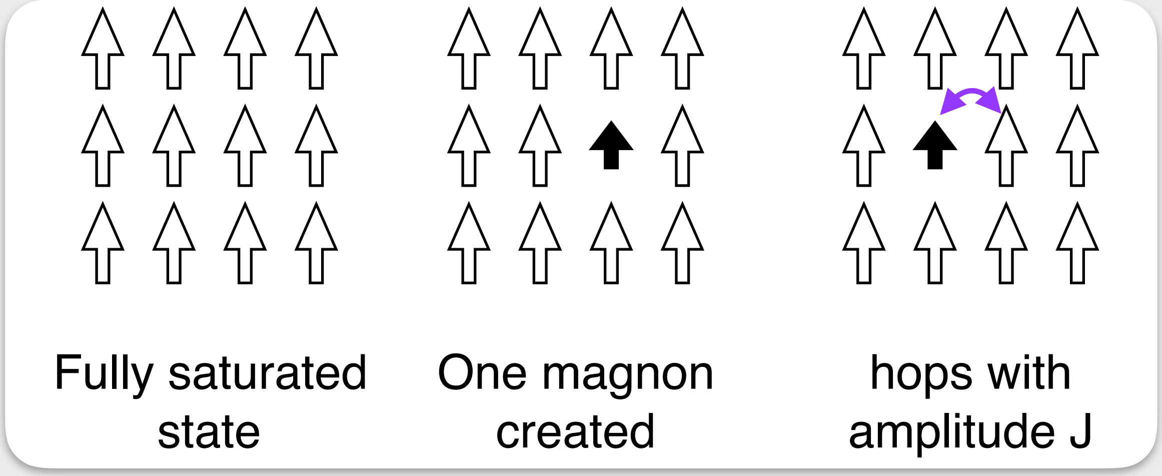


Theory:

H	Voigt $P^{\parallel}_{(0,0)}$	Voigt $P^{\parallel}_{(\pi,\pi)}$	⊥-directions	Voigt & Faraday $P^{\perp}_{(0,0)}$	Voigt & Faraday $P^{\perp}_{(\pi,\pi)}$
$H \parallel [100]$	(Q_0)	D_1	[010] and [001]	D_0	Q_1
$H \parallel [110]$	D_0	Q_1	$[\bar{1}10]$ and [001]	(Q_0)	D_1
$H \parallel [001]$	(Q_0)	Q_1	[100] and [010]	D_0	D_1

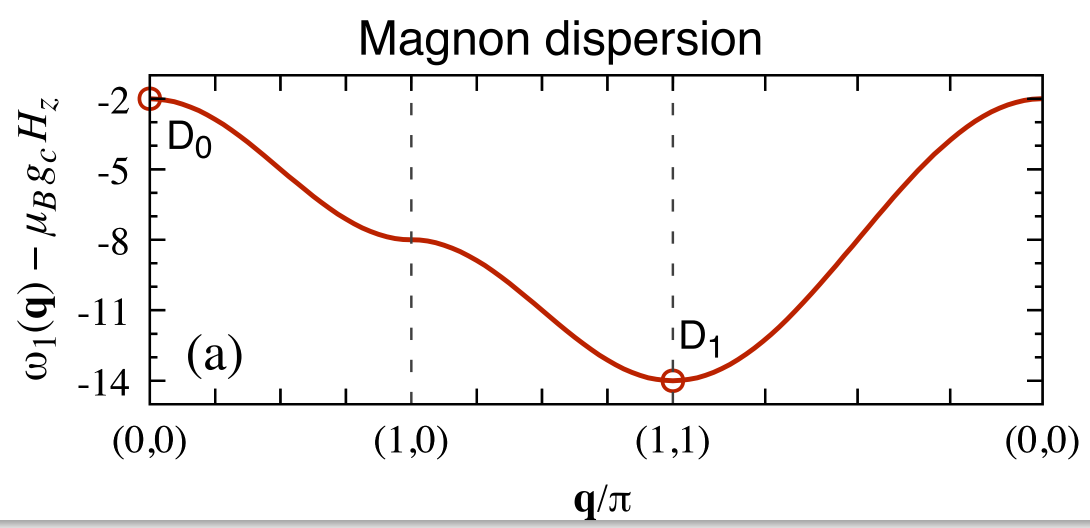
One-magnon excitations in [001] field

$$\mathcal{H} = J \sum_{(i,j)} \left(S_i^x S_j^x + S_i^y S_j^y\right) + J_z \sum_{(i,j)} S_i^z S_j^z + \Lambda \sum_i (S_i^z)^2 - \mu_{\rm B} g_c H_z \sum_i S_i^z \ ,$$

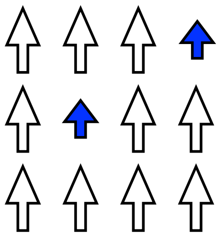


$$|\mathrm{D}(\mathbf{q})\rangle = \sum_j e^{i\mathbf{q}\cdot\mathbf{r}_j} S_j^- |\mathrm{GS}\rangle$$

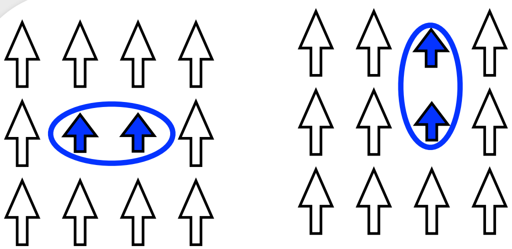
$$\omega_1(\mathbf{q}) = \mu_{\rm B} g_c H_z - 2\Lambda - 6J_z + 3J(\cos q_x + \cos q_y)$$



Two-magnon excitations in [001] field



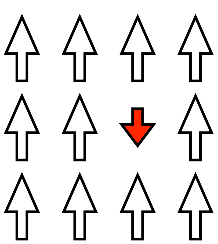
Two magnon can freely hop around, interaction arises when they are neighbors: 2 magnon continuum



B_1 bound states

M. Wortis, Phys. Rev. **132**, 85 (1963),
J. Hanus, Phys. Rev. Lett. **11**, 336 (1963).

$$\omega_{B_1}^{[001]} = 2\mu_B g_c H_z - 4\Lambda - 11 J_z$$

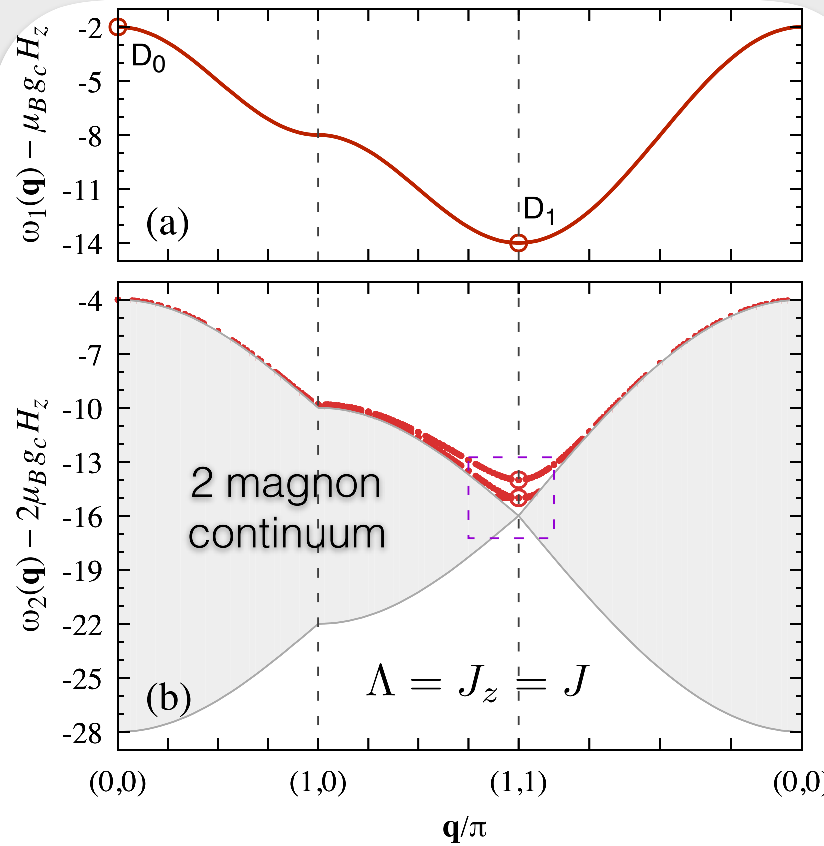


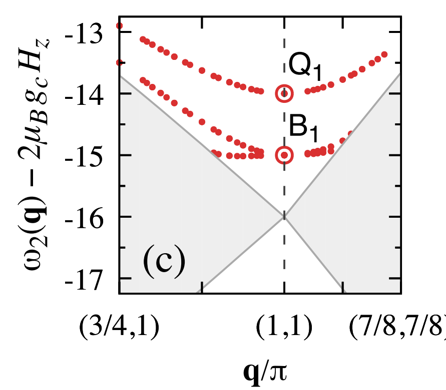
Q_1 bound states, Sz=-1/2 exact at **q**=(π,π)

$$|Q_1\rangle = \sum_j (-1)^j S_j^- S_j^- |GS\rangle$$

R. Silberglitt and J. B. Torrance, Jr., Phys. Rev. B **2**, 772 (1970)

$$\omega_{Q_1}^{[001]} = 2\mu_B g_c H_z - 2\Lambda - 12 J_z$$





Two-magnon excitations in [001] field

Two magnon (anti)bound states appear in the vicinity of **q**=(π,π),

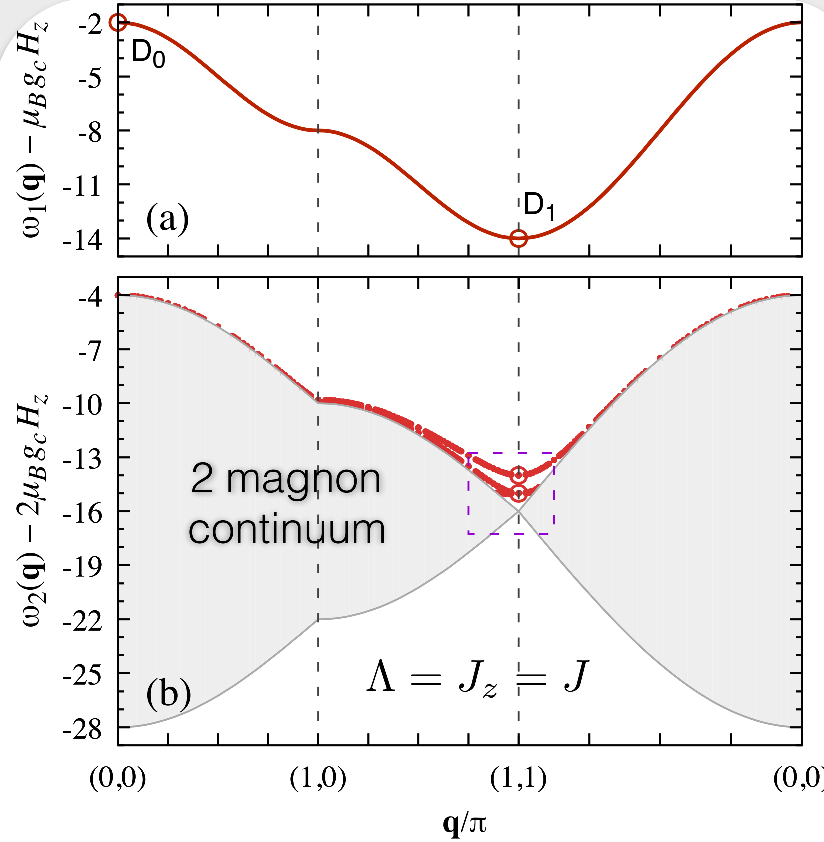
How can we see it?
How can we excite two-magnon with light?

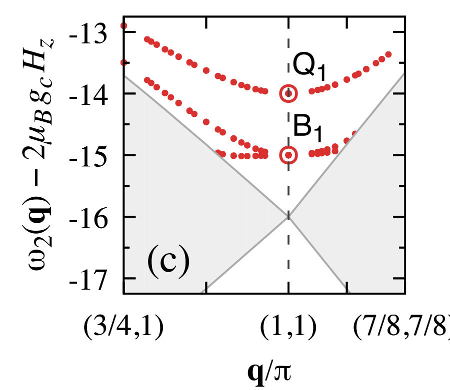
$$P_j^x \propto -\cos 2\kappa\, \overline{S_j^x S_j^z} - (-1)^j \sin 2\kappa\, \overline{S_j^y S_j^z}\ ,$$

$$P_j^y \propto \cos 2\kappa\, \overline{S_j^y S_j^z} - (-1)^j \sin 2\kappa\, \overline{S_j^x S_j^z}\ ,$$

$$P_j^z \propto \cos 2\kappa \left[(\hat{S}_j^y)^2 - (\hat{S}_j^x)^2\right] - (-1)^j \sin 2\kappa\, \overline{\hat{S}_j^x \hat{S}_j^y}\ ,$$

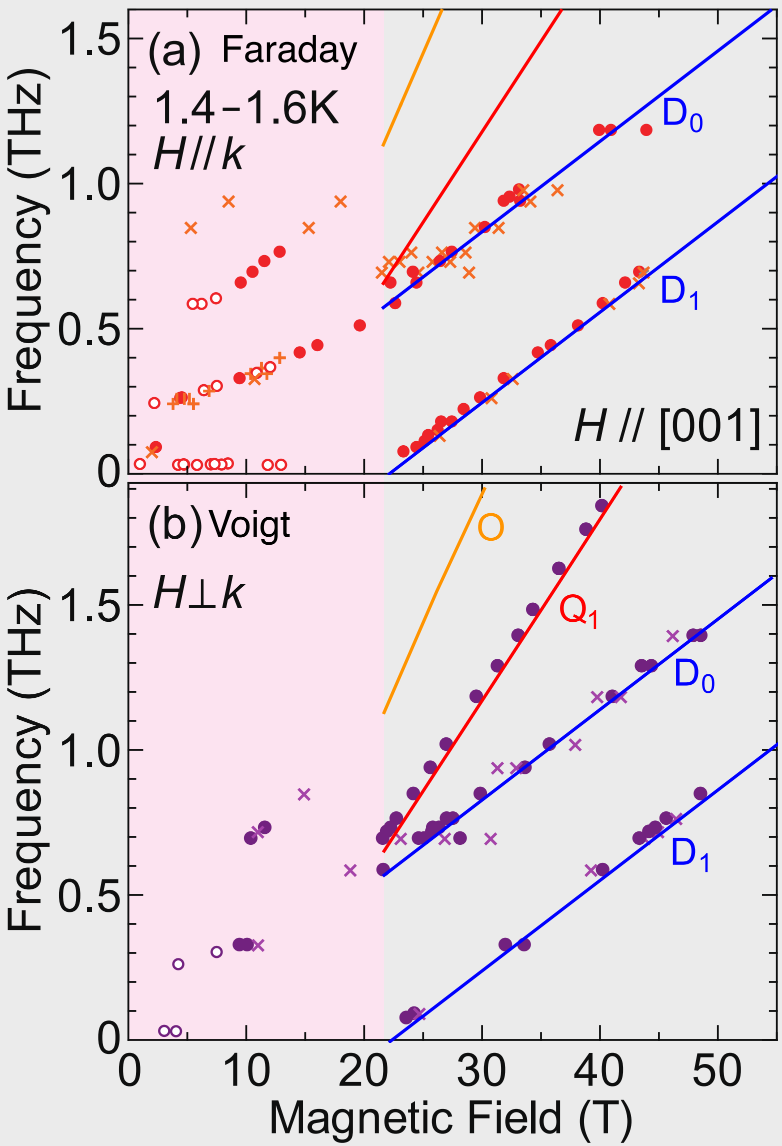
Q_0 at **q**=(0,0) Q_1 at **q**=(π,π)

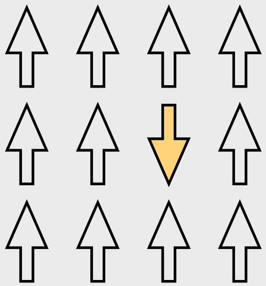




M. Akaki et al PRB **96**, 214406 (2017)

Octupole transition H ll [001]





one-magnon dipolar transition

$$\omega_{D_0} = g_z h^z - 2\Lambda$$

$$\omega_{D_1} = g_z h^z - 2\Lambda - 12J$$

two-magnon quadrupolar transition

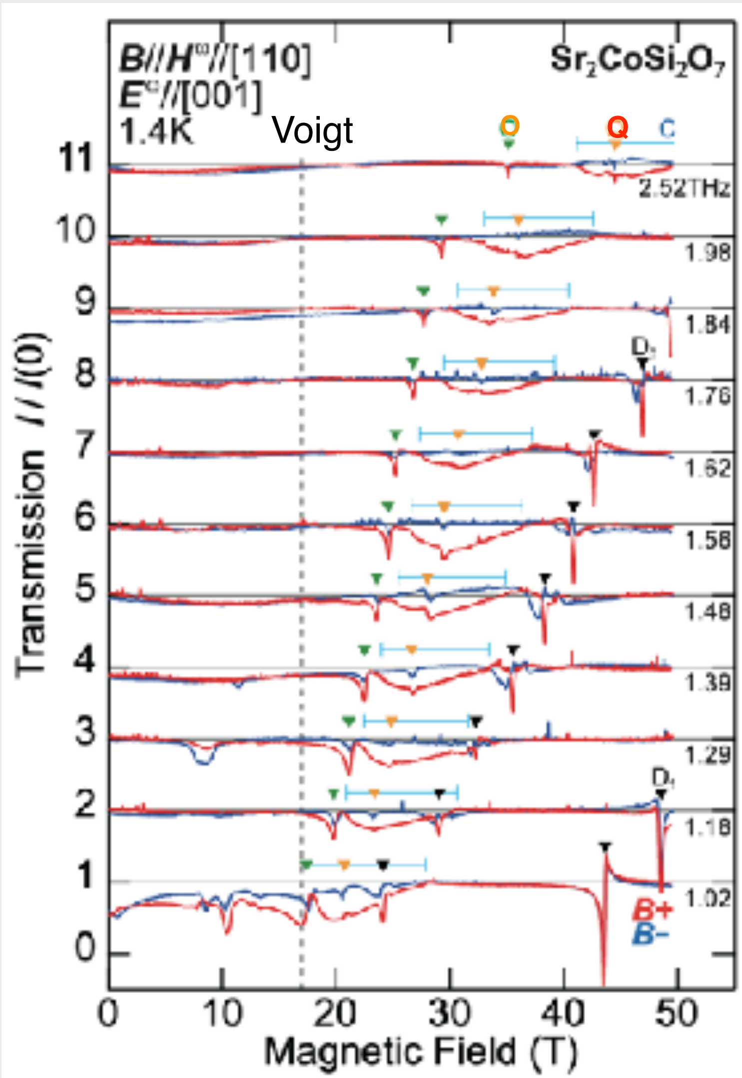
$$\omega_Q = 2g_z h^z - 12J - 2\Lambda$$

three-magnon octupolar transition

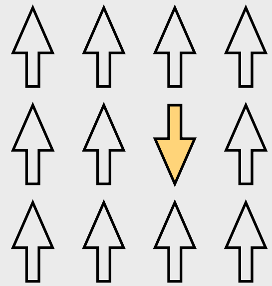
$$\omega_O = 3g_z h^z - 18J$$

Octupole transition H II [110]

Pulsed-field ESR measurements using polarized light



M. Akaki, Y. Narumi and M. Hagiwara unpublished



one-magnon dipolar transition

$$\omega_{D_0} = \mu_B g_x h^x + \Lambda - \frac{3\Lambda^2}{8\mu_B g_x h_x}$$

$$\omega_{D_1} = \mu_B g_x h^x + \Lambda - 12J - \frac{3\Lambda^2}{8\mu_B g_x h^x}$$

two-magnon quadrupolar transition

$$\omega_Q = 2\mu_B g_x h^x + \Lambda - 12J$$

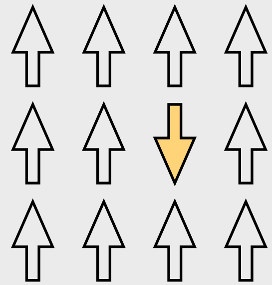
three-magnon octupolar transition

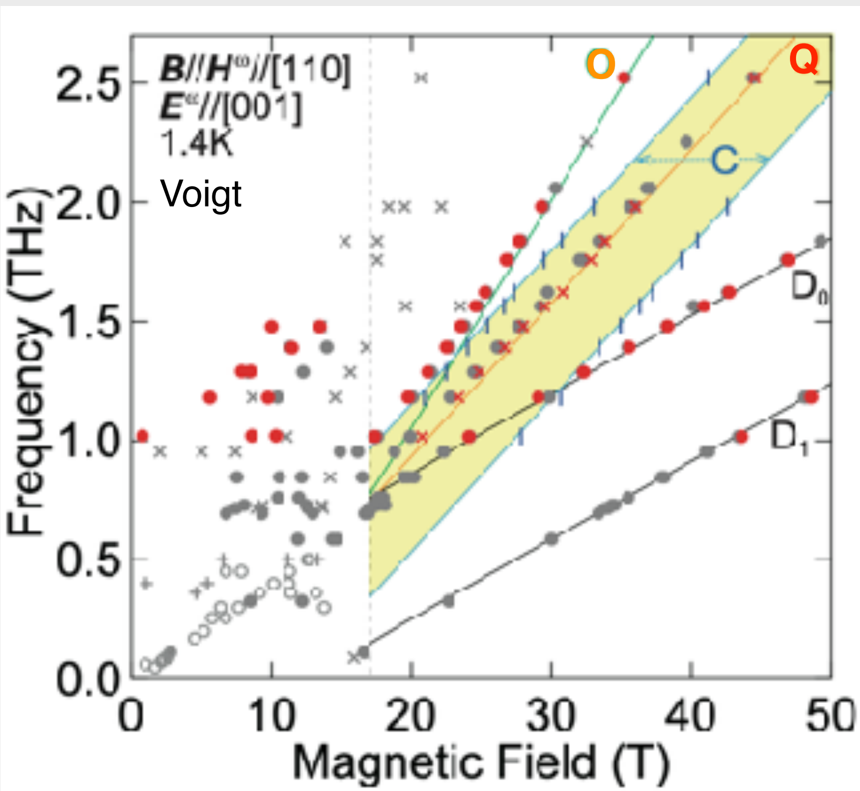
$$\omega_O = 3\mu_B g_x h^x - 18J + \frac{3\Lambda^2}{8\mu_B g_x h^x}$$

Octupole transition H ll [110]

M. Akaki, Y. Narumi and M. Hagiwara unpublished

Pulsed-field ESR measurements using polarized light





one-magnon dipolar transition

$$\omega_{D_0} = \mu_B g_x h^x + \Lambda - \frac{3\Lambda^2}{8\mu_B g_x h_x}$$

$$\omega_{D_1} = \mu_B g_x h^x + \Lambda - 12J - \frac{3\Lambda^2}{8\mu_B g_x h^x}$$

two-magnon quadrupolar transition

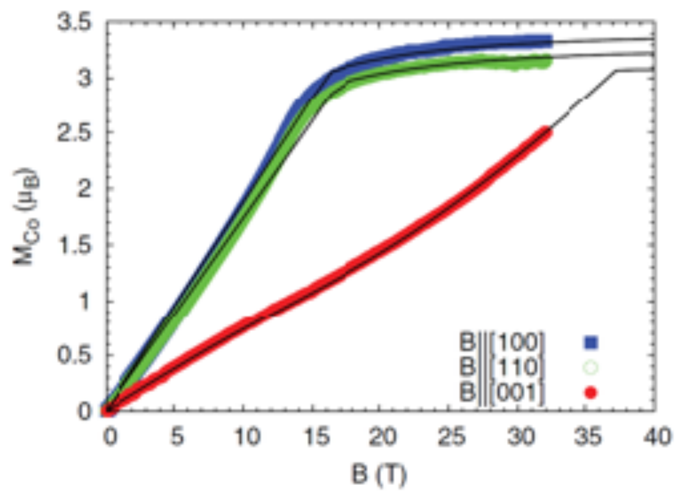
$$\omega_Q = 2\mu_B g_x h^x + \Lambda - 12J$$

three-magnon octupolar transition

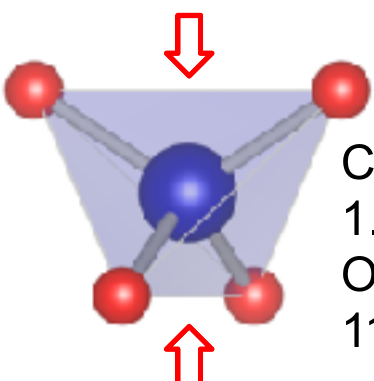
$$\omega_O = 3\mu_B g_x h^x - 18J + \frac{3\Lambda^2}{8\mu_B g_x h^x}$$

Comparing with $Ba_2CoGe_2O_7$

Anisotropy large Λ/J ≈ 6



V. Hutanu et al., PRB **89**, 064403 (2014)



Co-O length
1.98Å
O-Co-O angle
116.7°

$Ba_2CoGe_2O_7$
single crystal neutron, @RT
A. Sazonov *et al*., J. Appl. Cryst. **49**, 556 (2016).

h=0 the ground state is the planar antiferromagnetic state

$$|\Psi_A\rangle = e^{-i\varphi_A \hat{S}^z_A}|\Psi_{\rm SF}\rangle,$$
$$|\Psi_B\rangle = e^{-i\varphi_B \hat{S}^z_B}|\Psi_{\rm SF}\rangle,$$

with

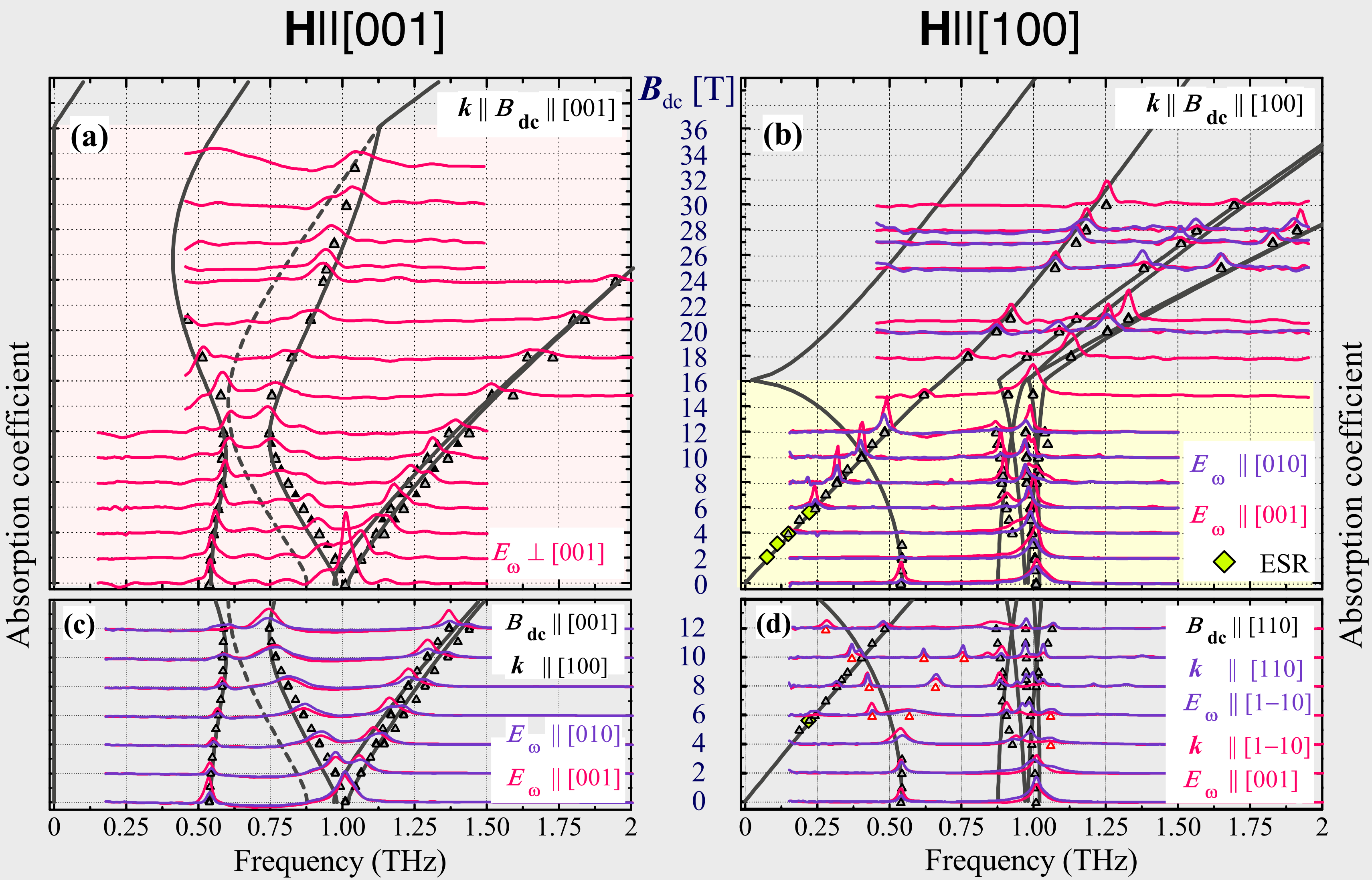
$$|\Psi_{\rm SF}\rangle = \frac{|\frac{3}{2}\rangle - i\sqrt{3}\eta|\frac{1}{2}\rangle - \sqrt{3}\eta|-\frac{1}{2}\rangle + i|-\frac{3}{2}\rangle}{\sqrt{6\eta^2+2}}$$

$$\langle \mathbf{S}_j\rangle = \frac{3\eta(\eta+1)}{3\eta^2+1}(\cos\varphi_j, \sin\varphi_j, 0)$$

$\eta \neq 1$ spin length shorter than 3/2

multipole degrees of freedom are mixed

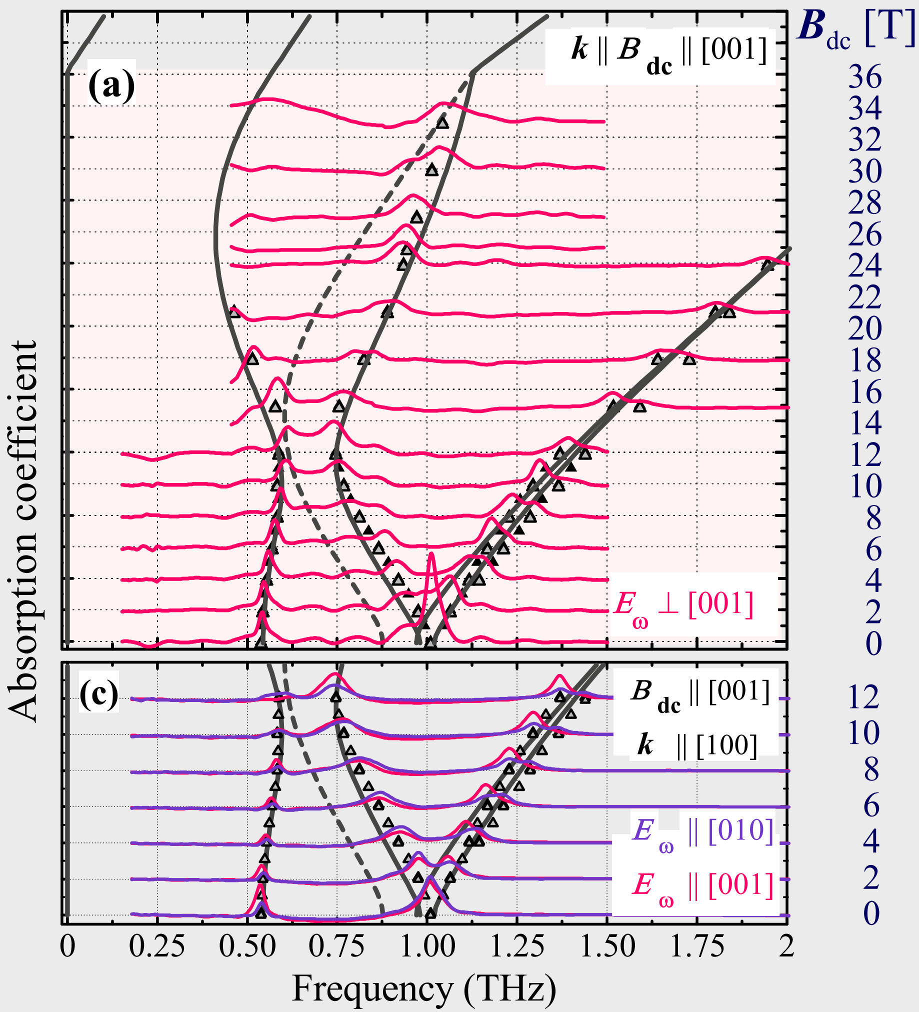
Comparing with $Ba_2CoGe_2O_7$



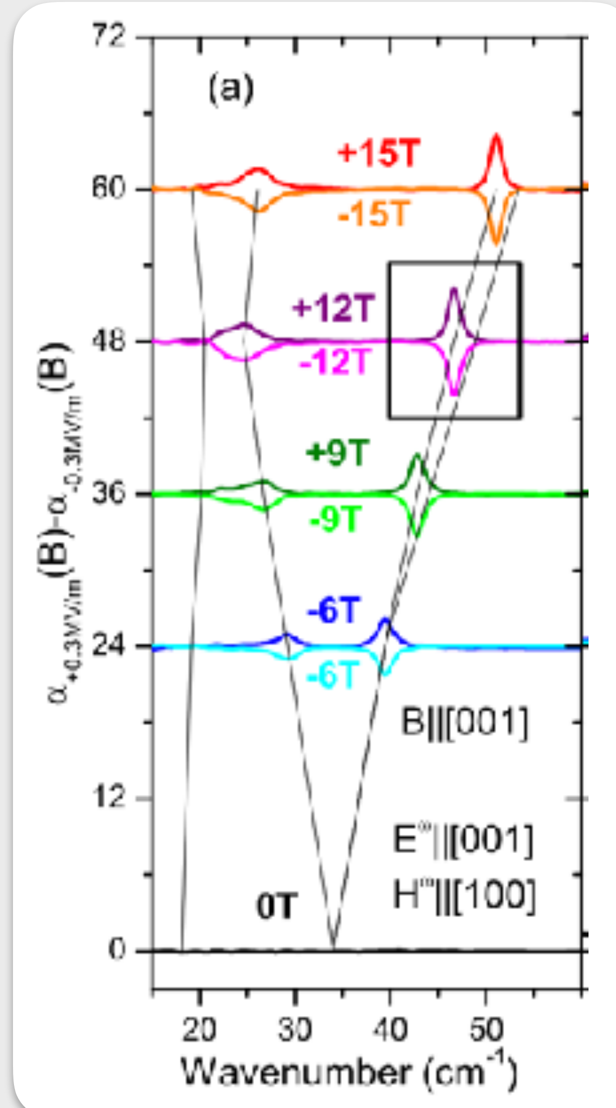
K. Penc et al PRL **108**, 257203 (2012)

Directional dichroism $Ba_2CoGe_2O_7$

H||[001]



J. Vít and S. Bordács unpublished



K. Penc et al PRL **108**, 257203 (2012)

Directional dichroism $Ba_2CoGe_2O_7$

J. Vít and S. Bordács unpublished

$$H^\omega \| [100] \rightarrow \mathrm{Im}\chi^{mm}_{xx}(\omega) \propto \sum_f |\langle f|S^x|0\rangle|^2 \delta(\omega - \omega_f)$$

$$E^\omega \| [001] \rightarrow \mathrm{Im}\chi^{ee}_{zz}(\omega) \propto \sum_f |\langle f|P^z|0\rangle|^2 \delta(\omega - \omega_f)$$

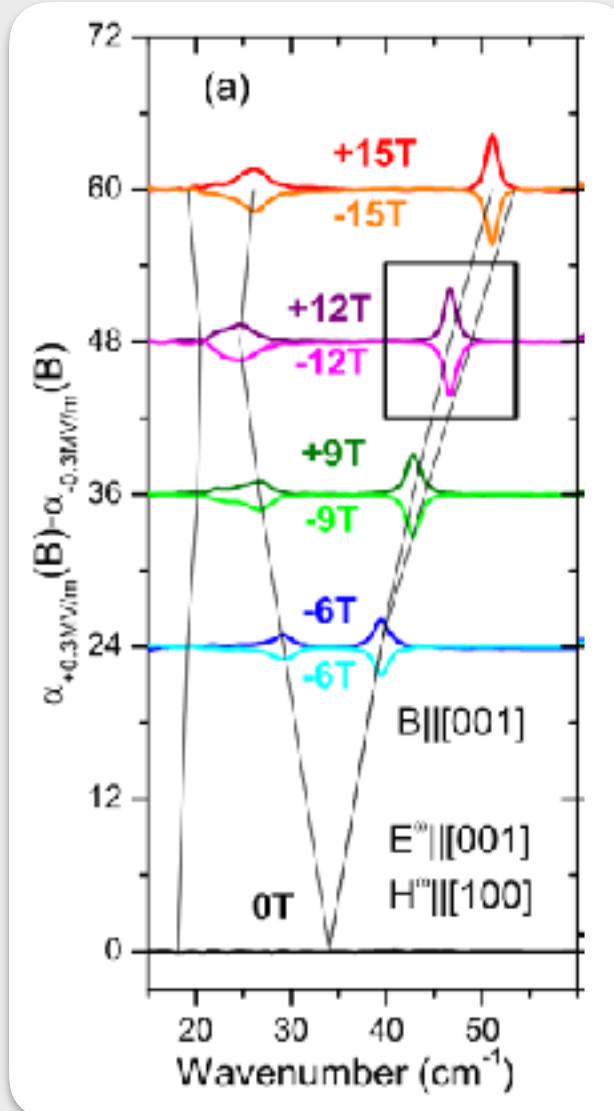
$$\mathrm{Im}\chi^{me}_{xz}(\omega) \propto \sum_f (\langle 0|S^x|f\rangle \langle f|P^z|0\rangle + \langle 0|P^z|f\rangle \langle f|S^x|0\rangle)\delta(\omega - \omega_f)$$

$$\alpha_\pm \propto \mathrm{Im}N_\pm \propto \mathrm{Im}(\sqrt{\epsilon_{zz}(\omega)\mu_x x(\omega)} \pm \chi^{me}_{xz}(\omega))$$

$$\downarrow$$

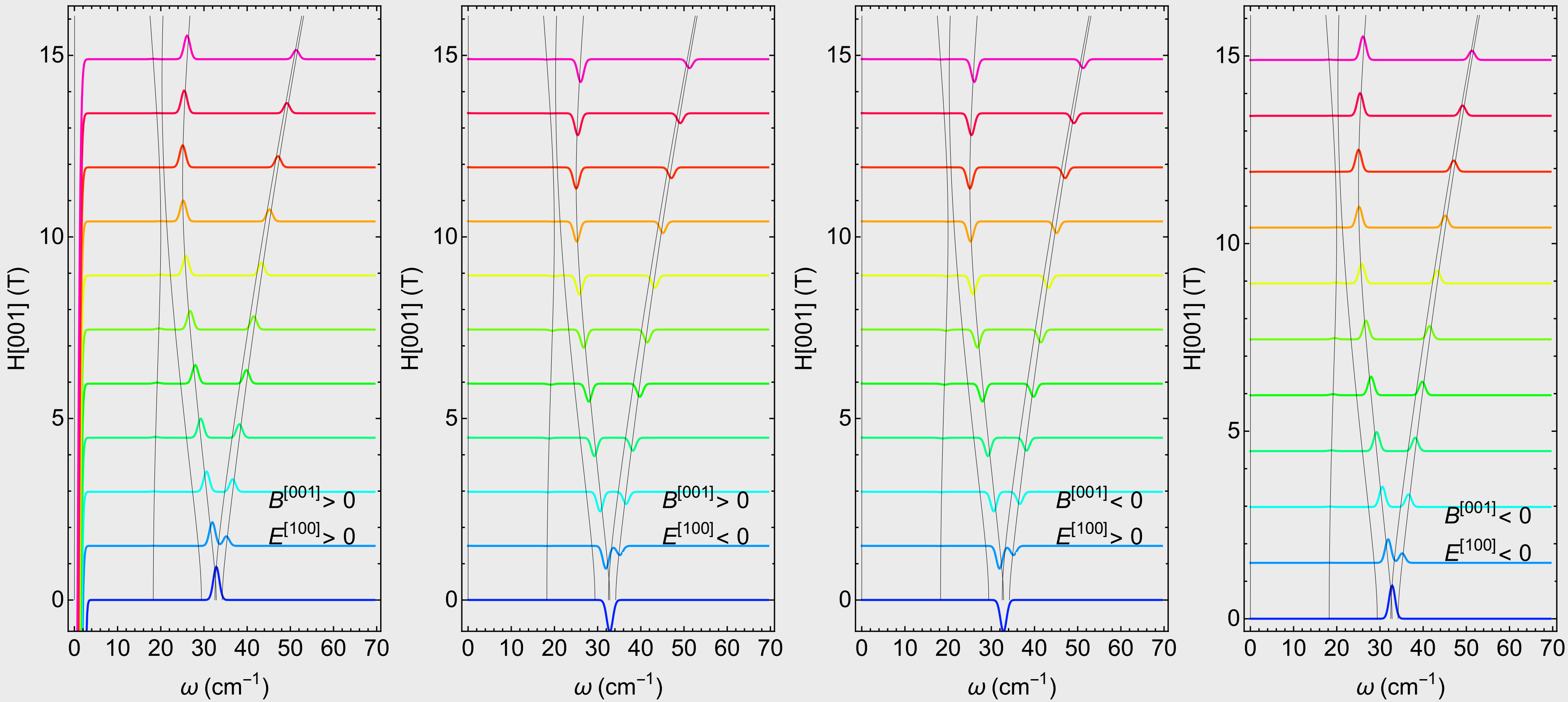
$$\alpha_+ - \alpha_- \propto 2\mathrm{Im}\chi^{me}_{xz}(\omega)$$

$E > 0$ $B > 0$ $\chi^{me} > 0$	$E > 0$ $B < 0$ $\chi^{me} < 0$
$E < 0$ $B > 0$ $\chi^{me} < 0$	$E < 0$ $B < 0$ $\chi^{me} > 0$



Directional dichroism $Ba_2CoGe_2O_7$

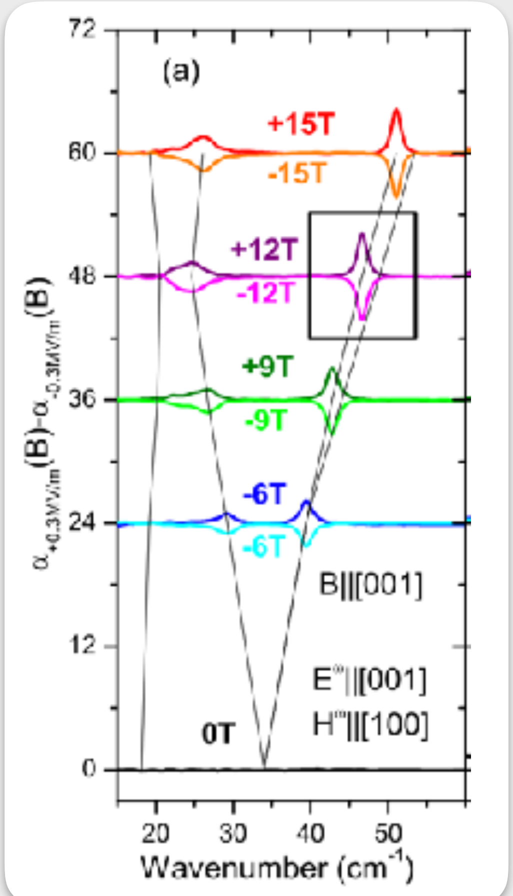
$E > 0 \quad B > 0$, $\chi^{me} > 0$	$E < 0 \quad B > 0$, $\chi^{me} < 0$	$E > 0 \quad B < 0$, $\chi^{me} < 0$	$E < 0 \quad B < 0$, $\chi^{me} > 0$



$$\mathrm{Im}\chi^{me}_{xz}(\omega) \propto \sum_f (\langle 0|S^x|f\rangle \langle f|P^z|0\rangle + \langle 0|P^z|f\rangle \langle f|S^x|0\rangle)\delta(\omega - \omega_f)$$

Anyway

$Ba_2CoGe_2O_7$



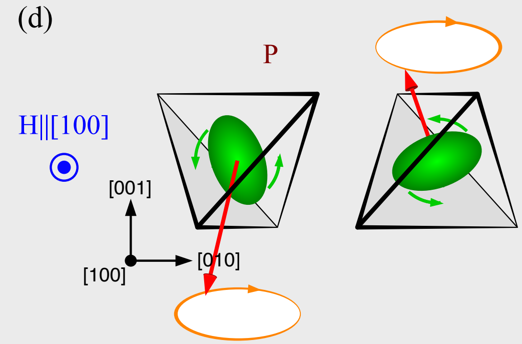
unpublished

directional dichroism

and

E field control of magnetic domains

$Sr_2CoGe_2O_7$



quadrupolar (octupolar) mode observed by ME coupling