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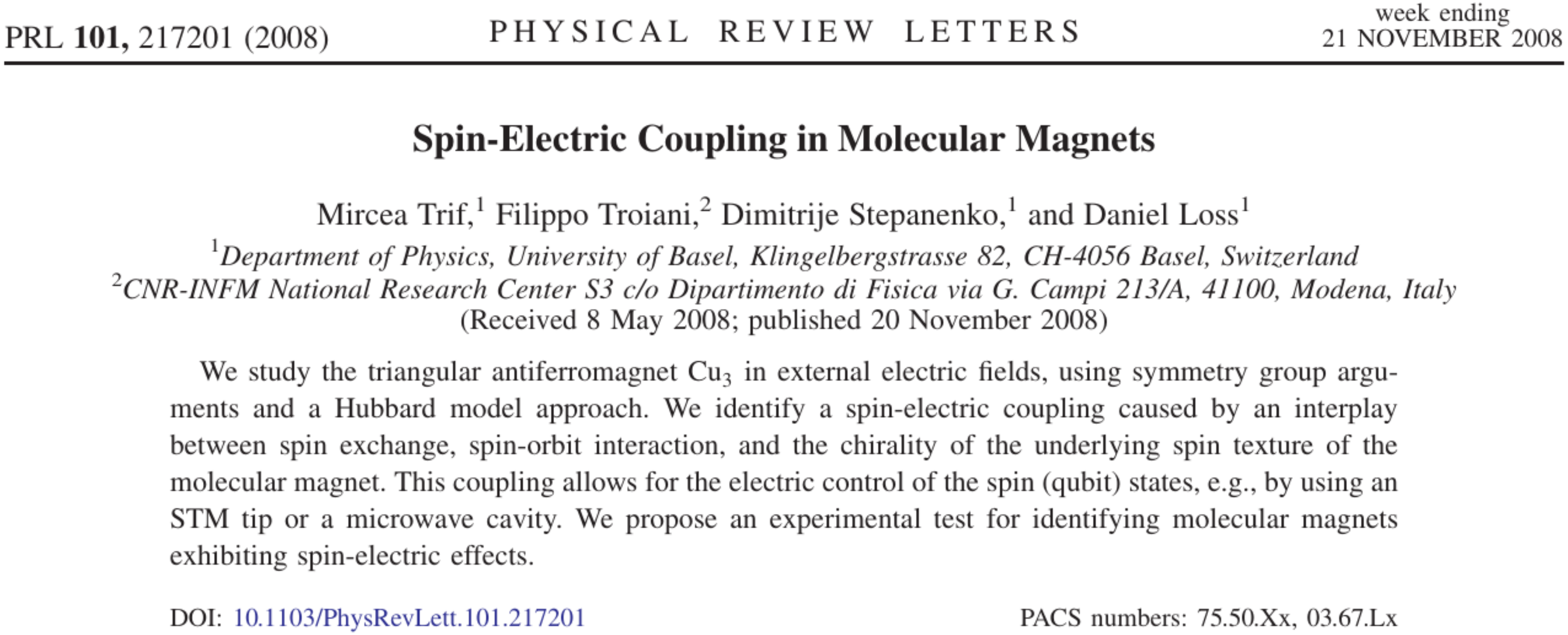
Spin chirality: a new concept in the arsenal of Molecular Magnetism

Athanassios K. Boudalis

JAM^2, Dourdan
27/11/2024

In medias res: a new idea for spin triangles



PRL **101,** 217201 (2008) PHYSICAL REVIEW LETTERS week ending 21 NOVEMBER 2008

Spin-Electric Coupling in Molecular Magnets

Mircea Trif,[1] Filippo Troiani,[2] Dimitrije Stepanenko,[1] and Daniel Loss[1]
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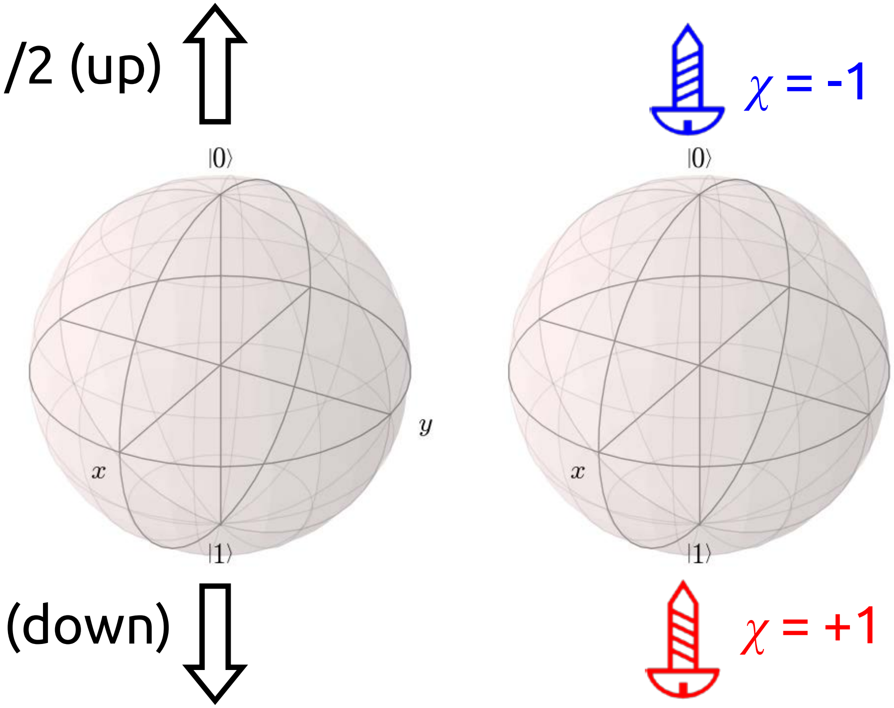
[2]*CNR-INFM National Research Center S3 c/o Dipartimento di Fisica via G. Campi 213/A, 41100, Modena, Italy*
(Received 8 May 2008; published 20 November 2008)

We study the triangular antiferromagnet Cu_3 in external electric fields, using symmetry group arguments and a Hubbard model approach. We identify a spin-electric coupling caused by an interplay between spin exchange, spin-orbit interaction, and the chirality of the underlying spin texture of the molecular magnet. This coupling allows for the electric control of the spin (qubit) states, e.g., by using an STM tip or a microwave cavity. We propose an experimental test for identifying molecular magnets exhibiting spin-electric effects.

DOI: 10.1103/PhysRevLett.101.217201 PACS numbers: 75.50.Xx, 03.67.Lx



M_S = -1/2 (up)

χ = -1

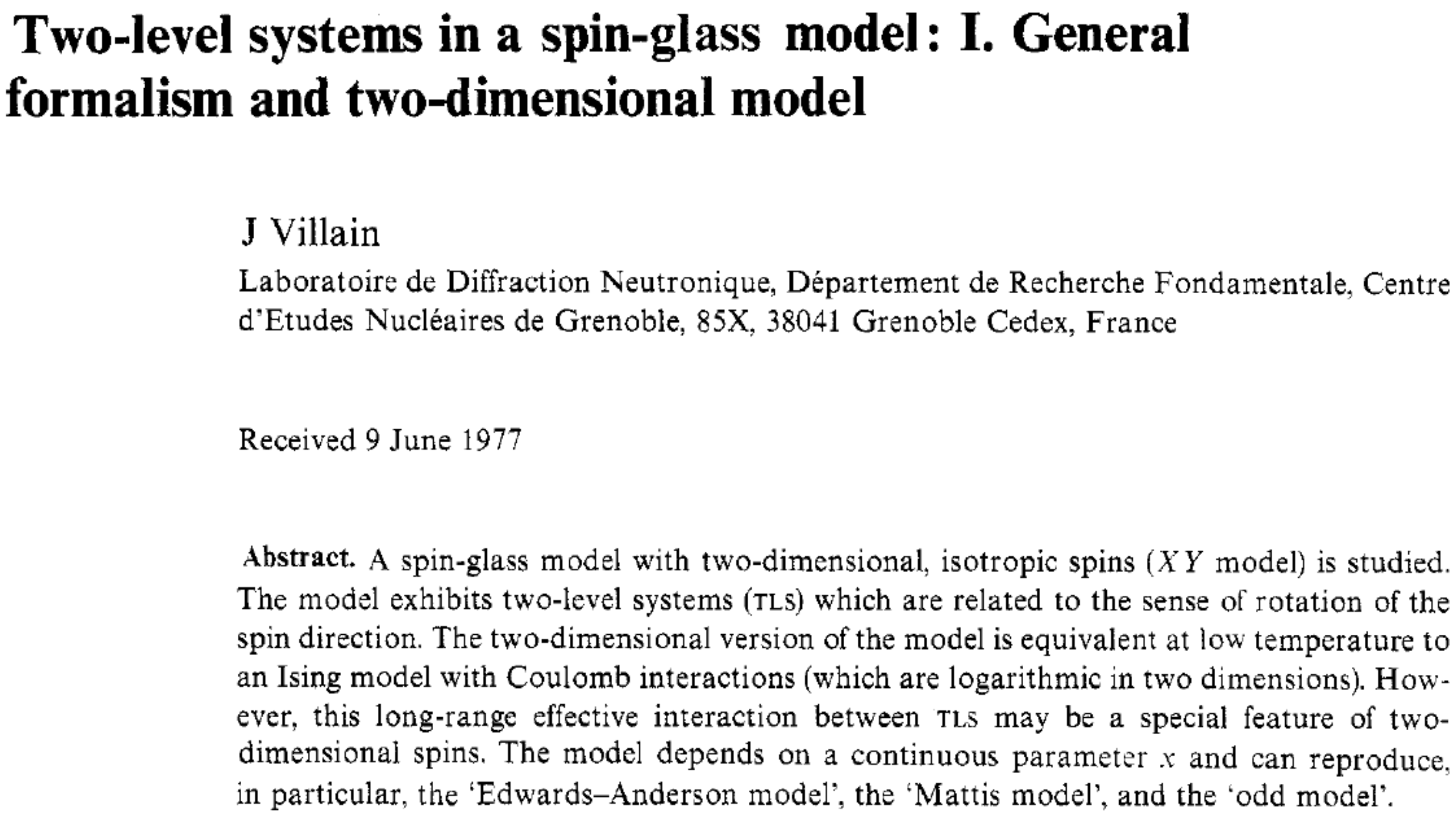
Spin projection

Spin chirality

M_S = +1/2 (down)

χ = +1

Ab ovo: a new quantum property



Two-level systems in a spin-glass model: I. General formalism and two-dimensional model

J Villain
Laboratoire de Diffraction Neutronique, Département de Recherche Fondamentale, Centre d'Etudes Nucléaires de Grenoble, 85X, 38041 Grenoble Cedex, France

Received 9 June 1977

Abstract. A spin-glass model with two-dimensional, isotropic spins (XY model) is studied. The model exhibits two-level systems (TLS) which are related to the sense of rotation of the spin direction. The two-dimensional version of the model is equivalent at low temperature to an Ising model with Coulomb interactions (which are logarithmic in two dimensions). However, this long-range effective interaction between TLS may be a special feature of two-dimensional spins. The model depends on a continuous parameter x and can reproduce, in particular, the 'Edwards–Anderson model', the 'Mattis model', and the 'odd model'.

Villain, *J. Phys. C: Solid State Phys.*, **1977**, *10*, 4793

Ab ovo: a new quantum property

Spin- and chirality-orderings of frustrated magnets — stacked-triangular anti-ferromagnets and spin glasses

Hikaru Kawamura

Vector chirality $$\kappa = \frac{2}{3\sqrt{3}} \sum_{<ij>} \vec{S}_i \times \vec{S}_j$$

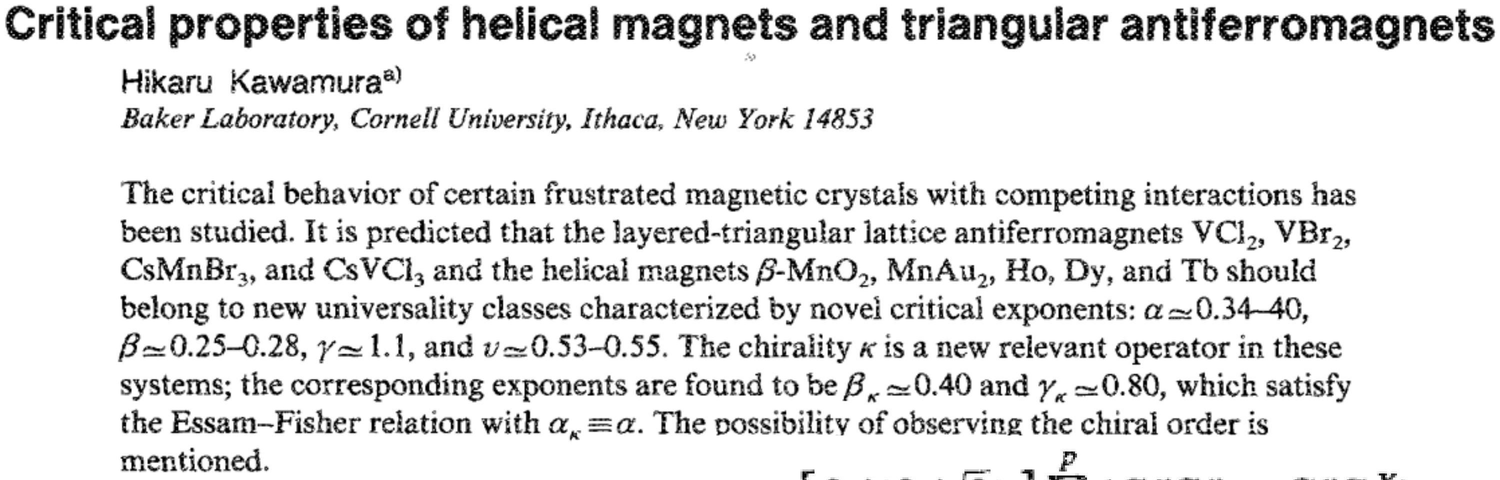
Scalar chirality $$\chi = \vec{S}_i \cdot \vec{S}_{i+\delta} \times \vec{S}_{i+\delta'}$$

Kawamura, *Can. J. Phys.*, **2001**, *79*, 1447

Ab ovo: a new quantum property

Vector chirality

$$\hat{\mathbf{K}} = \sum_{i,j=1-3} (\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j)$$



Critical properties of helical magnets and triangular antiferromagnets

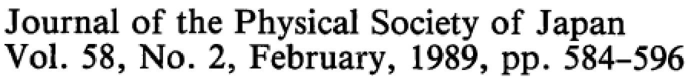
Hikaru Kawamura[a)]

Baker Laboratory, Cornell University, Ithaca, New York 14853

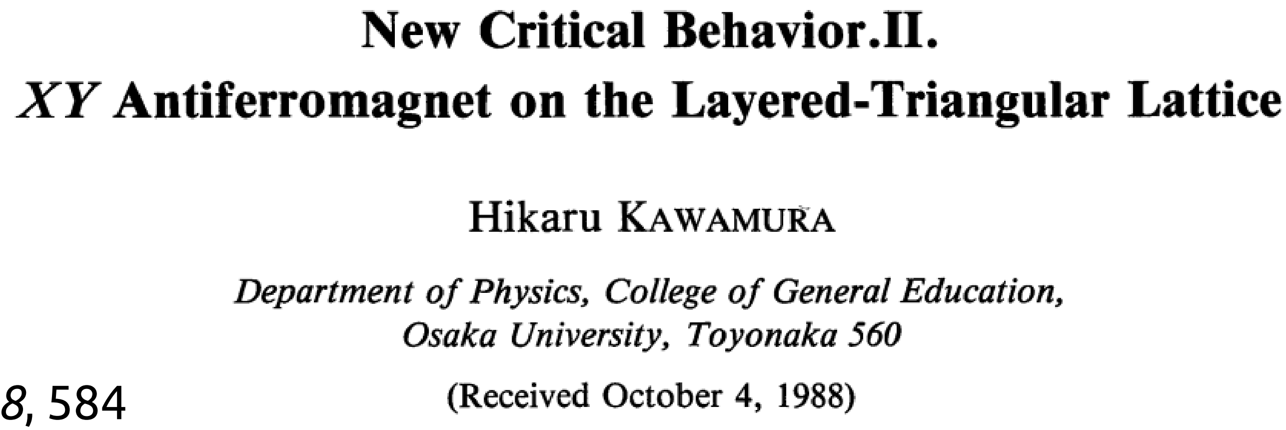
The critical behavior of certain frustrated magnetic crystals with competing interactions has been studied. It is predicted that the layered-triangular lattice antiferromagnets VCl_2, VBr_2, $CsMnBr_3$, and $CsVCl_3$ and the helical magnets β-MnO_2, $MnAu_2$, Ho, Dy, and Tb should belong to new universality classes characterized by novel critical exponents: $\alpha \simeq 0.34$–40, $\beta \simeq 0.25$–0.28, $\gamma \simeq 1.1$, and $\nu \simeq 0.53$–0.55. The chirality κ is a new relevant operator in these systems; the corresponding exponents are found to be $\beta_\kappa \simeq 0.40$ and $\gamma_\kappa \simeq 0.80$, which satisfy the Essam–Fisher relation with $\alpha_\kappa \equiv \alpha$. The possibility of observing the chiral order is mentioned.

Kawamura, *J. Appl. Phys.*, **1988**, *63*, 3086

$$\kappa_p = [2/(3/\sqrt{3})] \sum_{\langle ij \rangle}^{p} \underbrace{(S_i^x S_j^y - S_i^y S_j^x)}_{(\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j)_z}$$



Journal of the Physical Society of Japan
Vol. 58, No. 2, February, 1989, pp. 584–596



New Critical Behavior. II.
***XY* Antiferromagnet on the Layered-Triangular Lattice**

Hikaru KAWAMURA

Department of Physics, College of General Education,
Osaka University, Toyonaka 560

(Received October 4, 1988)

Kawamura, *J. Phys. Soc. Jpn.*, **1989**, *58*, 584

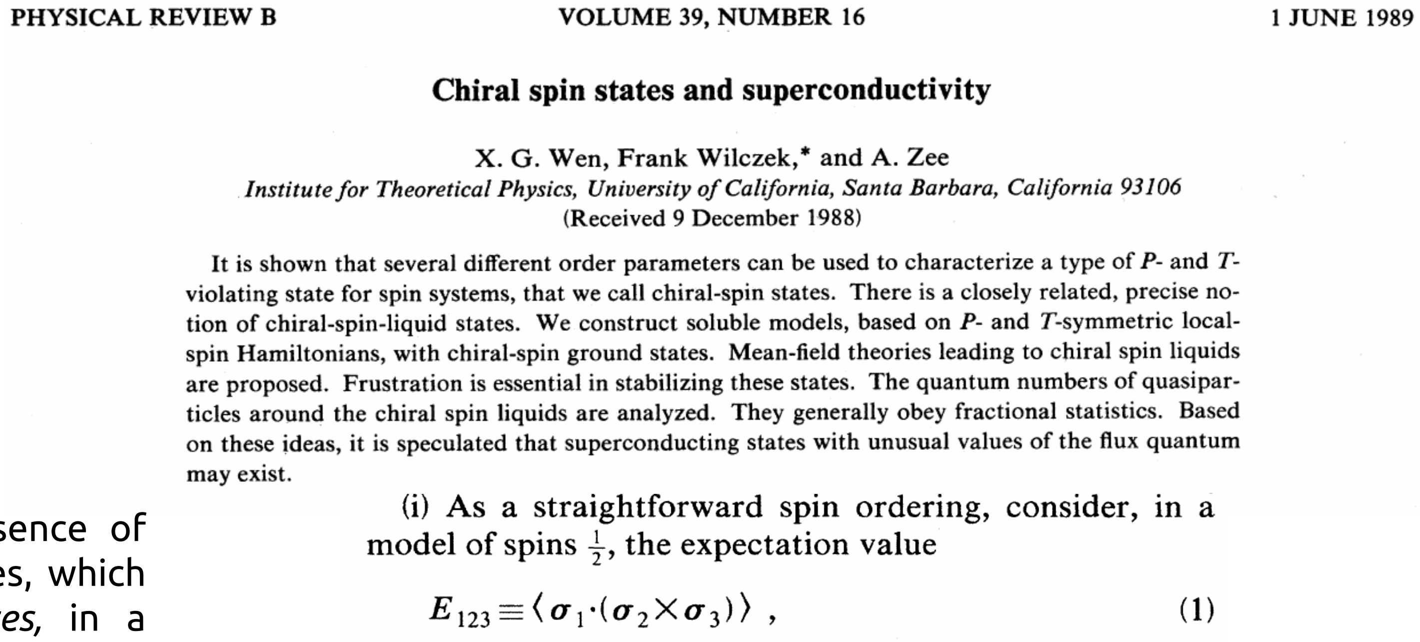
Ab ovo: a new quantum property

“Scalar” chirality

$$\hat{C}_z = \hat{\mathbf{S}}_1 \cdot (\hat{\mathbf{S}}_2 \times \hat{\mathbf{S}}_3)$$

Wen et al. PRB, **1989**, *39*, 11413

“…we characterize the common essence of the proposed *P*- and *T*-violating states, which we call generically *chiral spin states,* in a precise way. We do this, by defining a local order parameter.”



PHYSICAL REVIEW B VOLUME 39, NUMBER 16 1 JUNE 1989

Chiral spin states and superconductivity

X. G. Wen, Frank Wilczek,* and A. Zee
Institute for Theoretical Physics, University of California, Santa Barbara, California 93106

(Received 9 December 1988)

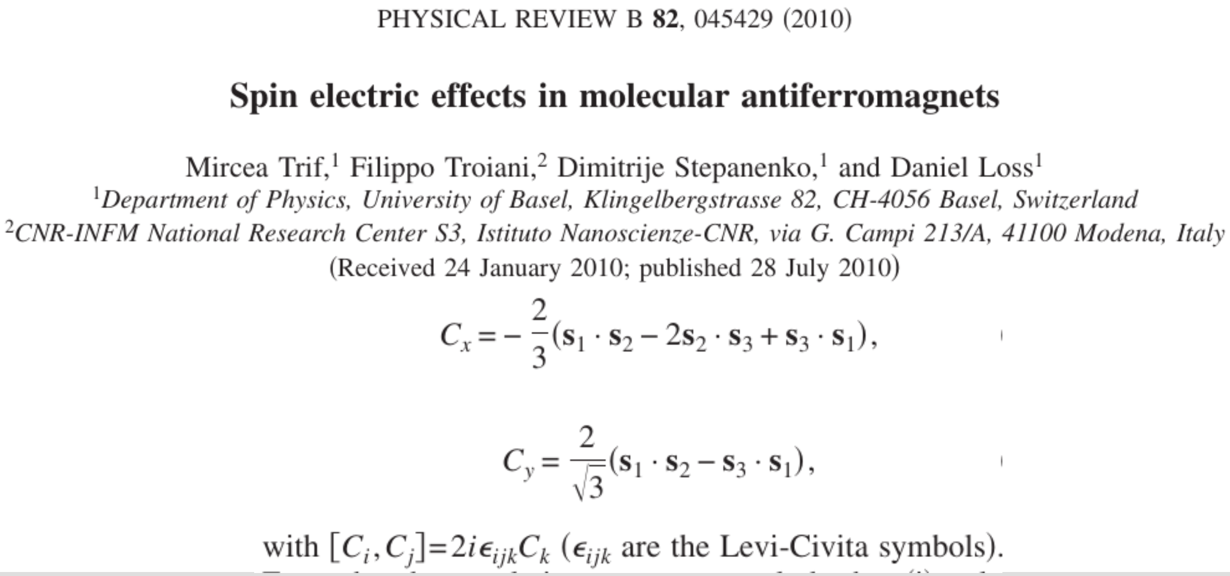
It is shown that several different order parameters can be used to characterize a type of *P*- and *T*-violating state for spin systems, that we call chiral-spin states. There is a closely related, precise notion of chiral-spin-liquid states. We construct soluble models, based on *P*- and *T*-symmetric local-spin Hamiltonians, with chiral-spin ground states. Mean-field theories leading to chiral spin liquids are proposed. Frustration is essential in stabilizing these states. The quantum numbers of quasiparticles around the chiral spin liquids are analyzed. They generally obey fractional statistics. Based on these ideas, it is speculated that superconducting states with unusual values of the flux quantum may exist.

(i) As a straightforward spin ordering, consider, in a model of spins $\frac{1}{2}$, the expectation value

$$E_{123} \equiv \langle \boldsymbol{\sigma}_1 \cdot (\boldsymbol{\sigma}_2 \times \boldsymbol{\sigma}_3) \rangle \; , \qquad (1)$$

C_x and C_y

Trif et al. PRB, **2010**, *82*, 045429



PHYSICAL REVIEW B **82**, 045429 (2010)

Spin electric effects in molecular antiferromagnets

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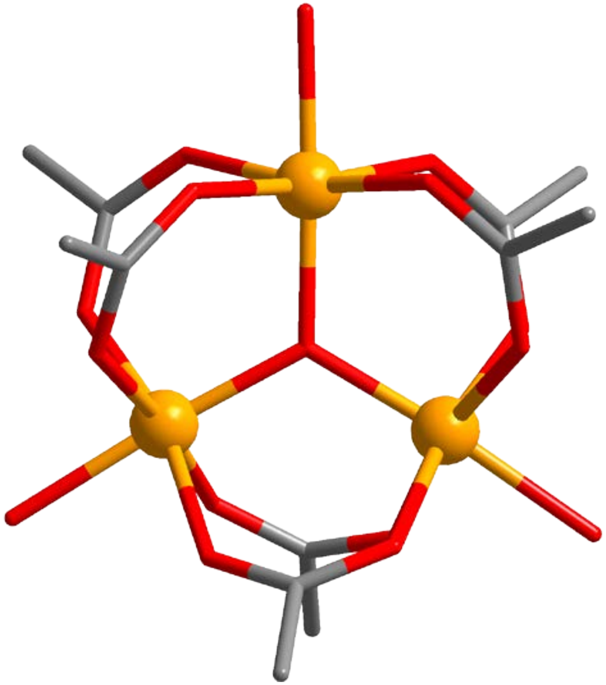
(Received 24 January 2010; published 28 July 2010)

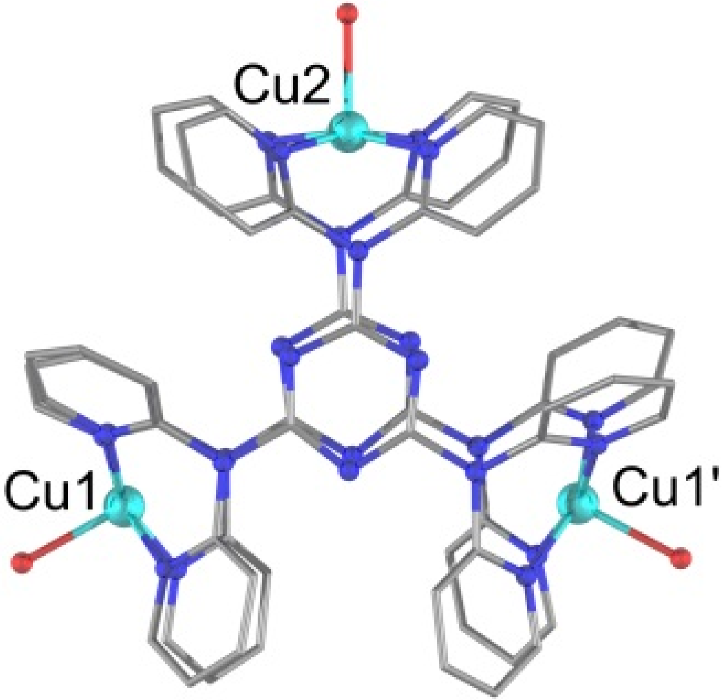
$$C_x = -\frac{2}{3}(\mathbf{s}_1 \cdot \mathbf{s}_2 - 2\mathbf{s}_2 \cdot \mathbf{s}_3 + \mathbf{s}_3 \cdot \mathbf{s}_1),$$

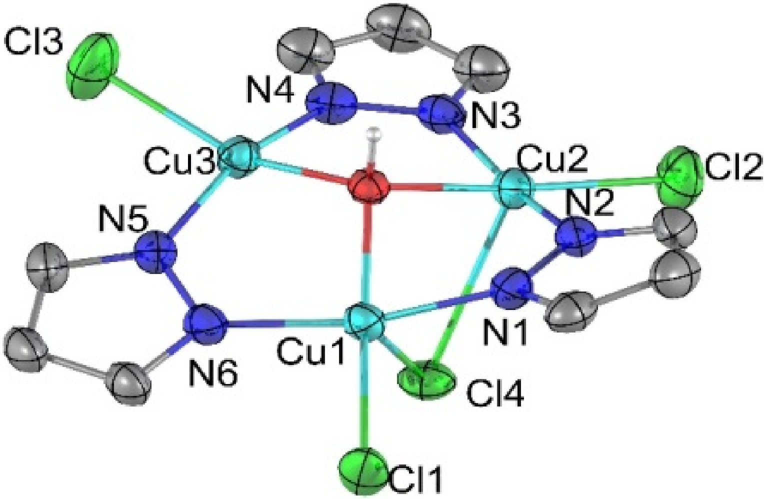
$$C_y = \frac{2}{\sqrt{3}}(\mathbf{s}_1 \cdot \mathbf{s}_2 - \mathbf{s}_3 \cdot \mathbf{s}_1),$$

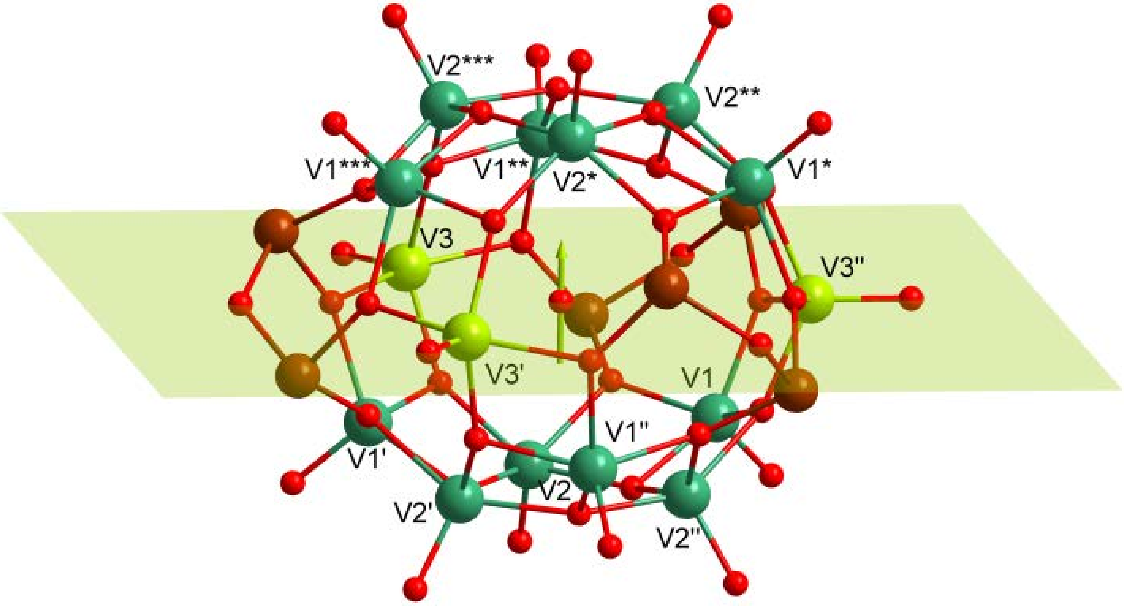
with $[C_i, C_j] = 2i\epsilon_{ijk}C_k$ (ϵ_{ijk} are the Levi-Civita symbols).

Spin-chiral chemical systems





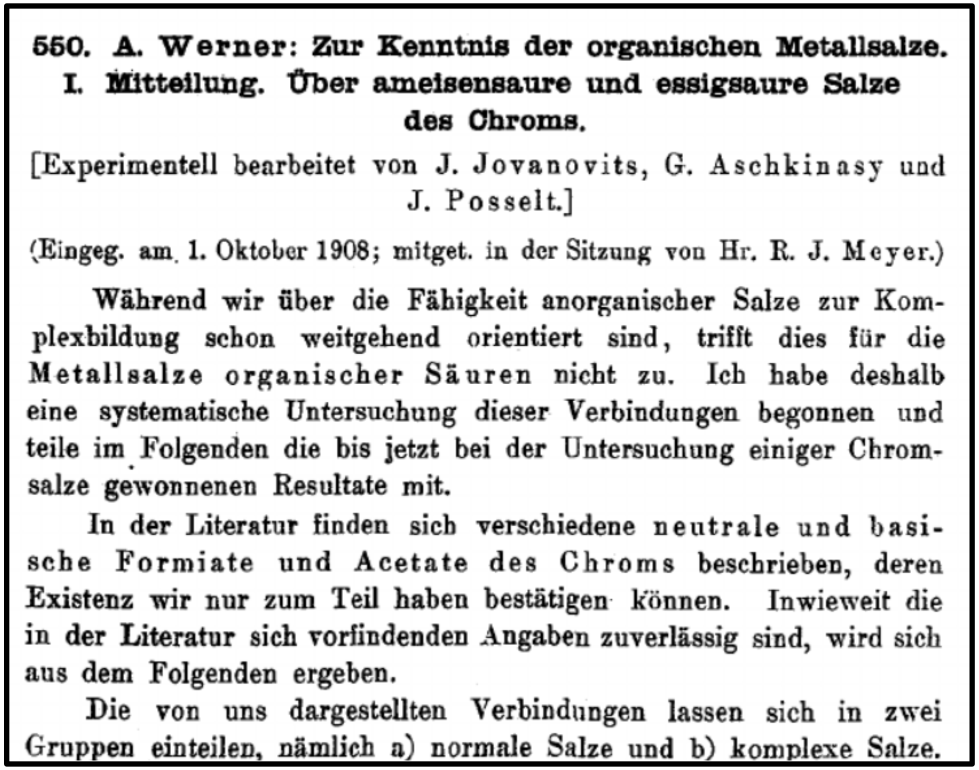




- S_i = 1/2, 3/2, 5/2…
- Antiferromagnetic
- ~D_{3h} or C_{3v} symmetry

An old story

Rudolf F. Weinland, *Berichte*, **1908**, *41*, 3236



550. A. Werner: Zur Kenntnis der organischen Metallsalze. I. Mitteilung. Über ameisensaure und essigsaure Salze des Chroms.

[Experimentell bearbeitet von J. Jovanovits, G. Aschkinasy und J. Posselt.]

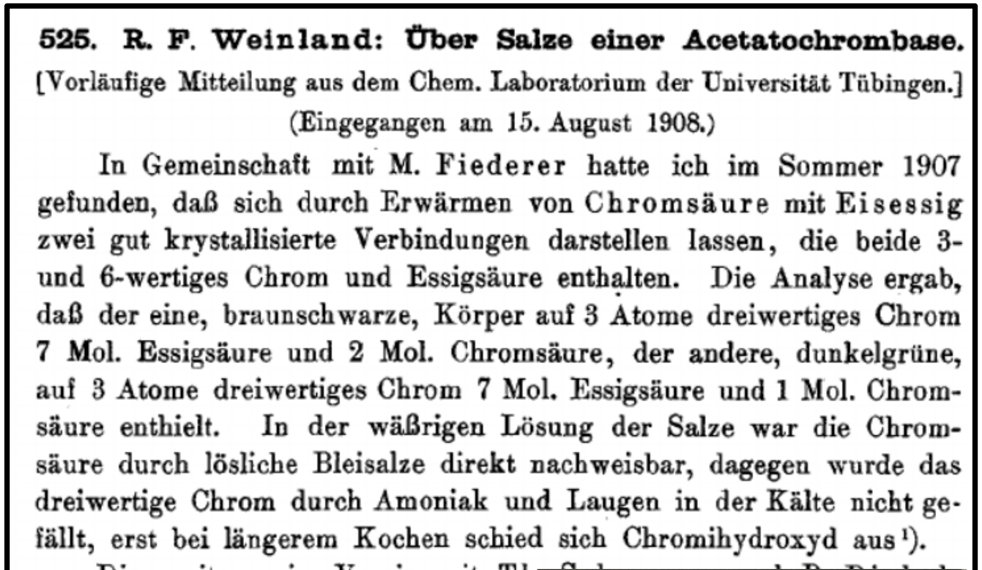
(Eingeg. am 1. Oktober 1908; mitget. in der Sitzung von Hr. R. J. Meyer.)

Während wir über die Fähigkeit anorganischer Salze zur Komplexbildung schon weitgehend orientiert sind, trifft dies für die Metallsalze organischer Säuren nicht zu. Ich habe deshalb eine systematische Untersuchung dieser Verbindungen begonnen und teile im Folgenden die bis jetzt bei der Untersuchung einiger Chromsalze gewonnenen Resultate mit.

In der Literatur finden sich verschiedene neutrale und basische Formiate und Acetate des Chroms beschrieben, deren Existenz wir nur zum Teil haben bestätigen können. Inwieweit die in der Literatur sich vorfindenden Angaben zuverlässig sind, wird sich aus dem Folgenden ergeben.

Die von uns dargestellten Verbindungen lassen sich in zwei Gruppen einteilen, nämlich a) normale Salze und b) komplexe Salze.

Alfred Werner, *Berichte*, **1908**, *41*, 3447



525. R. F. Weinland: Über Salze einer Acetatochrombase.

[Vorläufige Mitteilung aus dem Chem. Laboratorium der Universität Tübingen.]

(Eingegangen am 15. August 1908.)

In Gemeinschaft mit M. Fiederer hatte ich im Sommer 1907 gefunden, daß sich durch Erwärmen von Chromsäure mit Eisessig zwei gut krystallisierte Verbindungen darstellen lassen, die beide 3- und 6-wertiges Chrom und Essigsäure enthalten. Die Analyse ergab, daß der eine, braunschwarze, Körper auf 3 Atome dreiwertiges Chrom 7 Mol. Essigsäure und 2 Mol. Chromsäure, der andere, dunkelgrüne, auf 3 Atome dreiwertiges Chrom 7 Mol. Essigsäure und 1 Mol. Chromsäure enthielt. In der wäßrigen Lösung der Salze war die Chromsäure durch lösliche Bleisalze direkt nachweisbar, dagegen wurde das dreiwertige Chrom durch Amoniak und Laugen in der Kälte nicht gefällt, erst bei längerem Kochen schied sich Chromihydroxyd aus[1]).

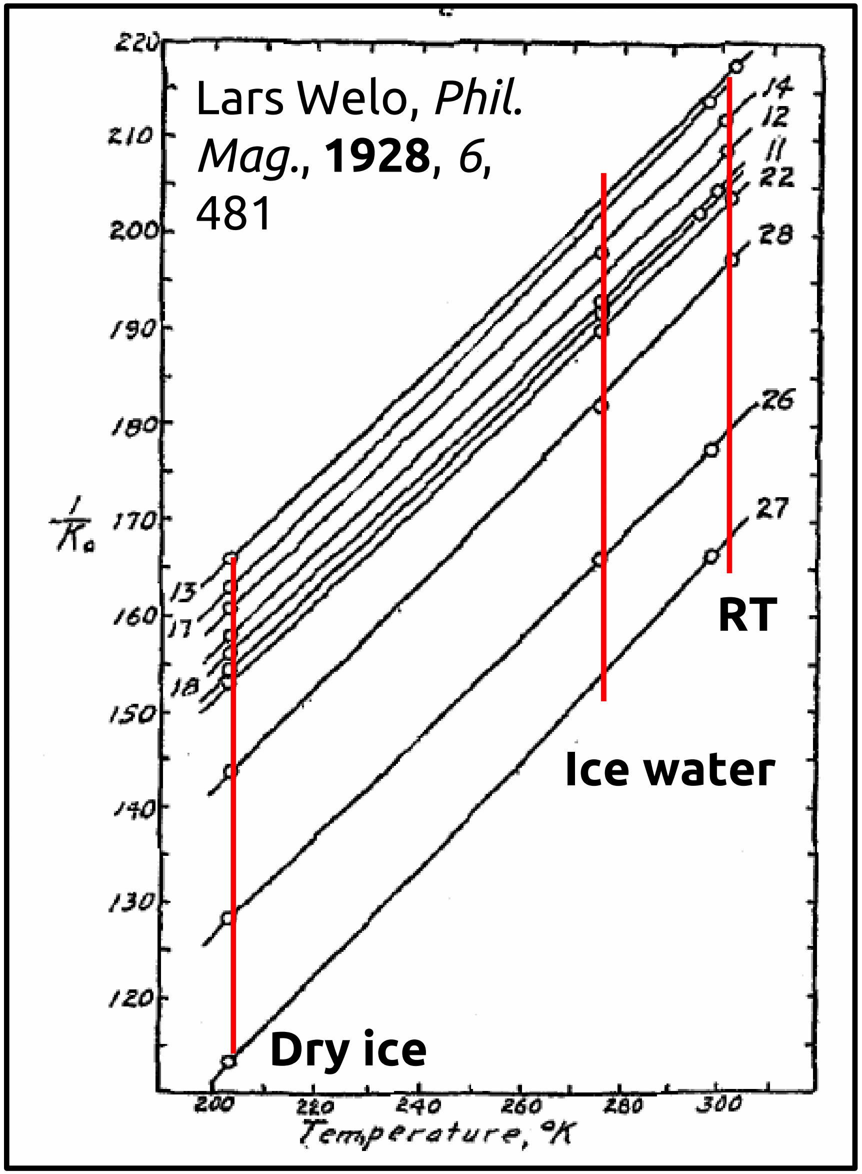
Die weitere, im Verein mit Th

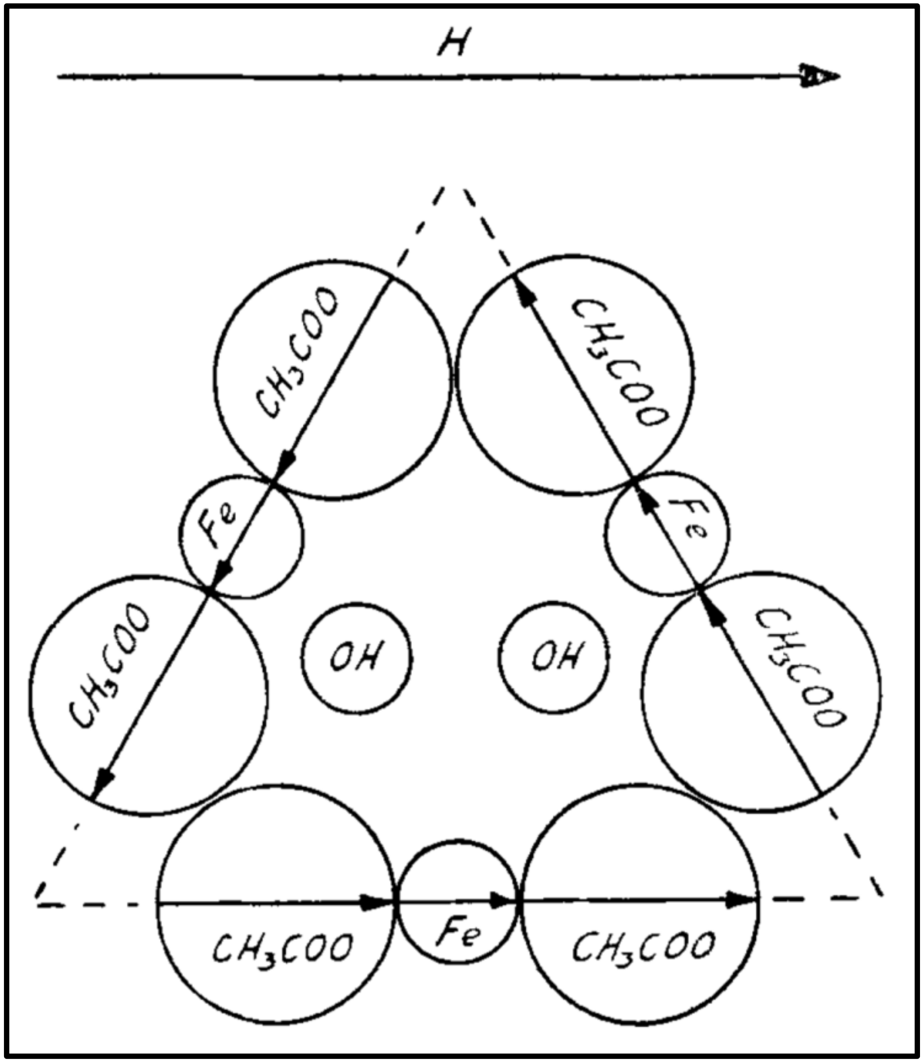
acker ausgeführte Untersuchung der V

mutlich Chromatacetate einer dreisäu

$\Big[Cr_3 \begin{matrix} (CH_3CO. \\ H_2O \end{matrix}$

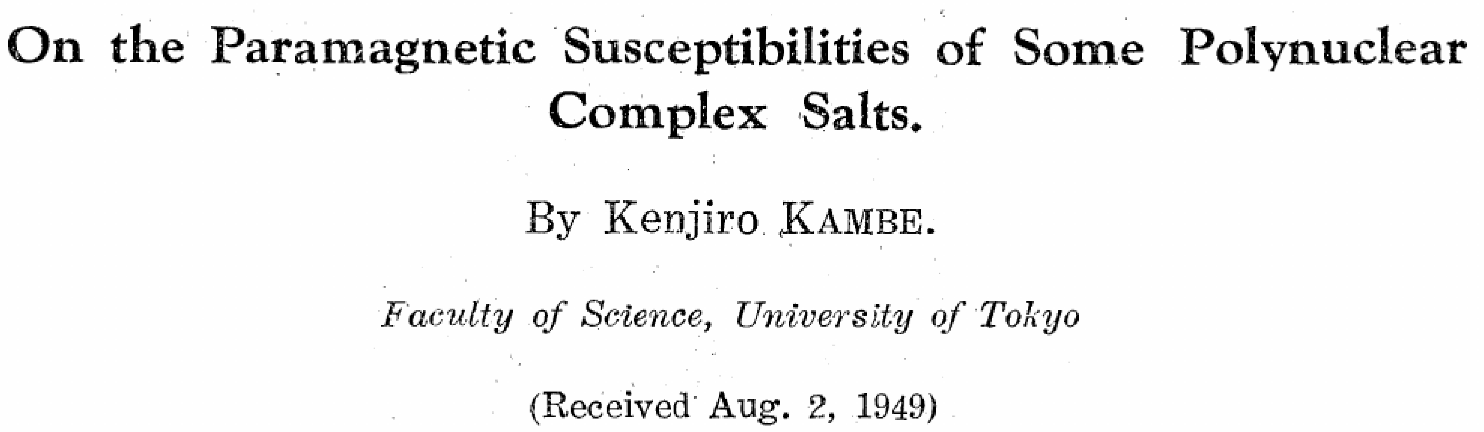
The beginnings of molecular magnetism?


Lars Welo, *Phil. Mag.*, **1928**, *6*, 481



“The problem of negative values of θ […] ***do[es] not appear to be directly related to the existence of polynuclear ions*** *[…] [O]rganic groups, CH_3COO, CHOO, C_6H_5COO, etc., are themselves very probably permanent electric dipoles. […] We may then adopt […] the assumption, that* ***the elementary magnets, that is, the iron and chromium ions, have permanent electric moments as well*** *[…]”*

The beginnings of molecular magnetism?

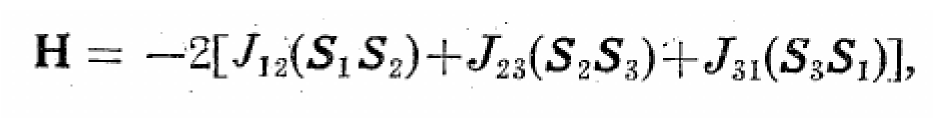


On the Paramagnetic Susceptibilities of Some Polynuclear Complex Salts.

By Kenjiro KAMBE.

Faculty of Science, University of Tokyo

(Received Aug. 2, 1949)



$$\mathbf{H} = -2[J_{12}(S_1S_2)+J_{23}(S_2S_3)+J_{31}(S_3S_1)],$$

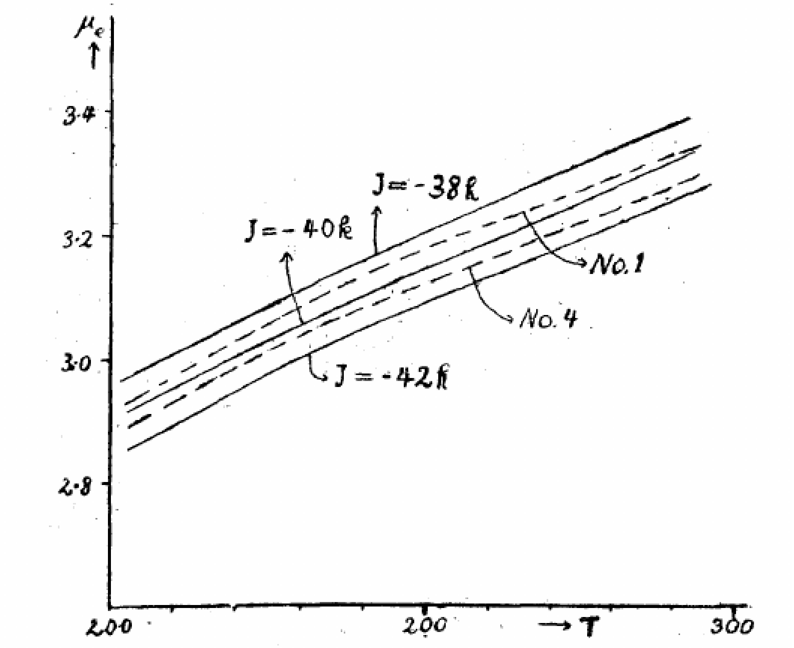
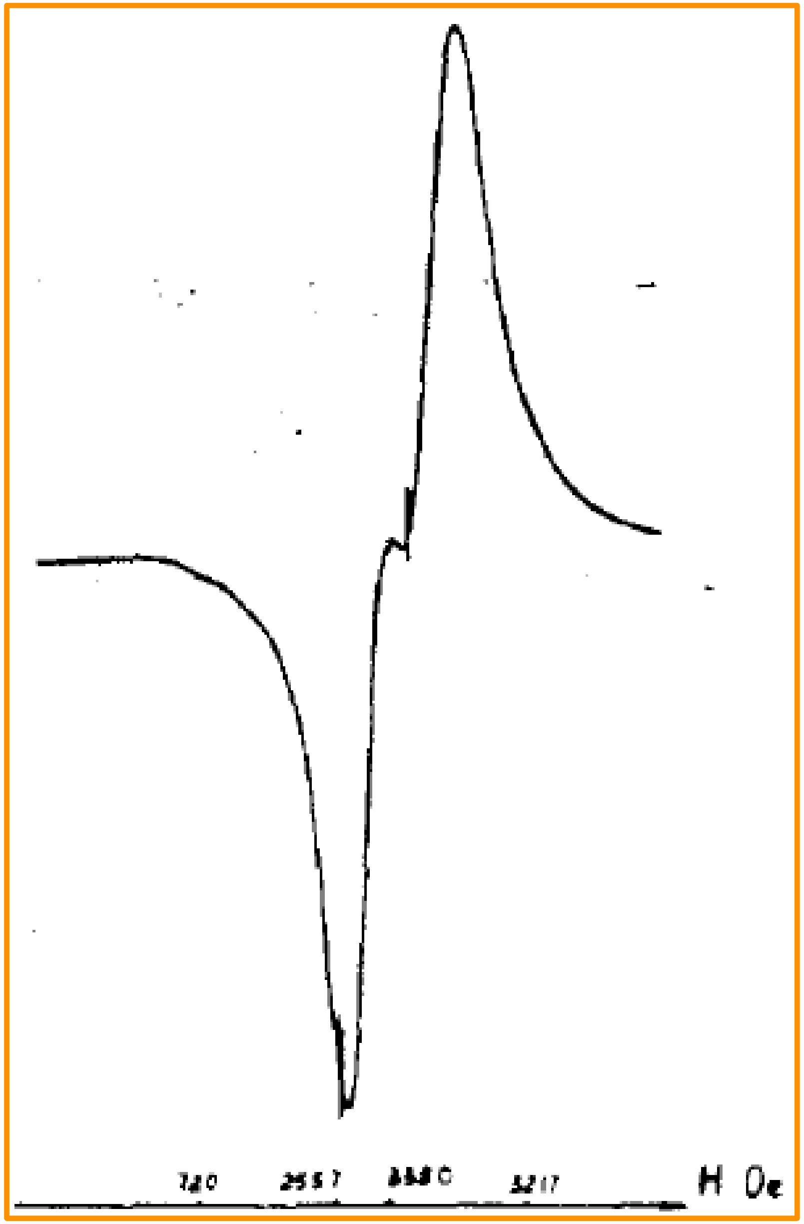


Fig. 1. Temperature variation of effective Bohr magnetons. For $S = 5/2$.

Kenjiro Kambe, J. Phys. Soc. Jpn, **1950**, *5*, 48

Basic magnetic structure

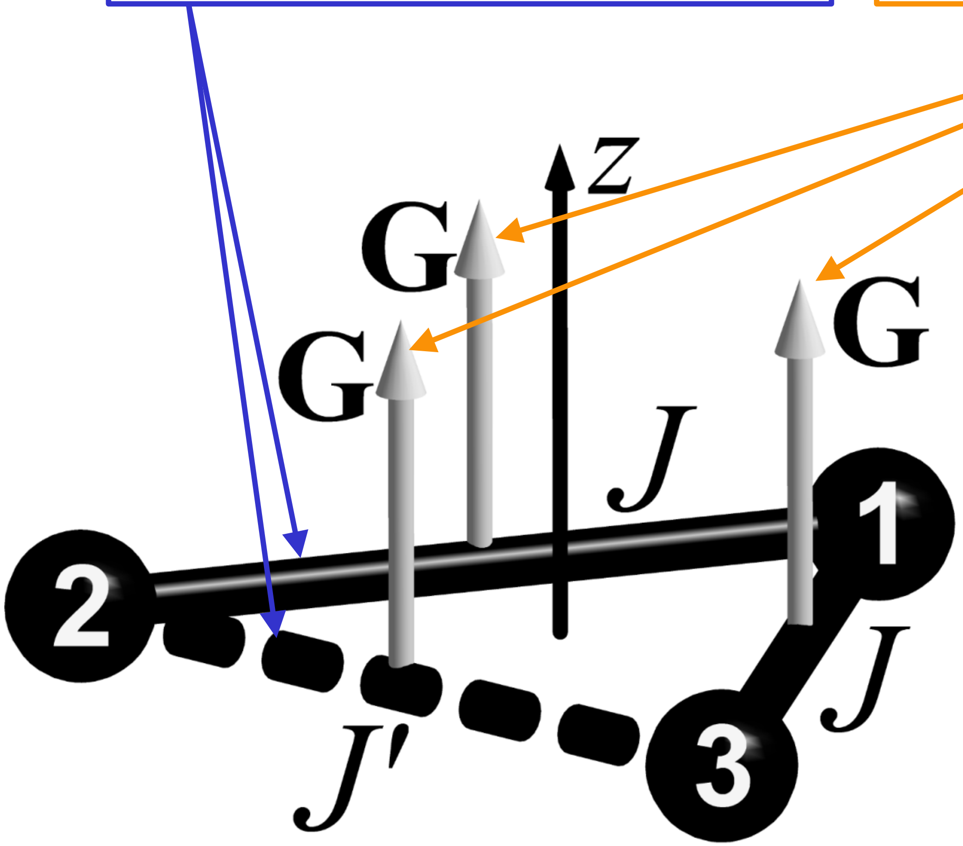


Fiz. Tverd. Tela, **1973**, *15*, 337
Solid State Commun., **1974**, *14*, 131

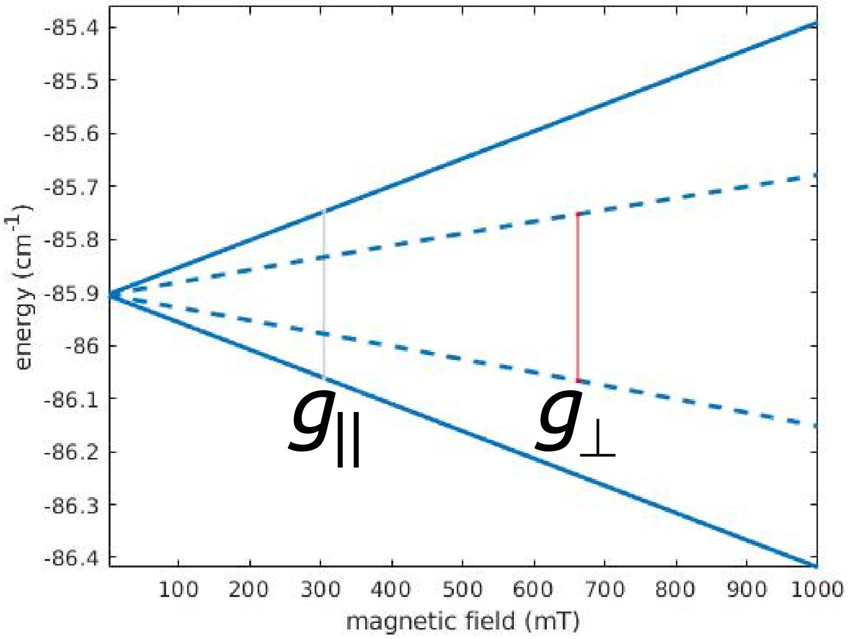
Basic magnetic structure

Heisenberg (isosceles) term + Dzyaloshinskii-Moriya term

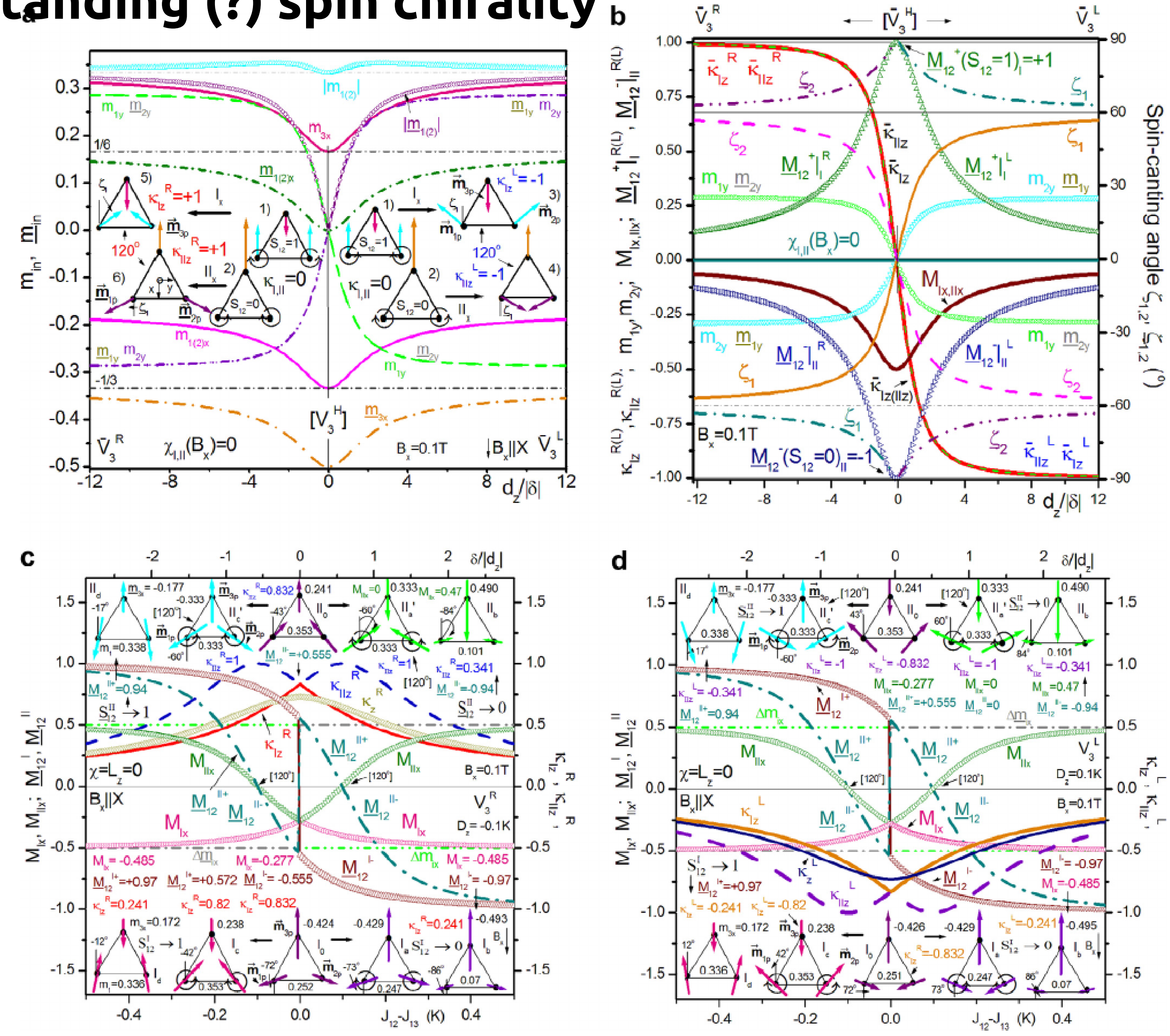
$$\hat{H} = \boxed{-2J(\hat{\mathbf{S}}_1\hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_1\hat{\mathbf{S}}_3) - 2J'\hat{\mathbf{S}}_2\hat{\mathbf{S}}_3} - \boxed{2\mathbf{G}(\hat{\mathbf{S}}_1\times\hat{\mathbf{S}}_2 + \hat{\mathbf{S}}_2\times\hat{\mathbf{S}}_3 + \hat{\mathbf{S}}_3\times\hat{\mathbf{S}}_1)}$$



$J \neq J'$, $\mathbf{G} \neq 0$



Understanding (?) spin chirality

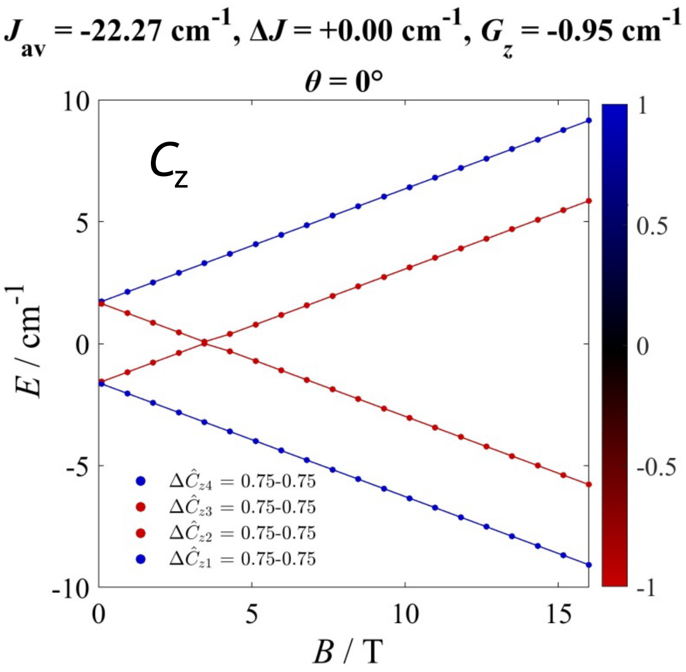


Belinsky, *Chem. Phys.*, **2014**, *435*, 62 & 95; *Inorg. Chem.*, **2016**, *55*, 4078 & 4091

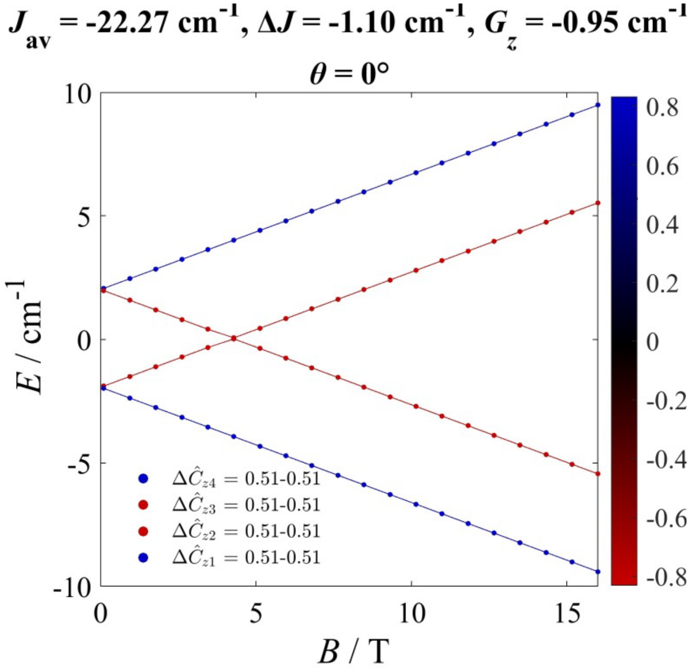
Exploration of spin-chiral states ($S_i = ½$)

- Set up spin Hamiltonian (terms and parameter values)
- Set up operators C_z and $\mathbf{K}_z$
  - Vector $$\hat{\mathbf{K}} = \frac{4}{\sqrt{3}} \sum_{i,j=1}^{3} \hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j \qquad \hat{\mathbf{K}}_z = \frac{4}{\sqrt{3}} \sum_{i,j=1}^{3} \left(\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j \right)_z = \frac{4}{\sqrt{3}} \sum_{i,j=1}^{3} \left(\hat{S}_{ix} \hat{S}_{jy} - \hat{S}_{iy} \hat{S}_{jx} \right)$$
  - Scalar $$\hat{C}_{123,z} = \frac{2}{\sqrt{3}} \boxed{\hat{\mathbf{S}}_1 \cdot (\hat{\mathbf{S}}_2 \times \hat{\mathbf{S}}_3)} \quad \hat{S}_{1x} \cdot \left(\hat{\mathbf{S}}_2 \times \hat{\mathbf{S}}_3 \right)_x + \hat{S}_{1y} \cdot \left(\hat{\mathbf{S}}_2 \times \hat{\mathbf{S}}_3 \right)_y + \hat{S}_{1z} \cdot \left(\hat{\mathbf{S}}_2 \times \hat{\mathbf{S}}_3 \right)_z$$
- Iterate over magnetic field values
  - Calculate expectation values $\langle \hat{O} \rangle$
  - Calculate standard deviations: $\Delta\hat{O} \equiv [\langle \hat{O}^2 \rangle - \langle \hat{O} \rangle^2]^{½}$

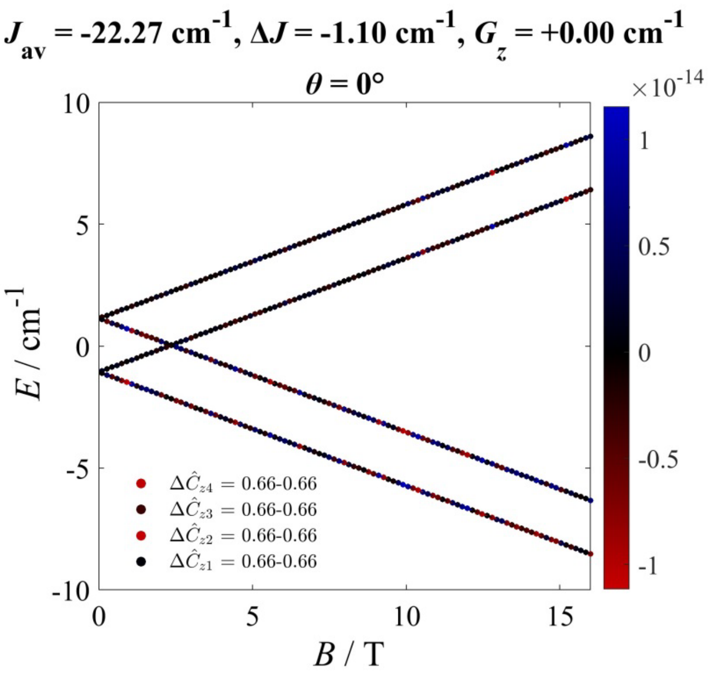
Exploration of spin-chiral states (S_i = ½)


J_{av} = -22.27 cm^{-1}, ΔJ = +0.00 cm^{-1}, G_z = -0.95 cm^{-1}

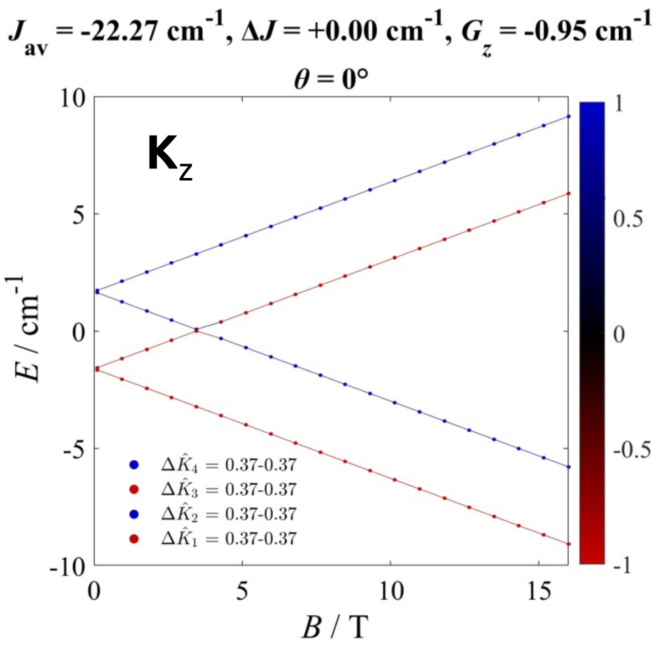
$\theta = 0°$


J_{av} = -22.27 cm^{-1}, ΔJ = -1.10 cm^{-1}, G_z = -0.95 cm^{-1}

$\theta = 0°$


J_{av} = -22.27 cm^{-1}, ΔJ = -1.10 cm^{-1}, G_z = +0.00 cm^{-1}

$\theta = 0°$


J_{av} = -22.27 cm^{-1}, ΔJ = +0.00 cm^{-1}, G_z = -0.95 cm^{-1}

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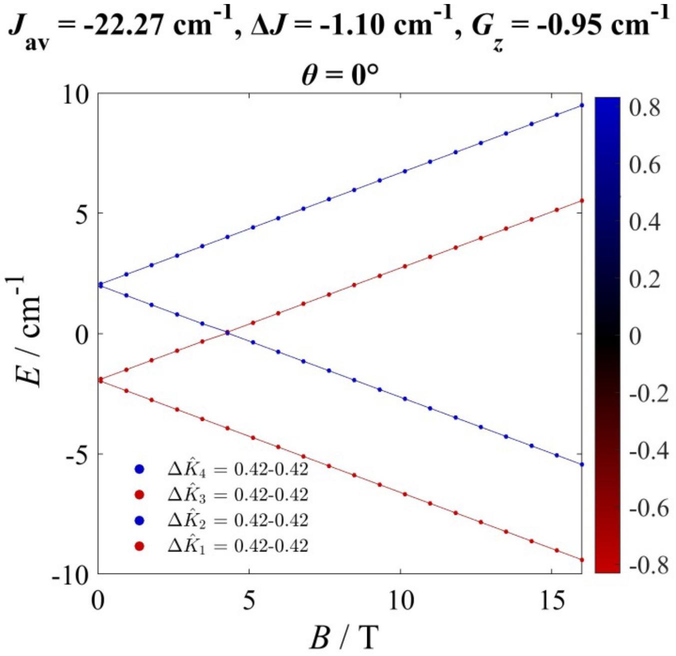
J_{av} = -22.27 cm^{-1}, ΔJ = -1.10 cm^{-1}, G_z = -0.95 cm^{-1}

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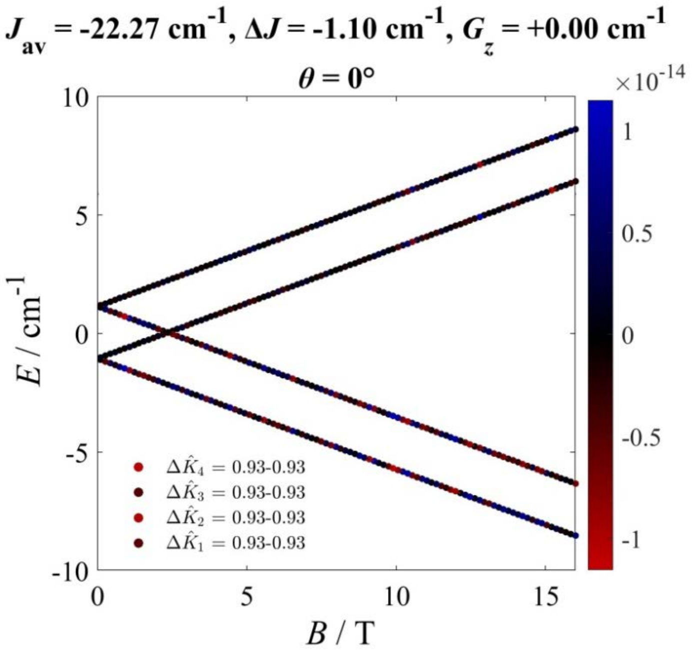
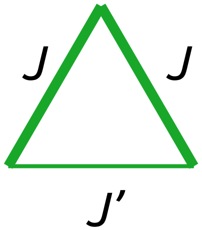
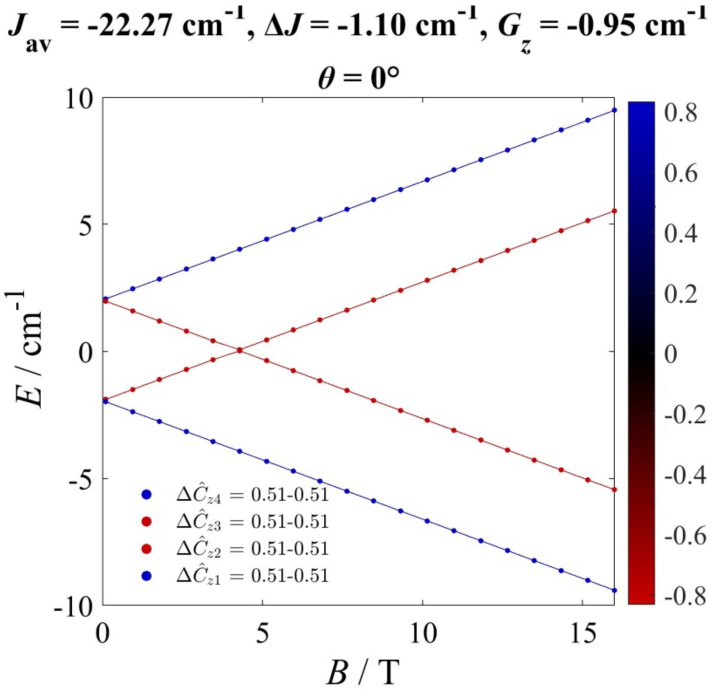

J_{av} = -22.27 cm^{-1}, ΔJ = -1.10 cm^{-1}, G_z = +0.00 cm^{-1}

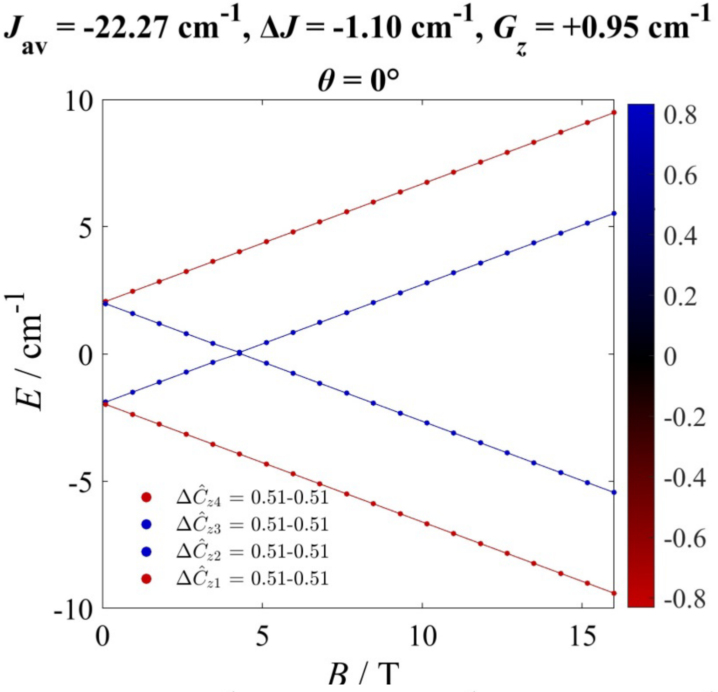
$\theta = 0°$

Exploration of spin-chiral states: sign of variables and C

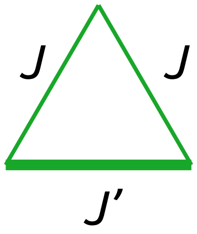


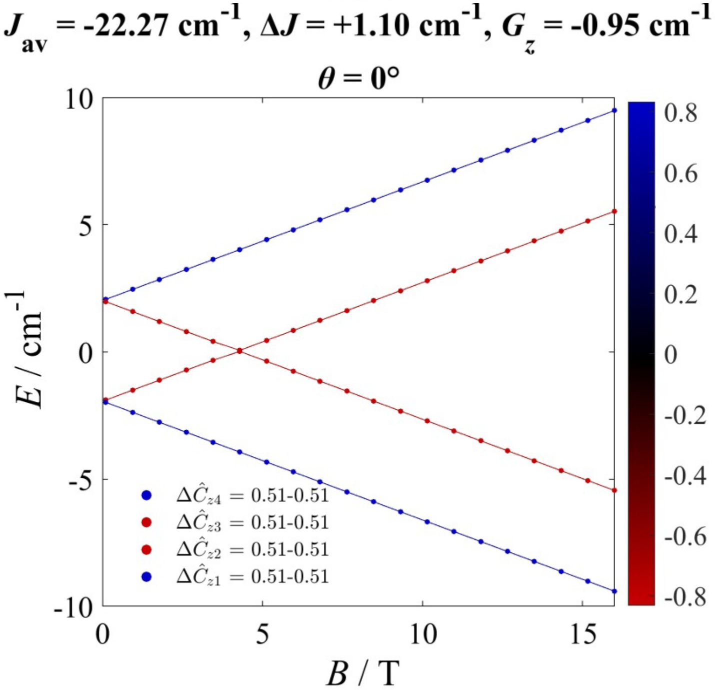

J_{av} = -22.27 cm^{-1}, ΔJ = -1.10 cm^{-1}, G_z = -0.95 cm^{-1}

$\theta = 0°$

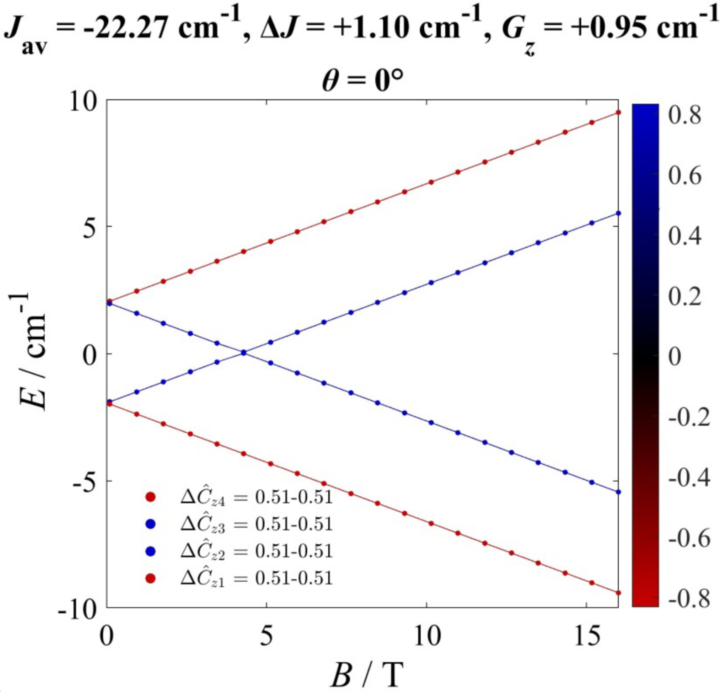

J_{av} = -22.27 cm^{-1}, ΔJ = -1.10 cm^{-1}, G_z = +0.95 cm^{-1}

$\theta = 0°$



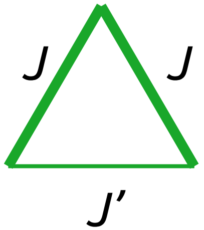

J_{av} = -22.27 cm^{-1}, ΔJ = +1.10 cm^{-1}, G_z = -0.95 cm^{-1}

$\theta = 0°$


J_{av} = -22.27 cm^{-1}, ΔJ = +1.10 cm^{-1}, G_z = +0.95 cm^{-1}

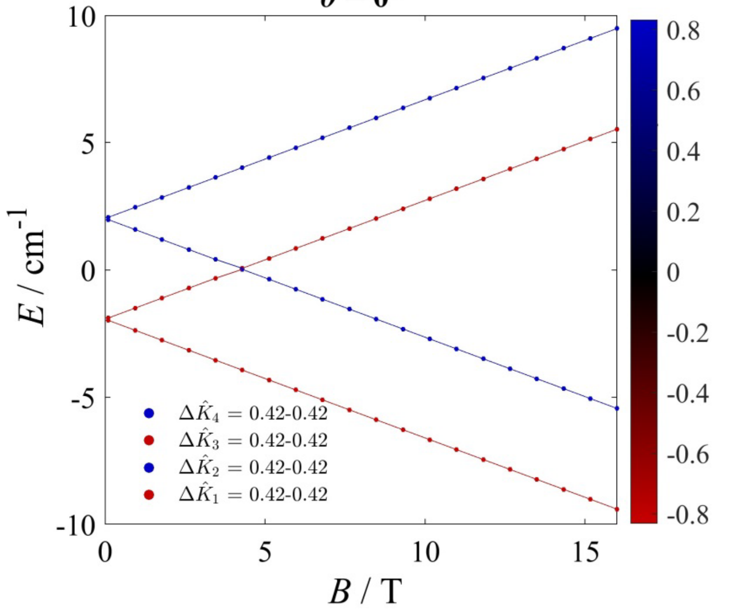
$\theta = 0°$

Exploration of spin-chiral states: sign of variables and K_z



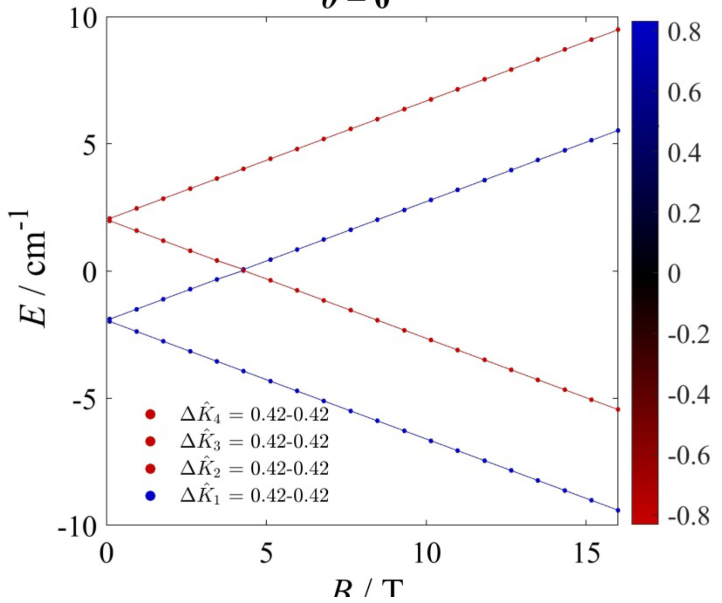
$J_{av} = -22.27\ \mathrm{cm}^{-1}$, $\Delta J = -1.10\ \mathrm{cm}^{-1}$, $G_z = -0.95\ \mathrm{cm}^{-1}$

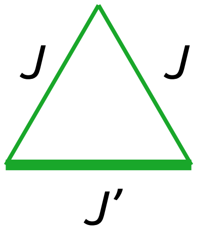
$\theta = 0°$



$J_{av} = -22.27\ \mathrm{cm}^{-1}$, $\Delta J = -1.10\ \mathrm{cm}^{-1}$, $G_z = +0.95\ \mathrm{cm}^{-1}$

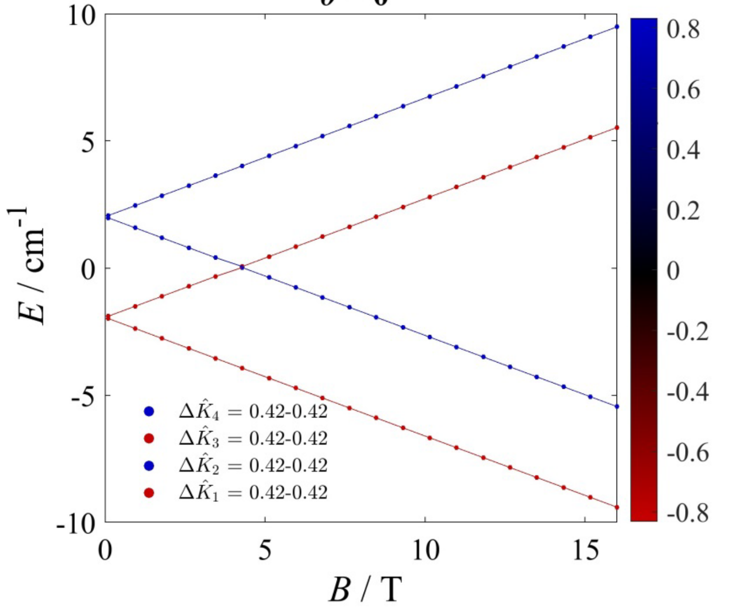
$\theta = 0°$





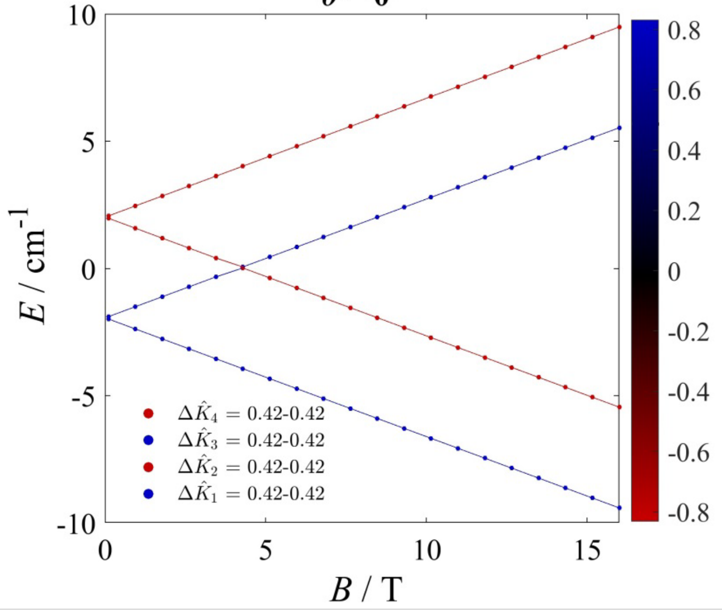
$J_{av} = -22.27\ \mathrm{cm}^{-1}$, $\Delta J = +1.10\ \mathrm{cm}^{-1}$, $G_z = -0.95\ \mathrm{cm}^{-1}$

$\theta = 0°$

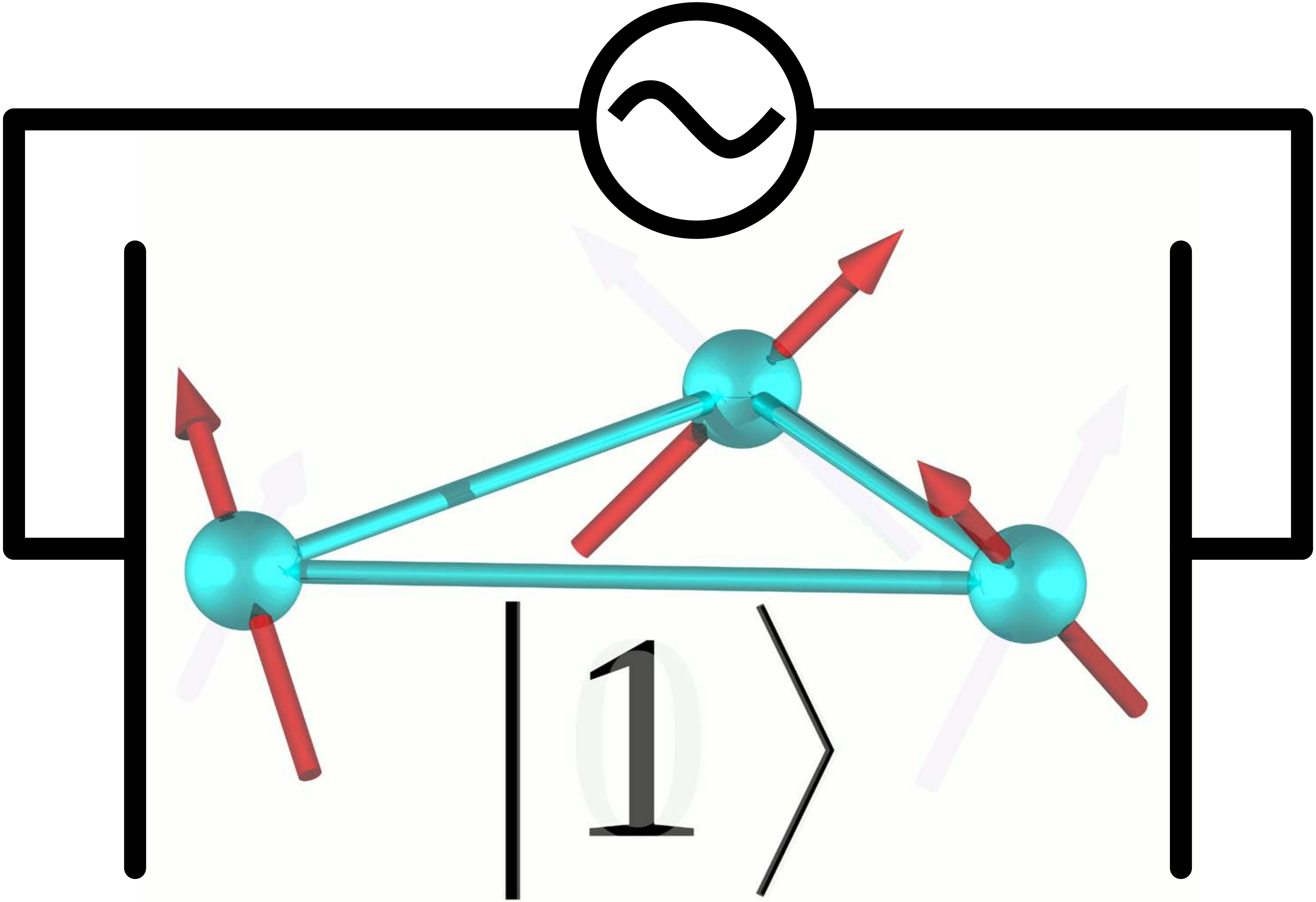


$J_{av} = -22.27\ \mathrm{cm}^{-1}$, $\Delta J = +1.10\ \mathrm{cm}^{-1}$, $G_z = +0.95\ \mathrm{cm}^{-1}$

$\theta = 0°$



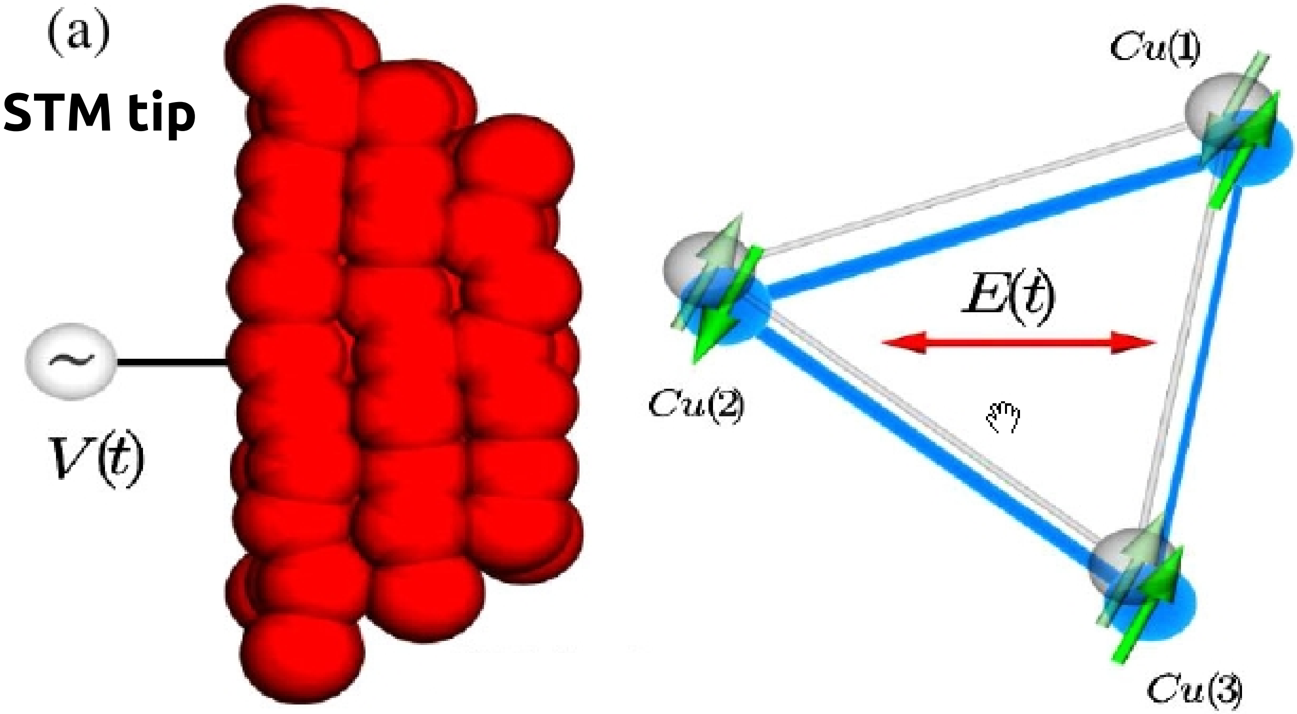
Spin-chiral transitions and magnetoelectricity



Excited by AC electric fields

Spin-chiral transitions and magnetoelectricity

Electric control



Trif et al., *Phys. Rev. Lett.*, **2008**, *101*, 217201

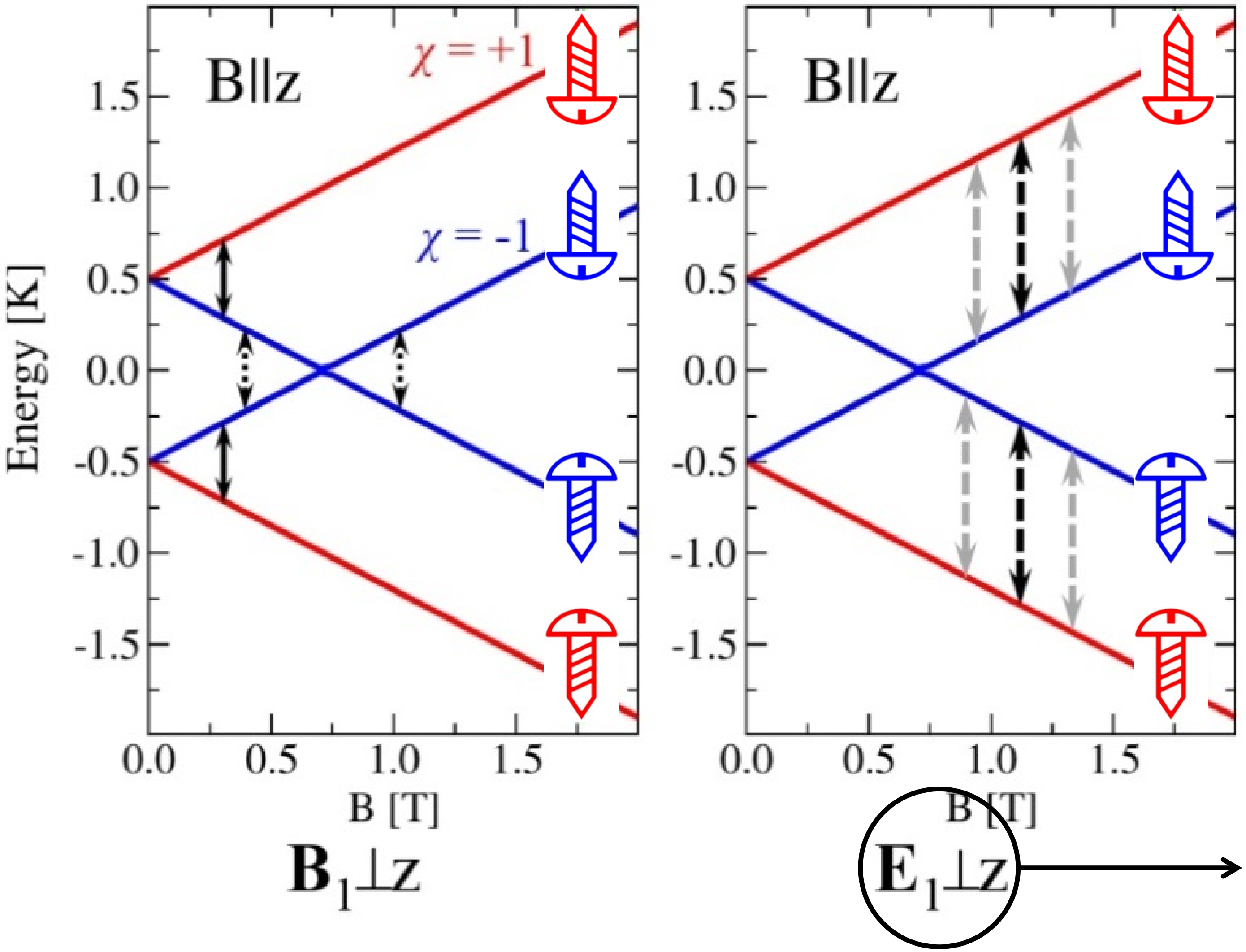
Trif et al., *Phys. Rev. B*, **2010**, *82*, 045429

Spin-chiral transitions and magnetoelectricity

Slow decoherence

Spin (projection) qubit

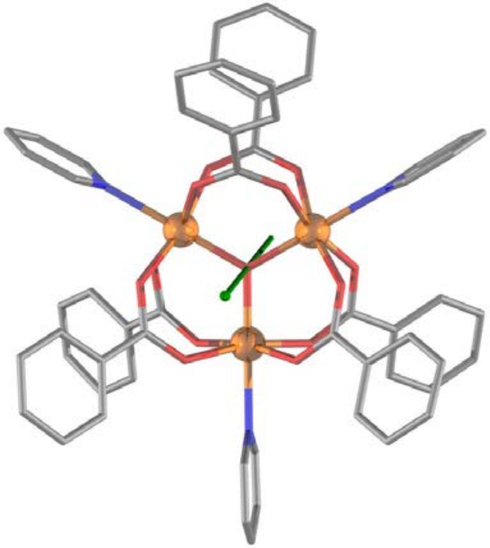
Spin chirality qubit



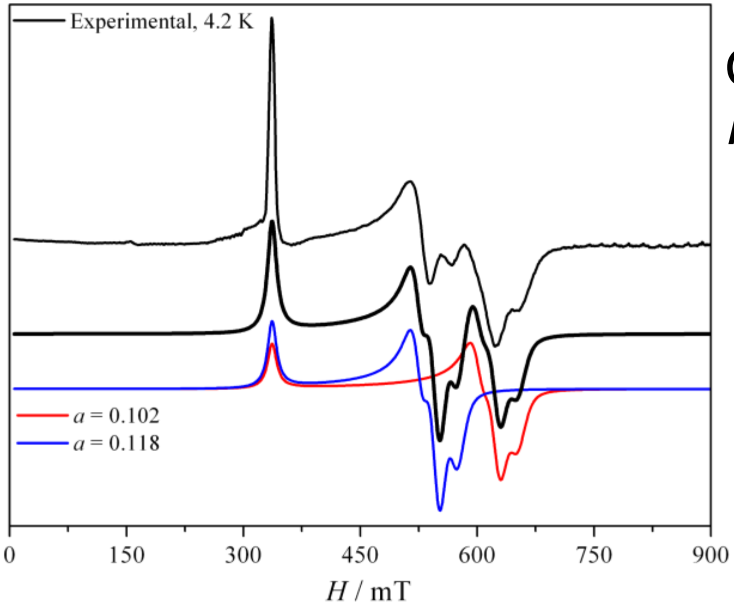
→ Magnetoelectric coupling

High-symmetry triangles (D_{3h})

Fe^{III}, Cr^{III}: same structure two different stories



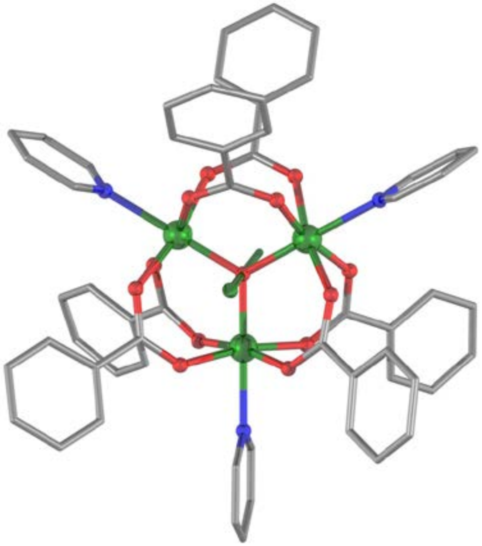
$[Fe_3O(O_2CPh)_6(py)_3]ClO_4{\cdot}py$



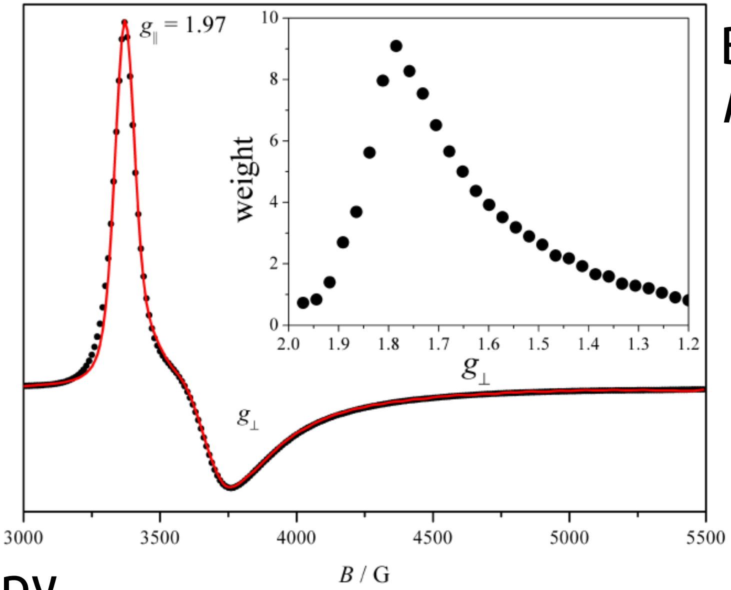
Georgopoulou et al.,
Inorg. Chem., **2017**, *56*, 762

G_z^{Fe} **~ 1.6-1.7 cm⁻¹**

ΔJ ~ 5 – 6 cm⁻¹



$[Cr_3O(O_2CPh)_6(py)_3]ClO_4{\cdot}0.5py$



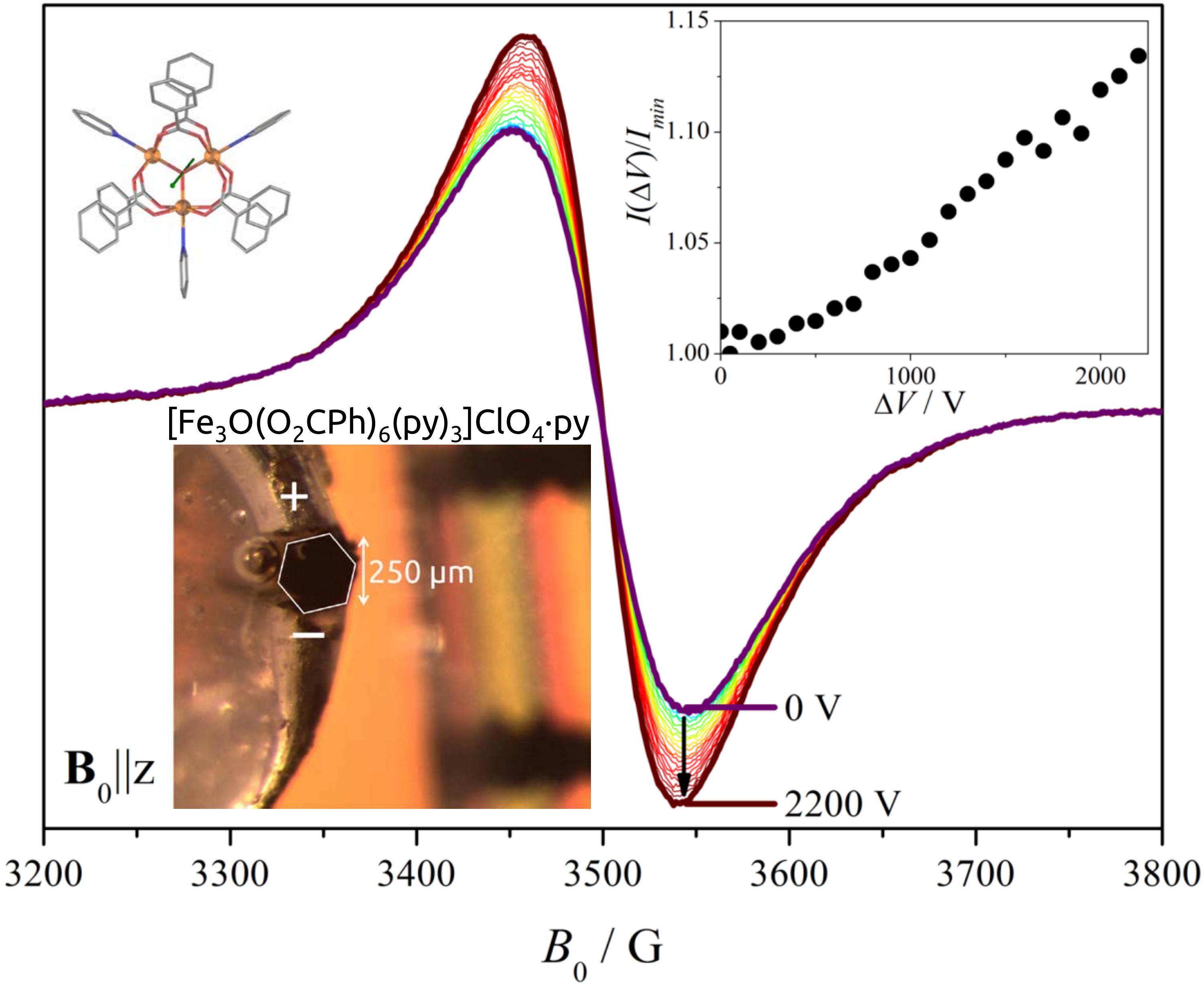
Boudalis et al.,
Inorg. Chem., **2018**, *57*, 13259

G_z^{Cr} ~ 0.041 cm⁻¹

ΔJ ~ 0.3 cm⁻¹

Magnetoelectric coupling in the Fe^{III} triangle

Under a static E-field



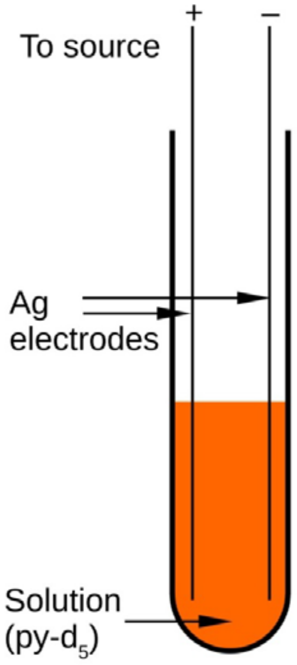
www.chemeurj.org

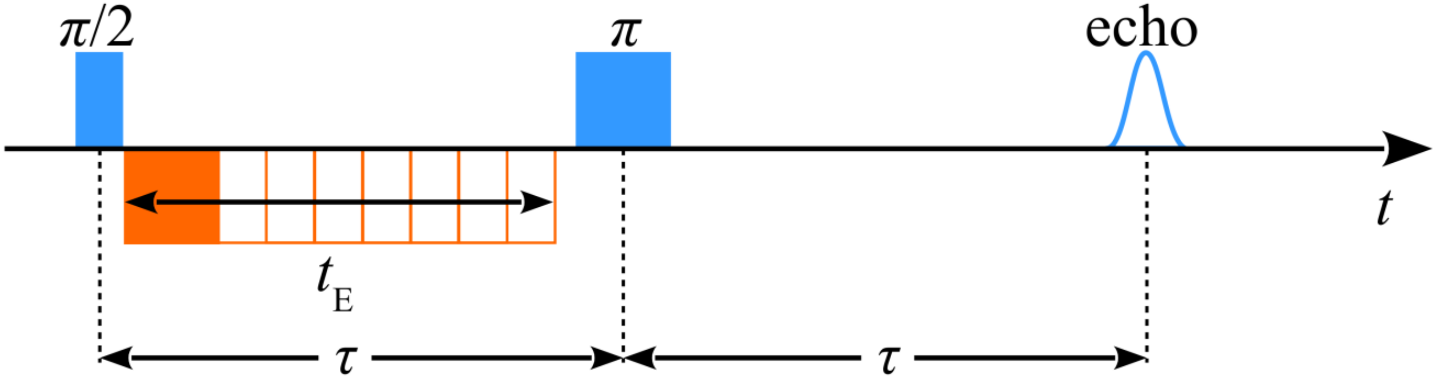
2018-24/56

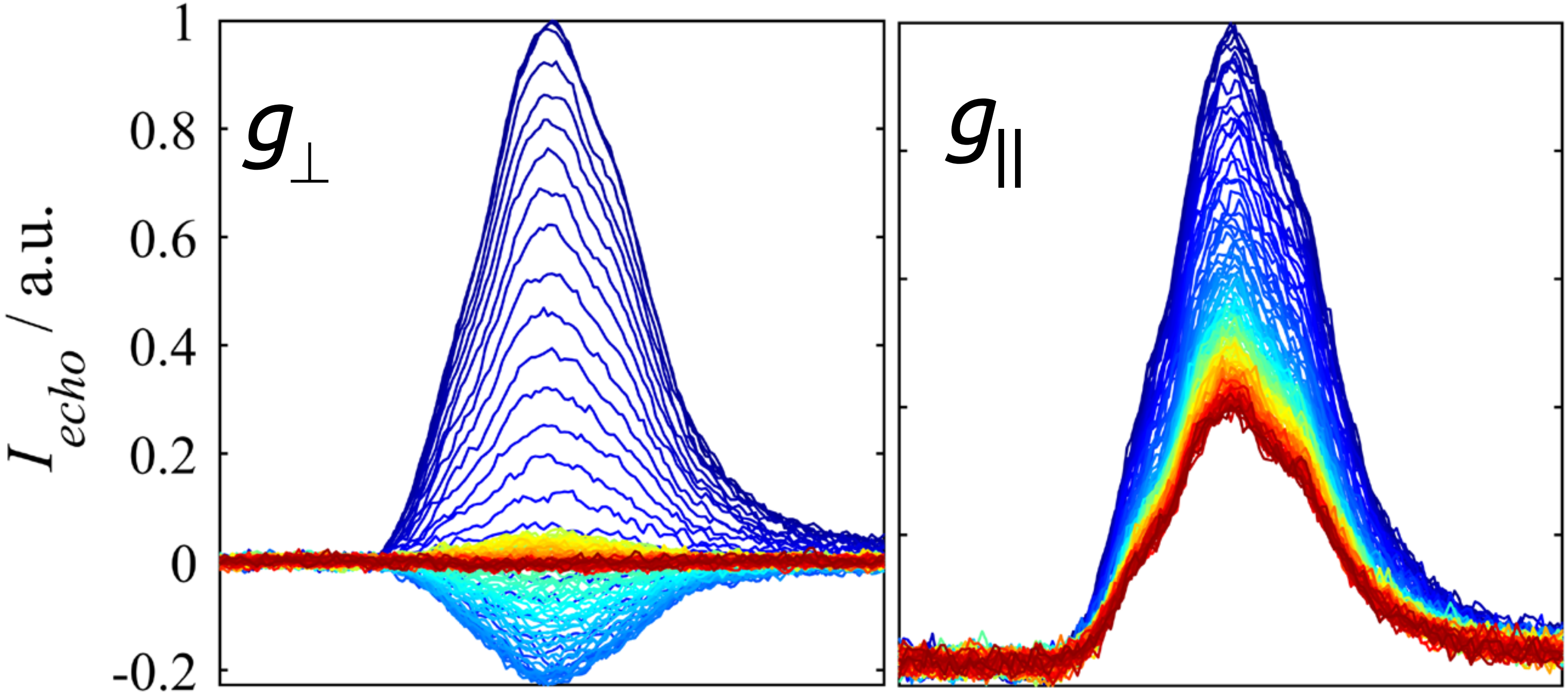
Cover Feature:
A. K. Boudalis et al.
First Demonstration of Magnetoelectric Coupling
in a Polynuclear Molecular Nanomagnet: Single-Crystal EPR Studies
of $[Fe_3O(O_2CPh)_6(py)_3]ClO_4$·py under Static Electric Fields

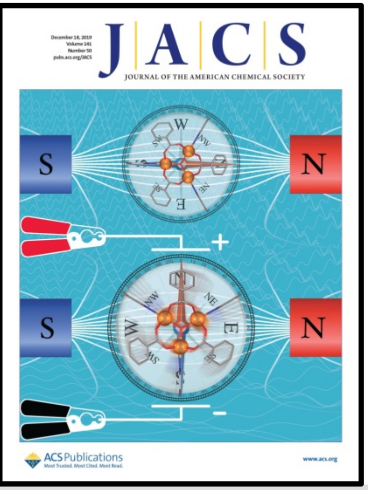
Boudalis et al., *Chem. Eur. J.*, **2018**, *24*, 14896

Magnetoelectric coupling in the Fe^{III} triangle





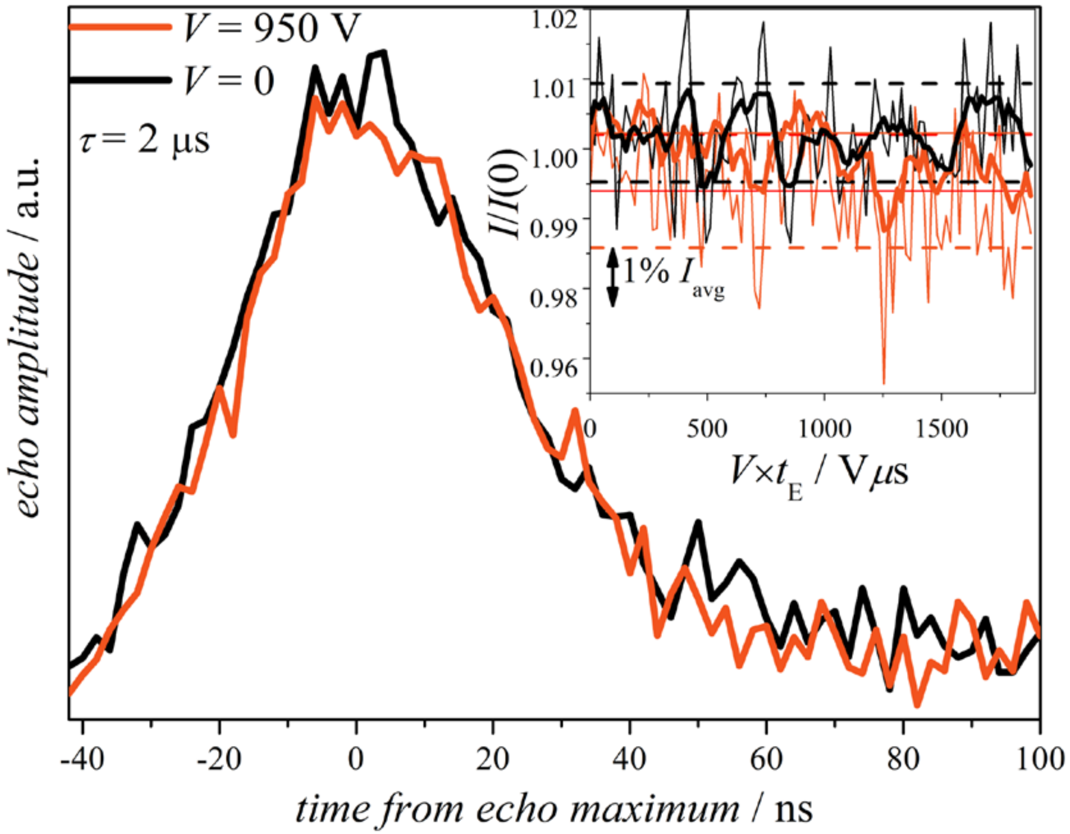


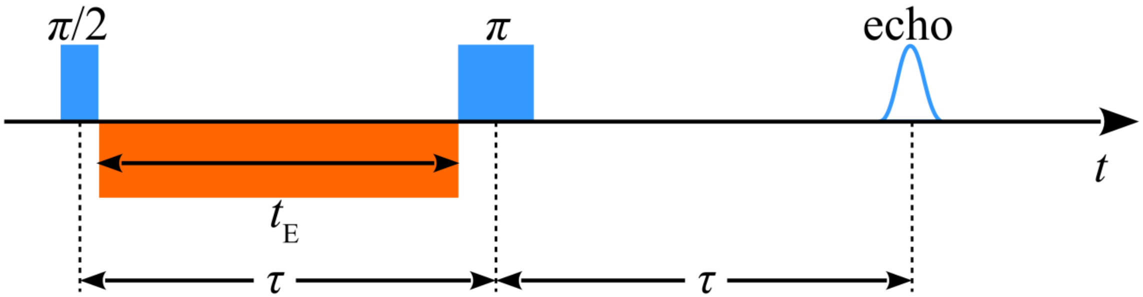


Boudalis et al., *J. Am. Chem. Soc.*, **2019**, *41*, 19765

Magnetoelectric coupling in the $\mathrm{Cr^{III}}$ triangle

None…





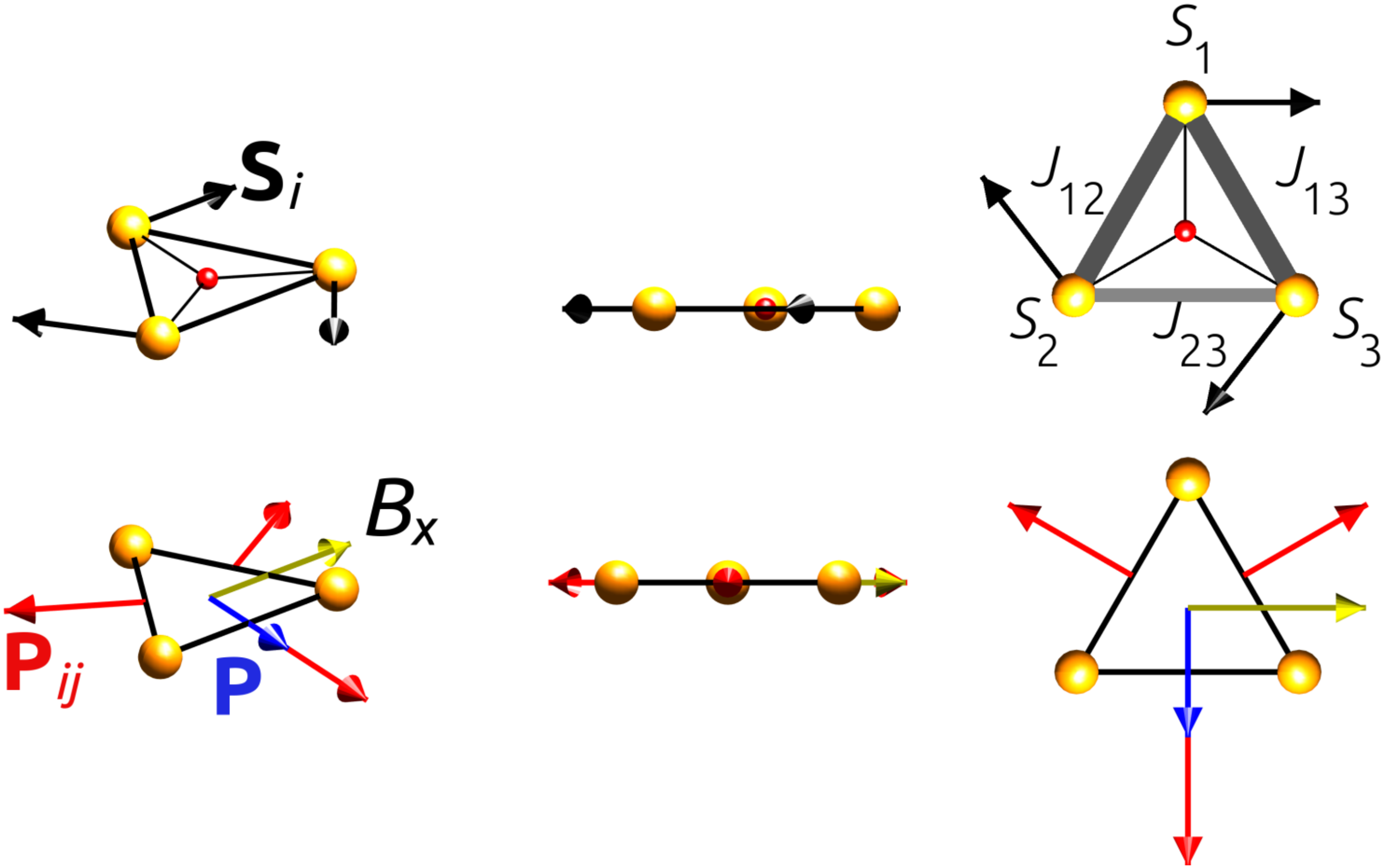
Boudalis et al., *J. Am. Chem. Soc.*, **2019**, *41*, 19765

Origins of ME coupling

Simulations per the KNB model

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

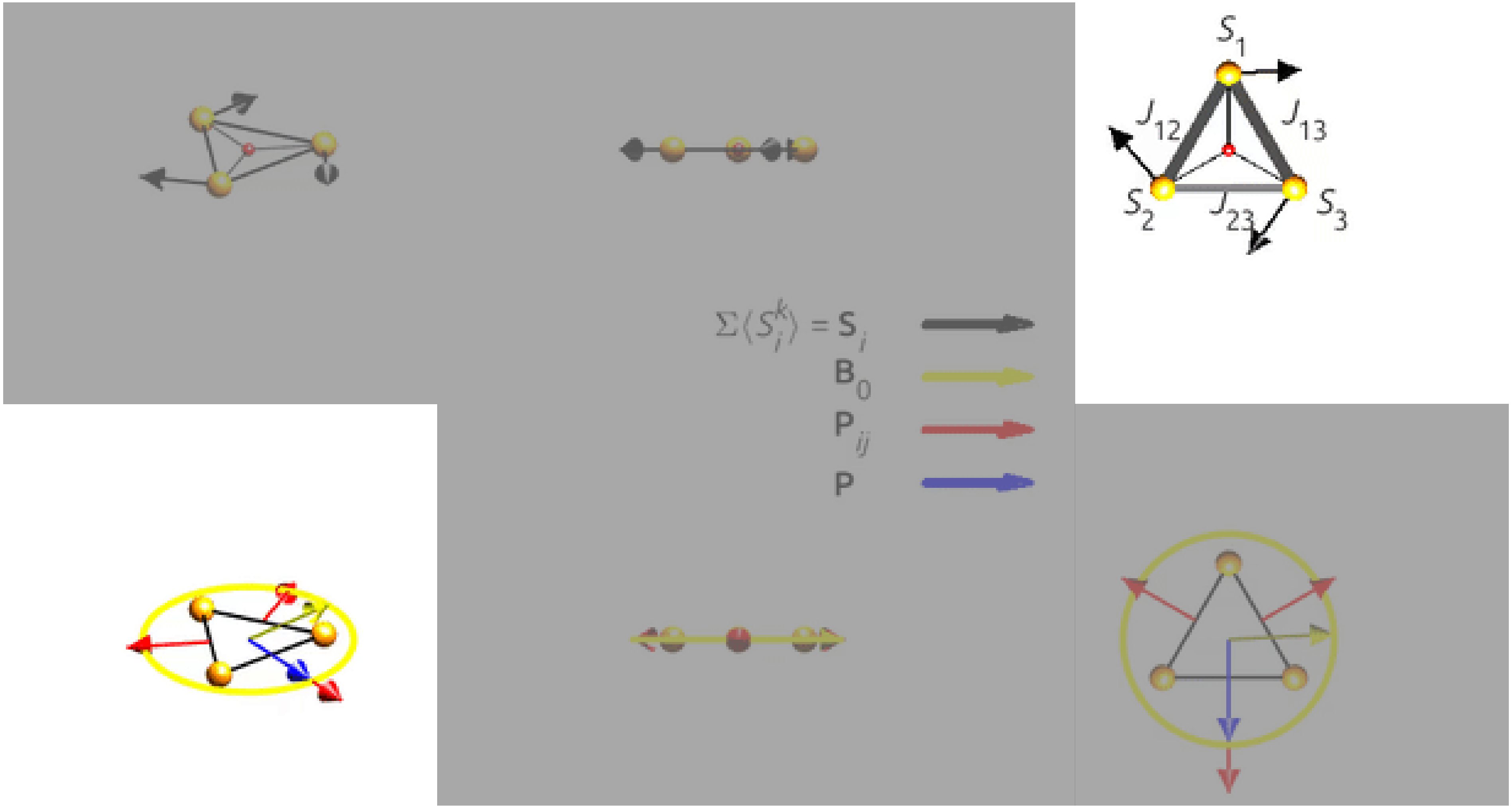
(Katsura-Nagaosa-Balatsky, *PRL*, **2005**, *95*, 057205)



Boudalis et al., *J. Am. Chem. Soc.*, **2019**, *41*, 19765

Origins of ME coupling

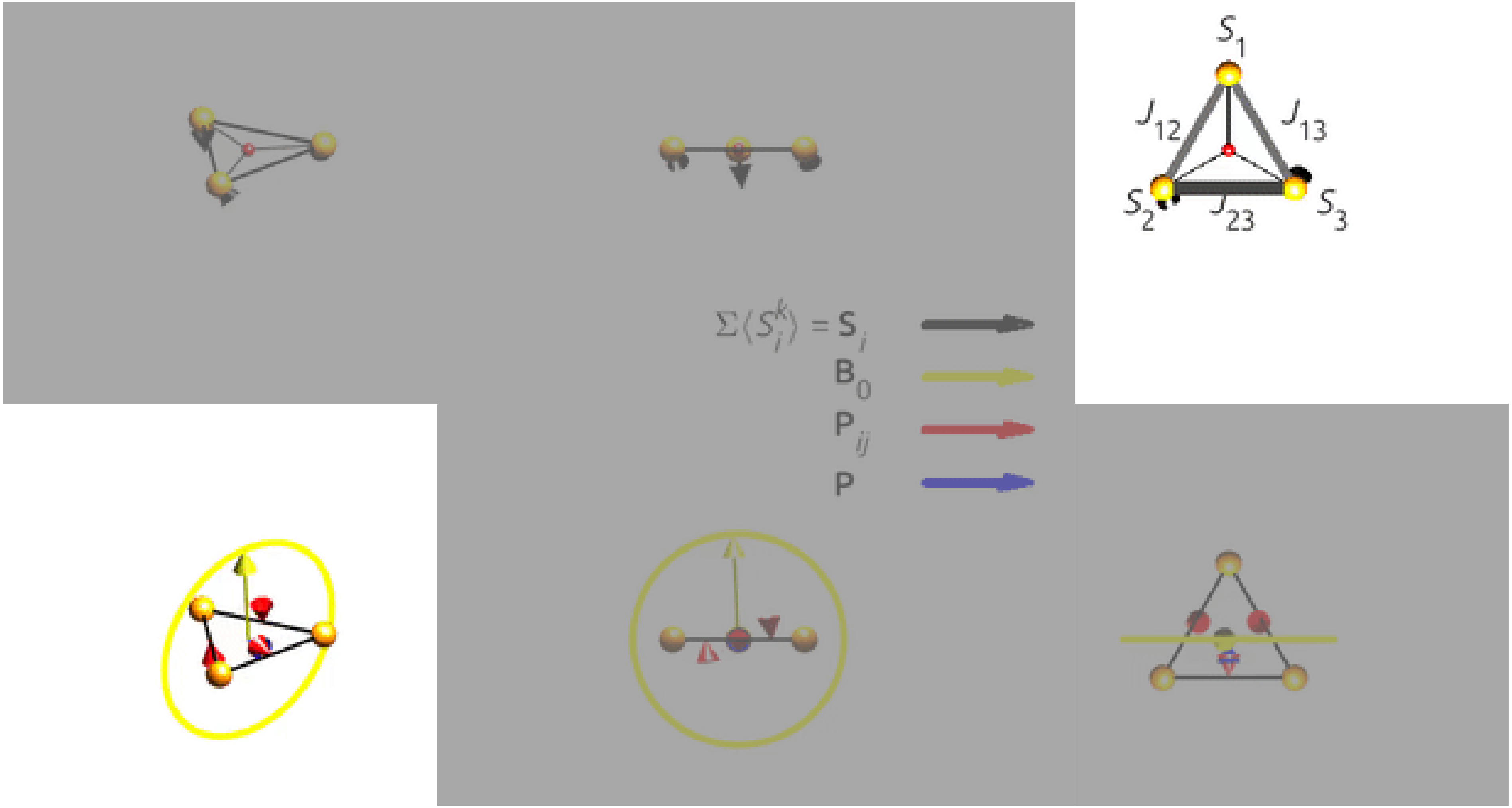
Simulations per the KNB model



Boudalis et al., *J. Am. Chem. Soc.*, **2019**, *41*, 19765

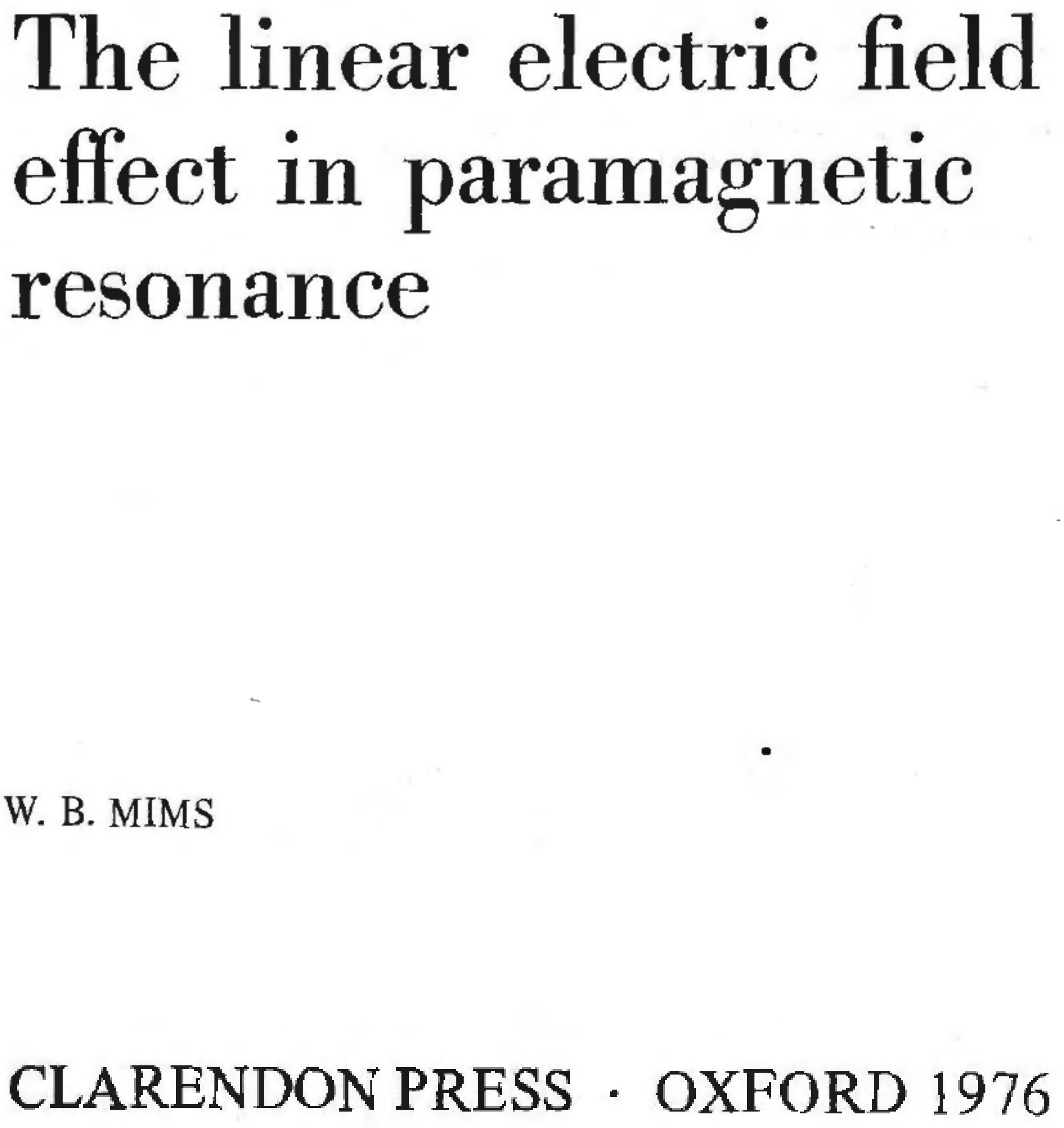
Origins of ME coupling

Simulations per the KNB model



Boudalis et al., *J. Am. Chem. Soc.*, **2019**, *41*, 19765

Other origins of ME coupling?



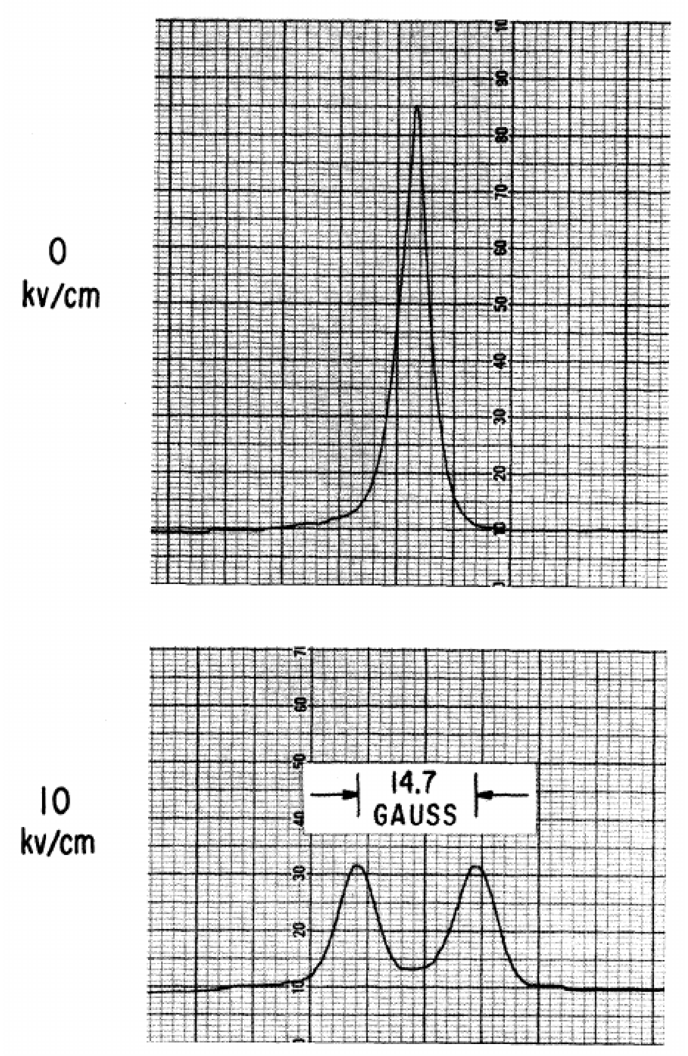
The linear electric field effect in paramagnetic resonance

W. B. MIMS

CLARENDON PRESS · OXFORD 1976

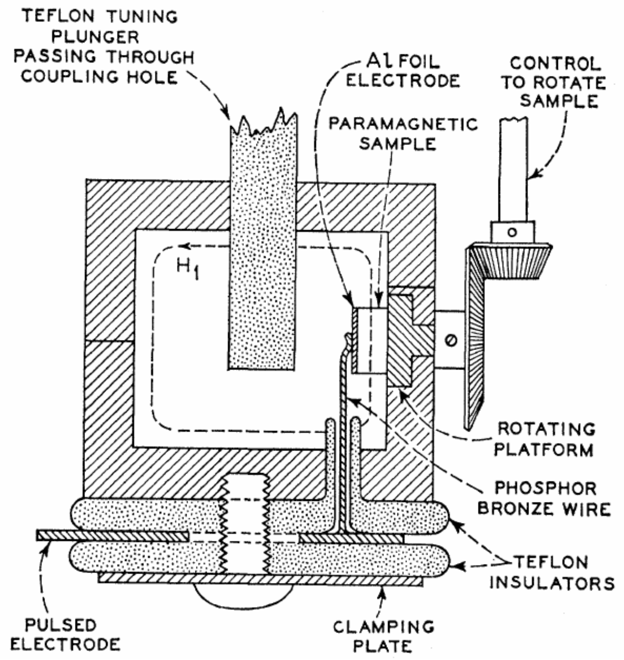
Other origins of ME coupling?

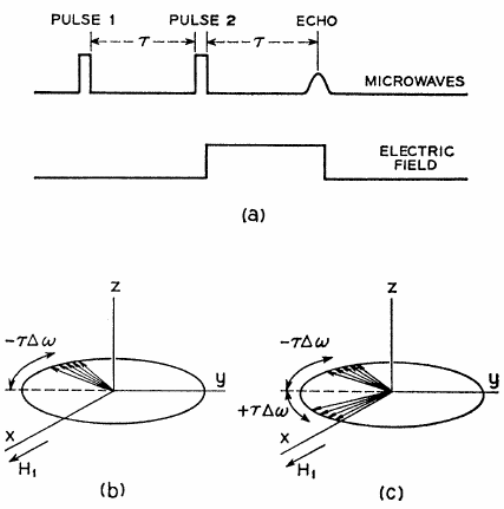
Fe^{I}:Si

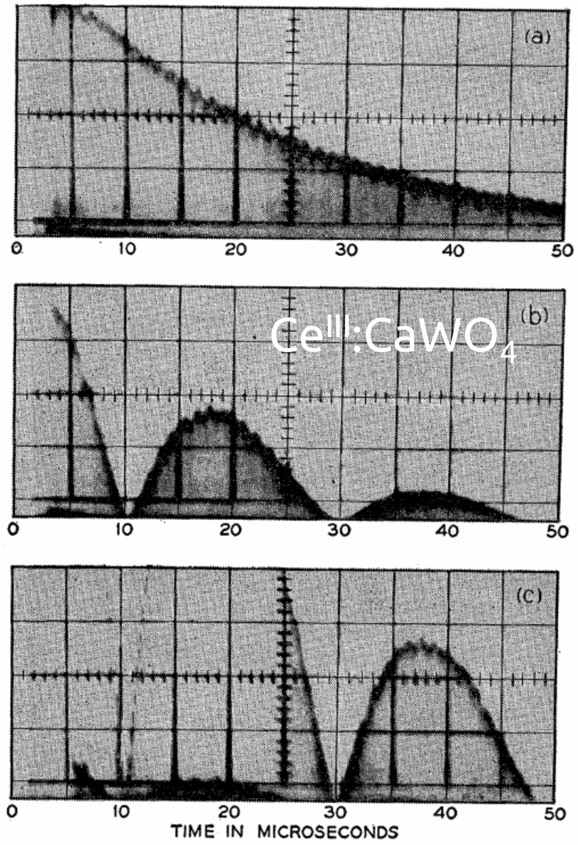


Ludwig & Woodberry, *PRL*, **1961**, *7*, 240

Other origins of ME coupling?

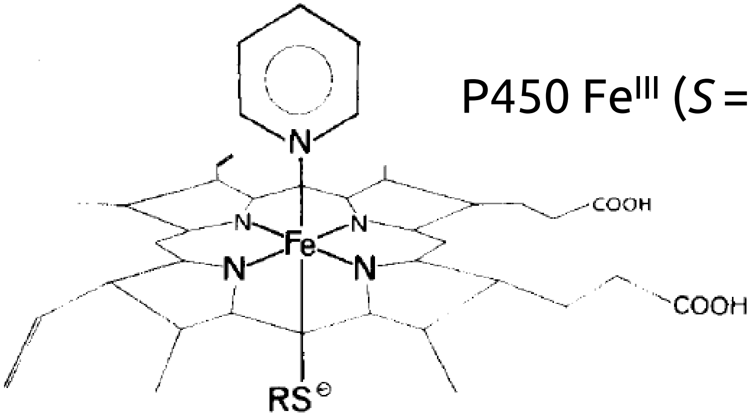






Ce^{III}:$CaWO_4$

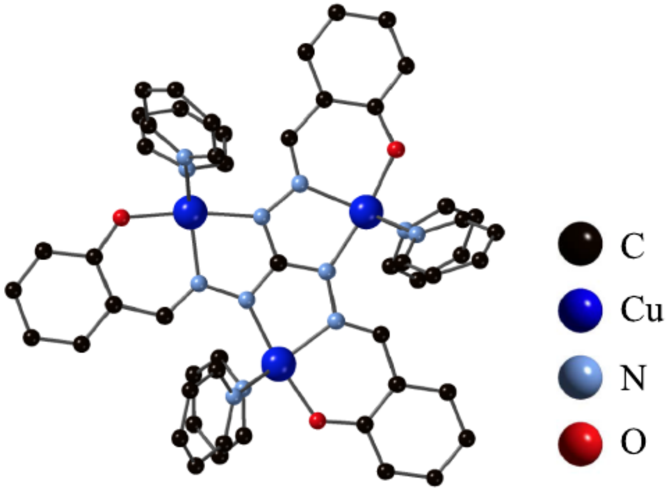
Mims, Phys Rev, **1964**, *133*, A835



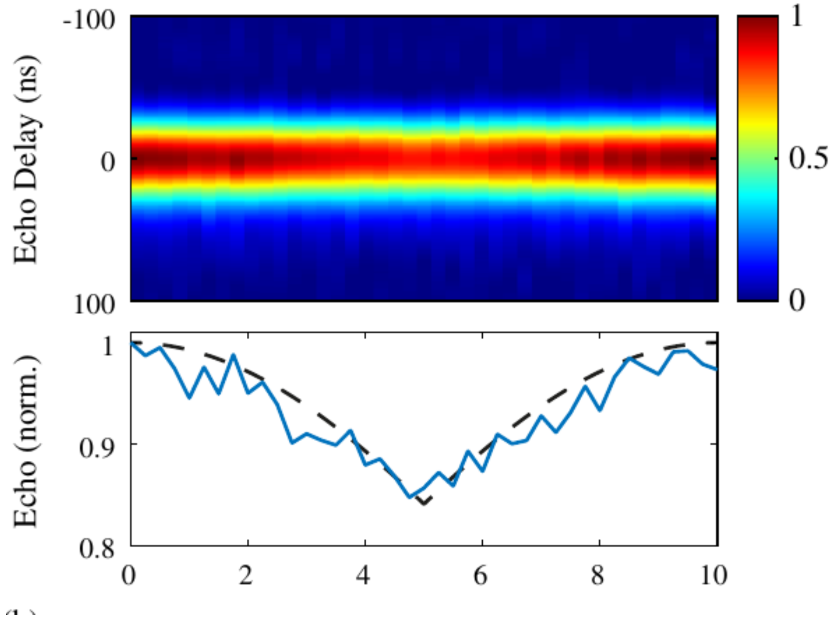
P450 Fe^{III} (S = ½)

Peisach & Mims, *PNAS*, **1973**, *70*, 2979
Mims & Peisach, *J. Chem. Phys.*, **1976**, *64*, 1074

Other origins of ME coupling?

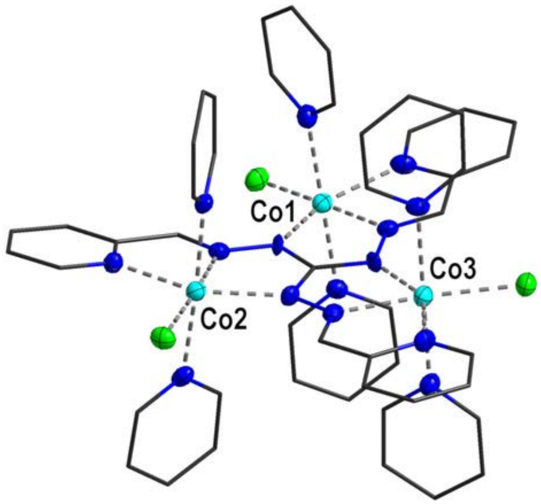


$[Cu_3(saltag)(py)_6]ClO_4$

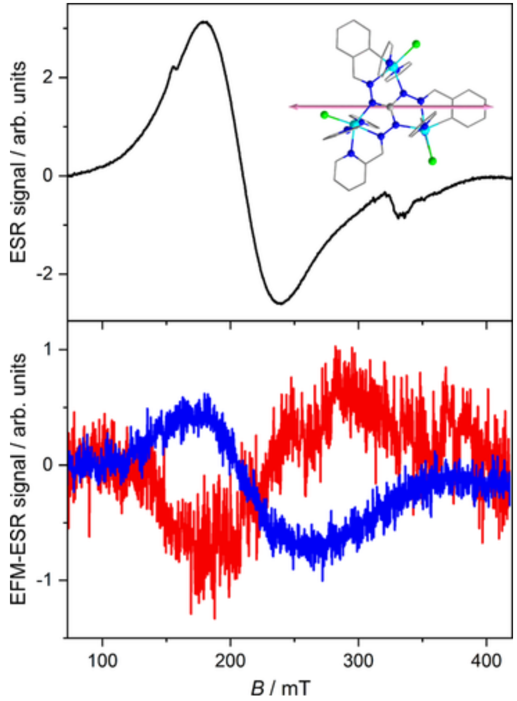


PRL, **2019**, *122*, 037202

Spin frustration removal (?)



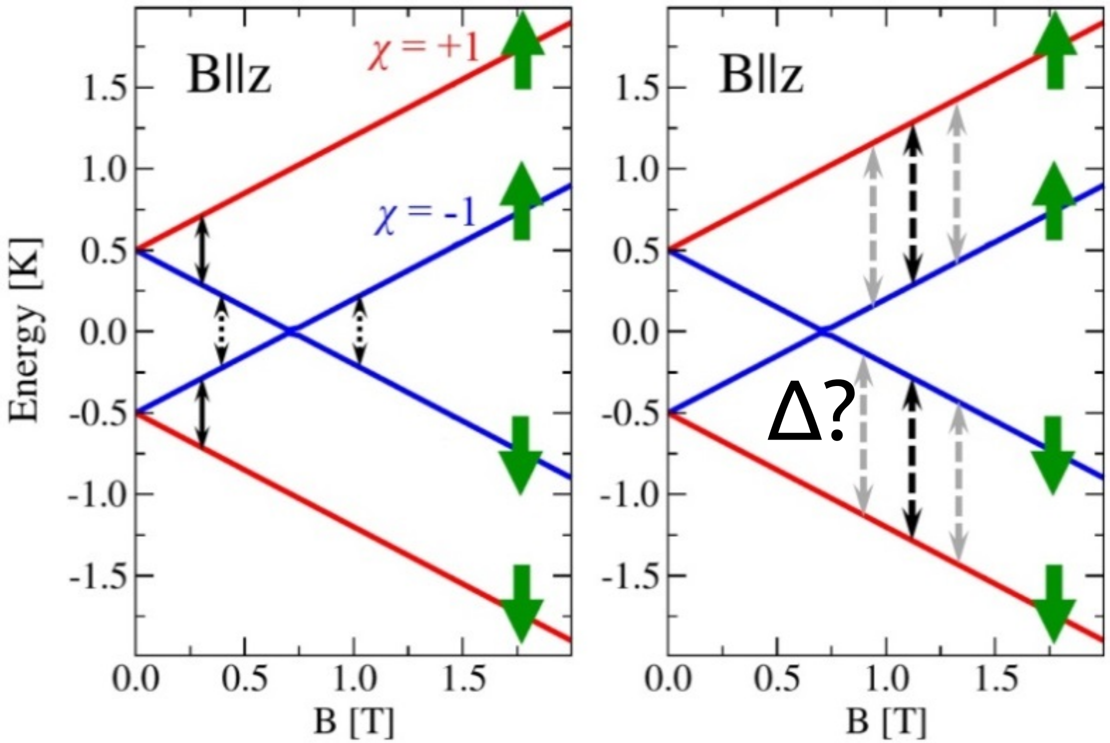
$[Co_3(pytag)(py)_6Cl_3]ClO_4 \cdot 3$ py



Angew. Chem. Int. Ed., **2021**, *60*, 8832

SOC-mediated *g*-factor modulation ?

Spin-electric excitations



$B_1 \perp z$ $E_1 \perp z$

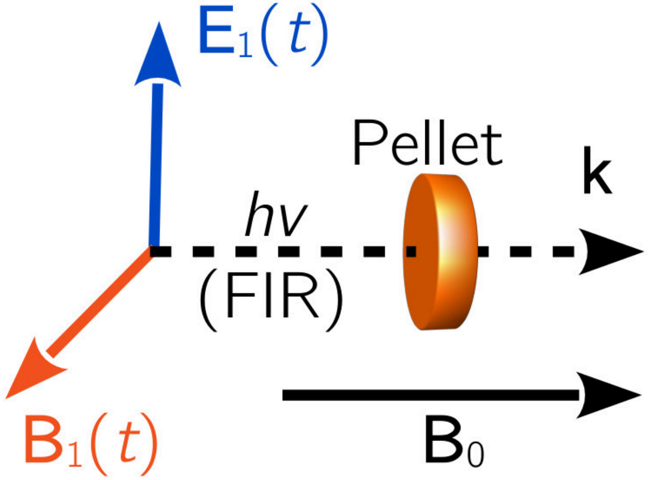
Fe_3

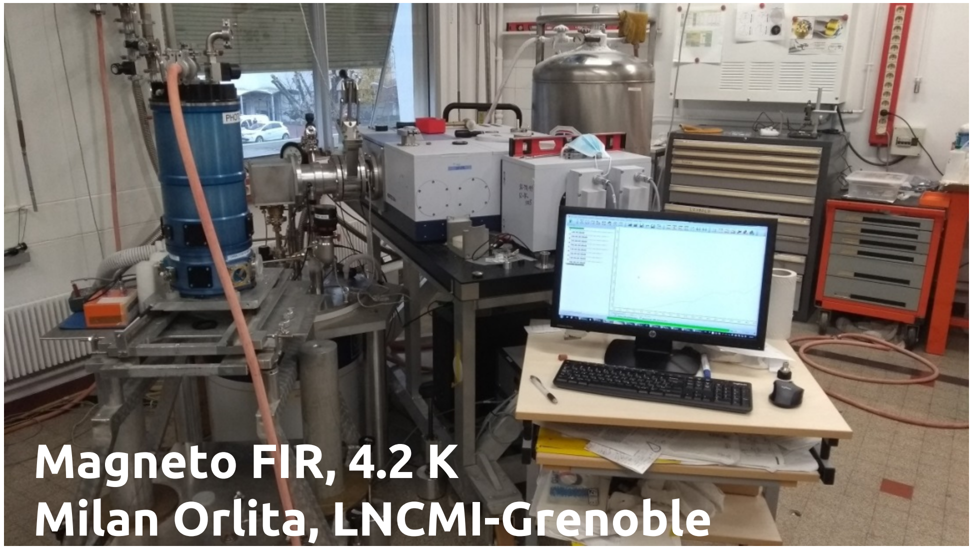
$G_z \sim$ 1.6-1.7 cm^{-1}

$\Delta J \sim 5 – 6$ cm^{-1}

50-55 cm^{-1} (1.5 THz)

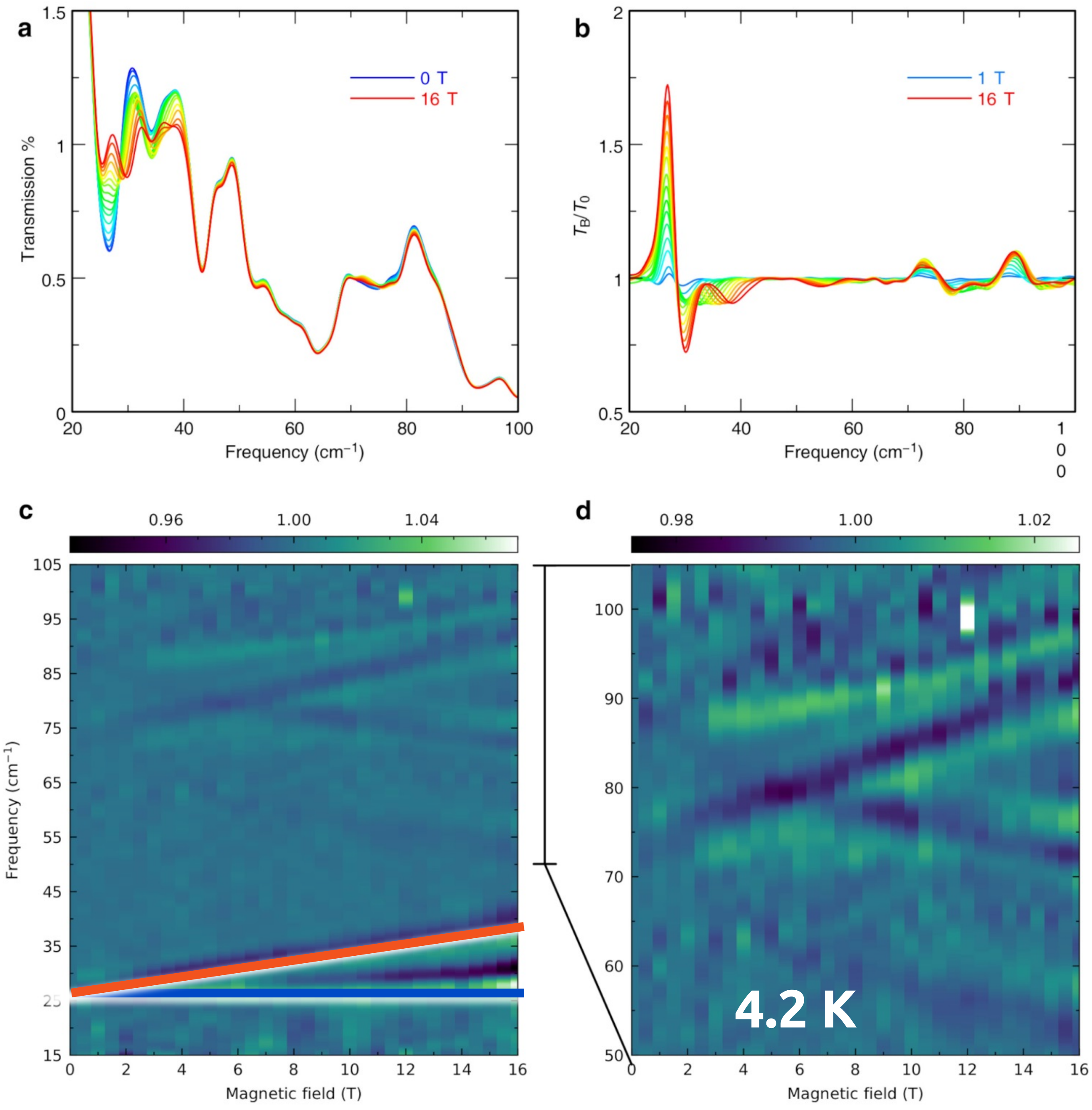
Far-IR

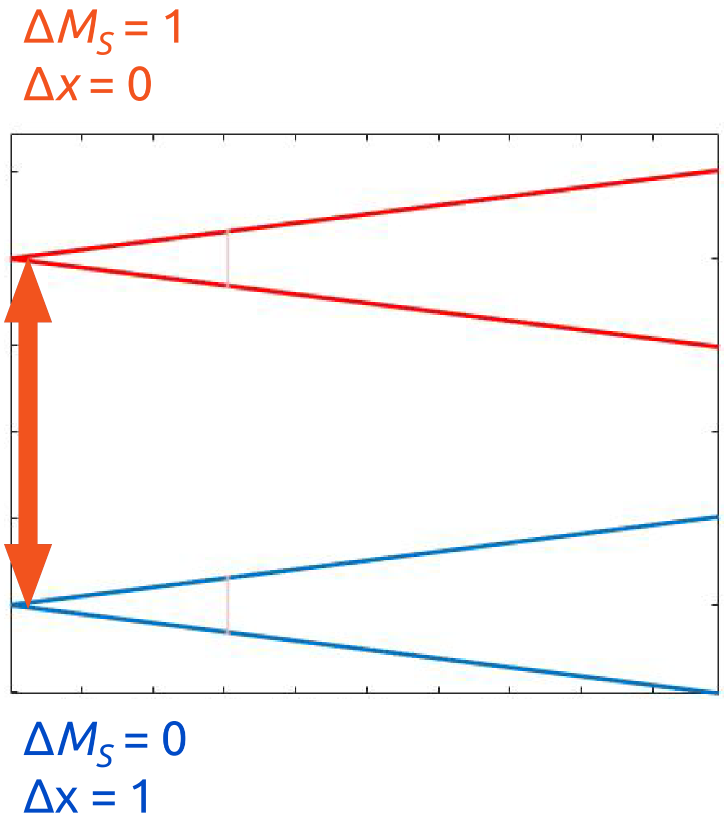




Magneto FIR, 4.2 K
Milan Orlita, LNCMI-Grenoble

Magneto-FIR spectra

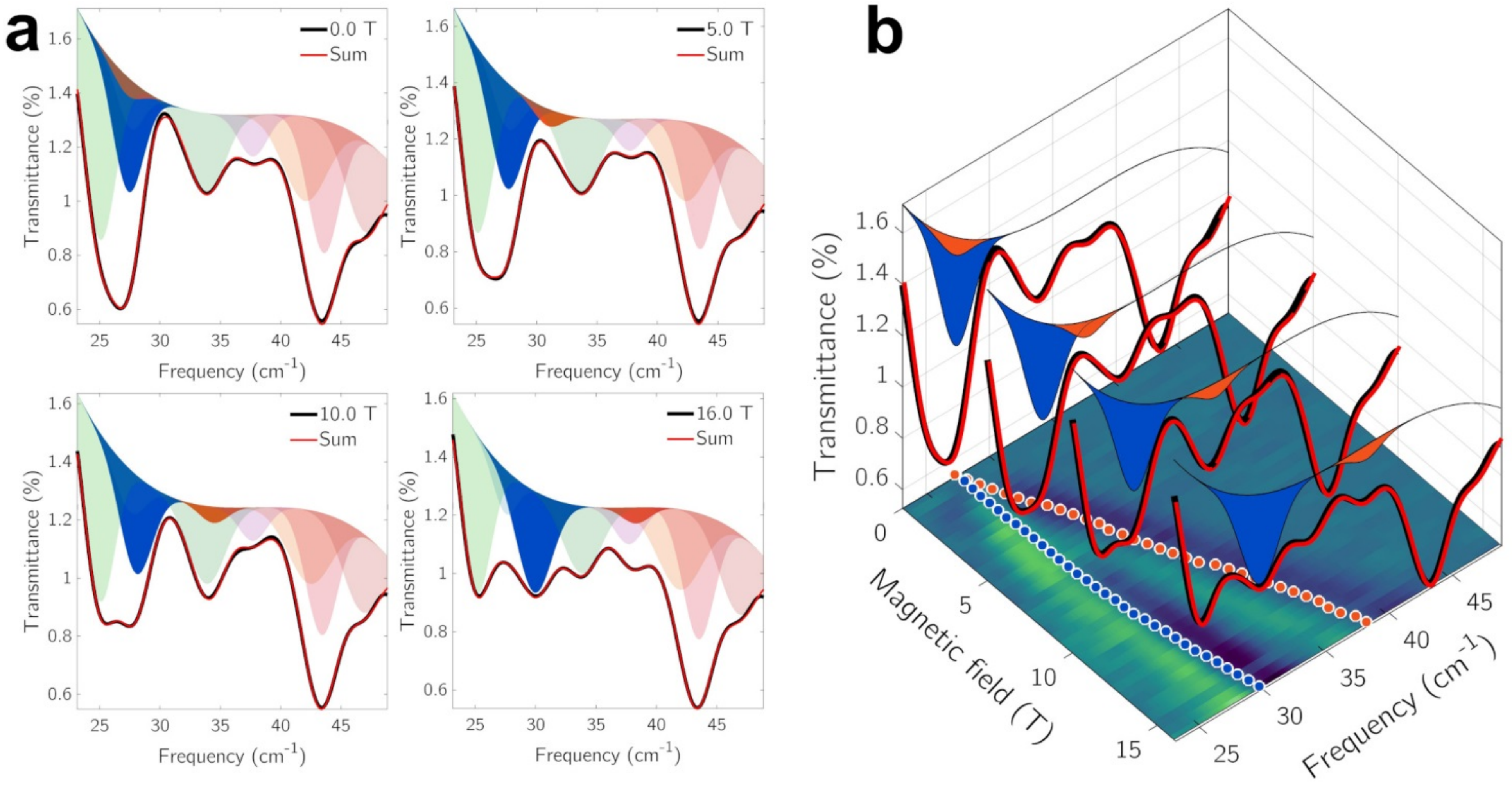




$\Delta M_S = 1$
$\Delta x = 0$

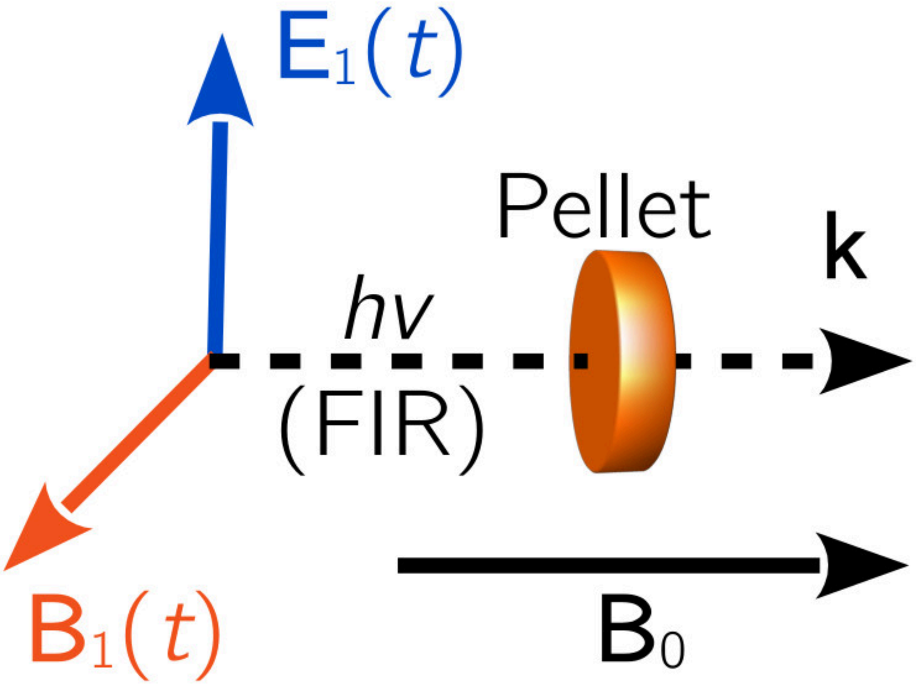
$\Delta M_S = 0$
$\Delta x = 1$

Empirical estimation of magnetic peak positions



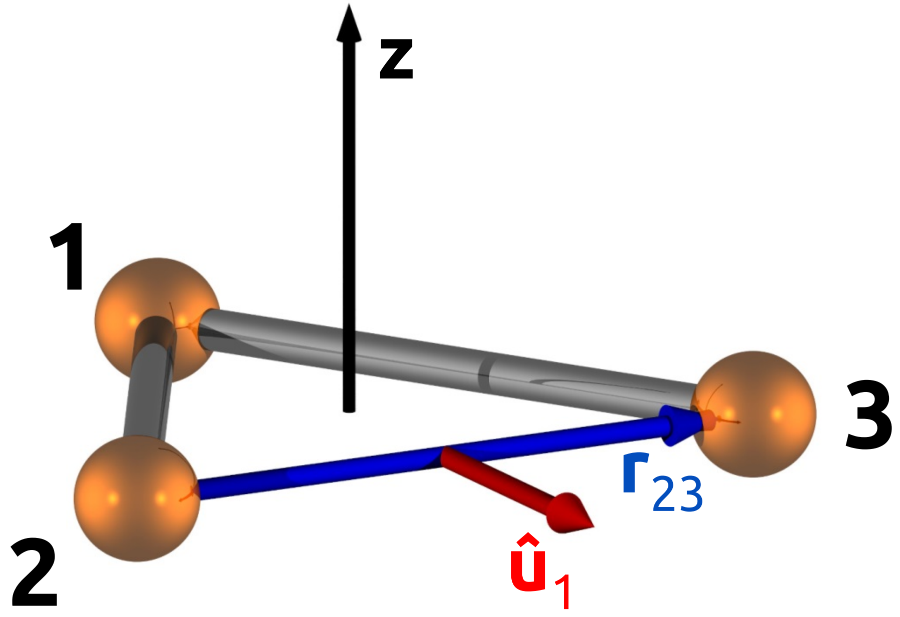
Interaction Hamiltonians

$$H_E = \kappa \sum_{i=1}^{3} \left(\mathbf{E}_1 \cdot \hat{\mathbf{u}}_k \right) \, \mathbf{s}_i \cdot \mathbf{s}_j$$

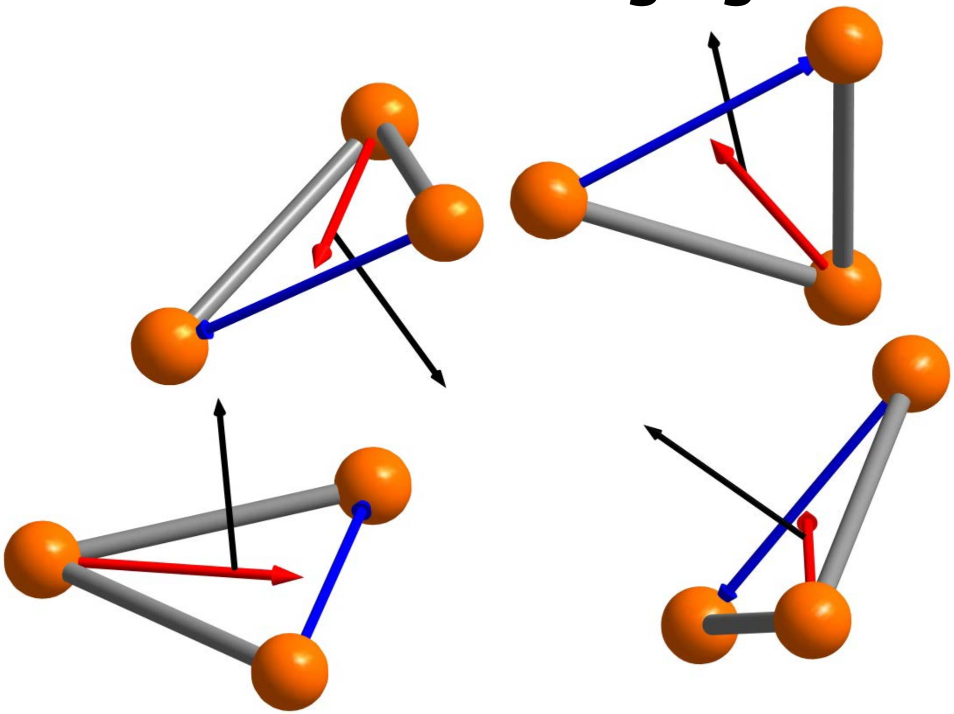


$$H_B = g \mu_B \mathbf{B}_1 \cdot \mathbf{S}$$

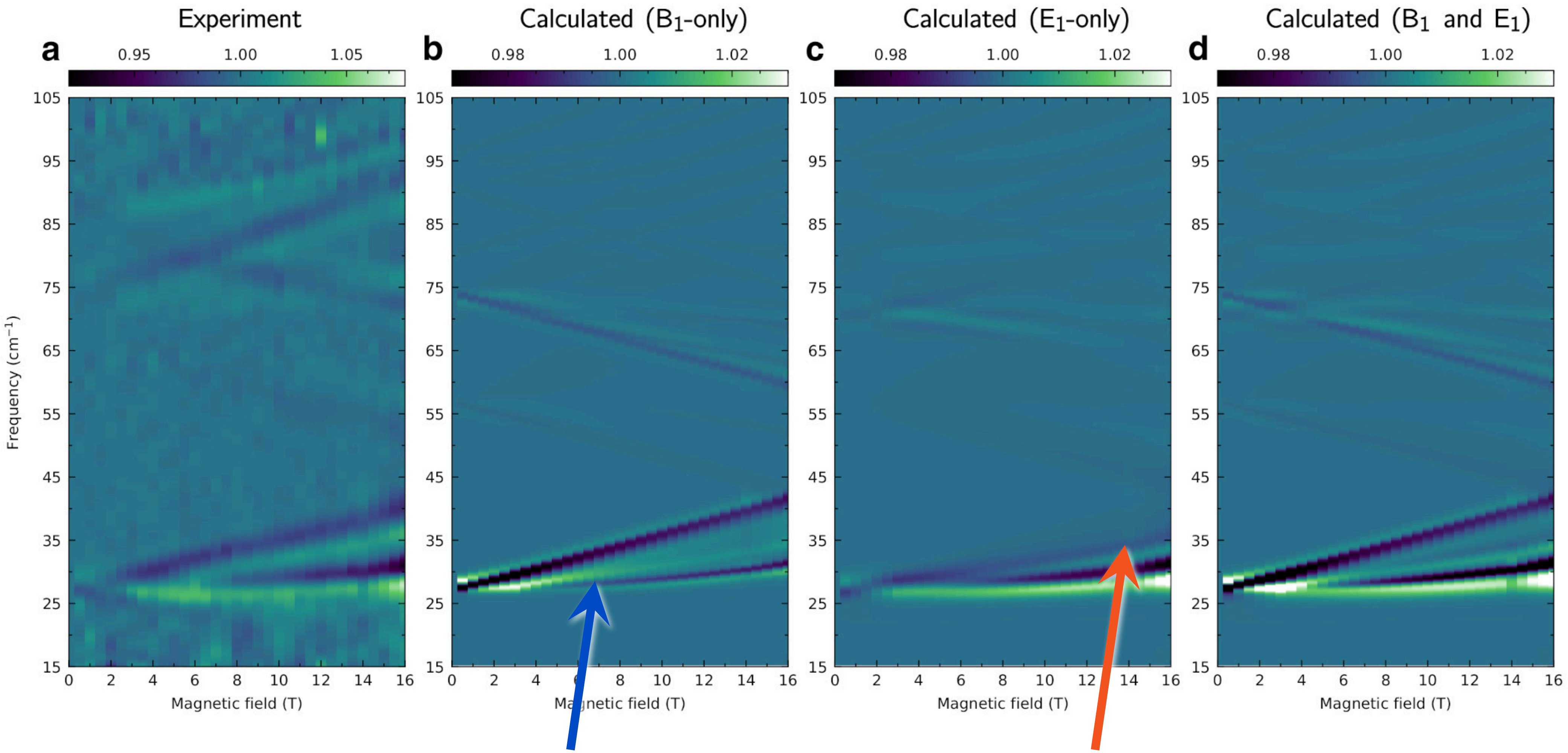
Filippo Troiani



Powder averaging



Magneto-FIR spectra



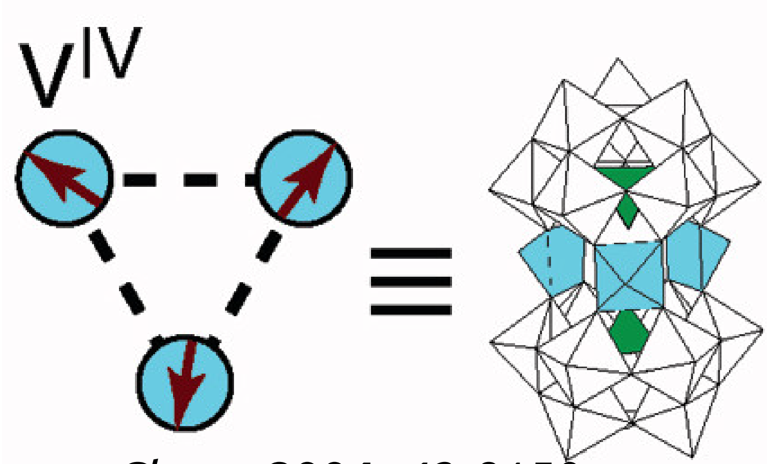
B_1 excites spin-electric transitions E_1 excites EPR transitions

Proper scaling of individual spectra…

$$\kappa \approx 4 \cdot 10^{-4}\ \mathrm{e\ nm}$$

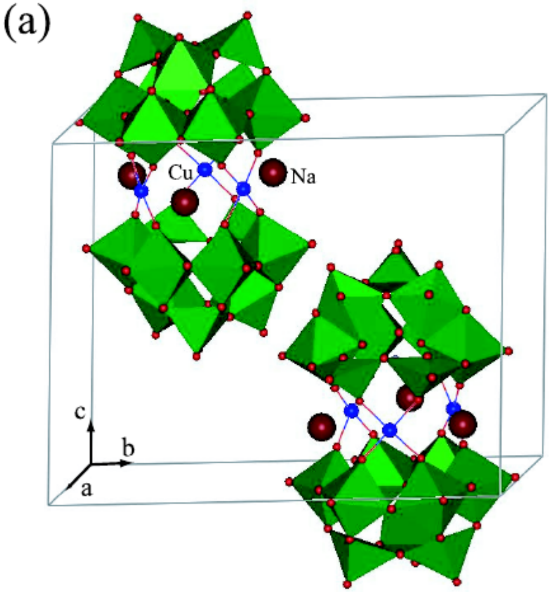
PRB, **2016,** *94*, 235423: ~ 10^{-4} e *a* ($Na_9[Cu_3Na_3(H_2O)_9(\alpha\text{-}AsW_9O_{33})_2]\cdot 26H_2O$)

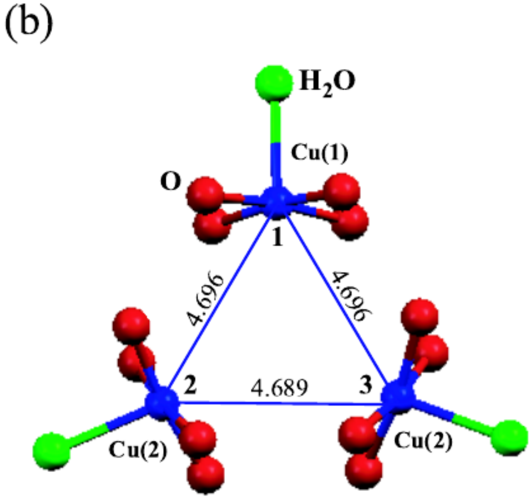
Previous theoretical models *vs* current results



Inorg. Chem., **2004**, *43*, 8150

$K_{11}H[(VO)_3(SbW_9O_{33})_2]\cdot 27H_2O$





PRL, **2006**, *96*, 107202

$Na_9[Cu_3Na_3(H_2O)_9(a\text{-}AsW_9O_{33})_2]\cdot 26H_2O$

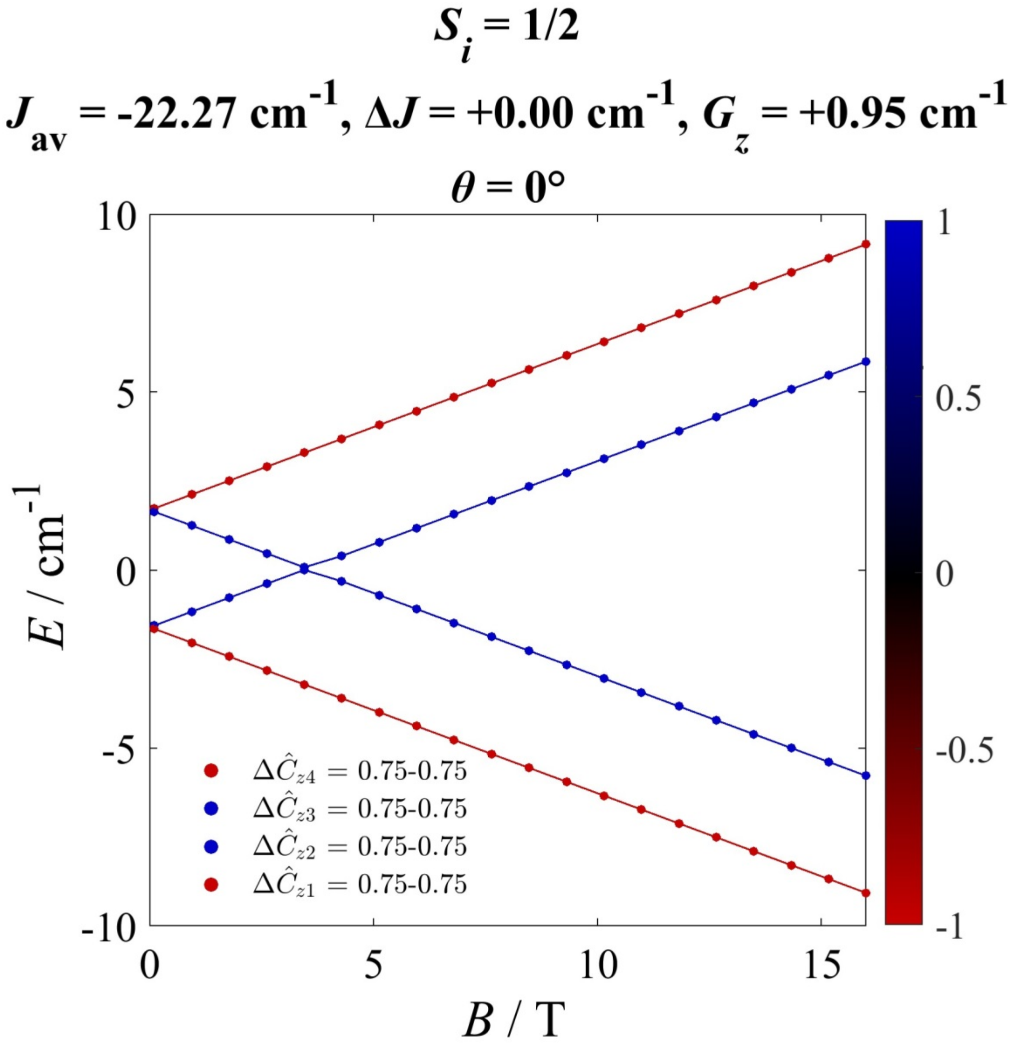
- $S_i = 1/2$
- $J_{ij} \sim 2\text{-}5\ \mathrm{cm}^{-1}$
- $J_{ij} >> \Delta J \sim 0$
- Doublet-doublet gap entirely due to DMI

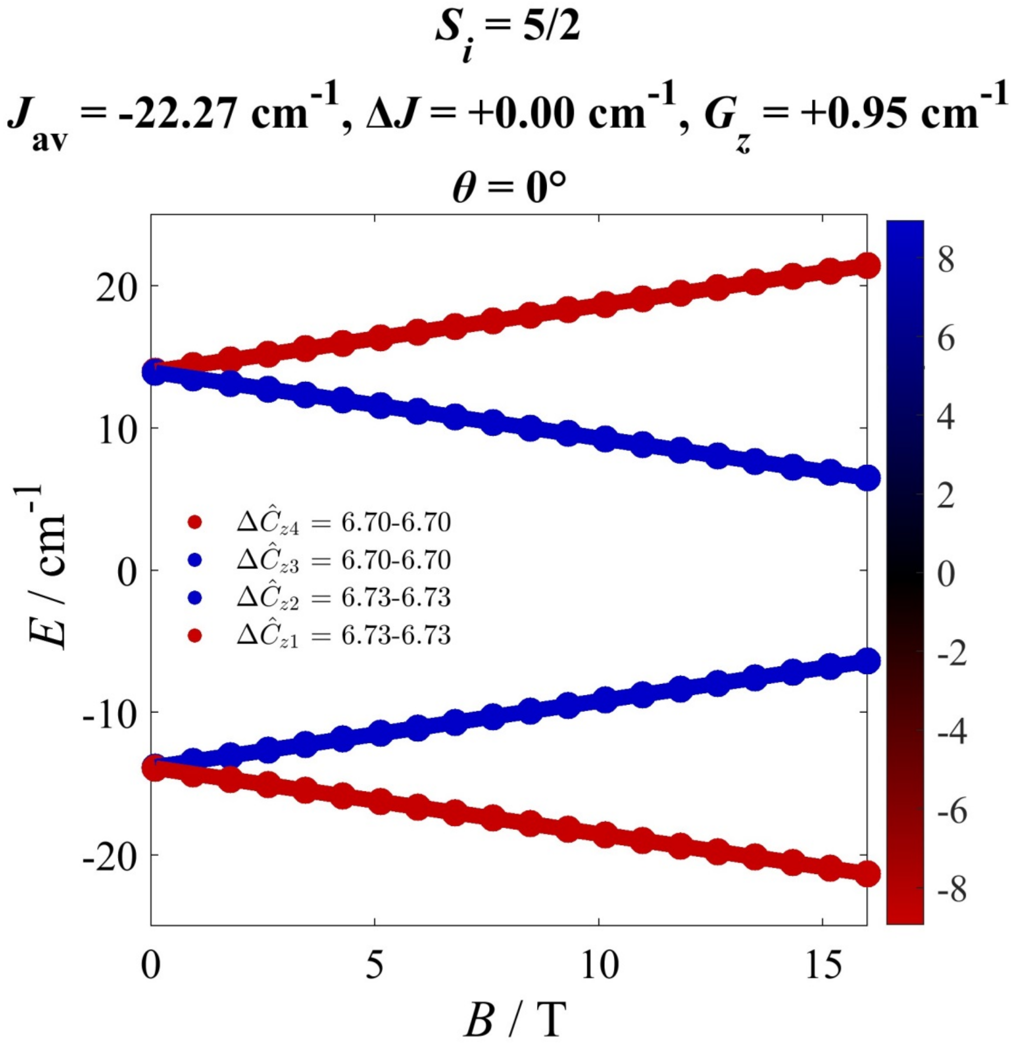
$S_i > ½$

S^2, C, **K**$_z$ are NOT good quantum numbers

S_i = 1/2 *vs* 5/2

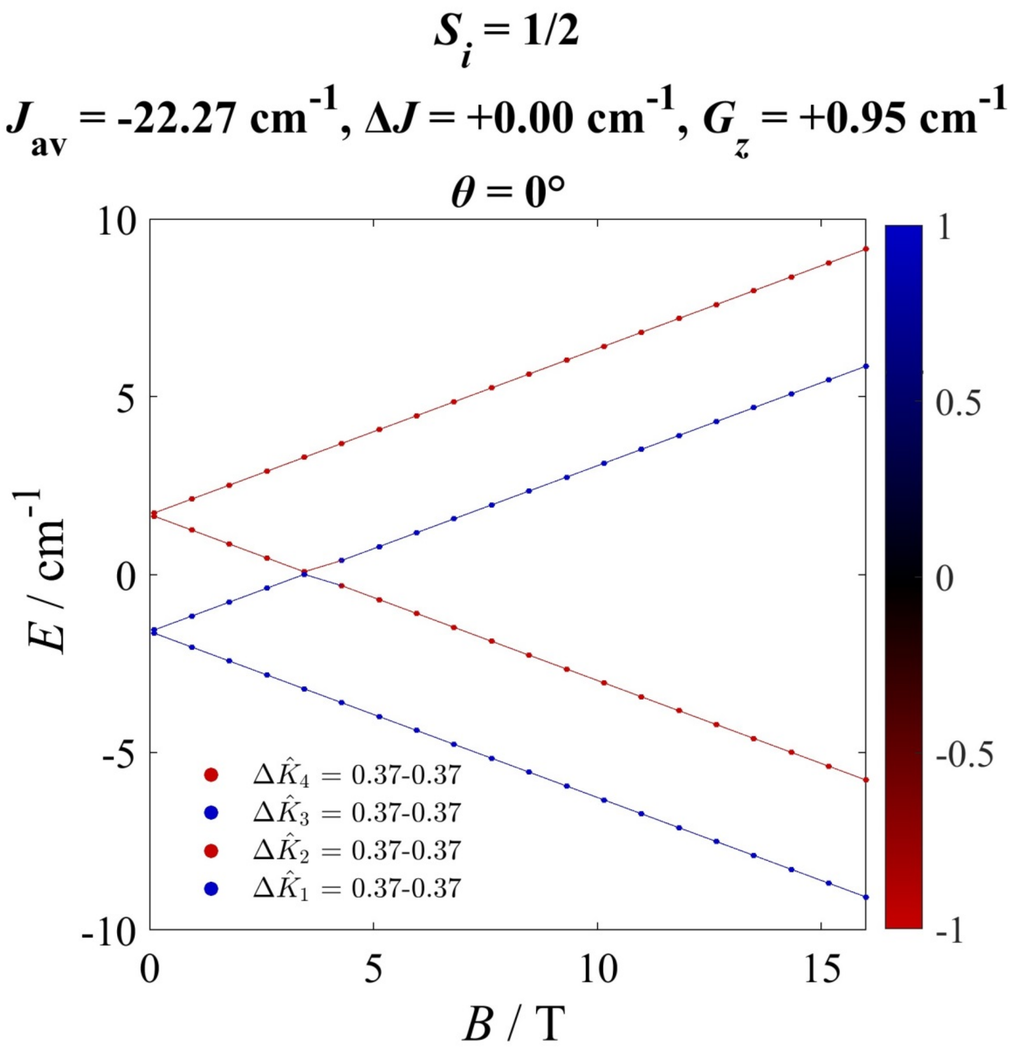
Scalar chirality





***S_i* = 1/2 *vs* 5/2**

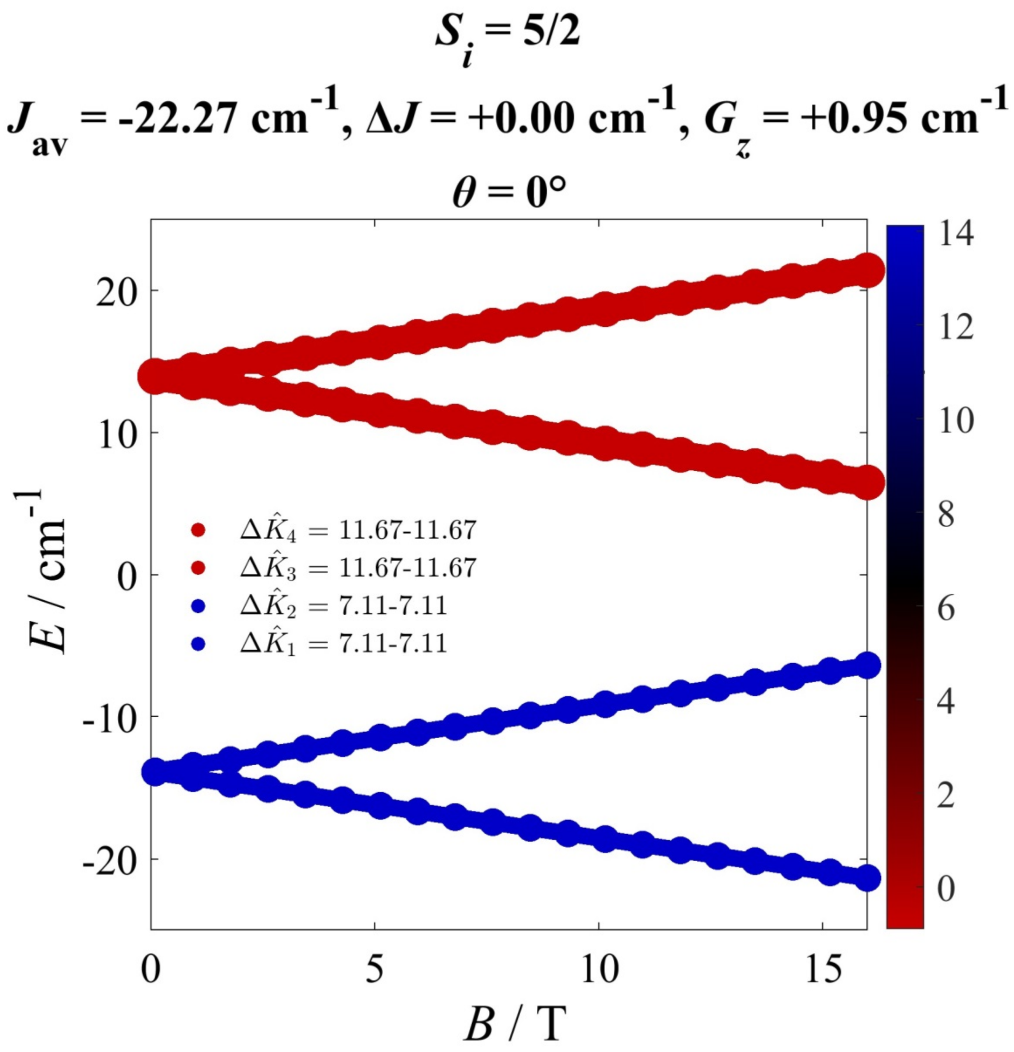
Vector chirality



$S_i = 1/2$

$J_{av} = -22.27\ \mathrm{cm}^{-1}$, $\Delta J = +0.00\ \mathrm{cm}^{-1}$, $G_z = +0.95\ \mathrm{cm}^{-1}$

$\theta = 0°$



$S_i = 5/2$

$J_{av} = -22.27\ \mathrm{cm}^{-1}$, $\Delta J = +0.00\ \mathrm{cm}^{-1}$, $G_z = +0.95\ \mathrm{cm}^{-1}$

$\theta = 0°$

Some new conclusions (more to come)

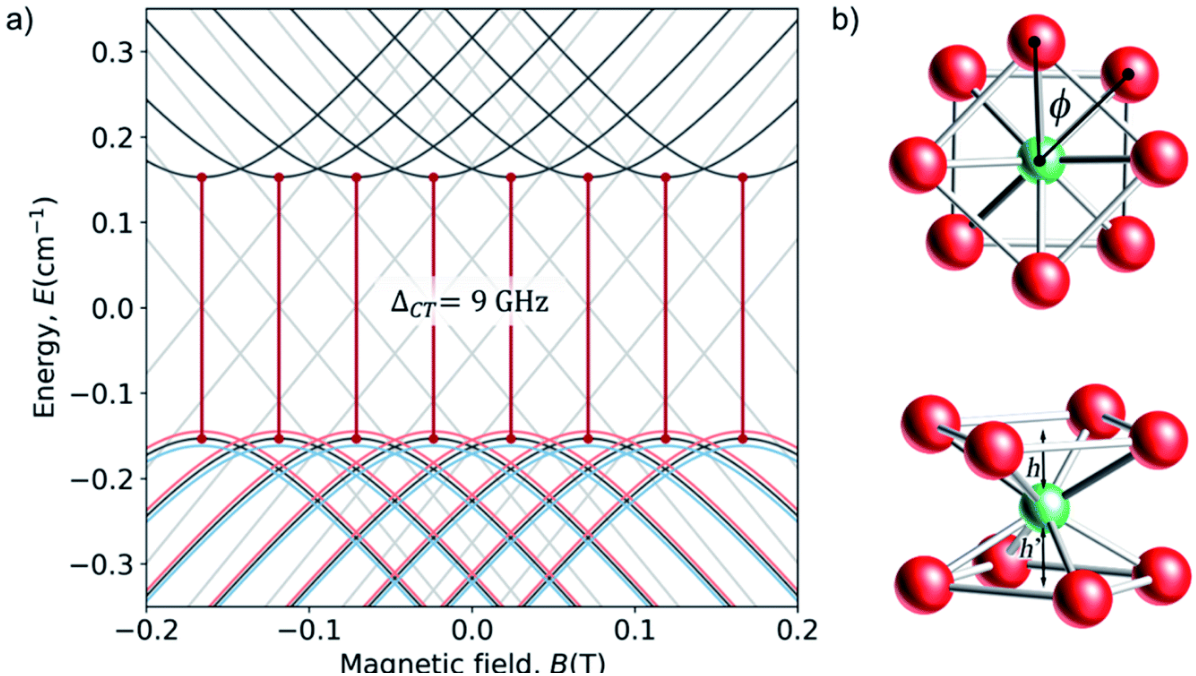
Large standard deviations of $\mathbf{S}^2$, C, $\mathbf{K}_{x/y/z}$
(i.e. $\Delta\hat{O} \equiv [\langle\hat{O}^2\rangle - \langle\hat{O}\rangle^2]^{1/2}$

Selection rules are relaxed

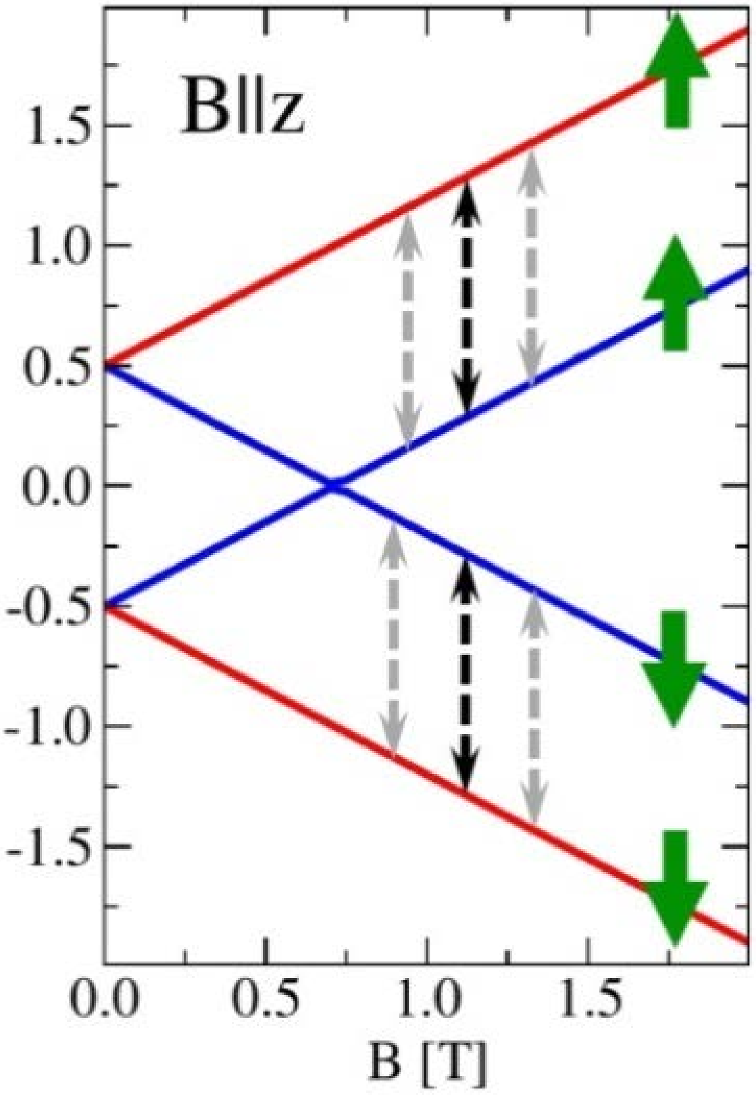
For $\theta \neq 0$:
E_1 partially excites EPR transitions
B_1 partially excites spin-electric transitions

$\Delta J \neq 0$ (isosceles distortion)
Spin-electric transitions allowed even if $|\Delta J| >> |G_z|$
But nuclear decoupling near the limit $|\Delta J| << |G_z|$

Perspectives: extended clock transitions

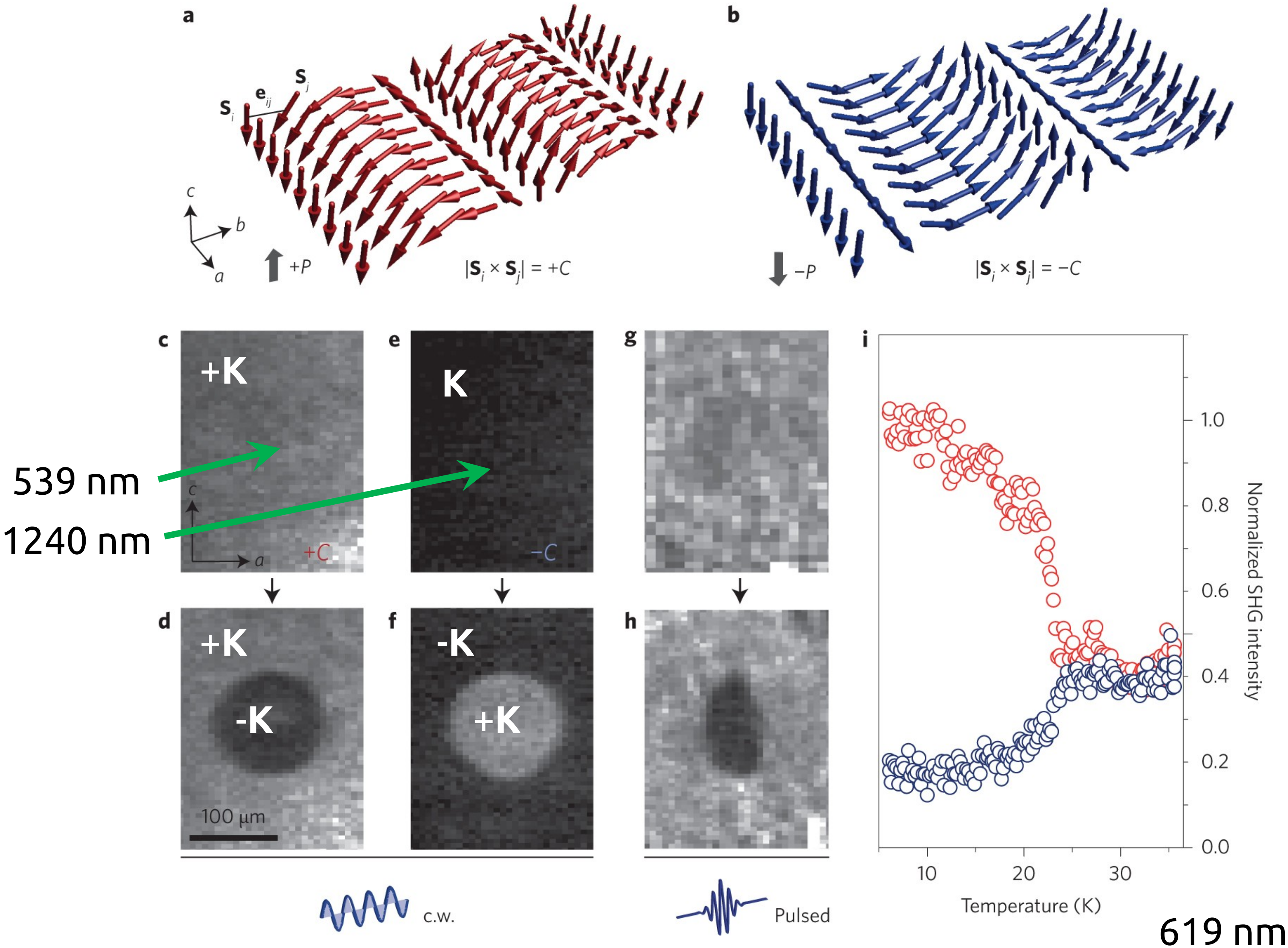


Chem. Sci., **2020**, *11*, 10718



Perspectives: readout

Second harmonic generation



539 nm

1240 nm

$TbMnO_3$

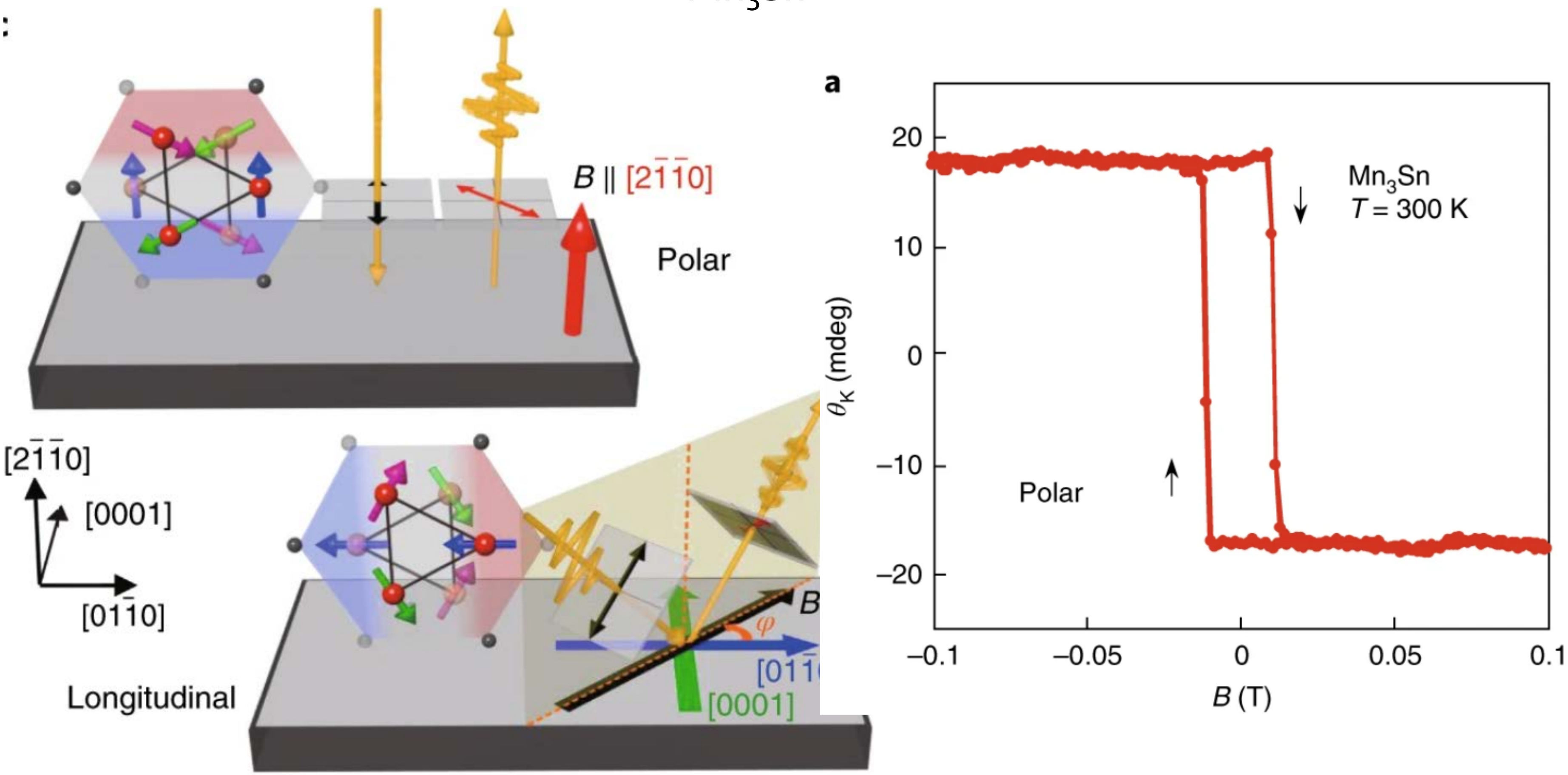
619 nm SHG readout

Nature Photonics, **2016**, *10*, 653

Perspectives: readout

Magneto-Optic Kerr Effect (MOKE)

Mn_3Sn



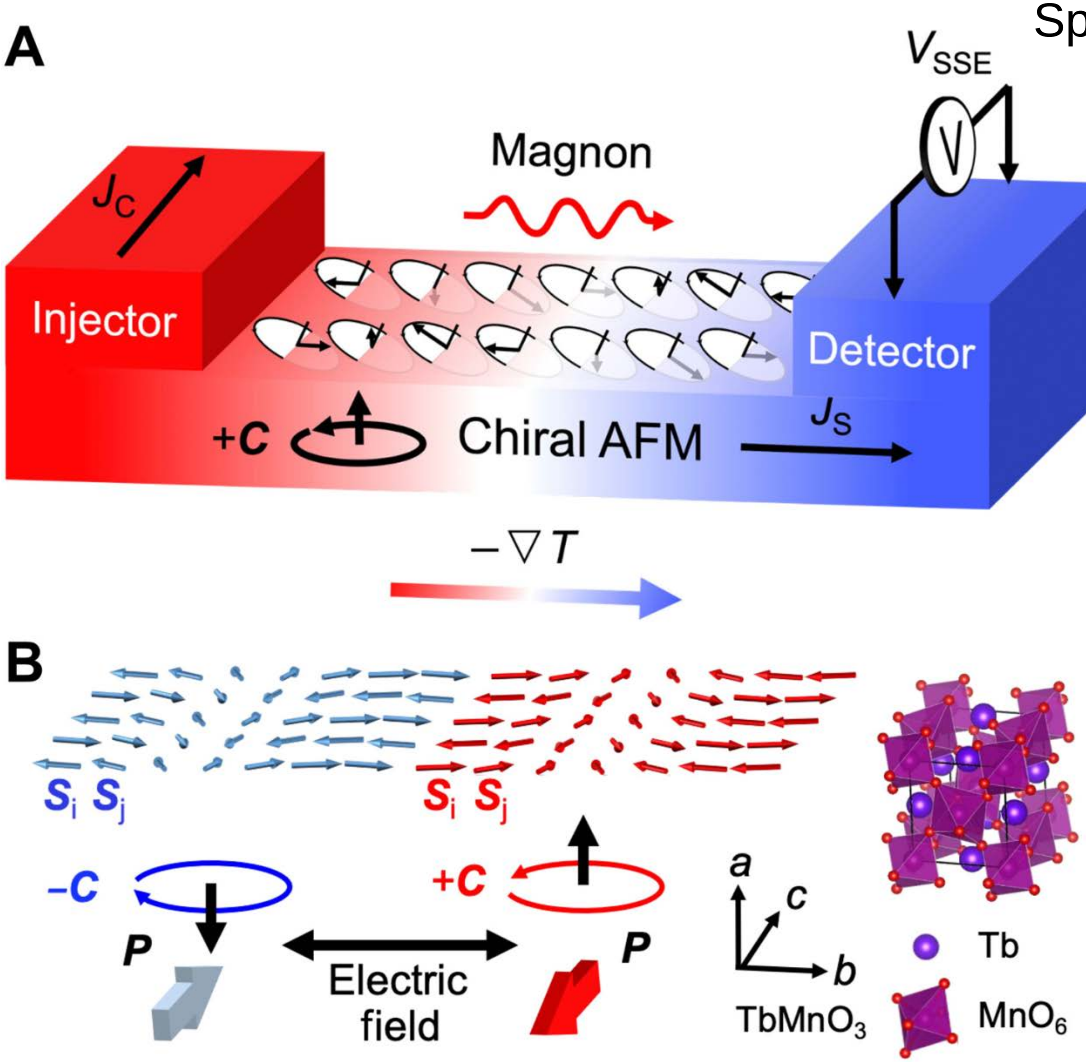
Nature Photonics, **2018**, *12*, 73

Perspectives: readout

Electrical

Spin Seebeck Effect voltage

TbMnO$_3$



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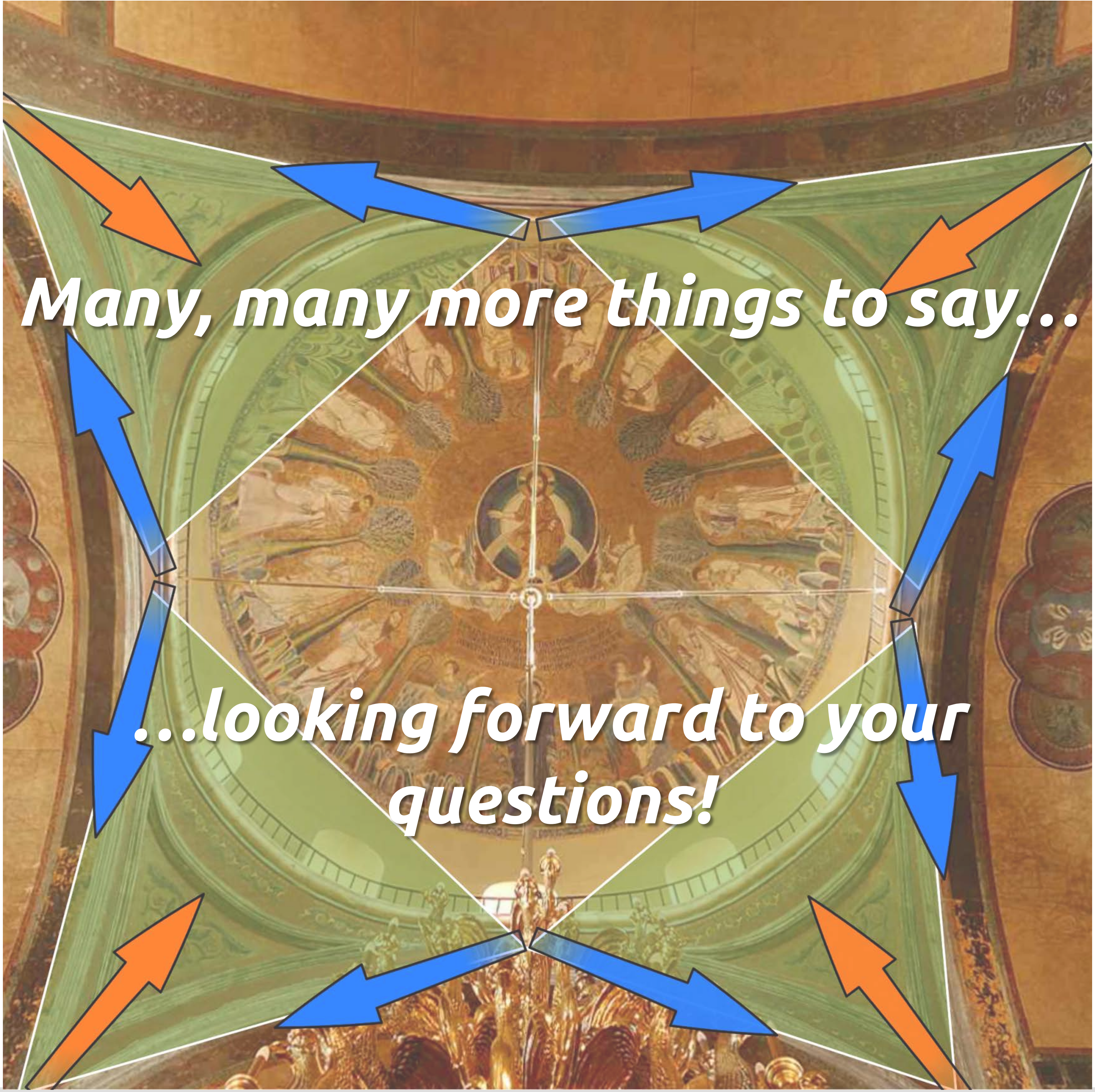

Many, many more things to say…

…looking forward to your questions!