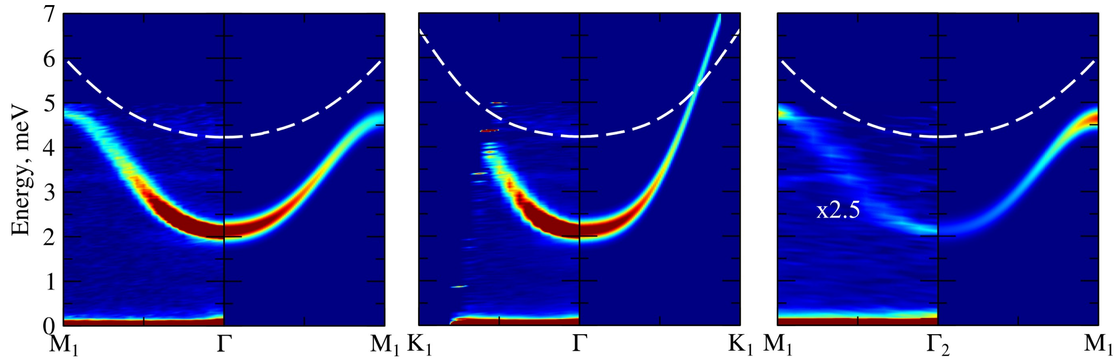
Magnetic spectrum of cobalt-based frustrated magnets



Pavel Maksimov

Joint Institute for Nuclear Research, Dubna

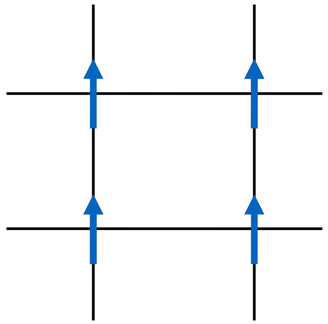
BLTP

Heisenberg model

$$\mathcal{H} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

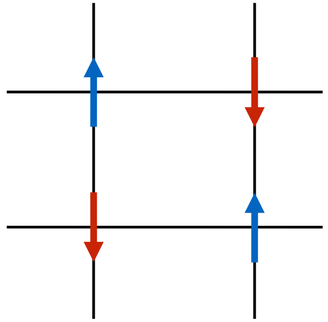
Ferromagnet

$J < 0$

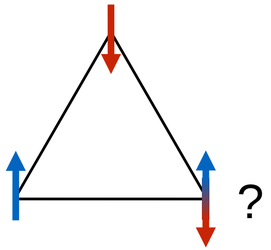


Antiferromagnet

$J > 0$



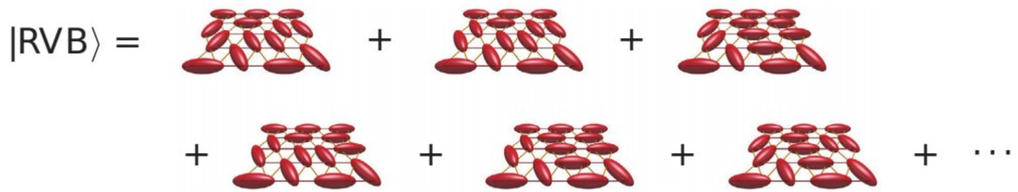
Frustrated magnets



G. H. Wannier, Physical Review 79, 357 (1950)

Classical degeneracy → Finite entropy at T=0

Resonating valence bond (RVB)



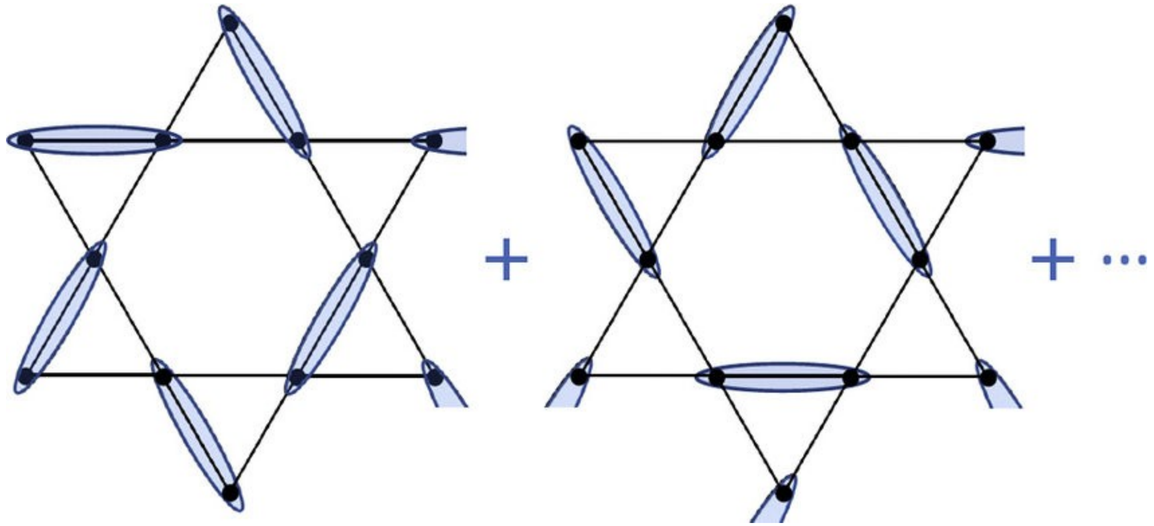
P. W. Anderson, Materials Research Bulletin 8, 153 (1973)

P. Fazekas and P. W. Anderson, Philosophical Magazine 30, 423 (1974)

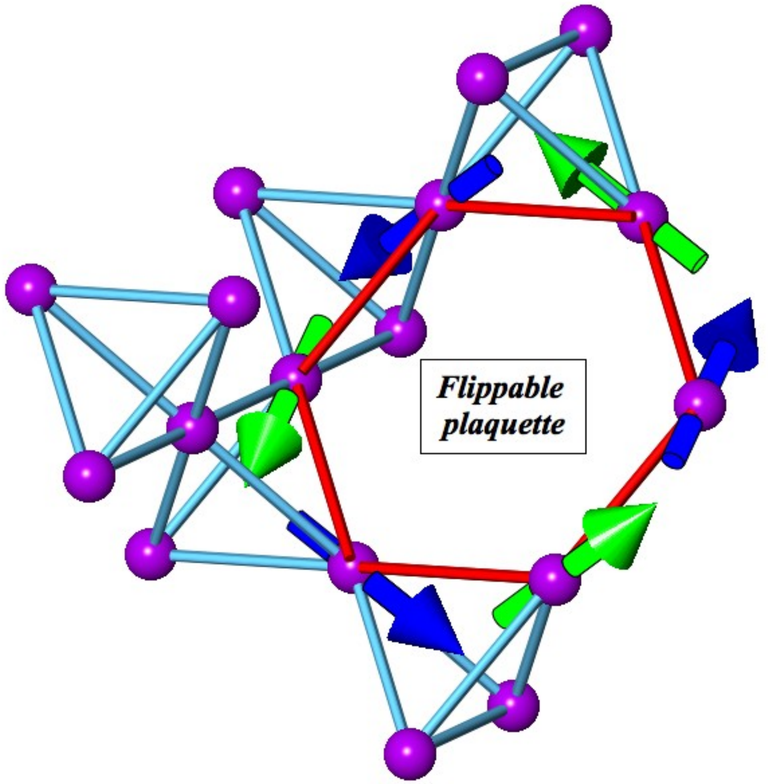
Spin liquids: fractionalized excitations, topological properties

L. Savary and L. Balents, Reports on Progress in Physics 80, 016502 (2017)

Frustration from geometry: kagome and pyrochlore lattices



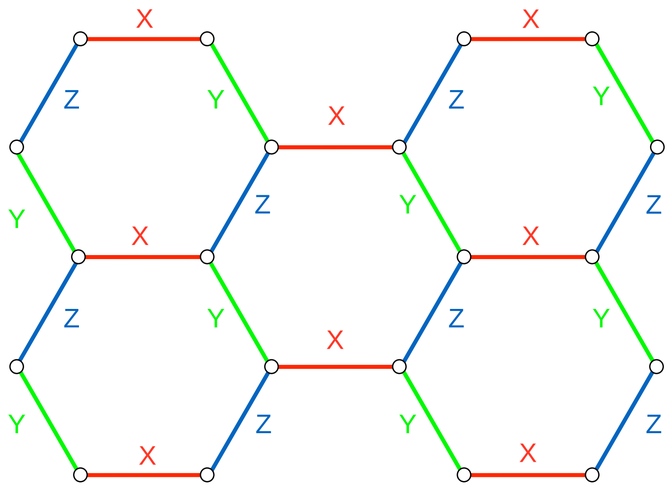
P. Mendels and F.Bert, Comptes Rendus Physique 17, 455 (2016)



M.J.P. Gingras and P.A. McClarty, Rep. Prog. Phys. 77, 056501 (2014)

Frustration from anisotropic interactions: Kitaev model

$$\mathcal{H} = K \sum_{\langle ij \rangle^{\gamma}} S_i^{\gamma} S_j^{\gamma} = K \sum_{\langle ij \rangle^{\mathrm{x}}} S_i^{x} S_j^{x} + K \sum_{\langle ij \rangle^{\mathrm{y}}} S_i^{y} S_j^{y} + K \sum_{\langle ij \rangle^{\mathrm{z}}} S_i^{z} S_j^{z}$$

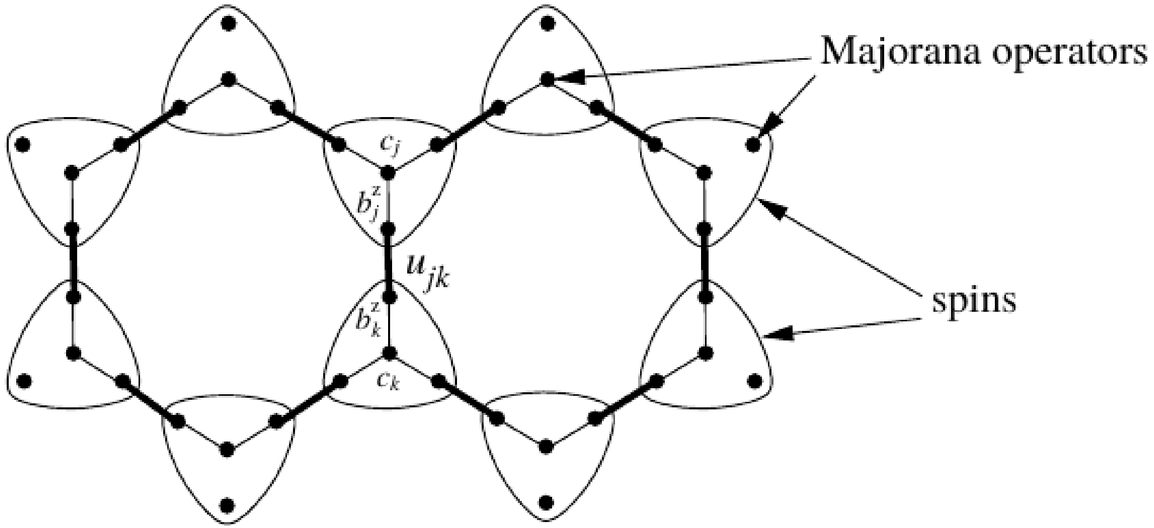


A. Kitaev, Annals of Physics 321, 2 (2006), January Special Issue

Exactly solvable for S=1/2, massive classical degeneracy

Kitaev model: Majorana fermions

$$\mathcal{H} = K \sum_{\langle ij \rangle^\gamma} S_i^\gamma S_j^\gamma = K \sum_{\langle ij \rangle^x} S_i^x S_j^x + K \sum_{\langle ij \rangle^y} S_i^y S_j^y + K \sum_{\langle ij \rangle^z} S_i^z S_j^z$$



Free fermion solution!

$$S^x = ib^x c, \; S^y = ib^y c, \; S^z = ib^z c$$

Majorana fermions:

$$c_j^2 = 1, \; c_i c_j = -c_j c_i, \; i \neq j$$

Non-abelian statistics in magnetic field: path to topological quantum computing

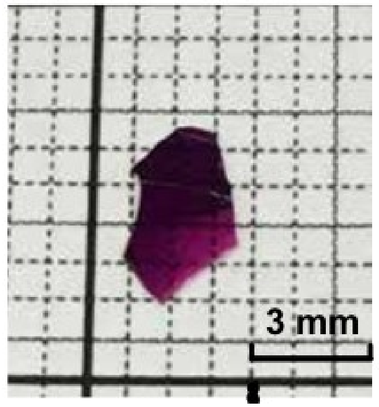
А. Китаев, А. Шень, М. Вялый

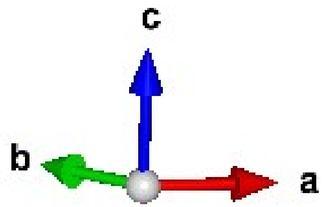
КЛАССИЧЕСКИЕ И КВАНТОВЫЕ ВЫЧИСЛЕНИЯ

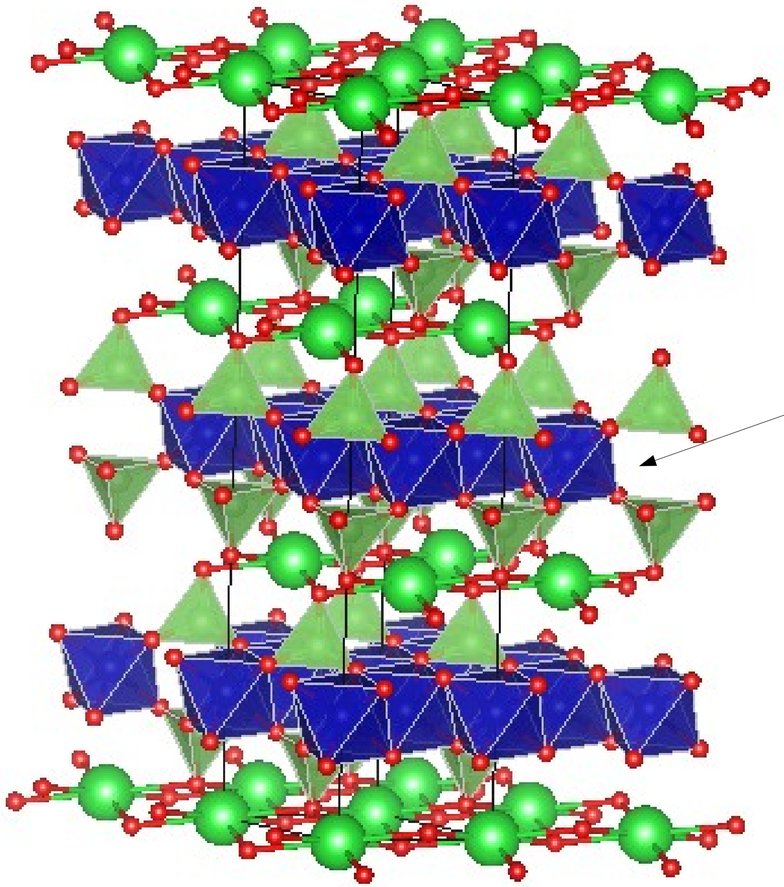
4. Анионы. Анионы — это особые возбуждения в двумерных квантовых системах, в частности, в двумерной электронной жидкости в магнитном поле. Один из авторов (А. К.) считает этот подход наиболее интересным (поскольку он же его и придумал [32]), поэтому опишем его более подробно.

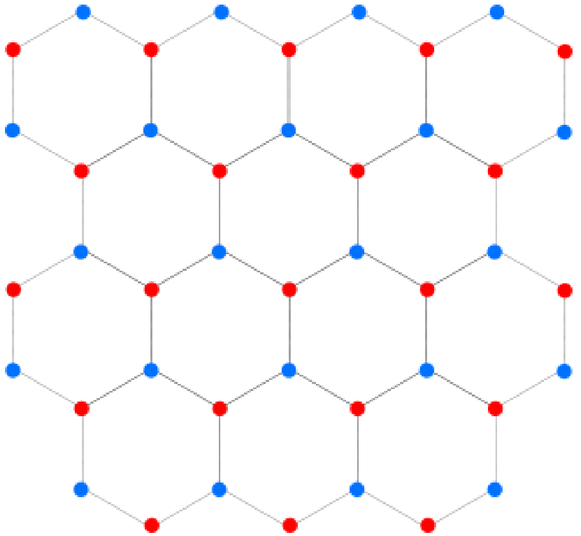
Honeycomb Kitaev magnet $BaCo_2(AsO_4)_2$

$BaCo_2(AsO_4)_2$: structure







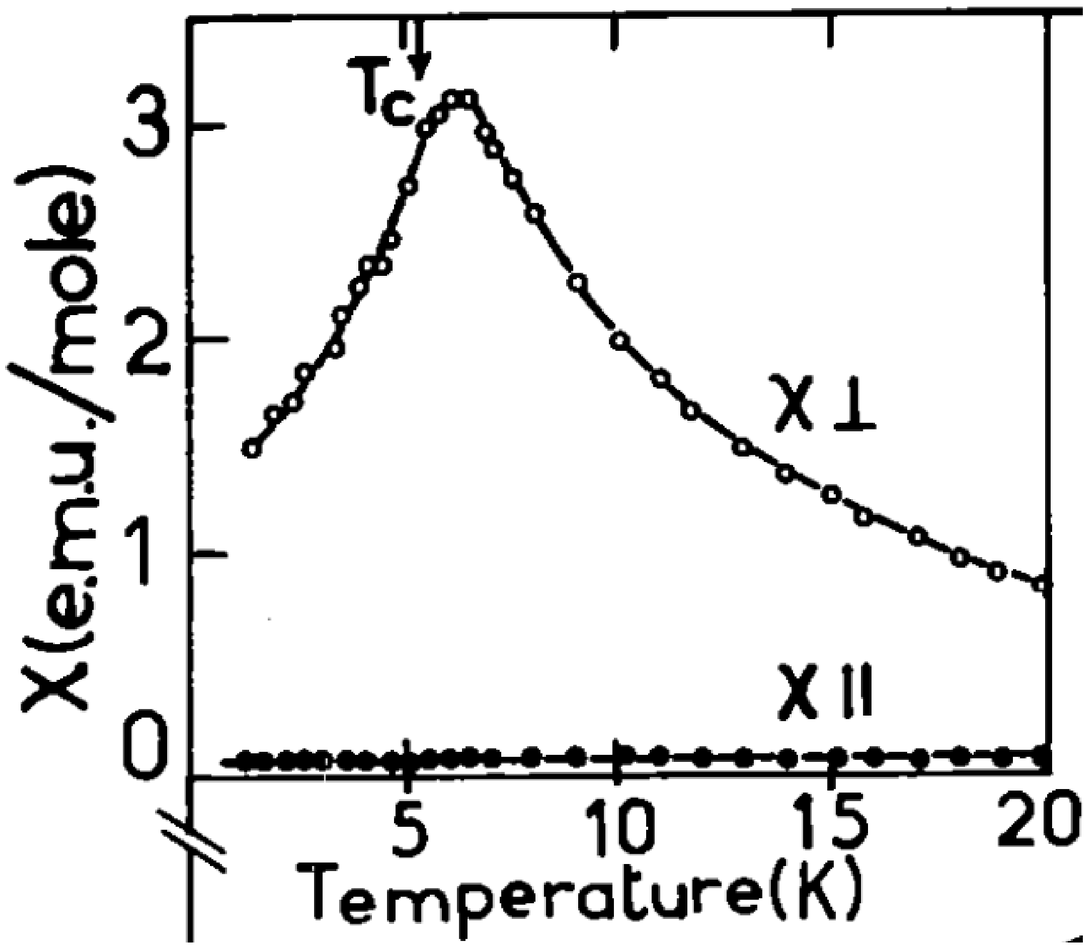


Honeycomb layers
(related to structure of mica)

S. Eymond and A. Durif, Mater. Res. Bull. 4 595 (1969)

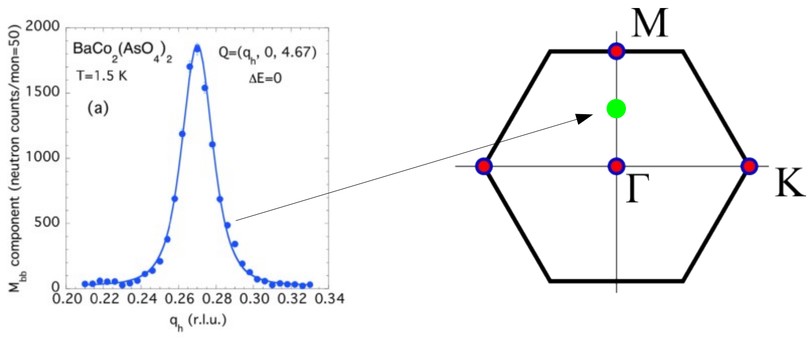
R. Zhong, T. Gao, N. P. Ong, and R. J. Cava, Sci. Adv. 6, eaay6953 (2020)

$BaCo_2(AsO_4)_2$: easy-plane anisotropy

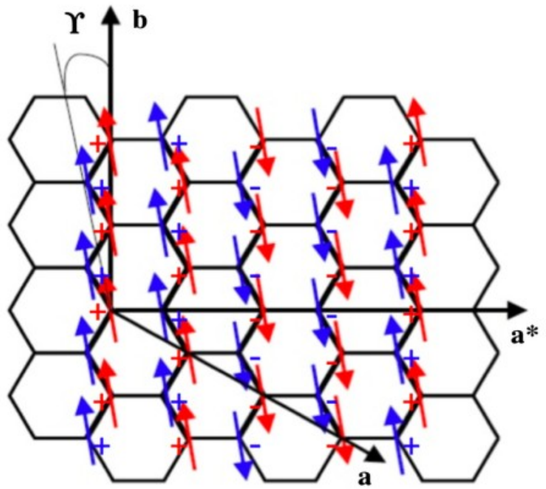


L.P. Regnault et al., Physica B+C 86, 660 (1977)

$BaCo_2(AsO_4)_2$: «double zigzag»+plateau

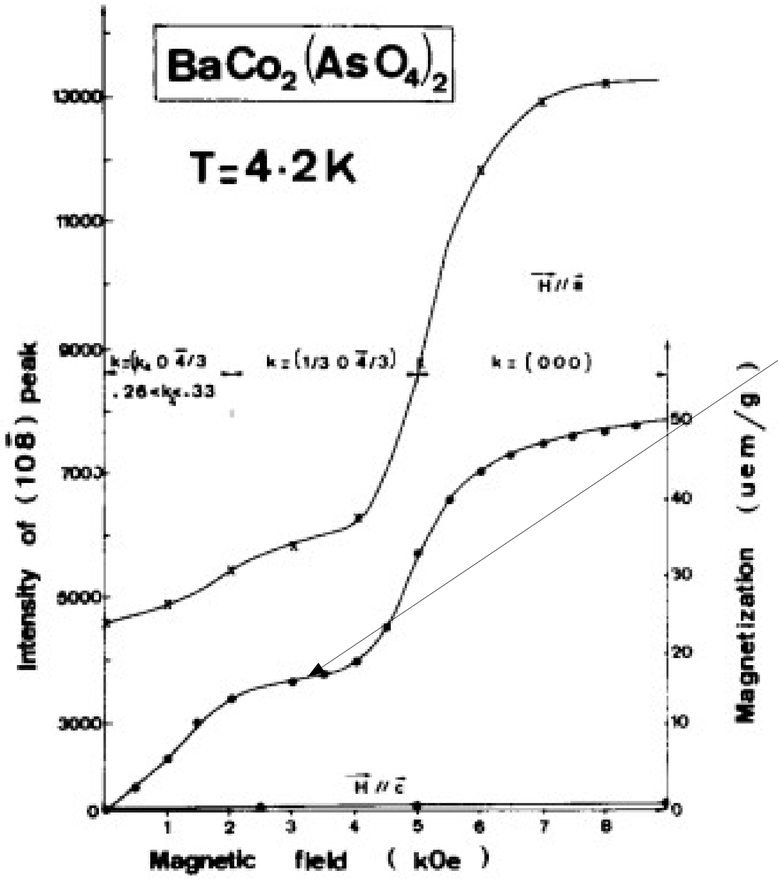


L.P. Regnault et al., Physica B+C 86, 660 (1977)



L. Regnault et al., Physica B: Condensed Matter 385-386, 425 (2006)

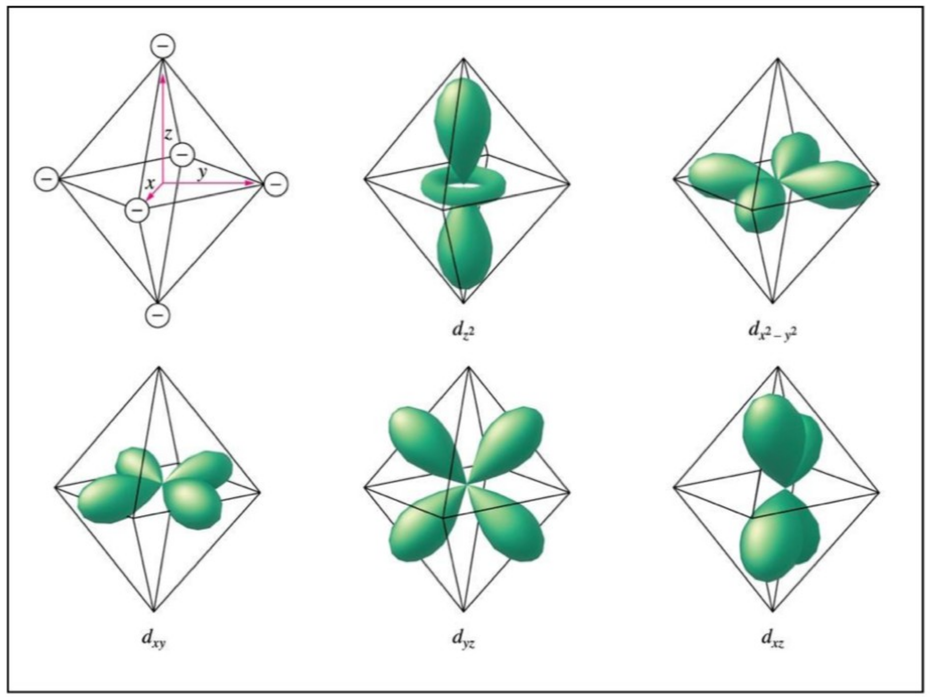
L.P. Regnault et al., Heliyon 4, e00507 (2018)

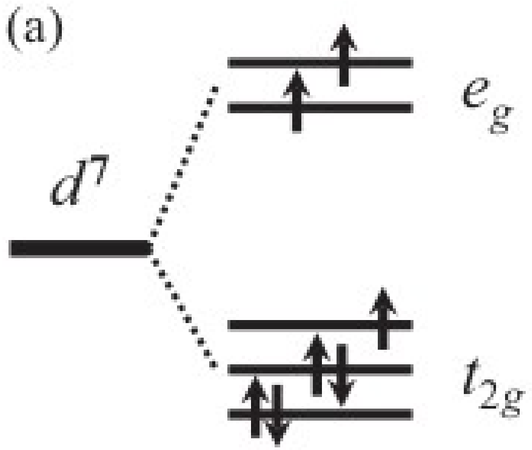


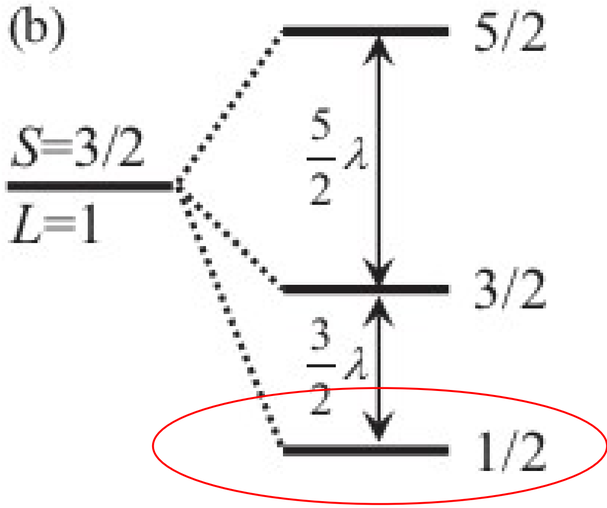
1/3 plateau

L.P. Regnault et al., Physica B+C 86, 660 (1977)

Electronic structure of d^7 ion - Co^{2+}



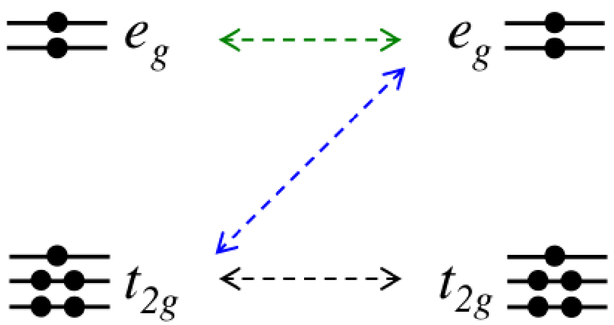


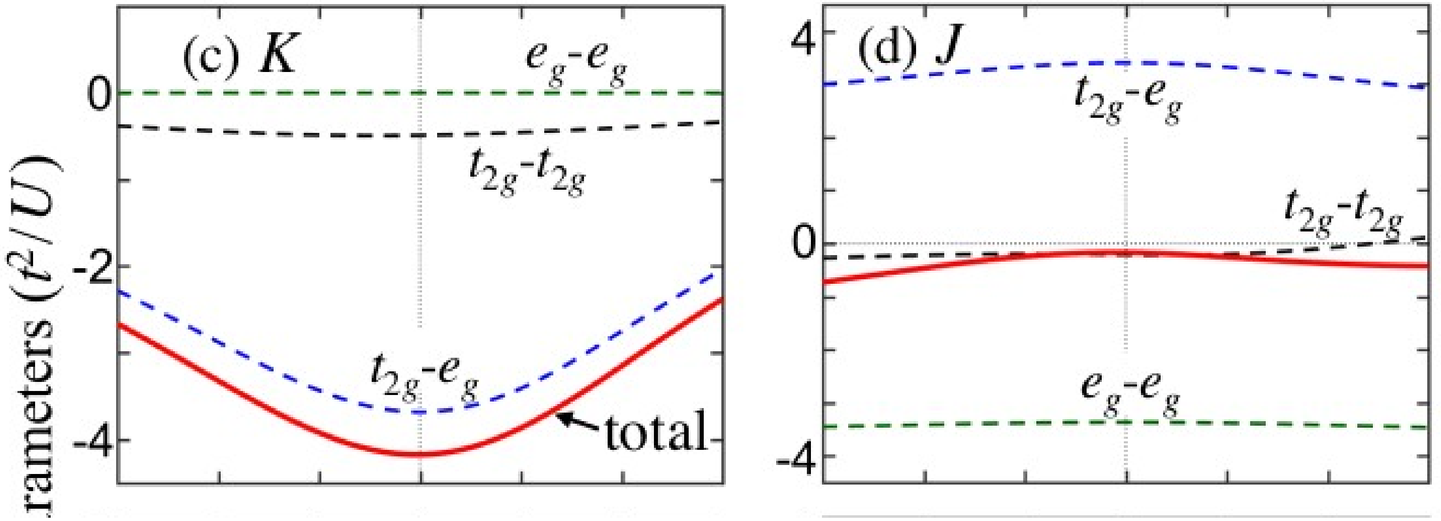


$$\psi_{+1}=c_1|-1,\tfrac{3}{2}\rangle+c_2|0,\tfrac{1}{2}\rangle+c_3|1,-\tfrac{1}{2}\rangle,$$
$$\psi_{-1}=c_1|1,-\tfrac{3}{2}\rangle+c_2|0,-\tfrac{1}{2}\rangle+c_3|-1,\tfrac{1}{2}\rangle,$$

Abragam A. and Pryce M.H.L, Proc. R. Soc. Lond. A206 173 (1951)
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)
R. Sano, Y. Kato and Y. Motome, Phys. Rev. B 97, 014408 (2018)

Exchanges of Co^{2+}





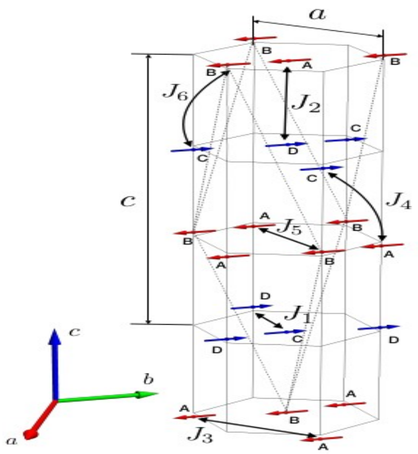
$$\mathcal{H} = \sum_{\langle ij \rangle^\gamma} J\mathbf{S}_i \cdot \mathbf{S}_j + KS_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right) + \Gamma'\left(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma\right)$$

H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)

R. Sano, Y. Kato and Y. Motome, Phys. Rev. B 97, 014408 (2018)

Kitaev materials with Co^{2+}

$CoTiO_3$

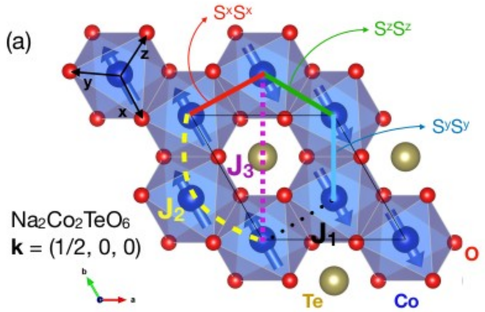


XXZ+(small bond-dependent)

Elliot, M. et al. Nat Commun 12, 3936 (2021)

$Na_2Co_2TeO_6$

$Na_3Co_2SbO_6$



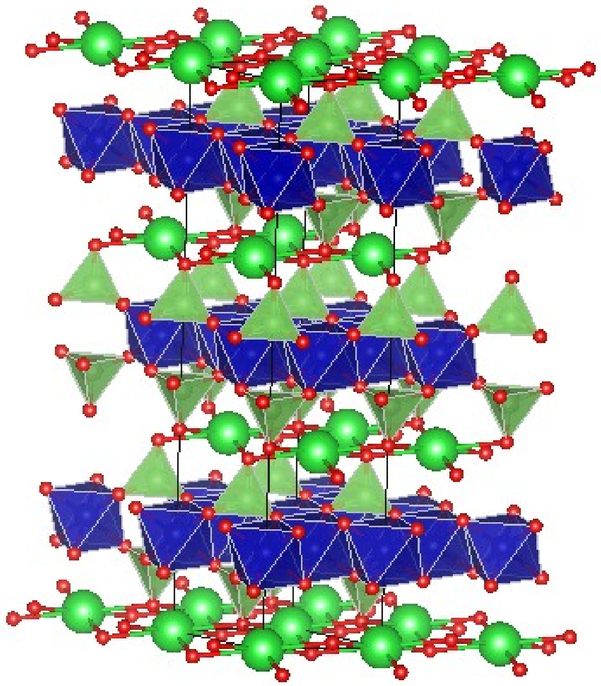
Large K, large J3

M. Songvilay et al., Phys. Rev. B 102, 224429 (2020)

C. Kim et al., J. Phys.: Condes. Matter 34 045802 (2022)

Ising chains: $CoNb_2O_6$, $BaCo_2V_2O_8$

$BaCo_2(AsO_4)_2$: *ab initio* exchange integrals



U	6 eV
J_1 (K)	-40.9
K_1 (K)	2.2
Γ_1 (K)	-1.7
Γ'_1 (K)	4.0
J_3 (K)	24.6
K_3 (K)	0.2
Γ_3 (K)	-6.0
Γ'_3 (K)	-2.3

DFT /A.Ushakov, Z.Pchelkina, S.Streltsov (IMP RAS)/ (total energy method)

Almost isotropic model with 3rd neighbor interaction

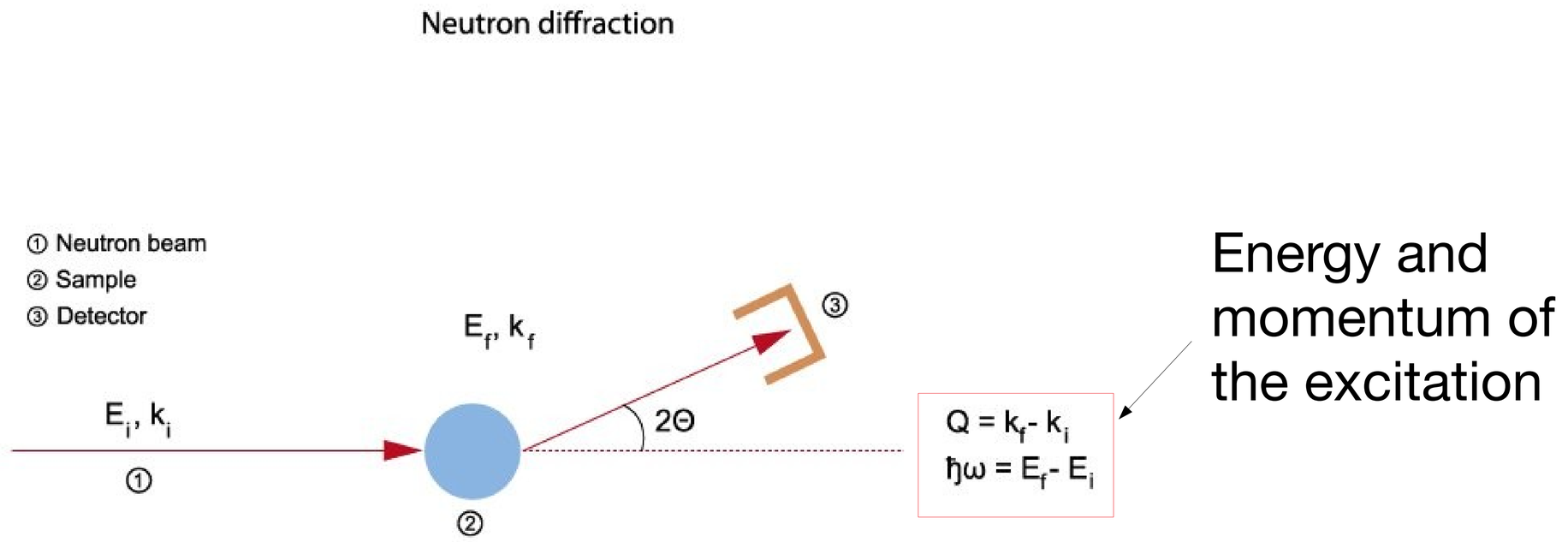
$J_3 > 0.5\ J_1$

Agreement with c S. Das et al., Phys. Rev. B 104, 134425 (2021)

PM et al., Phys. Rev. B 106, 165131 (2022)

P.A. Maksimov, Phys. Rev. B 108, L180405 (2023)

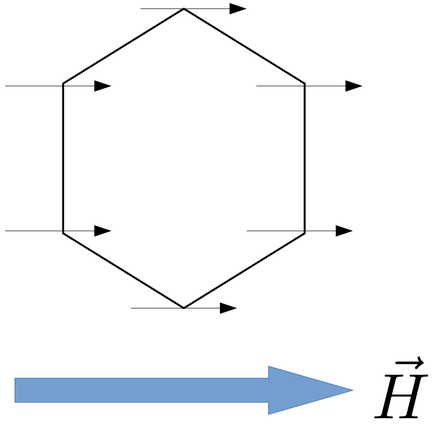
Neutron scattering



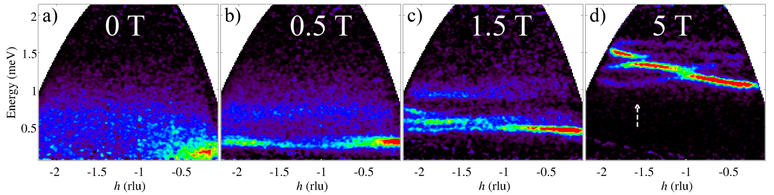
Energy and momentum of the excitation

Inelastic neutron scattering in $BaCo_2(AsO_4)_2$

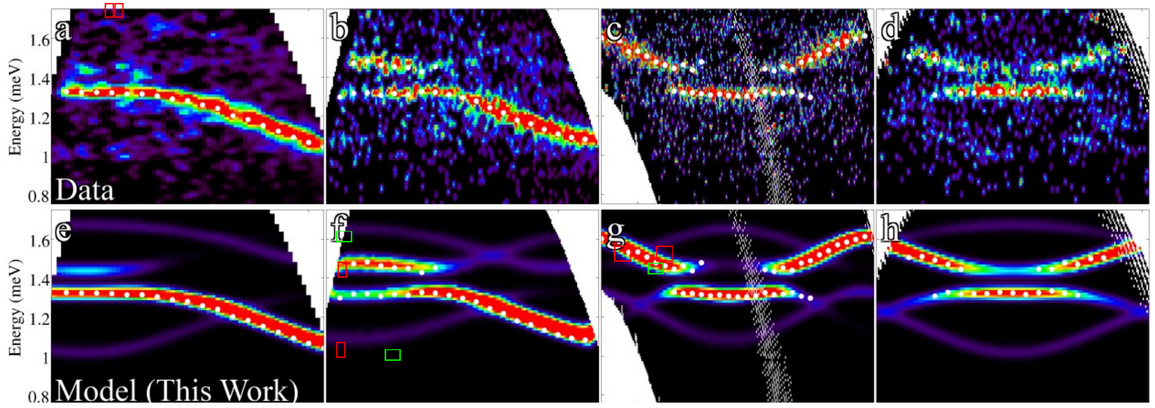
In polarized state spin-wave theory is almost exact → spectrum fit → exchange integrals



$$S^+ \approx a$$
$$S^- \approx a^\dagger$$
$$S^z = S - a^\dagger a$$



$Yb_2Ti_2O_7$ pyrochlore

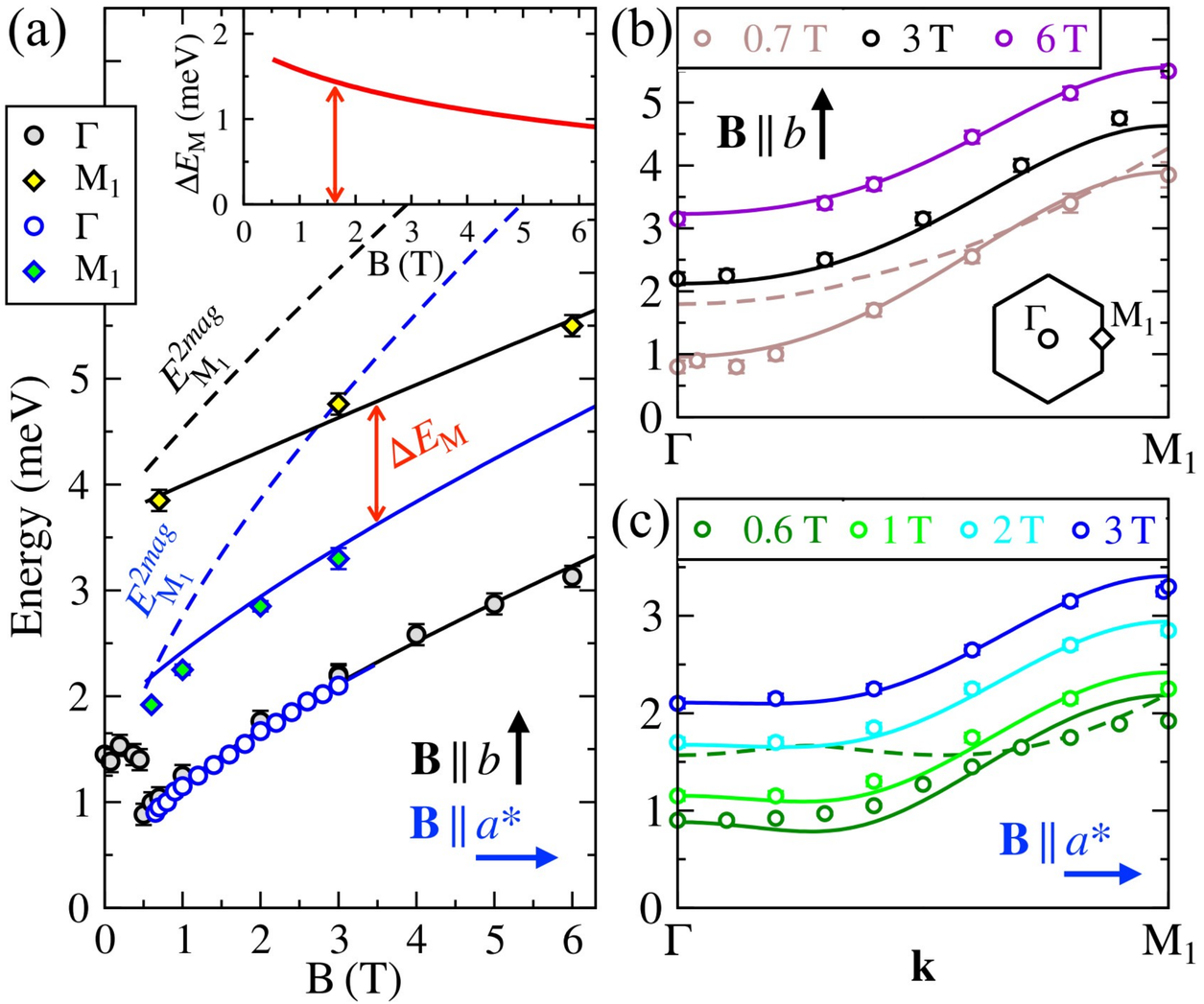


$$\mathcal{H}_{\text{Exchange}} = \sum_{\langle ij \rangle} \{ J_{zz} \mathsf{S}_i^z \mathsf{S}_j^z - J_{\pm} \left(\mathsf{S}_i^+ \mathsf{S}_j^- + \mathsf{S}_i^- \mathsf{S}_j^+ \right) + J_{\pm\pm} \left(\gamma_{ij} \mathsf{S}_i^+ \mathsf{S}_j^+ + \gamma_{ij}^* \mathsf{S}_i^- \mathsf{S}_j^- \right) + J_{z\pm} \left[\mathsf{S}_i^z \left(\zeta_{ij} \mathsf{S}_j^+ + \zeta_{ij}^* \mathsf{S}_i^- \right) + (i \leftrightarrow j) \right] \}$$

6 parameters: 4 exchanges + 2 g-factors

J. D. Thompson et al., Phys. Rev. Lett.119, 057203 (2017)

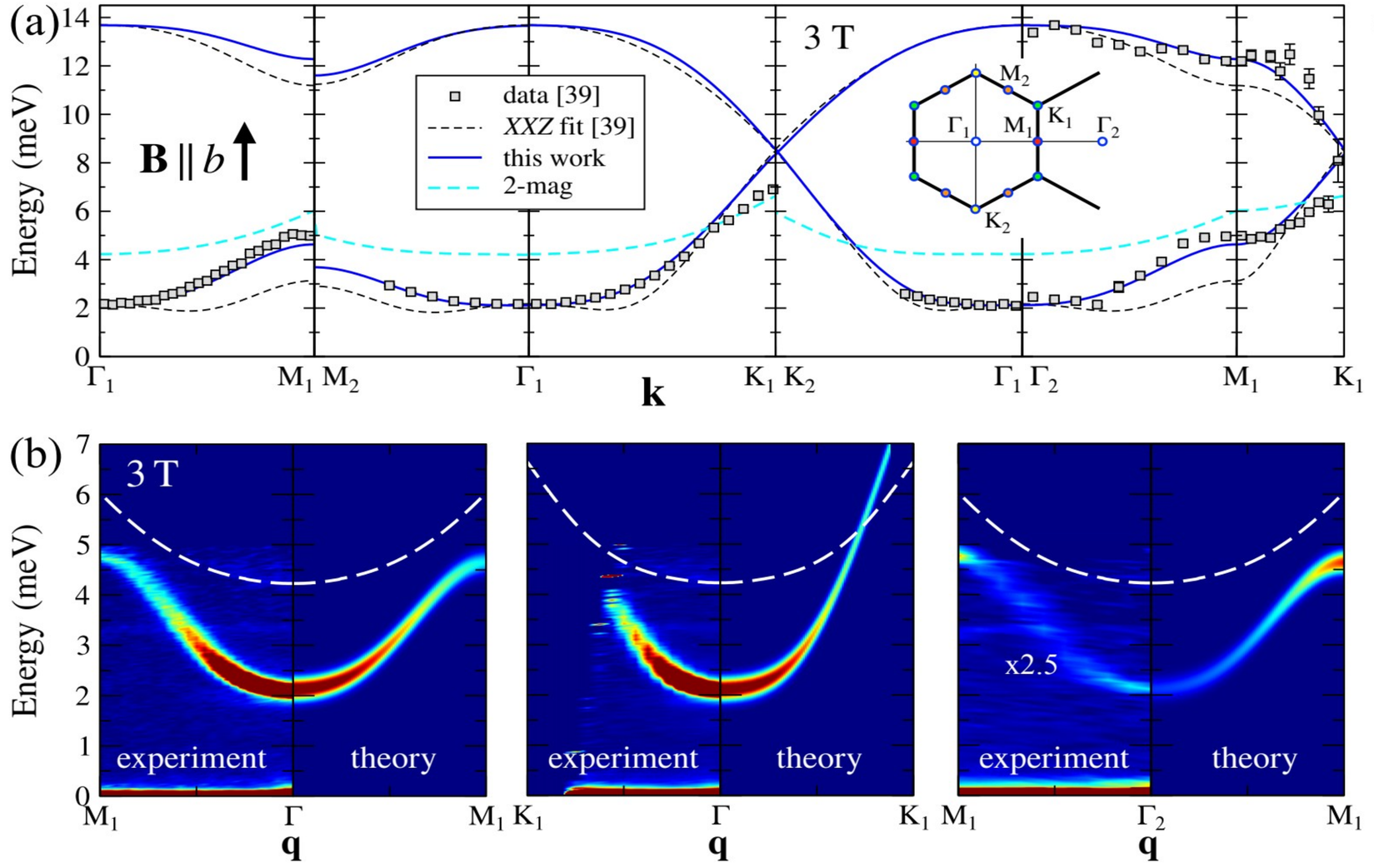
Inelastic neutron scattering in $BaCo_2(AsO_4)_2$



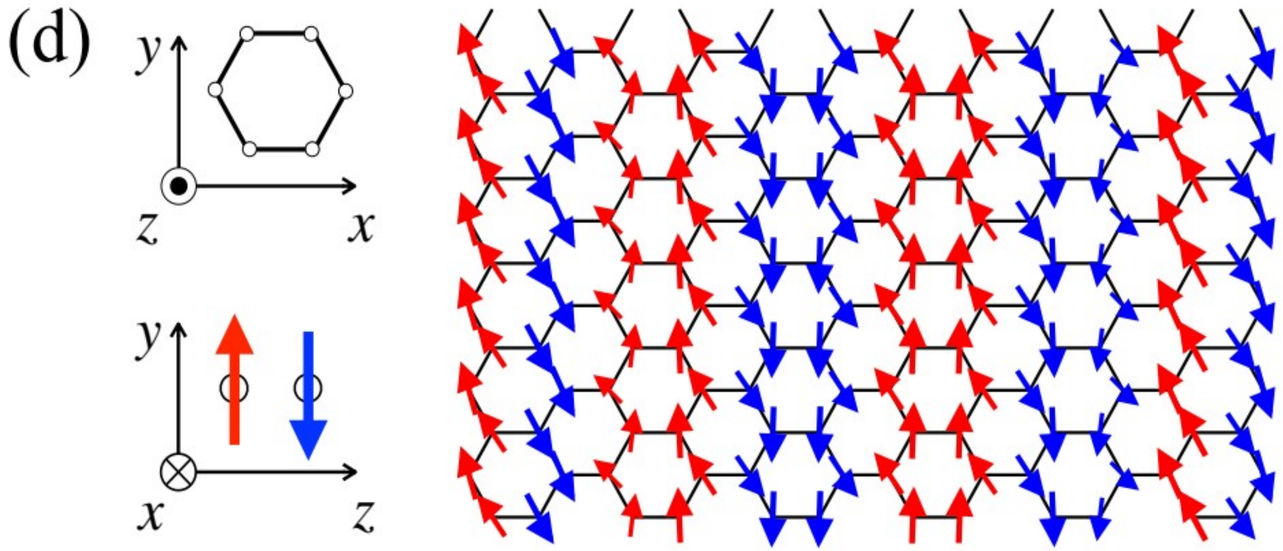
Dots – data from L.P. Regnault, Grenoble
Lines – linear spin-wave theory

$$\{J, K, \Gamma, \Gamma', J_3\} \approx \{-3.3, -5.6, 3.0, 0.6, 1.7\} \text{ meV}$$

Inelastic neutron scattering in $BaCo_2(AsO_4)_2$: fitting quality



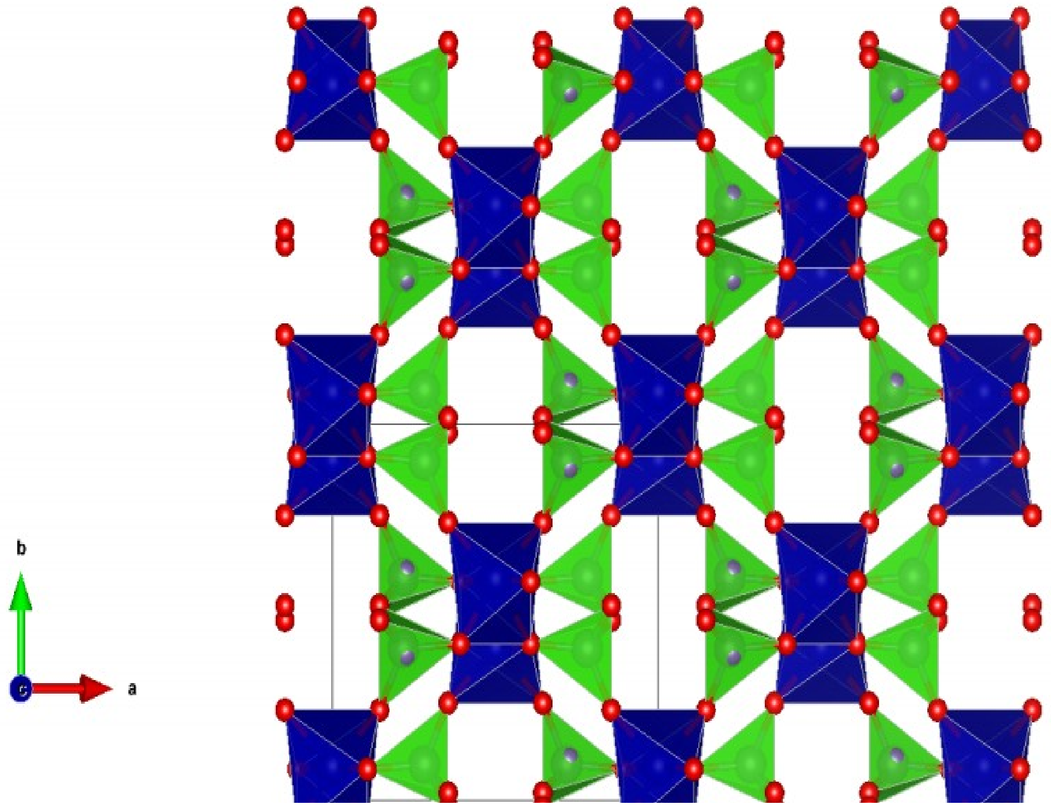
DMRG: double zigzag

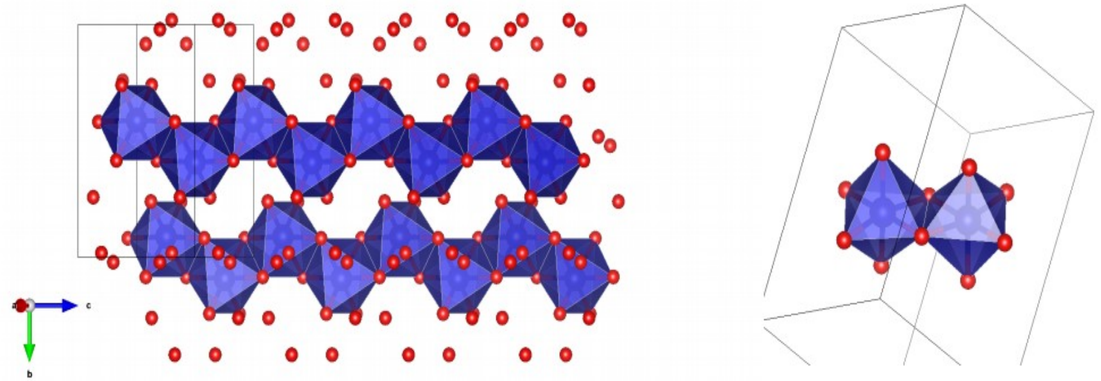


PM, Shengtao Jiang, L.P. Regnault, A.L. Chernyshev, to be published in PRL (2025)

Pyroxene $SrCoGe_2O_6$

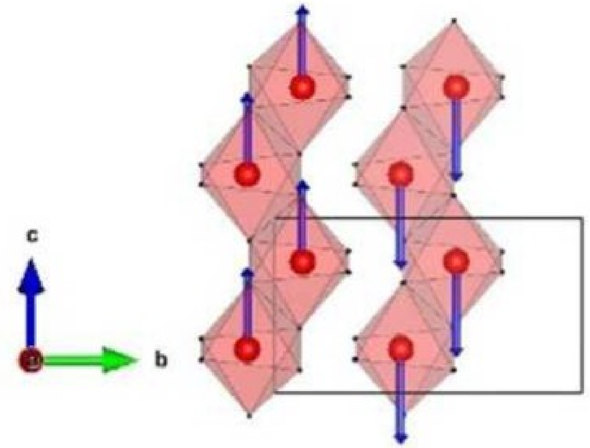
Pyroxene $SrCoGe_2O_6$

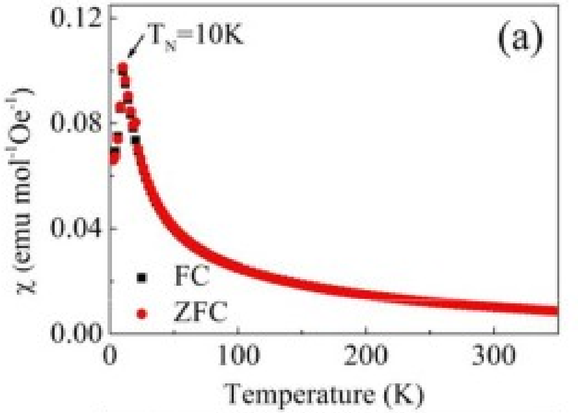


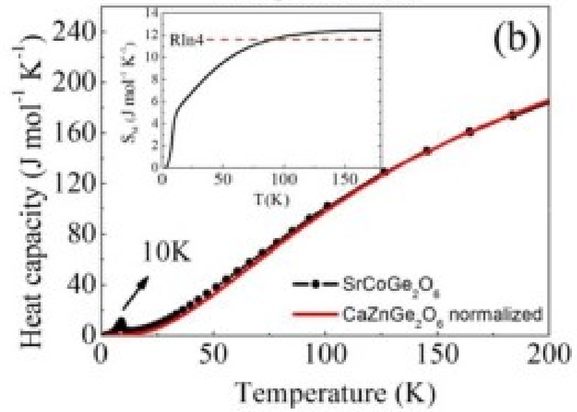


L. Ding, C. V. Colin, C. Darie and P. Bordet, J. Mater. Chem. C, 2016

Magnetic structure







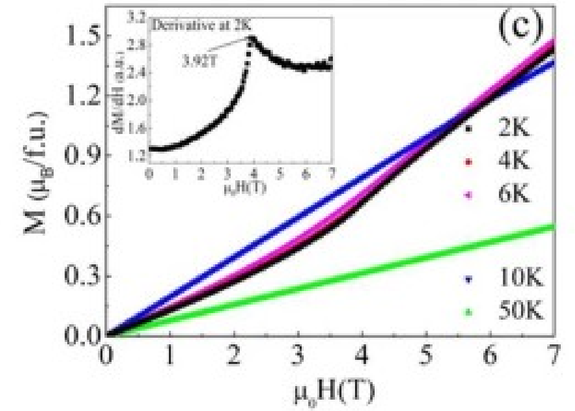
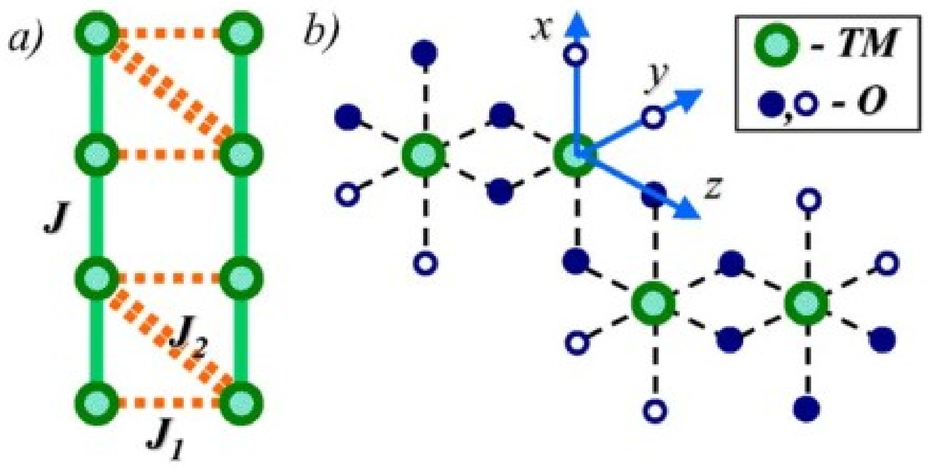
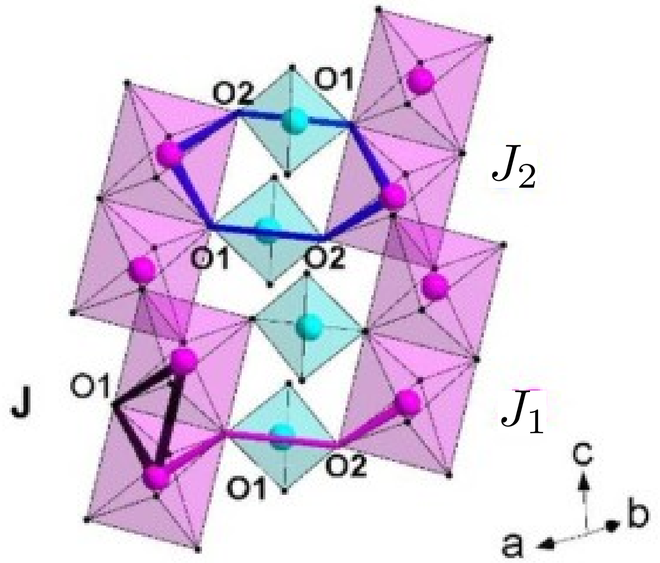


Fig. 8. Representation of the magnetic structure of $SrCoGe_2O_6$

L. Ding, C. V. Colin, C. Darie and P. Bordet, J. Mater. Chem. C, 2016

Ab initio calculations





DFT results:

J=-12 K (FM)

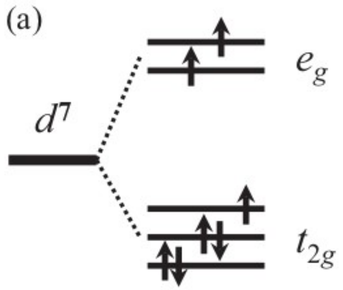
J1= 8 K (AF)

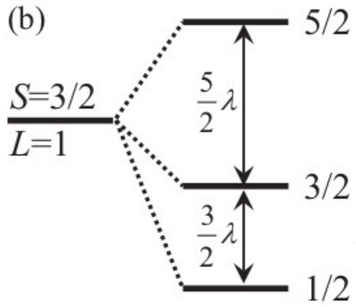
J2= 11 K (AF)

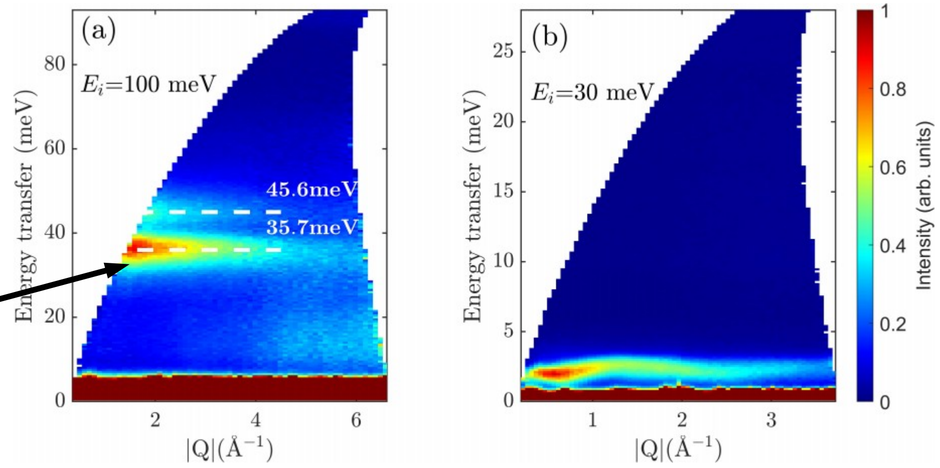
Strong inter-chain coupling → long-range order → spin-wave theory

Effective pseudospin ½ of Co^{2+}

Oak Ridge powder scattering data



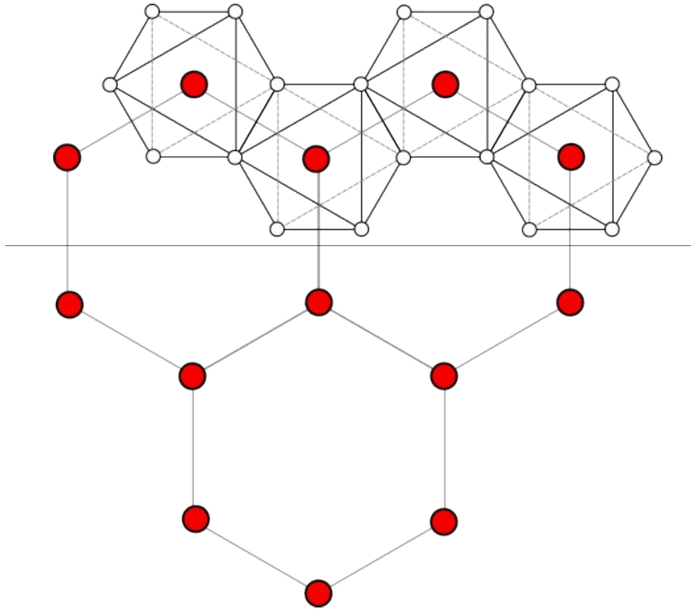


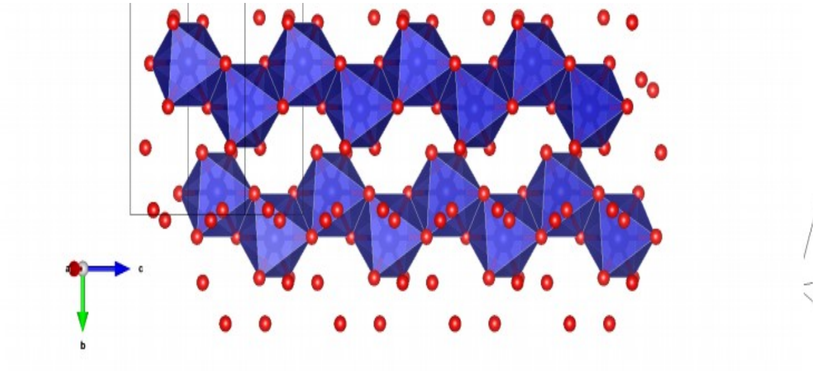


H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)

R. Sano, Y. Kato and Y. Motome, Phys. Rev. B 97, 014408 (2018)

Kitaev-Heisenberg ansatz

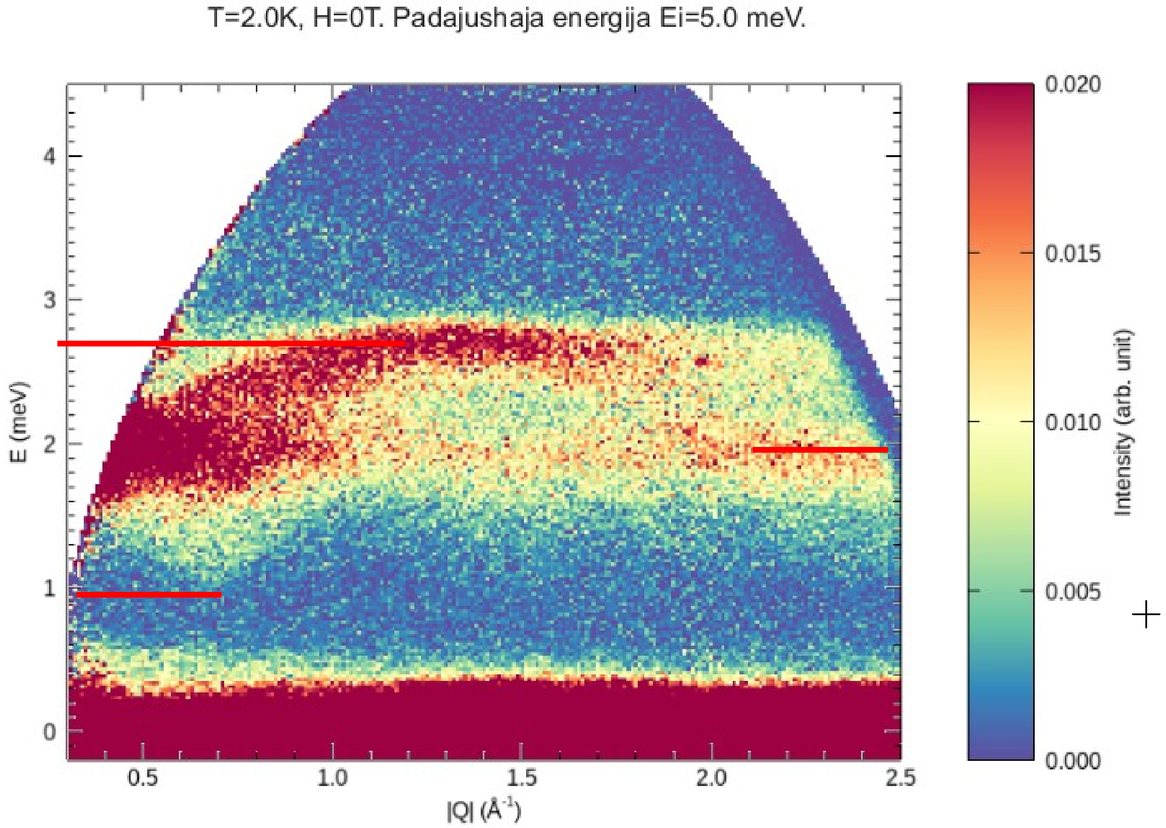


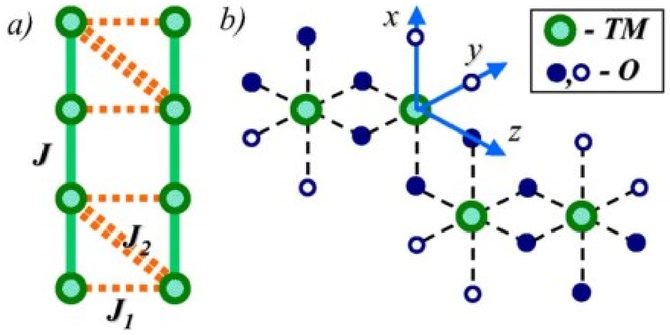


Only x and y bonds

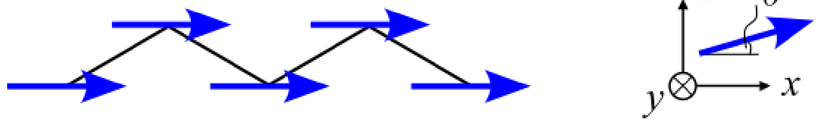
$$\mathrm{H}=\sum_{\langle ij\rangle^{\gamma}} J\mathbf{S}_i\cdot\mathbf{S}_j + KS_i^{\gamma}S_j^{\gamma} + \Gamma\left(S_i^{\alpha}S_j^{\beta}+S_i^{\beta}S_j^{\alpha}\right) + \Gamma'\left(S_i^{\alpha}S_j^{\gamma}+S_i^{\gamma}S_j^{\alpha}+S_i^{\gamma}S_j^{\beta}+S_i^{\beta}S_j^{\gamma}\right)$$

Fit criteria





$$\mathrm{H}=\sum_{\langle ij\rangle^\gamma} J\mathbf{S}_i\cdot\mathbf{S}_j+KS_i^\gamma S_j^\gamma+\Gamma\left(S_i^\alpha S_j^\beta+S_i^\beta S_j^\alpha\right)+\Gamma'\left(S_i^\alpha S_j^\gamma+S_i^\gamma S_j^\alpha+S_i^\gamma S_j^\beta+S_i^\beta S_j^\gamma\right)$$



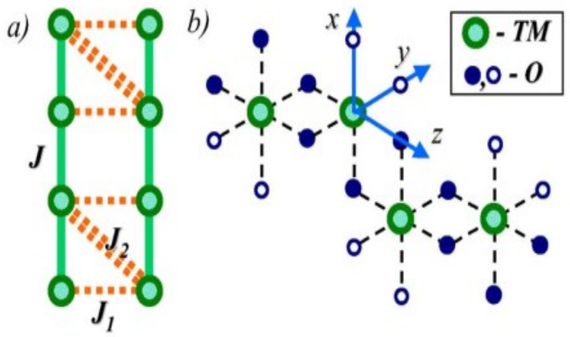
$$\tan 2\alpha=4\sqrt{2}\frac{1+r}{7r-2},\ r=-\frac{\Gamma}{K+\Gamma'}$$

Four critera (can be obtained analytically)

- $\varepsilon_1(0)$=1.0 meV
- ε_1(1/2,1/2,0)=1.9 meV
- ε_2(1/2,1/2,0)=2.7 meV
- Canting angle ~6°

J. Chaloupka and G. Khaliullin, Phys. Rev. B 92, 024413 (2015)

Full exchange model

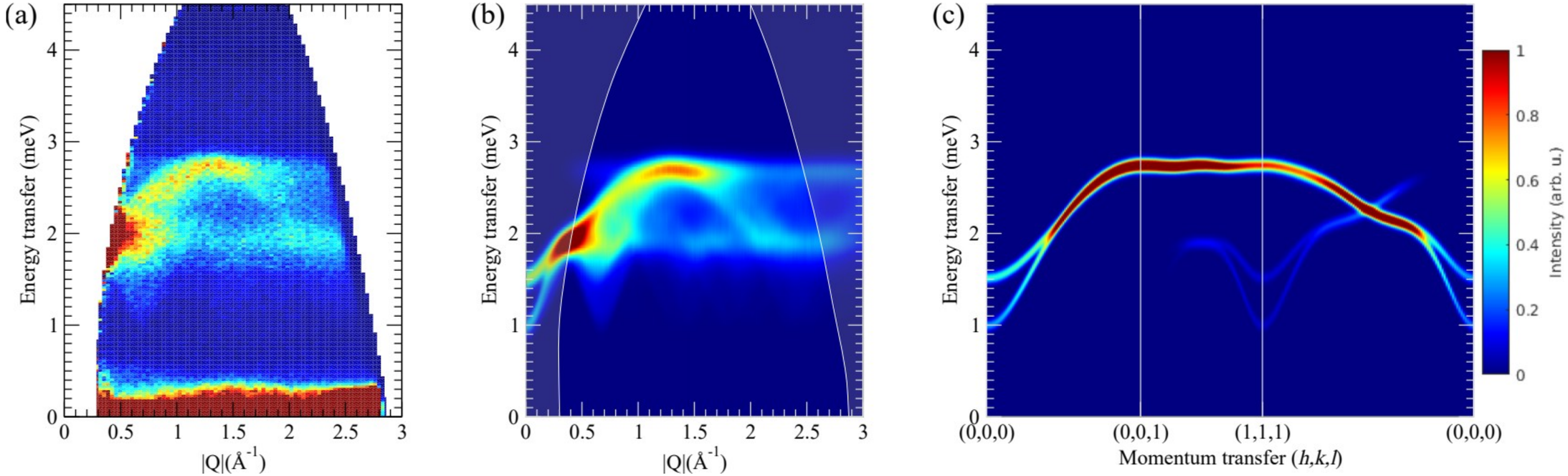


$$\mathrm{H}=\sum_{\langle ij\rangle^{\gamma}} J\mathbf{S}_i\cdot\mathbf{S}_j + KS_i^{\gamma}S_j^{\gamma} + \Gamma\left(S_i^{\alpha}S_j^{\beta}+S_i^{\beta}S_j^{\alpha}\right) + \Gamma'\left(S_i^{\alpha}S_j^{\gamma}+S_i^{\gamma}S_j^{\alpha}+S_i^{\gamma}S_j^{\beta}+S_i^{\beta}S_j^{\gamma}\right)$$

Table 1: Comparison of the exchanges from ab initio (GGA+U+SOC) calculations and neutron scattering fit (LSWT).

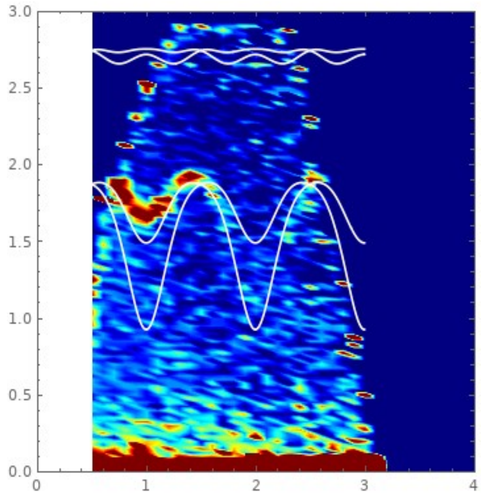
Method	J	K	$\lvert K/J\rvert$	Γ	Γ'	J_1	J_2
GGA+U+SOC	-1.20	1.12	0.93	-	-	0.74	1.06
LSWT	-0.87	0.83	0.96	0.43	-0.26	0.40	0.60

The exchanges are in units of meV.

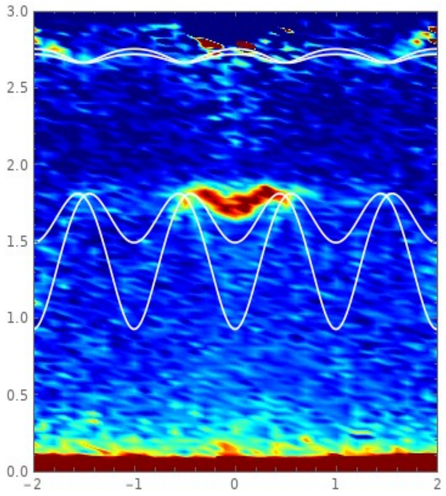


PM et al., PNAS 121 (43) e2409154121 (2024)

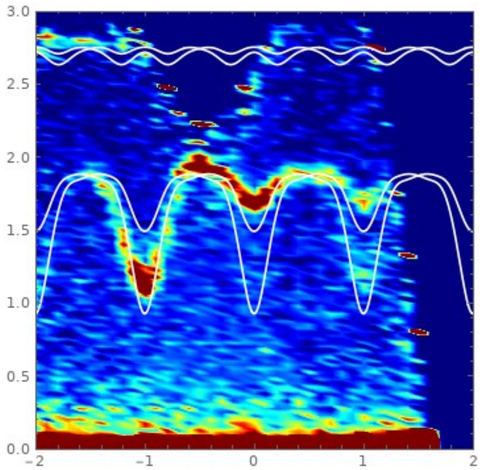
Single crystal neutron scattering



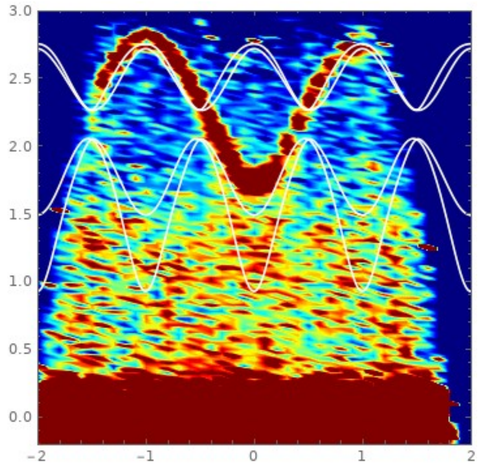
(h00)



(0k0)

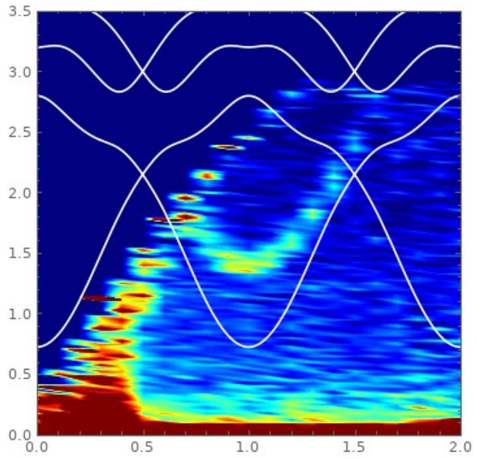


(hk0)



(00l)

H=0T



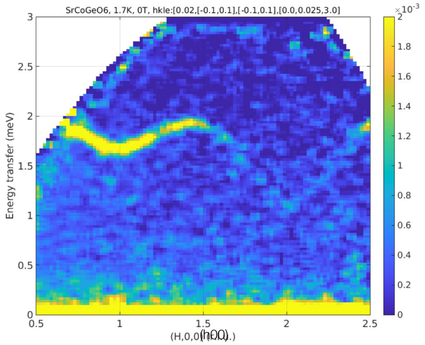
(h00)

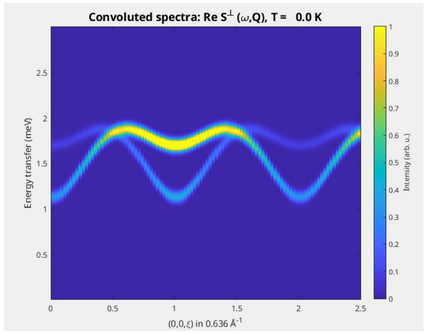
H=13.5T, H||c

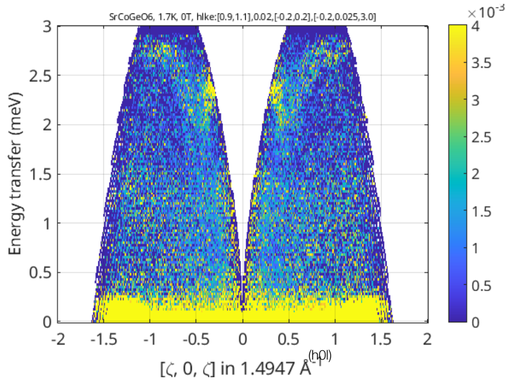
Unpublished data

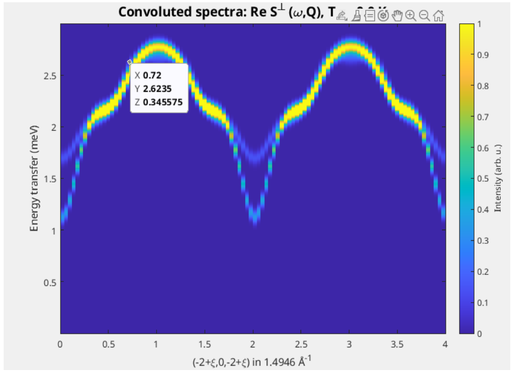
$$\hat{J}_{\text{chain}} = \begin{pmatrix} -1.12 & 0 & 0.13 \\ 0 & -0.1 & 0 \\ 0.13 & 0 & -0.85 \end{pmatrix}$$

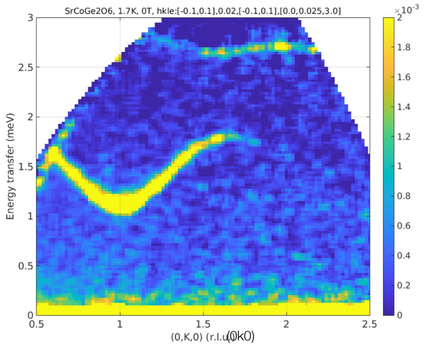
J1=0.2 meV, J2=0.8 meV

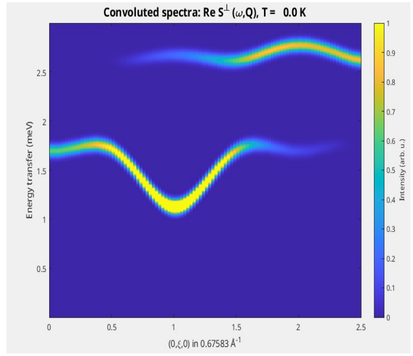


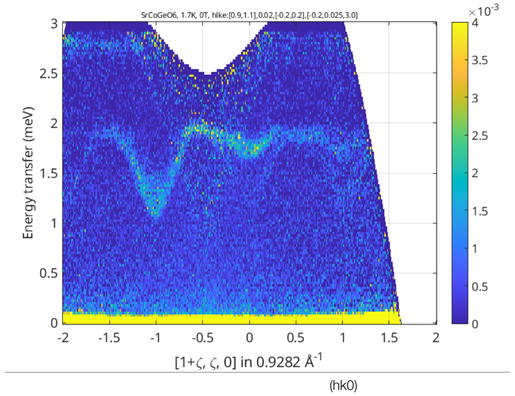


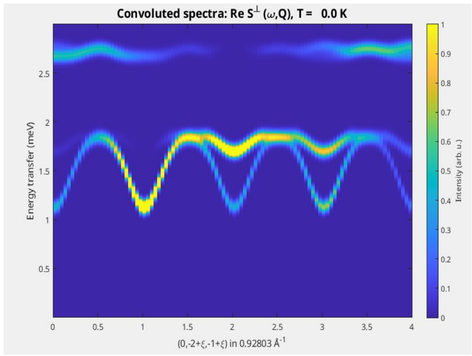


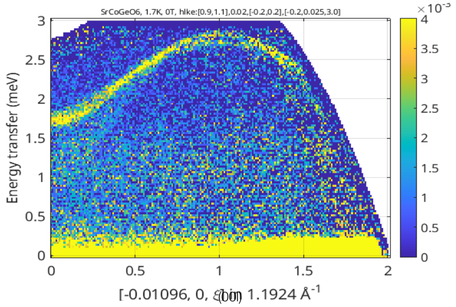


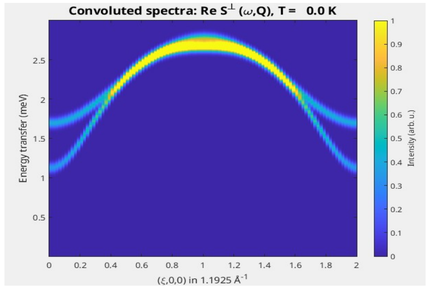


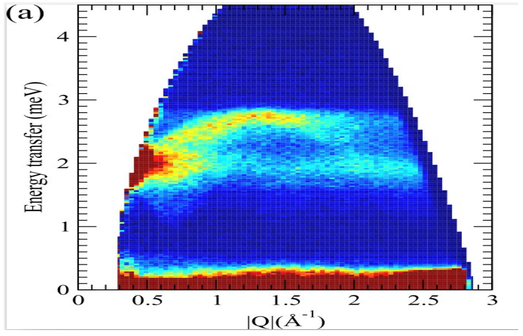


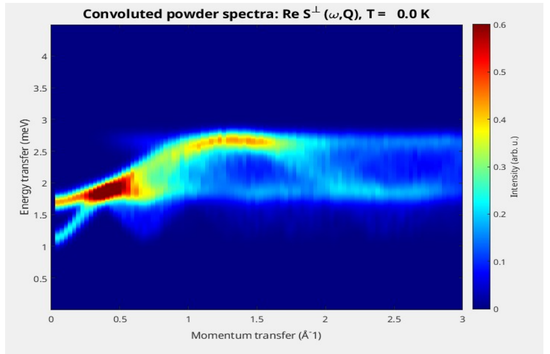








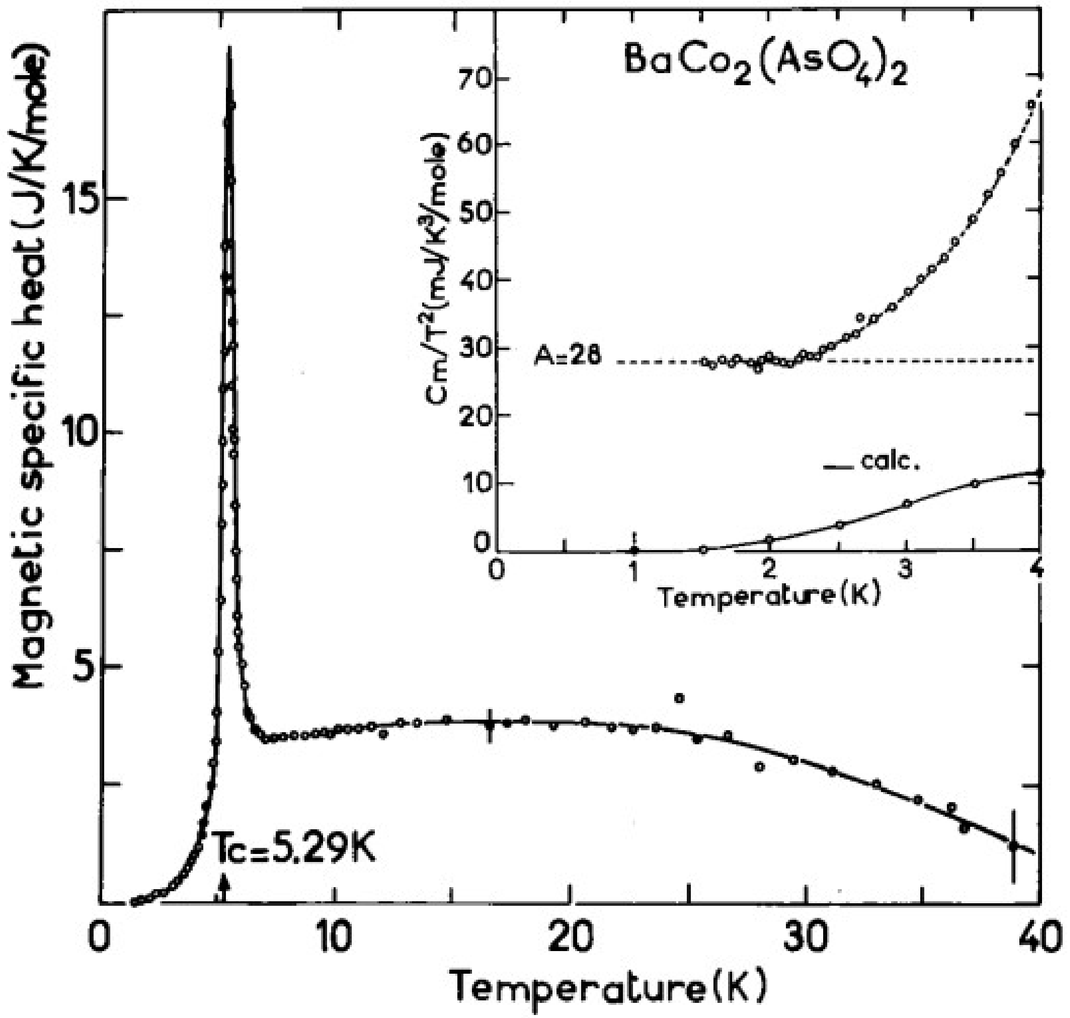




Conclusions

- Co2+ ions can have strongly anisotropic exchanges
- Need to be described by a 3x3 matrix → Kitaev-Heisenberg model, XYZ model, etc
- Inelastic neutron scattering can provide crucial info
- Linear spin-wave theory gets complicated
- Need as much information as possible

Specific heat

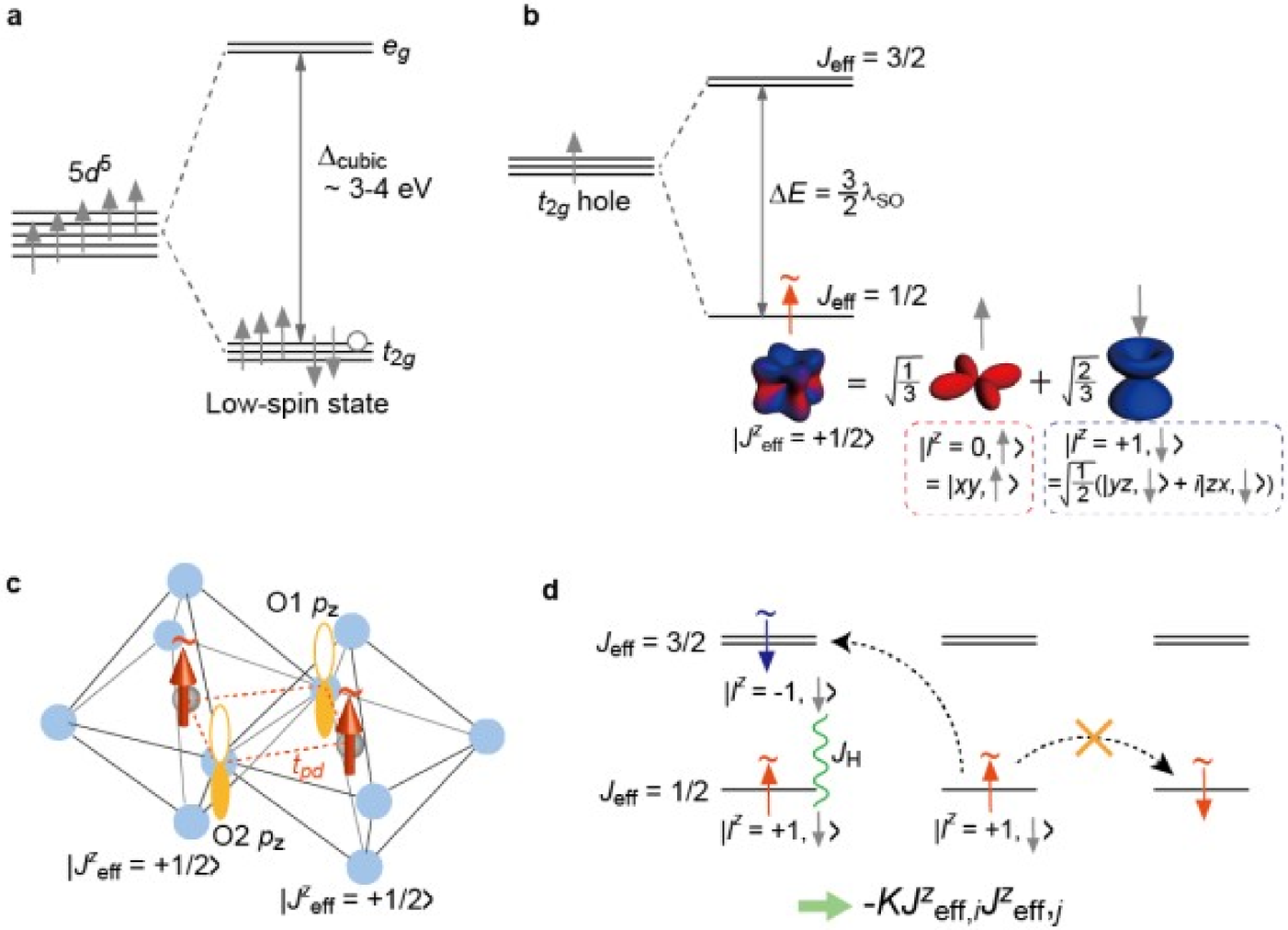


$$C_m - AT^2 = f(T)e^{-\Delta/T}$$

$$\Delta \approx 30K$$

L. Regnault and J. Rossat-Mignod, 1990

Kitaev exchange mechanism



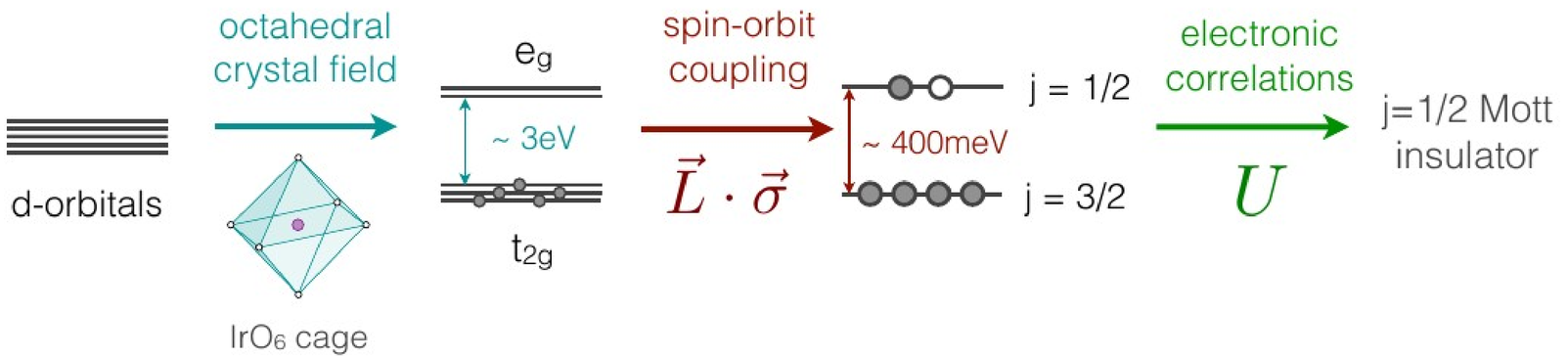
$$|j_{1/2}\rangle = \begin{cases} \frac{1}{\sqrt{3}}(-|xy,\uparrow\rangle - \mathrm{i}|xz,\downarrow\rangle - |yz,\downarrow\rangle) & (m_j = +\frac{1}{2}) \\ \frac{1}{\sqrt{3}}(|xy,\downarrow\rangle + i|xz,\uparrow\rangle - |yz,\uparrow\rangle) & (m_j = -\frac{1}{2}) \end{cases} \tag{4}$$

and

$$|j_{3/2}\rangle = \begin{cases} \frac{1}{\sqrt{2}}(-\mathrm{i}|xz,\uparrow\rangle - |yz,\uparrow\rangle) & (m_j = +\frac{3}{2}) \\ \frac{1}{\sqrt{6}}(2|xy,\uparrow\rangle - \mathrm{i}|xz,\downarrow\rangle - |yz,\downarrow\rangle) & (m_j = +\frac{1}{2}) \\ \frac{1}{\sqrt{6}}(2|xy,\downarrow\rangle - \mathrm{i}|xz,\uparrow\rangle + |yz,\uparrow\rangle) & (m_j = -\frac{1}{2}) \\ \frac{1}{\sqrt{2}}(-\mathrm{i}|xz,\downarrow\rangle + |yz,\downarrow\rangle) & (m_j = -\frac{3}{2}) \end{cases} . \tag{5}$$

Takagi, H., Takayama, T., Jackeli, G. et al., Nat Rev Phys 1, 264 (2019)

Pseudospin: d^5 ion+SOC



$$|j_{1/2}\rangle = \begin{cases} \frac{1}{\sqrt{3}}(-|xy,\uparrow\rangle - \mathrm{i}|xz,\downarrow\rangle - |yz,\downarrow\rangle) & (m_j = +\frac{1}{2}) \\ \frac{1}{\sqrt{3}}(|xy,\downarrow\rangle + i|xz,\uparrow\rangle - |yz,\uparrow\rangle) & (m_j = -\frac{1}{2}) \end{cases}$$

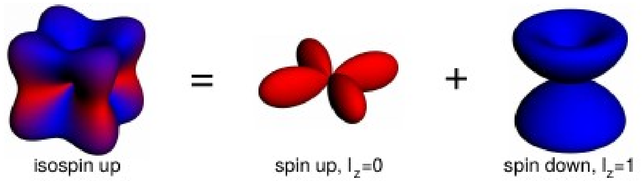
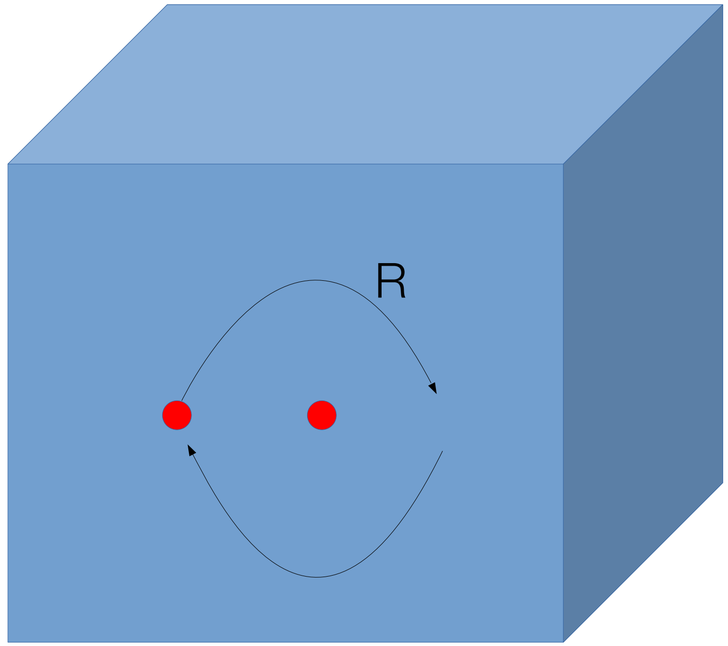


FIG. 1: (Color online) Density profile of a hole in the isospin up state (without tetragonal distortion). It is a superposition of a spin up hole density in $|xy\rangle$-orbital, $l_z = 0$, (middle) and spin down one in $(|yz\rangle + i|xz\rangle)$ state, $l_z = 1$, (right).

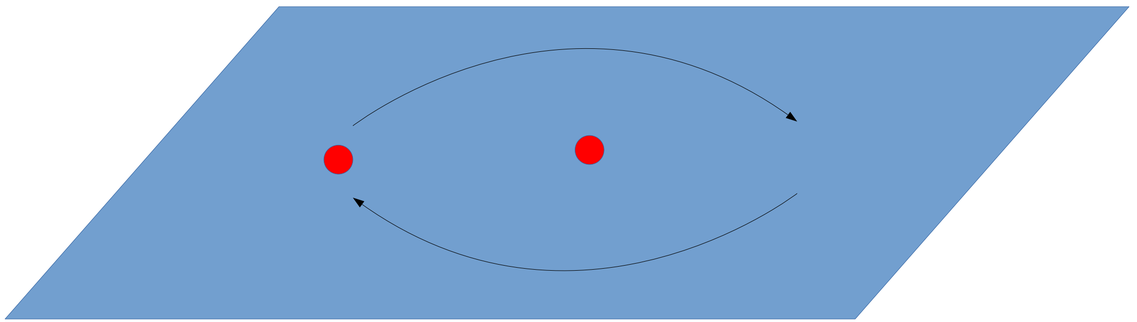
G. Jackeli and G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)

Topological quantum computing



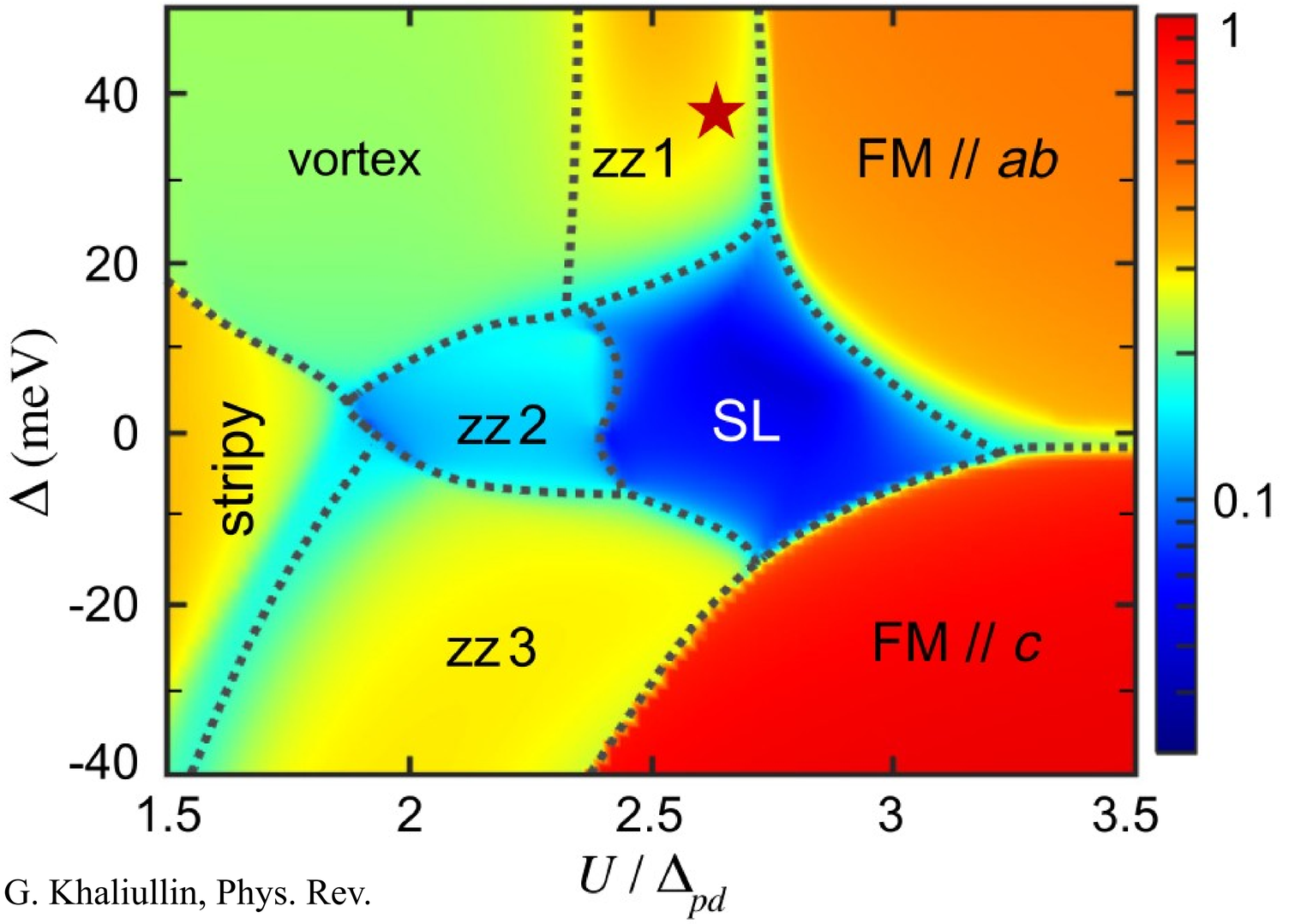
$$R^2 = 1$$

$$R = \pm 1$$



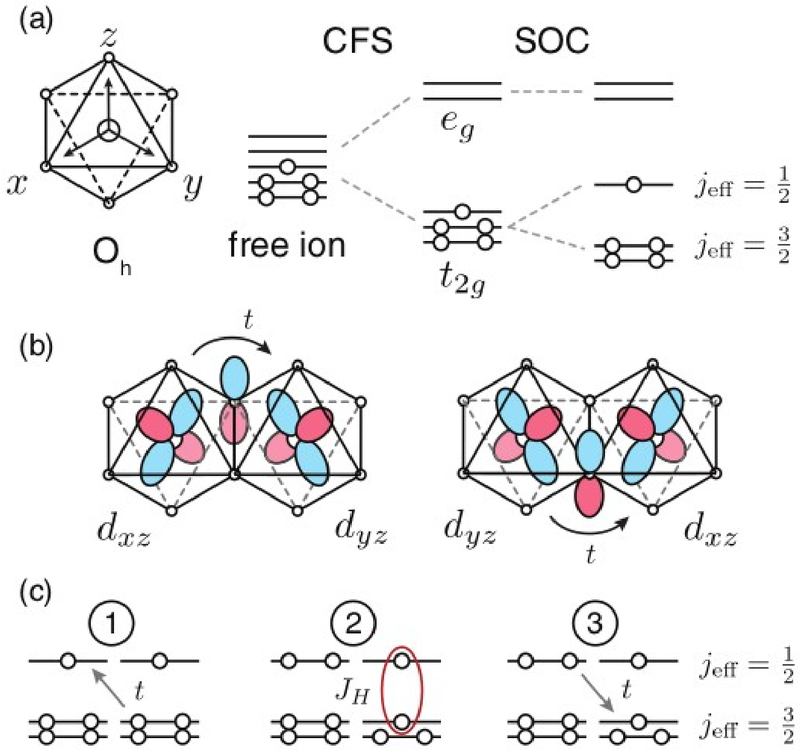
$$R = e^{i\theta}$$

Phase diagram of Co^{2+} honeycomb



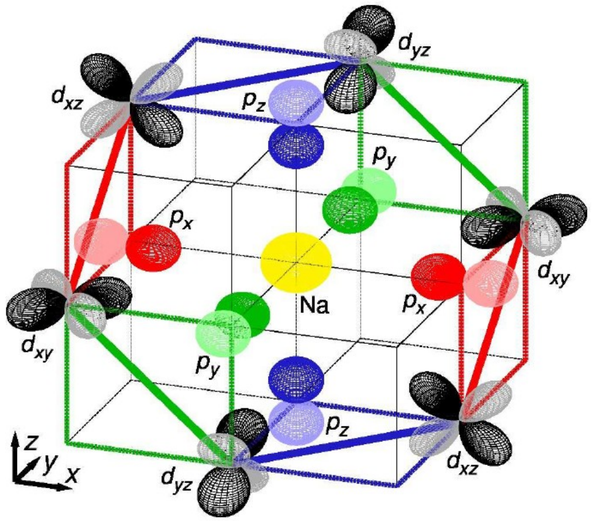
H. Liu, J. Chaloupka and G. Khaliullin, Phys. Rev. Lett. 125, 047201 (2020)

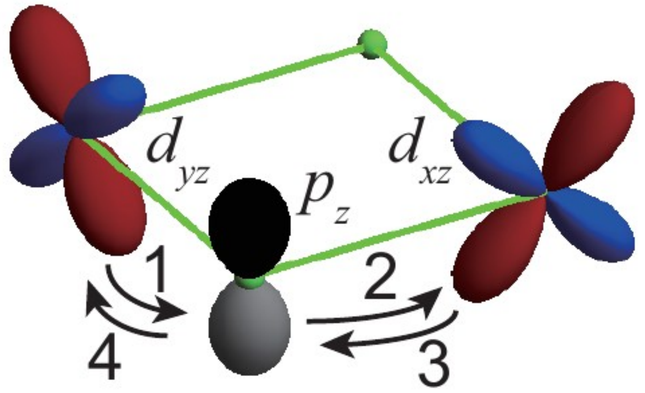
Kitaev materisals: d^5 ions



$$K \sim t^2 J_H / U^2$$

S.M. Winter et al., J. Phys.: Condens. Matter 29 493002 (2017)



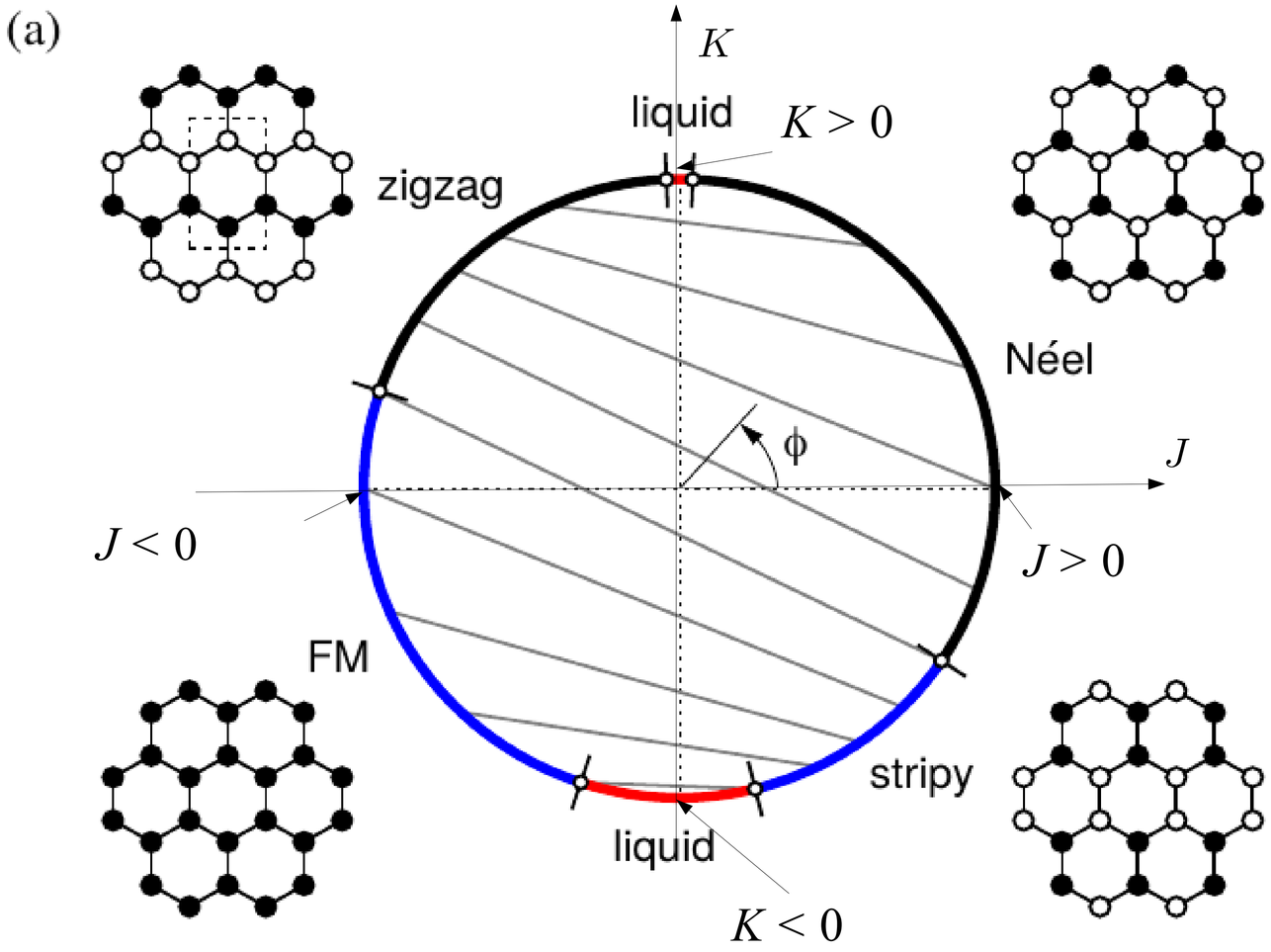


Взаимодействует только z-компонента псевдоспина

$$\mathcal{H} = K \sum_{\langle ij \rangle^\gamma} S_i^\gamma S_j^\gamma$$

G. Jackeli and G. Khaliullin, Phys. Rev. Lett. 102, 017205 (2009)

Фазовая диаграмма модели Китаева-Гейзенберга



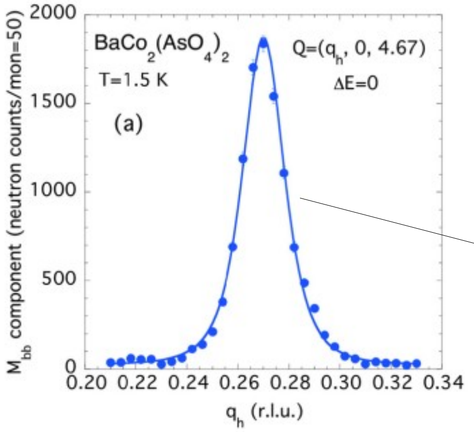
$$\mathcal{H} = \sum_{\langle ij \rangle^\gamma} J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma$$

$$J = \cos\phi, \;\; K = \sin\phi$$

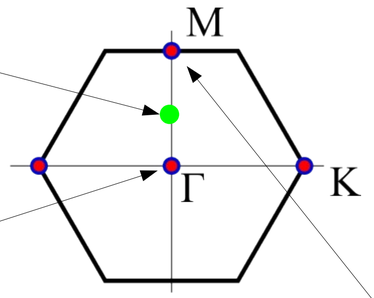
Большая часть фазовой диаграммы — упорядоченные фазы

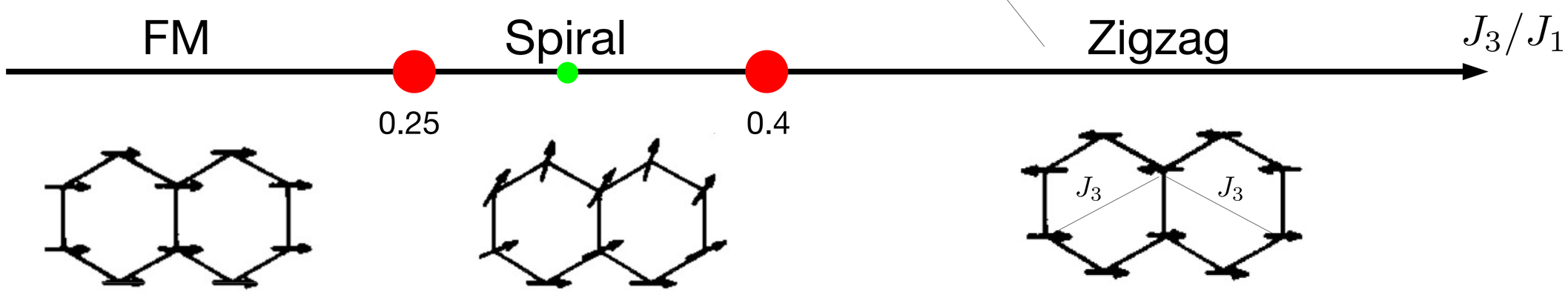
Jiří Chaloupka, George Jackeli, and Giniyat Khaliullin, Phys. Rev. Lett. 110, 097204 (2013)

$BaCo_2(AsO_4)_2$: J_1-J_3 model



$$\mathcal{H} = -J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle ij \rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j$$

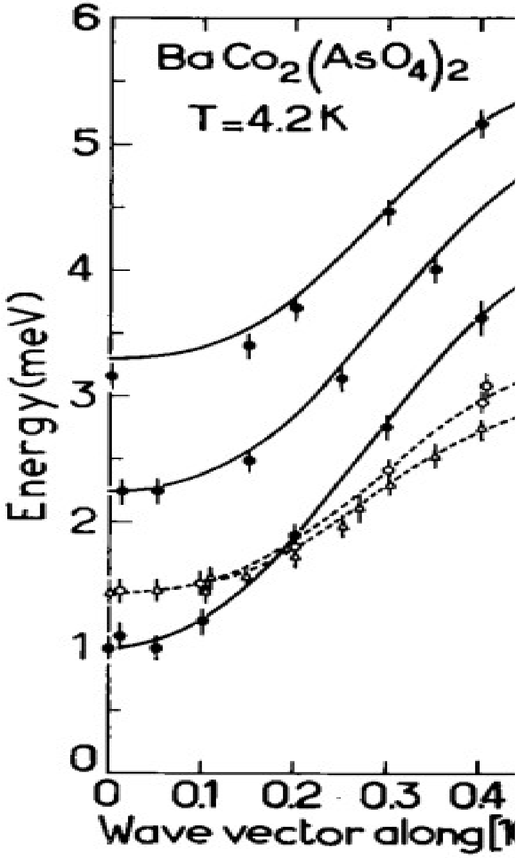




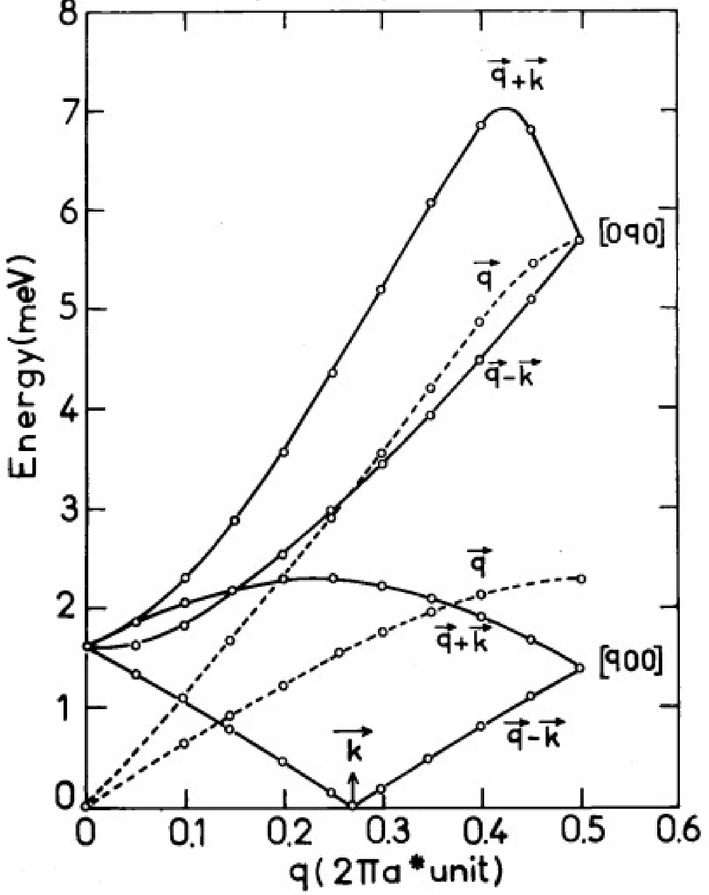
E. Rastelli et al., Physica B+C, 97 1 (1979)

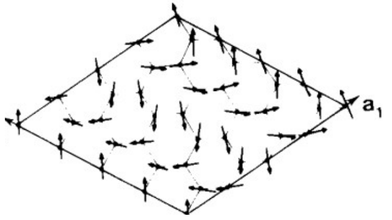
Magnetic excitations

Experiment



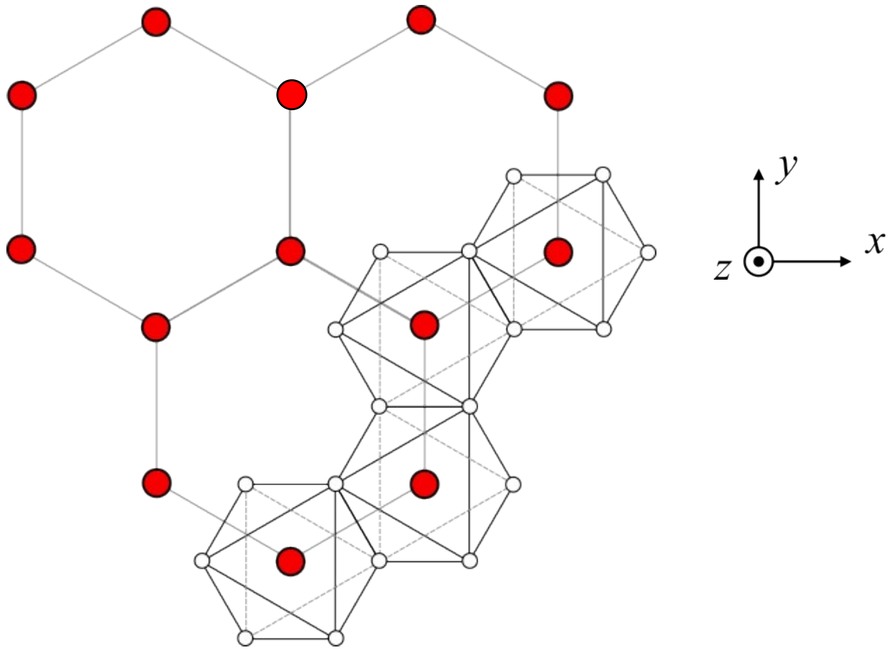
Theory for spiral state





L. Regnault and J. Rossat-Mignod, 1990

Эффективный обменный гамильтониан

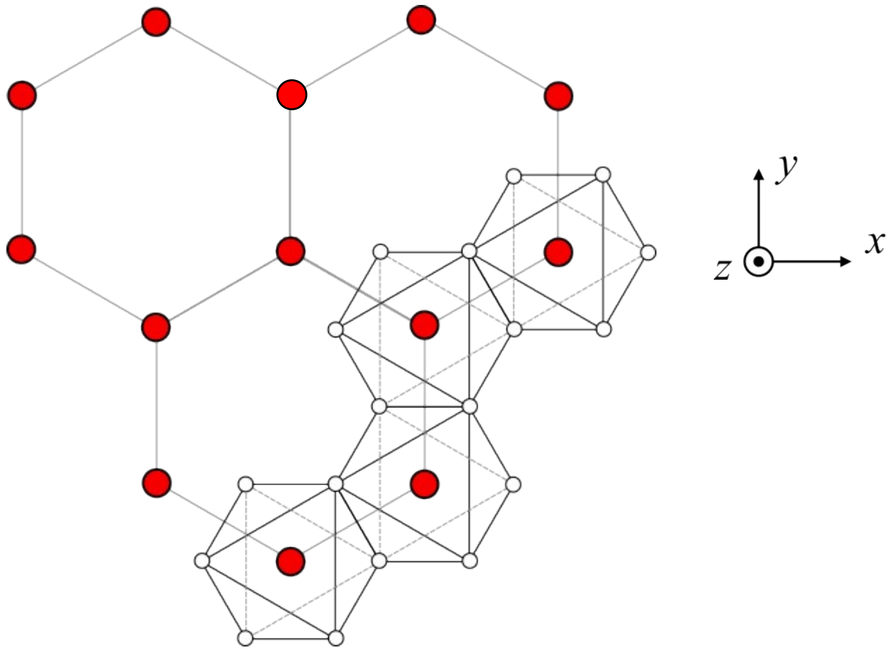


$$\hat{\mathcal{H}}_{ij} = \mathbf{S}_i^{\mathrm{T}} \begin{pmatrix} J_{xx} & J_{xy} & J_{xz} \\ J_{yx} & J_{yy} & J_{yz} \\ J_{zx} & J_{zy} & J_{zz} \end{pmatrix} \mathbf{S}_j$$

Взаимодействия, разрешенные симметрией решетки:

- поворот на π вокруг связи $x \rightarrow -x$, $z \rightarrow -z$
- инверсия относительно узла
- поворот на $2\pi/3$ в плоскости

Эффективный обменный гамильтониан



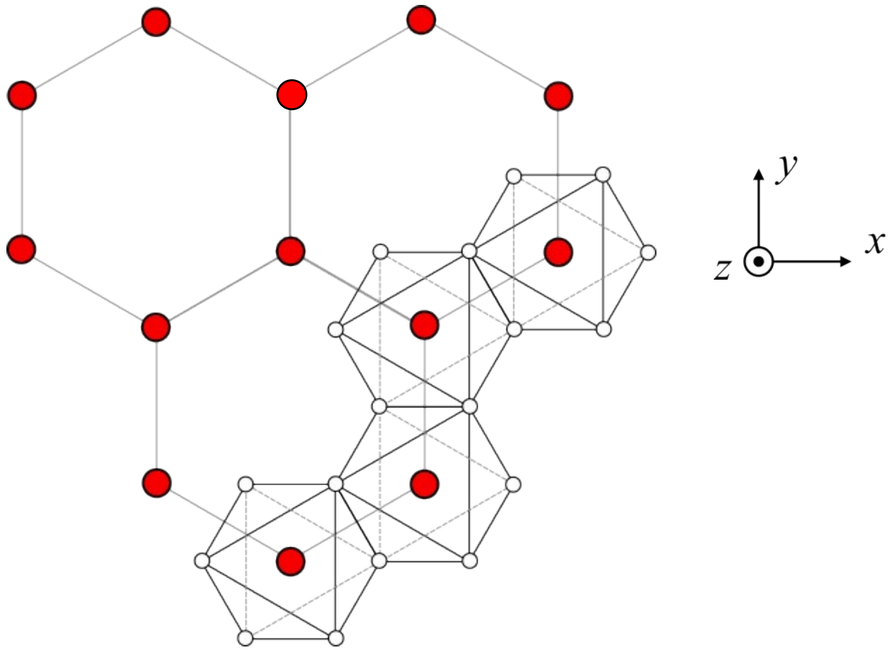
$$\hat{\mathcal{H}}_{ij} = \mathbf{S}_i^{\mathrm{T}} \begin{pmatrix} J_{xx} & \cancel{J_{xy}} & J_{xz} \\ \cancel{J_{yx}} & J_{yy} & \cancel{J_{yz}} \\ J_{zx} & \cancel{J_{zy}} & J_{zz} \end{pmatrix} \mathbf{S}_j$$

$$J_{xz} = J_{zx}$$

Взаимодействия, разрешенные симметрией решетки:

- поворот на π вокруг связи $x \rightarrow -x$, $z \rightarrow -z$
- инверсия относительно узла
- поворот на $2\pi/3$ в плоскости

Эффективный обменный гамильтониан



$$\hat{\mathcal{H}}_{ij} = \mathbf{S}_i^{\mathrm{T}} \begin{pmatrix} J_{xx} & \cancel{J_{xy}} & J_{xz} \\ \cancel{J_{yx}} & J_{yy} & \cancel{J_{yz}} \\ J_{zx} & \cancel{J_{zy}} & J_{zz} \end{pmatrix} \mathbf{S}_j$$

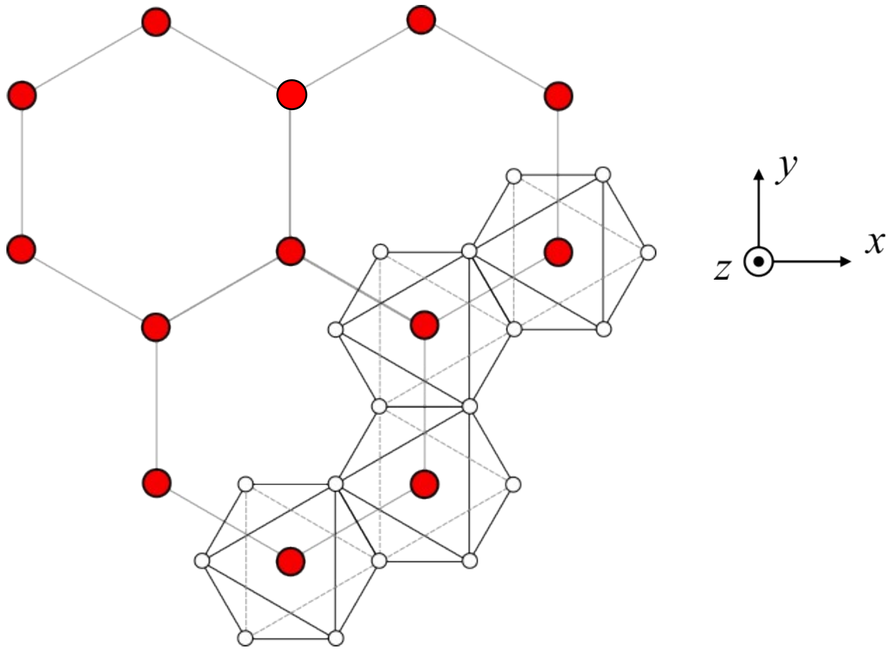
$$J_{xz} = J_{zx}$$

$$\mathrm{H}_{ij} = J\left(\Delta S_i^z S_j^z + S_i^x S_j^x + S_i^y S_j^y\right)$$
$$- 2J_{\pm\pm}\left(S_i^x S_j^x - S_i^y S_j^y\right) - J_{z\pm}\left(S_i^z S_j^x + S_i^x S_j^z\right)$$

Взаимодействия, разрешенные симметрией решетки:

- поворот на π вокруг связи $x \to -x$, $z \to -z$
- инверсия относительно узла
- поворот на $2\pi/3$ в плоскости

Эффективный обменный гамильтониан



$$\mathrm{H}=\sum_{\langle ij\rangle} J\left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right)$$ XXZ

Анизотропный обмен

$$-2J_{\pm\pm}\left(\left(S_i^x S_j^x - S_i^y S_j^y\right)c_\alpha - \left(S_i^x S_j^y + S_i^y S_j^x\right)s_\alpha\right)$$
$$-J_{z\pm}\left(\left(S_i^x S_j^z + S_i^z S_j^x\right)c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y\right)s_\alpha\right)$$

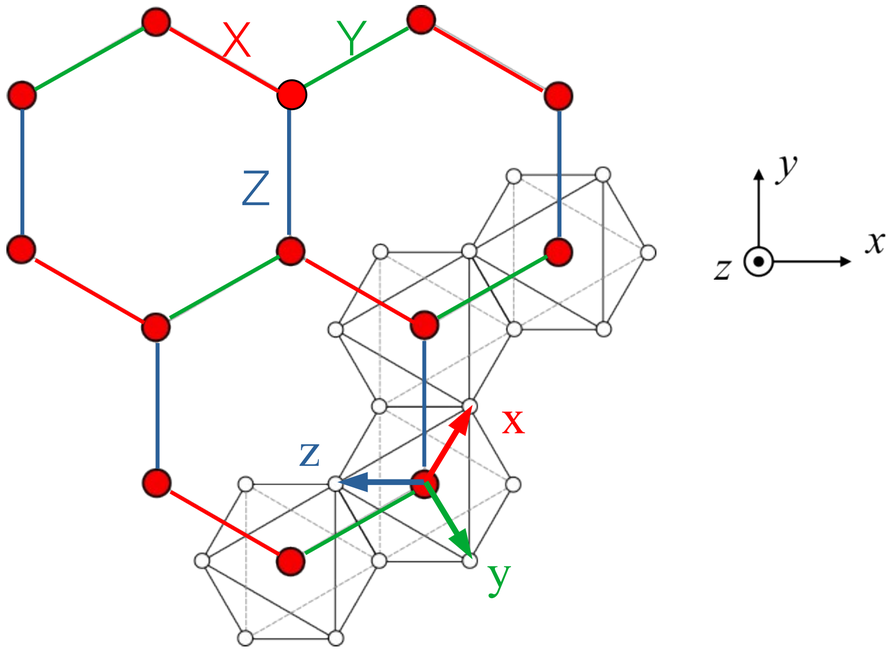
$$c_\alpha(s_\alpha) = \cos(\sin)\varphi_\alpha,\ \ \varphi_\alpha = \{0, 2\pi/3, -2\pi/3\}$$

J.G. Rau, E.K. Lee and H.-Y. Kee, Phys. Rev. Lett. 112, 077204 (2014)
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)
R. Sano, Y. Kato and Y. Motome, Phys. Rev. B 97, 014408 (2018)

Взаимодействия, разрешенные симметрией решетки:

- поворот на π вокруг связи $x \to -x$, $z \to -z$
- инверсия относительно узла
- поворот на $2\pi/3$ в плоскости

Различные формы анизотропного гамильтониана



$$\mathrm{H}=\sum_{\langle ij\rangle} J\left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right)$$
$$- 2J_{\pm\pm}\left(\left(S_i^x S_j^x - S_i^y S_j^y\right)c_\alpha - \left(S_i^x S_j^y + S_i^y S_j^x\right)s_\alpha\right)$$
$$- J_{z\pm}\left(\left(S_i^x S_j^z + S_i^z S_j^x\right)c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y\right)s_\alpha\right)$$

Расширенная модель Китаева-Гейзенберга – кубические оси

$$\mathcal{H} = \sum_{\langle ij\rangle^\gamma} J\mathbf{S}_i\cdot\mathbf{S}_j + KS_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right)$$
$$+\Gamma'\left(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma\right)$$

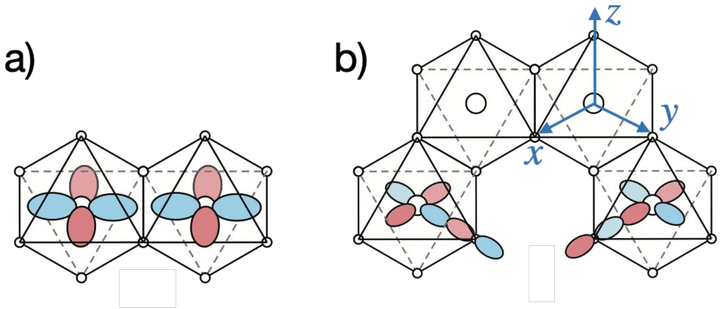
$$\mathrm{J}=\mathrm{J}_0 + \tfrac{1}{3}\left(K-\Gamma-2\Gamma'\right),$$
$$\Delta J = J_0 + \tfrac{1}{3}\left(K+2\Gamma+4\Gamma'\right),$$
$$2J_{\pm\pm} = \tfrac{1}{3}\left(-K-2\Gamma+2\Gamma'\right),$$
$$\sqrt{2}J_{z\pm} = \tfrac{1}{3}\left(2K-2\Gamma+2\Gamma'\right),$$

J.G. Rau, E.K. Lee and H.-Y. Kee, Phys. Rev. Lett. 112, 077204 (2014)
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)
R. Sano, Y. Kato and Y. Motome, Phys. Rev. B 97, 014408 (2018)

$BaCo_2(AsO_4)_2$: *ab initio* exchange integrals

DFT /A.Ushakov, Z.Pchelkina, S.Streltsov (IMP RAS)/
(total energy method)

a) b)

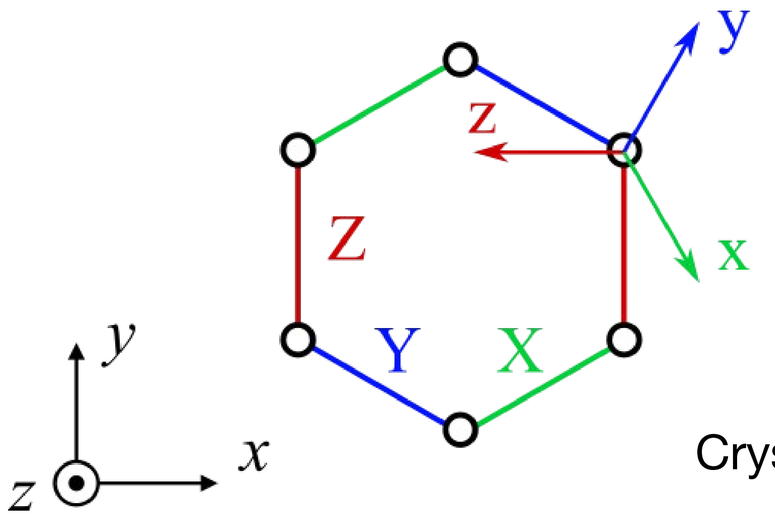


Direct hopping t `~ –300 meV larger than hopping through xz, yz orbitals, t ~ 50 meV
Strong third-neighbor hopping
t_3=124meV
ED /S.Winter, Y. Li (Wake Forest)/
(exact diagonalization)

U	5 eV	6 eV	7 eV
J_1 (K)	-61.0	-40.9	-37.6
K_1 (K)	0.3	2.2	5.3
Γ_1 (K)	-2.2	-1.7	-1.8
Γ_1' (K)	5.1	4.0	3.2
J_3 (K)	31.4	24.6	18.7
K_3 (K)	-0.2	0.2	-0.2
Γ_3 (K)	-4.5	-6.0	-4.5
Γ_3' (K)	-3.6	-2.3	-1.8

$J_{H,t_{2g}}$	0.7 eV		0.9 eV	
U	3.25 eV	5 eV	6 eV	7 eV
J_1 (K)	-107 (-127)	-37 (-57)	-18 (-38)	-8.8 (-29)
K_1 (K)	32	13	6.5	3.4
Γ_1 (K)	28 (35)	14 (21)	8.0 (15)	4.8 (12)
Γ_1' (K)	9.4 (16)	7 (14)	4.0 (11)	2.4 (9)
J_3 (K)	43	30	27	24
K_3 (K)	-0.6	-0.4	-0.3	-0.3
Γ_3 (K)	-20	-12	-10	-8.9
Γ_3' (K)	-21	-12	-11	-9.2

Kitaev model in «natural» reference frame



Cubic axes
(ligand octahedra)
{x,y,z}

$$\mathrm{H}=\sum_{\langle ij\rangle^\gamma} J\mathbf{S}_i\cdot\mathbf{S}_j + KS_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right) + \Gamma'\left(S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma\right)$$

Crystallographic axes
(lattice plane)
{*x,y,z*}

$$\mathrm{H}=\sum_{\langle ij\rangle} J\left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right)$$

XXZ

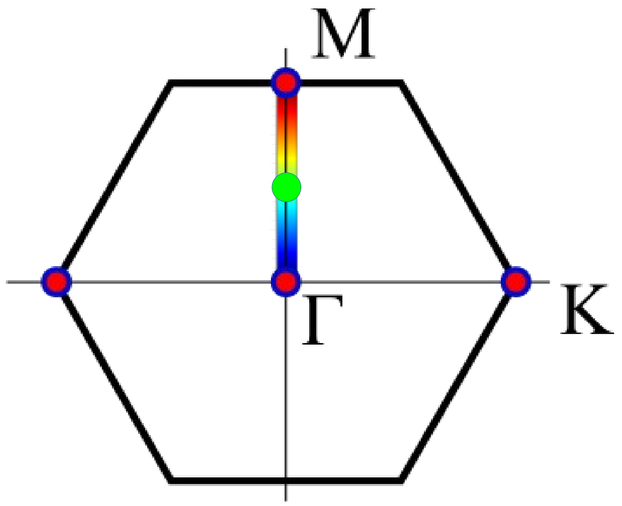
$$- 2J_{\pm\pm}\left(\left(S_i^x S_j^x - S_i^y S_j^y\right)c_\alpha - \left(S_i^x S_j^y + S_i^y S_j^x\right)s_\alpha\right)$$
$$- J_{z\pm}\left(\left(S_i^x S_j^z + S_i^z S_j^x\right)c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y\right)s_\alpha\right)$$

Pseudodipolar — bond-dependent

$$\mathrm{J}=\mathrm{J}_0+\tfrac{1}{3}\left(K-\Gamma-2\Gamma'\right),$$
$$\Delta J = J_0+\tfrac{1}{3}\left(K+2\Gamma+4\Gamma'\right),$$
$$2J_{\pm\pm} = \tfrac{1}{3}\left(-K-2\Gamma+2\Gamma'\right),$$
$$\sqrt{2}J_{z\pm} = \tfrac{1}{3}\left(2K-2\Gamma+2\Gamma'\right),$$

$$c_\alpha \equiv \cos\phi_\alpha$$
$$\phi_\alpha = \{0, 2\pi/3, -2\pi/3\}$$

J_1-J_3+XXZ model

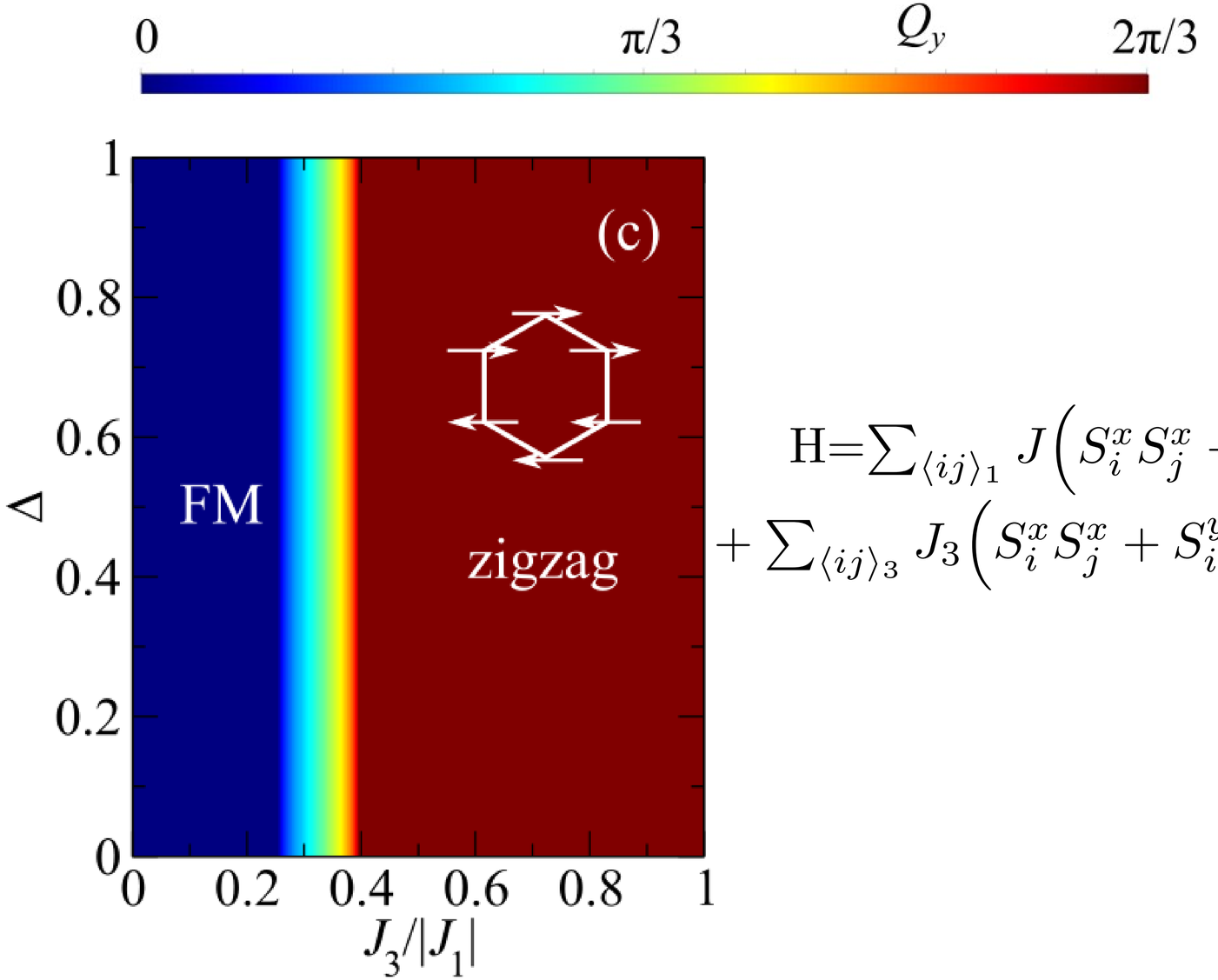


Luttinger-Tisza method:

$$\mathcal{H} = \sum_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} J_{\mathbf{q}} \mathbf{S}_{-\mathbf{q}}$$

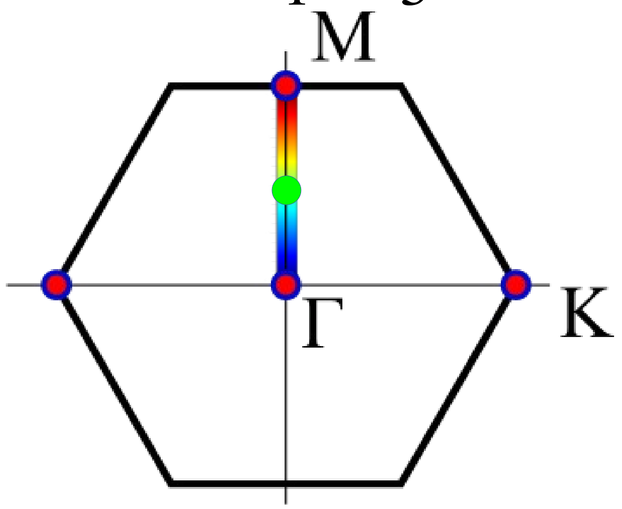
$$\mathbf{S}_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} e^{i\mathbf{q}\mathbf{r}_i}$$

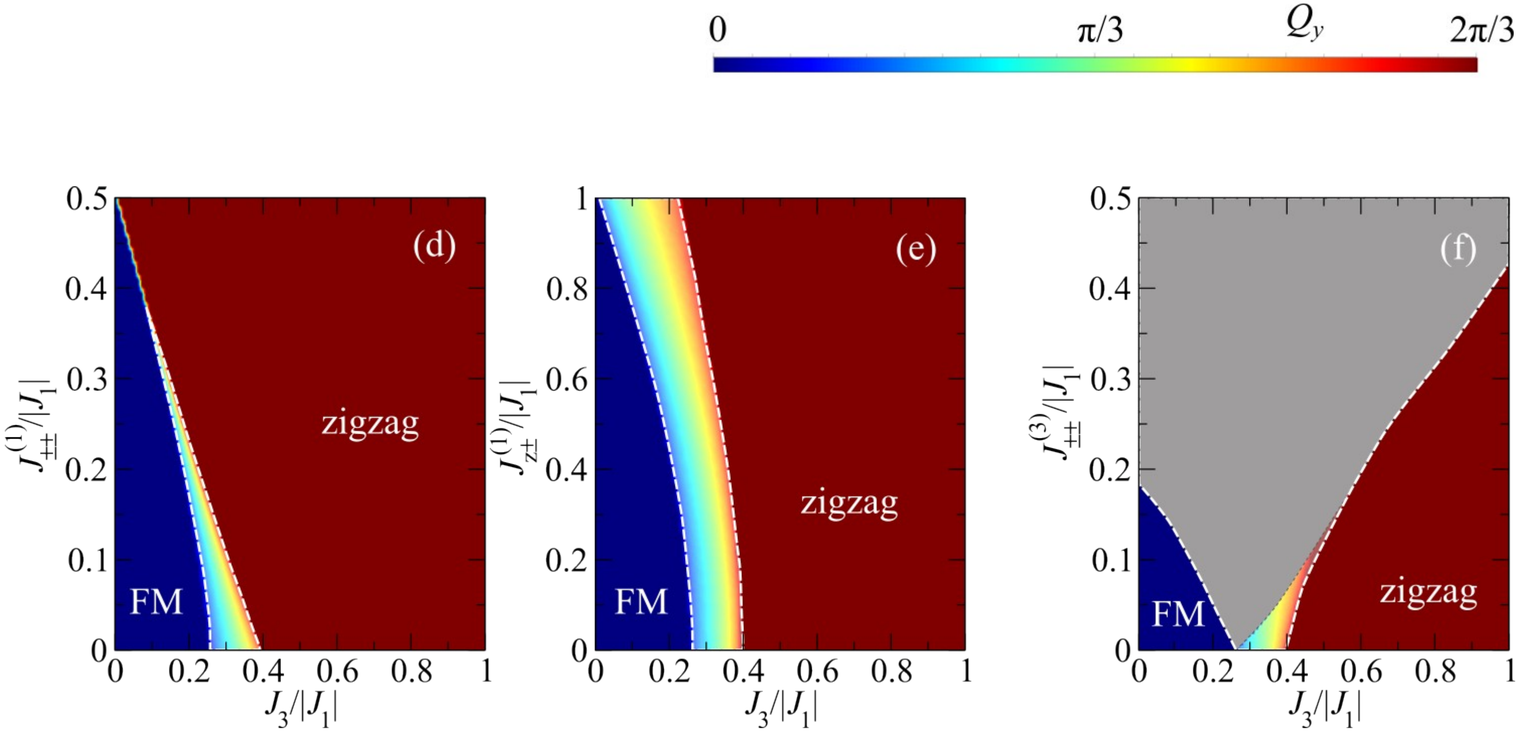
$$\sum_i |\mathbf{S}_i|^2 = NS^2$$



$$\mathrm{H} = \sum_{\langle ij \rangle_1} J \left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z \right) + \sum_{\langle ij \rangle_3} J_3 \left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z \right)$$

J_1-J_3+XXZ model+bond-dependent terms





All bond-dependent terms suppress (0,Q_y) spiral at large J_3

$$\mathrm{H}=\sum_{\langle ij\rangle} J\left(S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z\right)$$
$$-2J_{\pm\pm}\left(\left(S_i^x S_j^x - S_i^y S_j^y\right)c_\alpha - \left(S_i^x S_j^y + S_i^y S_j^x\right)s_\alpha\right)$$
$$-J_{z\pm}\left(\left(S_i^x S_j^z + S_i^z S_j^x\right)c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y\right)s_\alpha\right)$$

BaCo2(AsO4)2

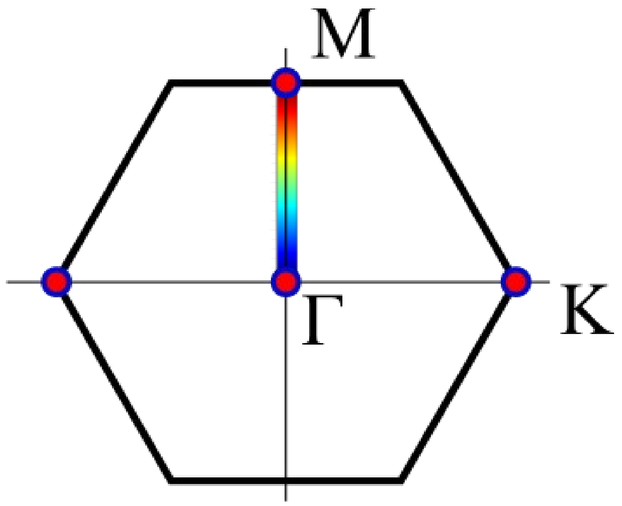
TABLE VI. Local exchange couplings for $BaCo_2(AsO_4)_2$.

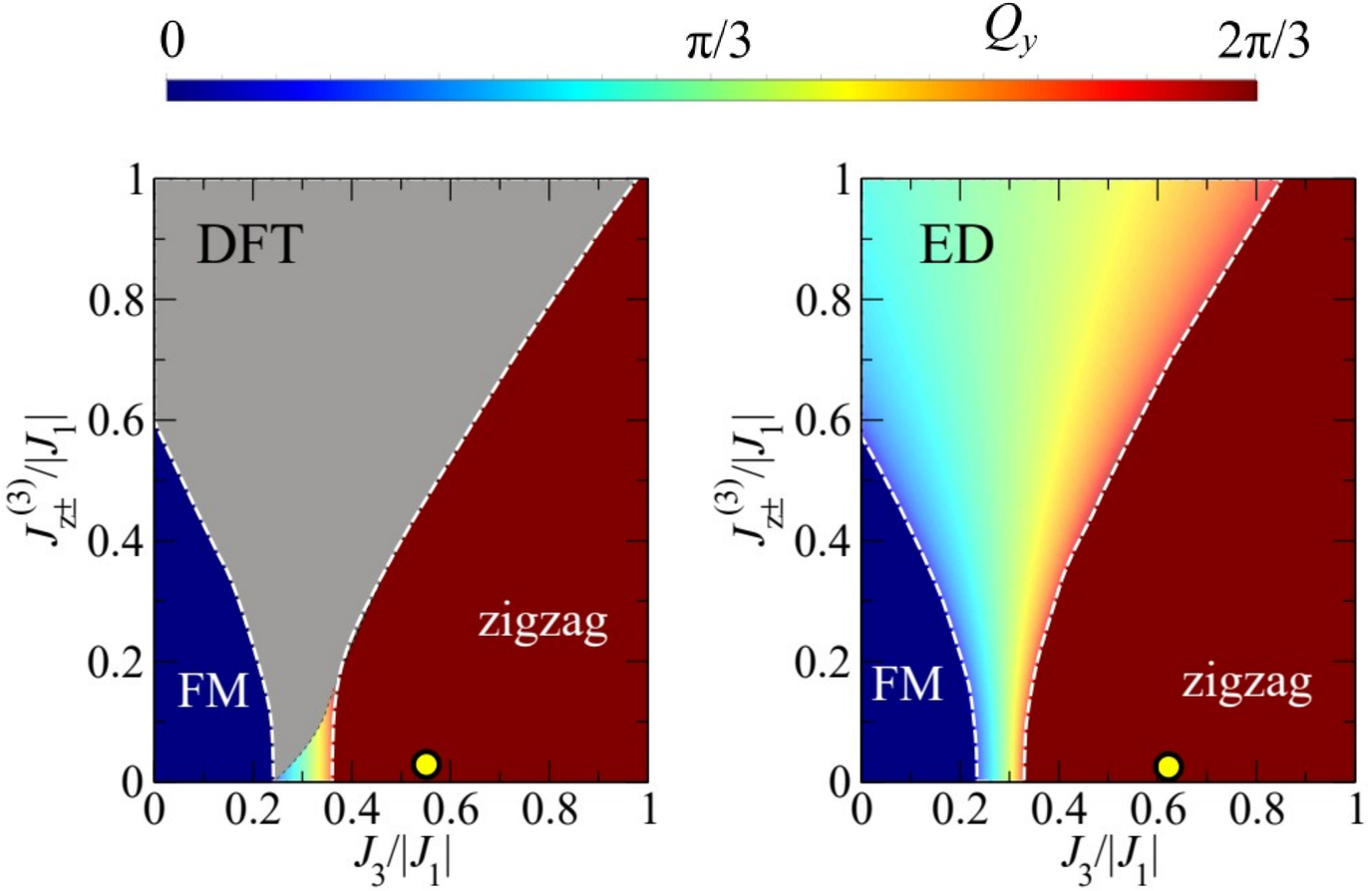
$BaCo_2(AsO_4)_2$	Bond-1	Bond-3
J	−12.40	5.51
K	2.06	−0.30
Γ	4.05	−1.44
η	−0.84	0.00
Γ'_1	1.82	−1.42
Γ'_2	4.89	−1.45

$BaCo_2(AsO_4)_2$	Bond-1	Bond-3
J^{XY}	−14.5 meV	7.1 meV
J^{Z}	−3.8 meV	2.3 meV
D	1.5 meV	≈0
E	−1.7 meV	≈0

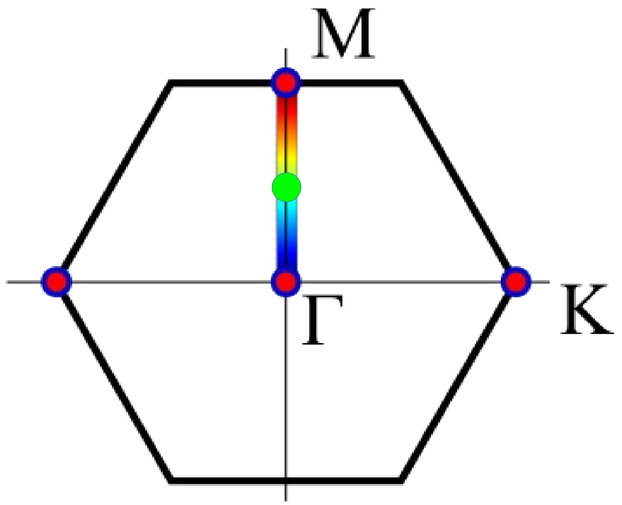
S. Das et al., Phys. Rev. B 104, 134425 (2021)

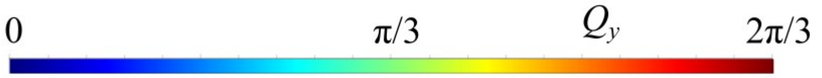
Ab initio ground state





Minimal model for $BaCo_2(AsO_4)_2$

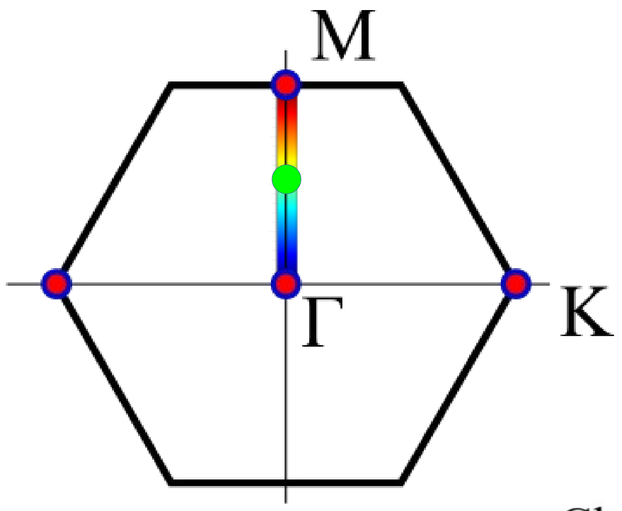




$$\mathrm{H}_{\mathrm{min}} = \sum_{\langle ij \rangle_1} J_1 \left(S_i^x S_j^x + S_i^y S_j^y \right) + J_3 \sum_{\langle ij \rangle_3} \left(S_i^x S_j^x + S_i^y S_j^y \right)$$
$$- J_{z\pm}^{(3)} \left(\left(S_i^x S_j^z + S_i^z S_j^x \right) c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y \right) s_\alpha \right),$$

$$\mathrm{J}_{z\pm} = \frac{\sqrt{2}}{3} \left(K - \Gamma + \Gamma' \right)$$

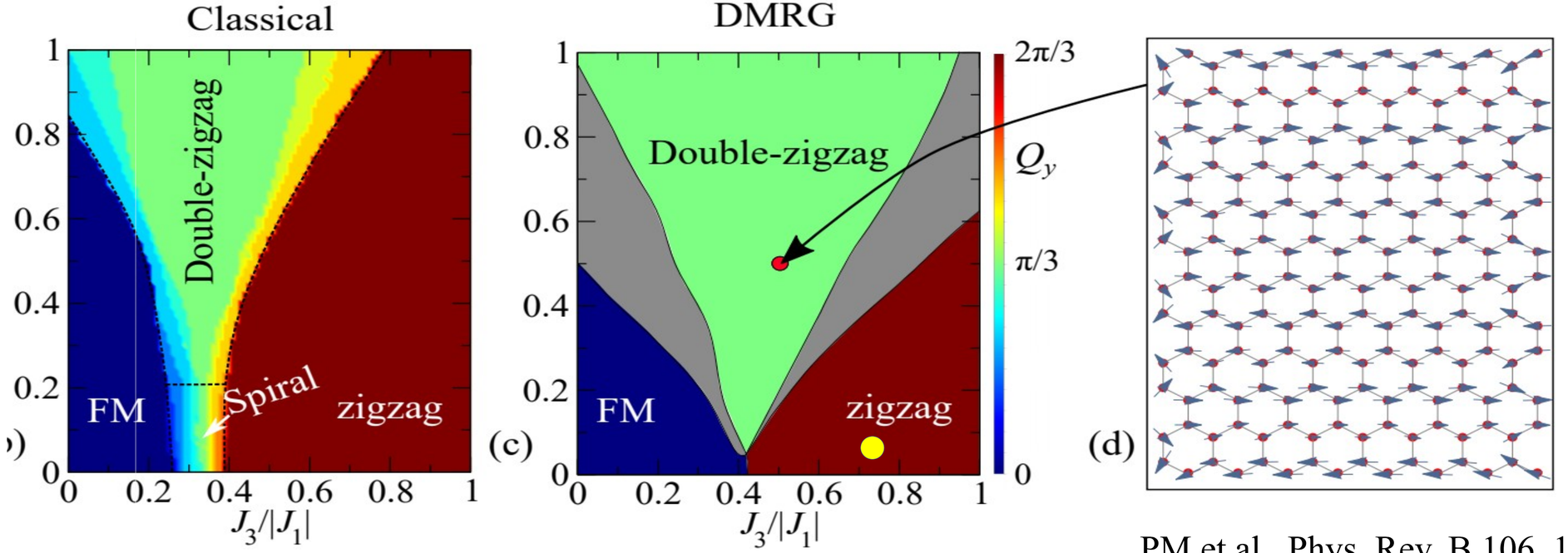
Double zigzag in $BaCo_2(AsO_4)_2$



$$\mathrm{H}_{\min} = \sum_{\langle ij\rangle_1} J_1 \left(S_i^x S_j^x + S_i^y S_j^y\right) + J_3 \sum_{\langle ij\rangle_3} \left(S_i^x S_j^x + S_i^y S_j^y\right)$$
$$- J_{z\pm}^{(3)} \left(\left(S_i^x S_j^z + S_i^z S_j^x\right) c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y\right) s_\alpha\right),$$

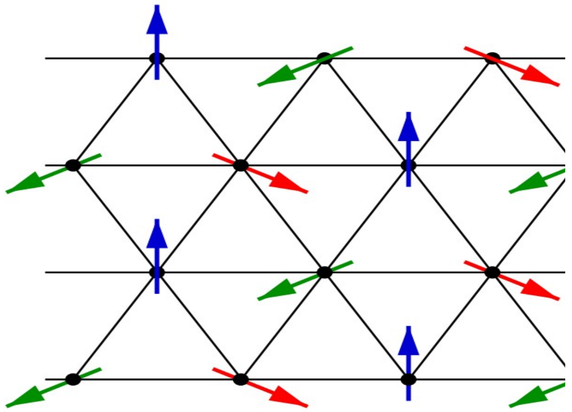
$$\mathrm{J}_{z\pm} = \frac{\sqrt{2}}{3}\left(K - \Gamma + \Gamma'\right)$$

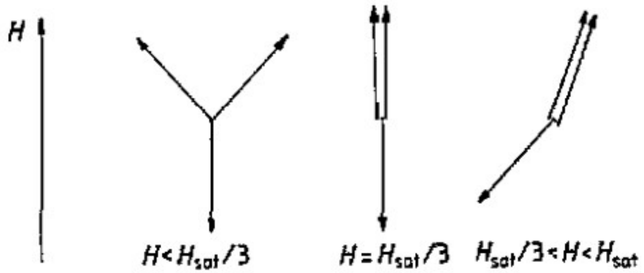
S=1/2

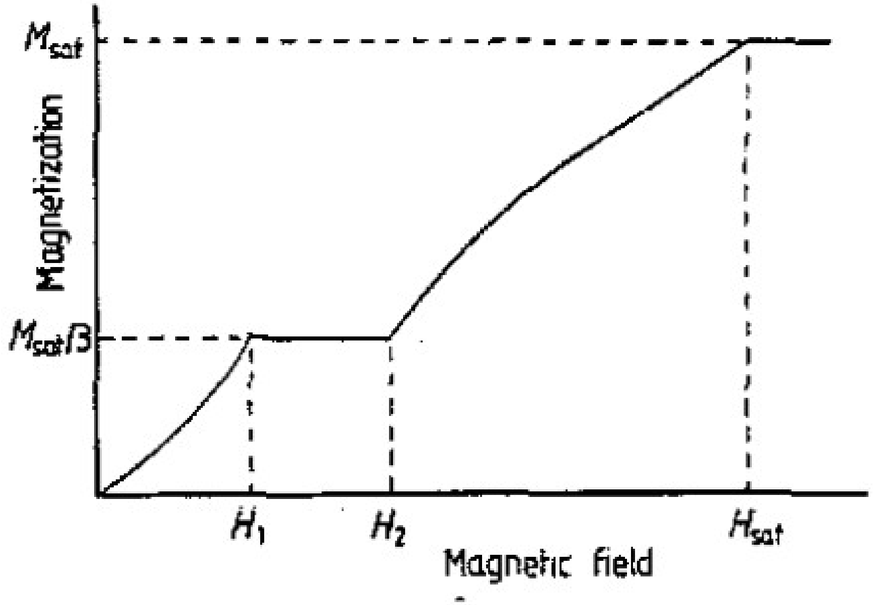


PM et al., Phys. Rev. B 106, 165131 (2022)

Quantum fluctuations

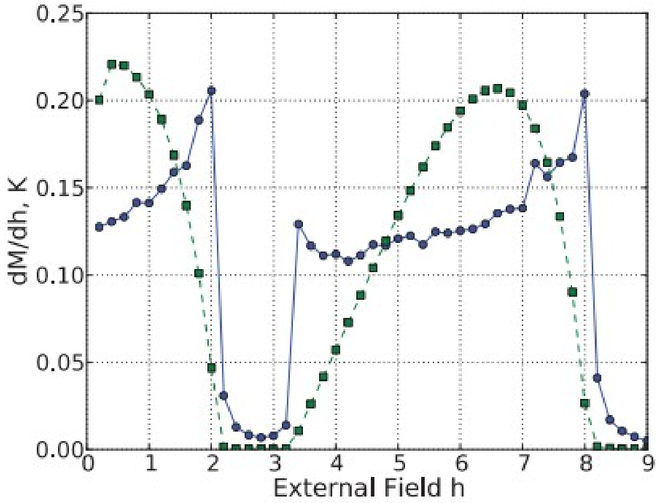






Biquadratic coupling

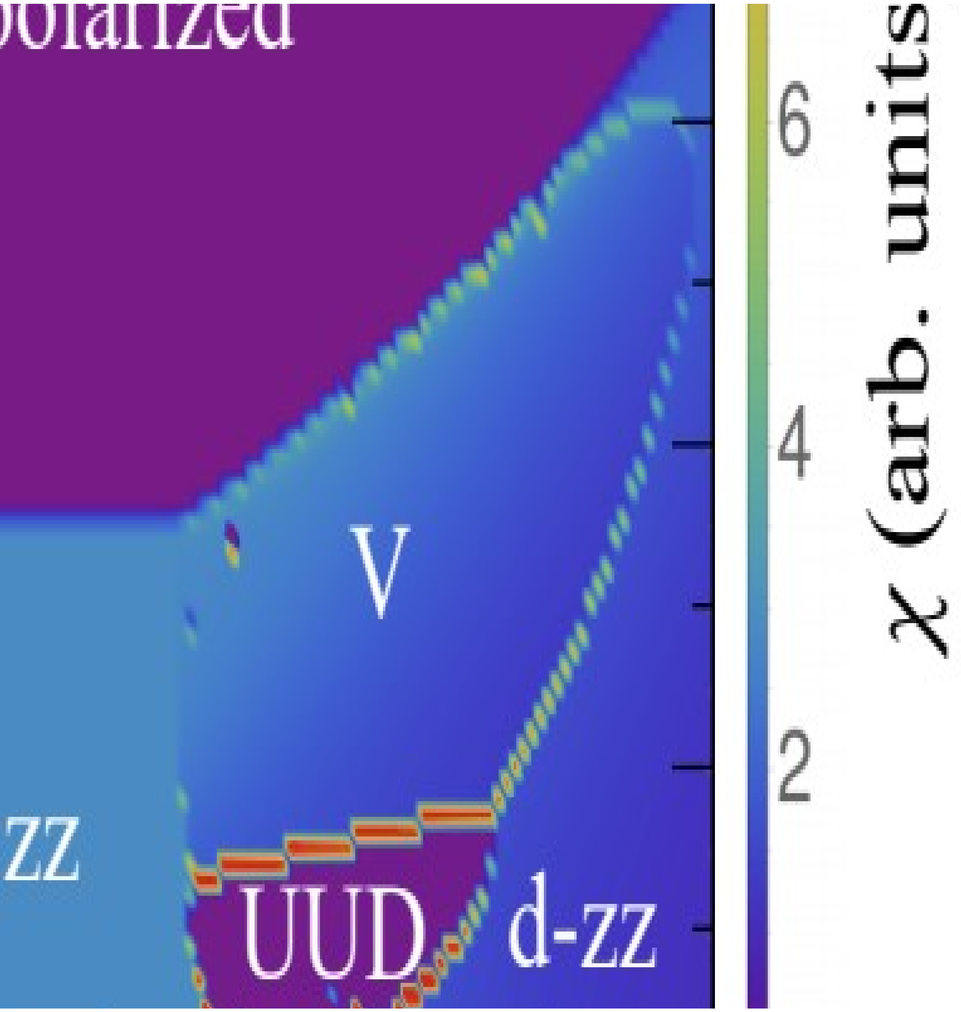
$$B \sum_{\langle ij \rangle} (\mathbf{S}_i \mathbf{S}_j)^2$$



AV Chubukov and D I Golosov, J. Phys.: Condens. Matter 3, 69 (1991)

Christian Griset, Shane Head, Jason Alicea, and Oleg A. Starykh
Phys. Rev. B 84, 245108 (2011)

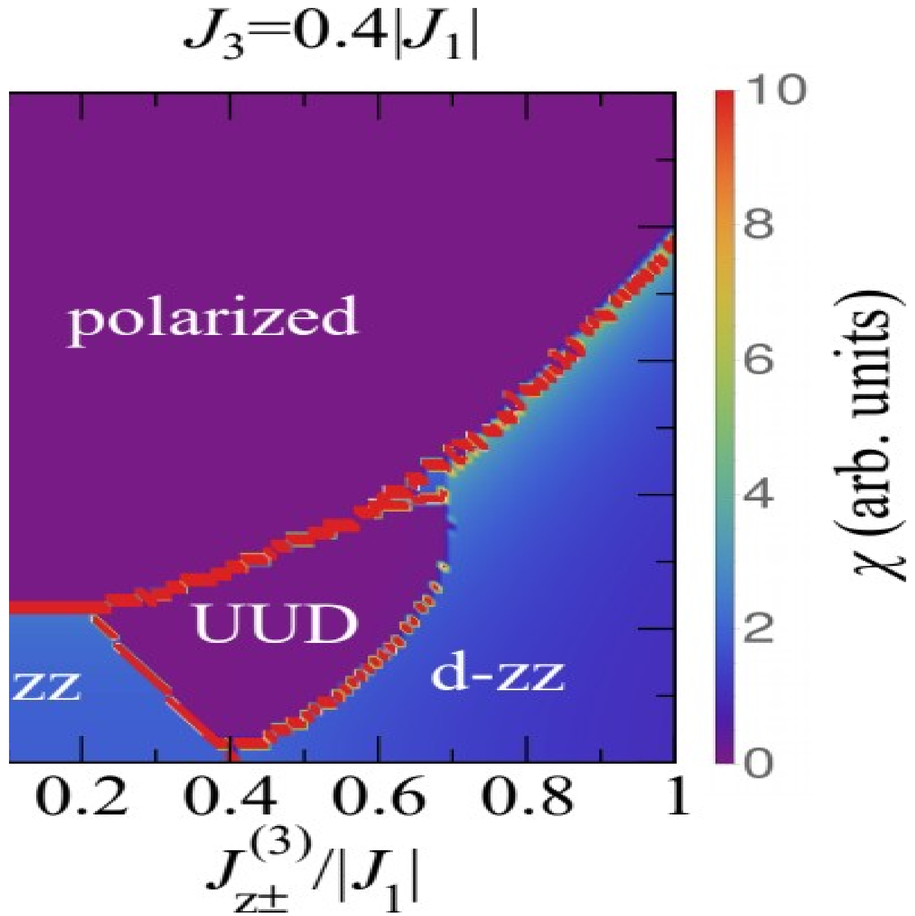
Quantum fluctuations



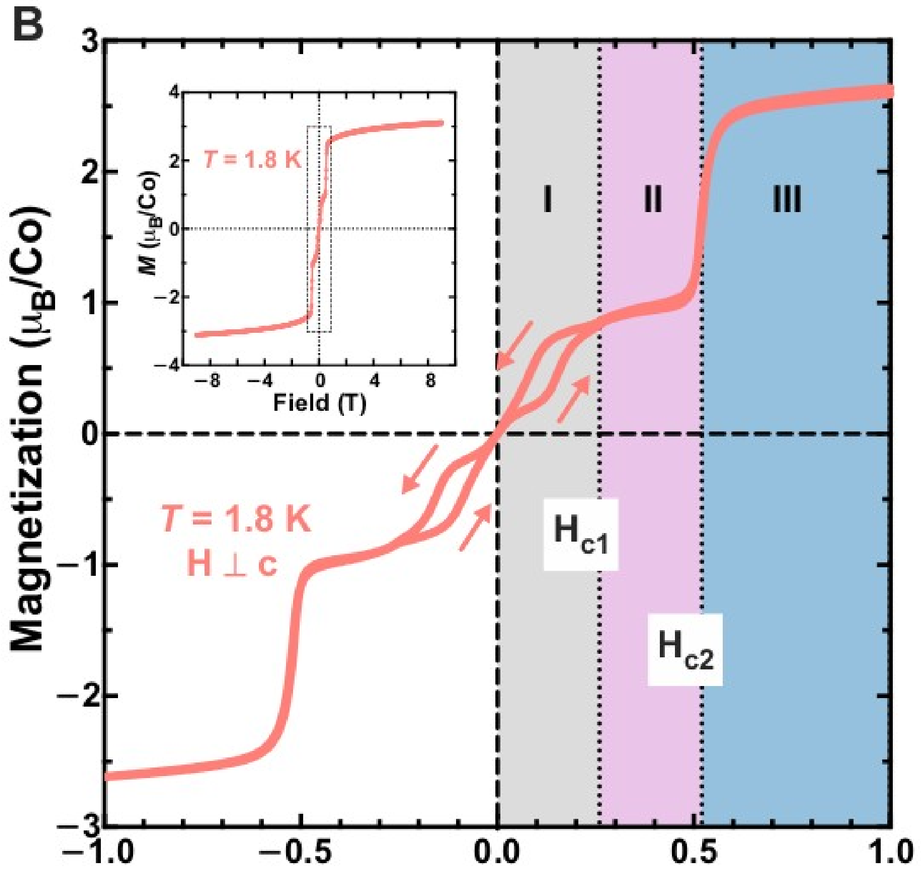
Biquadratic coupling

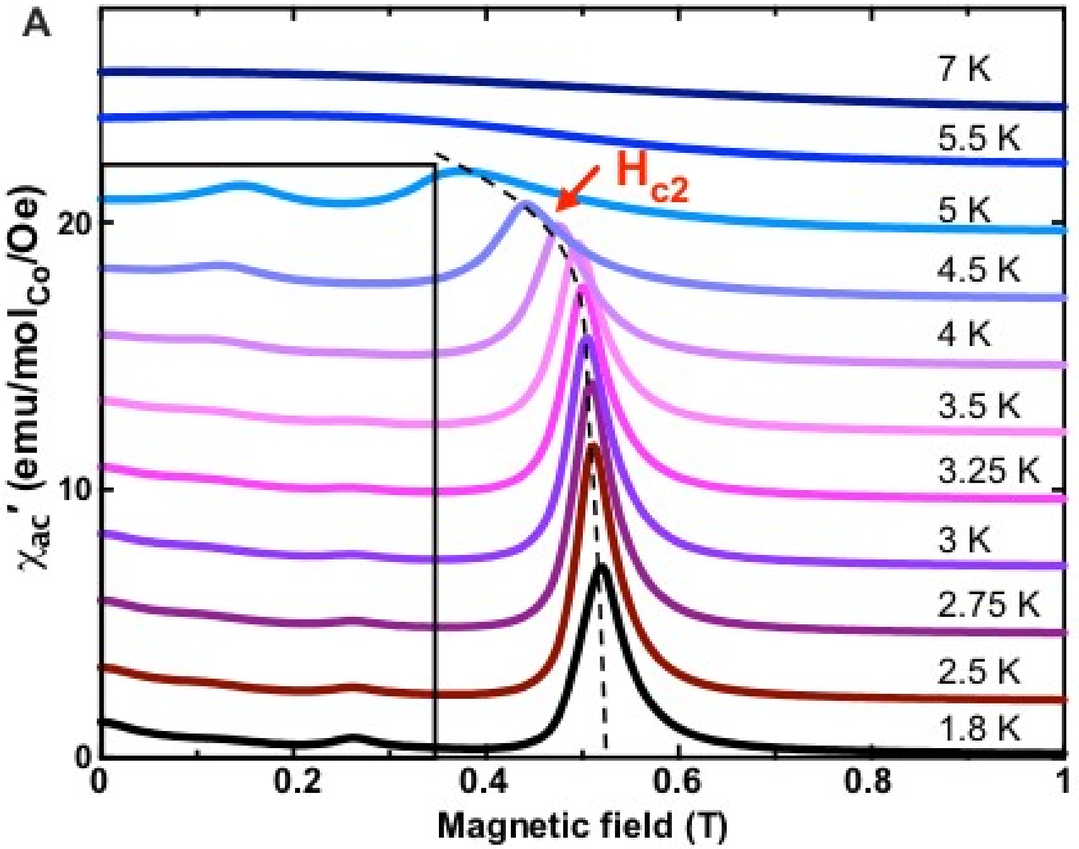
$$-B\sum_{\langle ij\rangle}(\mathbf{S}_i\mathbf{S}_j)^2$$

$$B = 0.04|J_1|$$



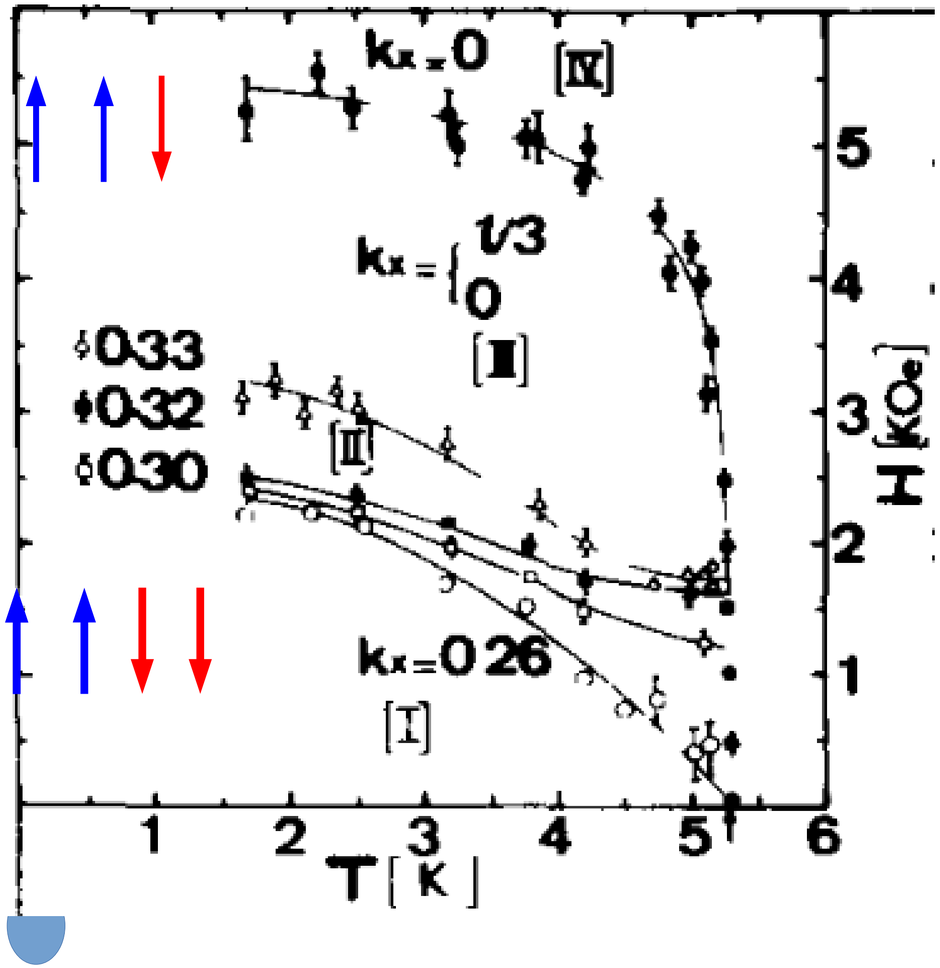
Field-induced magnetic transitions in $BaCo_2(AsO_4)_2$





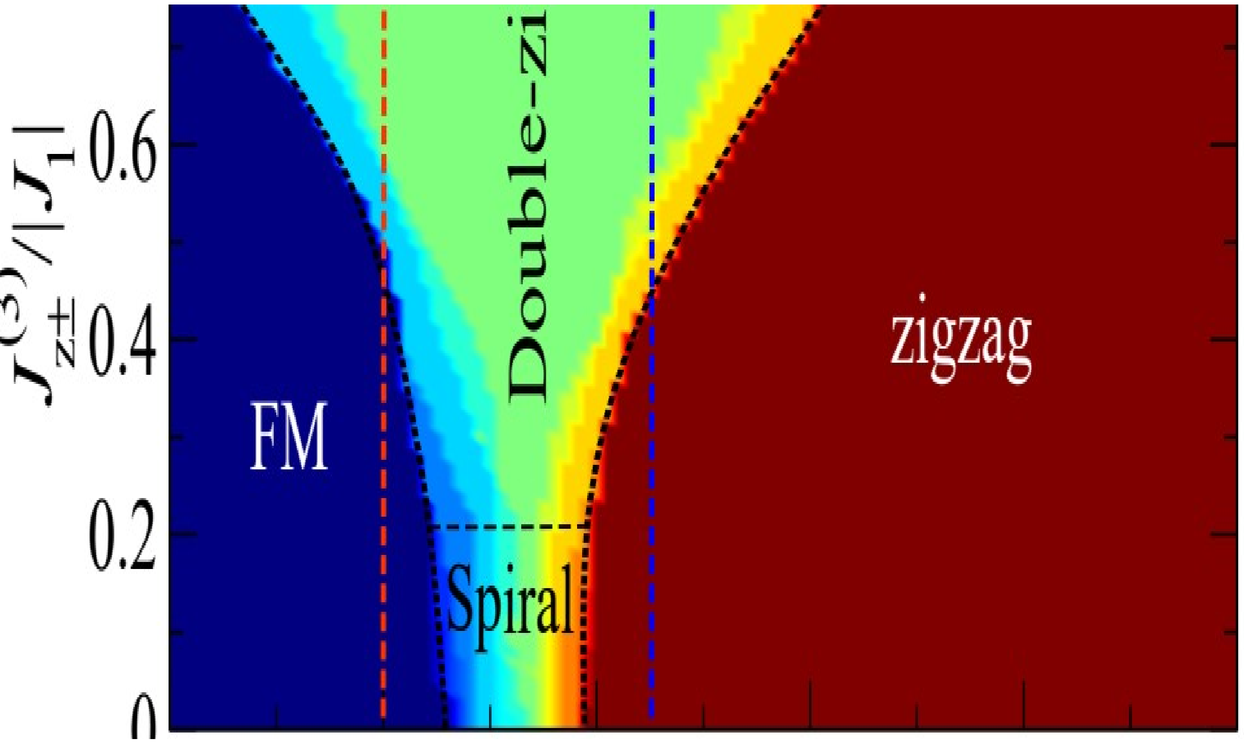
R. Zhong, T. Gao, N. P. Ong, and R. J. Cava, Sci. Adv. 6, eaay6953 (2020)

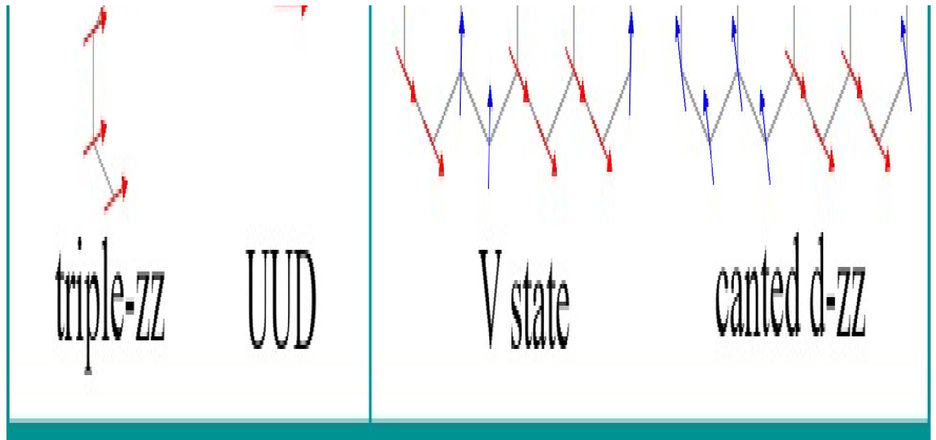
Field induced magnetic transitions in $BaCo_2(AsO_4)_2$

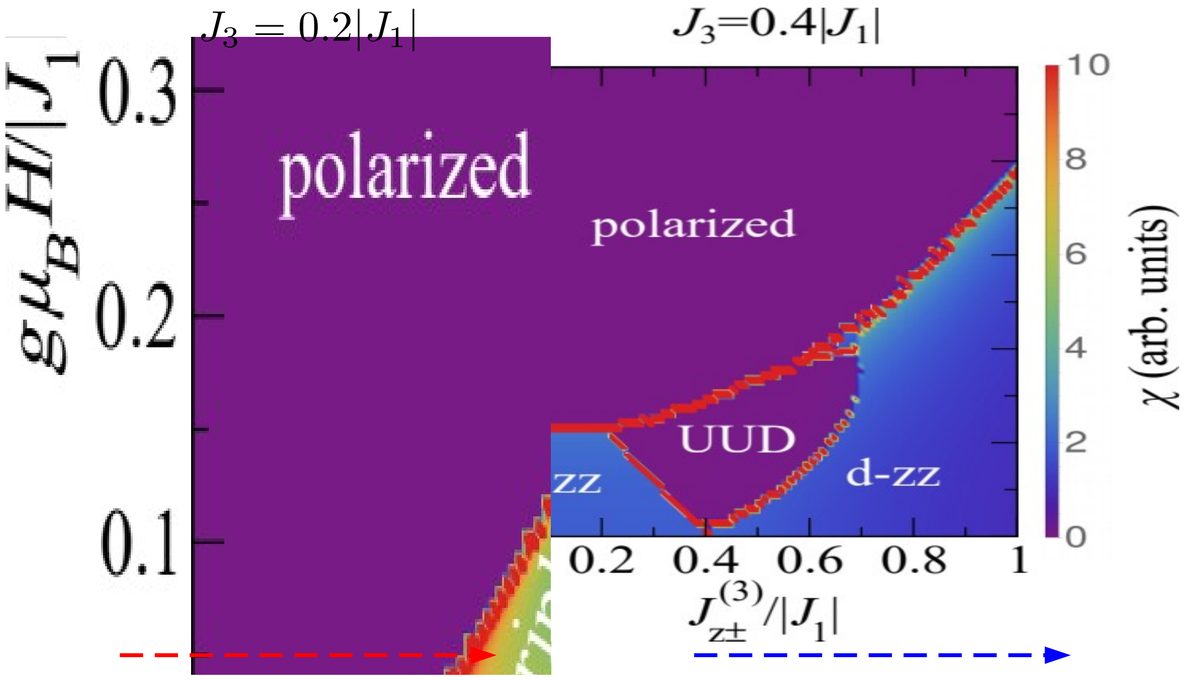


L.P. REGNAULT and J. ROSSAT-MIGNOD, Journal of Magnetism and Magnetic Materials 14,194 (1979)

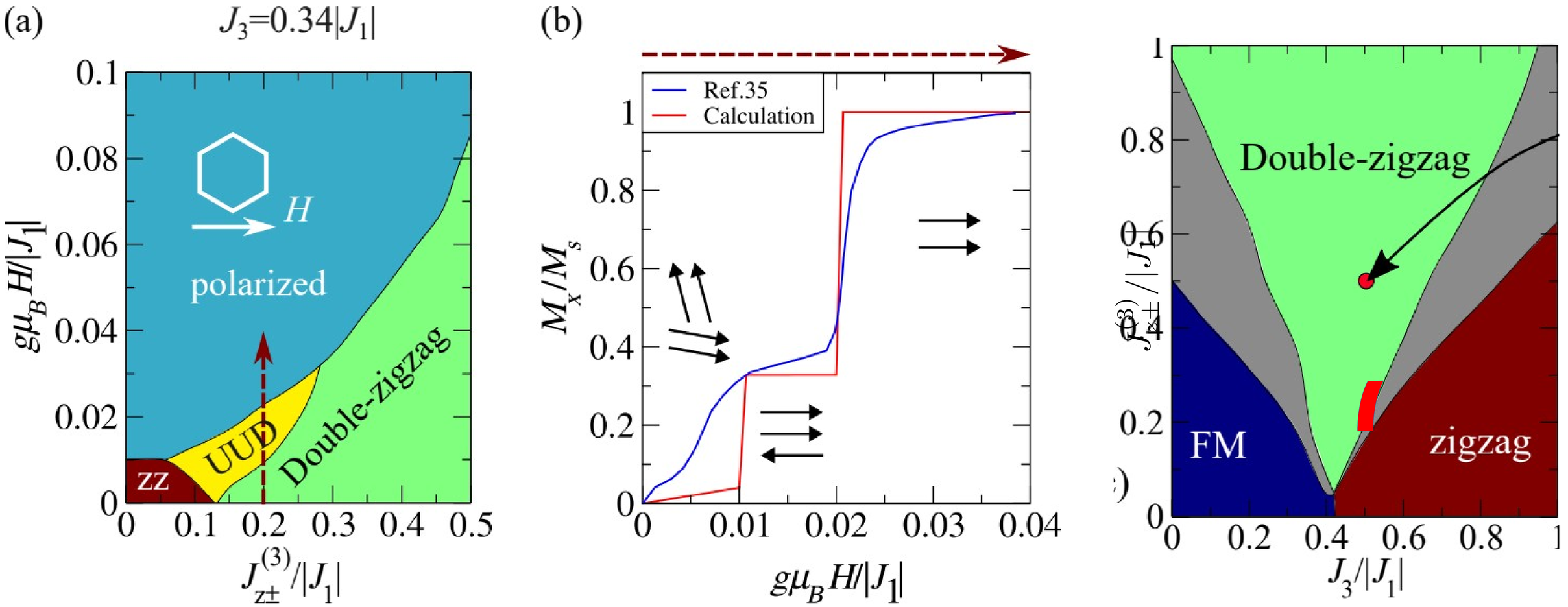
Field-induced transitions of the minimal model





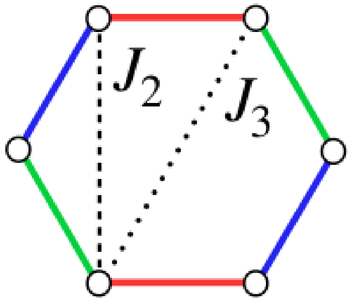


Field transitions in $BaCo_2(AsO_4)_2$



R. Zhong, T. Gao, N. P. Ong, and R. J. Cava, Sci. Adv. 6, eaay6953 (2020)

Inelastic neutron scattering in $BaCo_2(AsO_4)_2$: fitting



$$\mathrm{H}=\sum_{\langle ij\rangle_n} J_n\left(S_i^xS_j^x+S_i^yS_j^y+\Delta_nS_i^zS_j^z\right)$$
$$-2J_{\pm\pm}\left(\left(S_i^xS_j^x-S_i^yS_j^y\right)c_\alpha-\left(S_i^xS_j^y+S_i^yS_j^x\right)s_\alpha\right)$$
$$-J_{z\pm}\left(\left(S_i^xS_j^z+S_i^zS_j^x\right)c_\alpha+\left(S_i^yS_j^z+S_i^zS_j^y\right)s_\alpha\right)$$

n=1,2,3

Spin-wave theory

$$S^+\approx a$$
$$S^-\approx a^\dagger$$
$$S^z=S-a^\dagger a$$

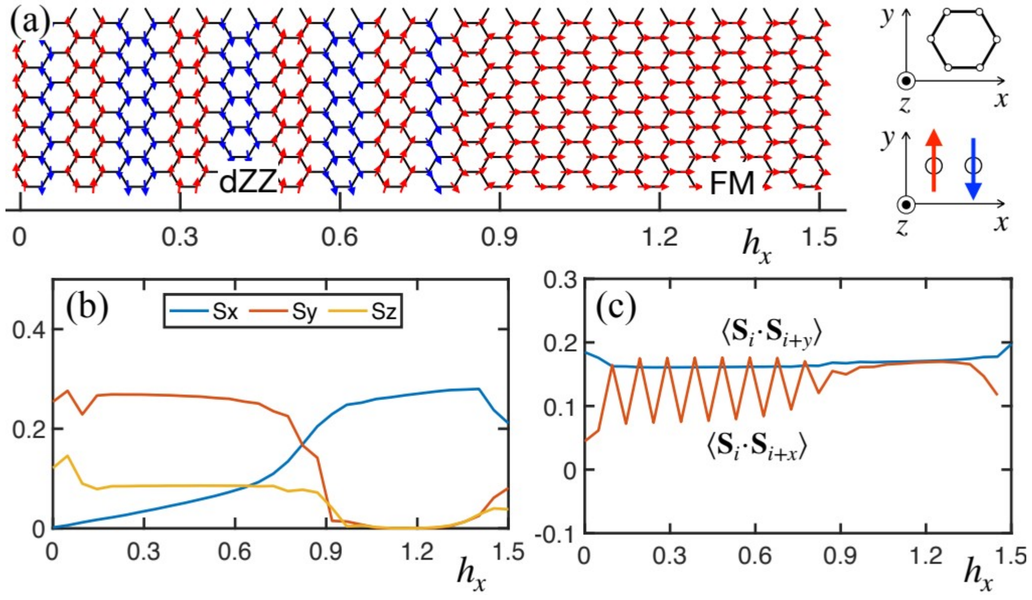
7 equations →

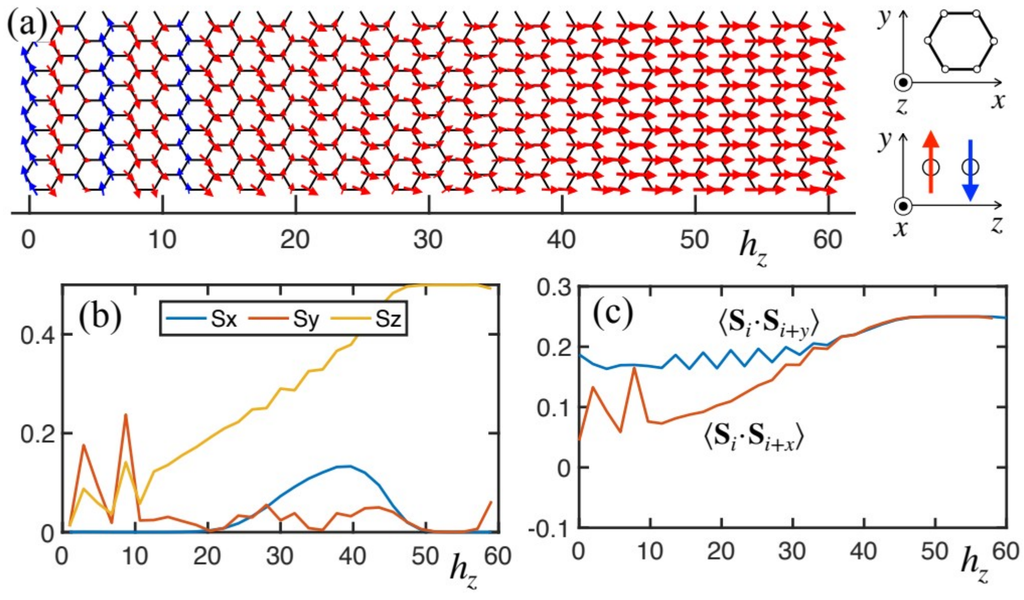
Free parameters

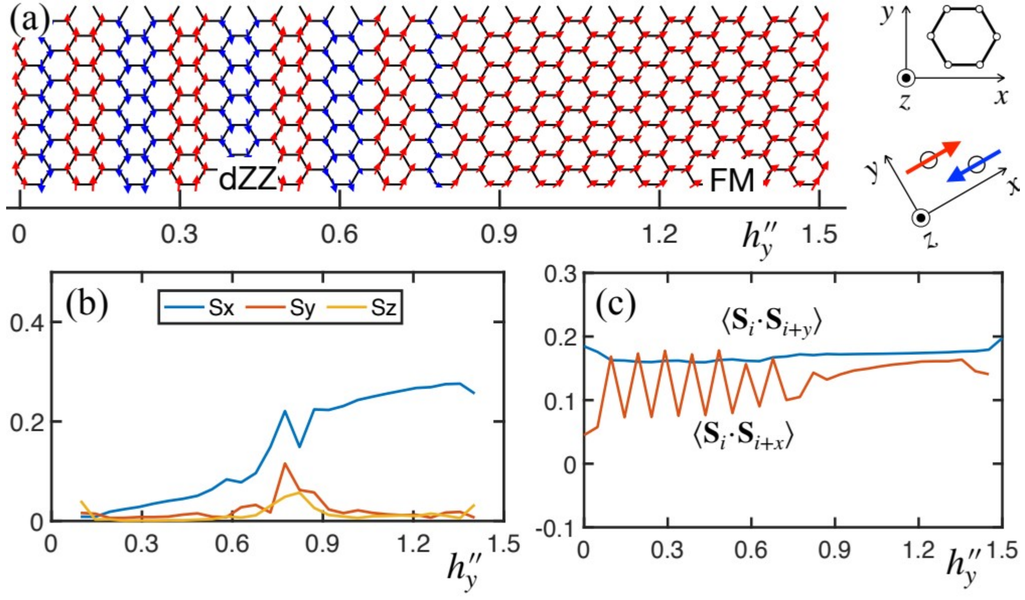
$$\{\mathrm{J}_1,\Delta_1,\mathrm{J}_{\pm\pm},\mathrm{J}_{z\pm},\mathrm{J}_3,\Delta_3,g_b\}$$

Fixed J_2=-0.21 meV, g_{a^*}=4.8

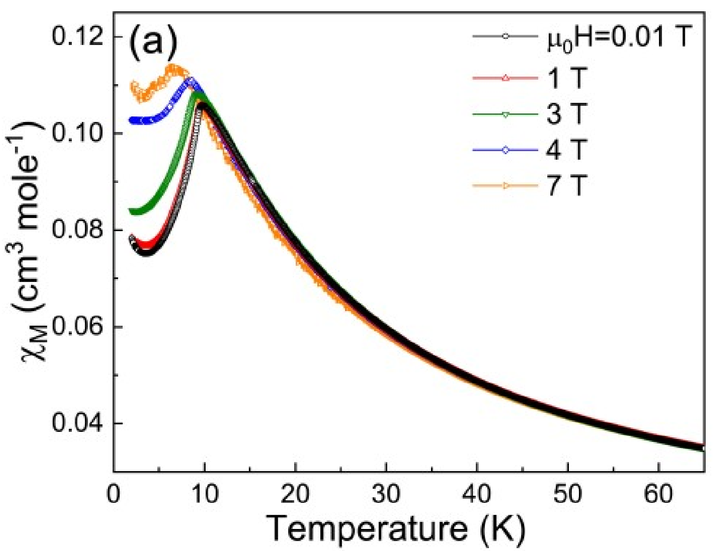
$$\{\mathrm{J}_1,\Delta_1,\mathrm{J}_{\pm\pm},\mathrm{J}_{z\pm}\}=\{-6.54,0.36,0.15,-3.76\}\ (\mathrm{meV})$$
$$\{\mathrm{J}_2,\mathrm{J}_3,\Delta_3\}=\{-0.21,1.70,0.03\}\ (\mathrm{meV})$$

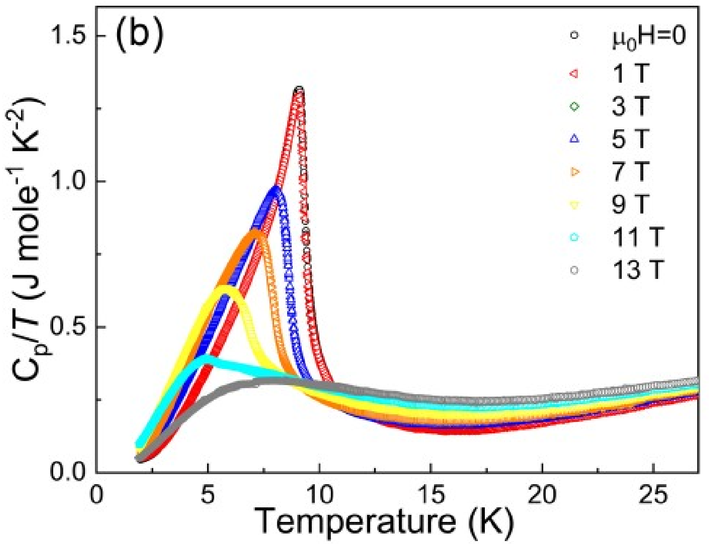

(a)
dZZ
FM
h_x
(b)
Sx
Sy
Sz
h_x
(c)
$\langle \mathbf{S}_i \cdot \mathbf{S}_{i+y} \rangle$
$\langle \mathbf{S}_i \cdot \mathbf{S}_{i+x} \rangle$
h_x

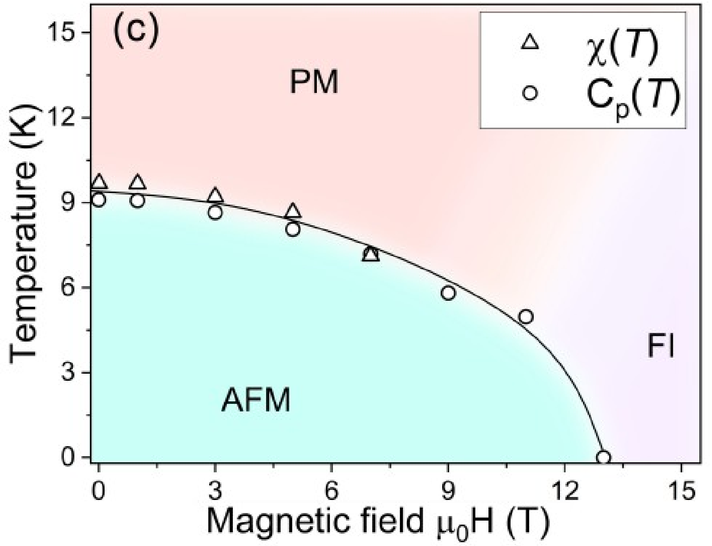

(a)
h_z
(b)
Sx
Sy
Sz
h_z
(c)
$\langle \mathbf{S}_i \cdot \mathbf{S}_{i+y} \rangle$
$\langle \mathbf{S}_i \cdot \mathbf{S}_{i+x} \rangle$
h_z


(a)
dZZ
FM
h_y''
(b)
Sx
Sy
Sz
h_y''
(c)
$\langle \mathbf{S}_i \cdot \mathbf{S}_{i+y} \rangle$
$\langle \mathbf{S}_i \cdot \mathbf{S}_{i+x} \rangle$
h_y''

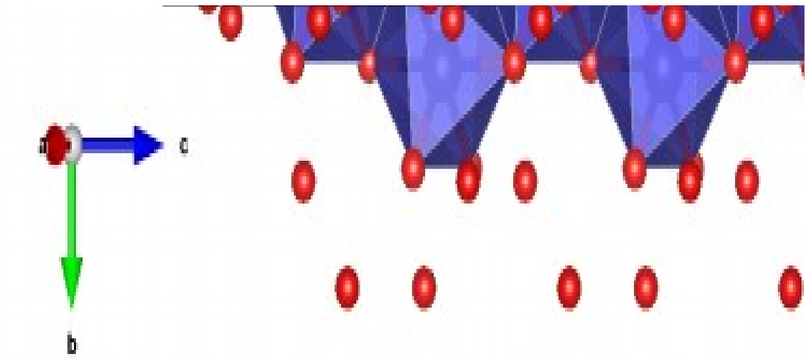
Magnetic order







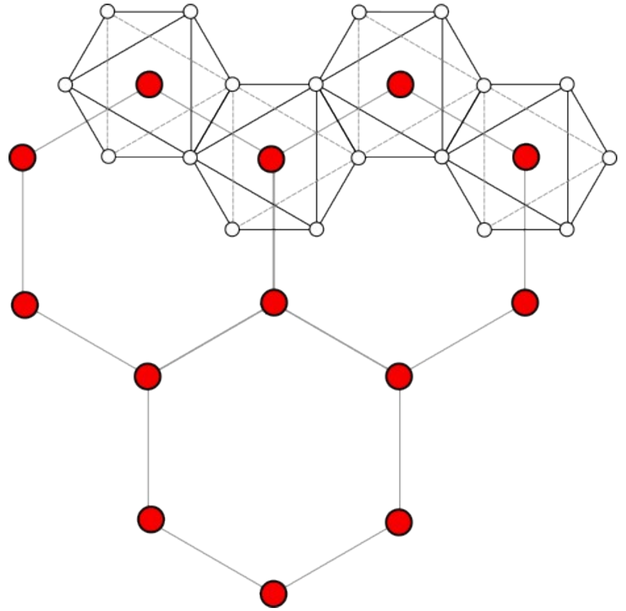
Intra-chain exchanges



Glide symmetry of chains:
c/2 shift+ reflection

$$J^{(m)} = \begin{pmatrix} J^{xx} & (-1)^m J^{xy} & J^{xz} \\ (-1)^m J^{xy} & J^{yy} & (-1)^m J^{yz} \\ J^{xz} & (-1)^m J^{yz} & J^{zz} \end{pmatrix}$$

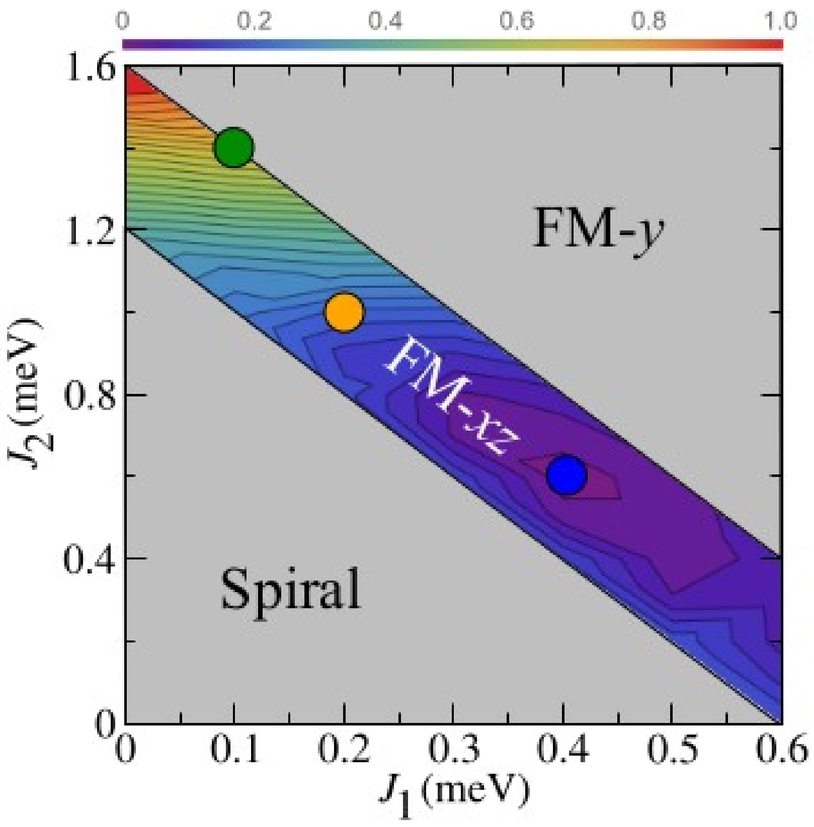
Generic model has 6 parameters

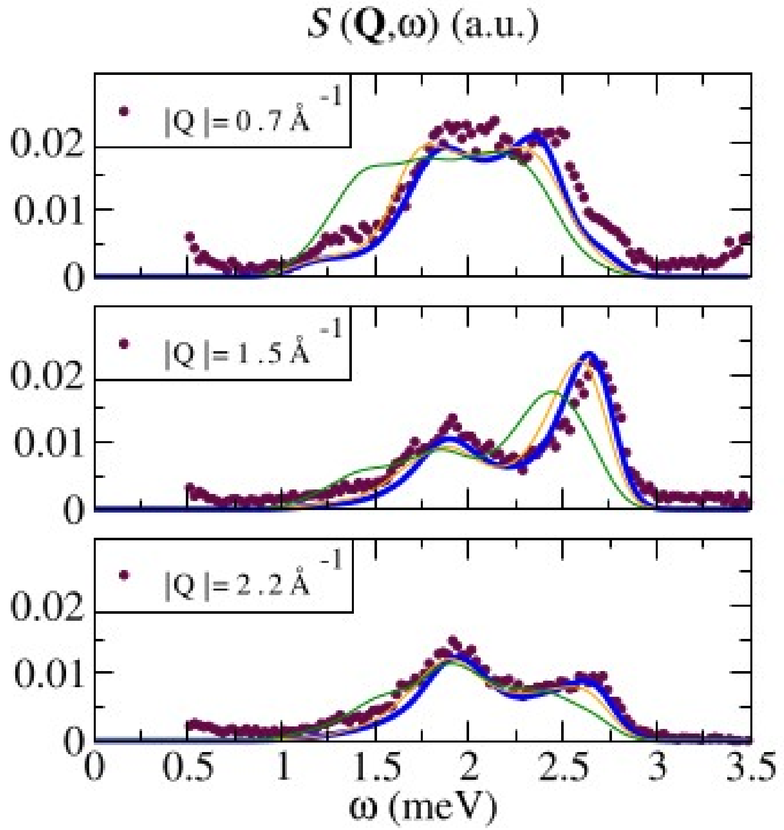


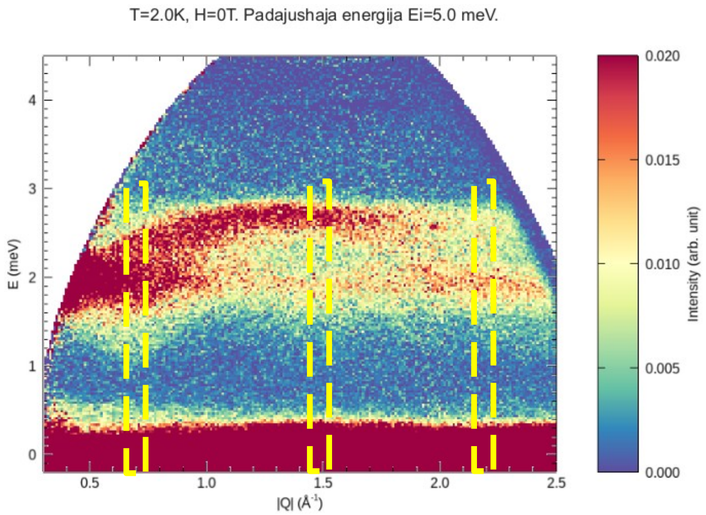
$$\begin{aligned} \mathrm{J}_{xx} &= J + \tfrac{1}{6}\left(K - 4\Gamma - 2\Gamma'\right), \\ J_{yy} &= J + K/2 - \Gamma', \\ J_{zz} &= J + \tfrac{1}{3}\left(K + 2\Gamma + 4\Gamma'\right), \\ J_{xy} &= \tfrac{1}{2\sqrt{3}}\left(K + 2\Gamma - 2\Gamma'\right), \\ J_{xz} &= \tfrac{1}{3\sqrt{2}}\left(-K + \Gamma - \Gamma'\right), \\ J_{yz} &= -\tfrac{1}{\sqrt{6}}\left(K - \Gamma + \Gamma'\right). \end{aligned}$$

Michele Fava , Radu Coldea and S. A. Parameswaran , PNAS 117, 25219 (2019)

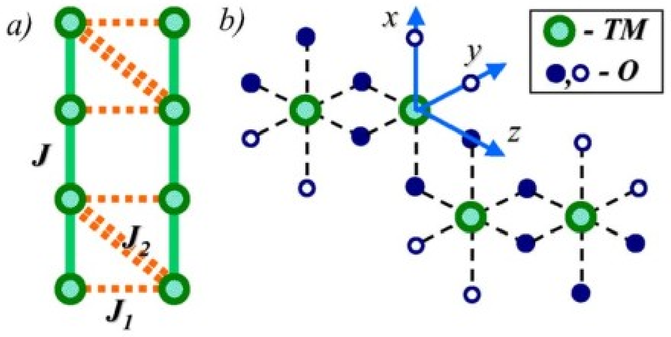
Fit quality



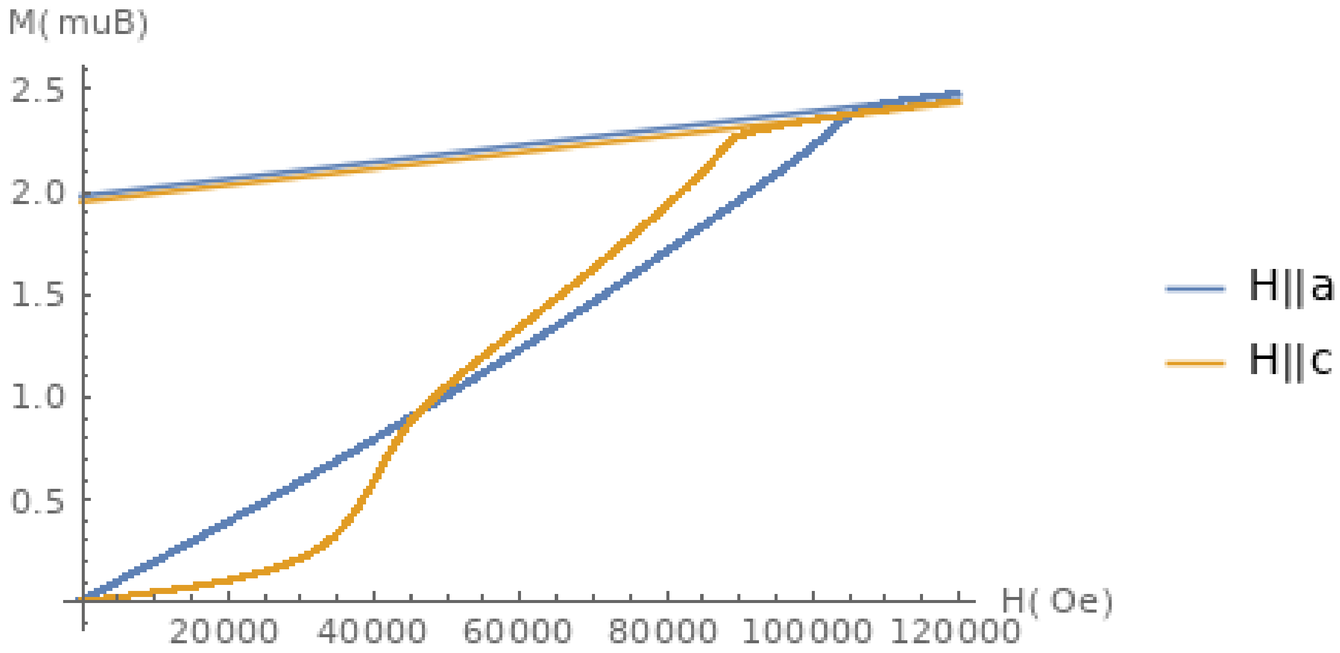




$(J_1, J_2) =$ (0.4,0.6) (0.2,1.0) (0.1,1.4)

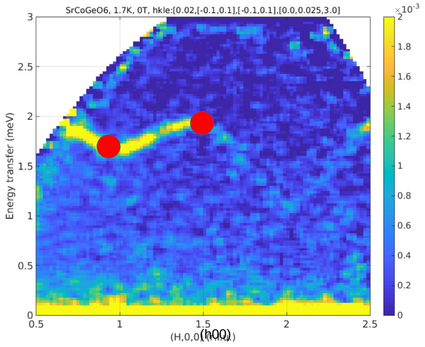


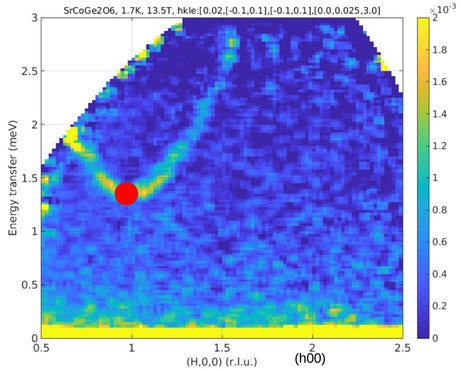
First step: Fitting magnetization

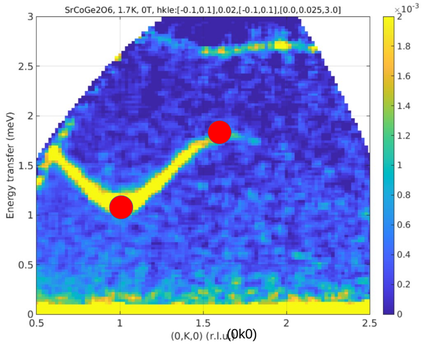


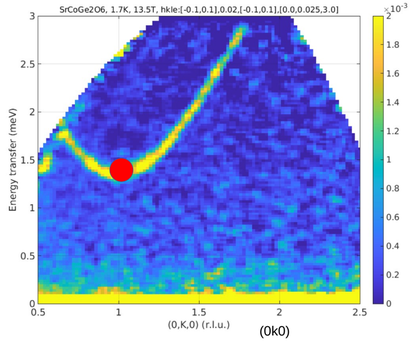
ga=3.96
gc=3.9
Need to correct for Van Vleck contribution

Fitting procedure





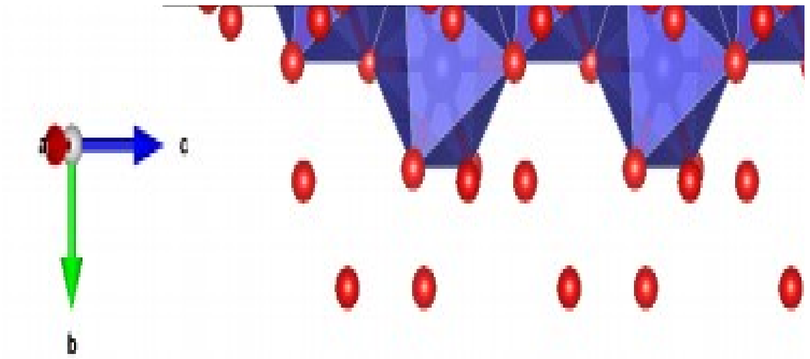




- Identify several different high-symmetry points
- Optimize spin-wave energies
  (depend only on several parameters)
- Obtain (Jxx,Jyy,Jzz,J1,J2)
- Jxz depends on the 0T canting angle

$$\tan 2\theta = \frac{2J^{xz}}{J^{xx} - J^{zz}}$$

Intra-chain exchanges



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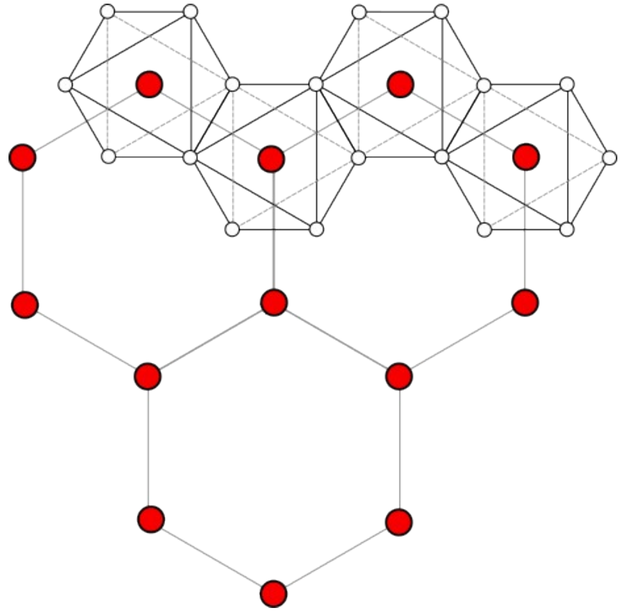

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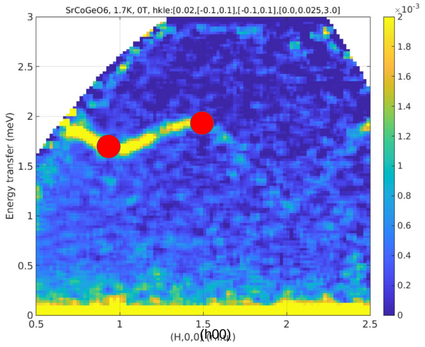
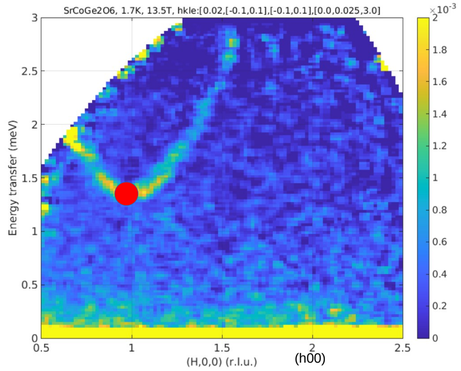
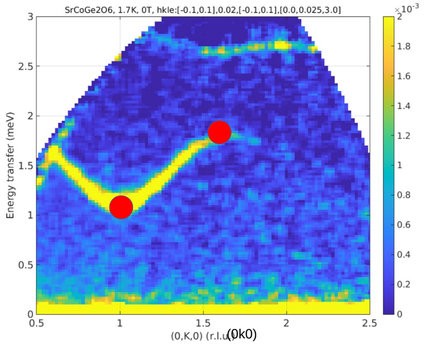
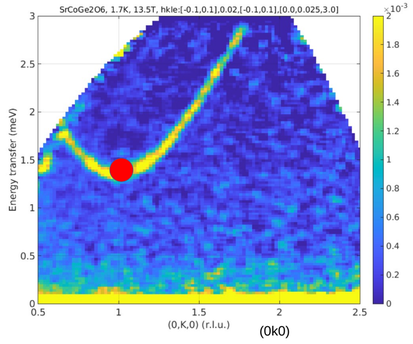
Fitting procedure









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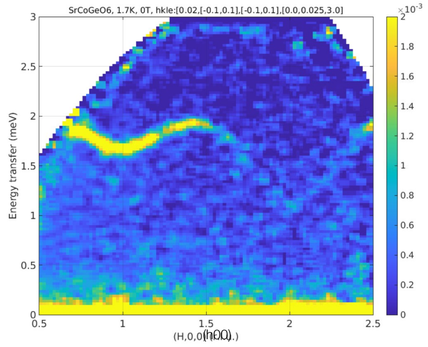
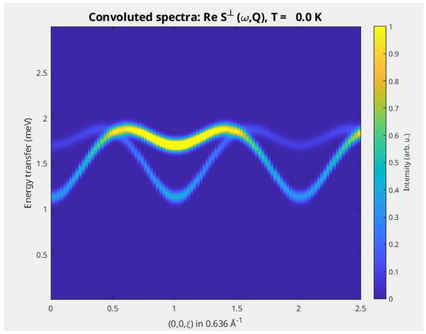
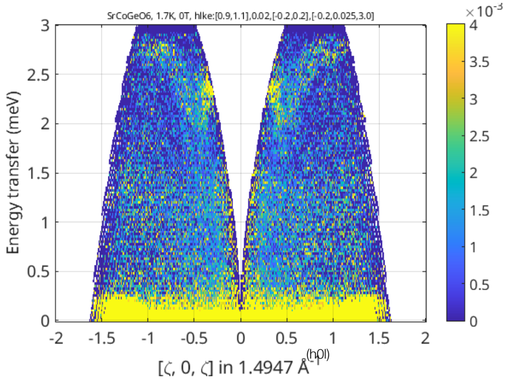
H–O

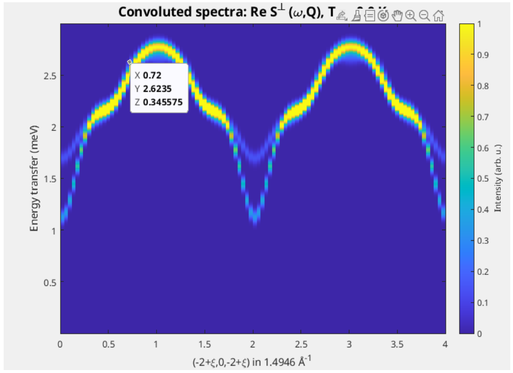
$$\hat{J}_{\text{chain}} = \begin{pmatrix} -1.12 & 0 & 0.13 \\ 0 & -0.1 & 0 \\ 0.13 & 0 & -0.85 \end{pmatrix}$$

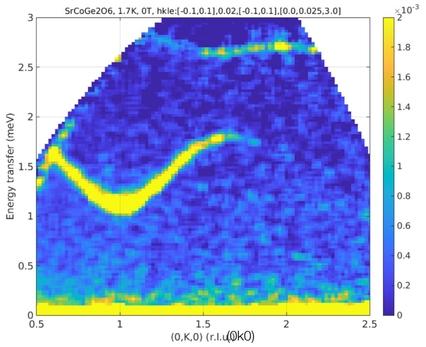
J1=0.2 meV, J2=0.8 meV

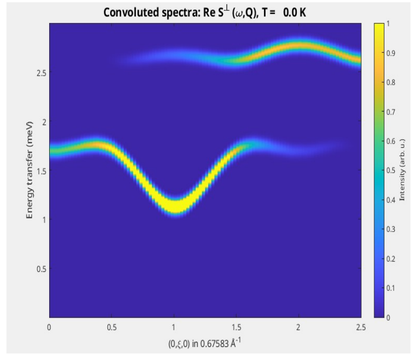


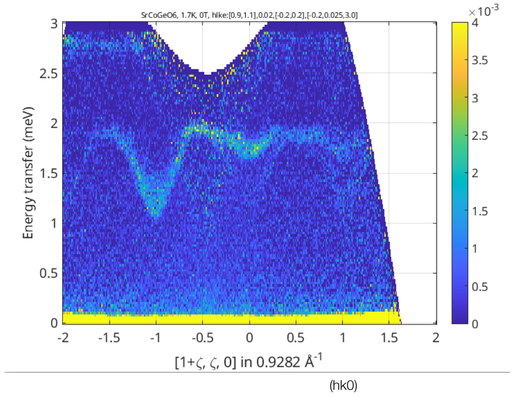


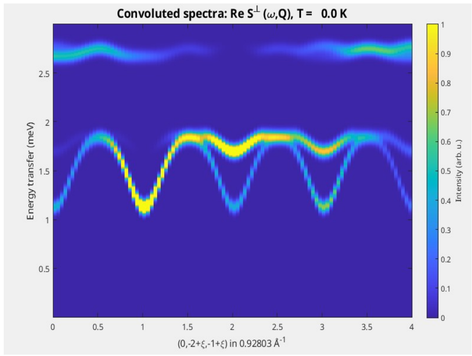


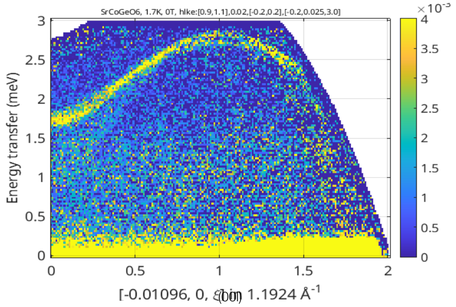


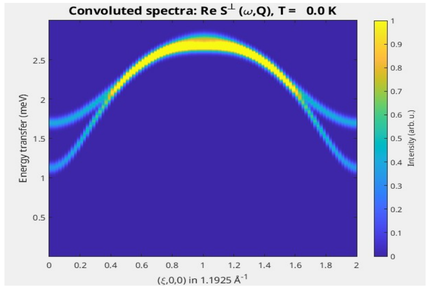


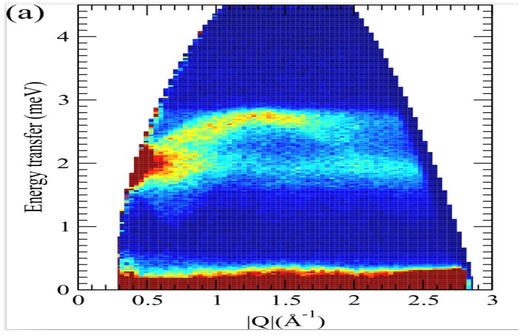


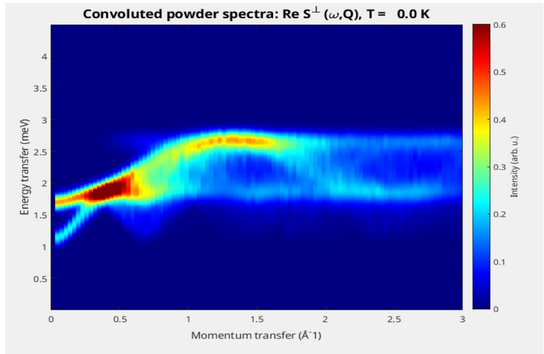




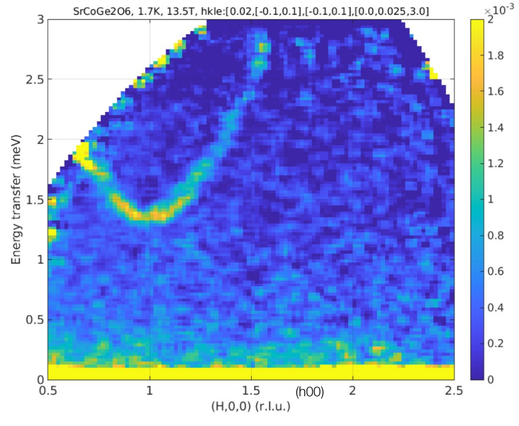


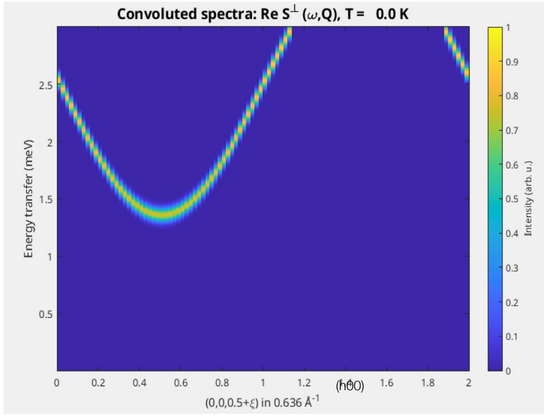


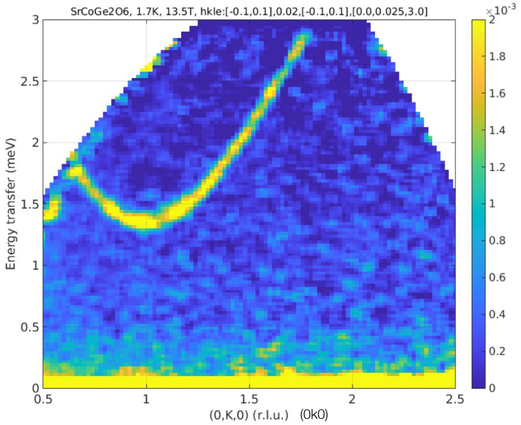


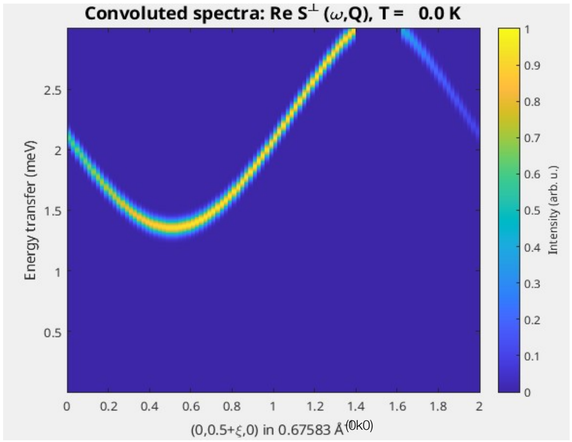


H=13.5T, H||c









H=8T, H||b