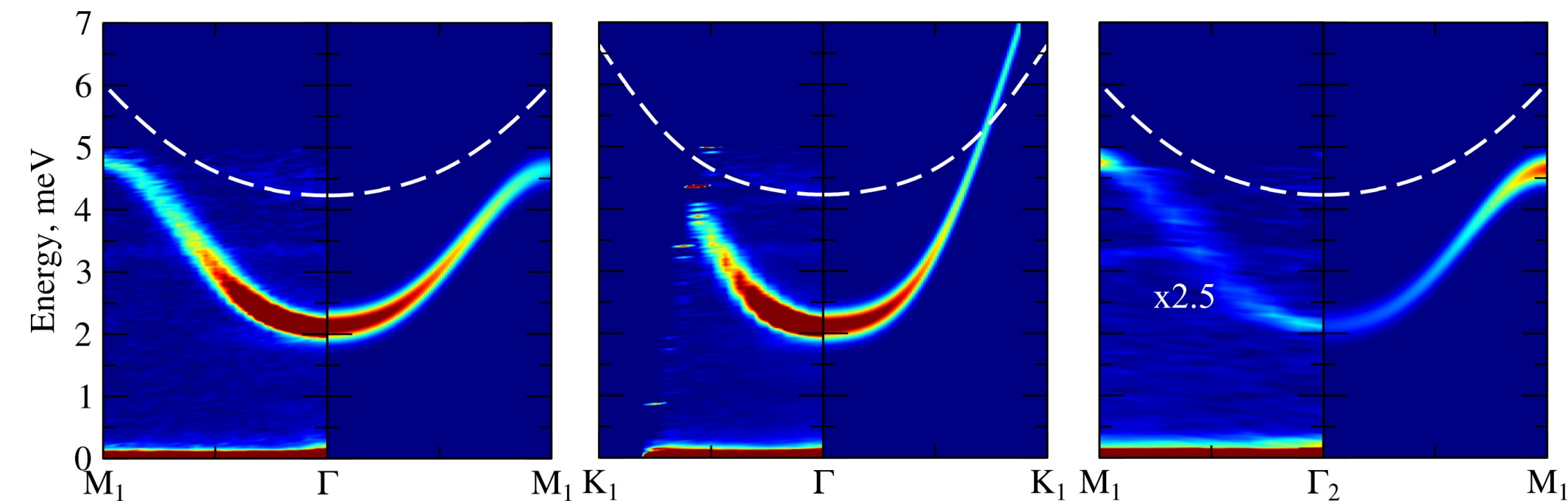


Magnetic spectrum of cobalt-based frustrated magnets



Pavel Maksimov

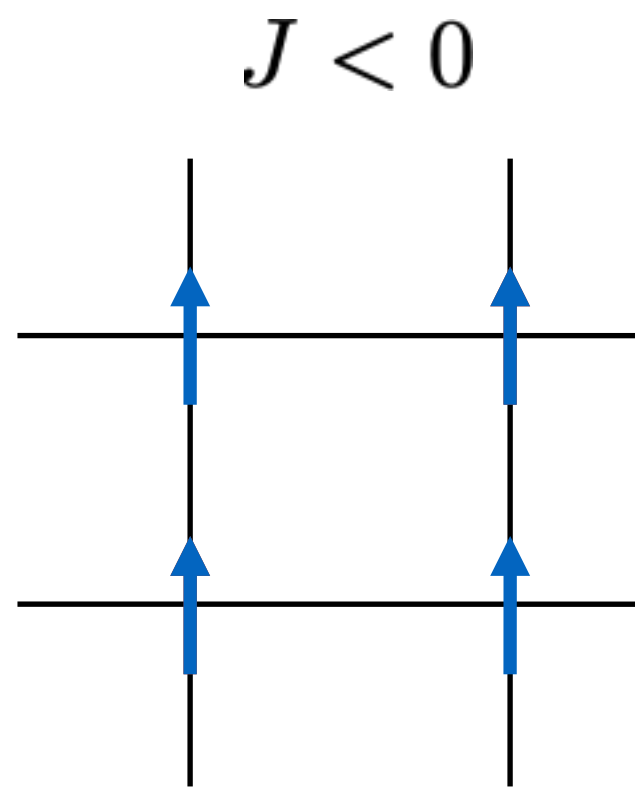
Joint Institute for Nuclear Research, Dubna



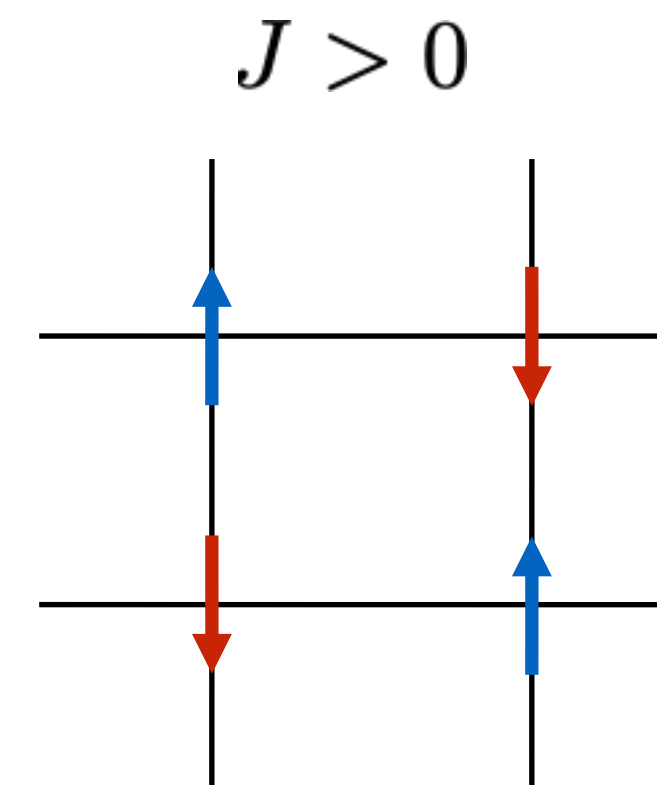
Heisenberg model

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

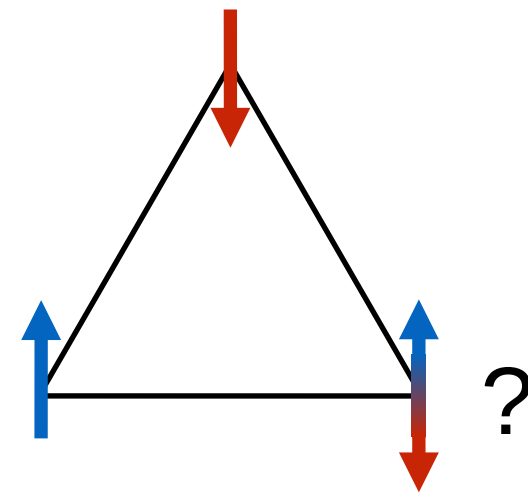
Ferromagnet



Antiferromagnet



Frustrated magnets



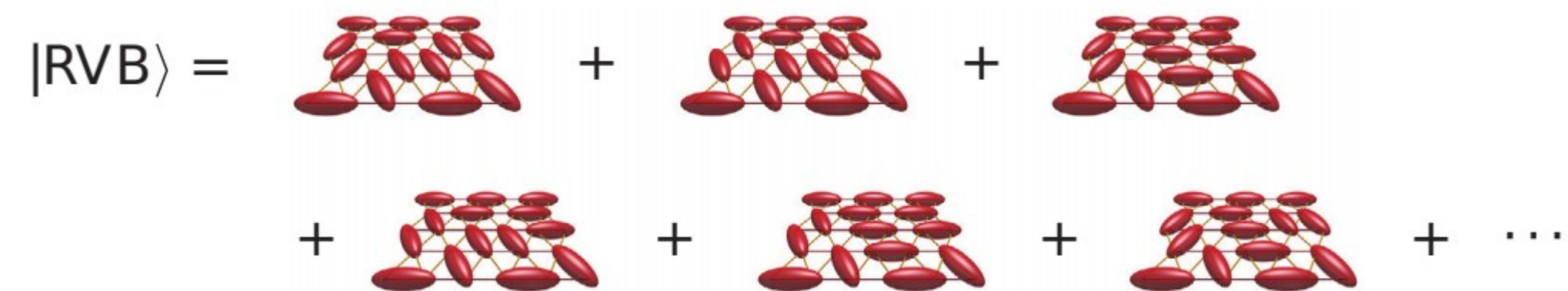
G. H. Wannier, *Physical Review* 79, 357 (1950)

Classical degeneracy $\rightarrow$ Finite entropy at $T=0$

Resonating valence bond (RVB)

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

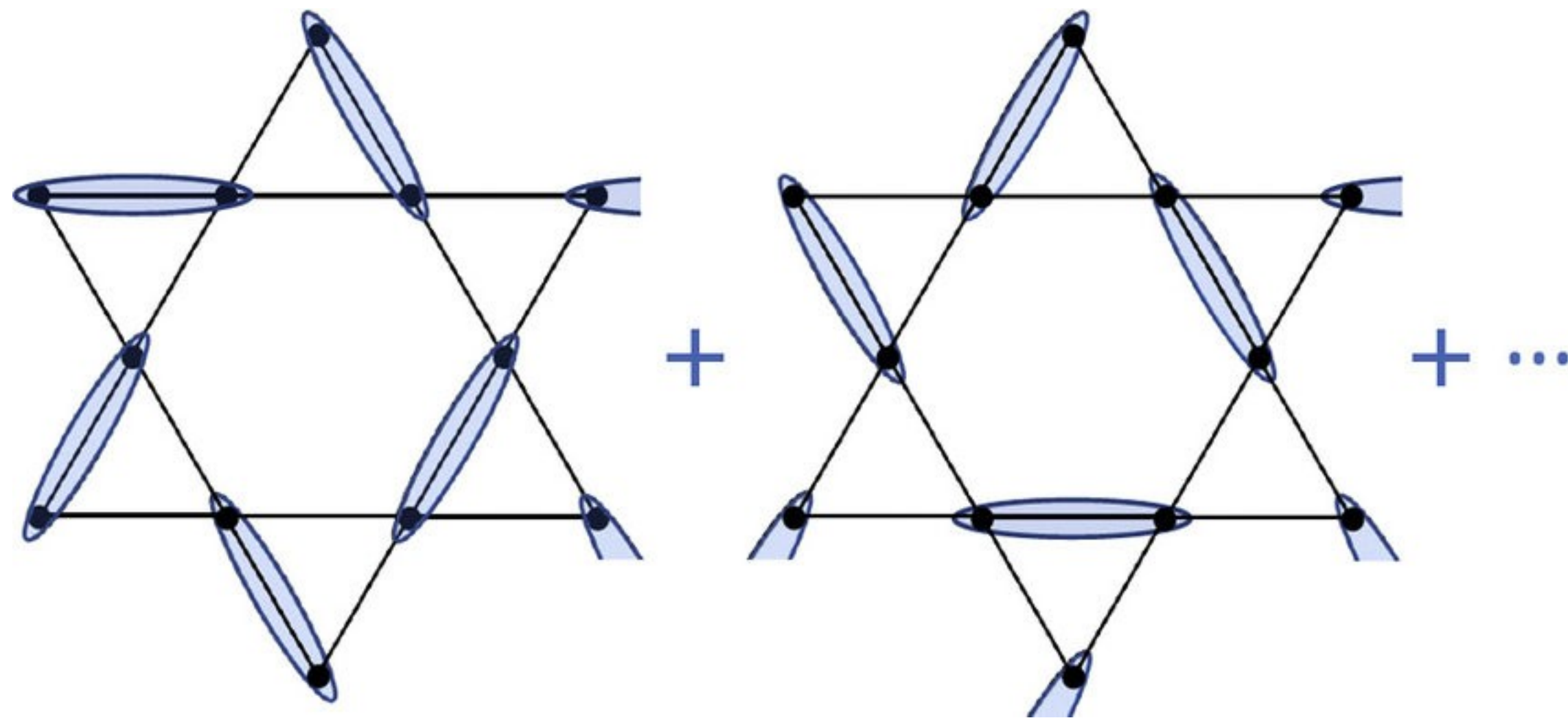
P. Fazekas and P. W. Anderson, *Philosophical Magazine* 30, 423 (1974)



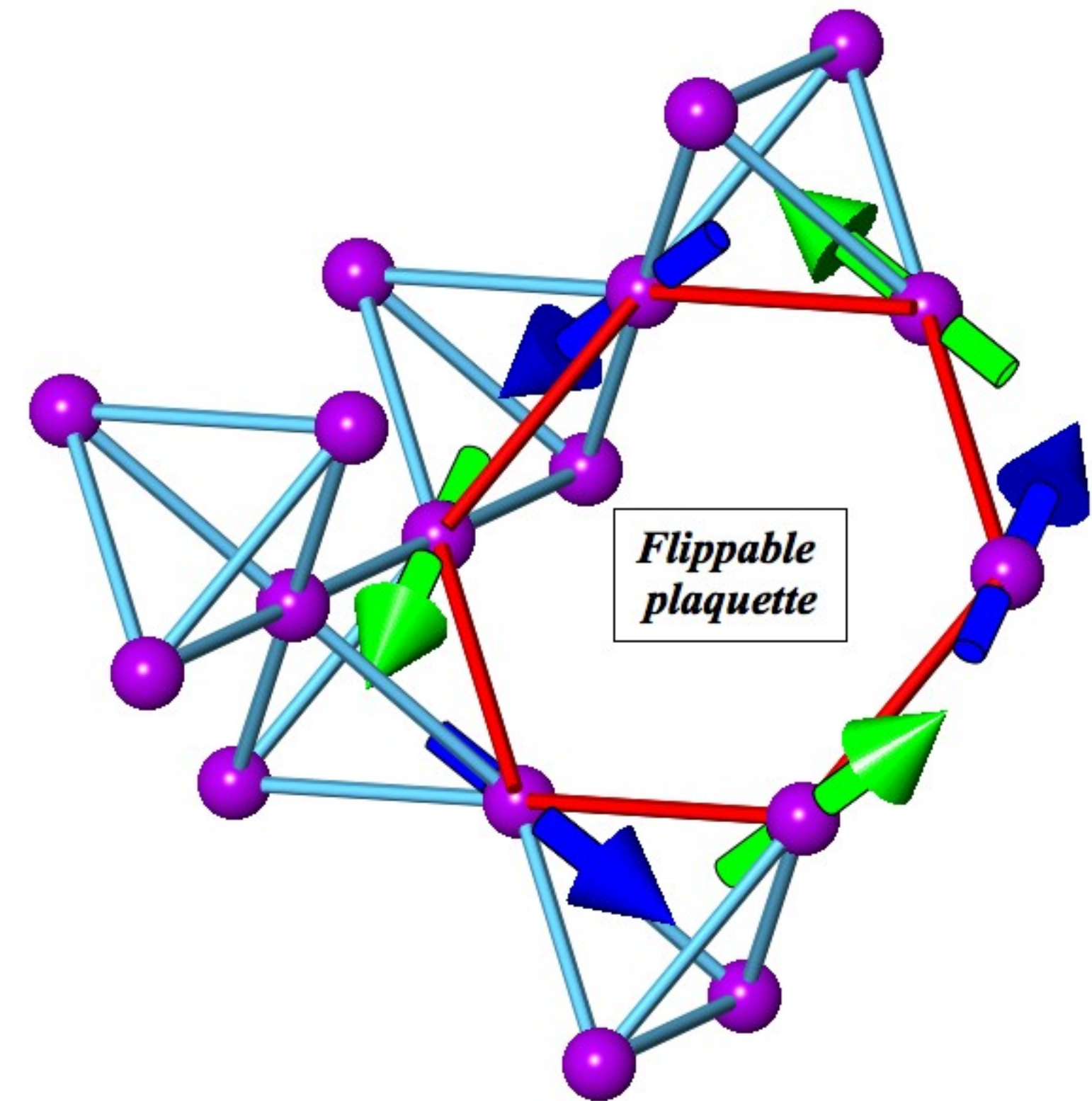
Spin liquids: fractionalized excitations, topological properties

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

Frustration from geometry: kagome and pyrochlore lattices



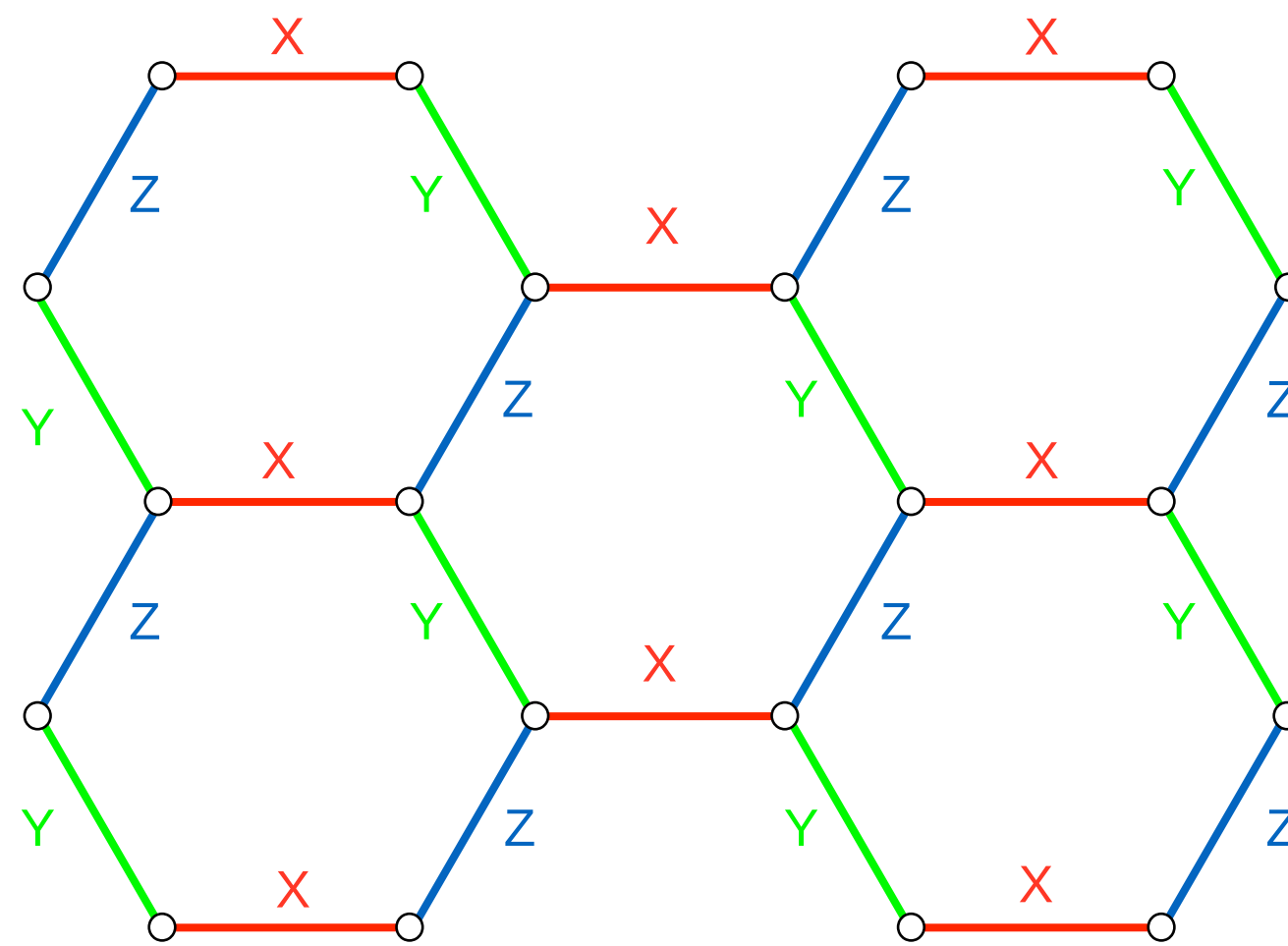
P. Mendels and F.Bert, Comptes Rendus Physique 17, 455 (2016)



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

Frustration from anisotropic interactions: Kitaev model

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



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

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

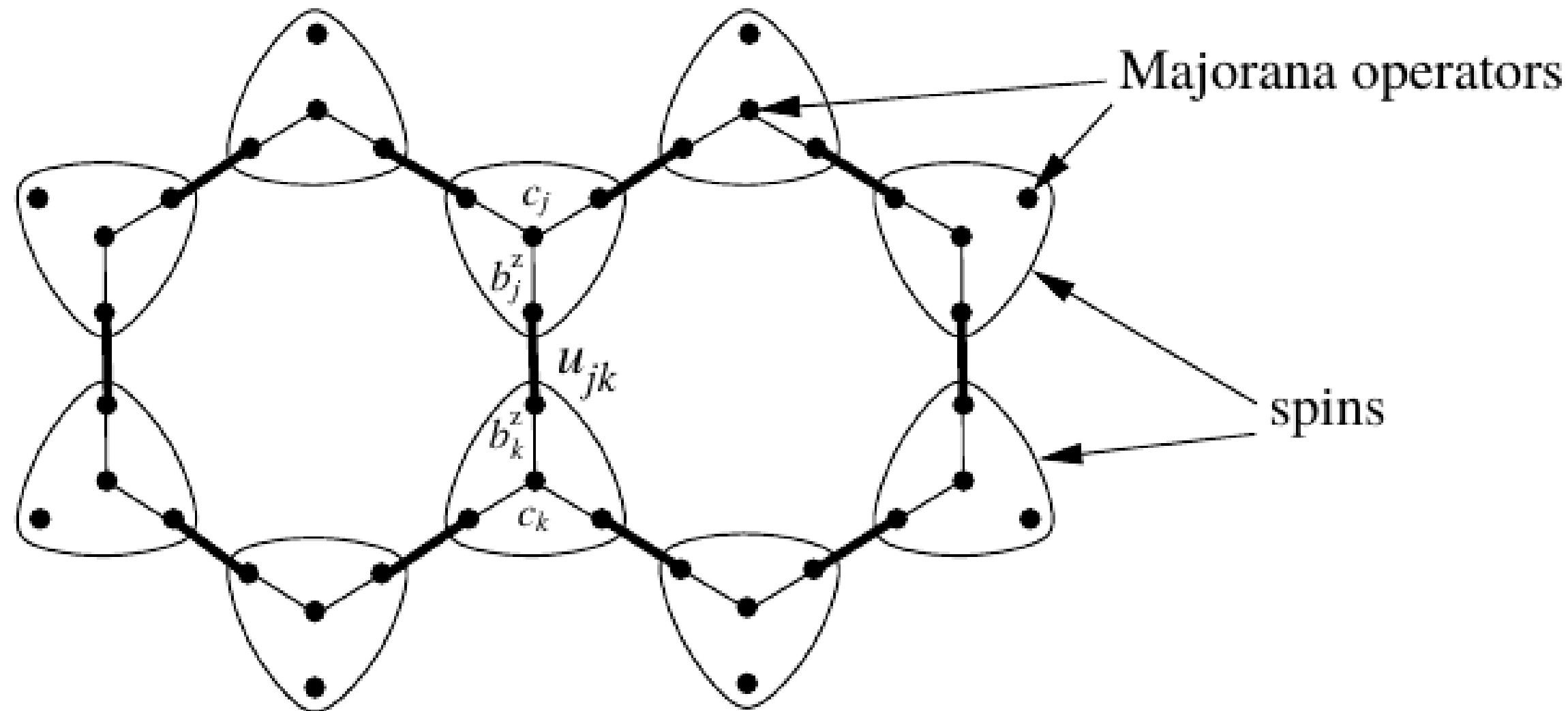
Kitaev model: Majorana fermions

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

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

Majorana fermions:
 $c_j^2 = 1, \quad c_i c_j = -c_j c_i, \quad i \neq j$

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



Free fermion solution!

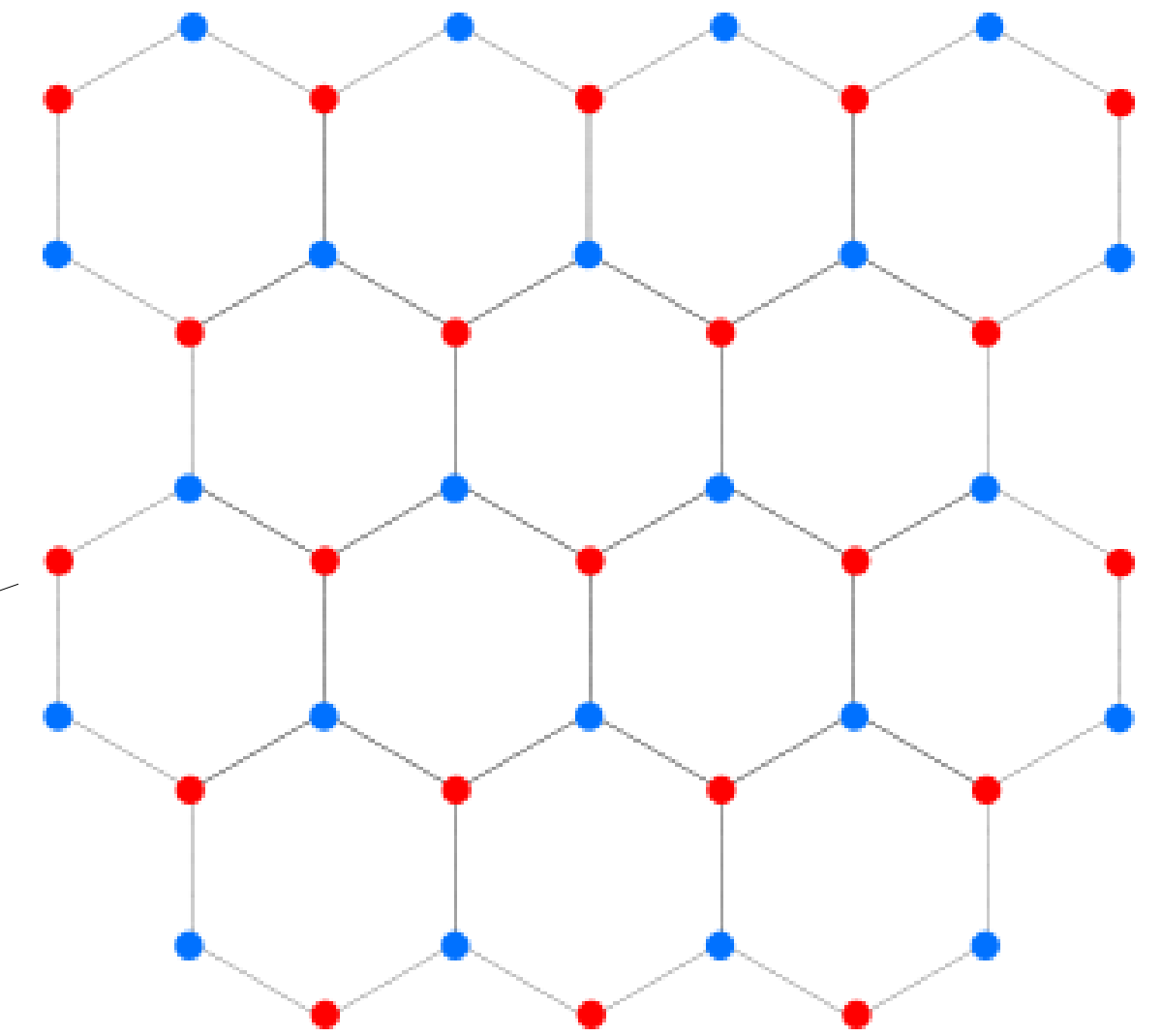
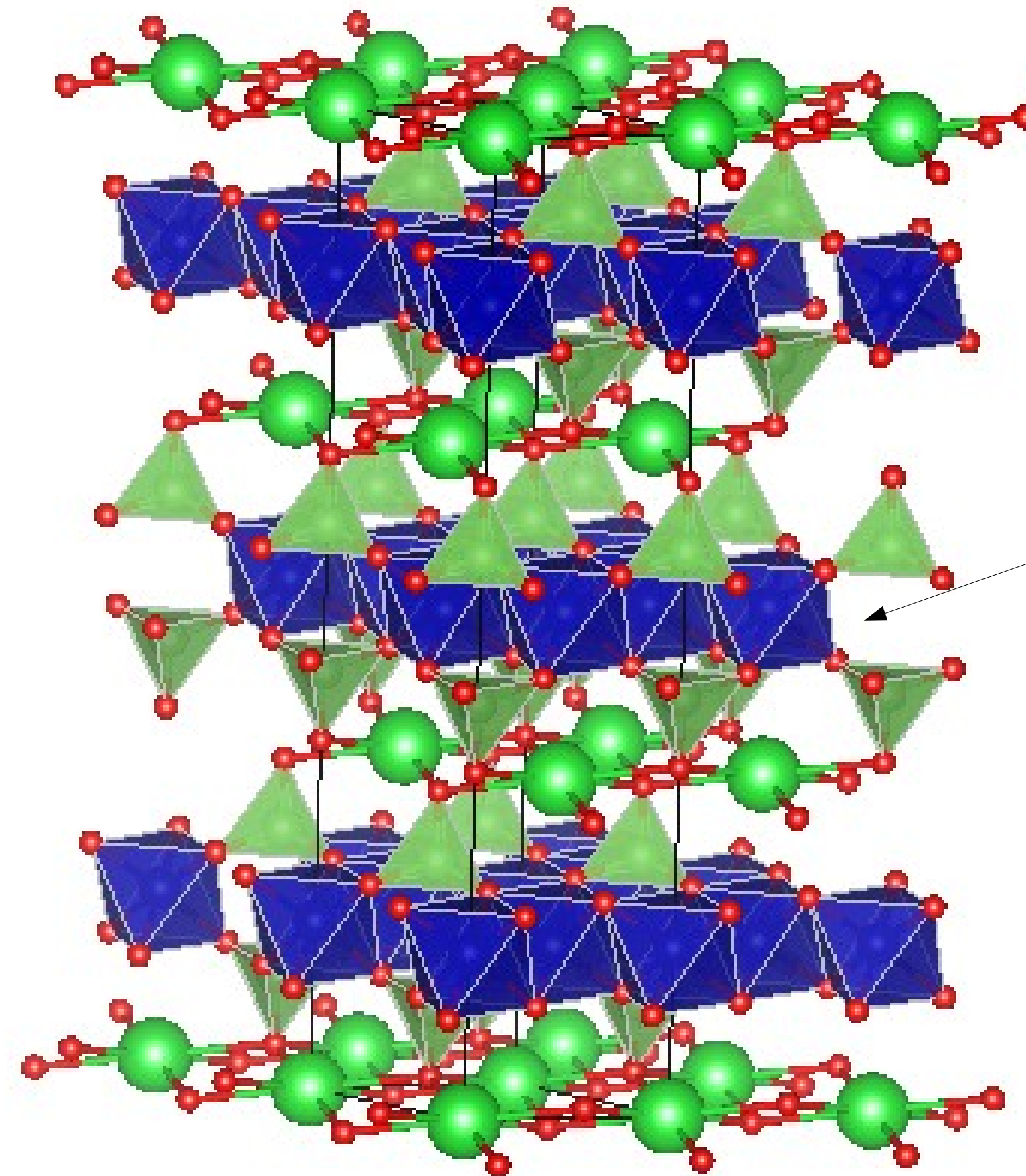
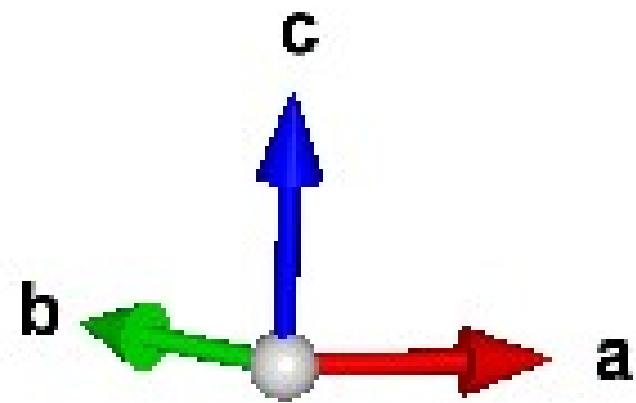
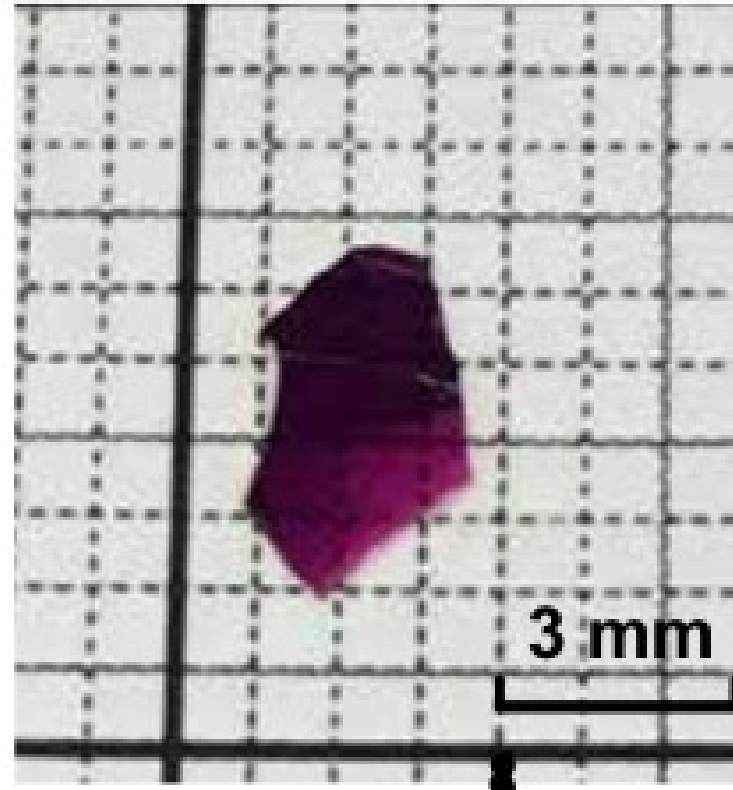
А. Китаев, А. Шень, М. Вялый

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 $\text{BaCo}_2(\text{AsO}_4)_2$

$\text{BaCo}_2(\text{AsO}_4)_2$: structure



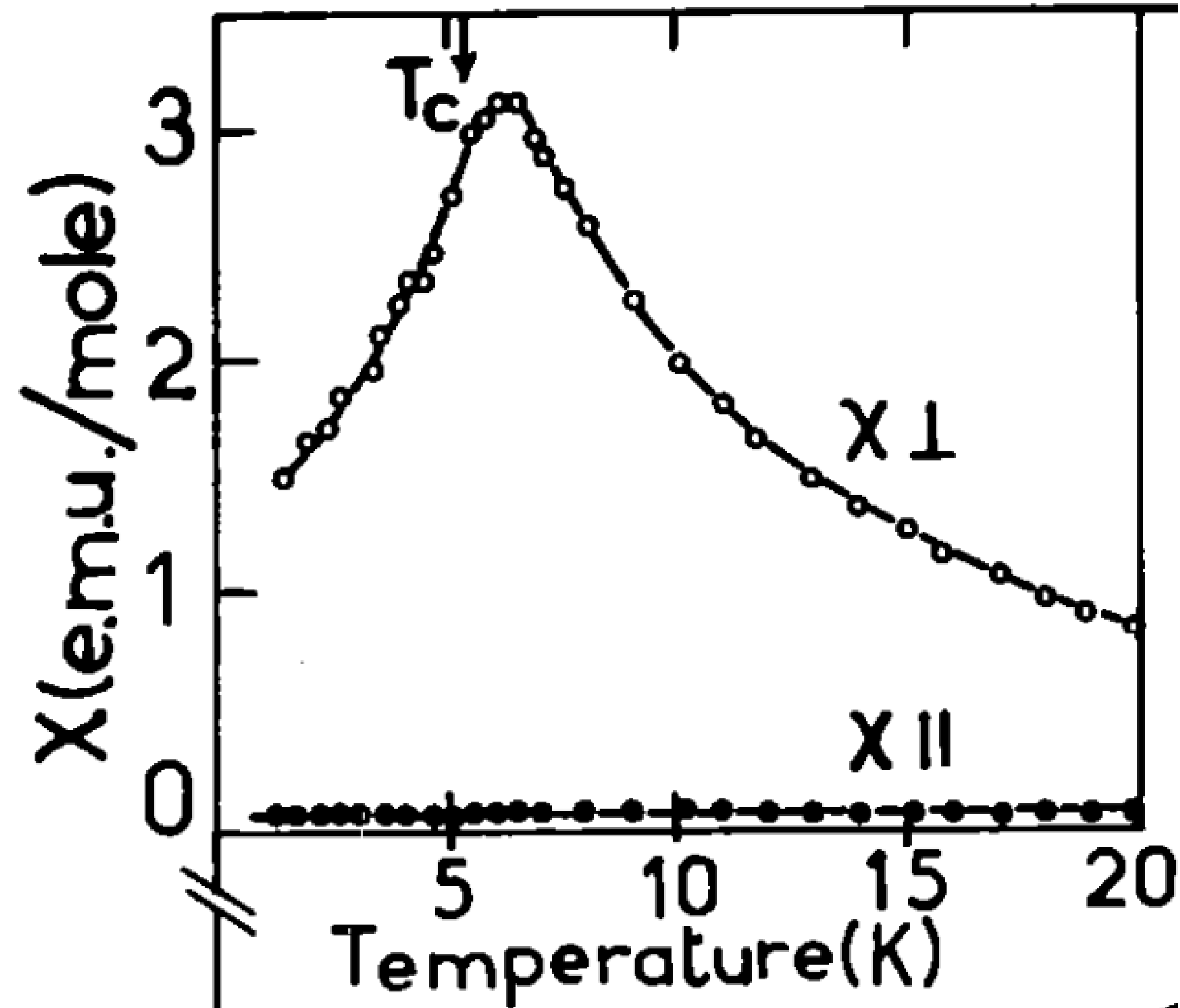
Honeycomb layers
(related to structure of mica)



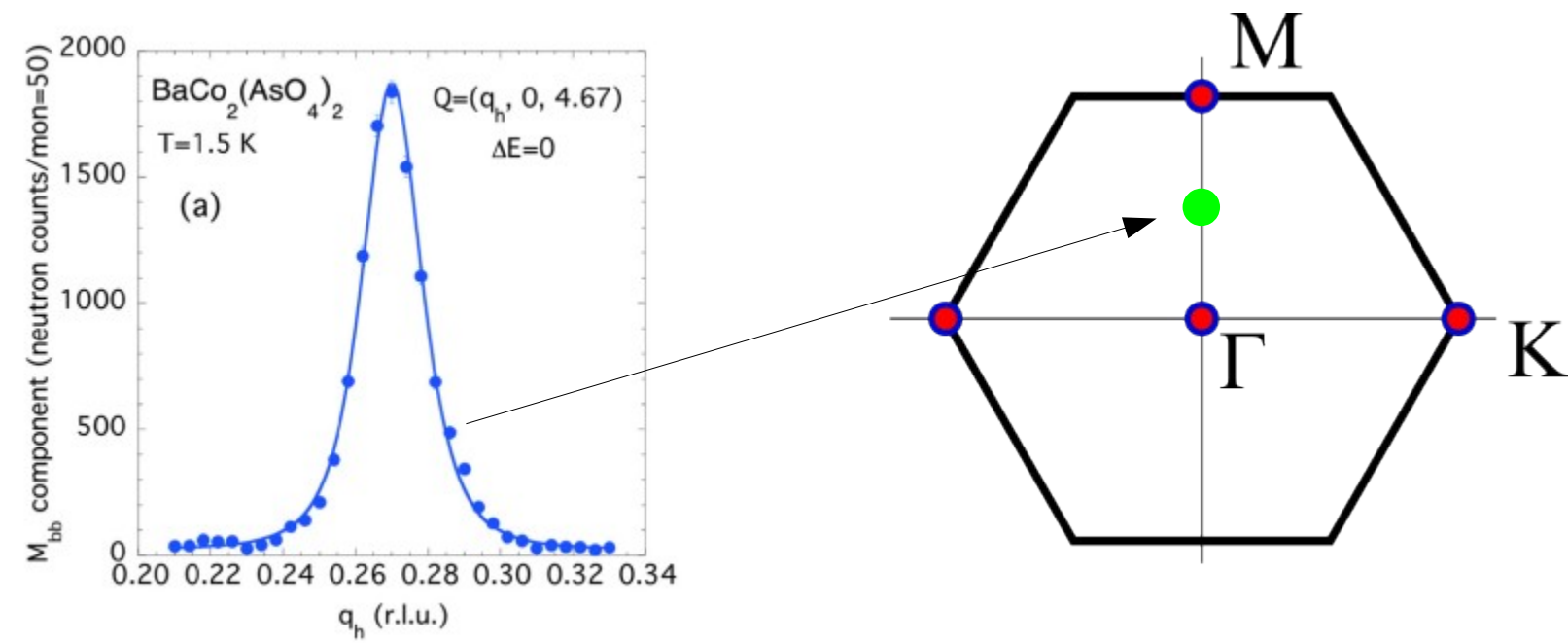
S. Eymond and A. Durif, Mater. Res. Bull. 4 595 (1969)

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

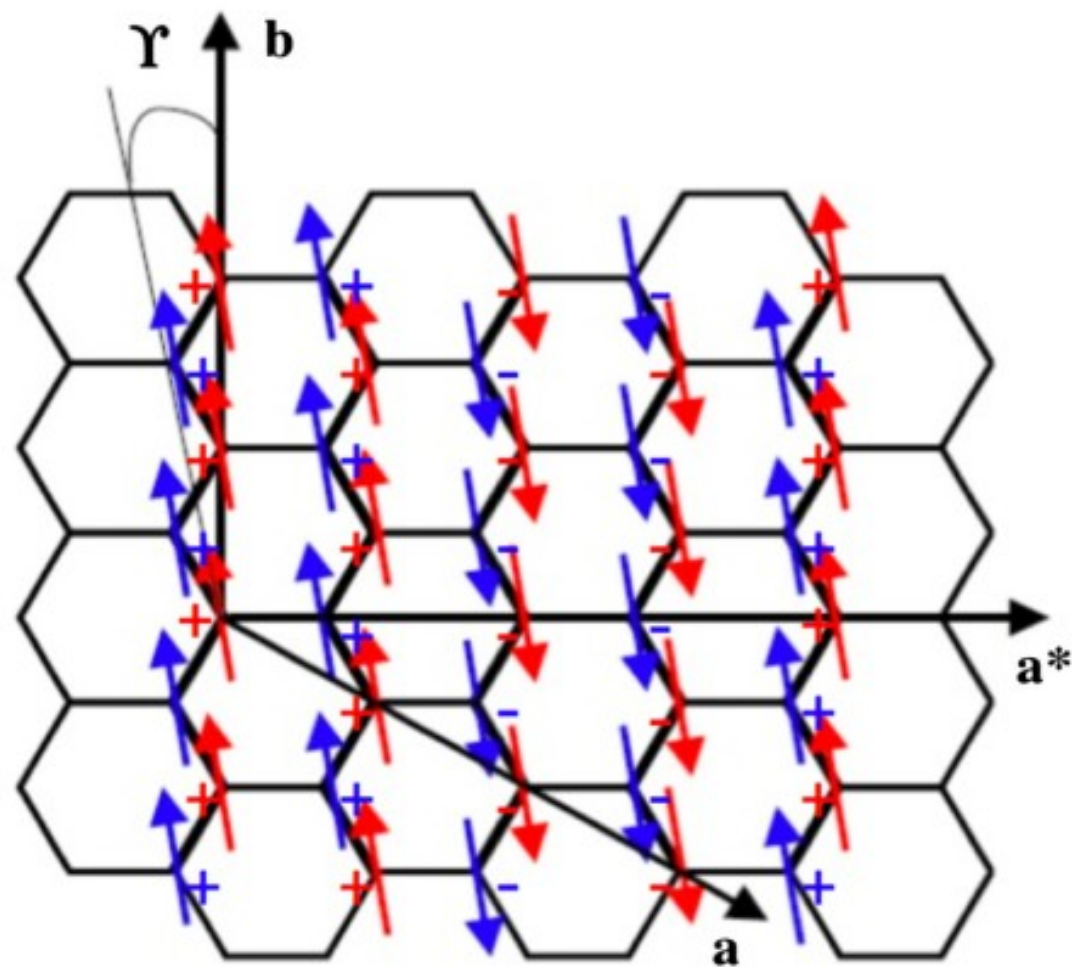
$\text{BaCo}_2(\text{AsO}_4)_2$: easy-plane anisotropy



BaCo₂(AsO₄)₂: «double zigzag»+plateau

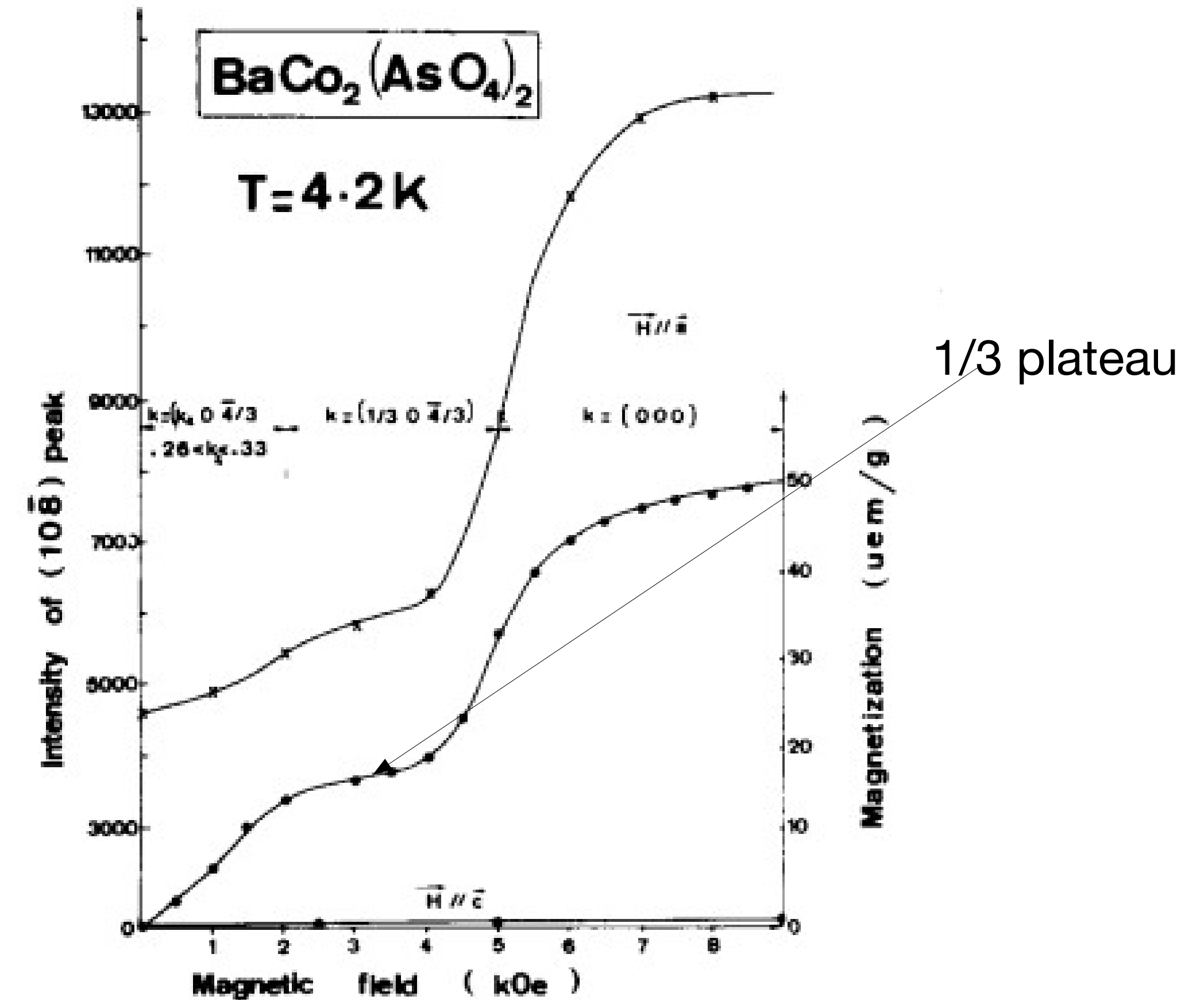


L.P. Regnault et al., Physica B+C 86, 660 (1977)



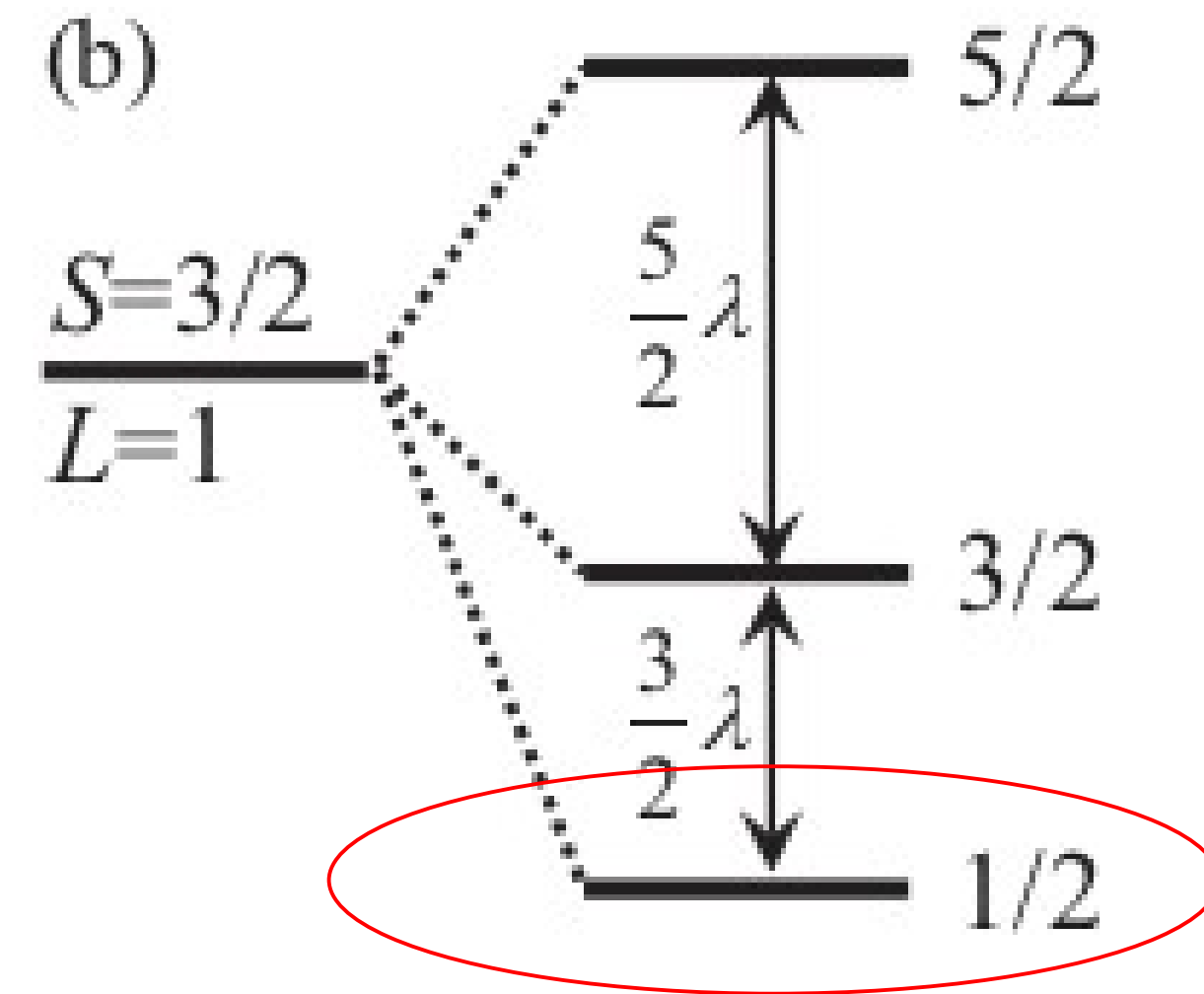
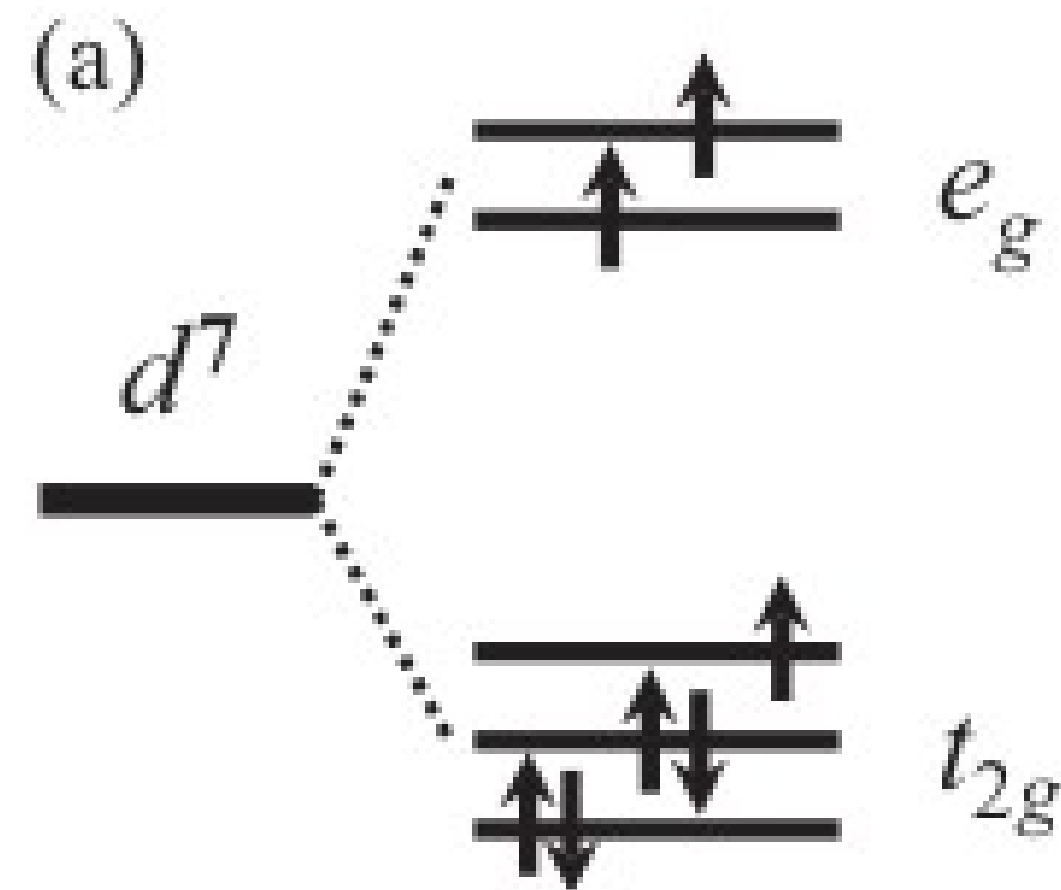
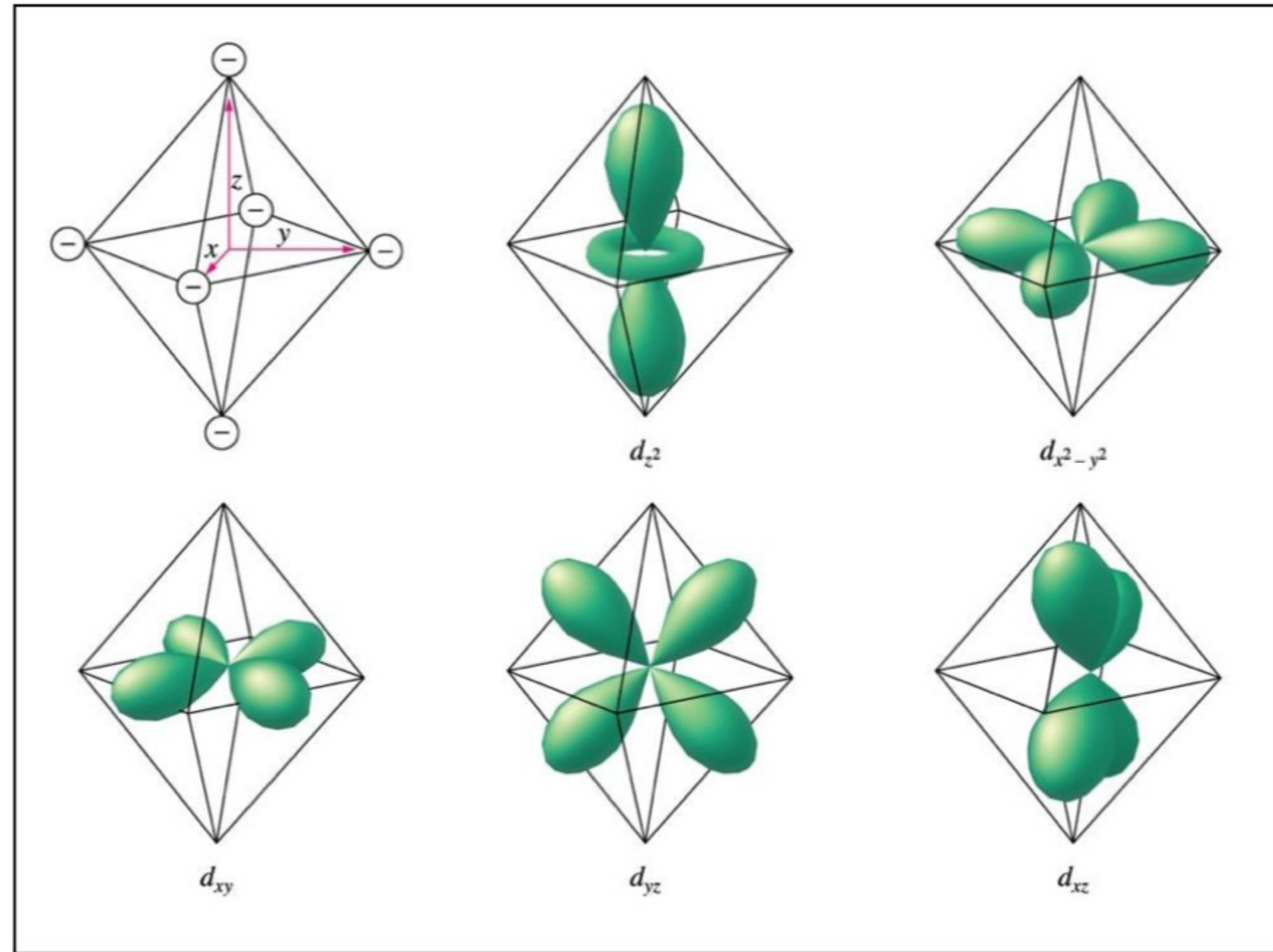
L. Regnault et al., Physica B: Condensed Matter 385-386, 425 (2006)

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



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

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



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

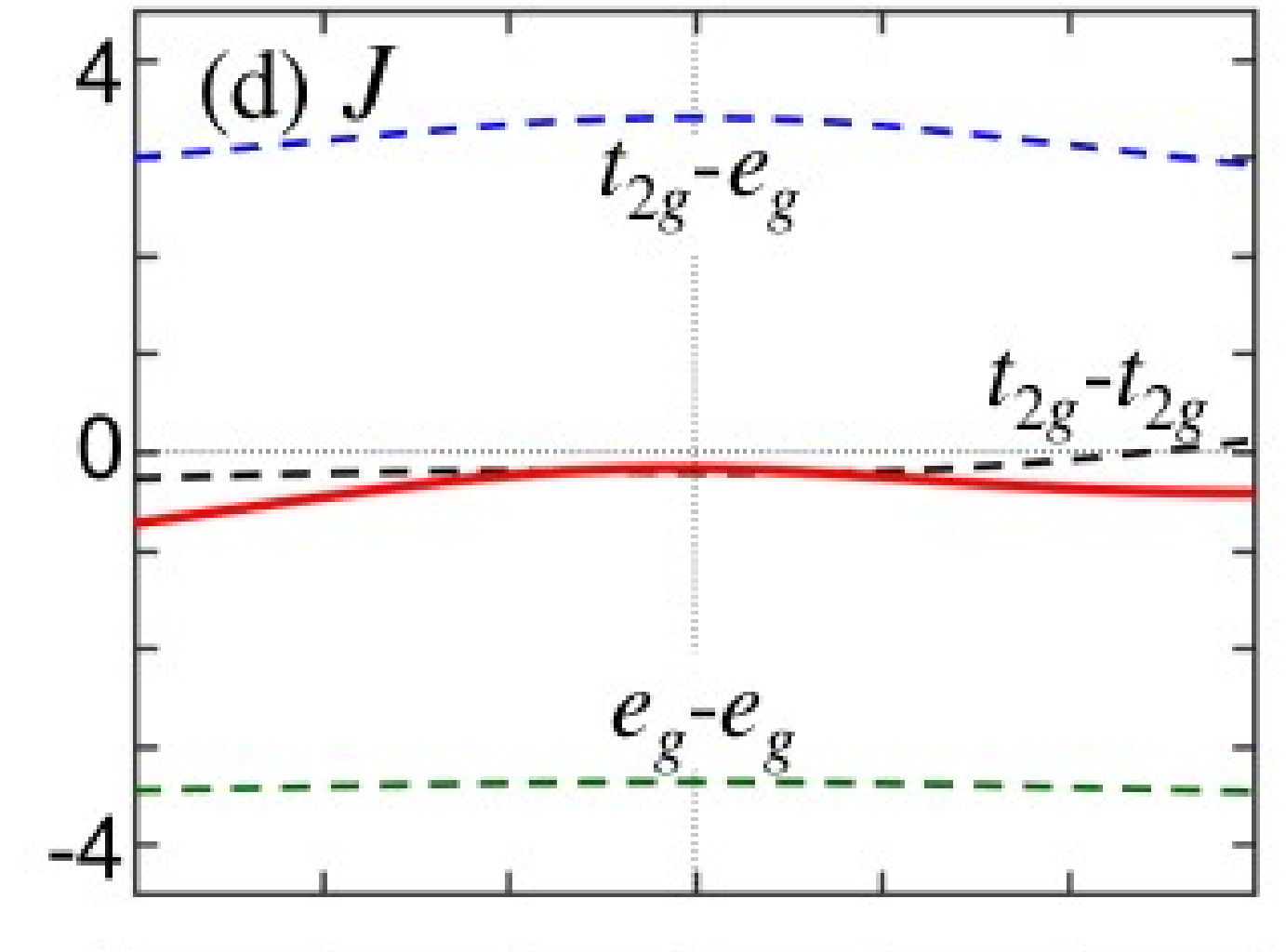
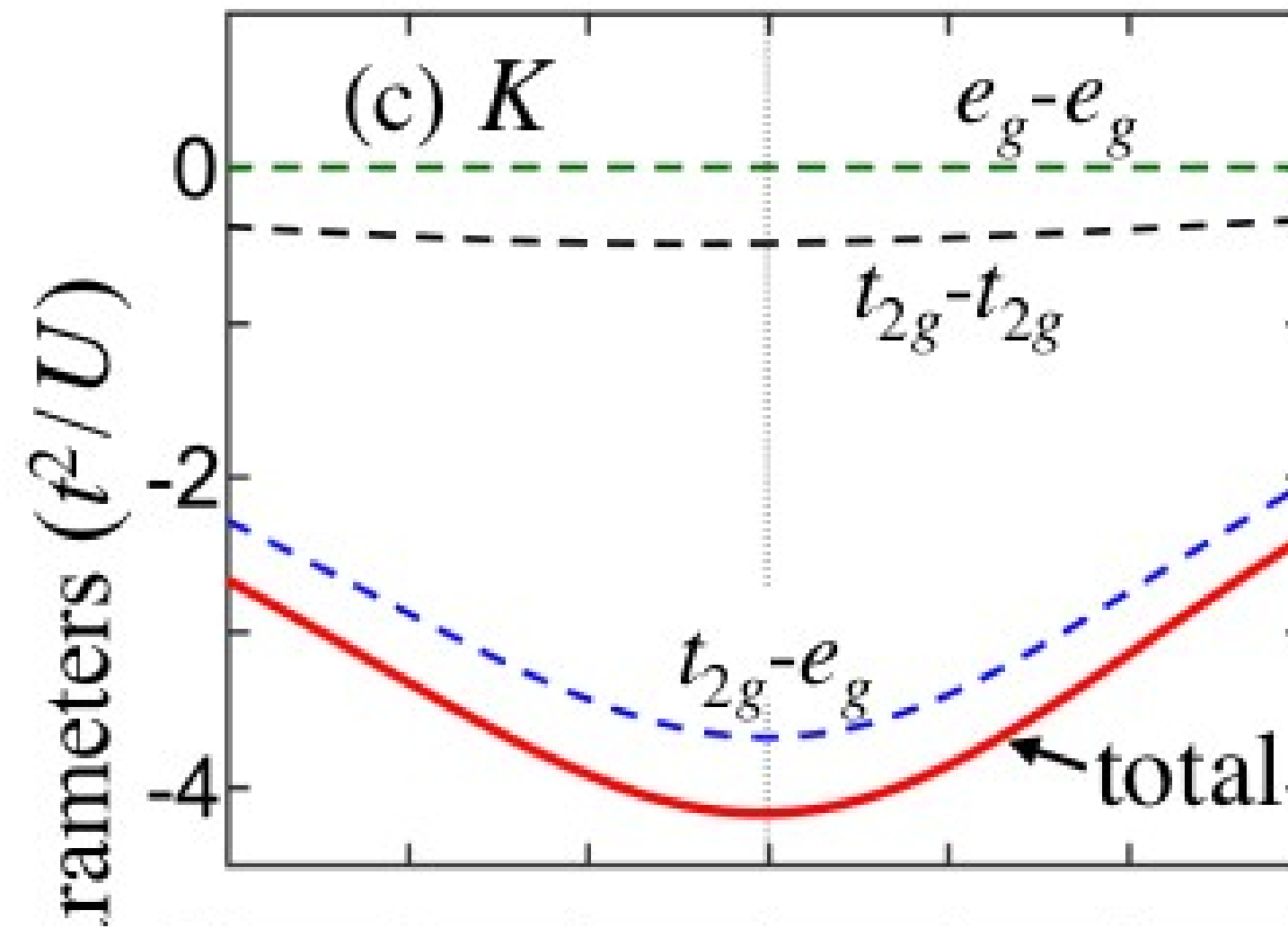
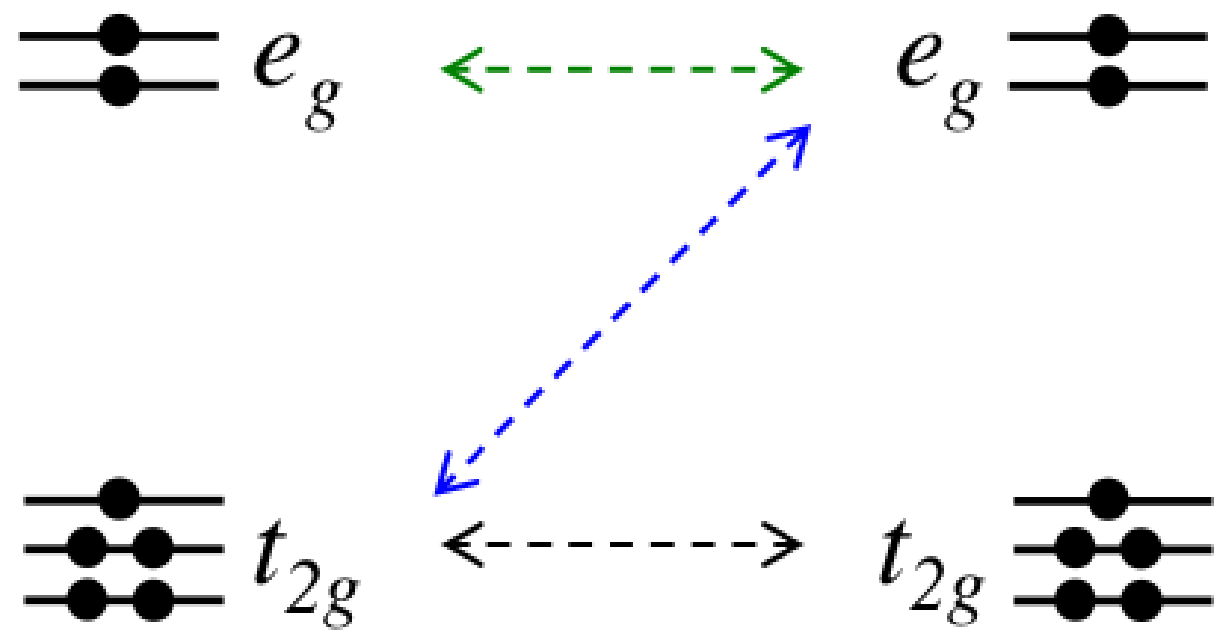
$$\psi_{-1} = c_1 | 1, -\frac{3}{2} \rangle + c_2 | 0, -\frac{1}{2} \rangle + c_3 | -1, \frac{1}{2} \rangle,$$

Abraham 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+}



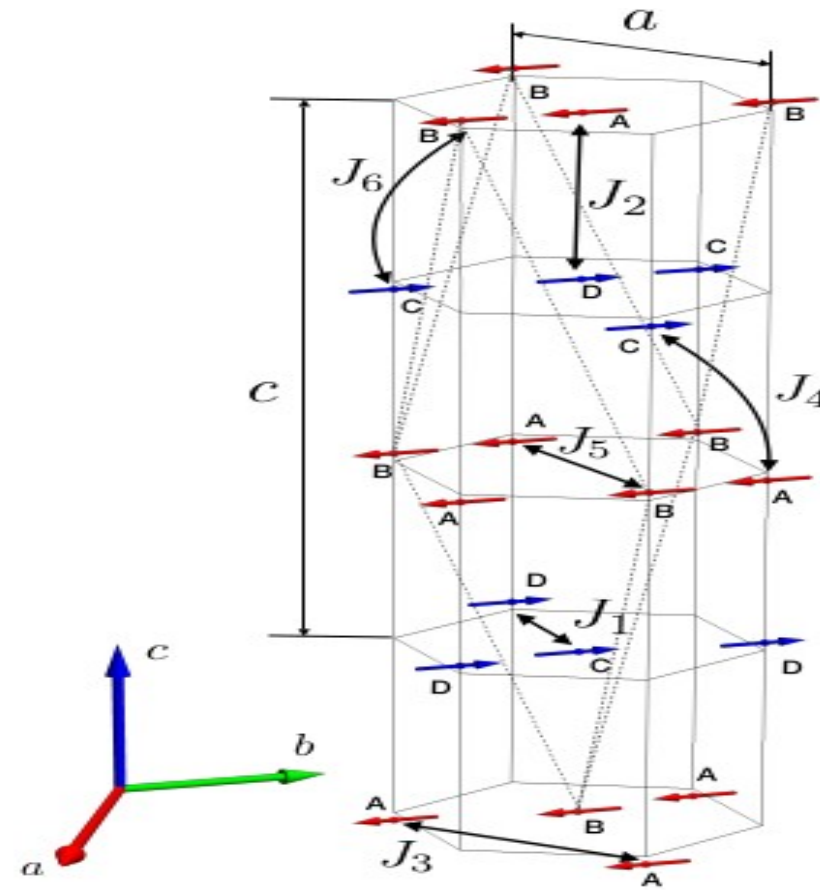
$$\mathcal{H} = \sum_{\langle ij \rangle^\gamma} J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma \left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \right) + \Gamma' \left(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma \right)$$

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

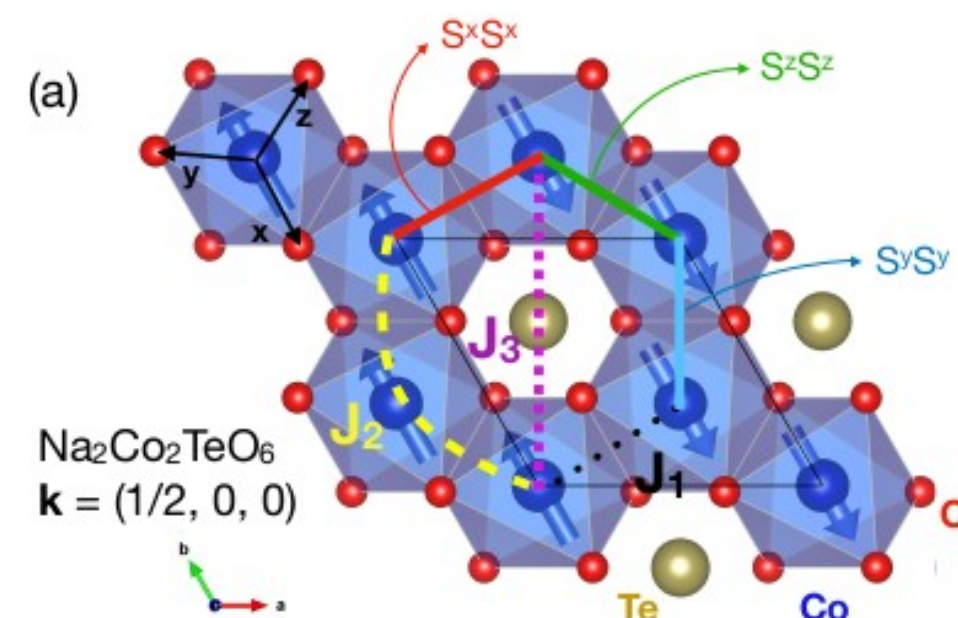


XXZ+(small bond-dependent)

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

$\text{Na}_2\text{Co}_2\text{TeO}_6$

$\text{Na}_3\text{Co}_2\text{SbO}_6$



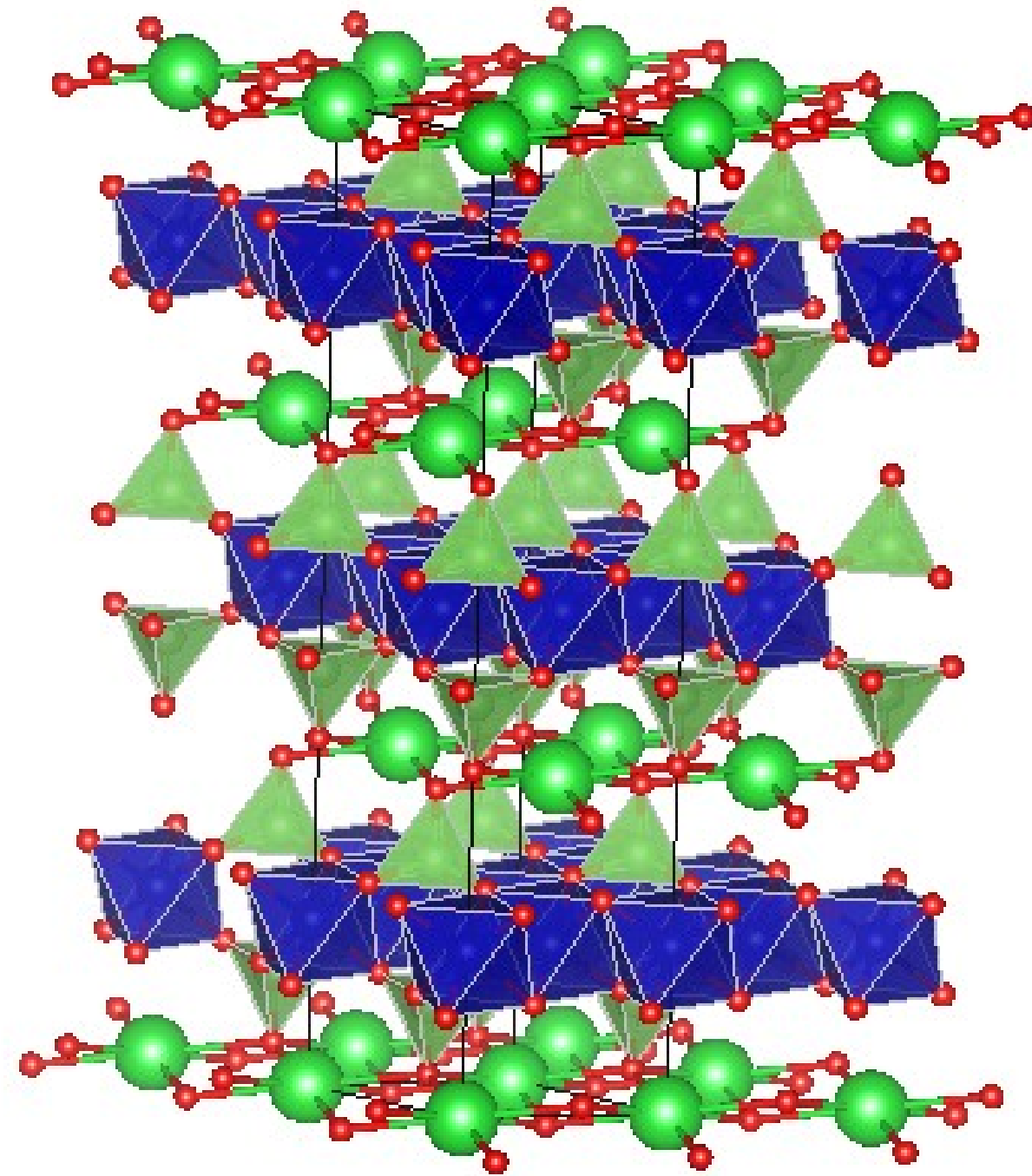
Large K, large J_3

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 , $\text{BaCo}_2\text{V}_2\text{O}_8$

BaCo₂(AsO₄)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**

$$J_3 > 0.5 J_1$$

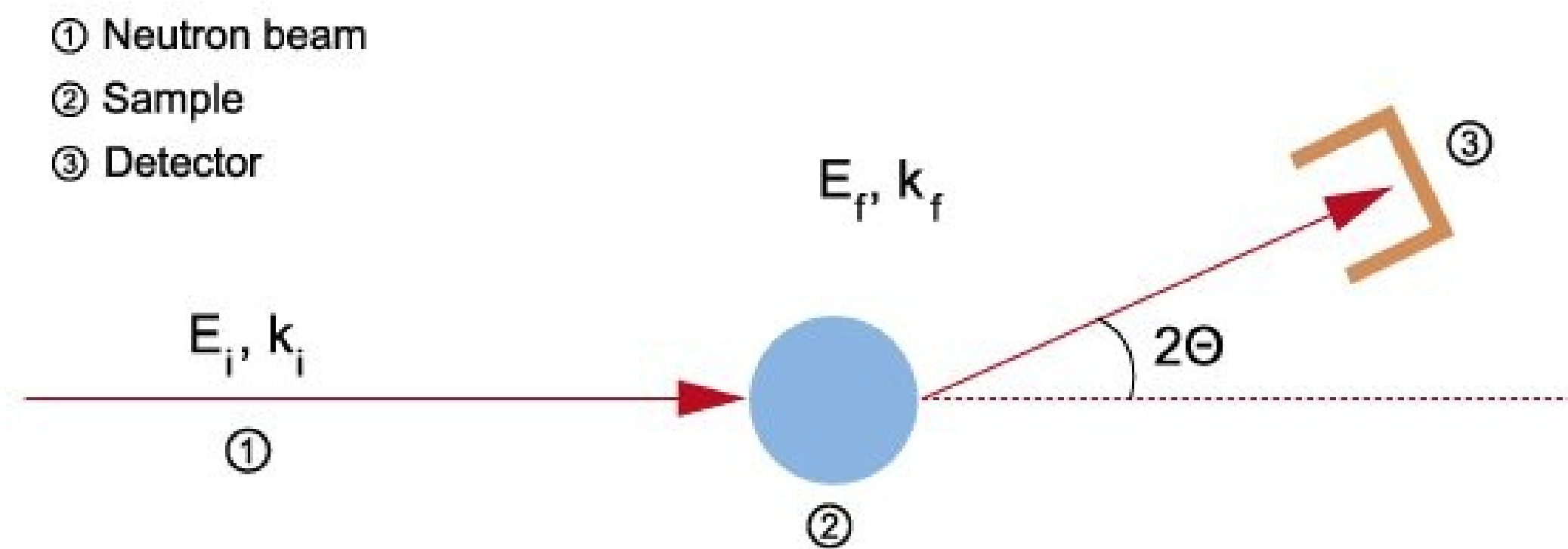
Agreement with c S. Das et al., Phys. Rev. B 104, 134425 (2021)

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

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

Neutron scattering

Neutron diffraction

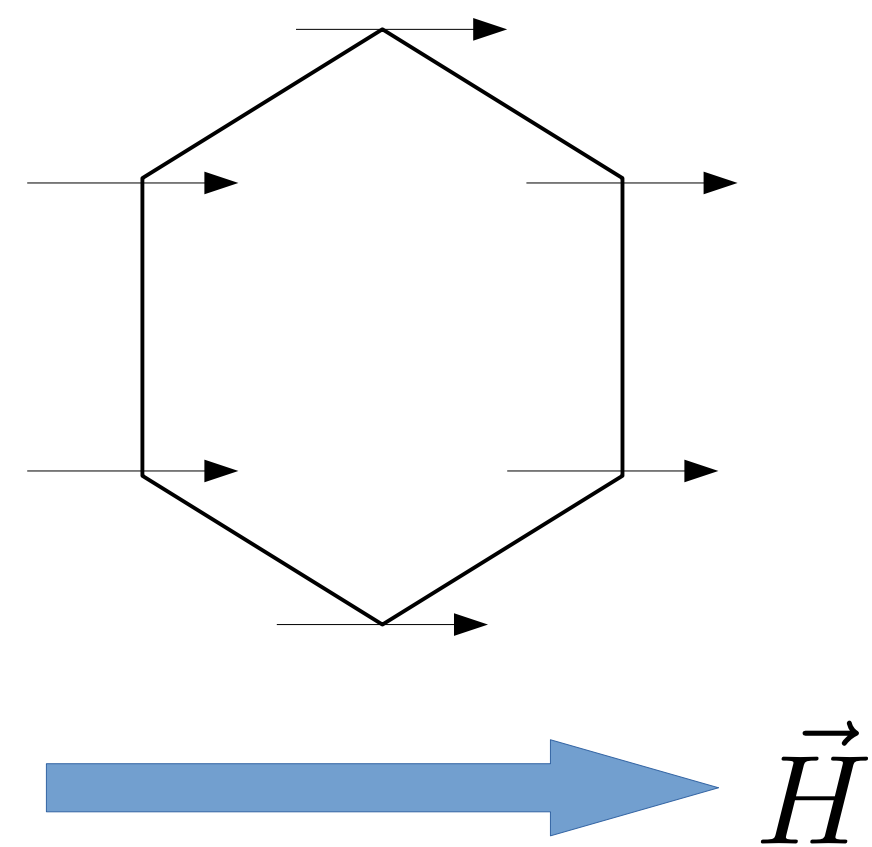


$$Q = k_f - k_i$$
$$\hbar\omega = E_f - E_i$$

Energy and
momentum of
the excitation

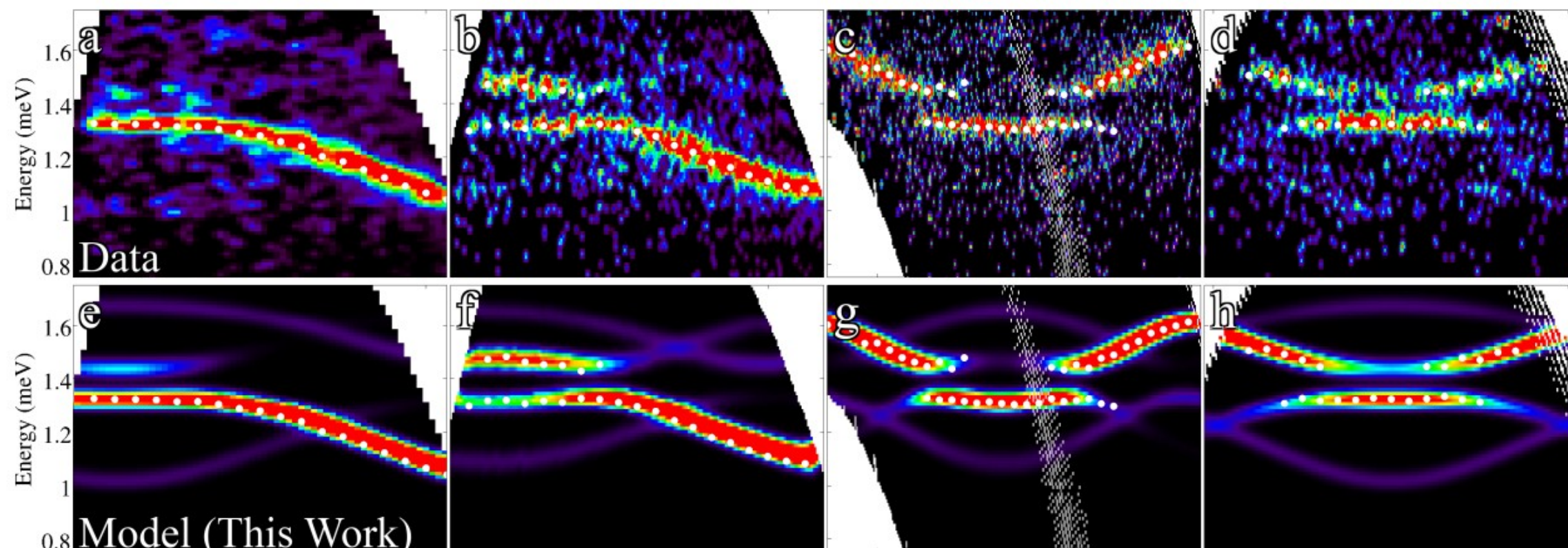
Inelastic neutron scattering in $\text{BaCo}_2(\text{AsO}_4)_2$

In polarized state spin-wave theory is almost exact $\rightarrow$ spectrum fit $\rightarrow$ exchange integrals



$$\begin{aligned} S^+ &\approx a \\ S^- &\approx a^\dagger \\ S^z &= S - a^\dagger a \end{aligned}$$

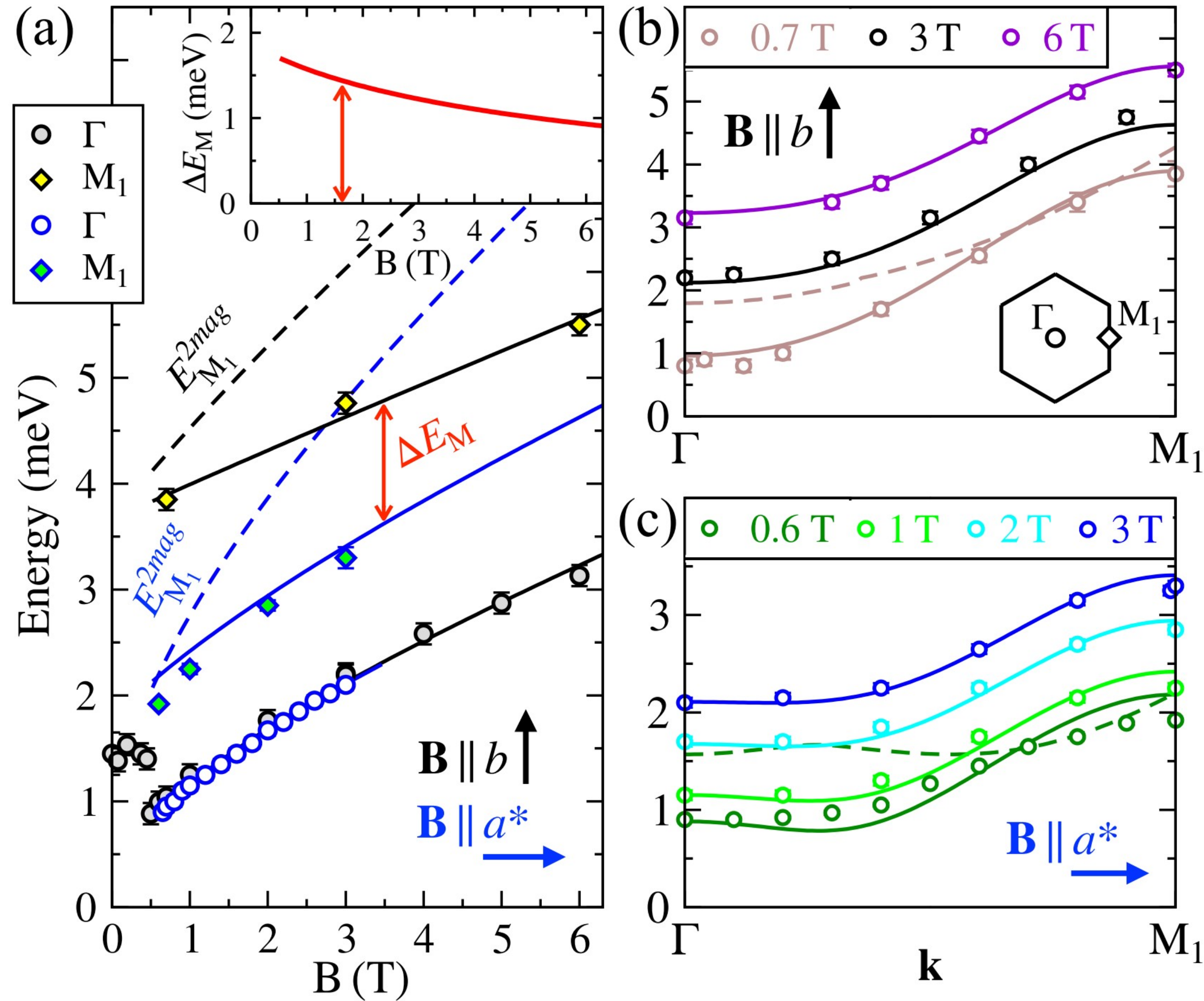
$\text{Yb}_2\text{Ti}_2\text{O}_7$ pyrochlore



$$\begin{aligned} \mathcal{H}_{\text{Exchange}} = \sum_{\langle ij \rangle} \{ & J_{zz} S_i^z S_j^z - J_{\pm} (S_i^+ S_j^- + S_i^- S_j^+) \\ & + J_{\pm\pm} (\gamma_{ij} S_i^+ S_j^+ + \gamma_{ij}^* S_i^- S_j^-) \\ & + J_{z\pm} [S_i^z (\zeta_{ij} S_j^+ + \zeta_{ij}^* S_i^-) + (i \leftrightarrow j)] \} \end{aligned}$$

6 parameters: 4 exchanges + 2 g-factors

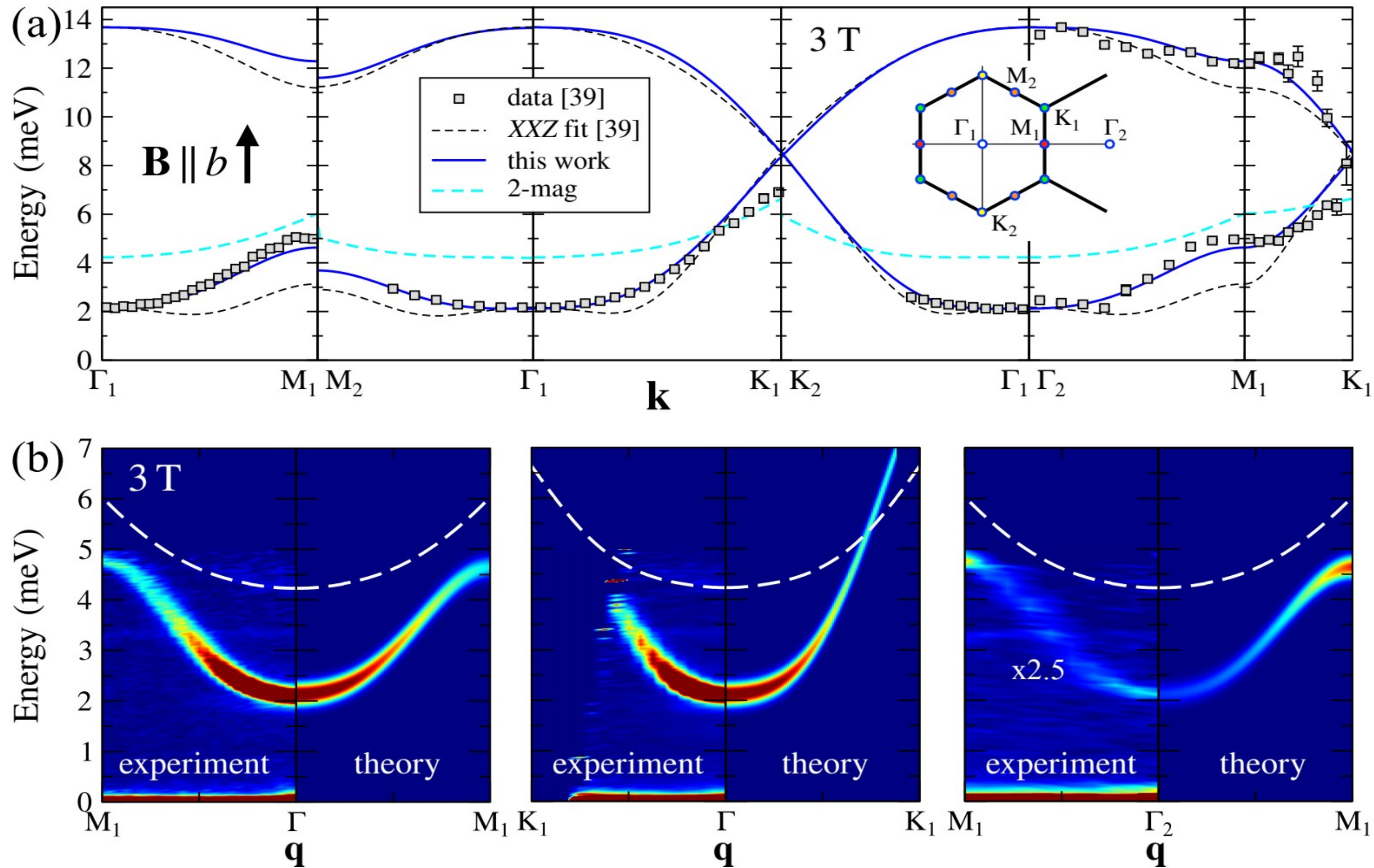
Inelastic neutron scattering in $\text{BaCo}_2(\text{AsO}_4)_2$



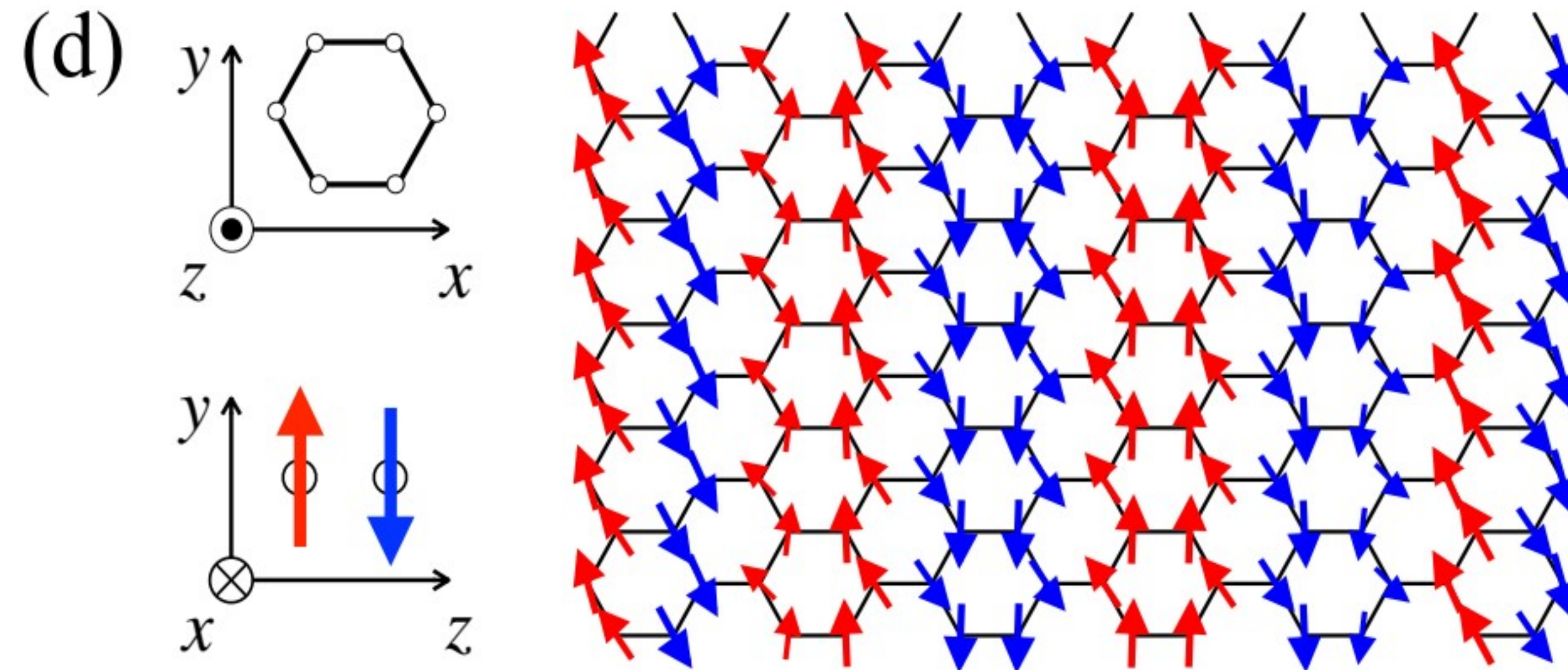
Dots – data from L.P. Regnault, Grenoble
Lines – linear spin-wave theory

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

Inelastic neutron scattering in $\text{BaCo}_2(\text{AsO}_4)_2$: fitting quality

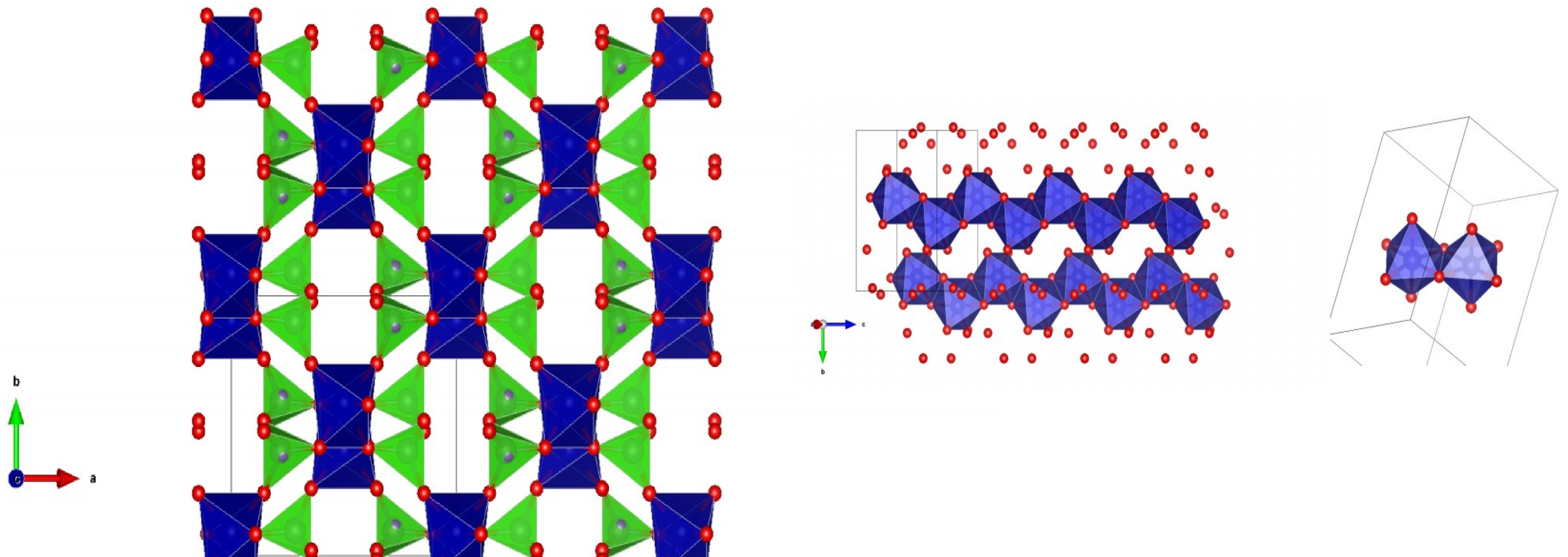


DMRG: double zigzag



Pyroxene $\text{SrCoGe}_2\text{O}_6$

Pyroxene $\text{SrCoGe}_2\text{O}_6$



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

Magnetic structure

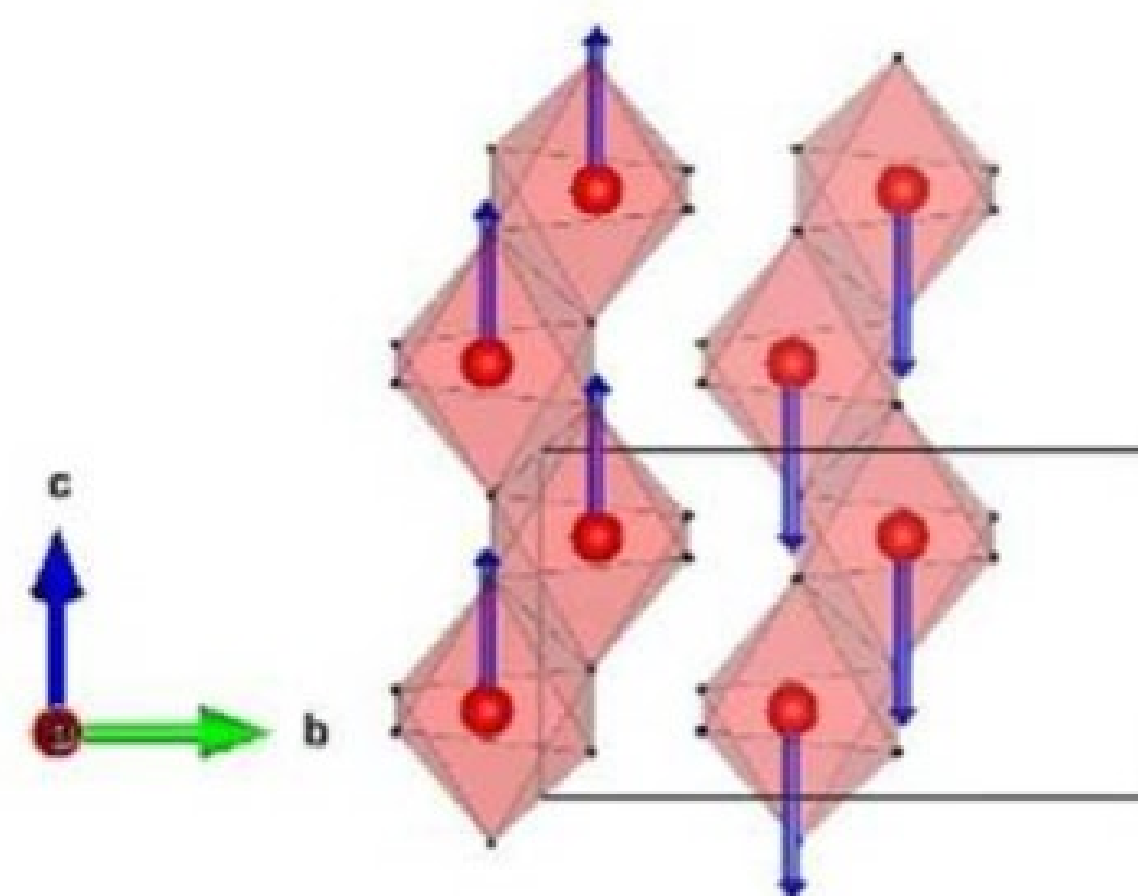
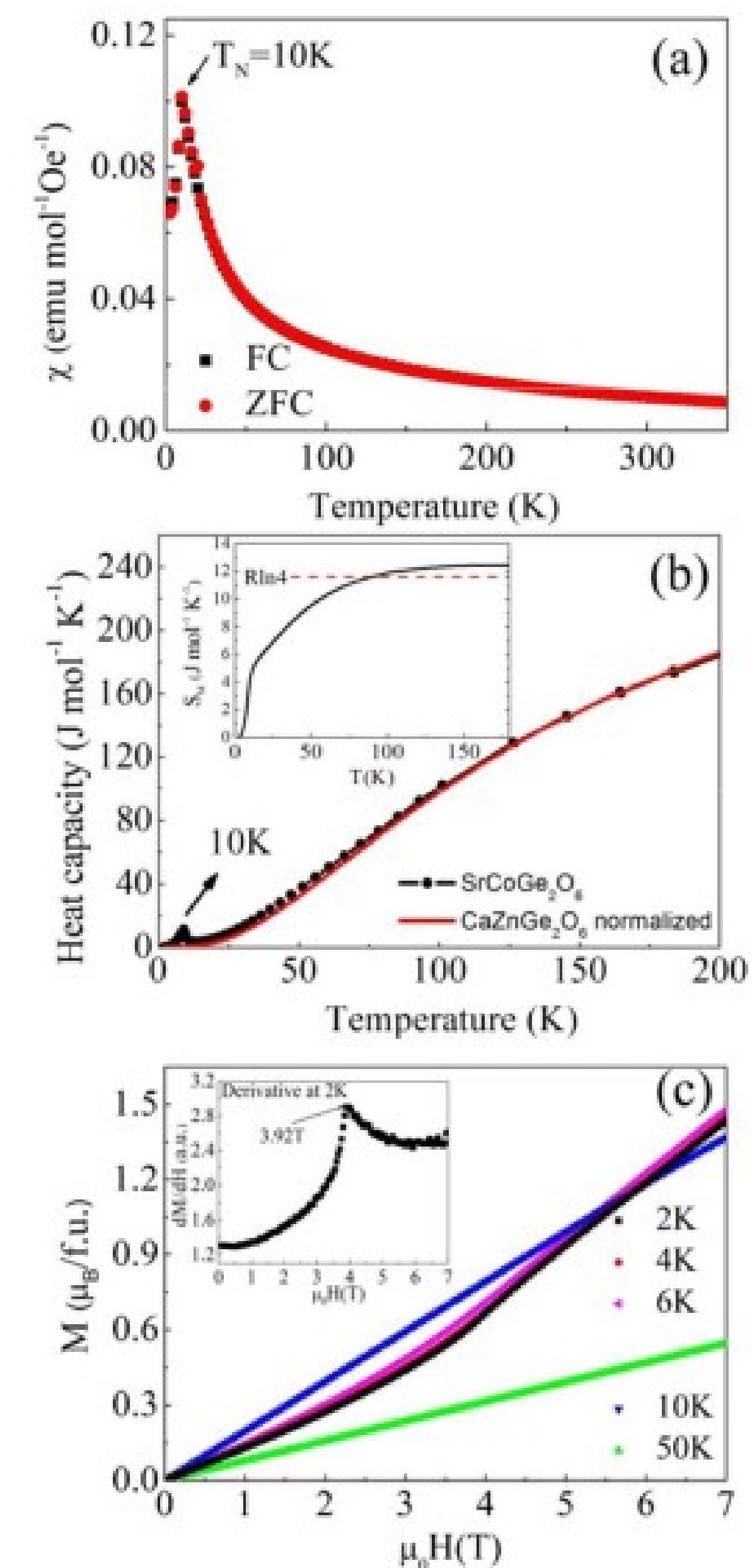
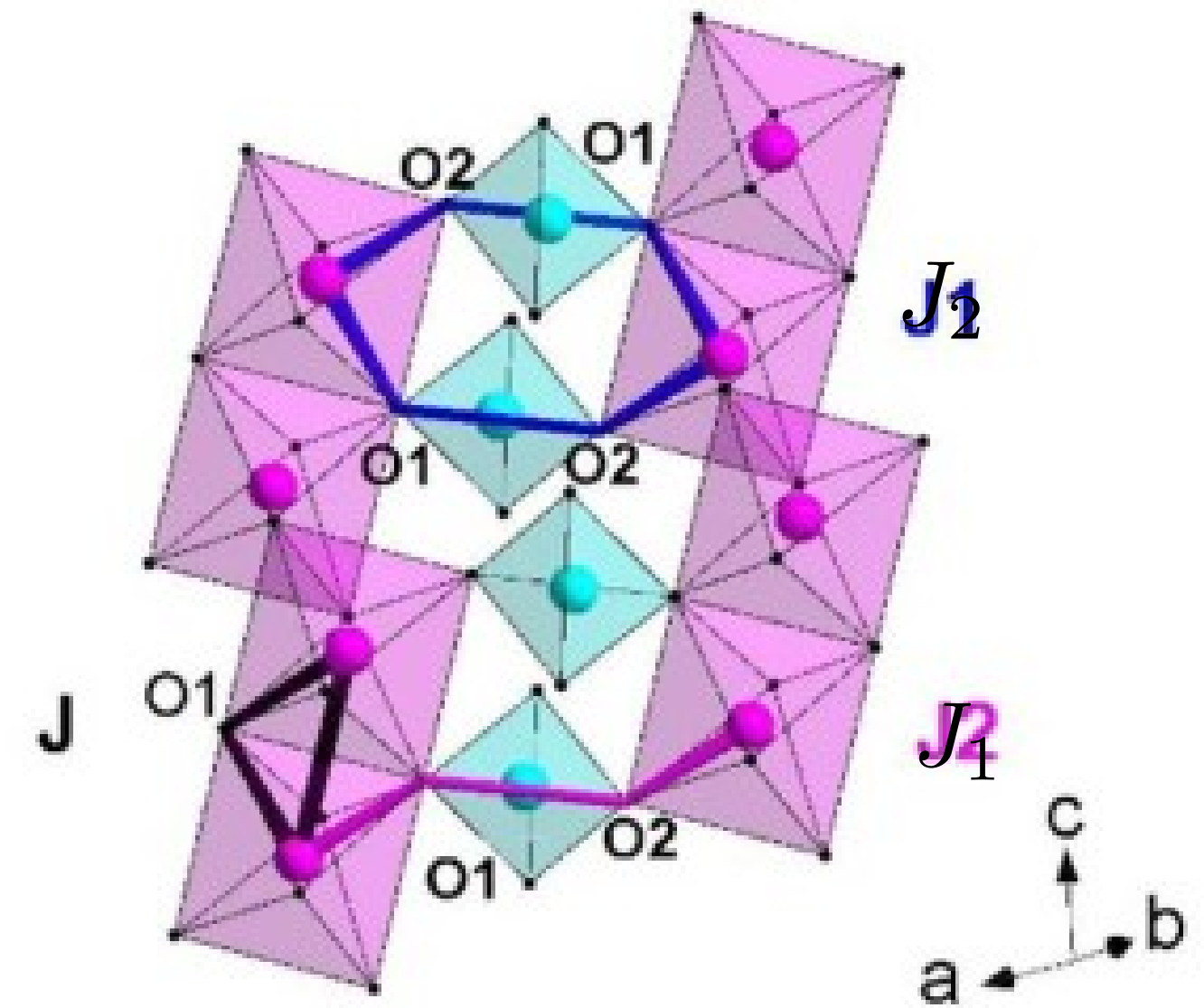
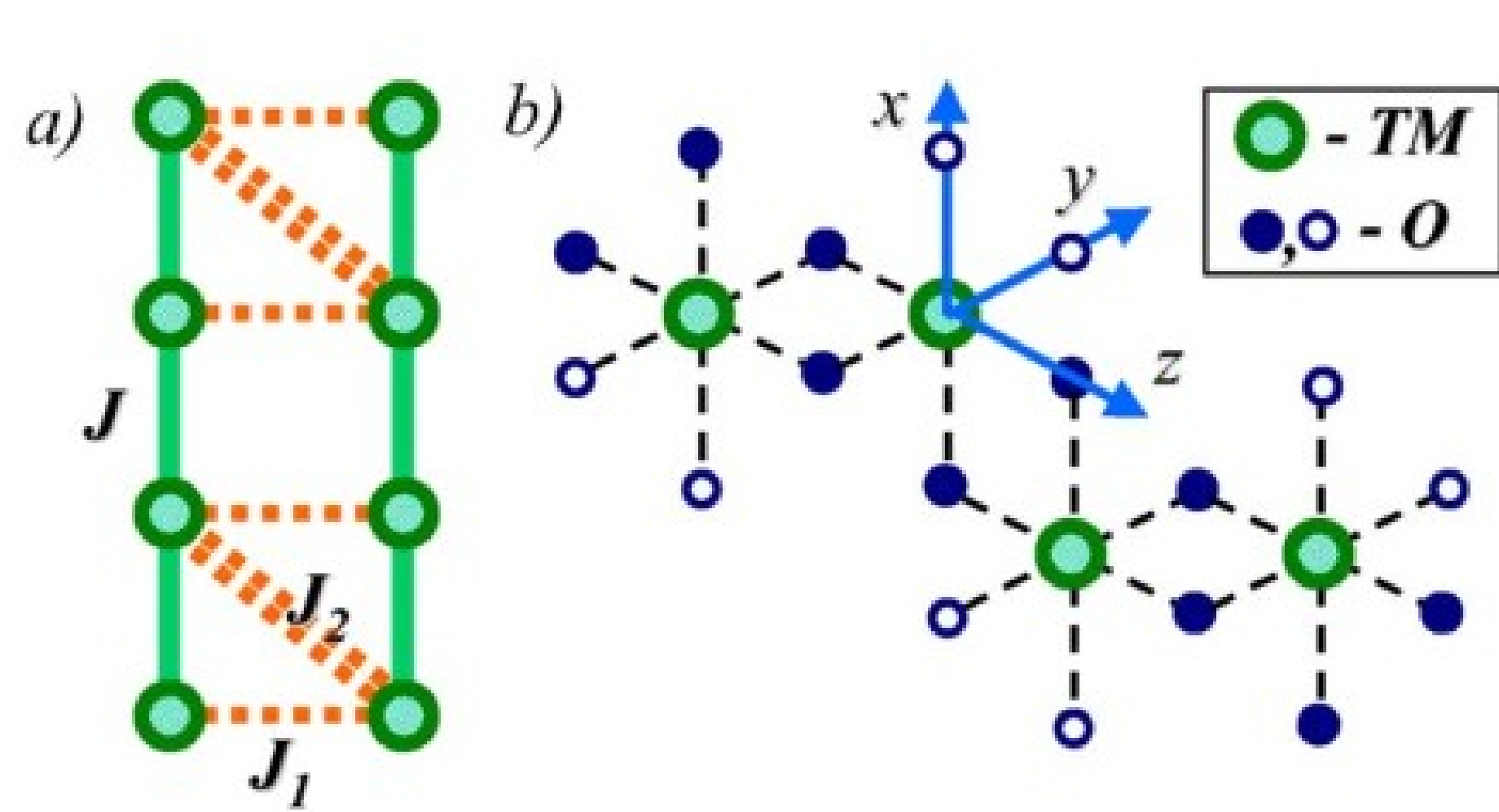


Fig. 8. Representation of the magnetic structure of $\text{SrCoGe}_2\text{O}_6$



Ab initio calculations



DFT results:

$J = -12$ K (FM)

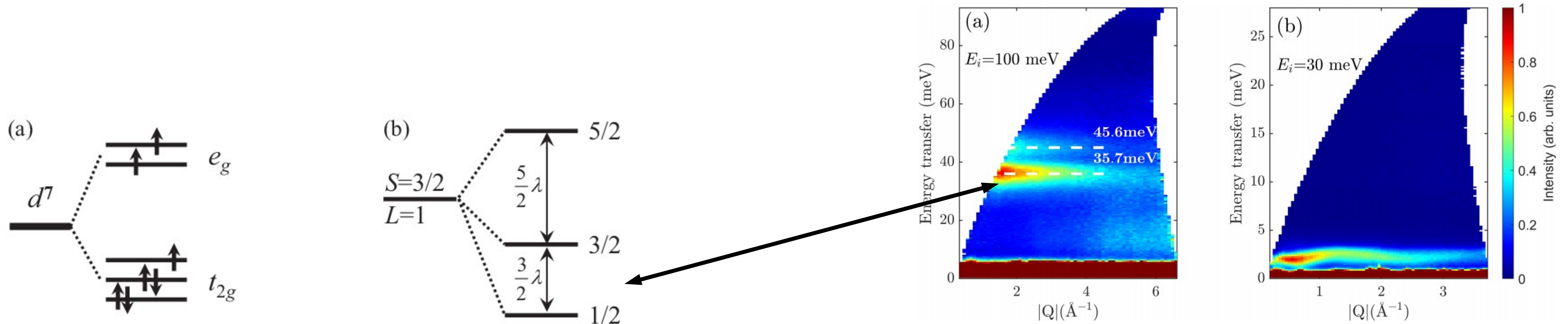
$J_1 = 8$ K (AF)

$J_2 = 11$ K (AF)

Strong inter-chain coupling $\rightarrow$ long-range order $\rightarrow$ spin-wave theory

Effective pseudospin $\frac{1}{2}$ of Co^{2+}

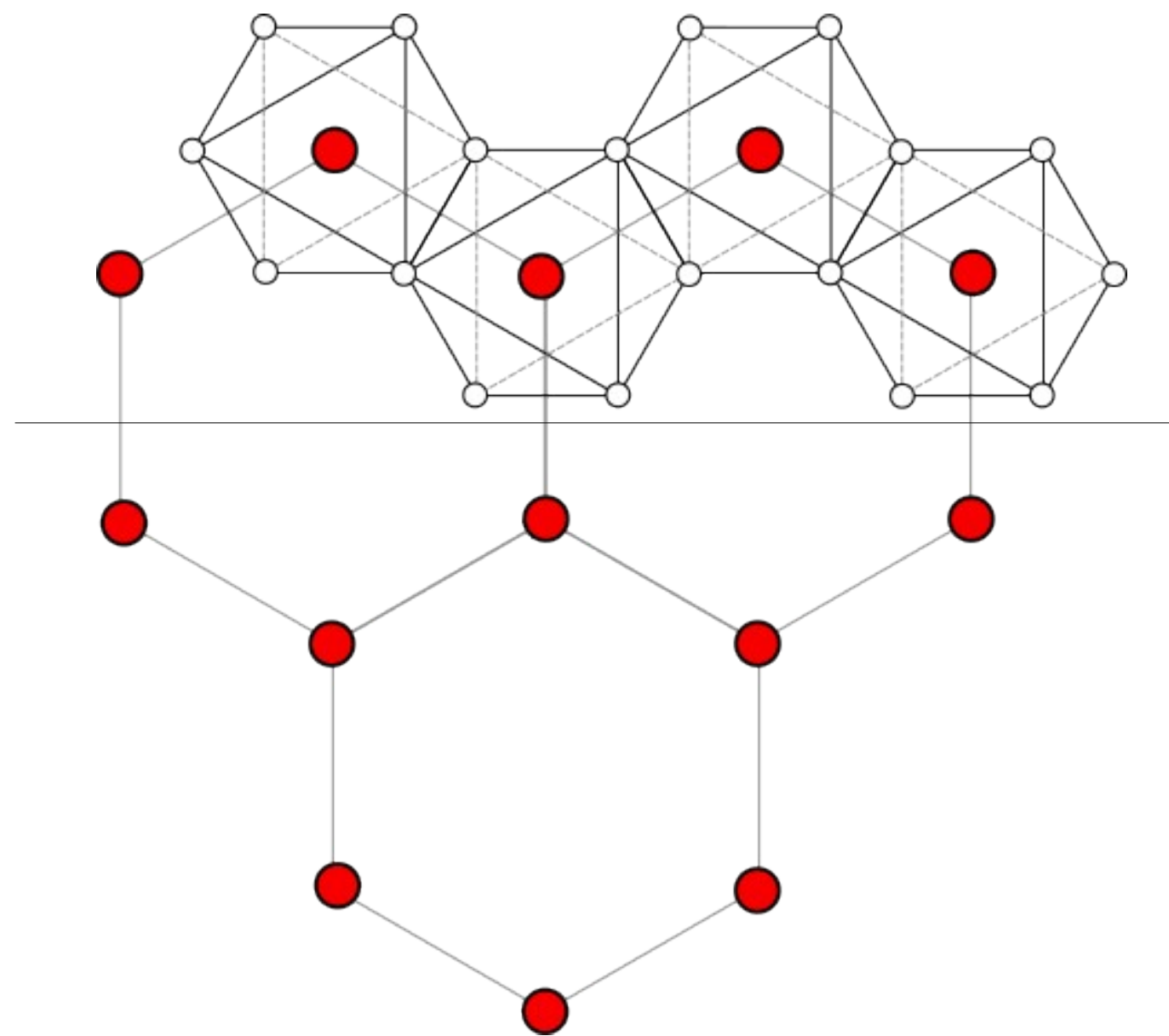
Oak Ridge powder scattering data



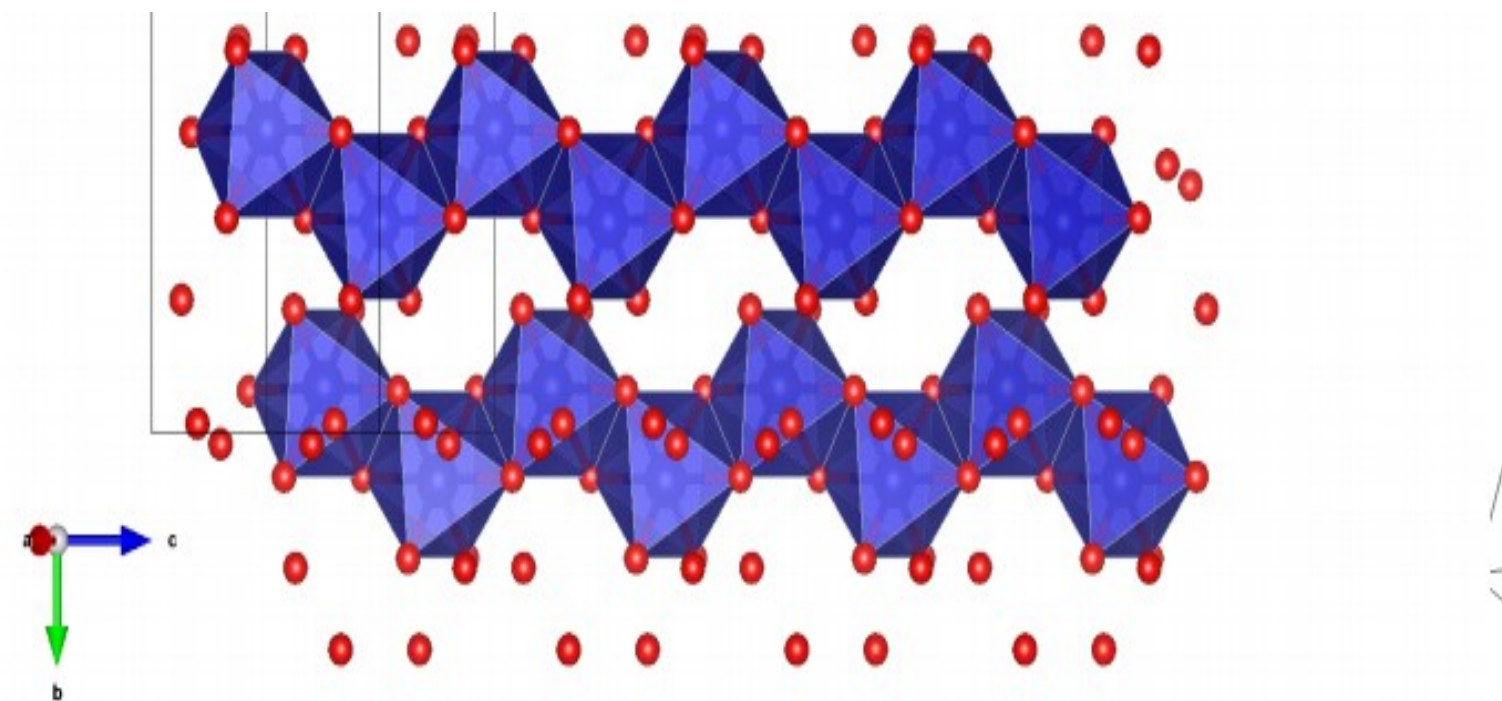
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)

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

Kitaev-Heisenberg ansatz

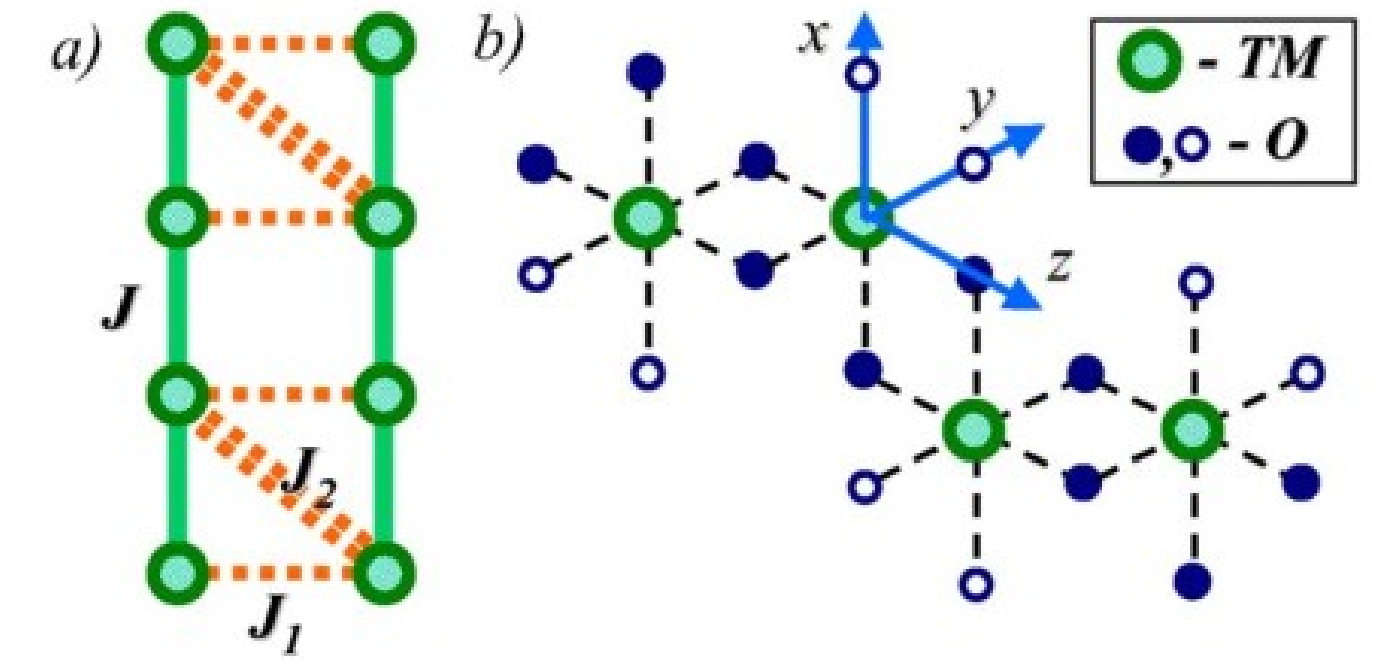
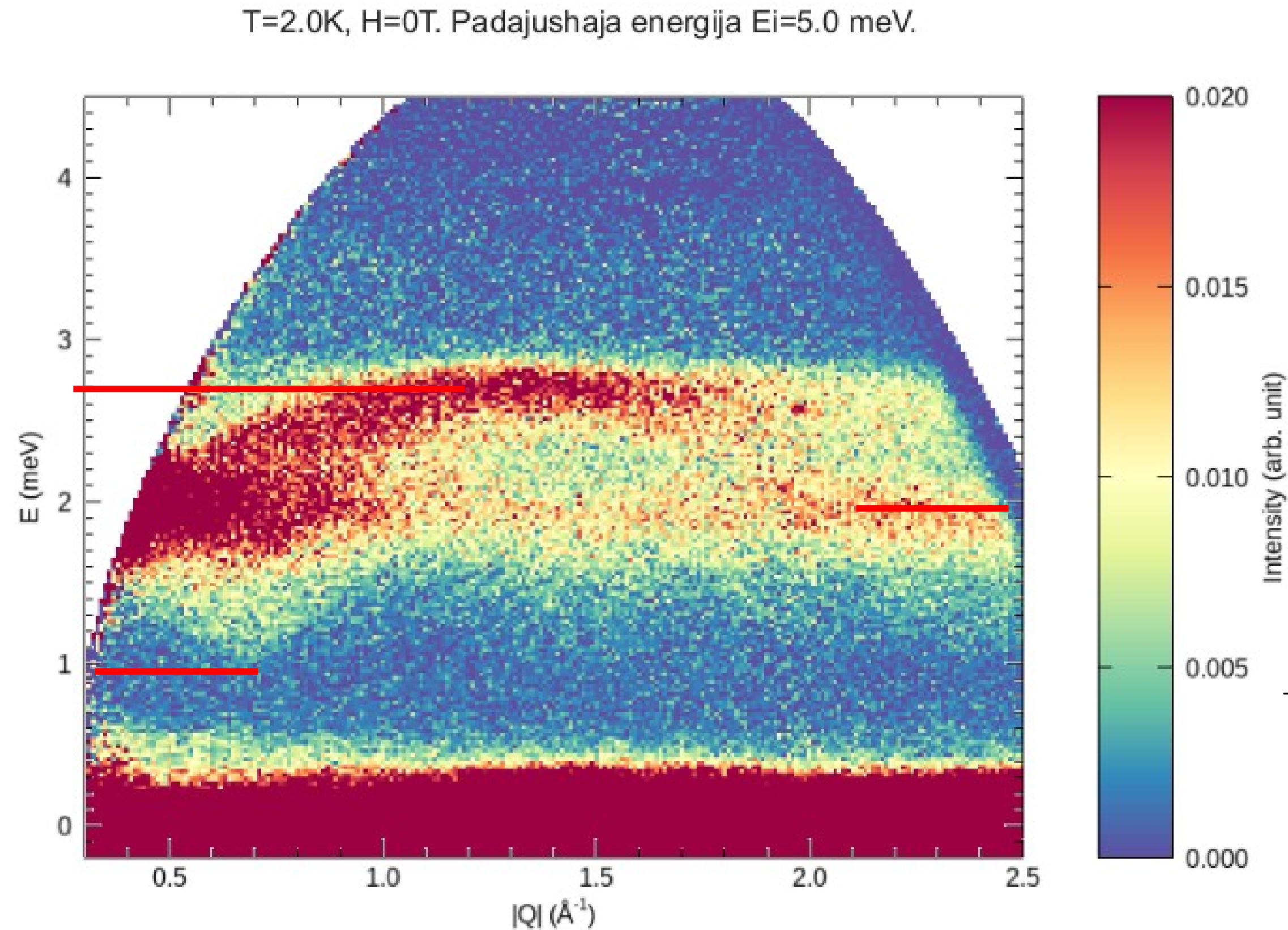


$$H = \sum_{\langle ij \rangle \gamma} J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma \left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha \right) + \Gamma' \left(S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma \right)$$

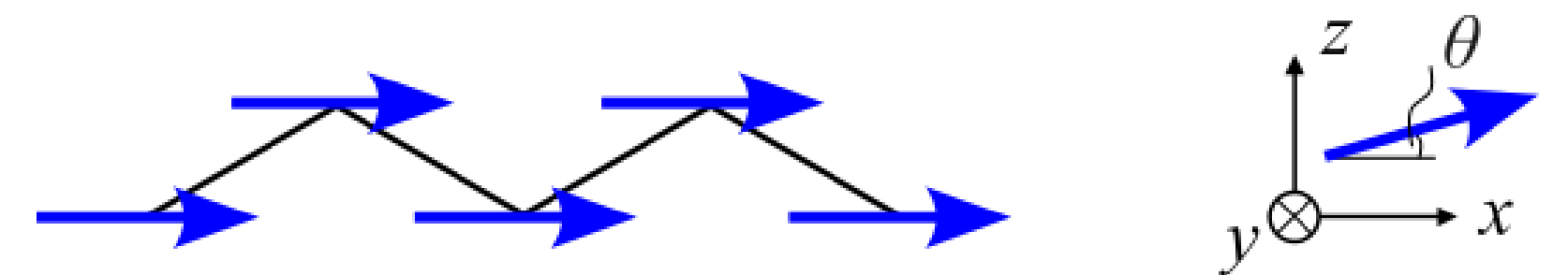


Only x and y bonds

Fit criteria



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

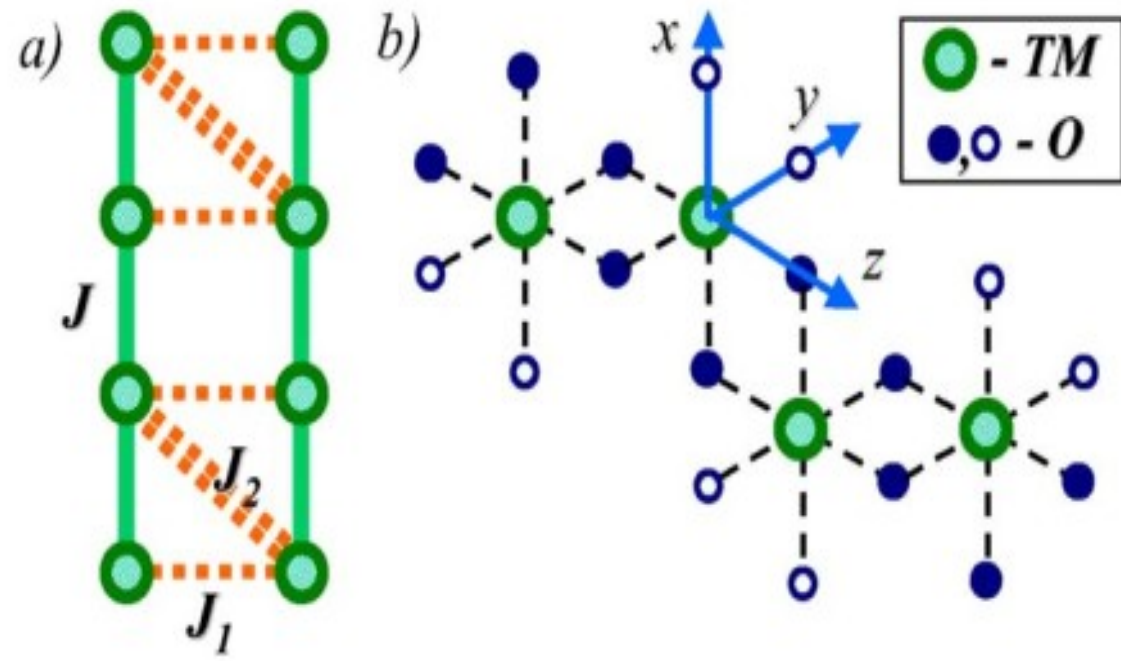


$$\tan 2\alpha = 4\sqrt{2} \frac{1+r}{7r-2}, \quad r = -\frac{\Gamma}{K + \Gamma'}$$

Four criteria (can be obtained analytically)

- $\varepsilon_1(0)=1.0$ meV
- $\varepsilon_1(1/2,1/2,0)=1.9$ meV
- $\varepsilon_2(1/2,1/2,0)=2.7$ meV
- Canting angle $\sim 6^\circ$

Full exchange model

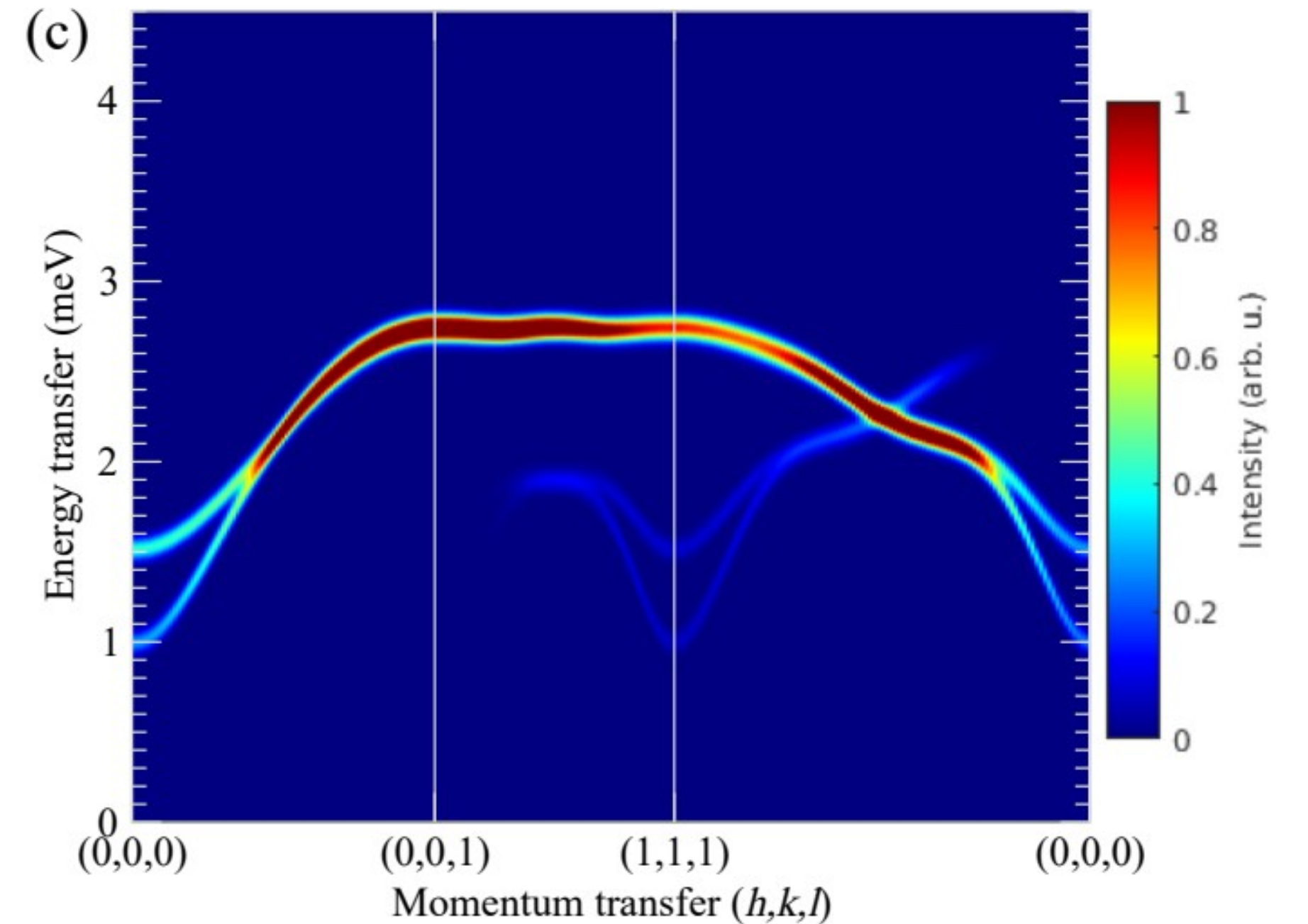
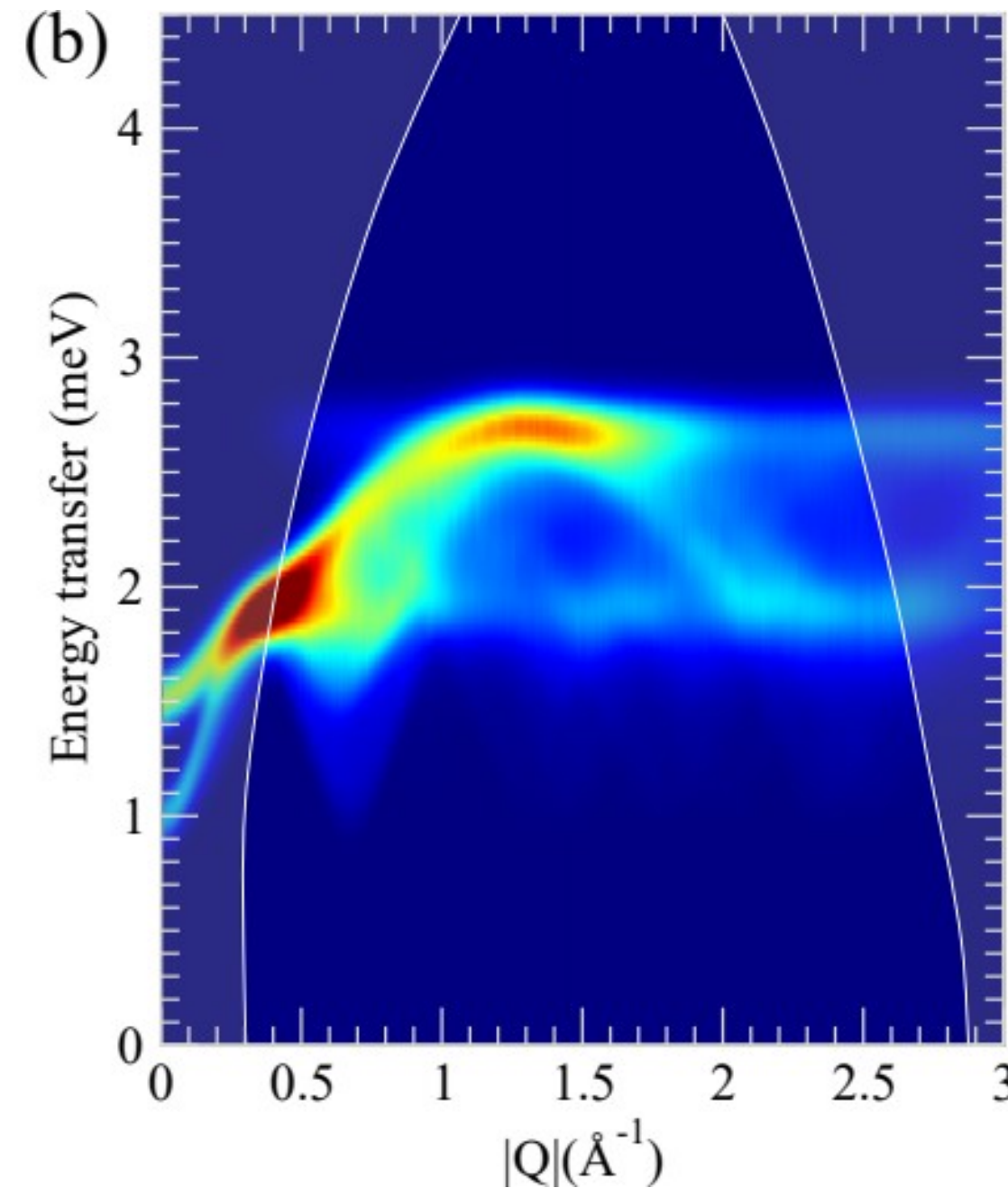
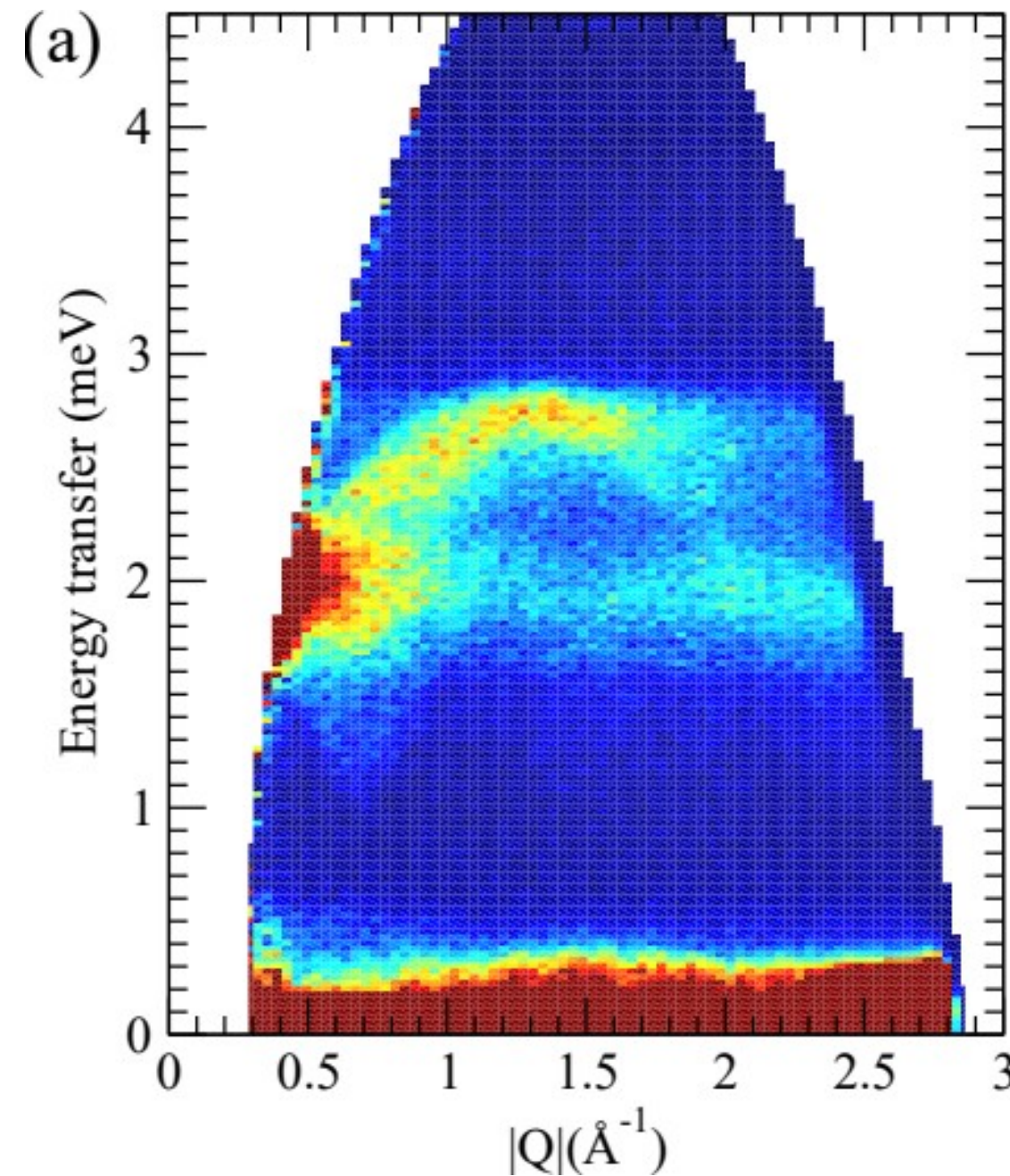


$$H = \sum_{\langle ij \rangle \gamma} J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma' (S_i^\alpha S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\beta S_j^\gamma)$$

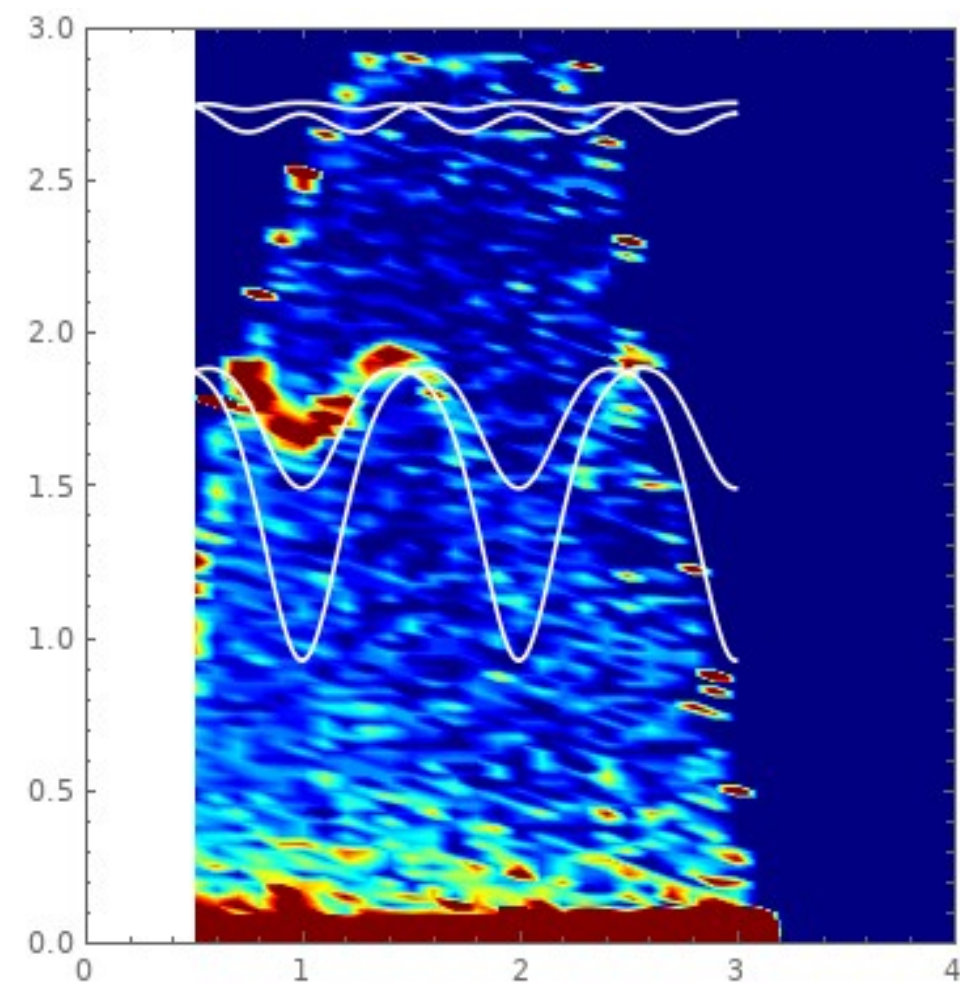
Table 1: Comparison of the exchanges from ab initio (GGA+U+SOC) calculations and neutron scattering fit (LSWT).

Method	J	K	$ K/J $	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

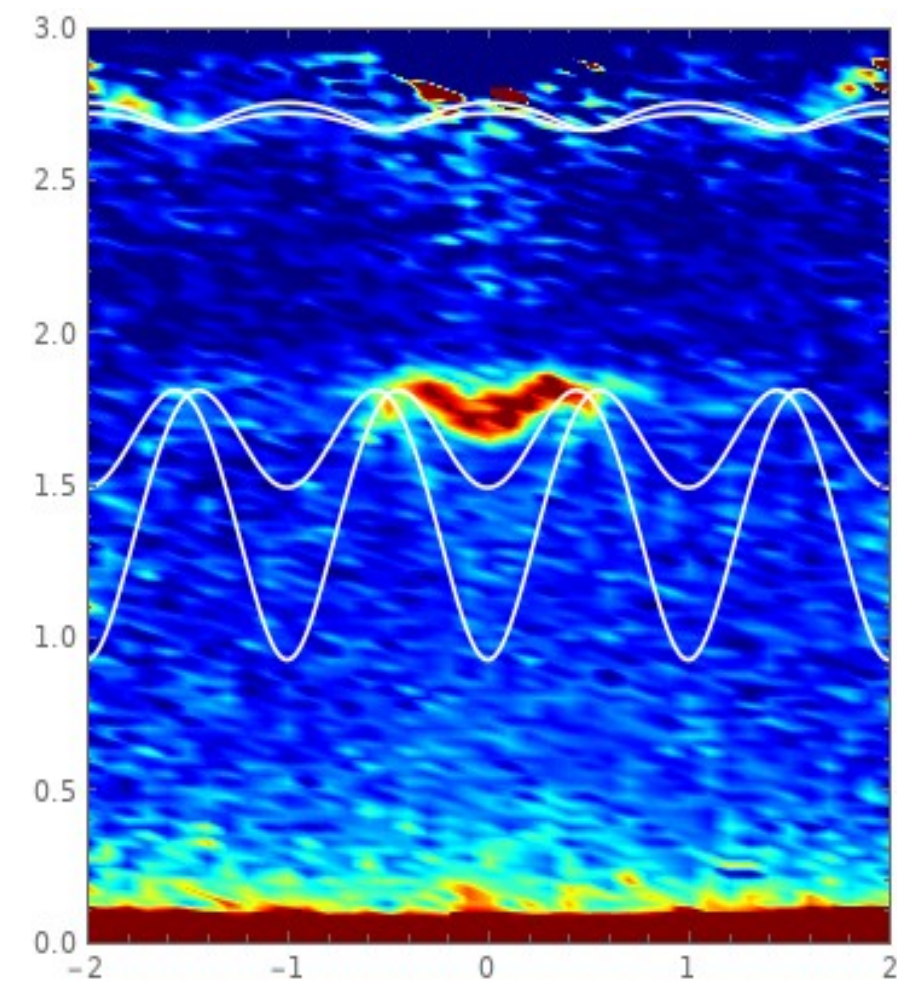
The exchanges are in units of meV.



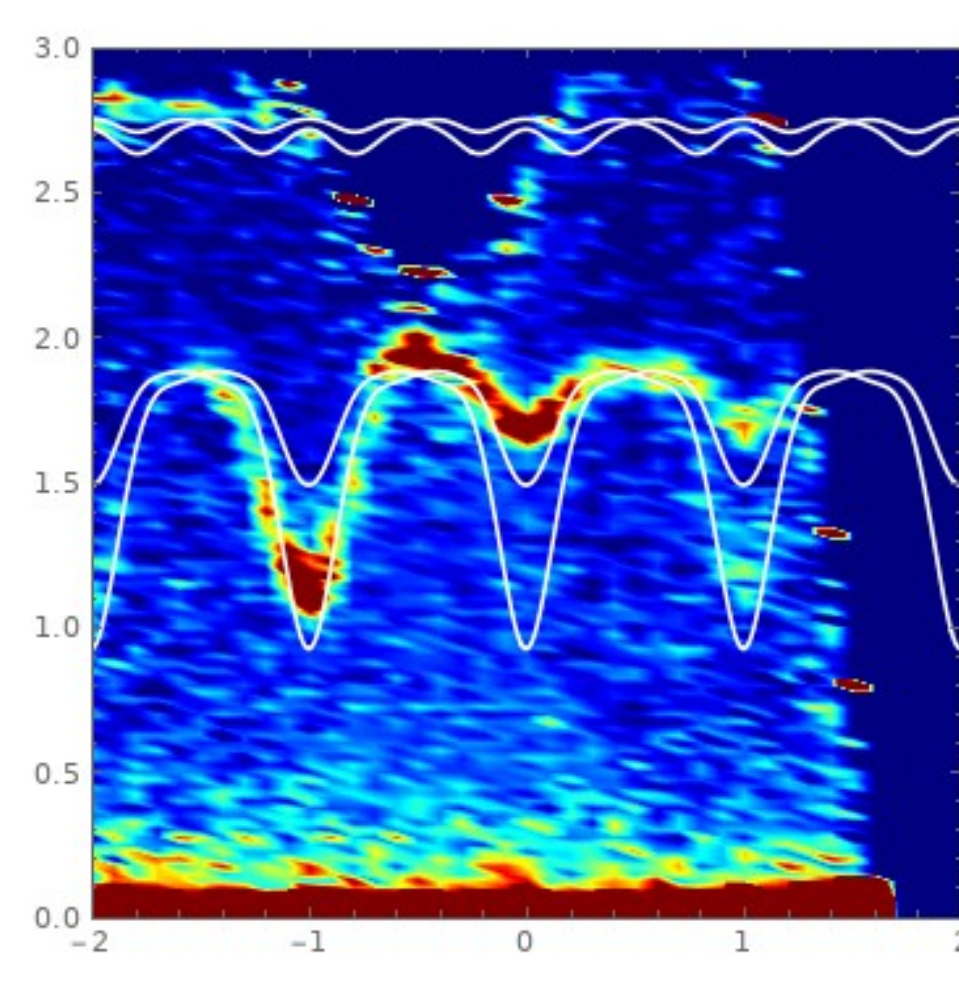
Single crystal neutron scattering



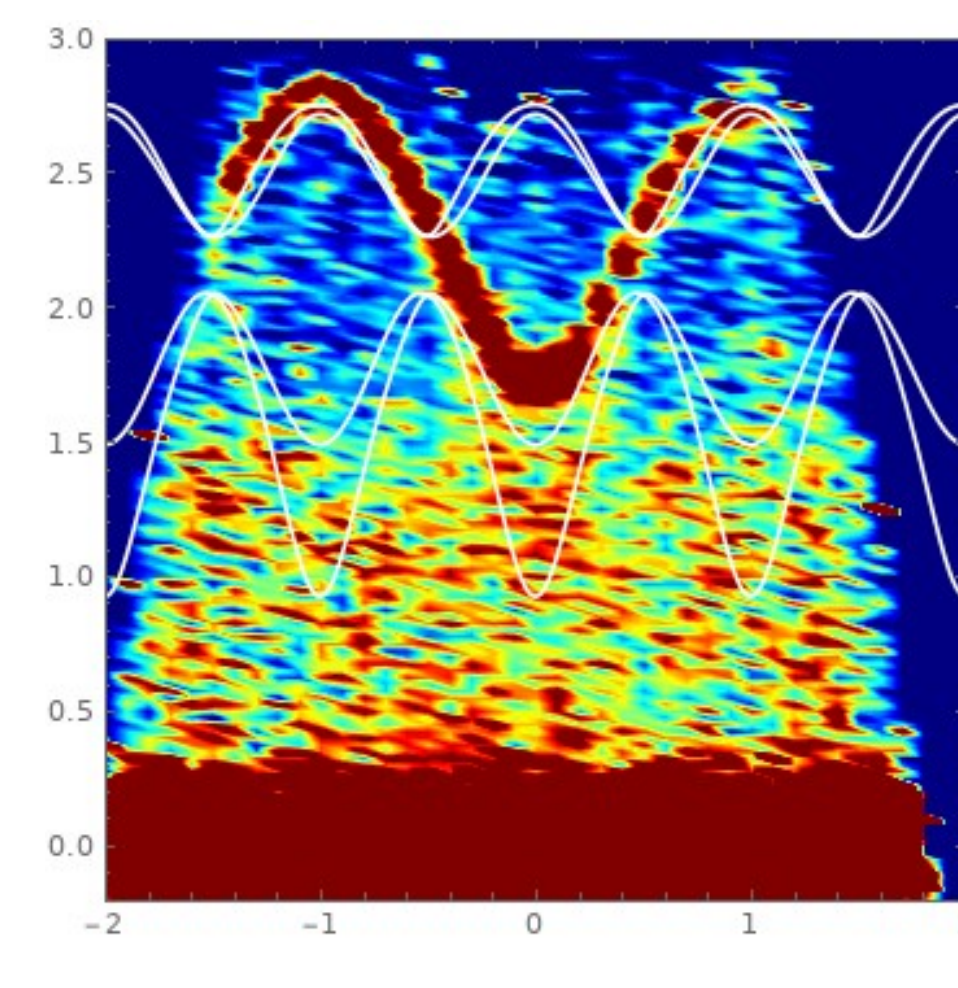
$(h00)$



$(0k0)$

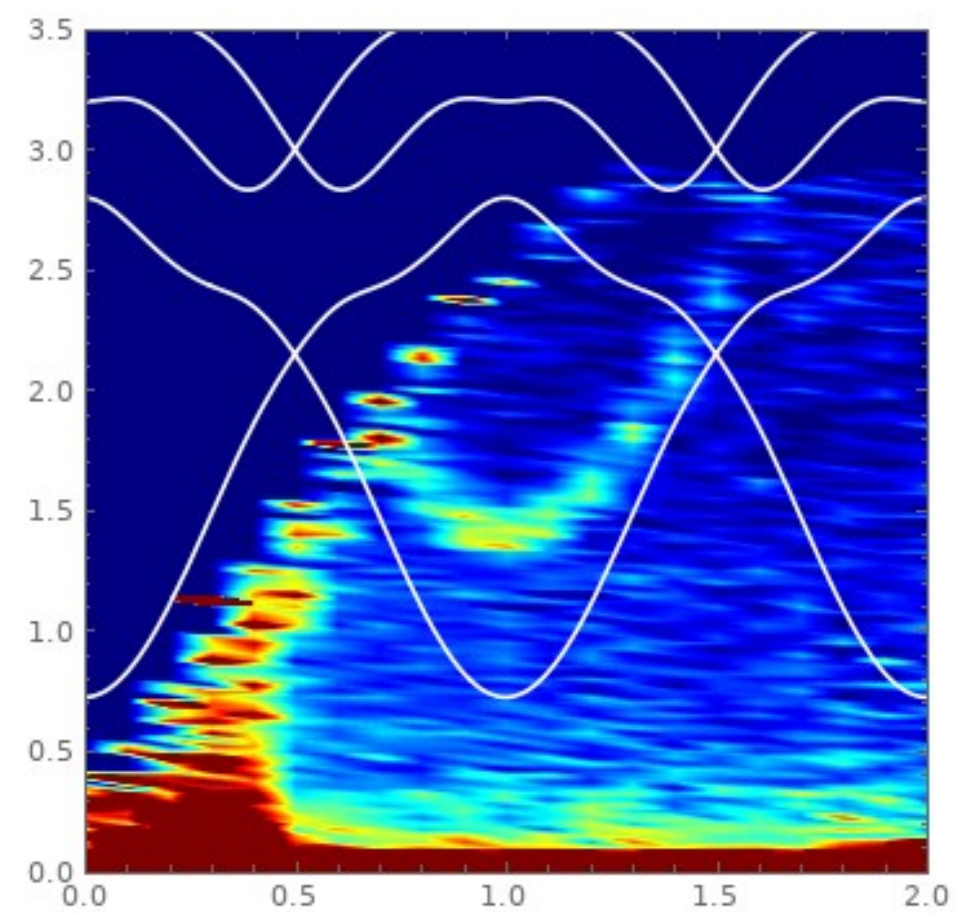


$(hk0)$



$(00l)$

$H=0T$



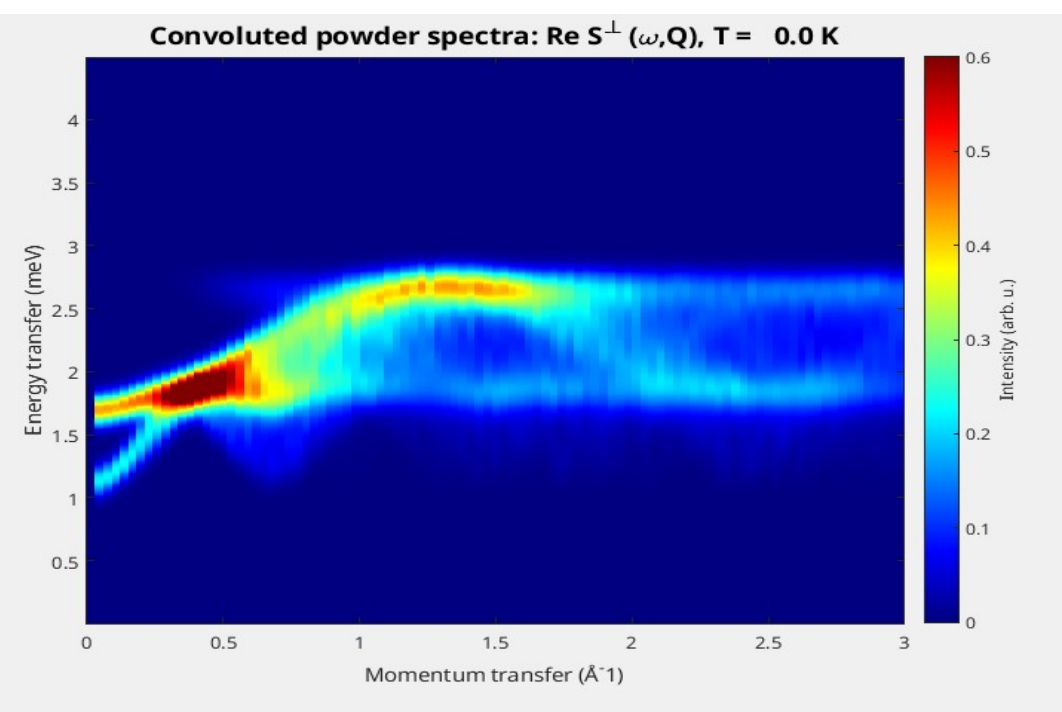
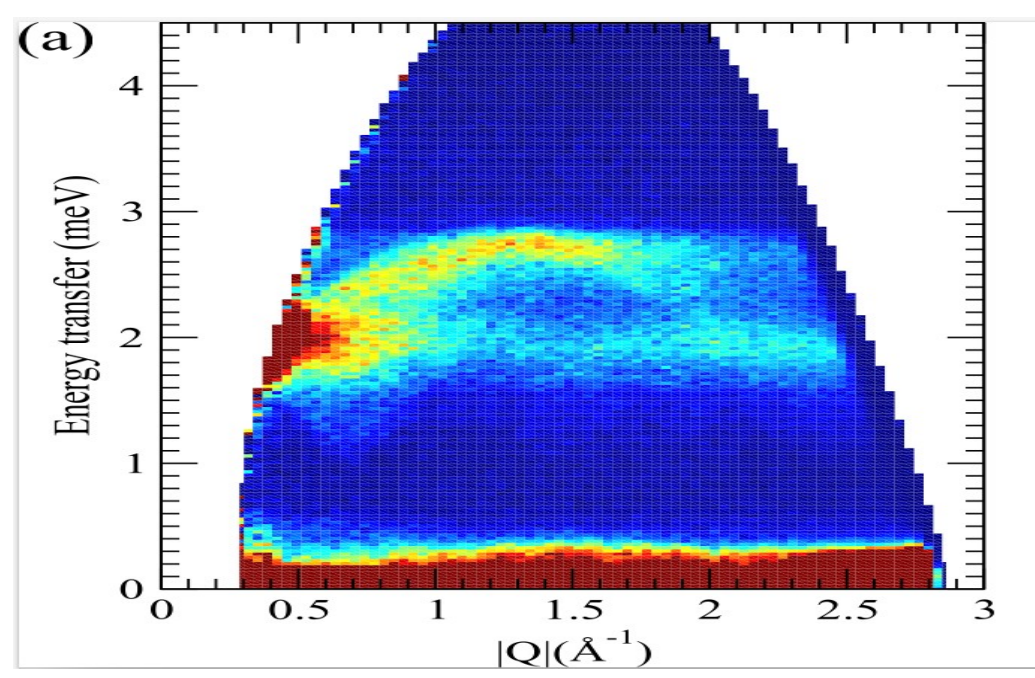
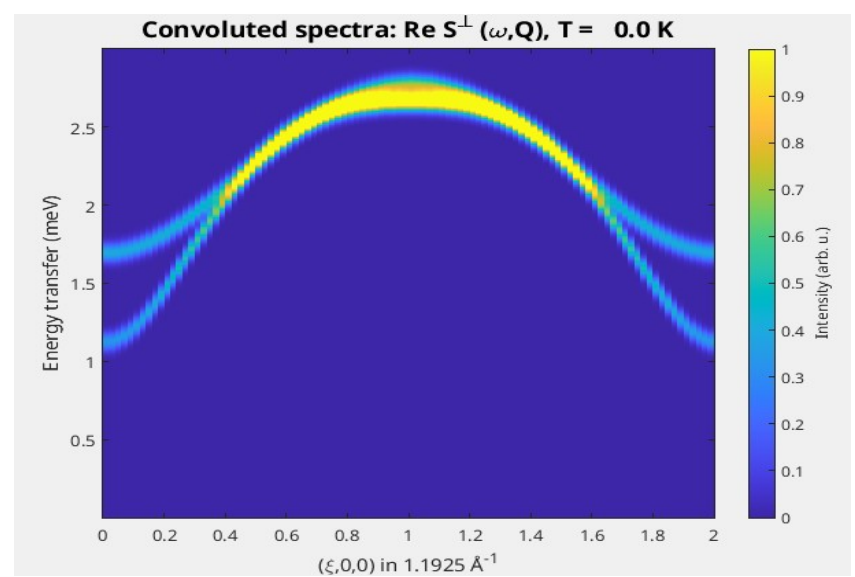
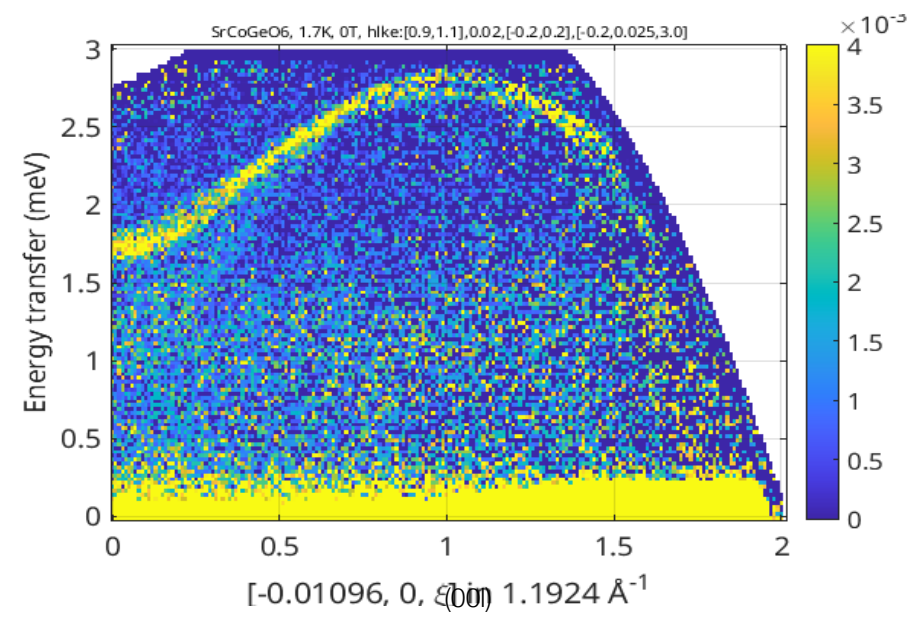
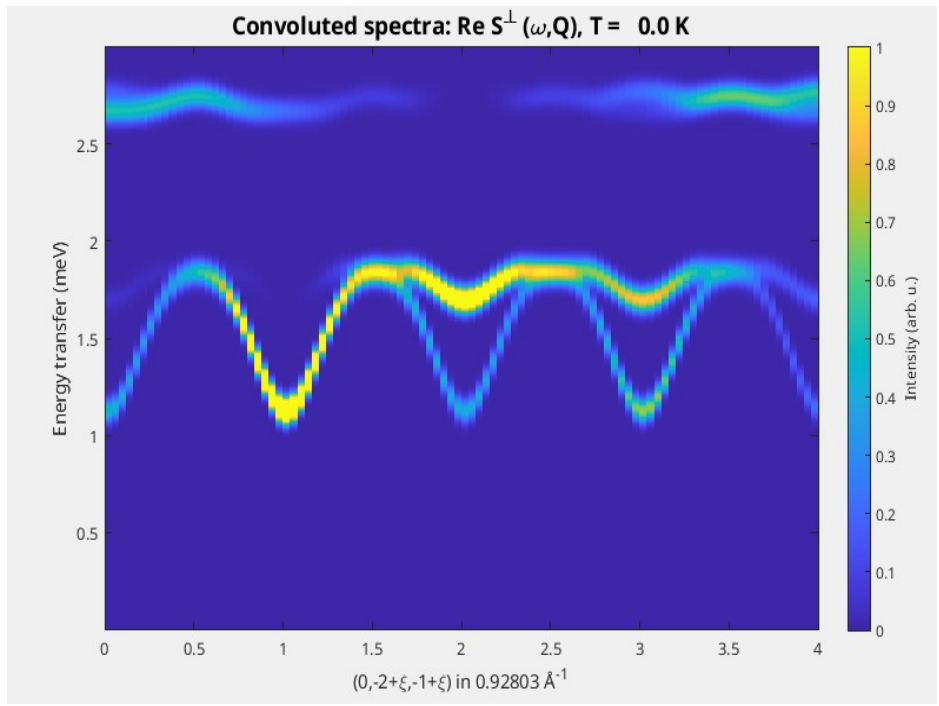
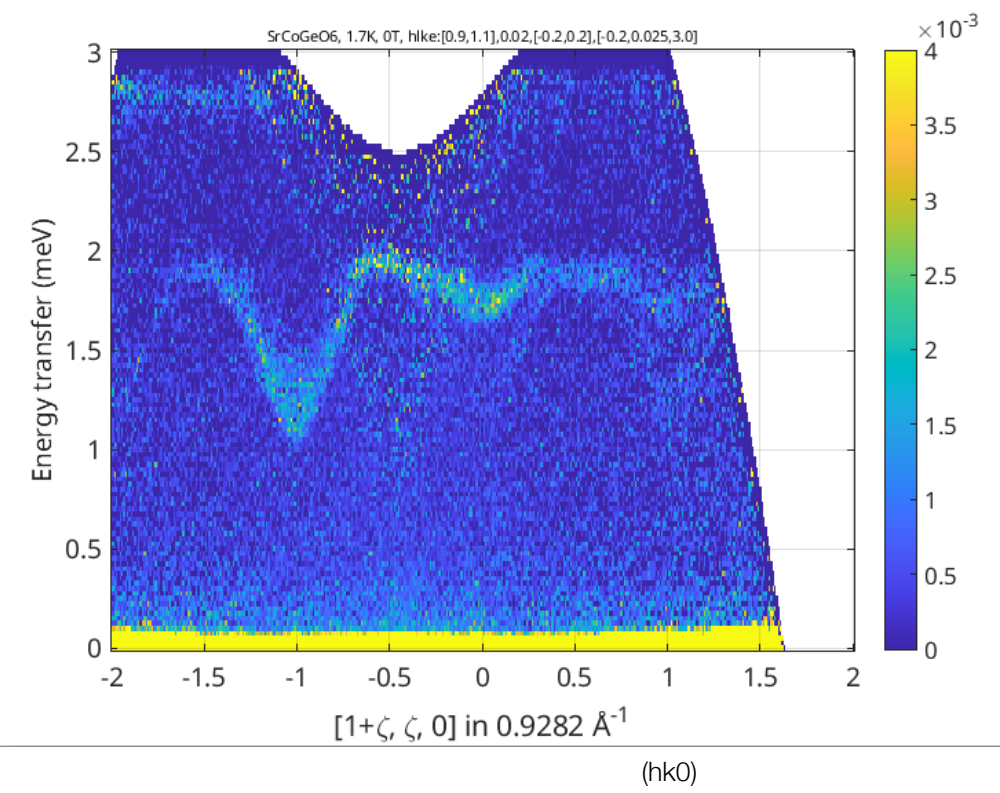
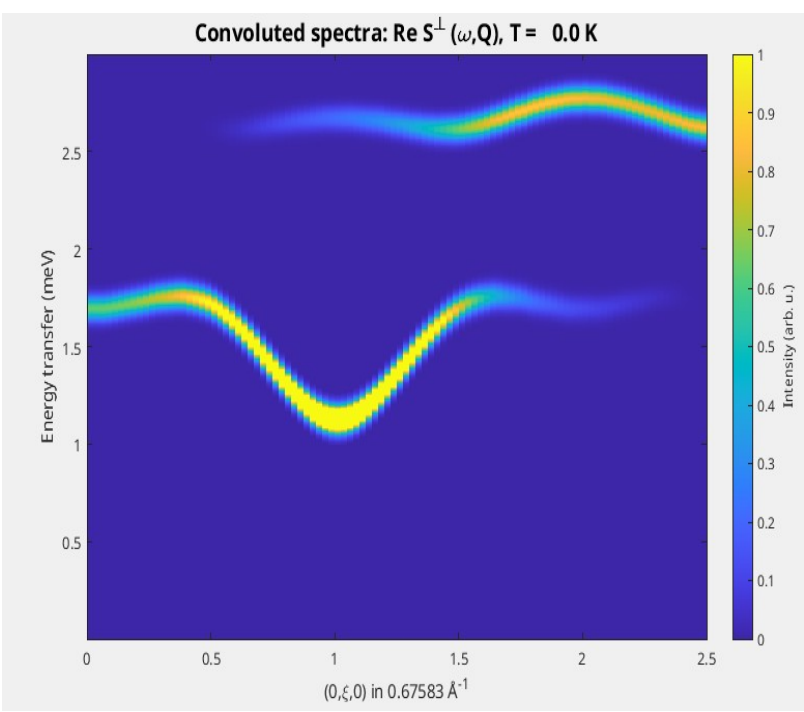
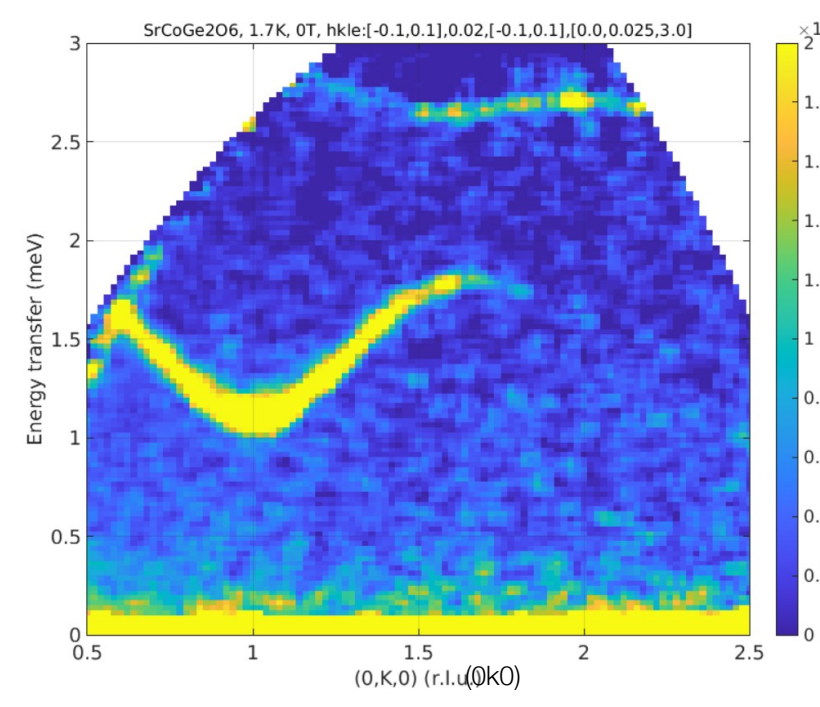
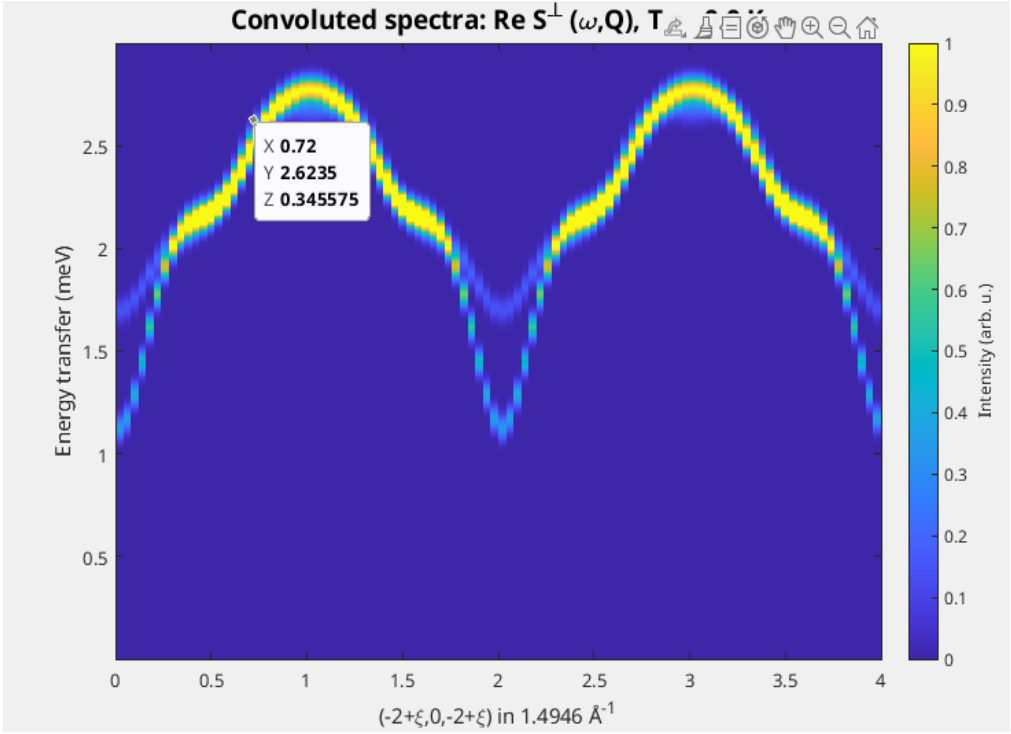
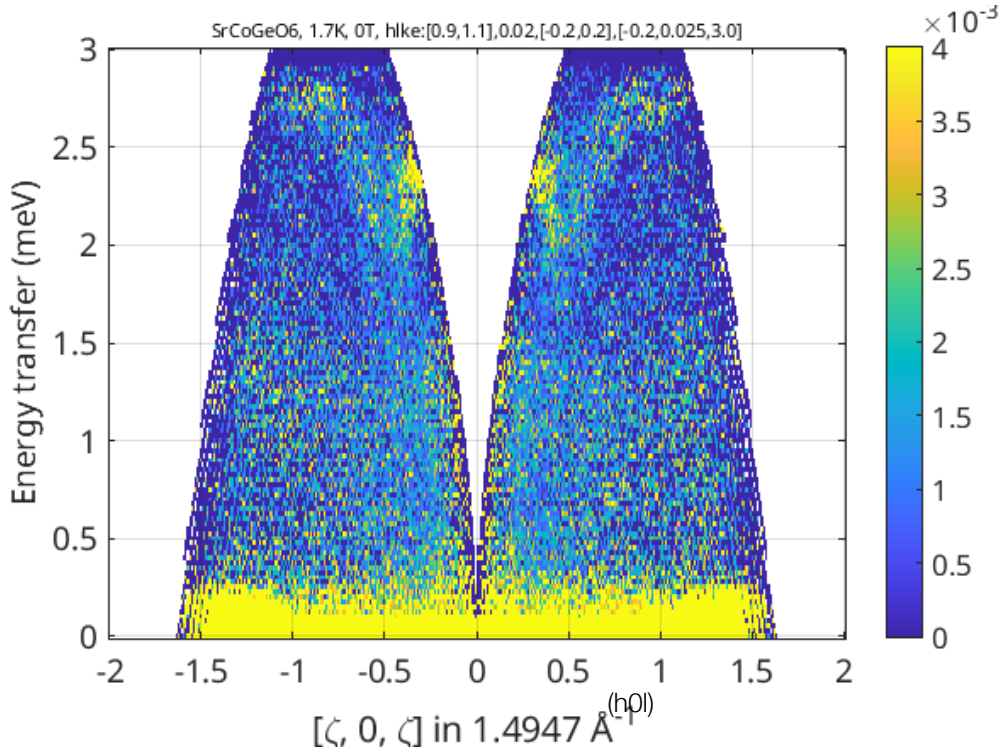
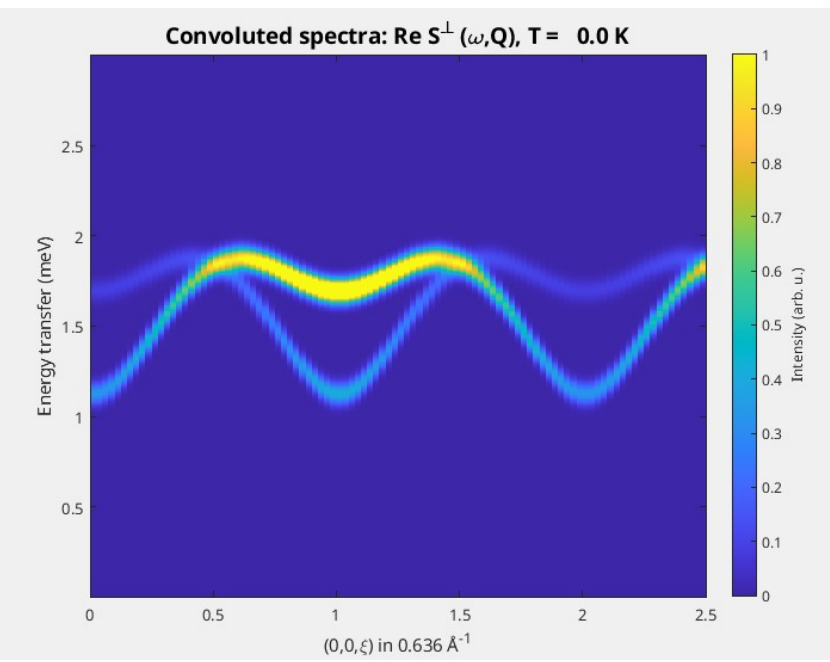
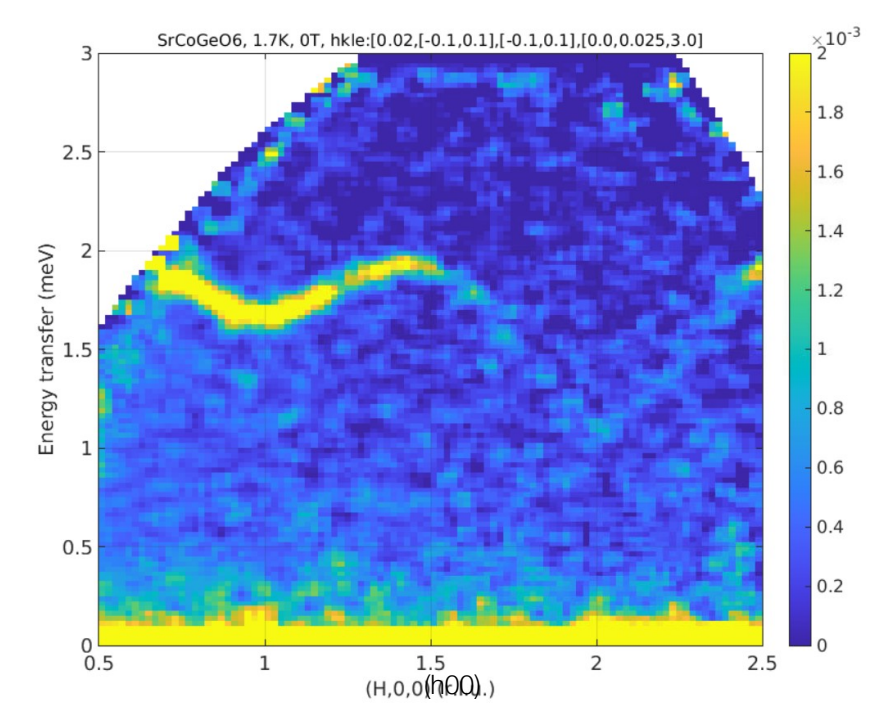
$(h00)$

$H=13.5T, H\parallel c$

Unpublished data

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

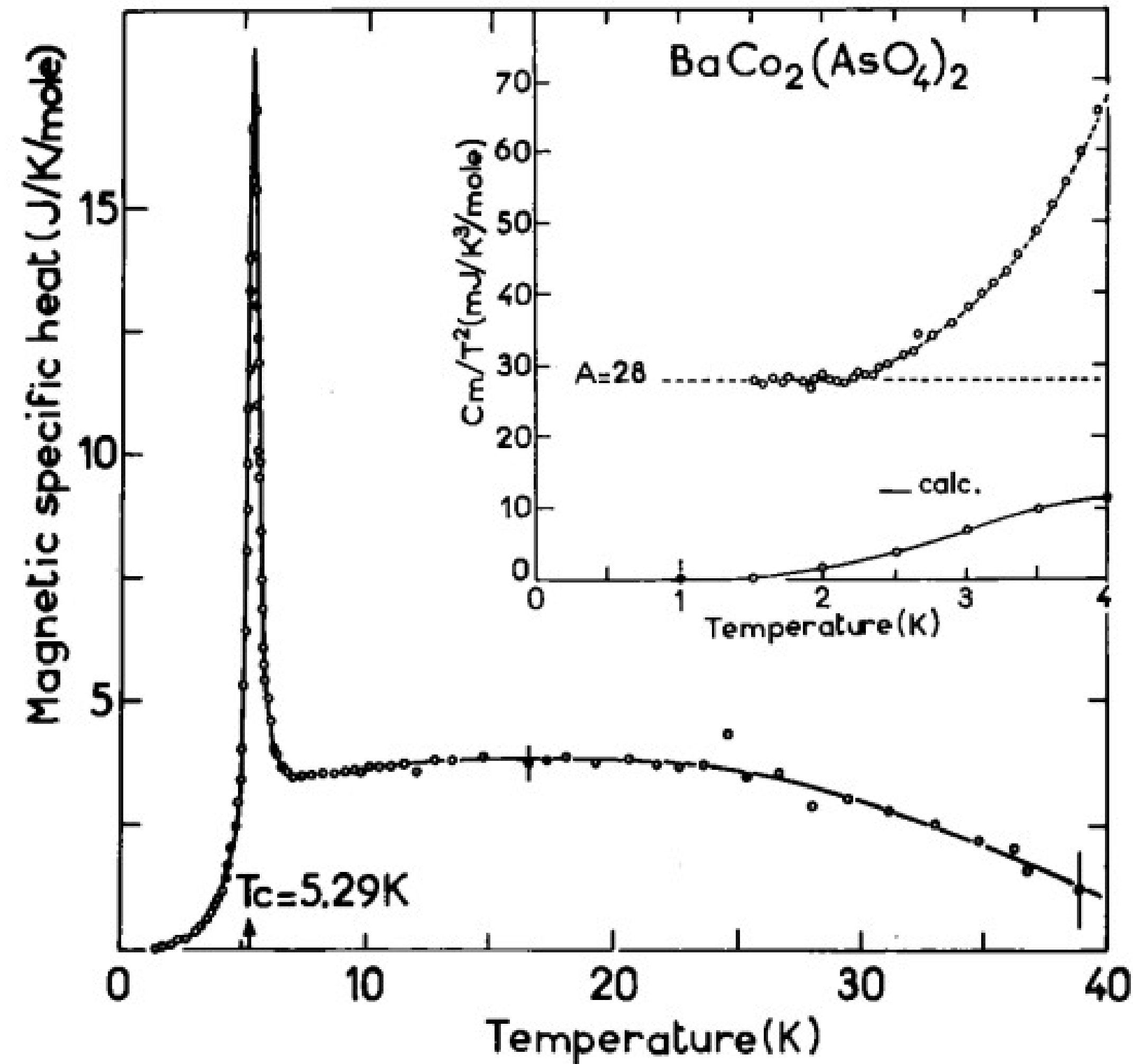
J1=0.2 meV, J2=0.8 meV



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

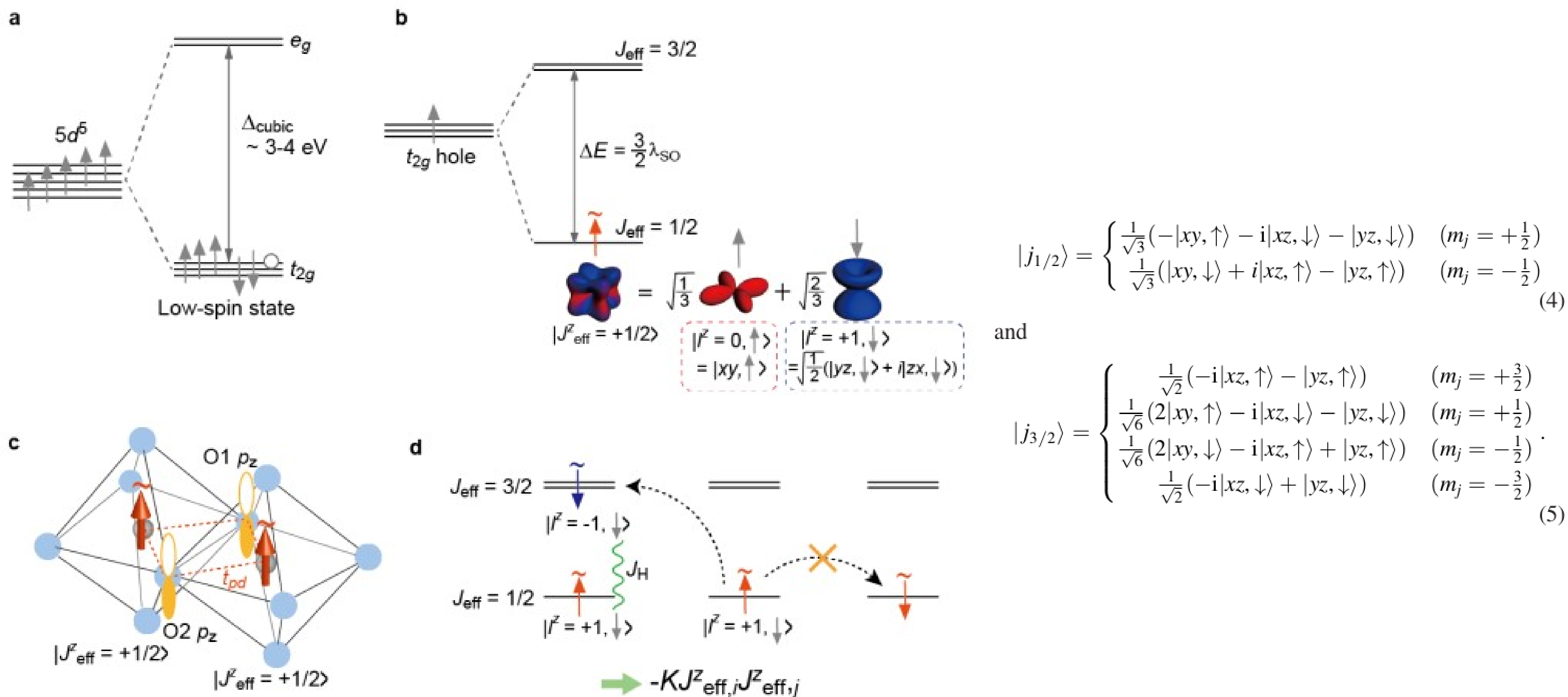
Specific heat



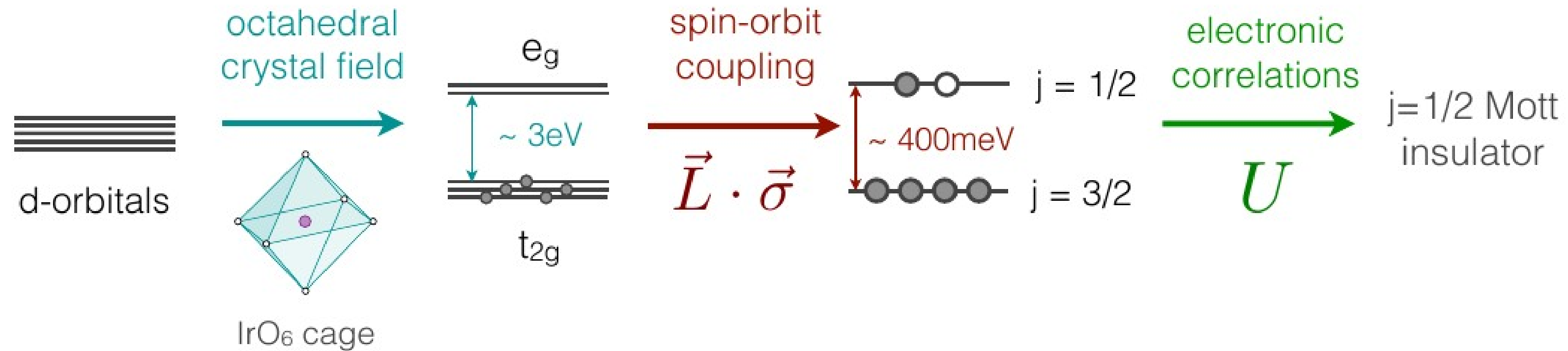
$$C_m - AT^2 = f(T)e^{-\Delta/T}$$

$$\Delta \approx 30\text{K}$$

Kitaev exchange mechanism



Pseudospin: d⁵ ion+SOC



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

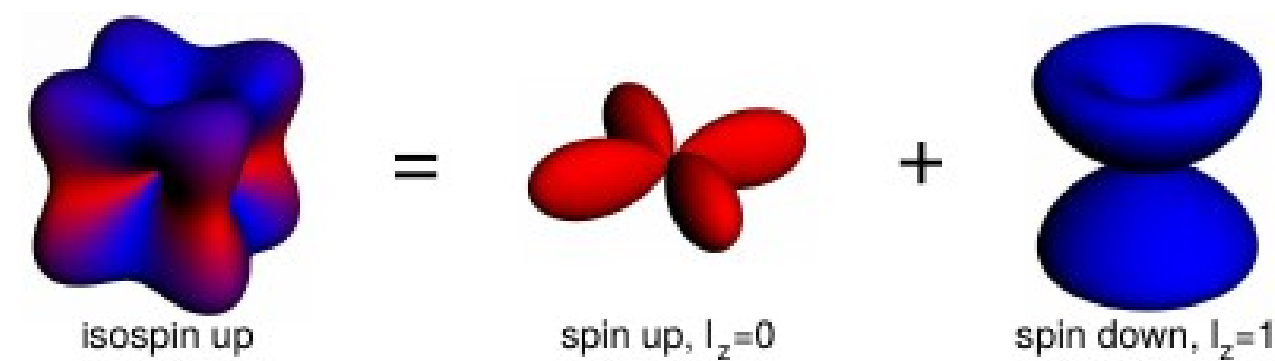
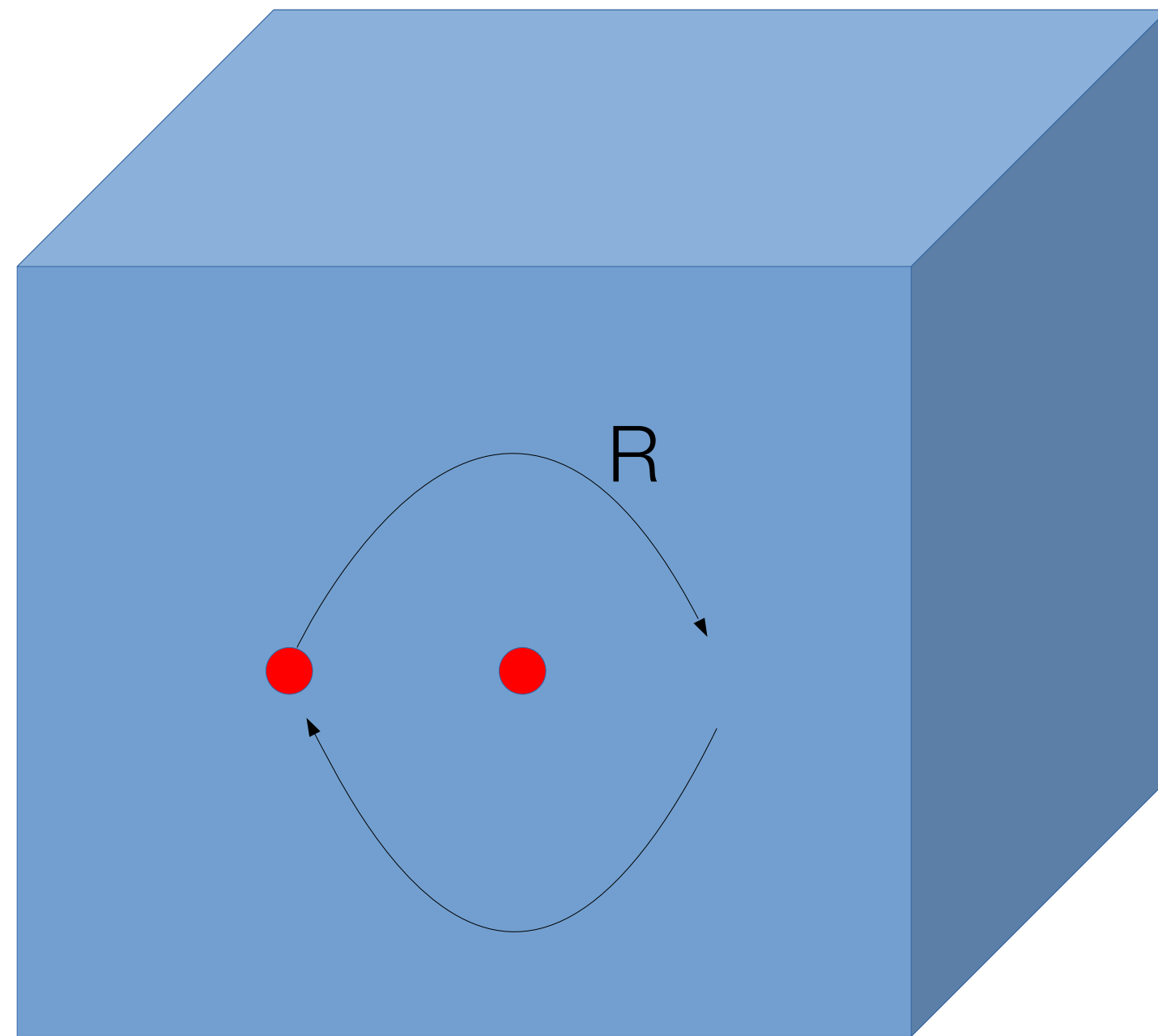
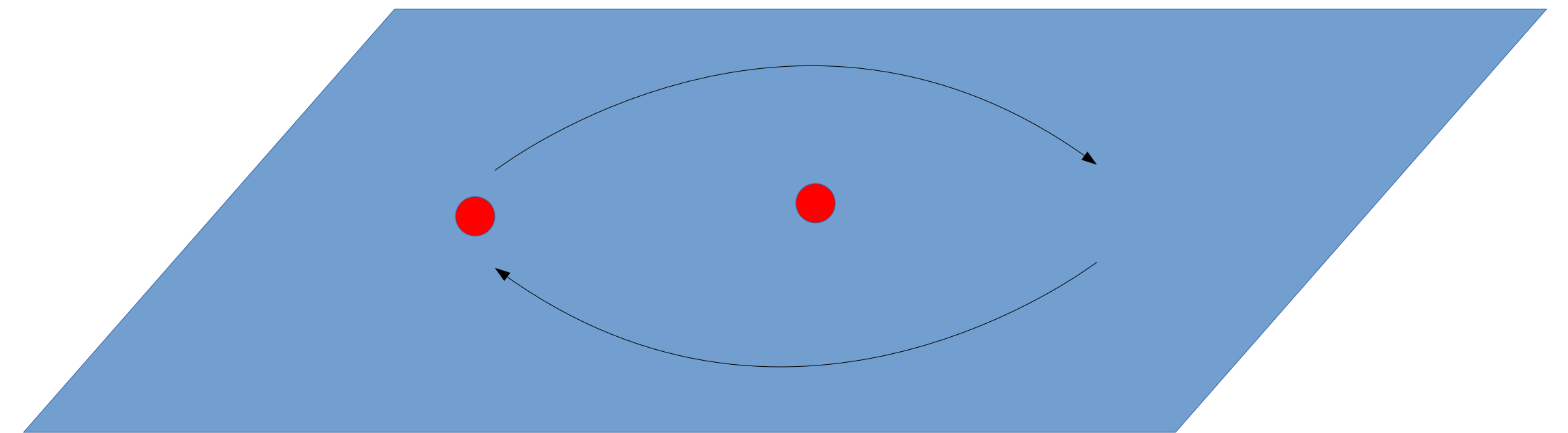


FIG. 1: (Color online) Density profile of a hole in the isospin up state (without tetragonal distortion). It is a superposition of a spin up hole density in $|xy\rangle$ -orbital, $l_z = 0$, (middle) and spin down one in $(|yz\rangle + i|xz\rangle)$ state, $l_z = 1$, (right).

Topological quantum computing

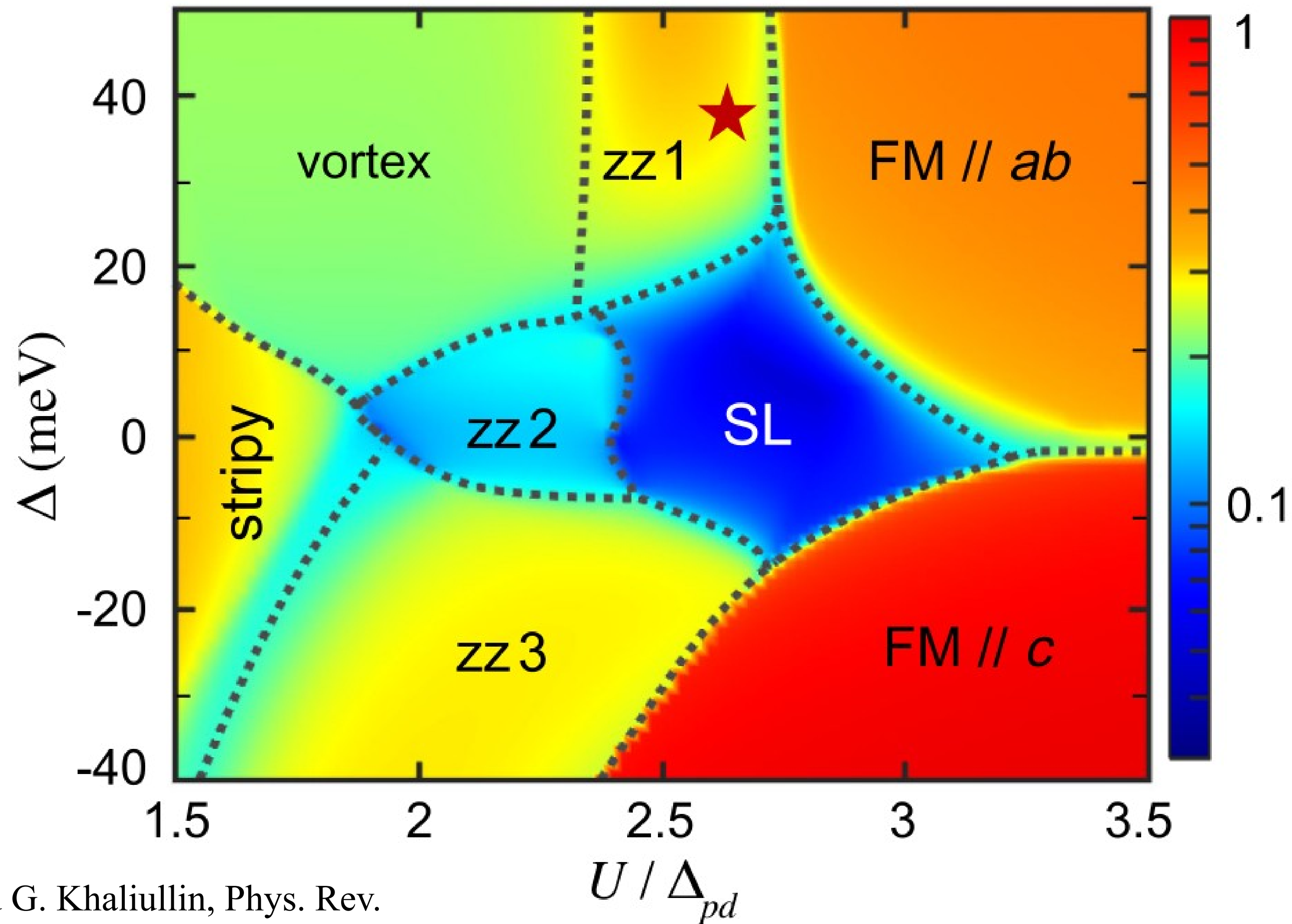


$$R^2 = 1$$
$$R = \pm 1$$

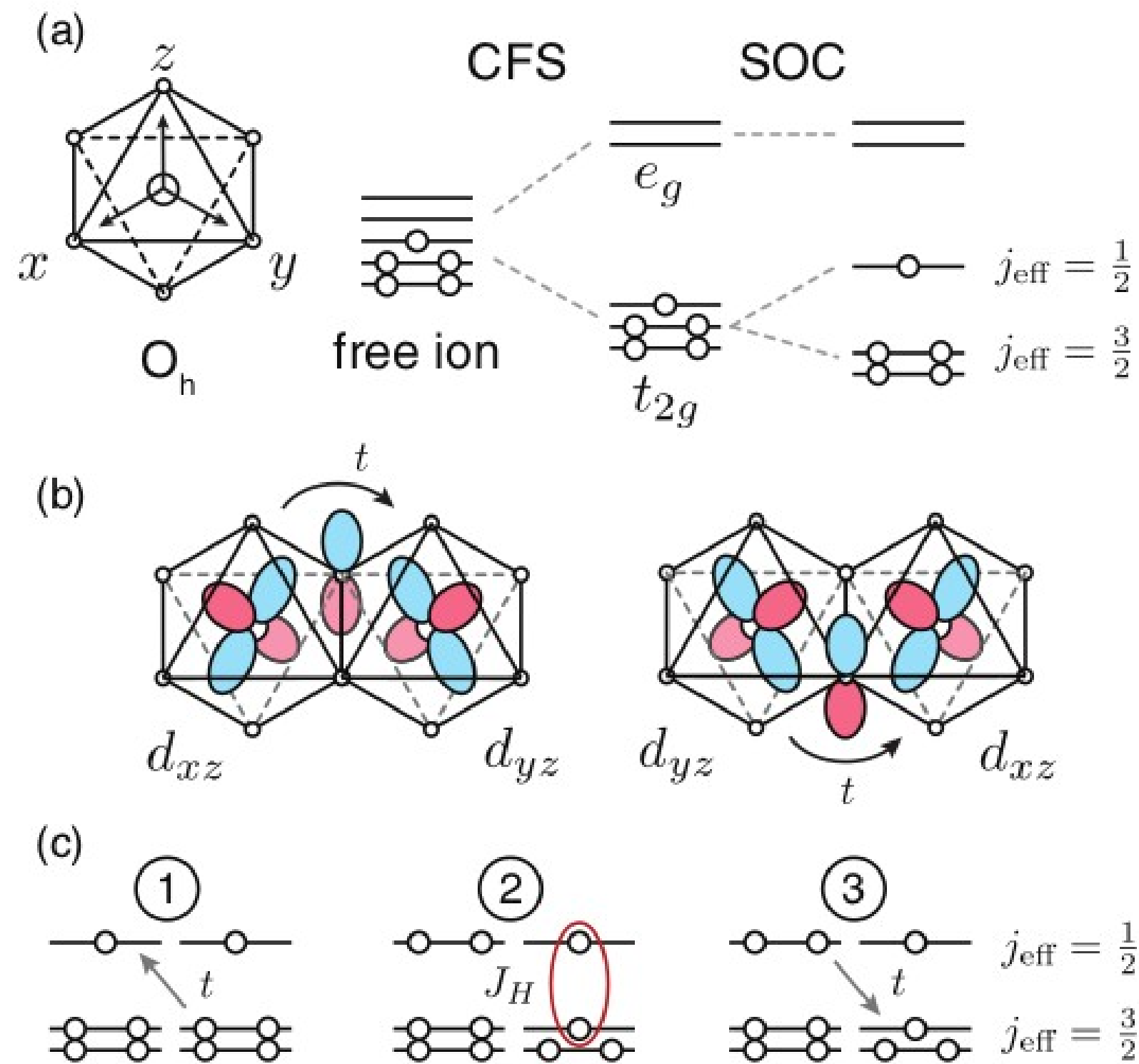


$$R = e^{i\theta}$$

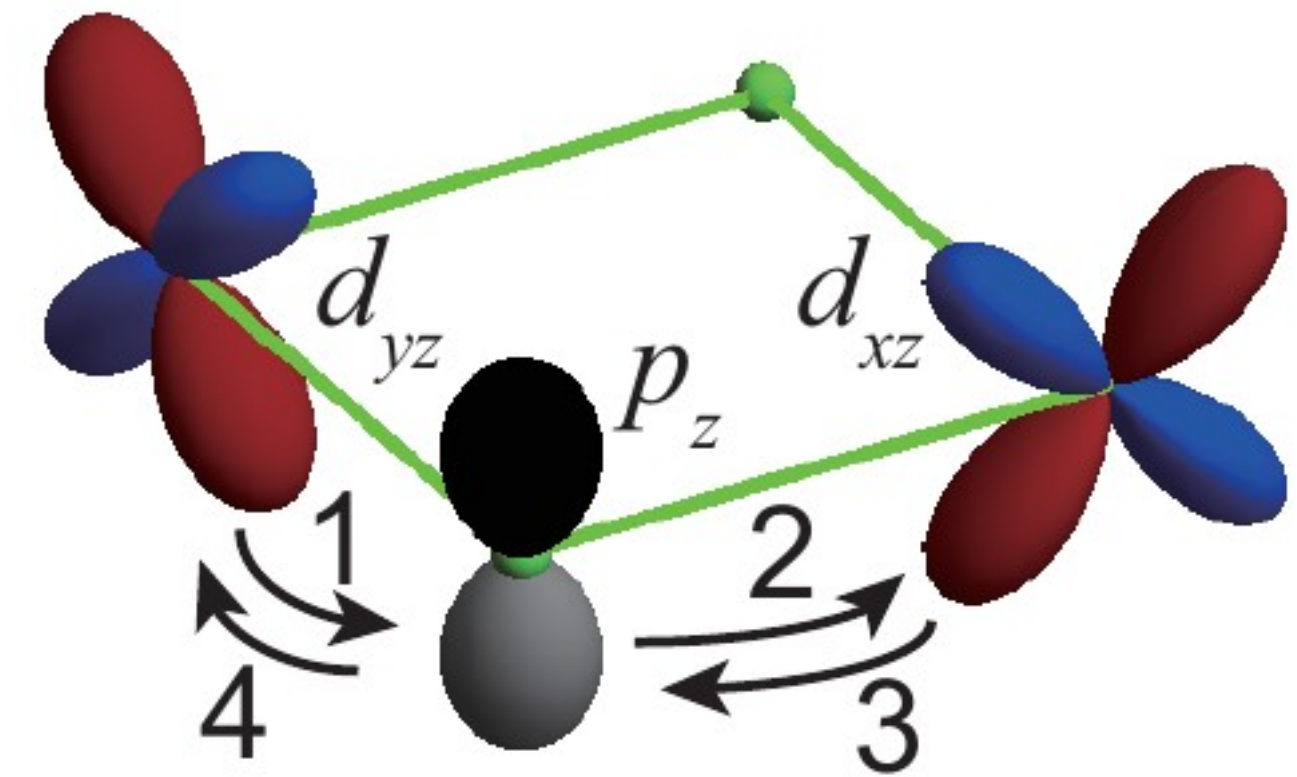
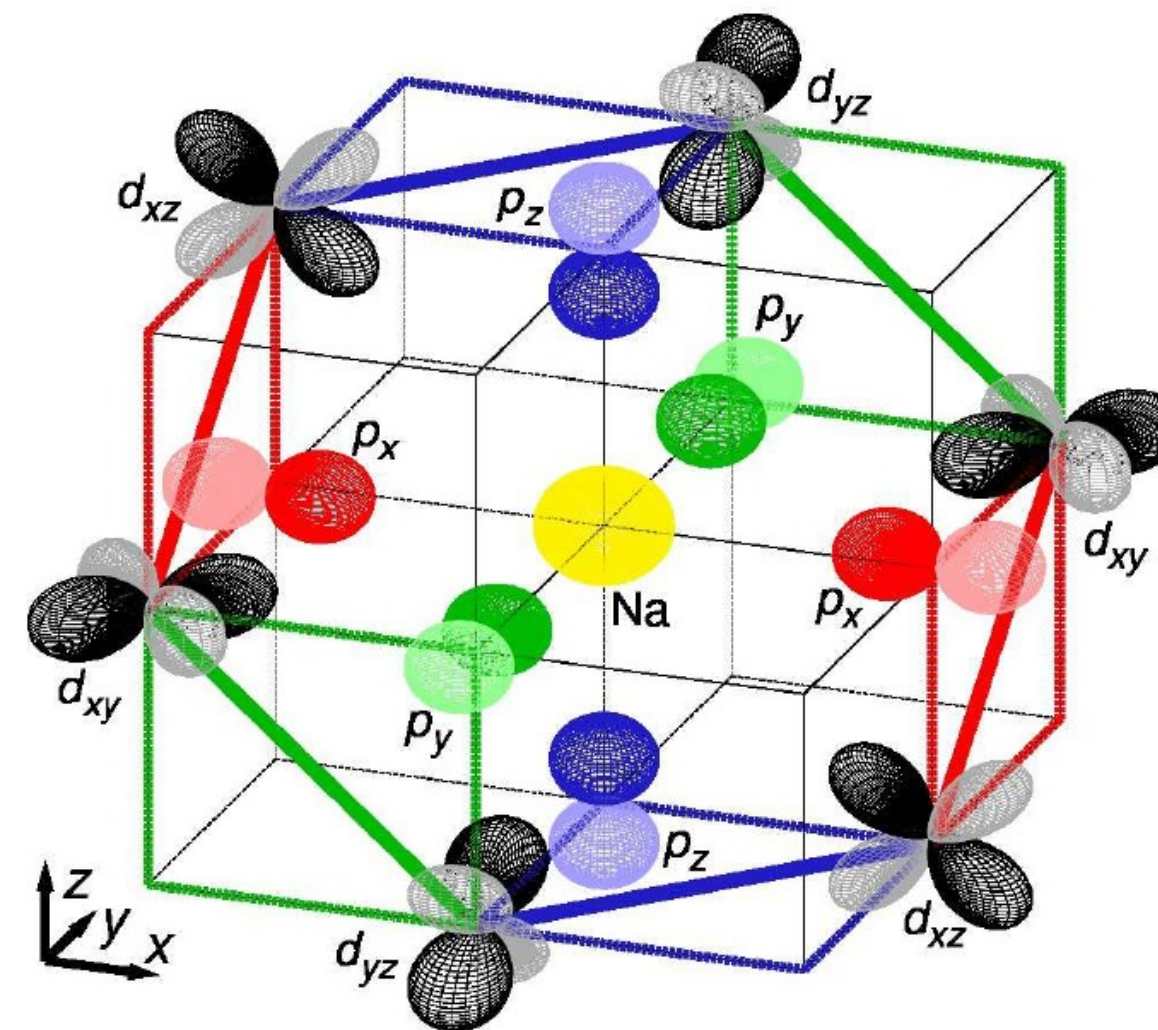
Phase diagram of Co^{2+} honeycomb



Kitaev materials: d^5 ions



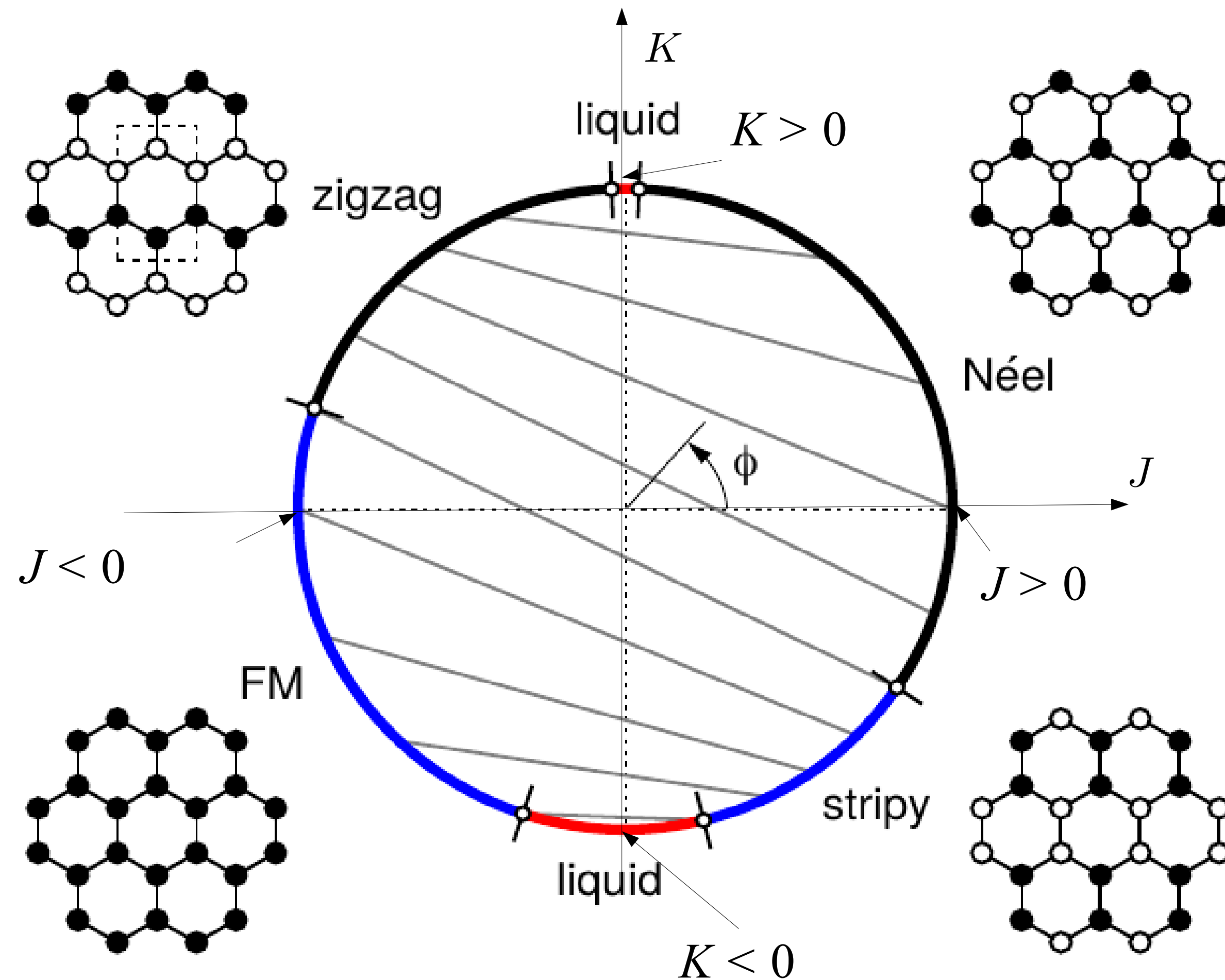
$$K \sim t^2 J_H / U^2$$



Взаимодействует только
z-компонента псевдоспина

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

Фазовая диаграмма модели Китаева-Гейзенберга

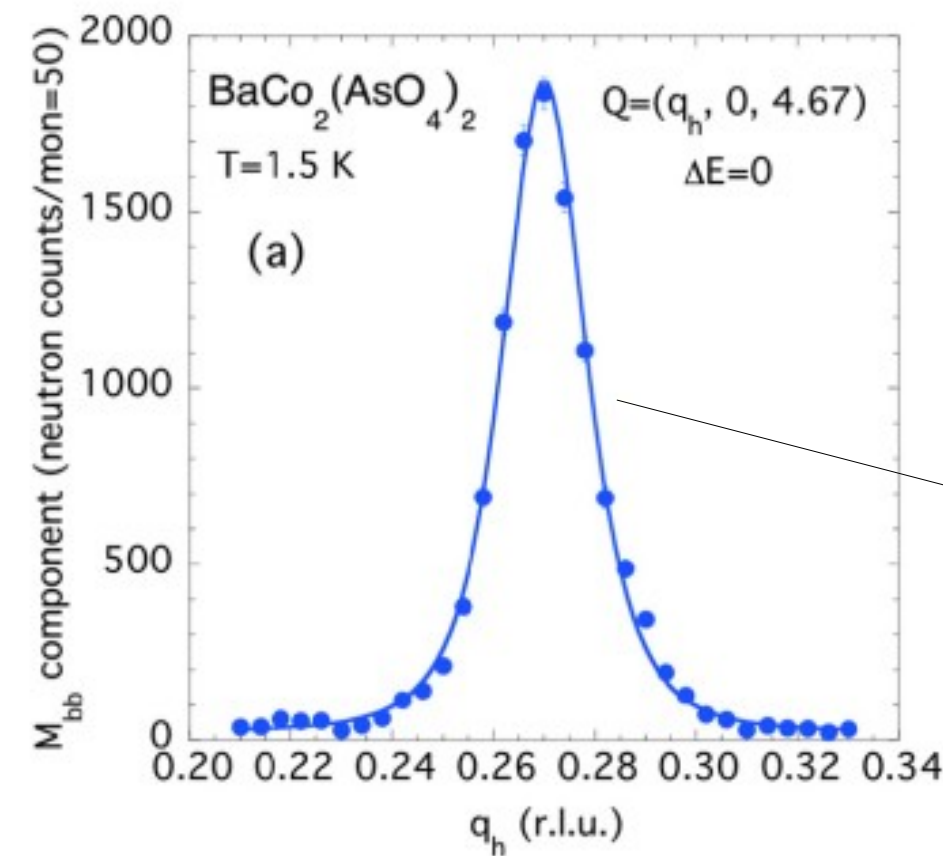


$$\mathcal{H} = \sum_{\langle ij \rangle^\gamma} J \mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma$$

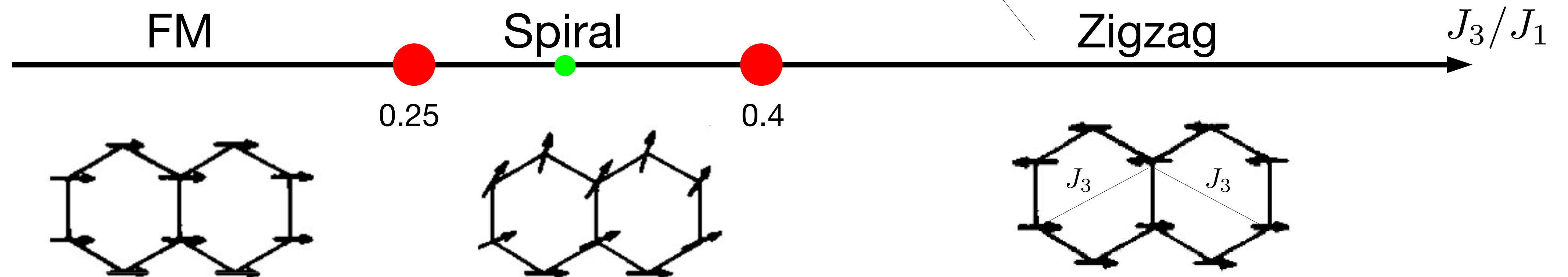
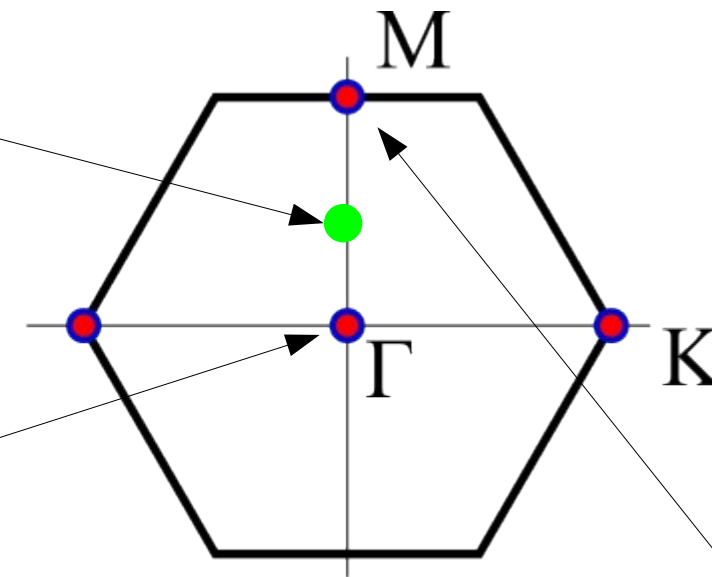
$$J = \cos \phi, \quad K = \sin \phi$$

Большая часть фазовой диаграммы — упорядоченные фазы

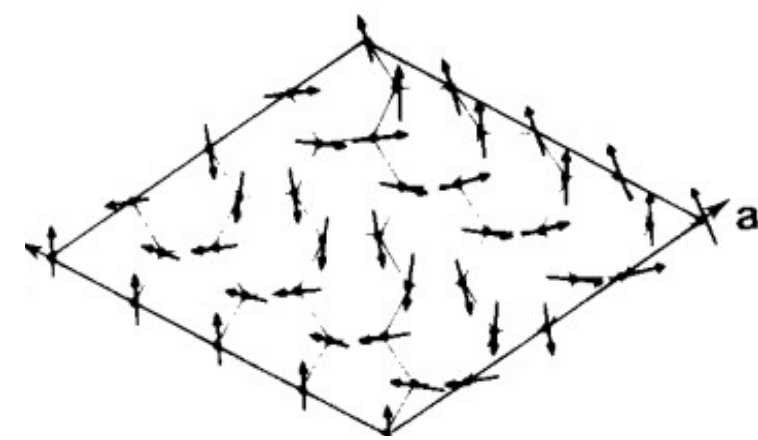
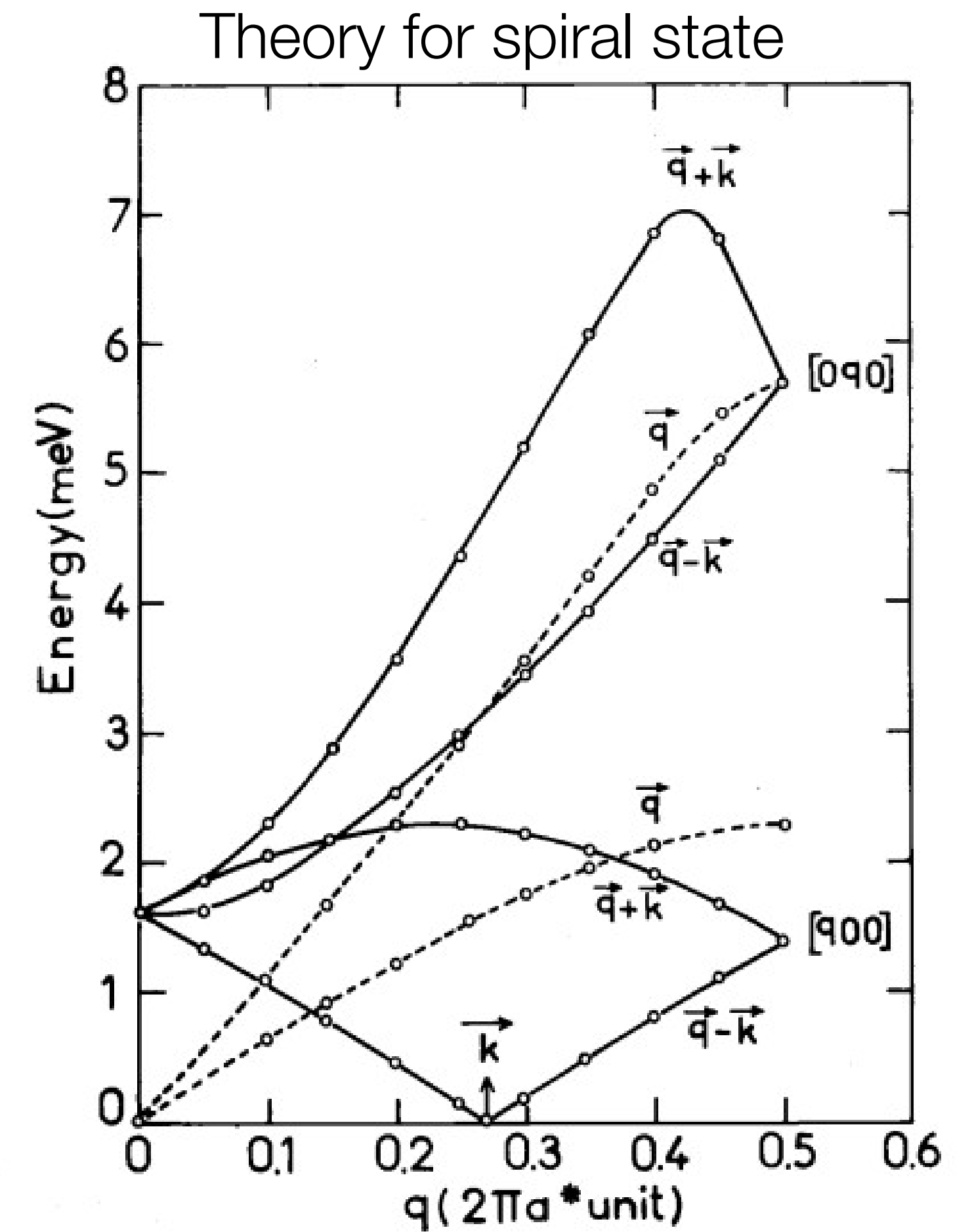
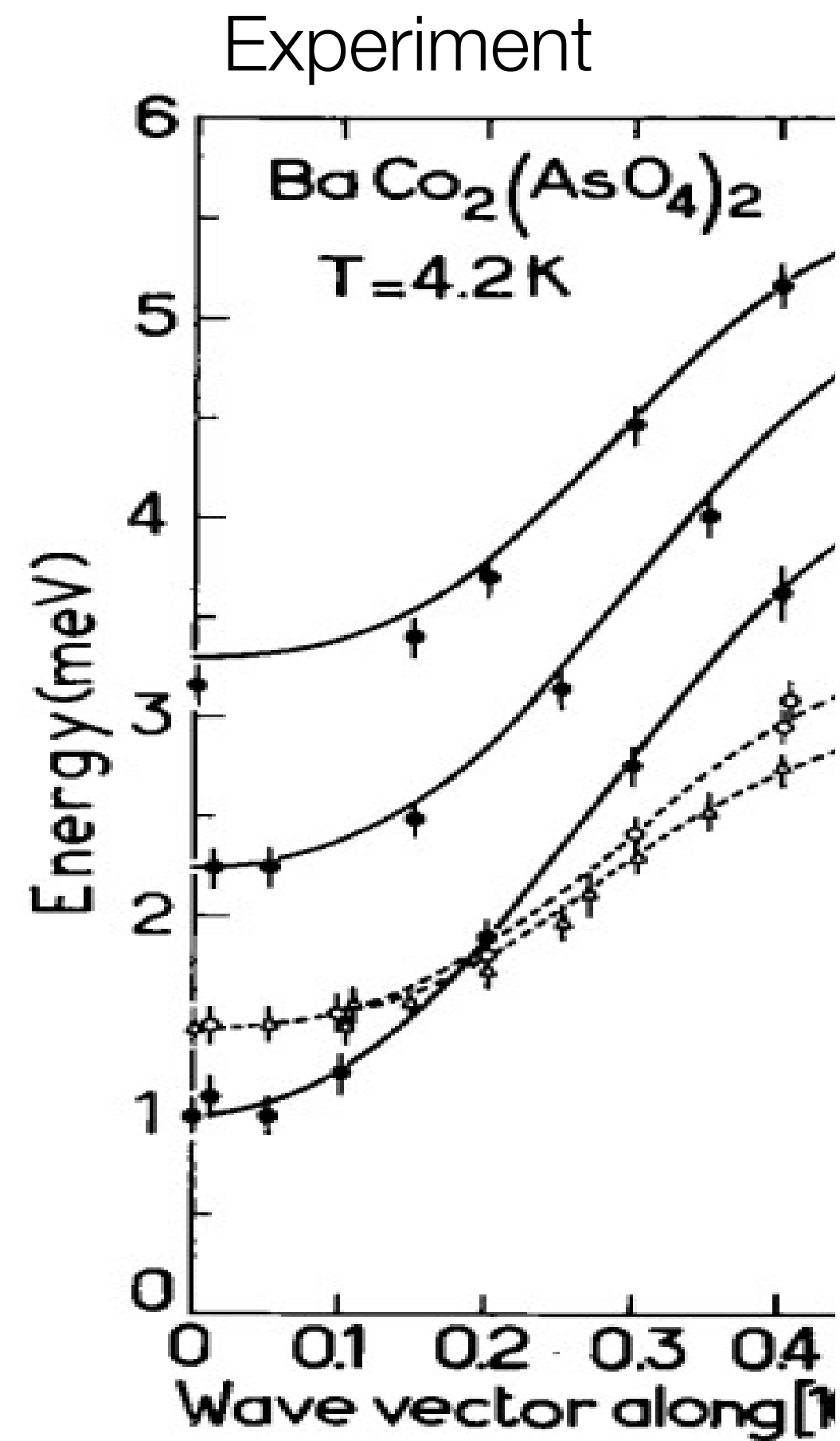
BaCo₂(AsO₄)₂: J_1 - J_3 model



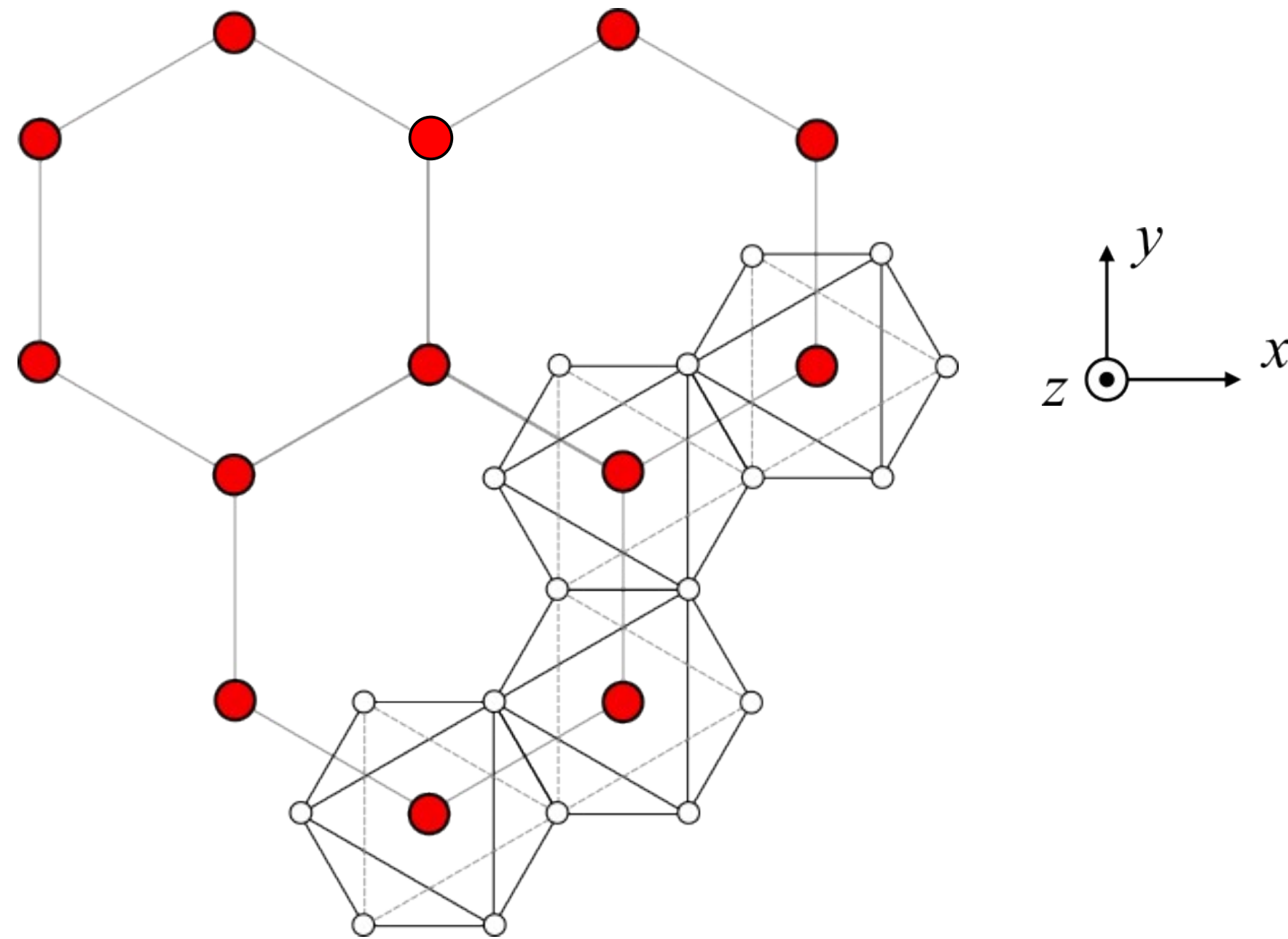
$$\mathcal{H} = -J_1 \sum_{\langle ij \rangle_1} \mathbf{S}_i \cdot \mathbf{S}_j + J_3 \sum_{\langle ij \rangle_3} \mathbf{S}_i \cdot \mathbf{S}_j$$



Magnetic excitations



Эффективный обменный гамильтониан

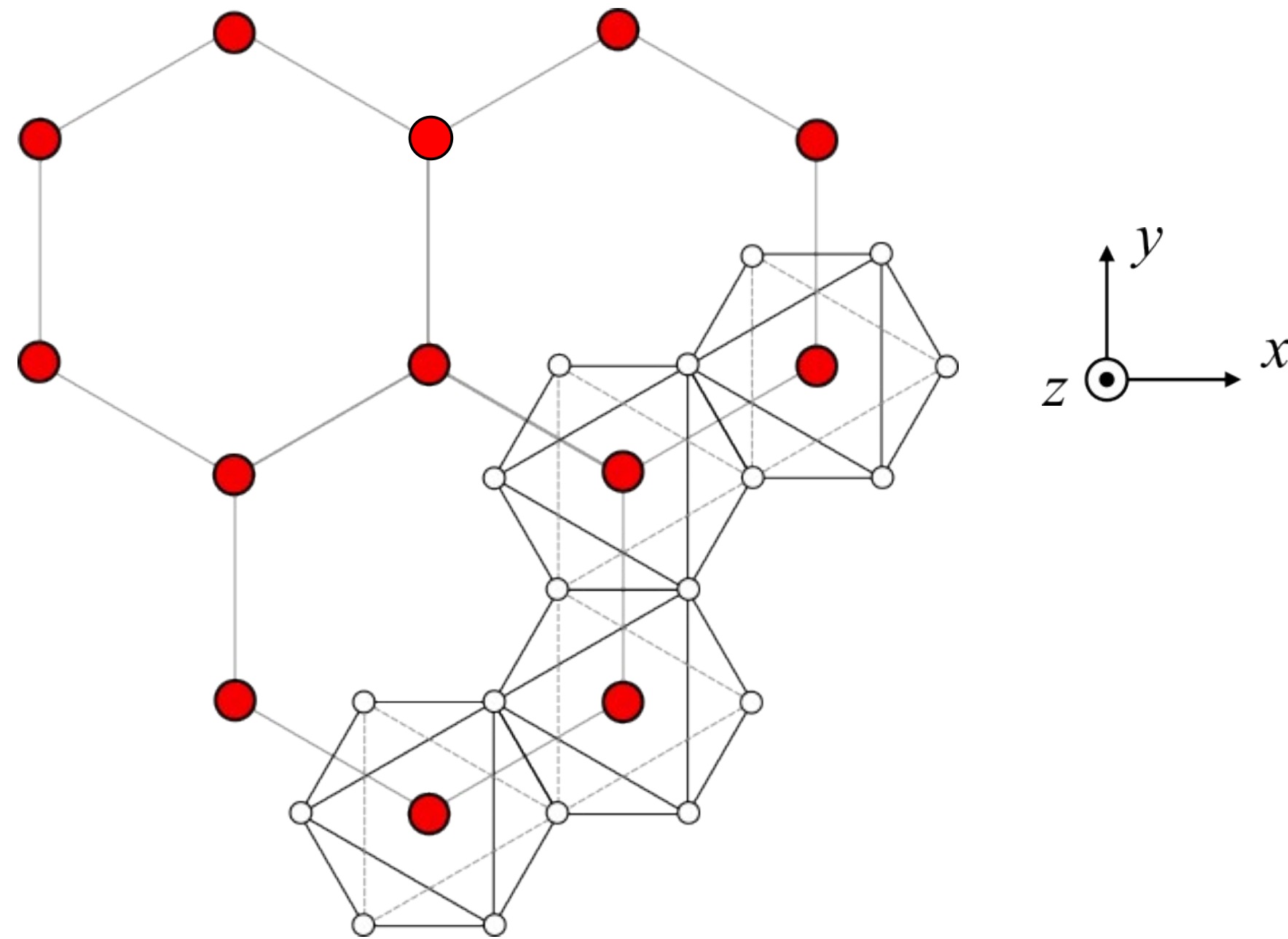


$$\hat{\mathcal{H}}_{ij} = \mathbf{S}_i^T \begin{pmatrix} J_{xx} & J_{xy} & J_{xz} \\ J_{yx} & J_{yy} & J_{yz} \\ J_{zx} & J_{zy} & J_{zz} \end{pmatrix} \mathbf{S}_j$$

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



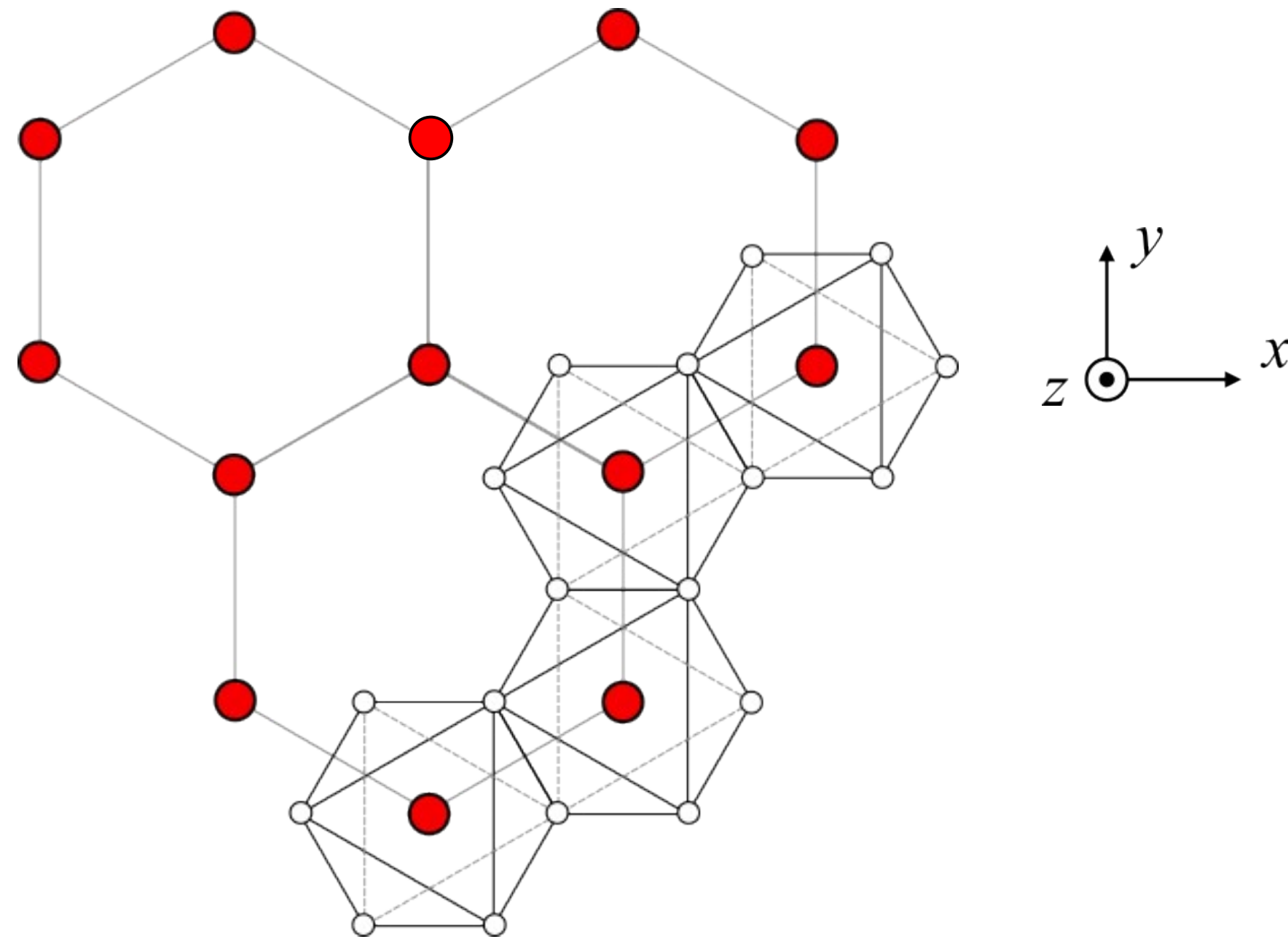
$$\hat{\mathcal{H}}_{ij} = \mathbf{S}_i^T \begin{pmatrix} J_{xx} & \cancel{J_{xy}} & \boxed{J_{xz}} \\ \cancel{J_{yx}} & J_{yy} & \cancel{J_{yz}} \\ \boxed{J_{zx}} & \cancel{J_{zy}} & J_{zz} \end{pmatrix} \mathbf{S}_j$$

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

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



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

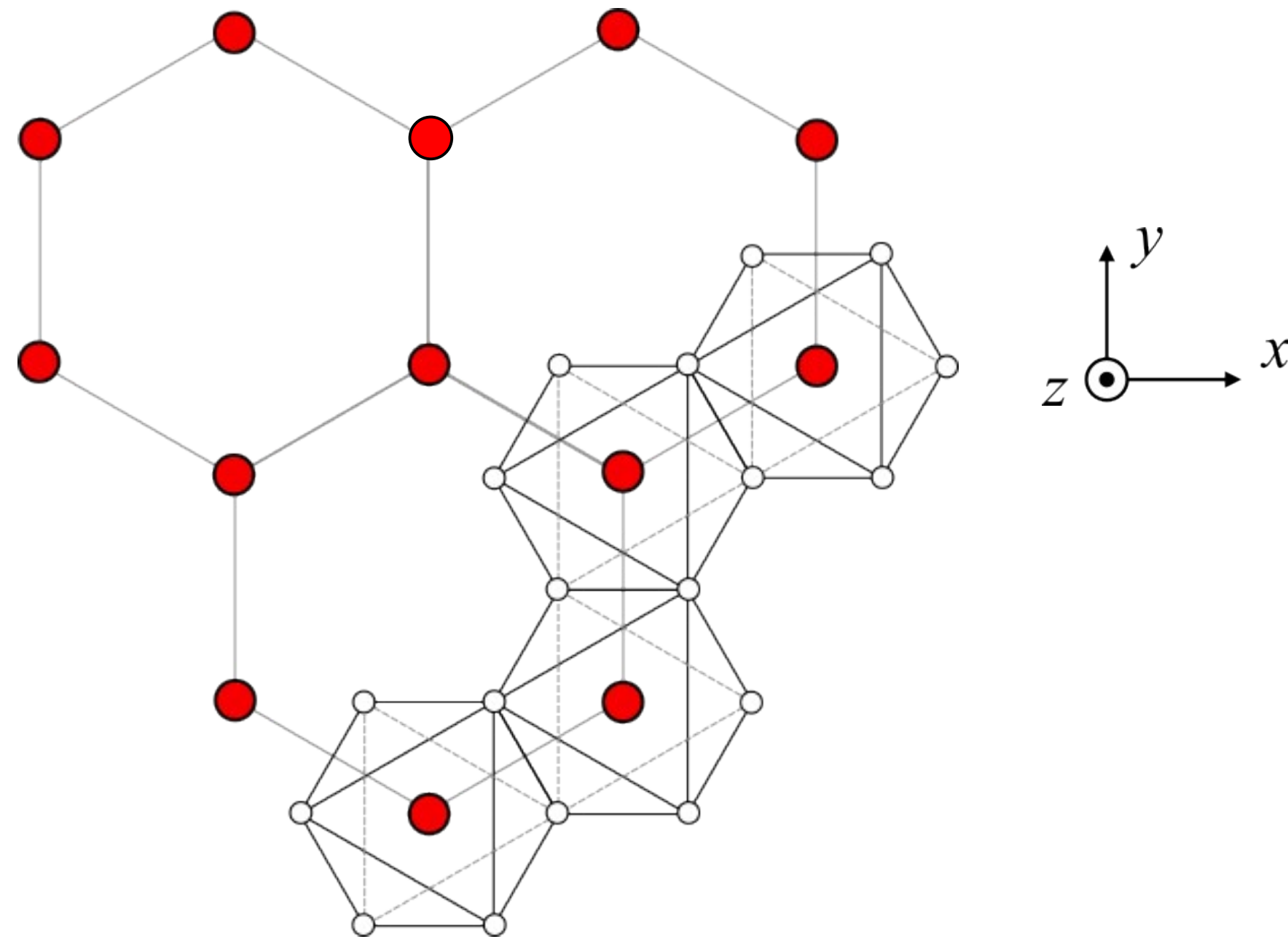
$$J_{xz} = J_{zx}$$

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

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



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

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

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

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

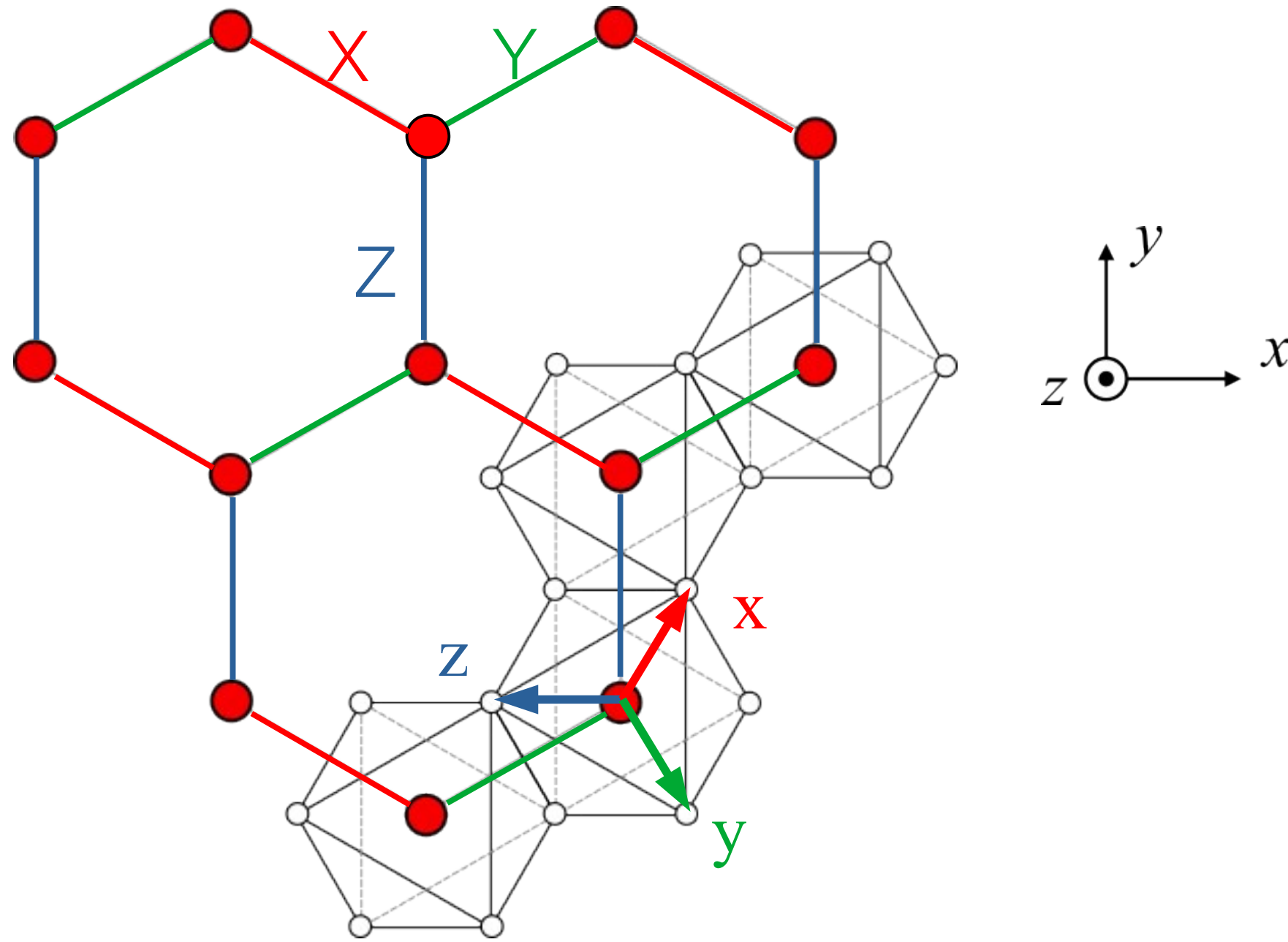
АНИЗОТРОП
НЫЙ ОБМЕН

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

Различные формы анизотропного гамильтониана



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

Расширенная модель Китаева-Гейзенберга – кубические оси

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

J.G. Rau, E.K. Lee and H.-Y. Kee, Phys. Rev. Lett. 112, 077204 (2014)

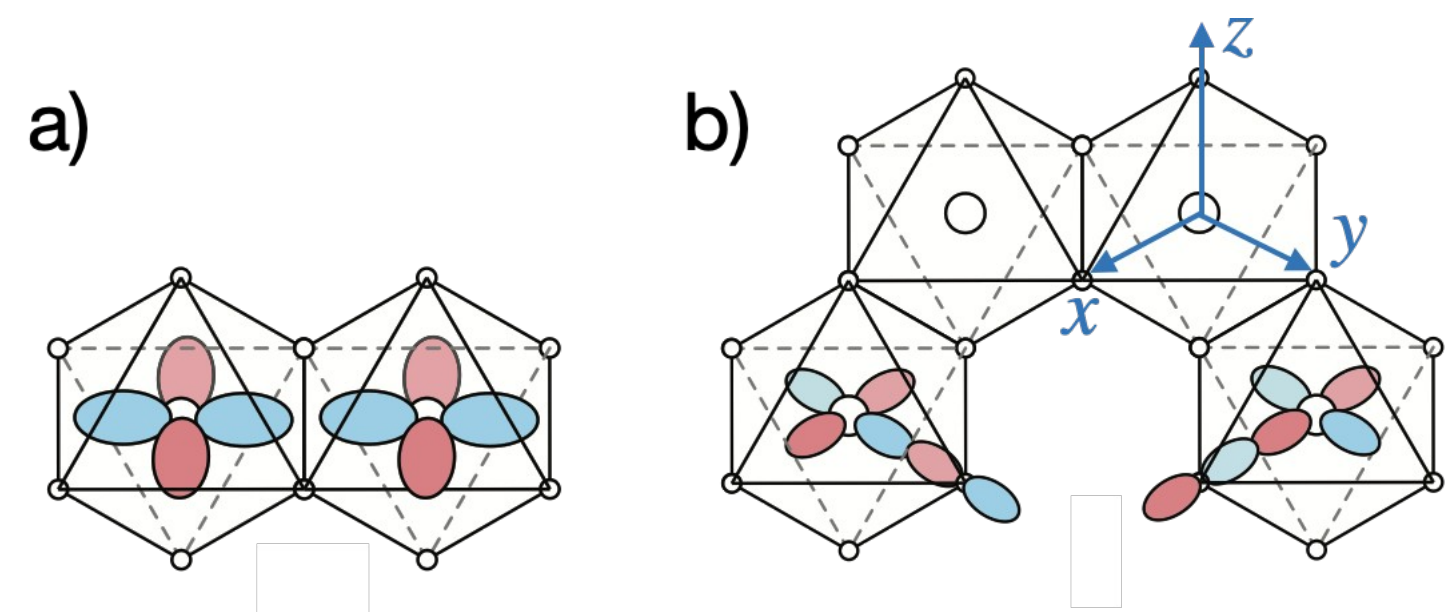
H. Liu and G. Khaliullin, Phys. Rev. B 97, 014407 (2018)

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

$$\begin{aligned}
 J &= J_0 + \frac{1}{3} (K - \Gamma - 2\Gamma'), \\
 \Delta J &= J_0 + \frac{1}{3} (K + 2\Gamma + 4\Gamma'), \\
 2J_{\pm\pm} &= \frac{1}{3} (-K - 2\Gamma + 2\Gamma'), \\
 \sqrt{2}J_{z\pm} &= \frac{1}{3} (2K - 2\Gamma + 2\Gamma'),
 \end{aligned}$$

BaCo₂(AsO₄)₂: *ab initio* exchange integrals

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



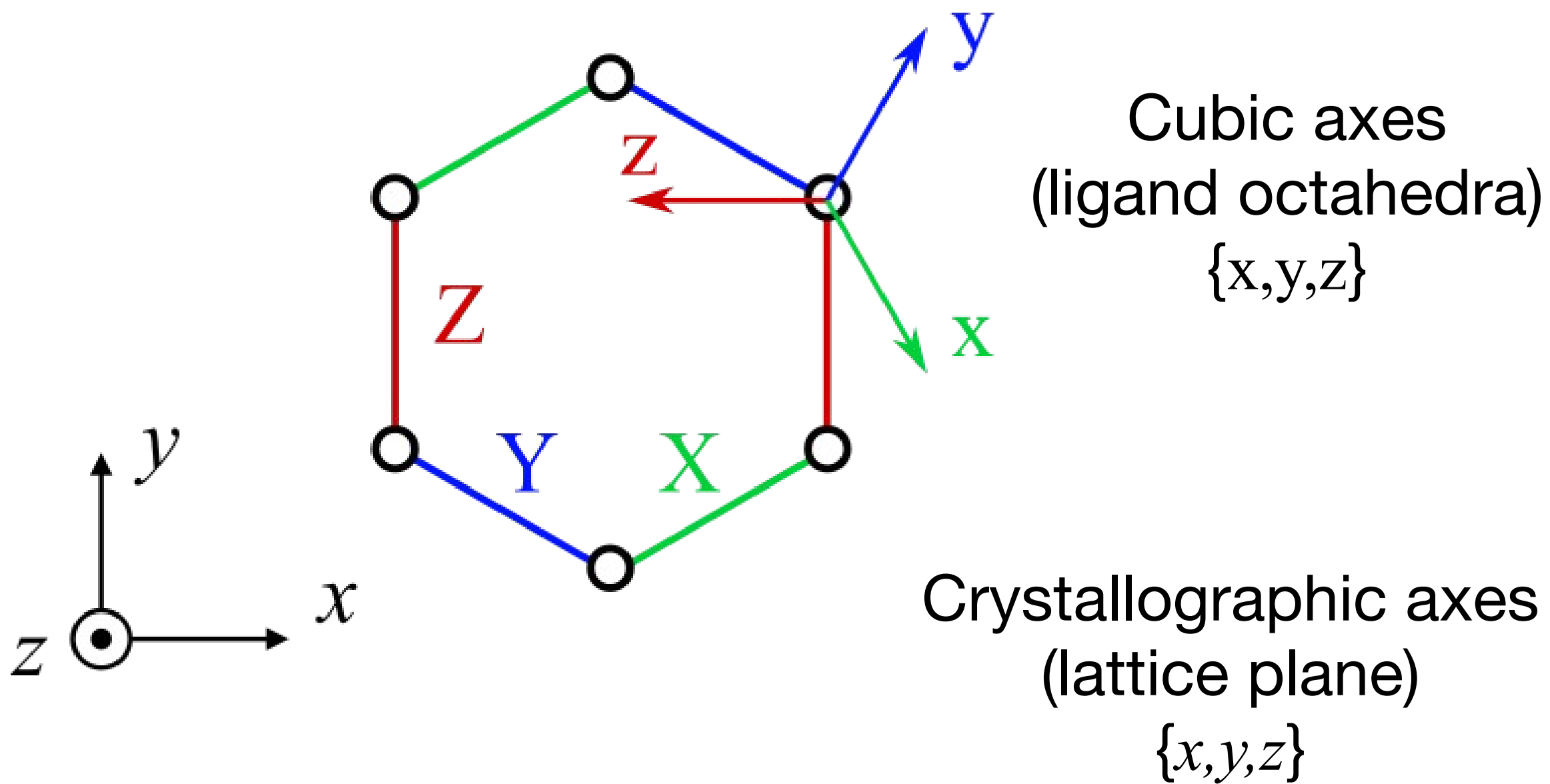
Direct hopping $t \sim -300$ meV larger than hopping
through xz, yz orbitals, $t \sim 50$ meV
Strong third-neighbor hopping
 $t_3=124$ meV

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



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

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

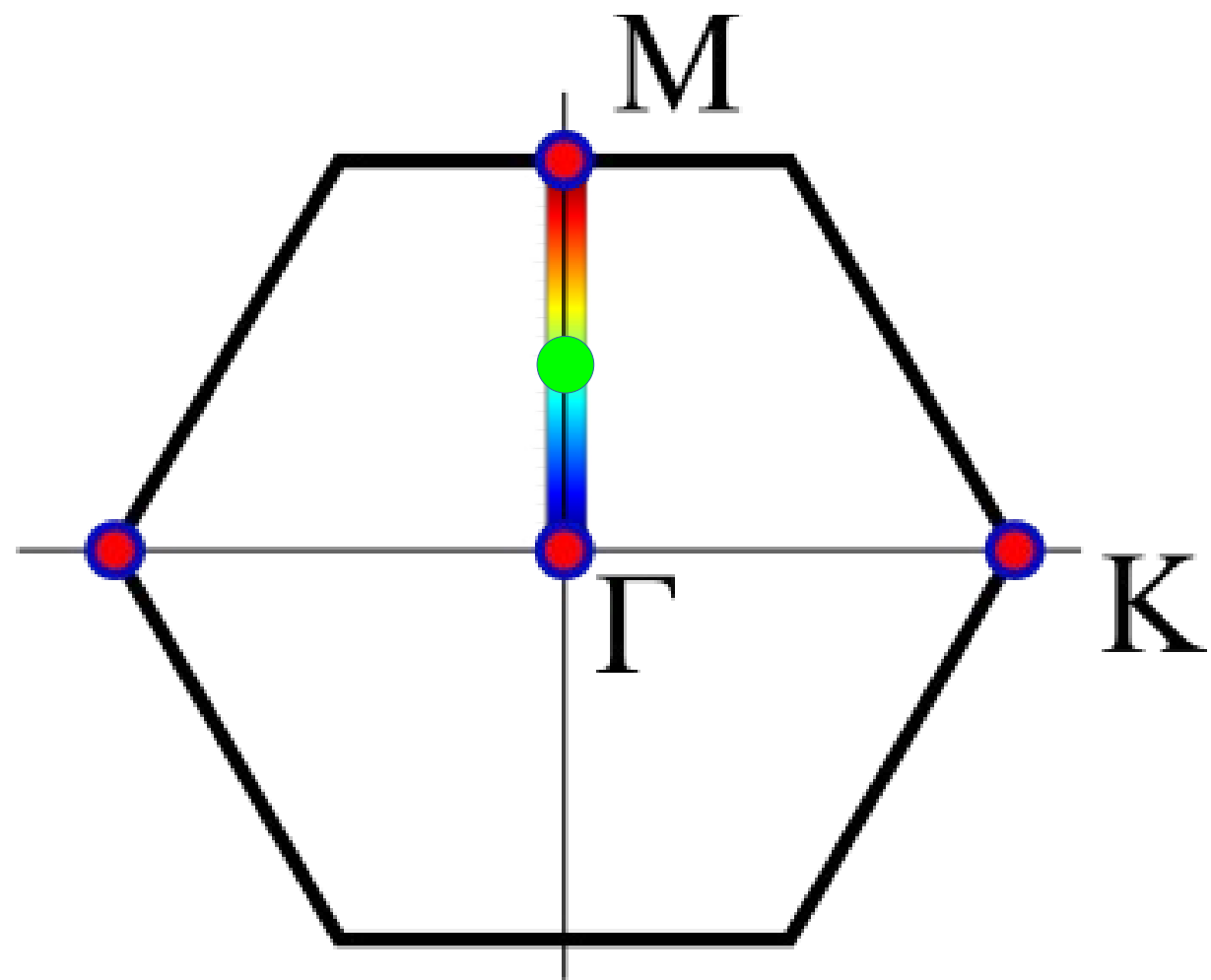
XXZ

$$\begin{aligned} J &= J_0 + \frac{1}{3} (K - \Gamma - 2\Gamma'), \\ \Delta J &= J_0 + \frac{1}{3} (K + 2\Gamma + 4\Gamma'), \\ 2J_{\pm\pm} &= \frac{1}{3} (-K - 2\Gamma + 2\Gamma'), \\ \sqrt{2}J_{z\pm} &= \frac{1}{3} (2K - 2\Gamma + 2\Gamma'), \end{aligned}$$

Pseudodipolar —
bond-dependent

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

J_1 - J_3 +XXZ model

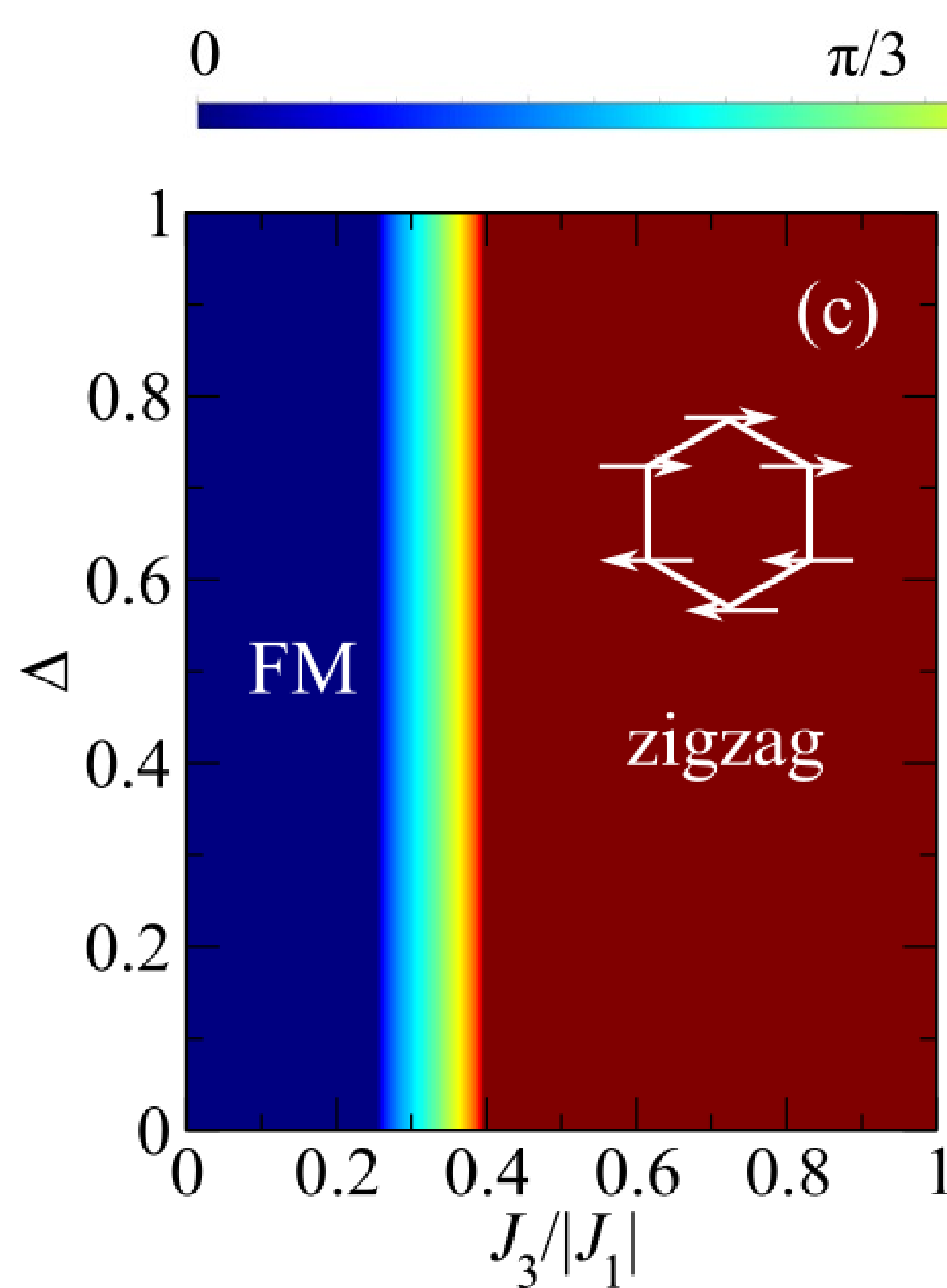


Luttinger-Tisza method:

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

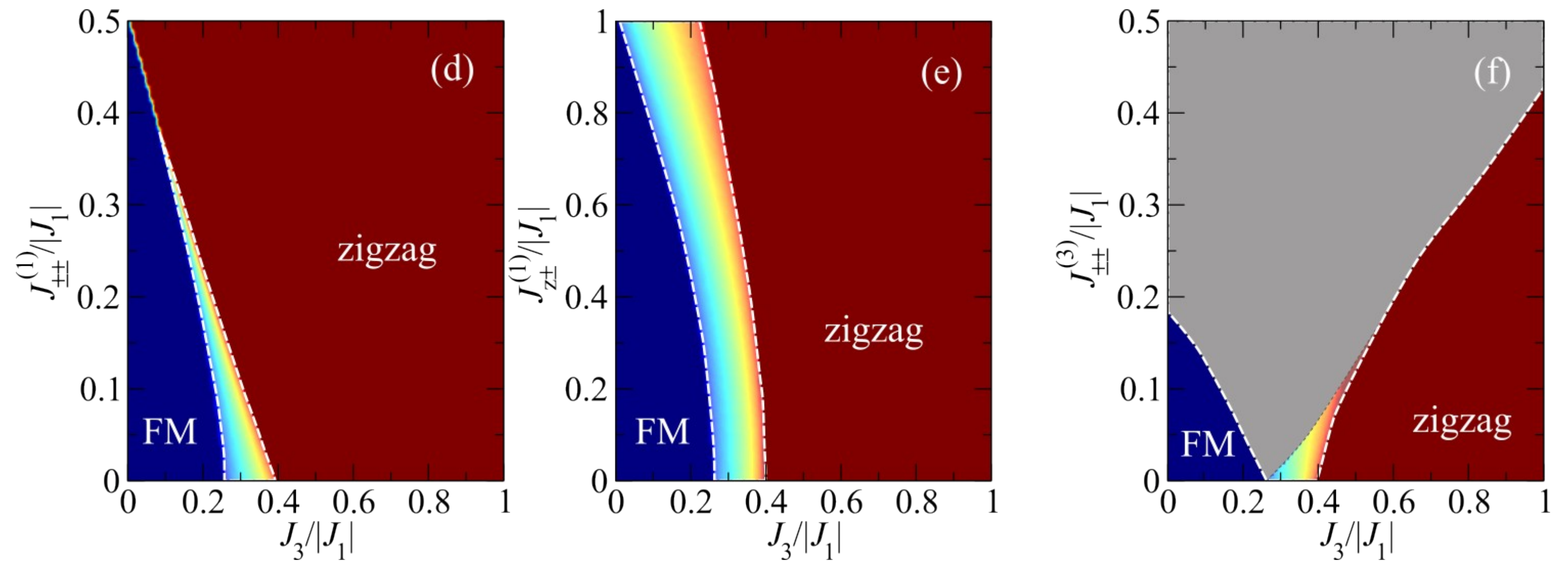
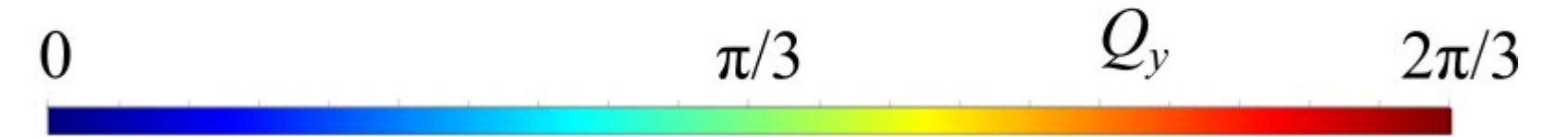
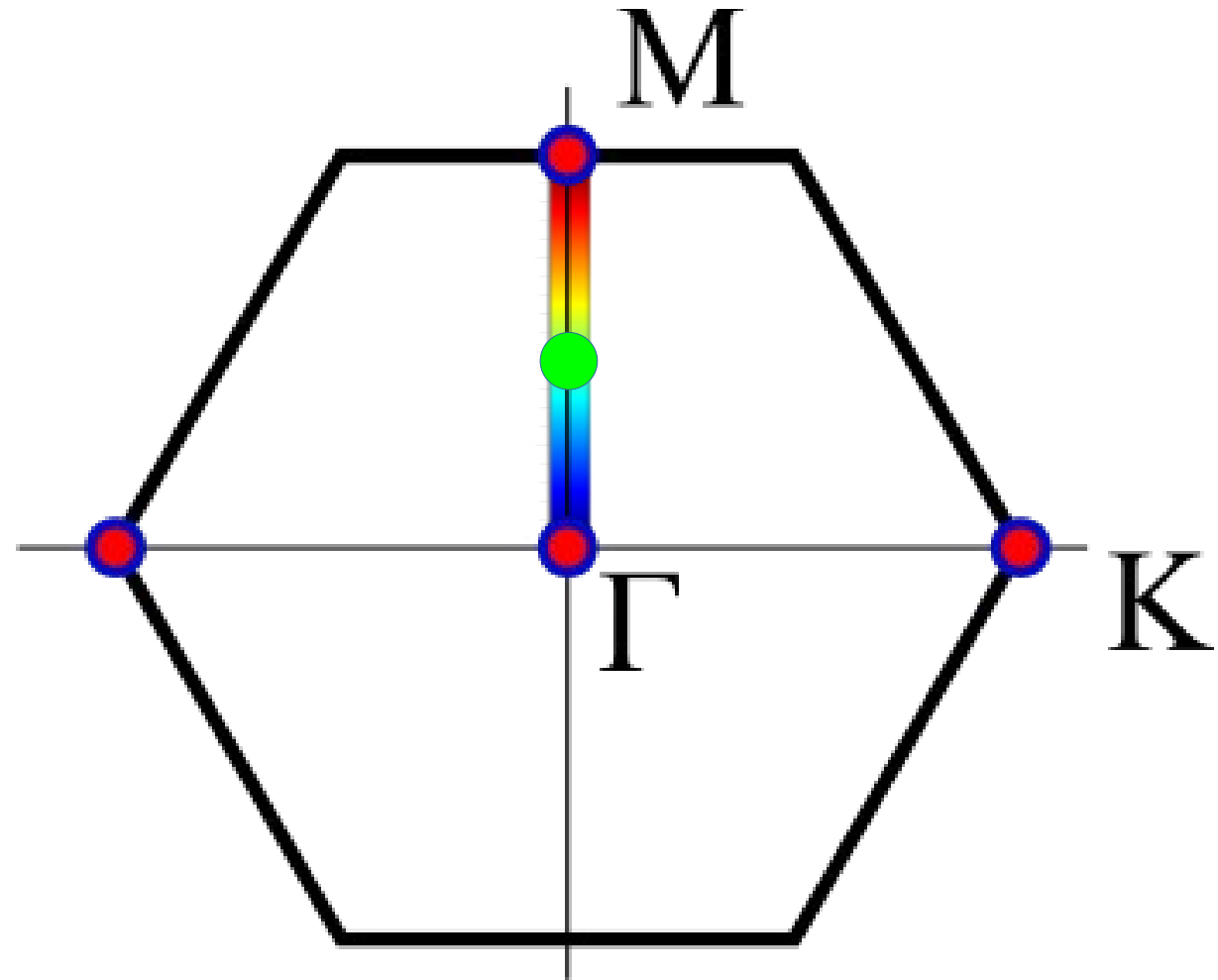
$$\mathbf{S}_i = \frac{1}{\sqrt{N}} \sum_{\mathbf{q}} \mathbf{S}_{\mathbf{q}} e^{i\mathbf{q}\mathbf{r}_i}$$

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



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

J_1 - J_3 +XXZ model+bond-dependent terms



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

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

BaCo₂(AsO₄)₂

TABLE VI. Local exchange couplings for BaCo₂(AsO₄)₂.

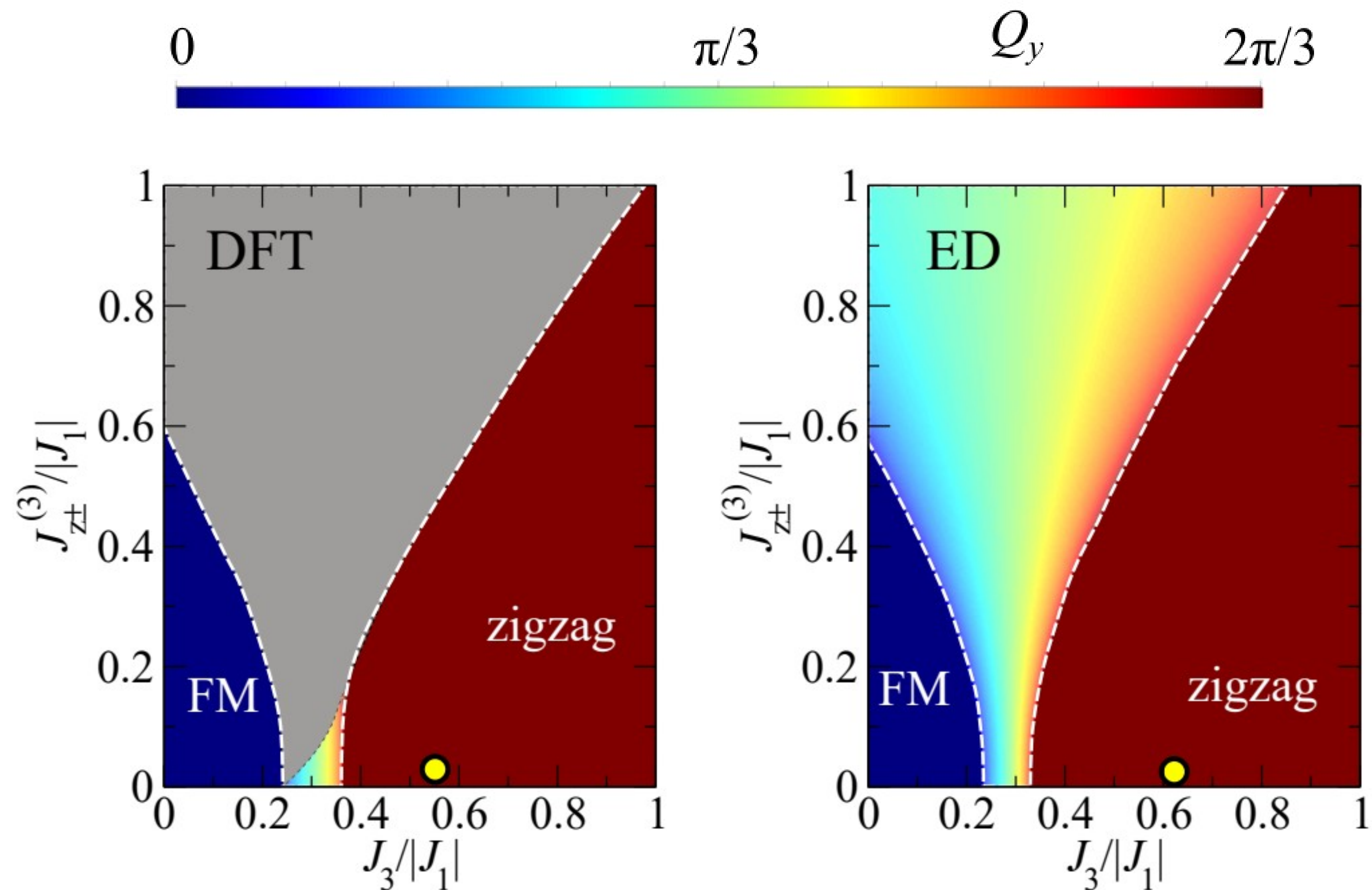
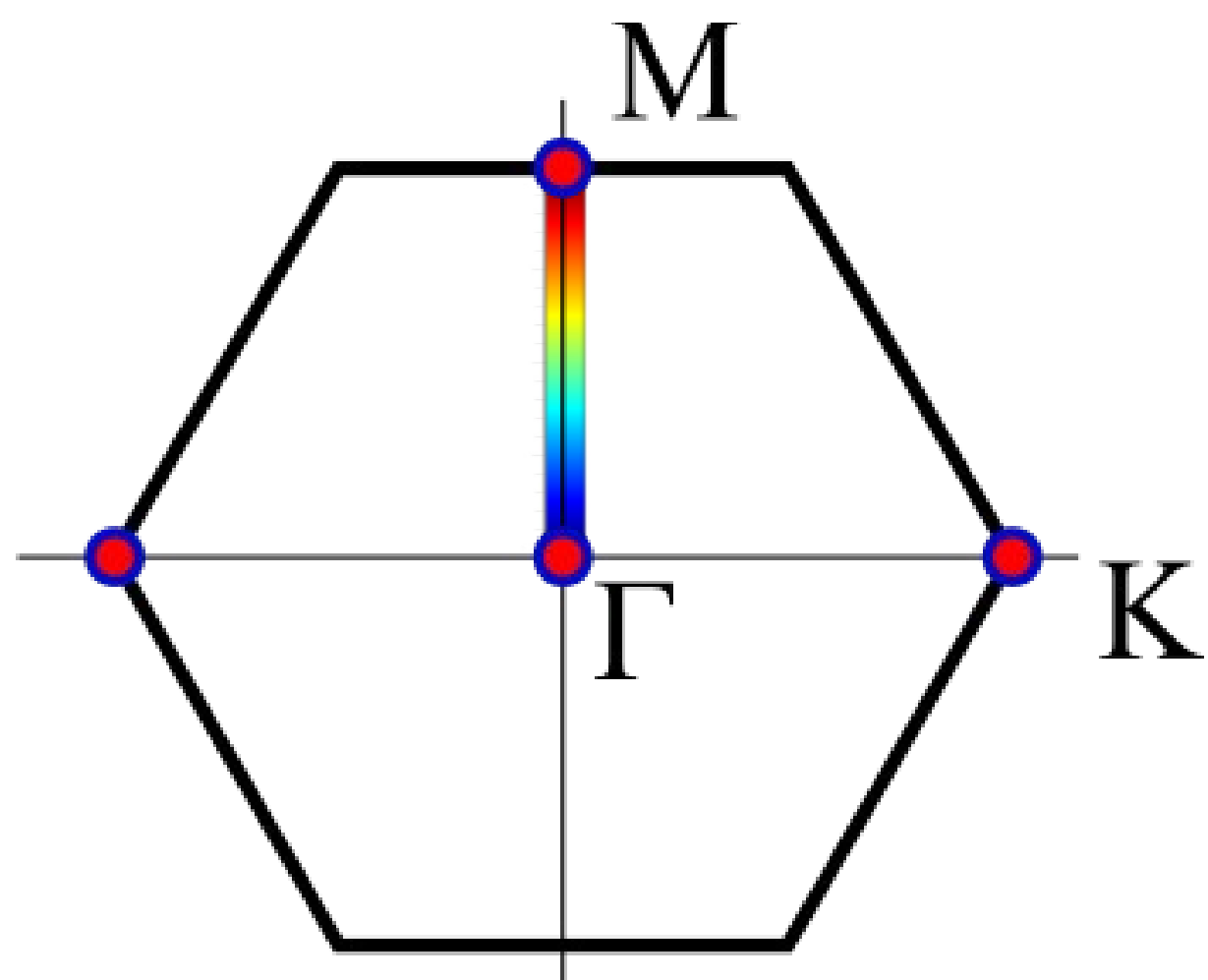
BaCo ₂ (AsO ₄) ₂	Bond-1	Bond-3
J	−12.40	5.51
K	2.06	−0.30
Γ	4.05	−1.44
η	−0.84	0.00
Γ'_1	1.82	−1.42
Γ'_2	4.89	−1.45

- - -

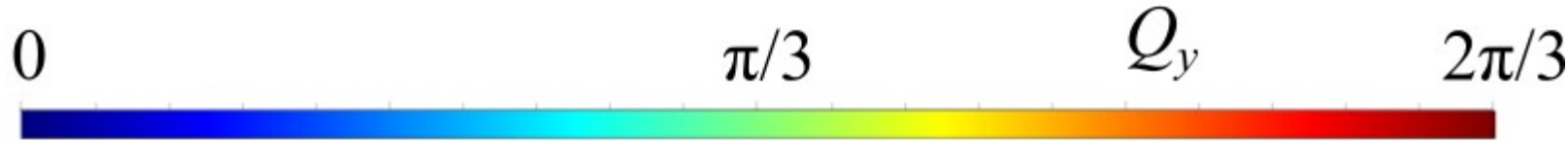
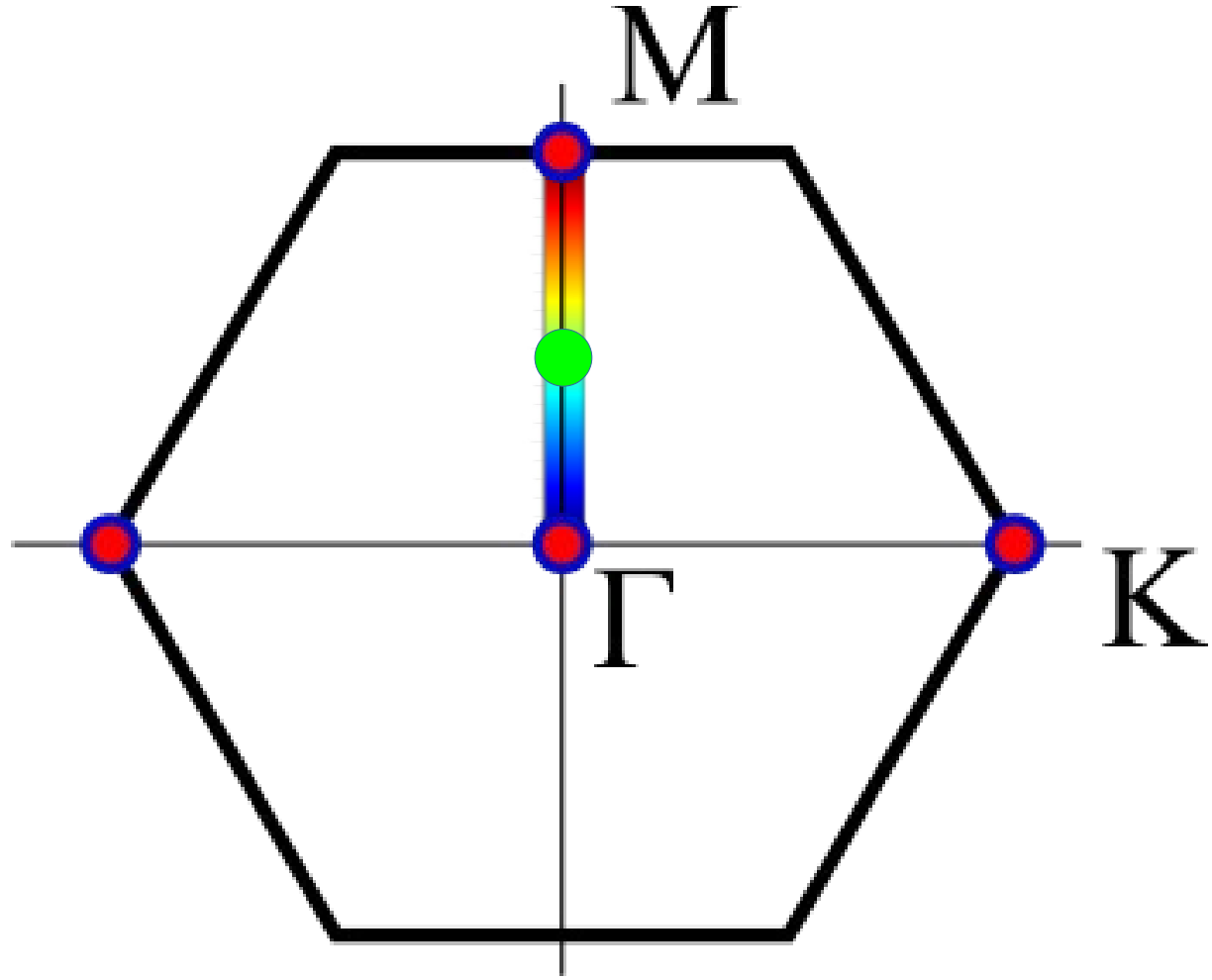
BaCo ₂ (AsO ₄) ₂	Bond-1	Bond-3
J^{XY}	−14.5 meV	7.1 meV
J^Z	−3.8 meV	2.3 meV
D	1.5 meV	≈0
E	−1.7 meV	≈0

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

Ab initio ground state

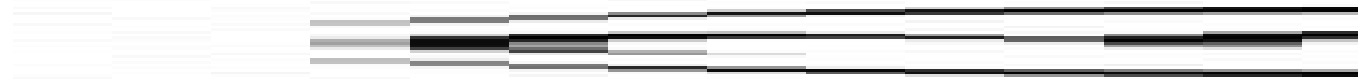


Minimal model for BaCo₂(AsO₄)₂

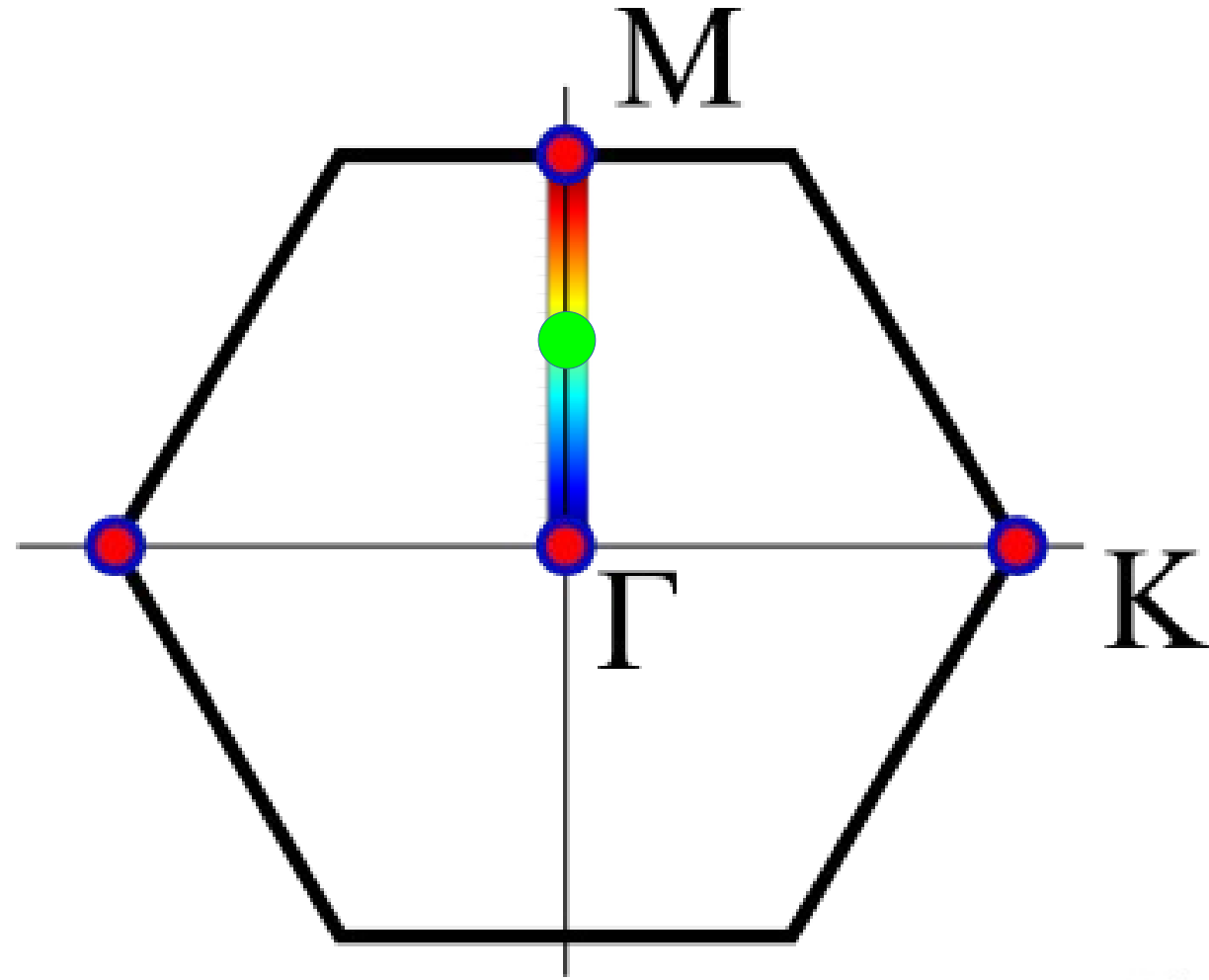


$$\begin{aligned}
 H_{\min} = & \sum_{\langle ij \rangle_1} J_1 \left(S_i^x S_j^x + S_i^y S_j^y \right) + J_3 \sum_{\langle ij \rangle_3} \left(S_i^x S_j^x + S_i^y S_j^y \right) \\
 & - J_{z\pm}^{(3)} \left(\left(S_i^x S_j^z + S_i^z S_j^x \right) c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y \right) s_\alpha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

$$J_{z\pm} = \frac{\sqrt{2}}{3} (K - \Gamma + \Gamma')$$

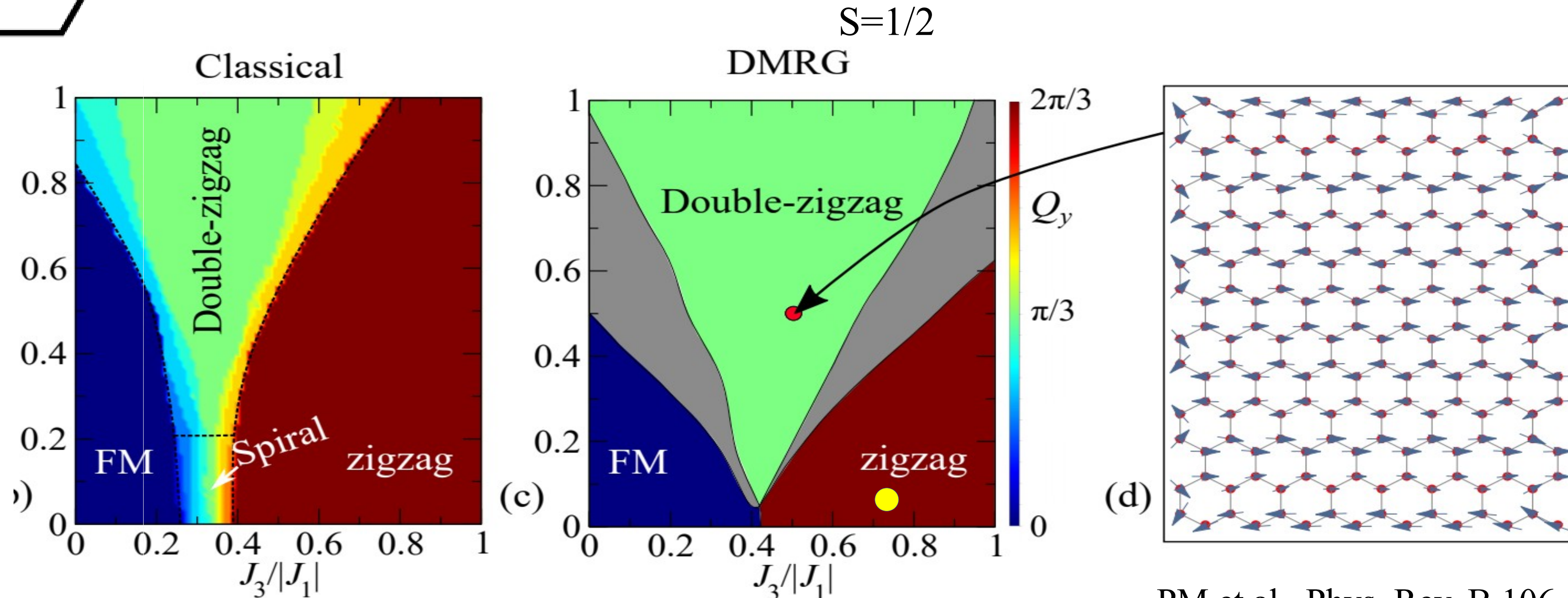


Double zigzag in $\text{BaCo}_2(\text{AsO}_4)_2$



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

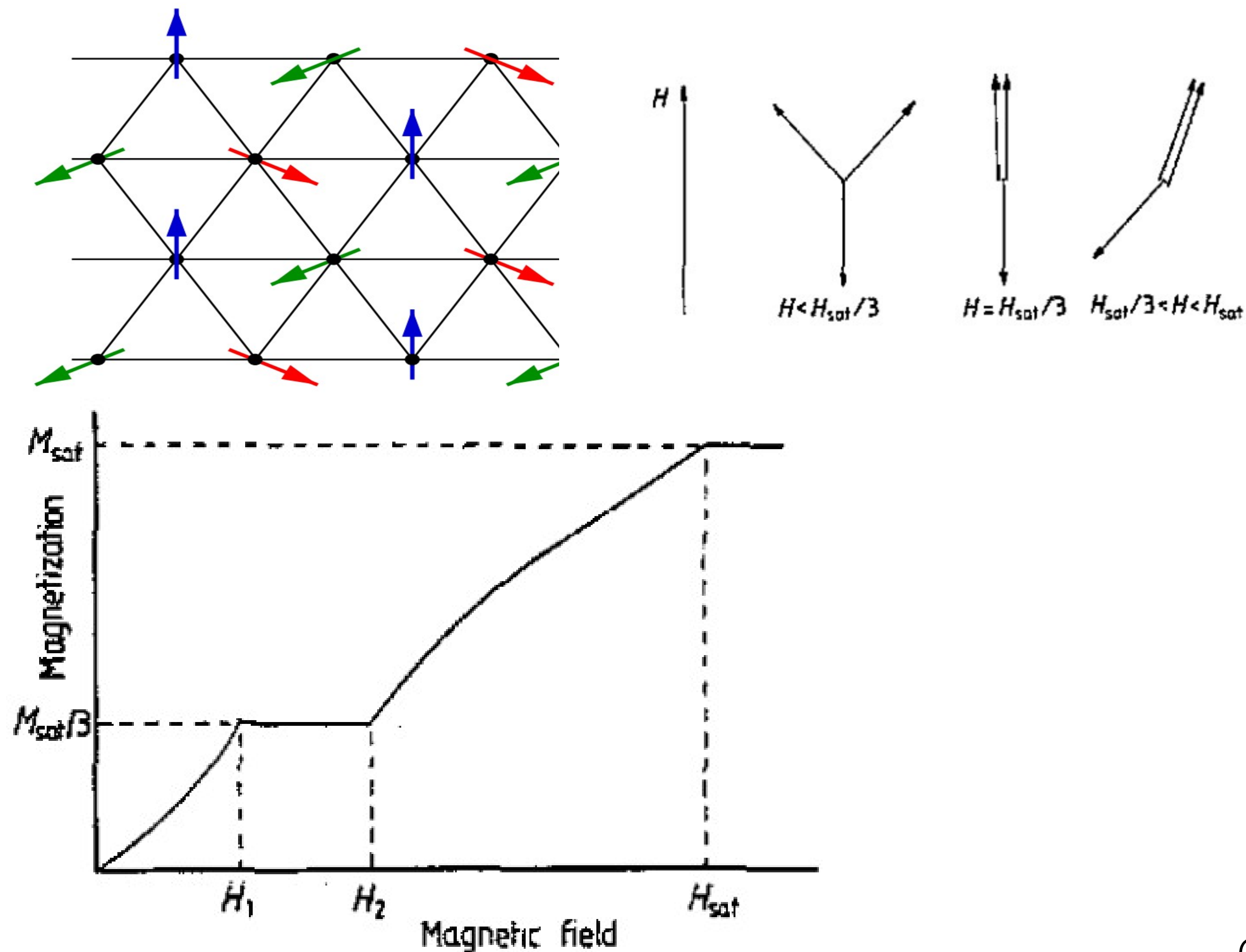
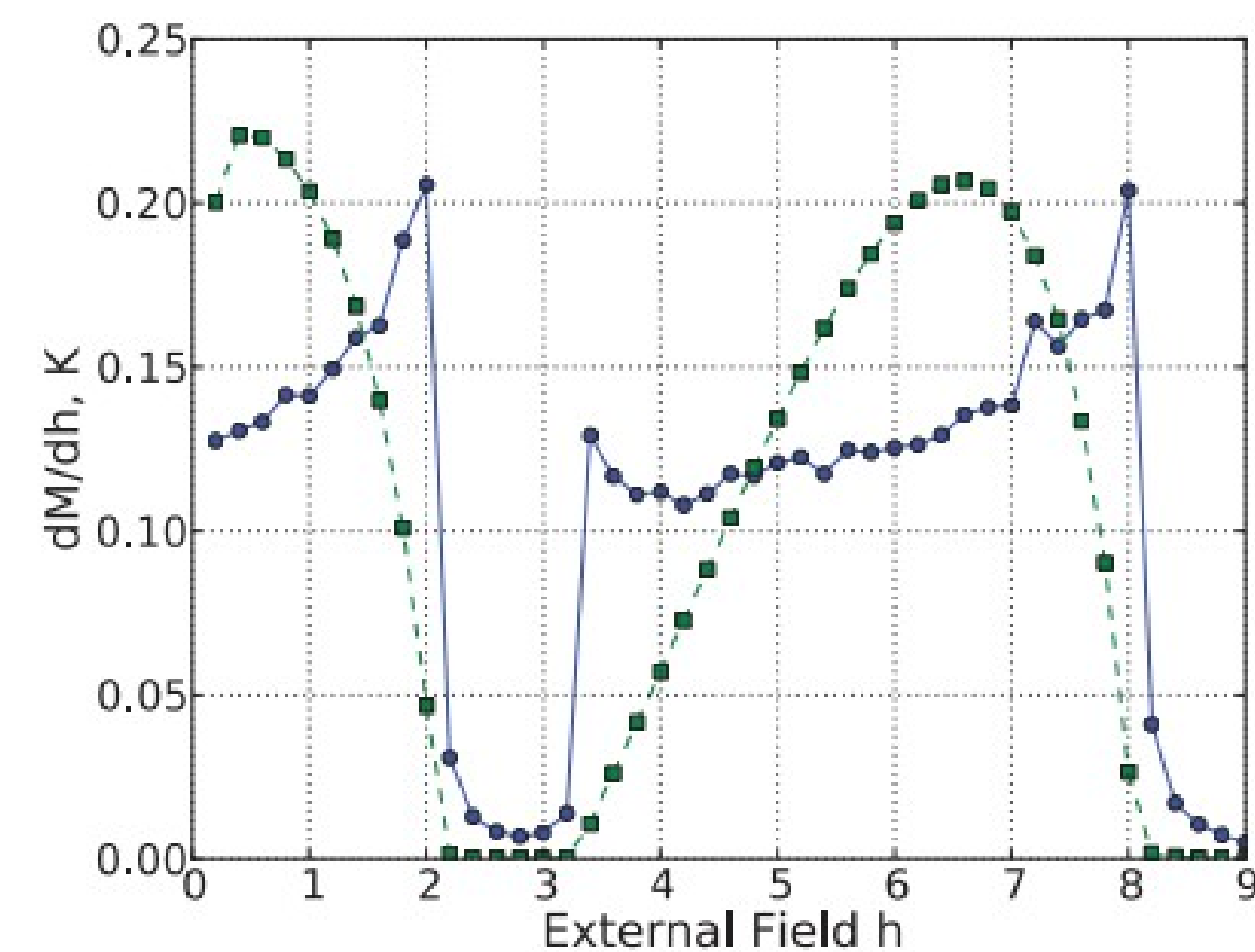
$$J_{z\pm} = \frac{\sqrt{2}}{3} (K - \Gamma + \Gamma')$$



Quantum fluctuations

Biquadratic coupling

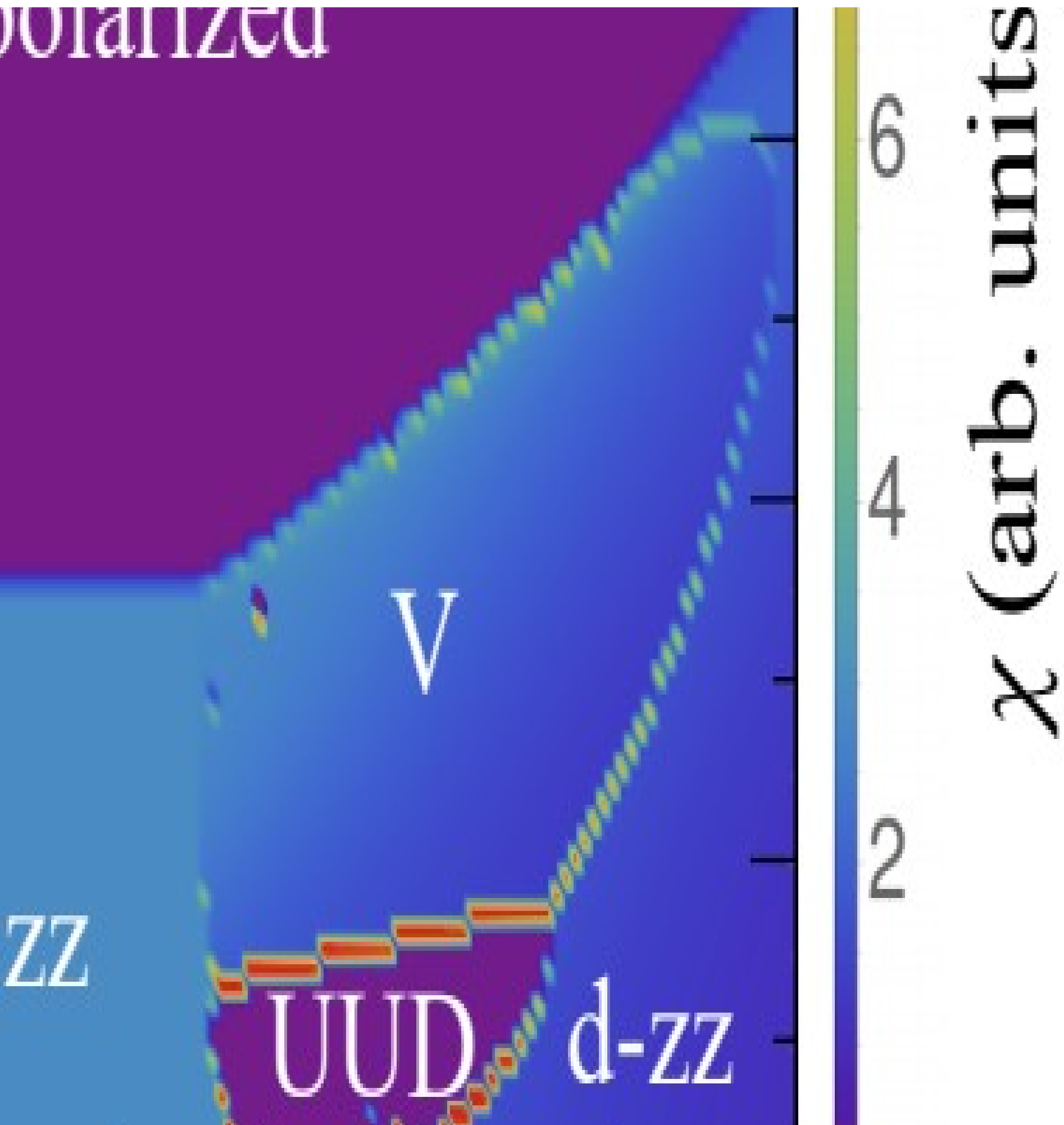
$$B \sum_{\langle ij \rangle} (\mathbf{S}_i \mathbf{S}_j)^2$$



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

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

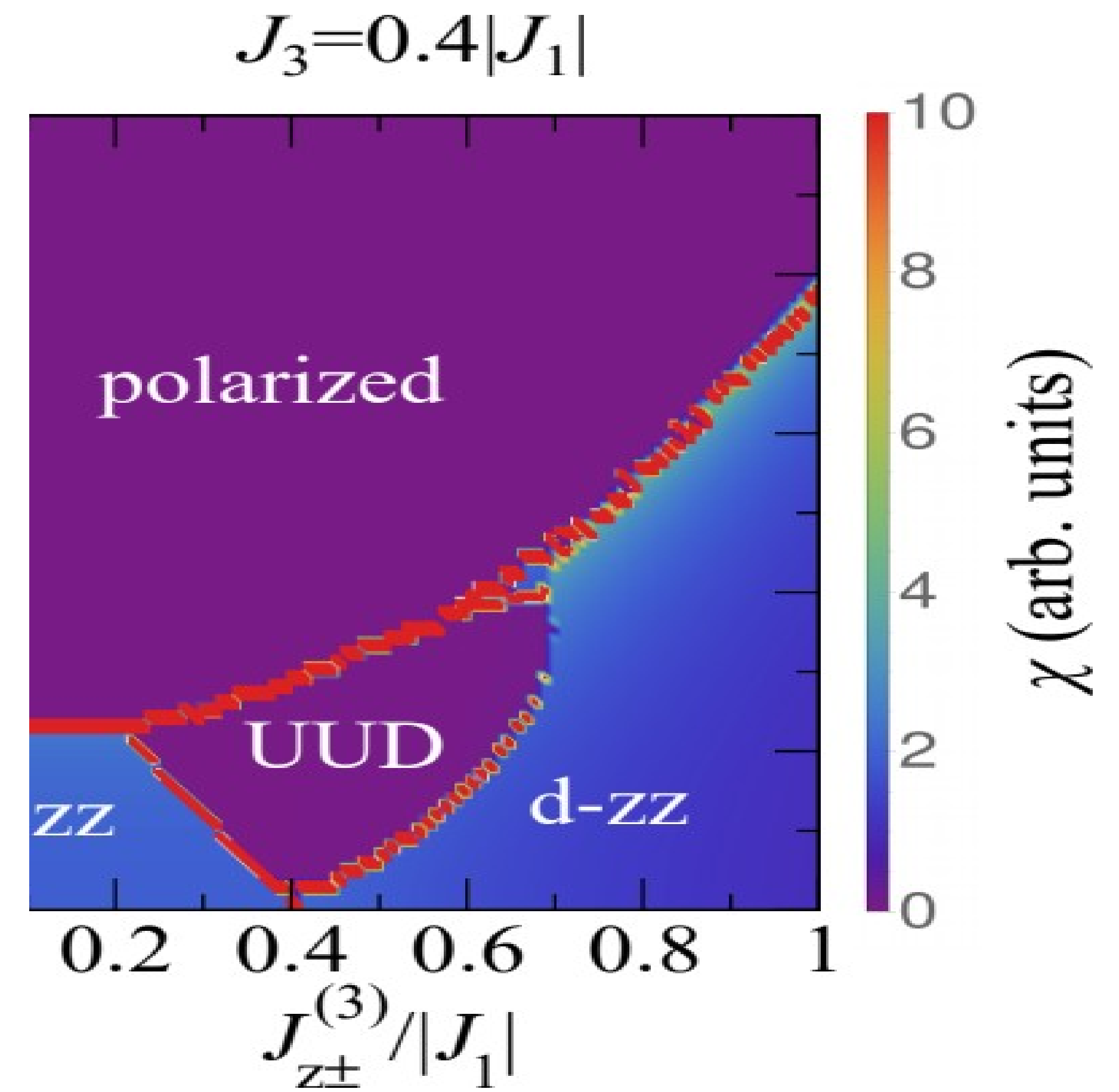
Quantum fluctuations



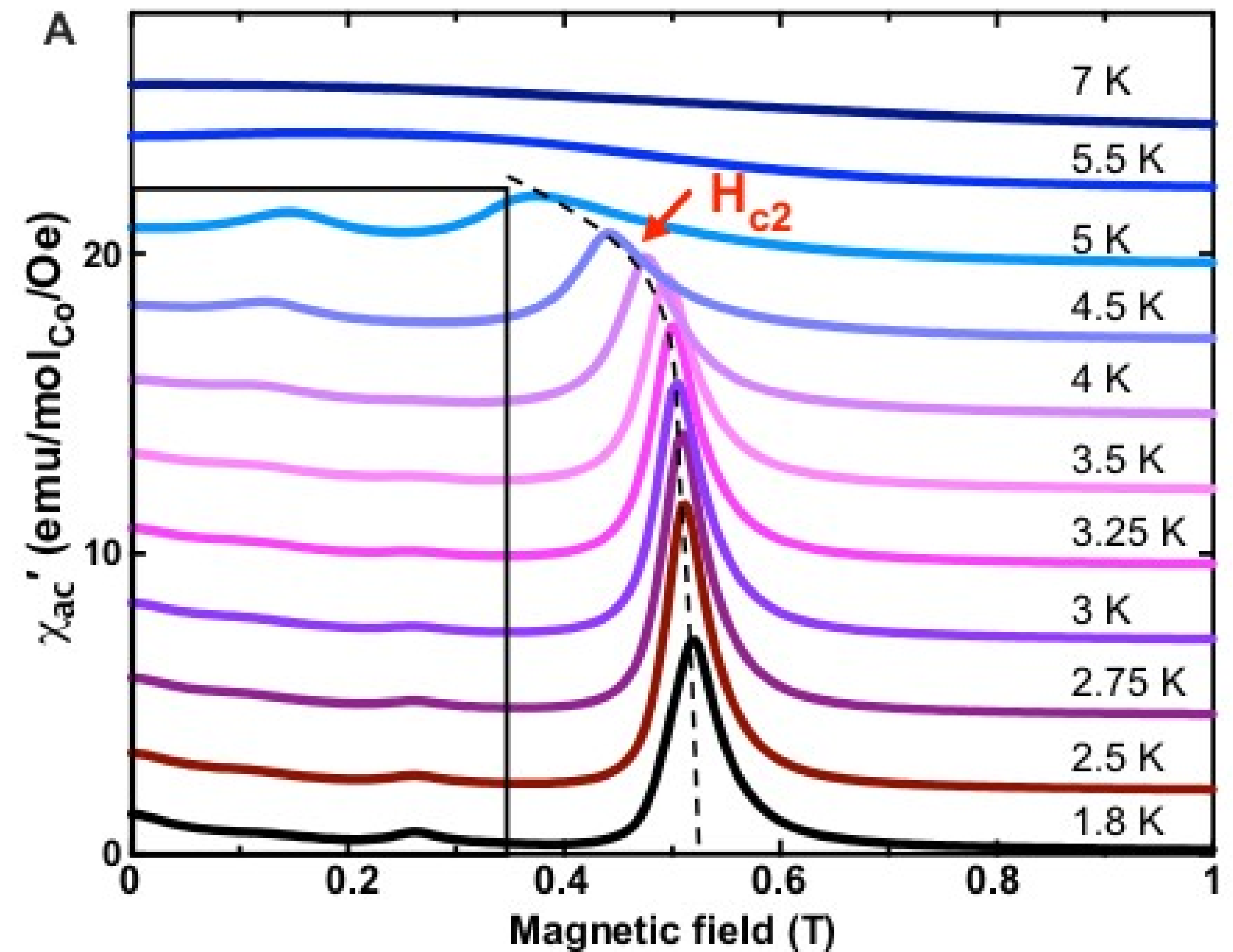
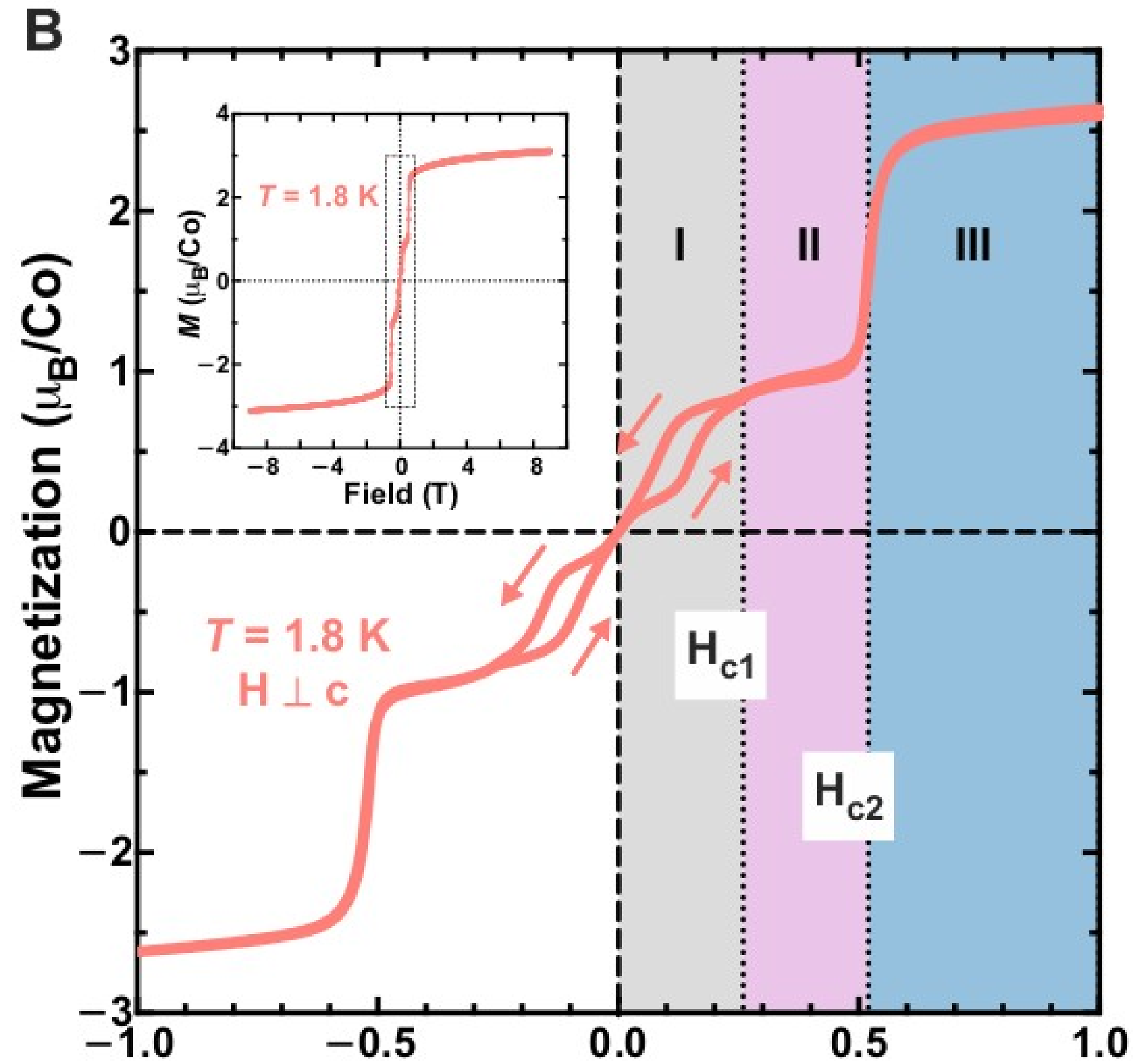
Biquadratic coupling

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

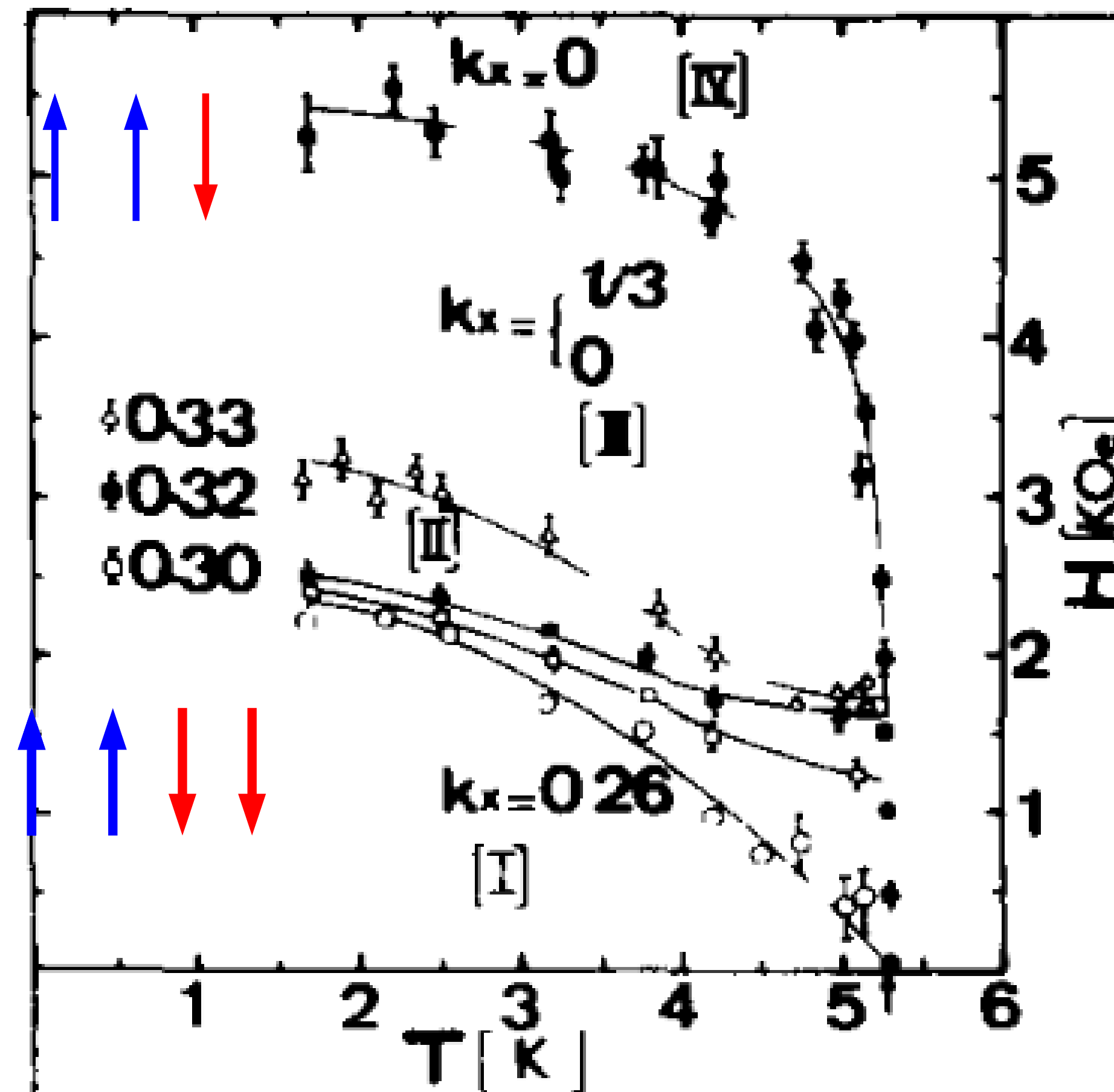
$B = 0.04|J_1|$



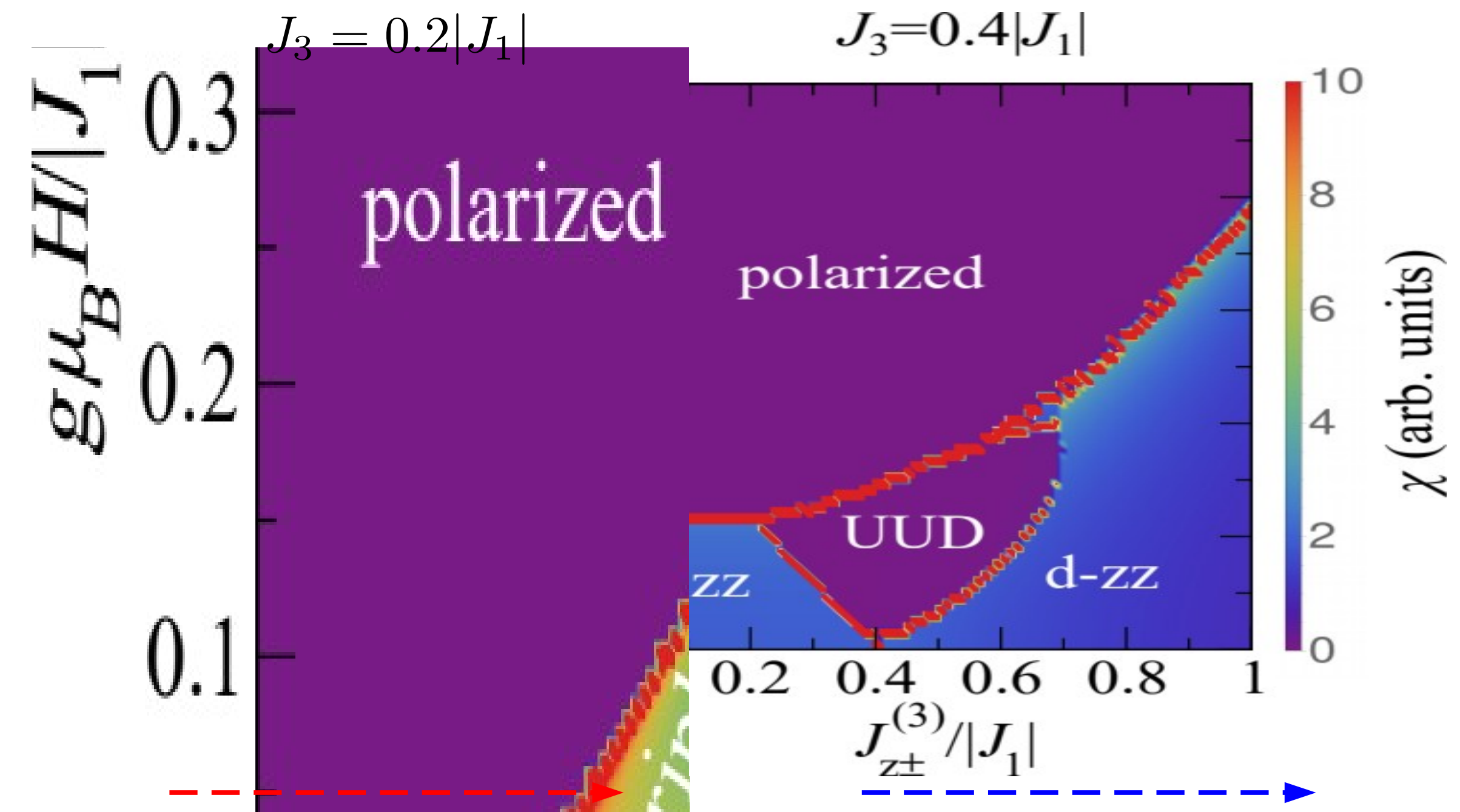
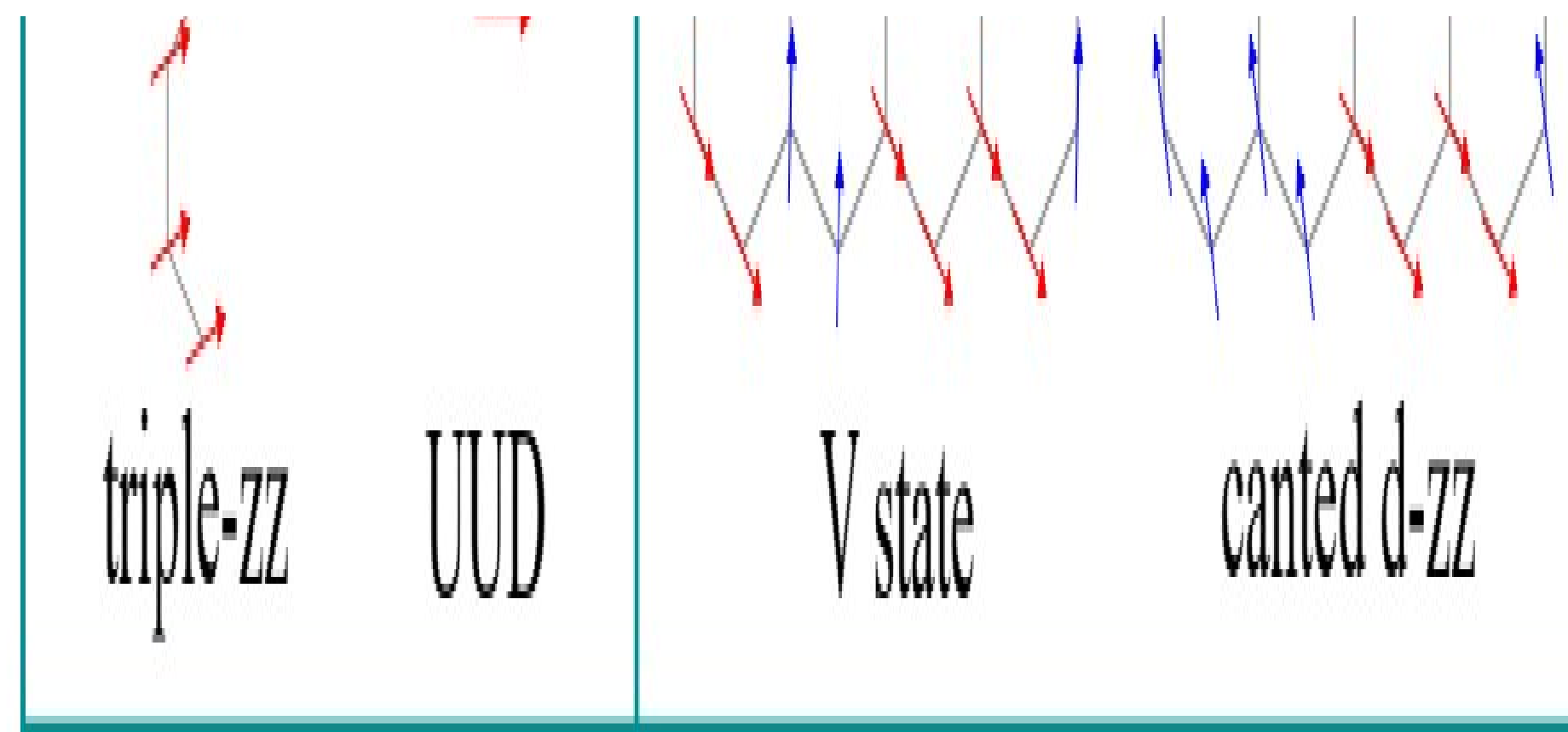
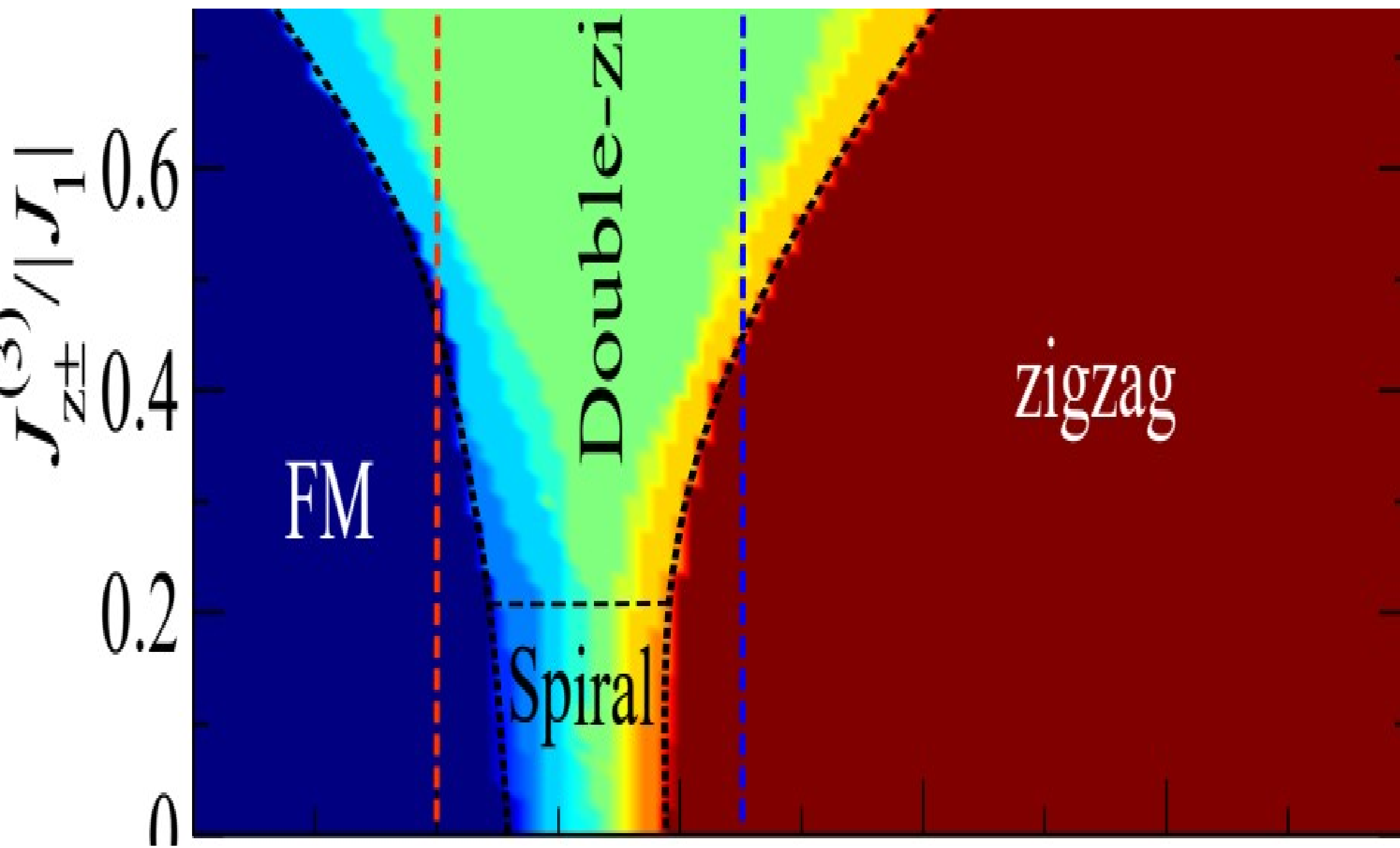
Field-induced magnetic transitions in $\text{BaCo}_2(\text{AsO}_4)_2$



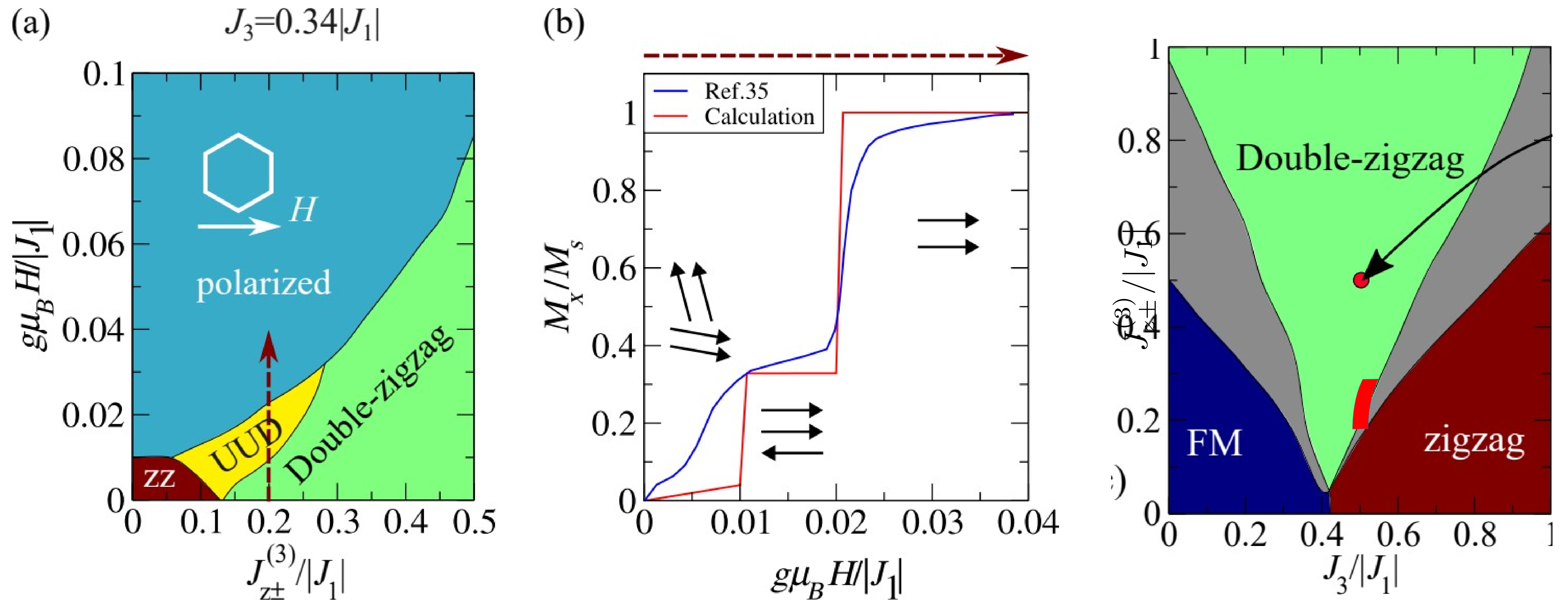
Field induced magnetic transitions in $\text{BaCo}_2(\text{AsO}_4)_2$



Field-induced transitions of the minimal model

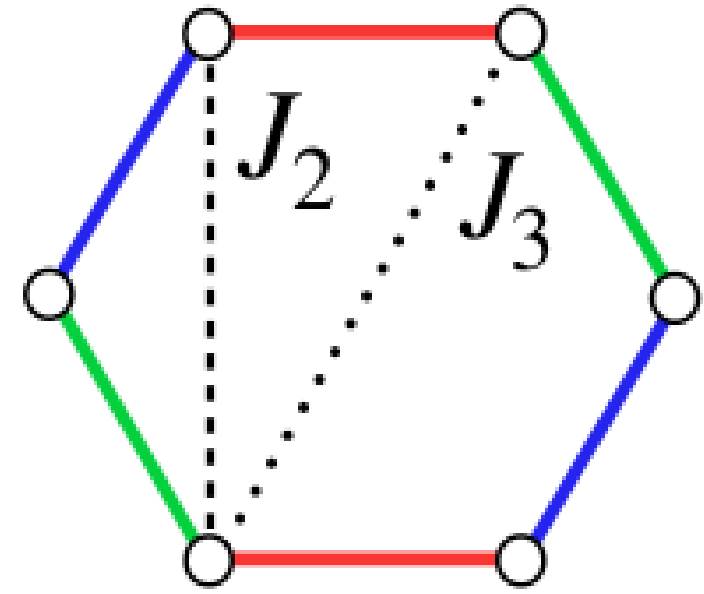


Field transitions in $\text{BaCo}_2(\text{AsO}_4)_2$



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

Inelastic neutron scattering in $\text{BaCo}_2(\text{AsO}_4)_2$: fitting



Spin-wave theory

$$S^+ \approx a$$

$$S^- \approx a^\dagger$$

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

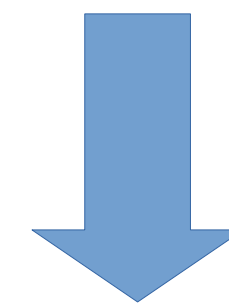
$$\begin{aligned}
 H = & \sum_{\langle ij \rangle_n} J_n \left(S_i^x S_j^x + S_i^y S_j^y + \Delta_n S_i^z S_j^z \right) \\
 & - 2J_{\pm\pm} \left(\left(S_i^x S_j^x - S_i^y S_j^y \right) c_\alpha - \left(S_i^x S_j^y + S_i^y S_j^x \right) s_\alpha \right) \\
 & - J_{z\pm} \left(\left(S_i^x S_j^z + S_i^z S_j^x \right) c_\alpha + \left(S_i^y S_j^z + S_i^z S_j^y \right) s_\alpha \right)
 \end{aligned} \quad n=1,2,3$$

7 equations $\rightarrow$

Free parameters

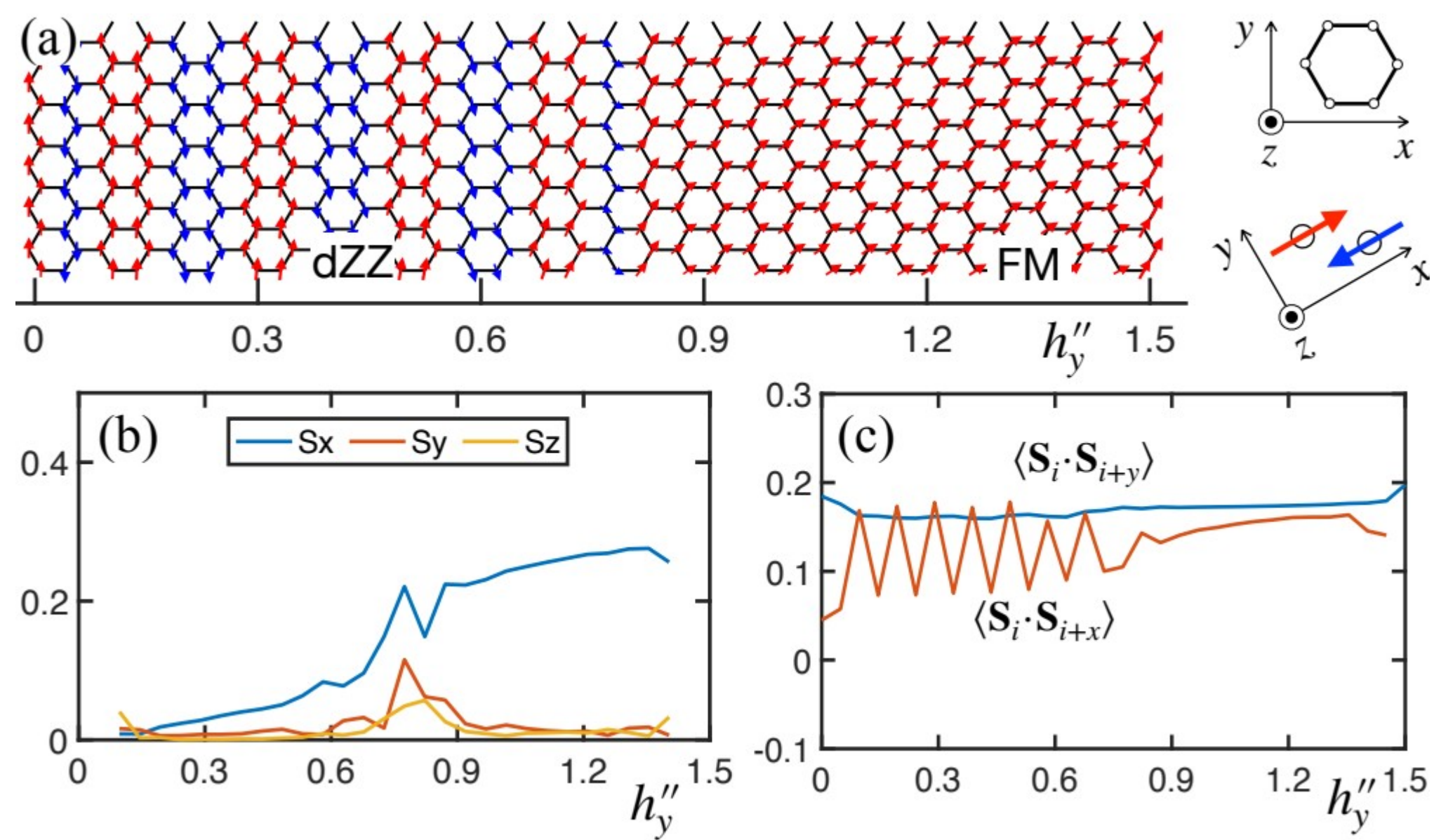
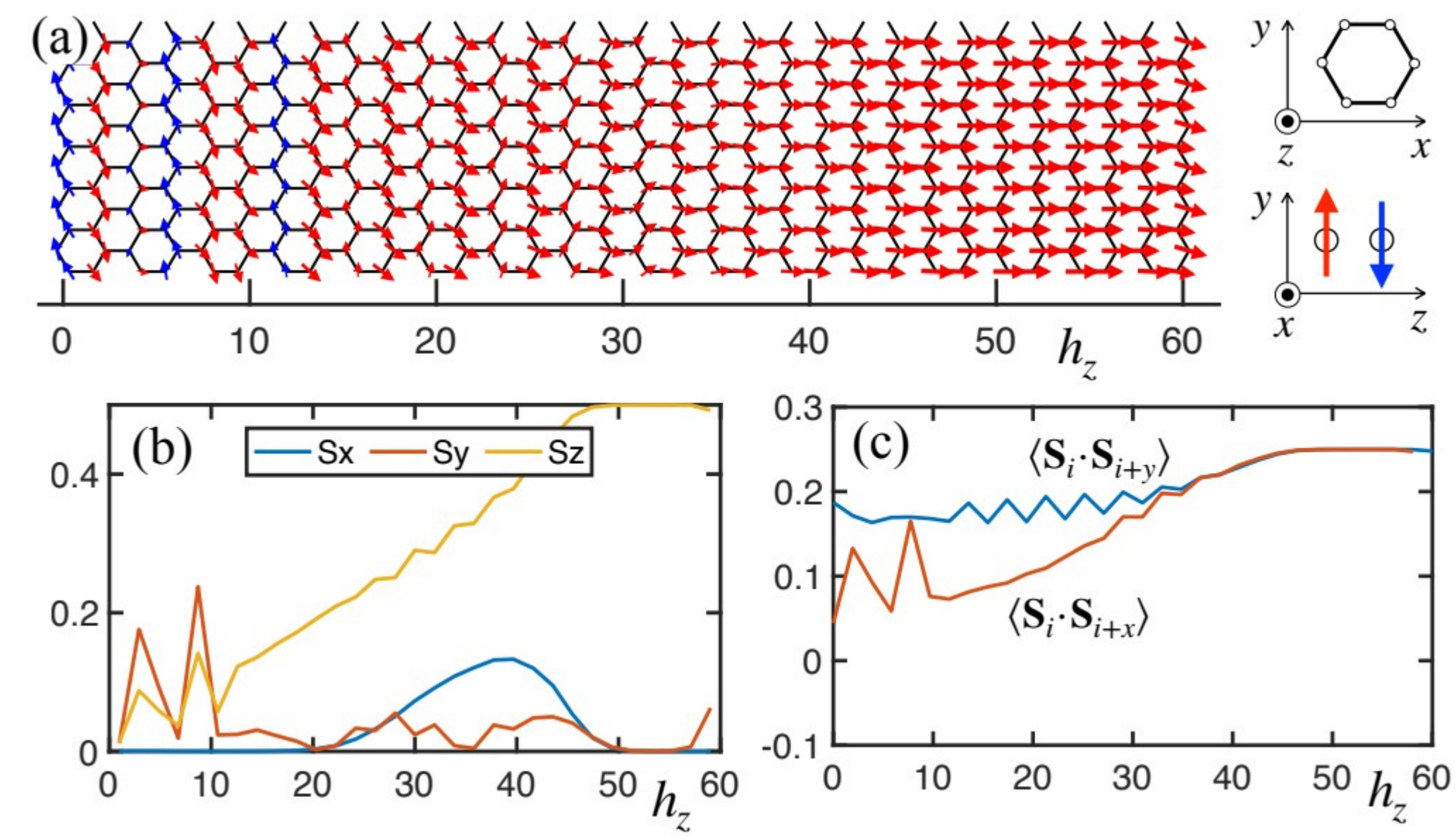
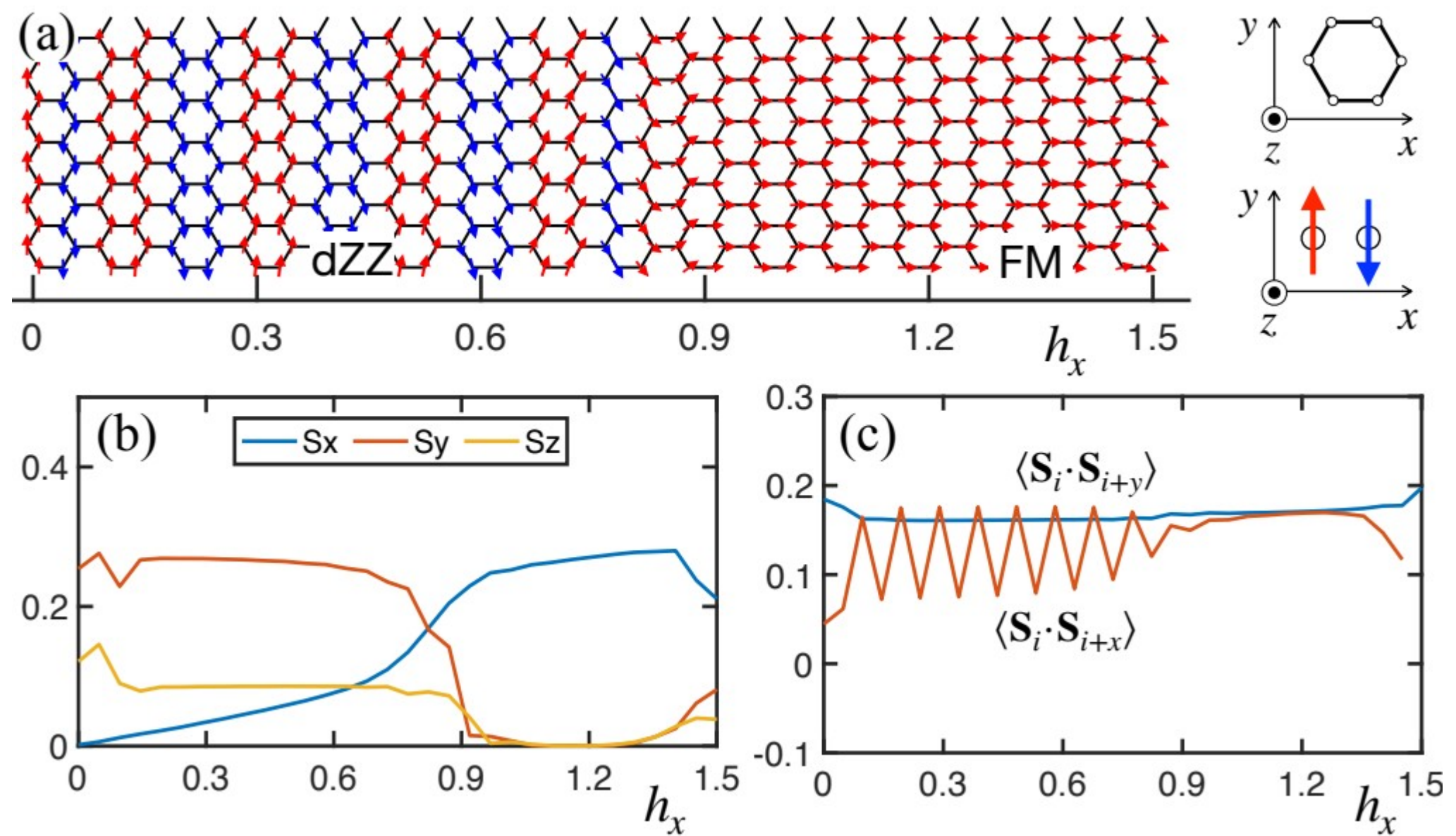
$$\{J_1, \Delta_1, J_{\pm\pm}, J_{z\pm}, J_3, \Delta_3, g_b\}$$

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

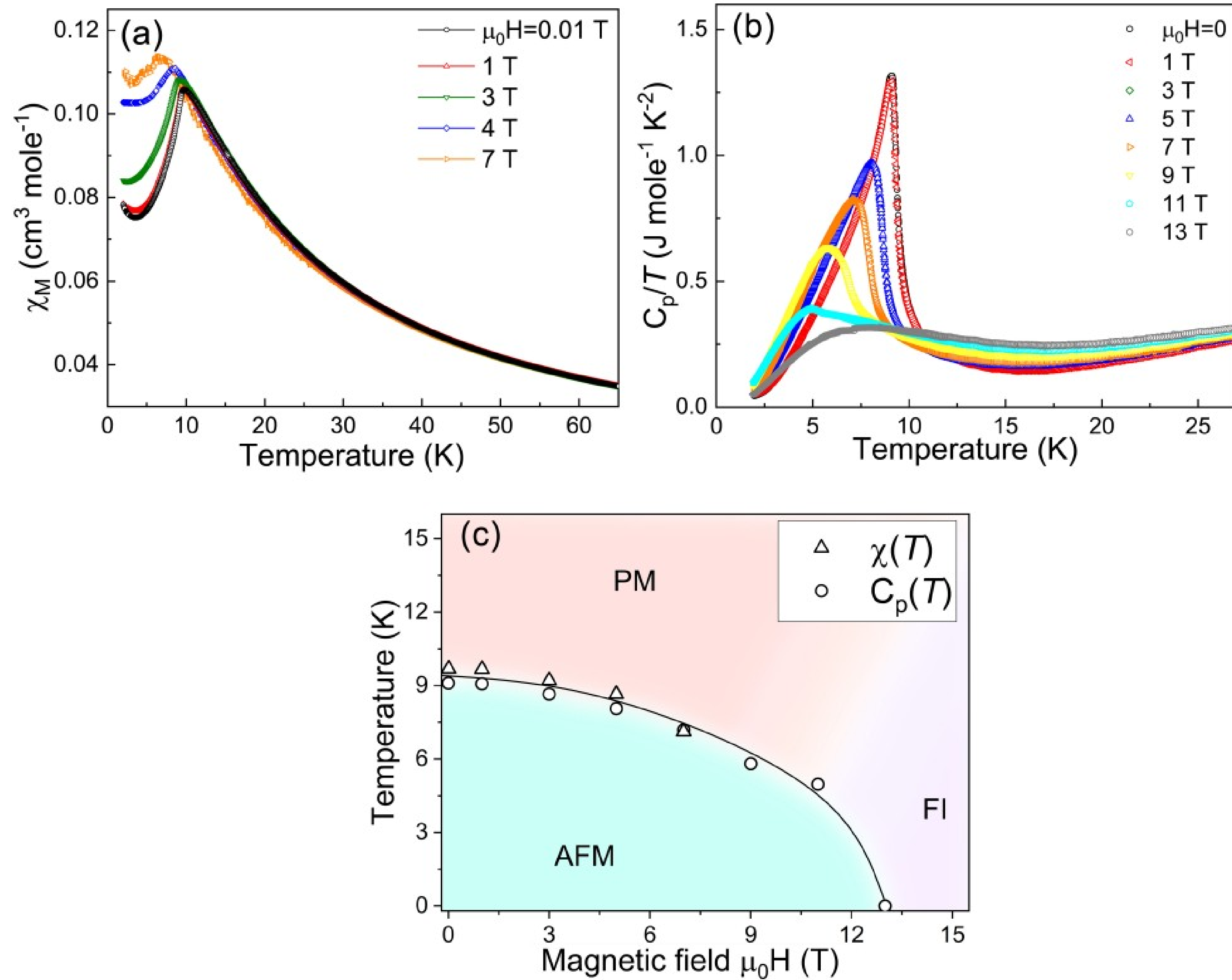


$$\{J_1, \Delta_1, J_{\pm\pm}, J_{z\pm}\} = \{-6.54, 0.36, 0.15, -3.76\} \text{ (meV)}$$

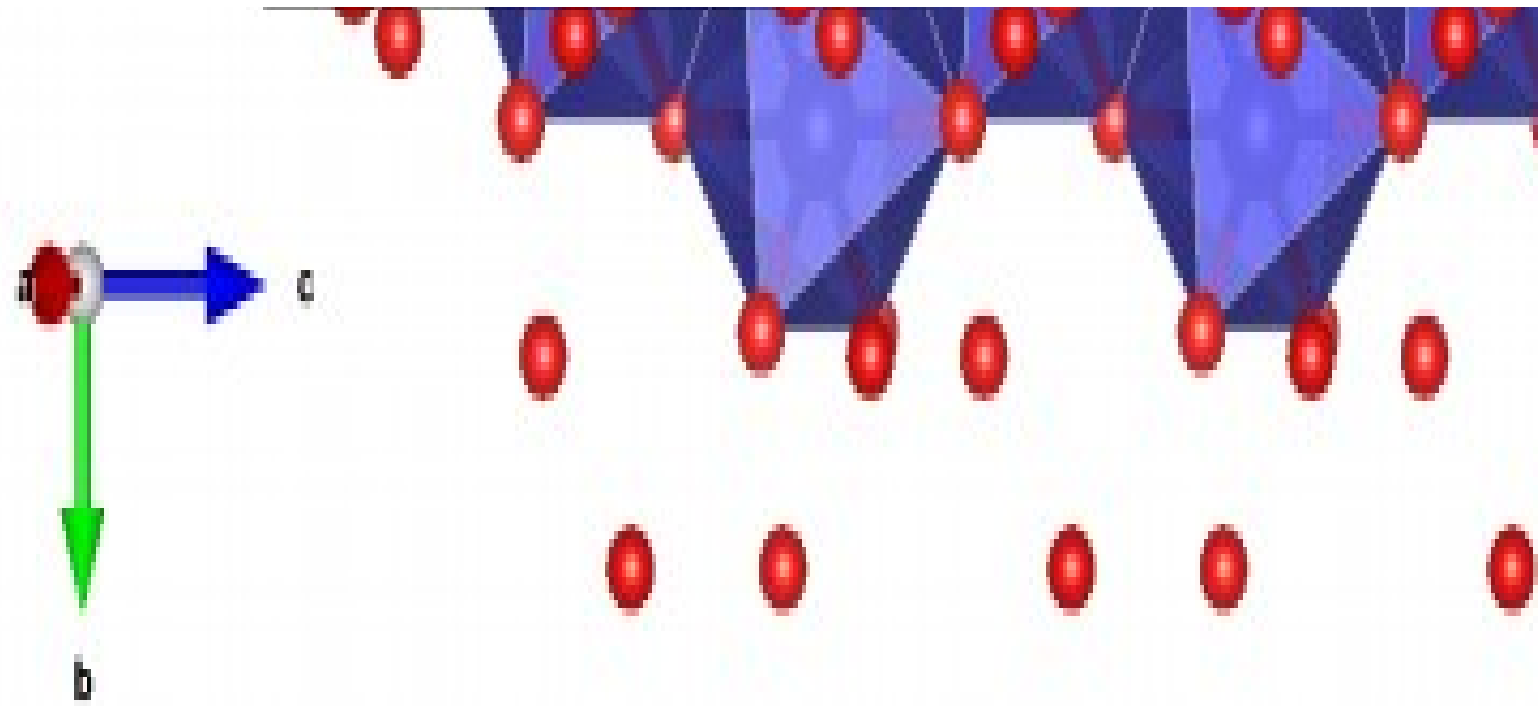
$$\{J_2, J_3, \Delta_3\} = \{-0.21, 1.70, 0.03\} \text{ (meV)}$$



Magnetic order



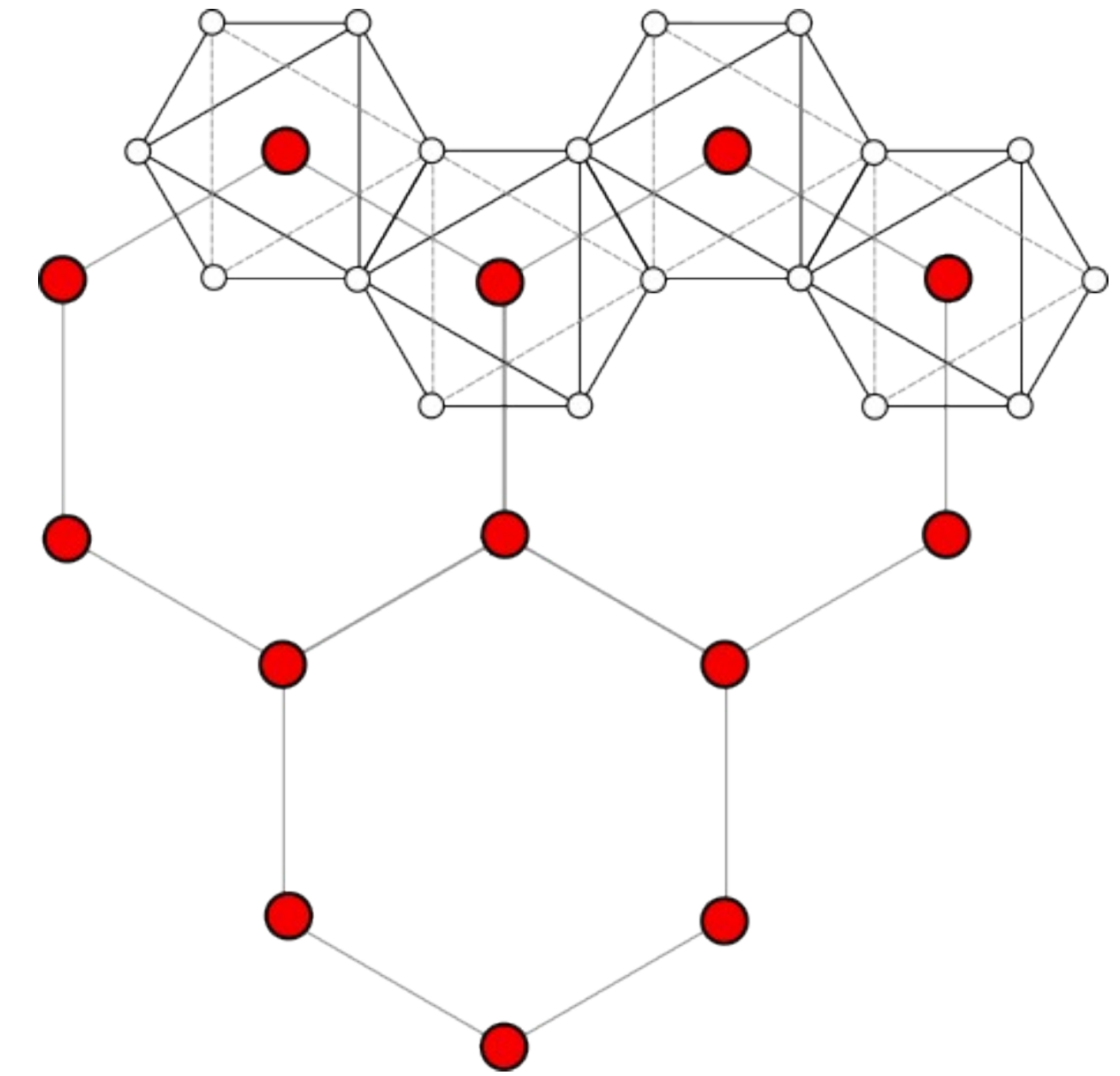
Intra-chain exchanges



Glide symmetry of chains:
c/2 shift+ reflection

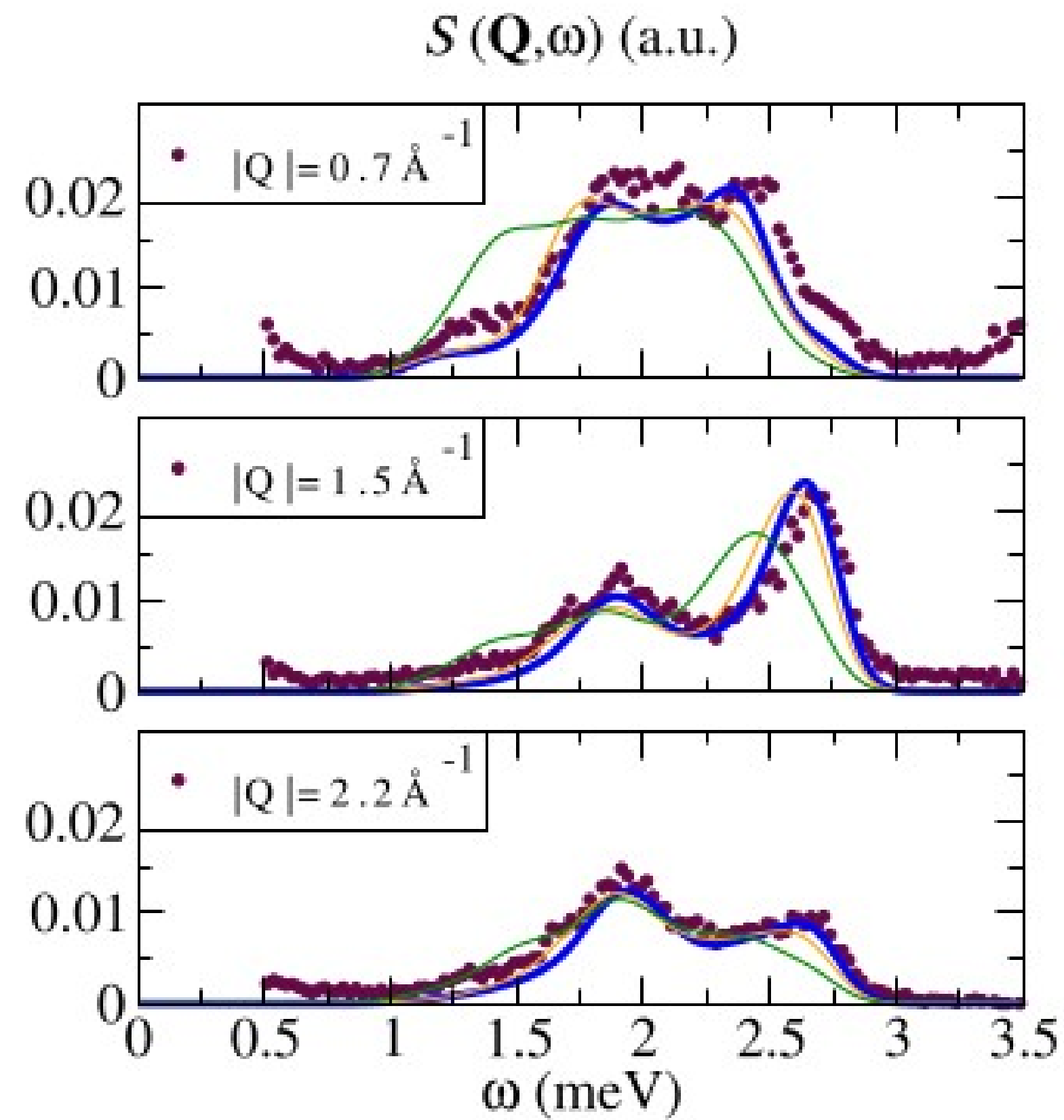
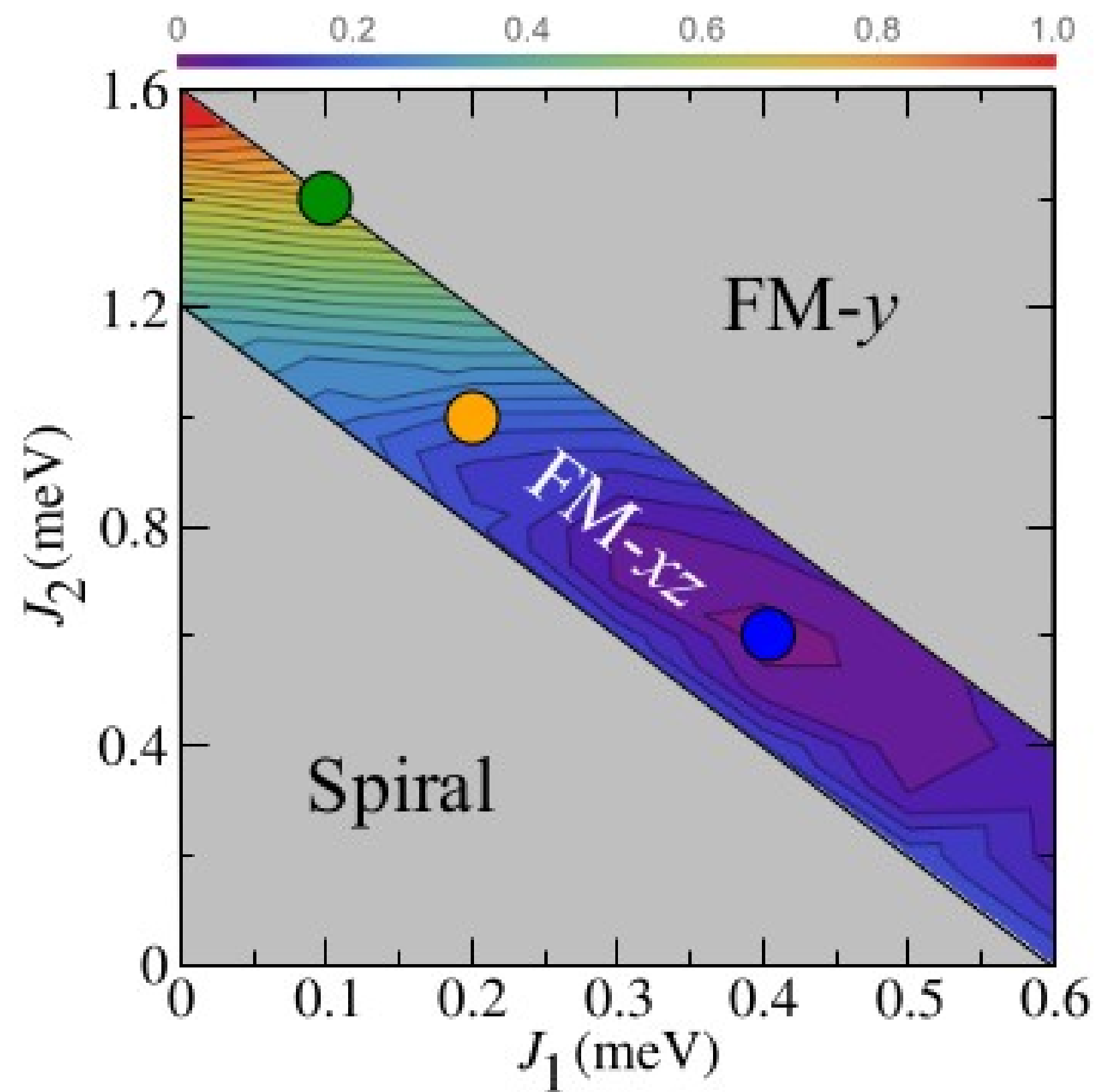
$$J^{(m)} = \begin{pmatrix} J^{xx} & (-1)^m J^{xy} & J^{xz} \\ (-1)^m J^{xy} & J^{yy} & (-1)^m J^{yz} \\ J^{xz} & (-1)^m J^{yz} & J^{zz} \end{pmatrix}$$

Generic model has 6
parameters

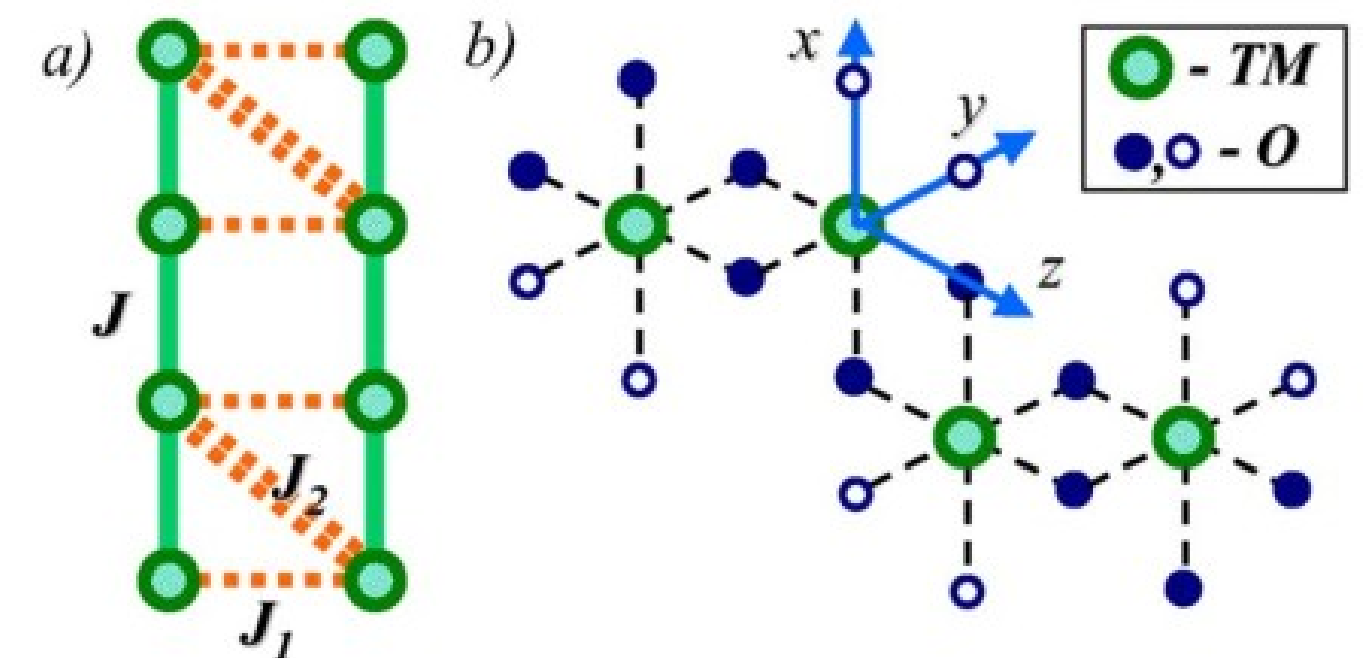
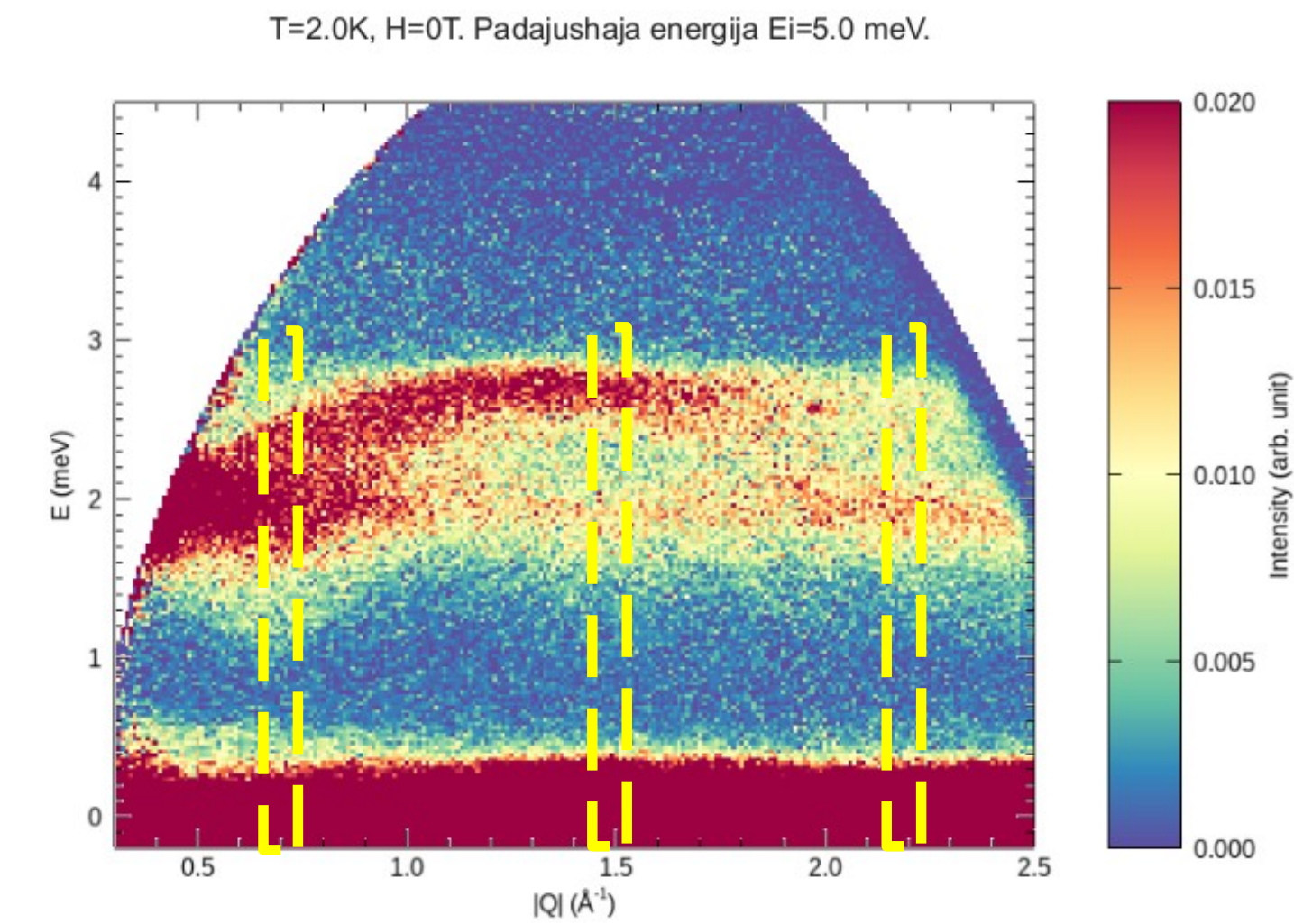


$$\begin{aligned} J_{xx} &= J + \frac{1}{6} (K - 4\Gamma - 2\Gamma'), \\ J_{yy} &= J + K/2 - \Gamma', \\ J_{zz} &= J + \frac{1}{3} (K + 2\Gamma + 4\Gamma'), \\ J_{xy} &= \frac{1}{2\sqrt{3}} (K + 2\Gamma - 2\Gamma'), \\ J_{xz} &= \frac{1}{3\sqrt{2}} (-K + \Gamma - \Gamma'), \\ J_{yz} &= -\frac{1}{\sqrt{6}}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

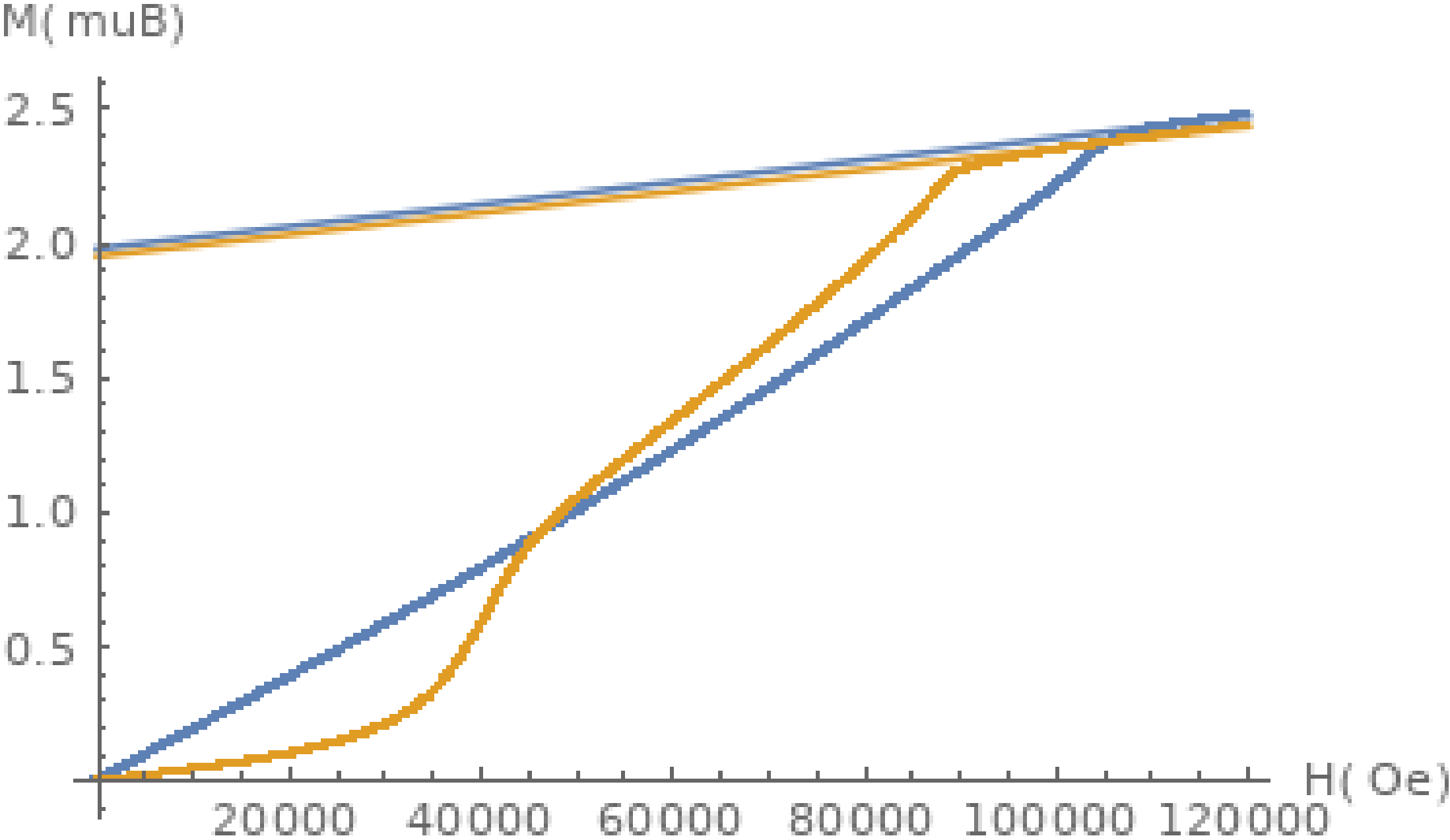
Fit quality



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



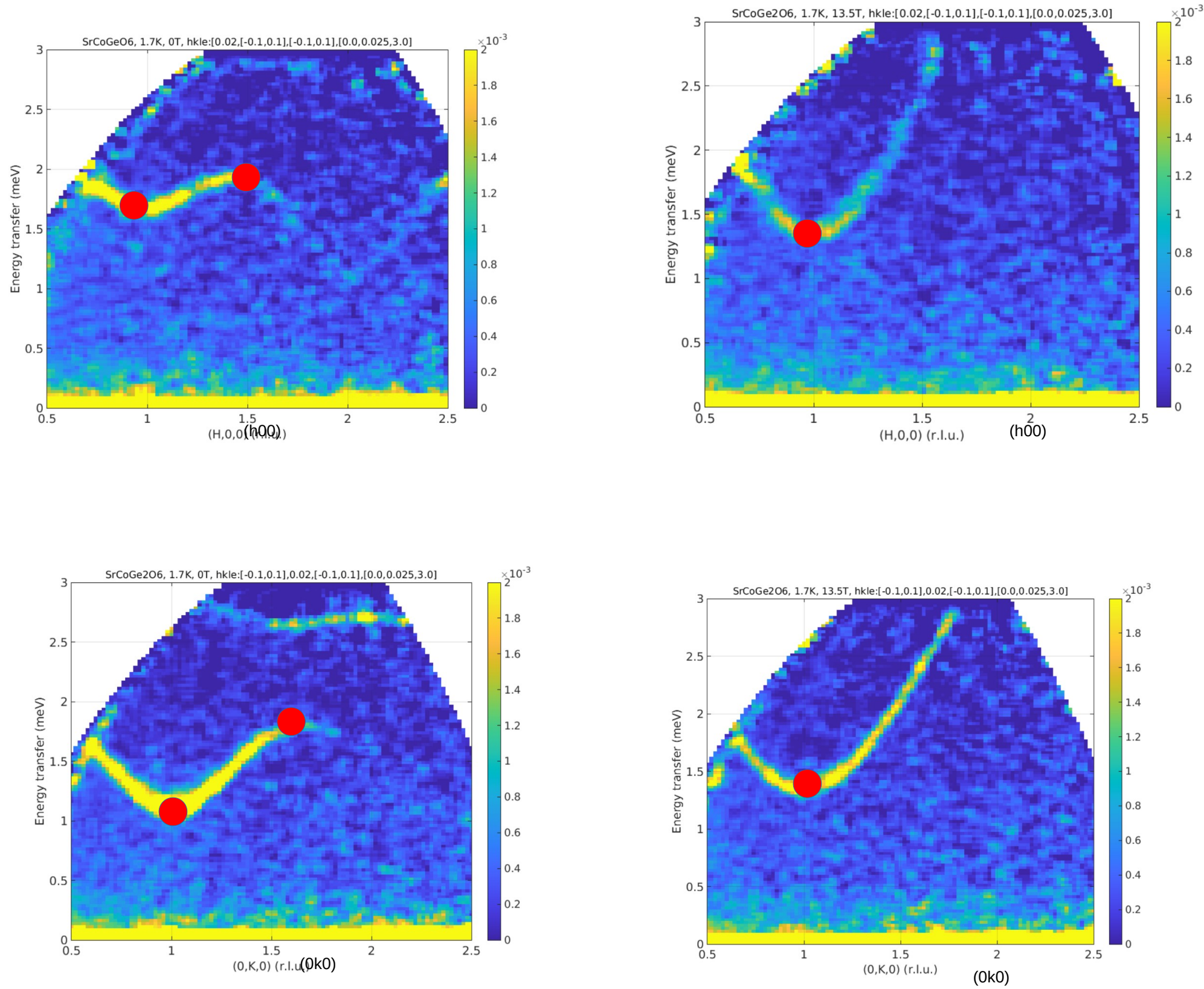
First step: Fitting magnetization



$H \parallel a$
 $H \parallel c$

$g_a=3.96$
 $g_c=3.9$
Need to correct for Van Vleck contribution

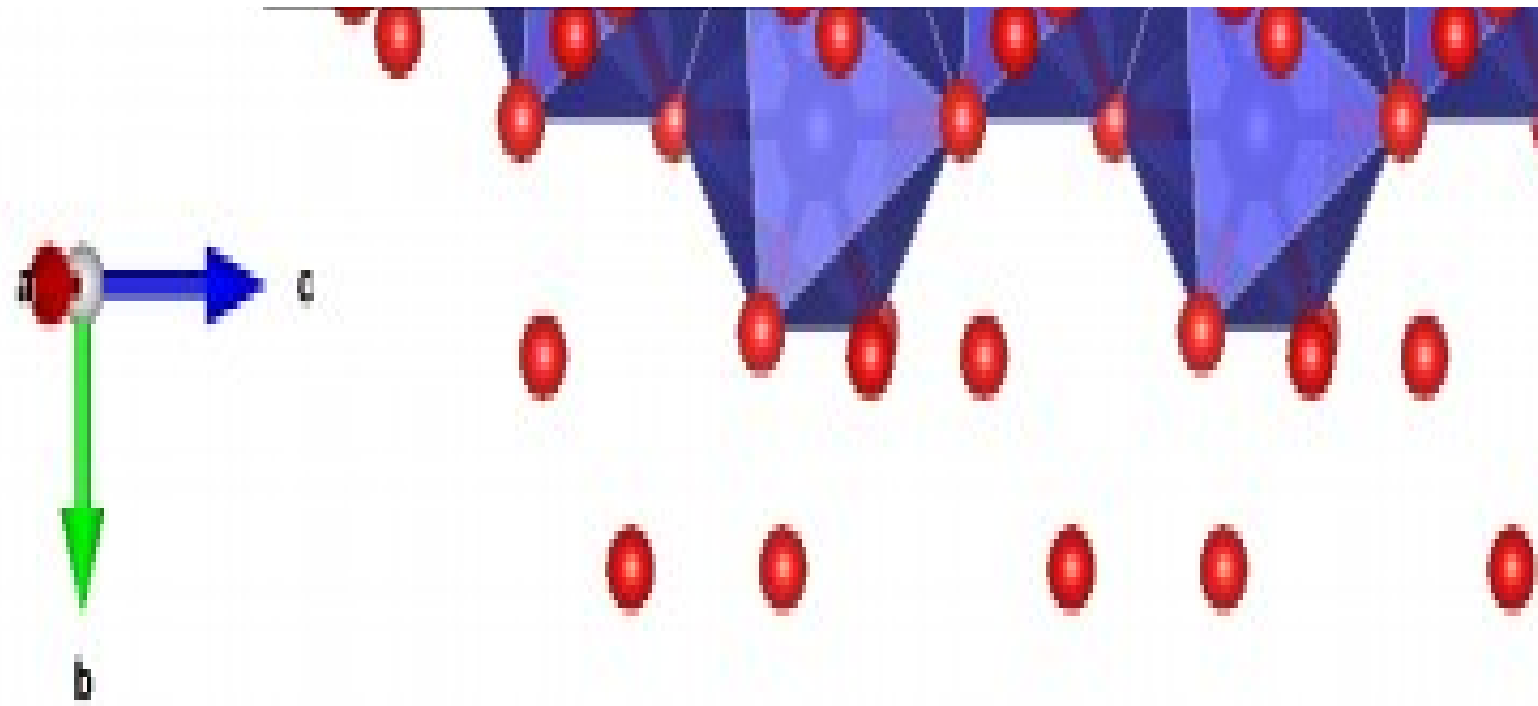
Fitting procedure



- 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

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

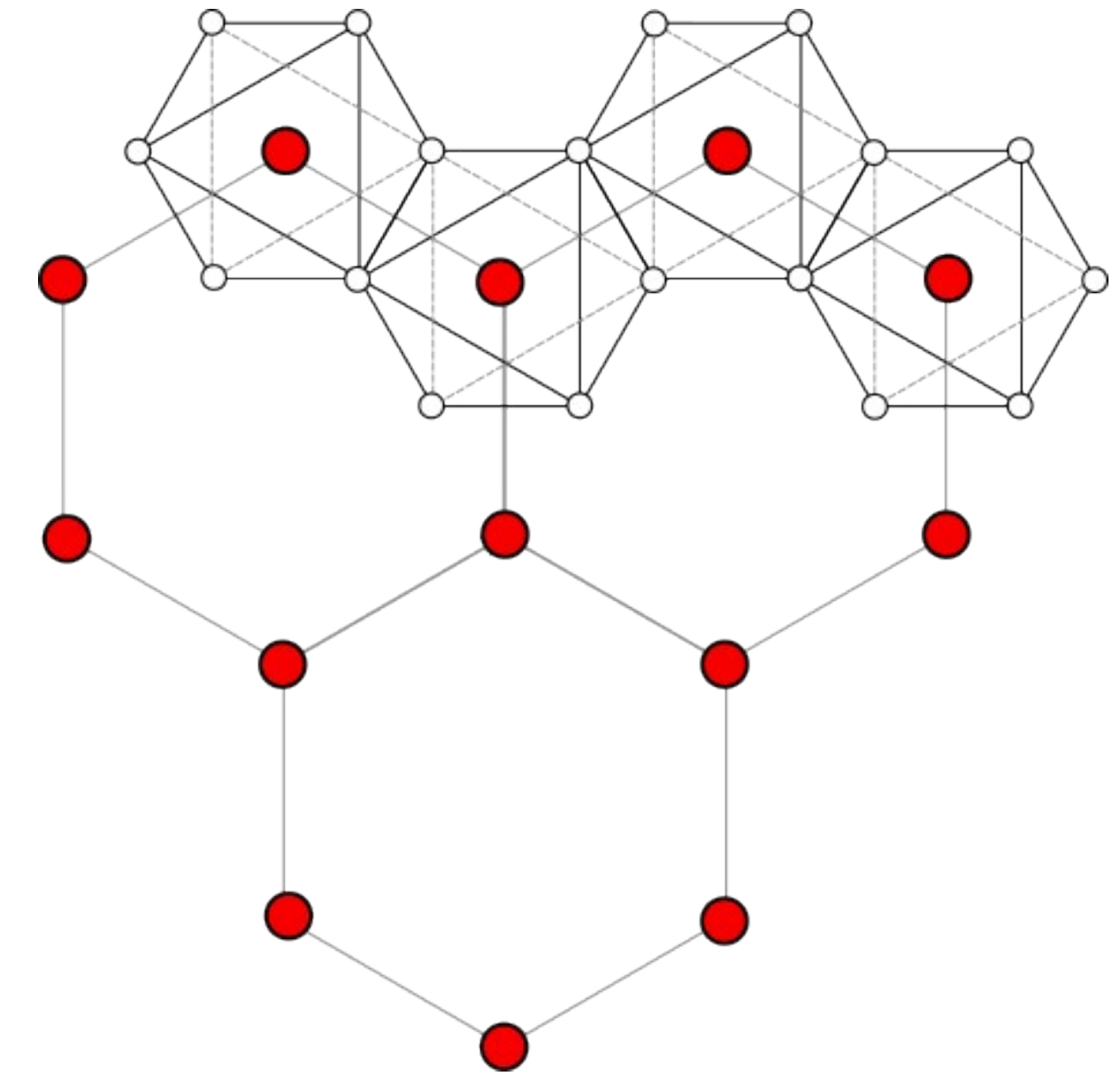
Intra-chain exchanges



Glide symmetry of chains:
c/2 shift+ reflection

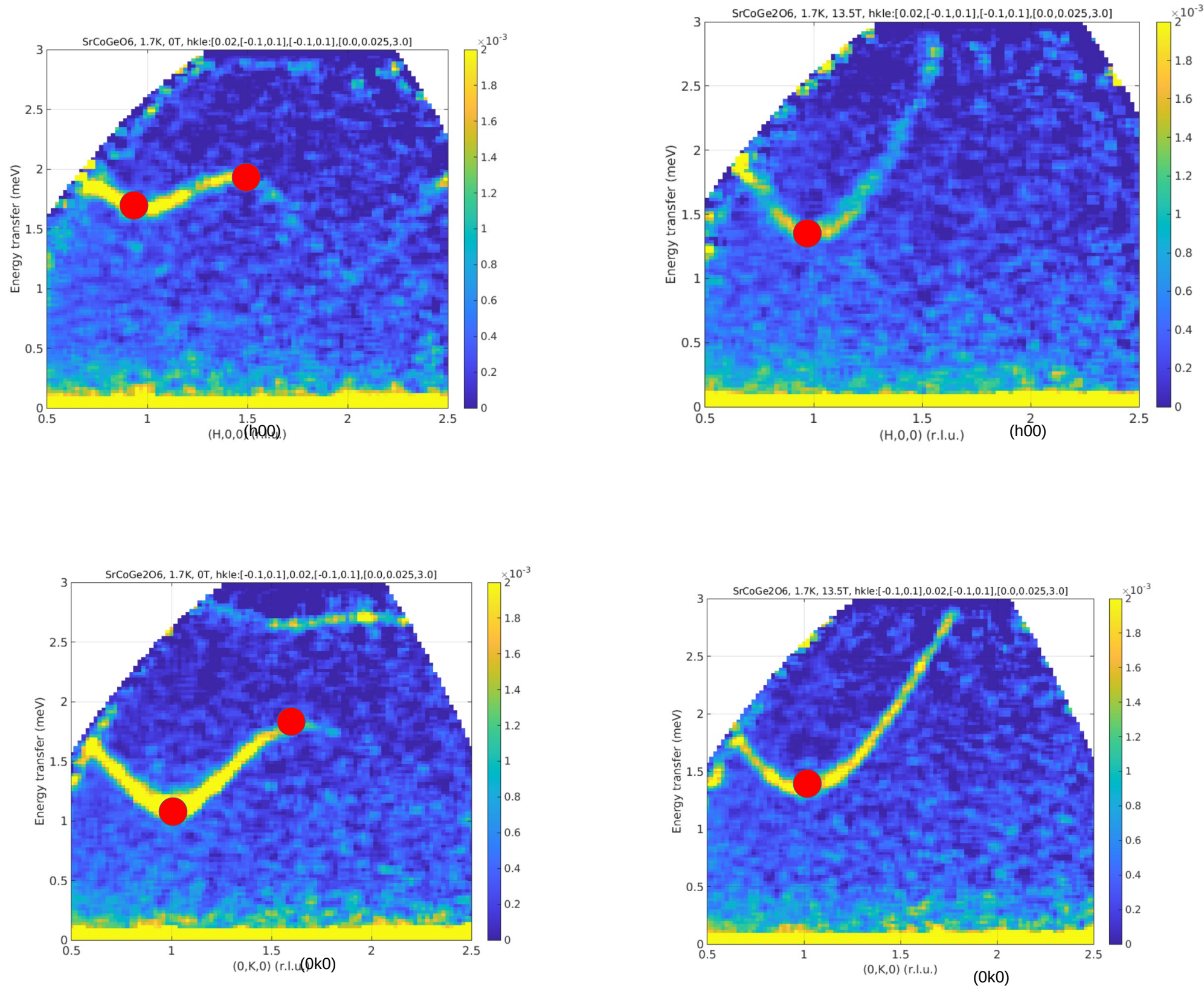
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Generic model has 6
parameters



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

Fitting procedure



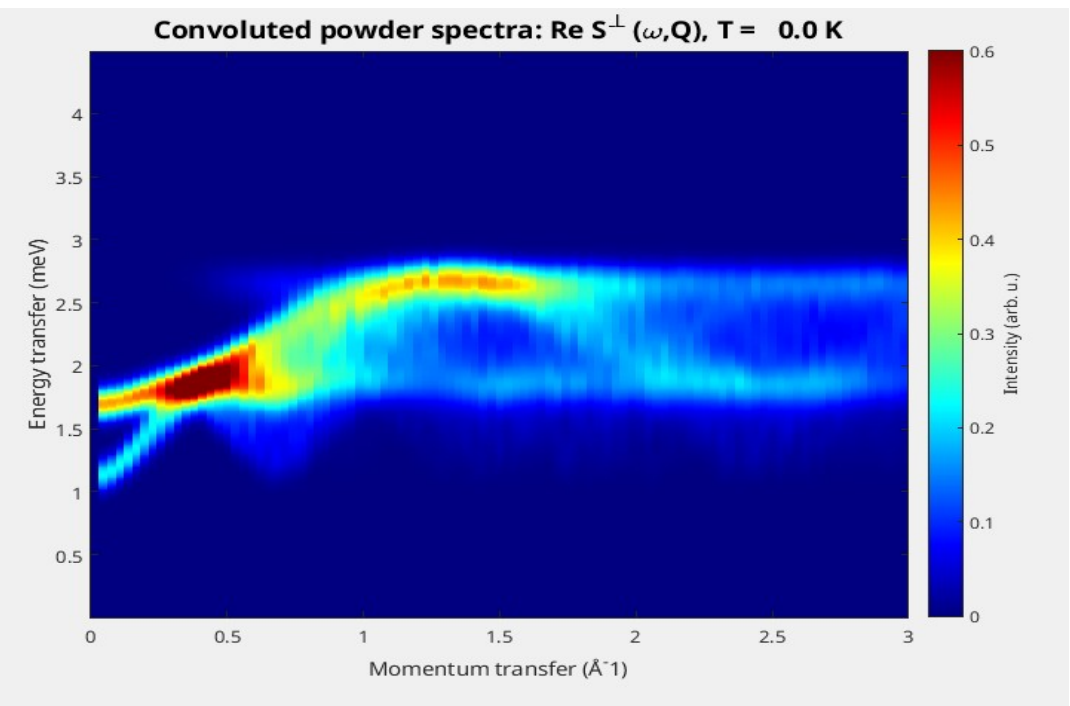
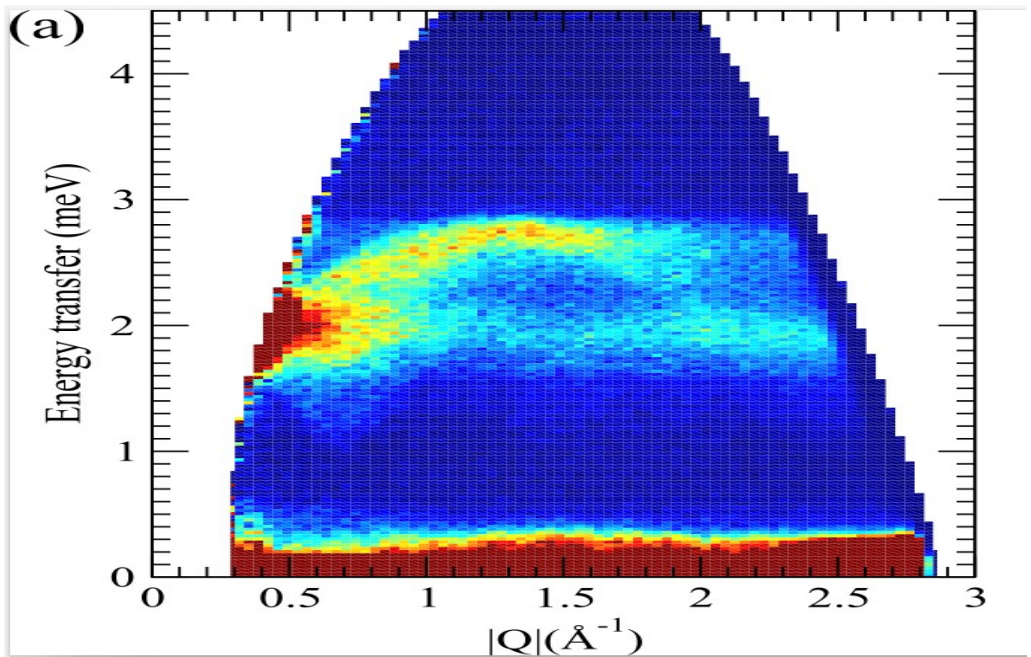
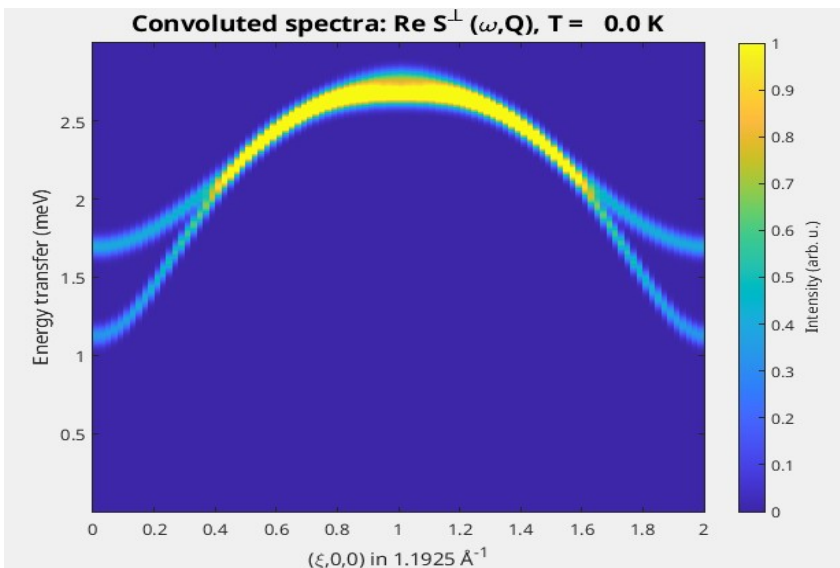
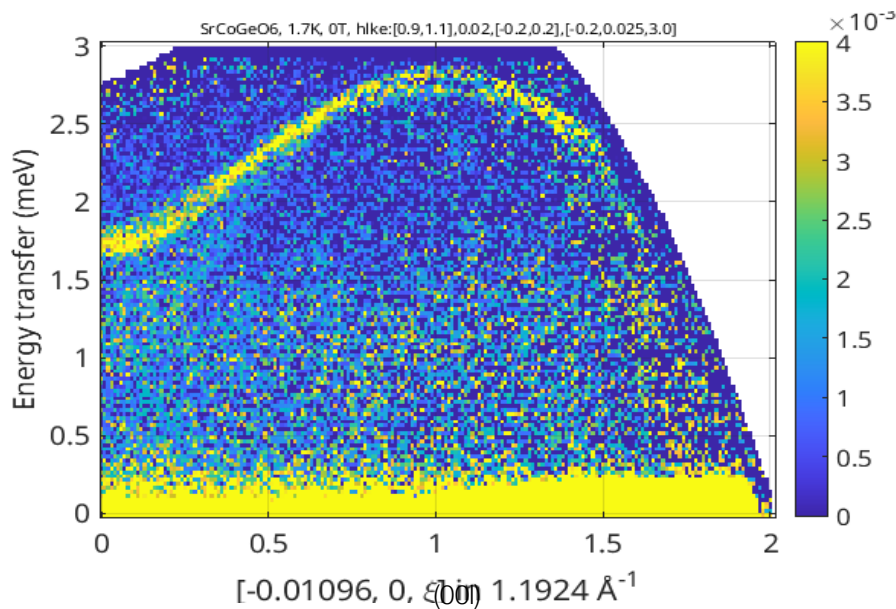
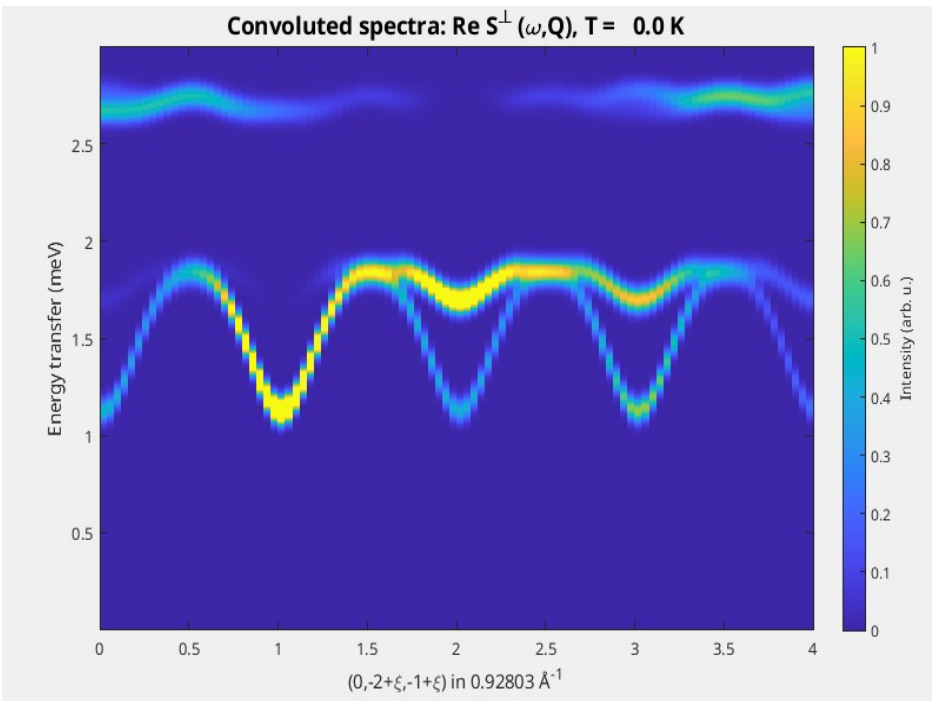
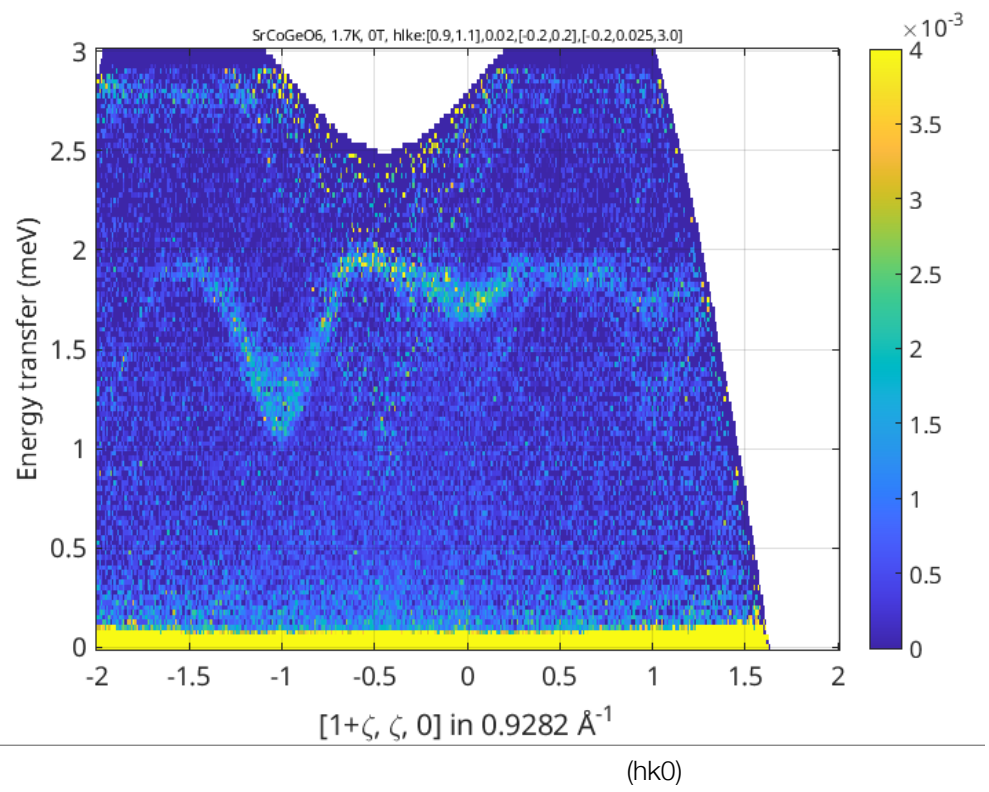
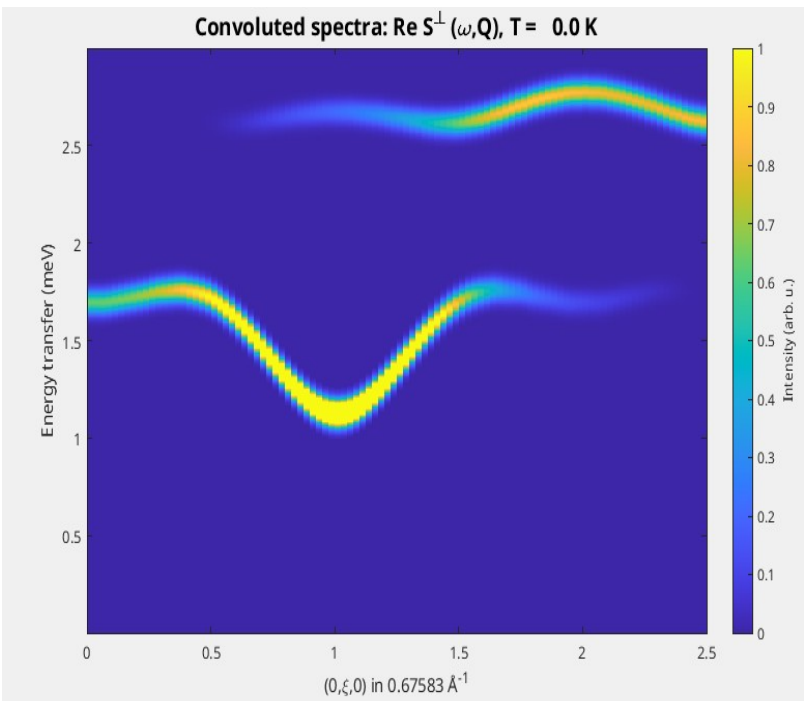
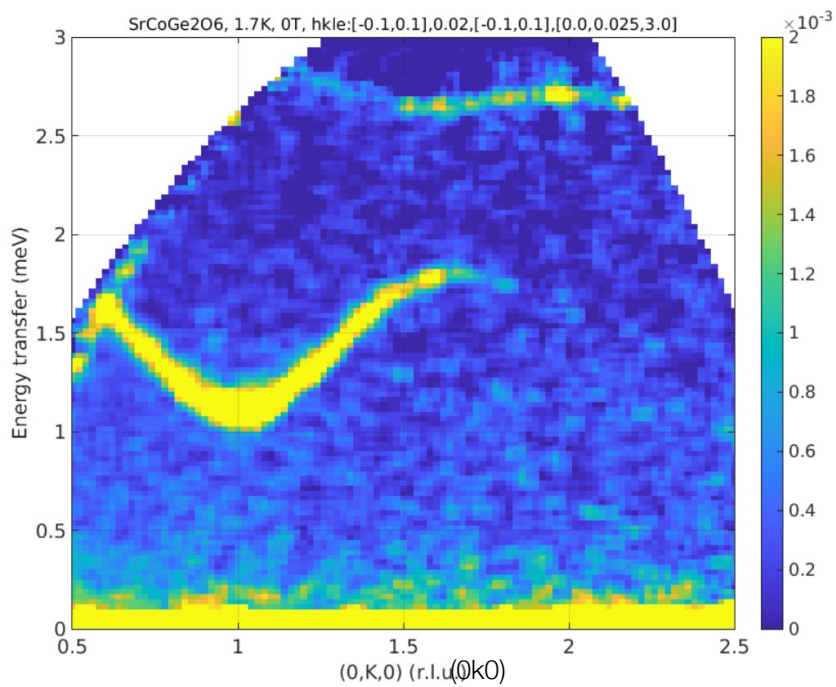
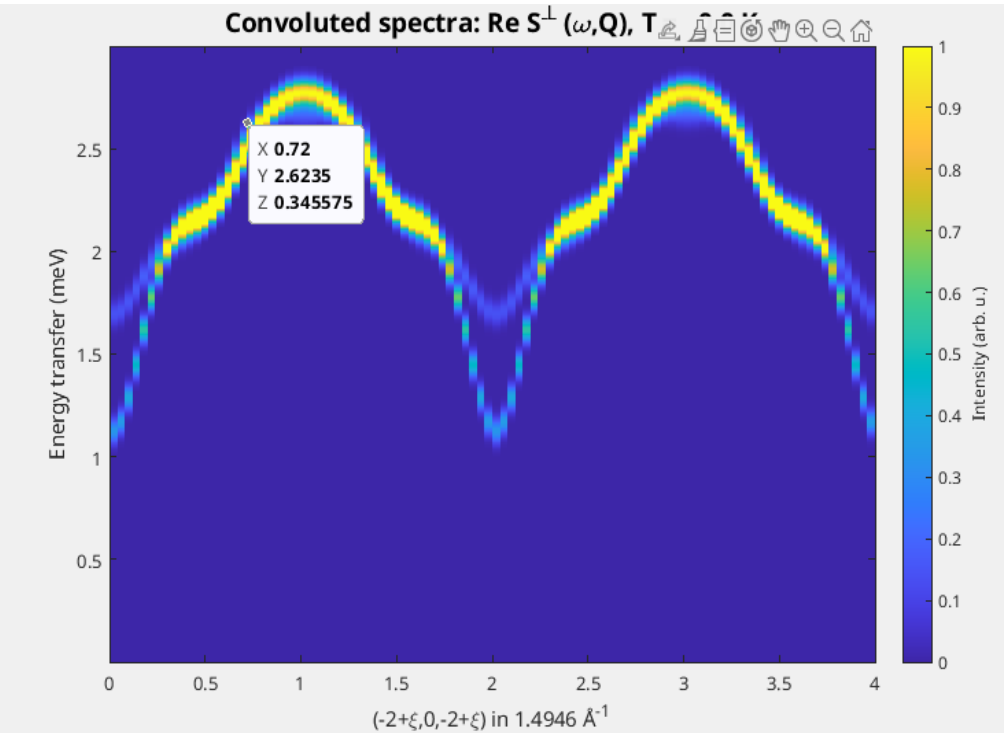
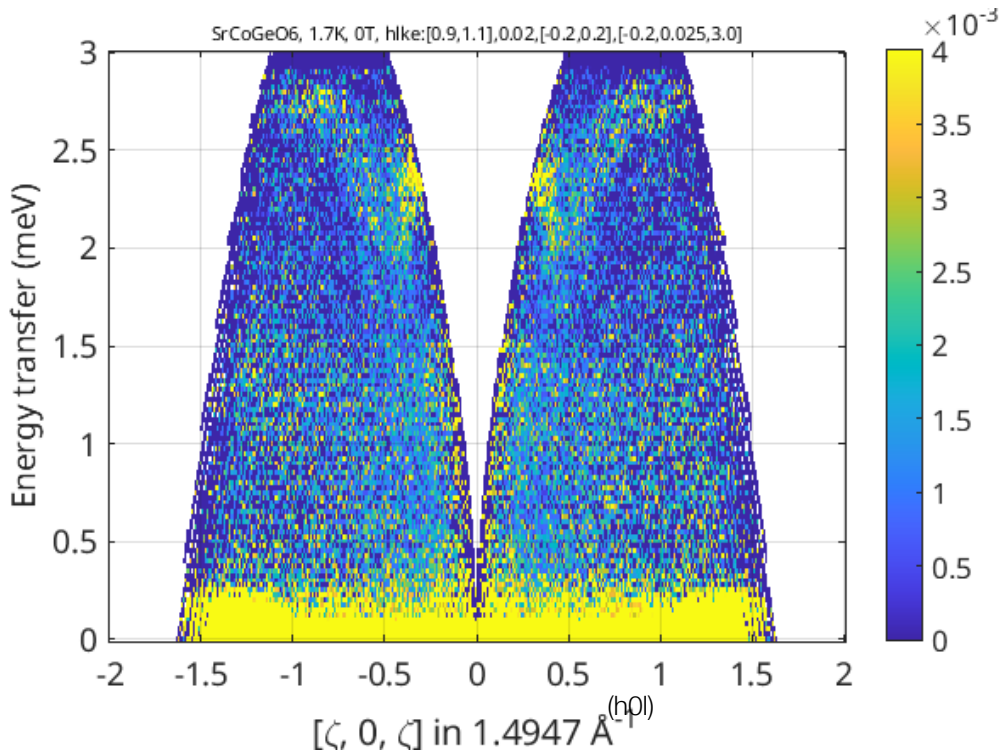
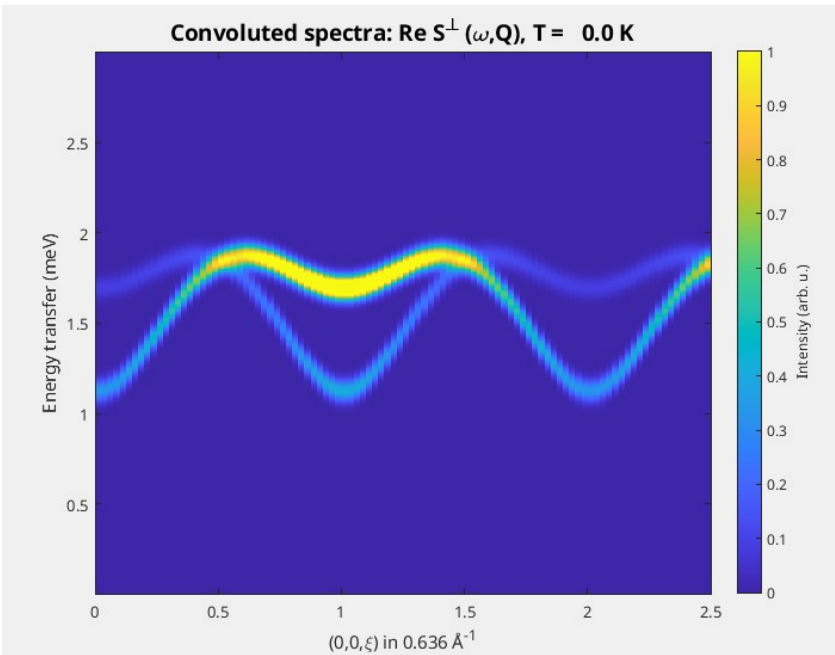
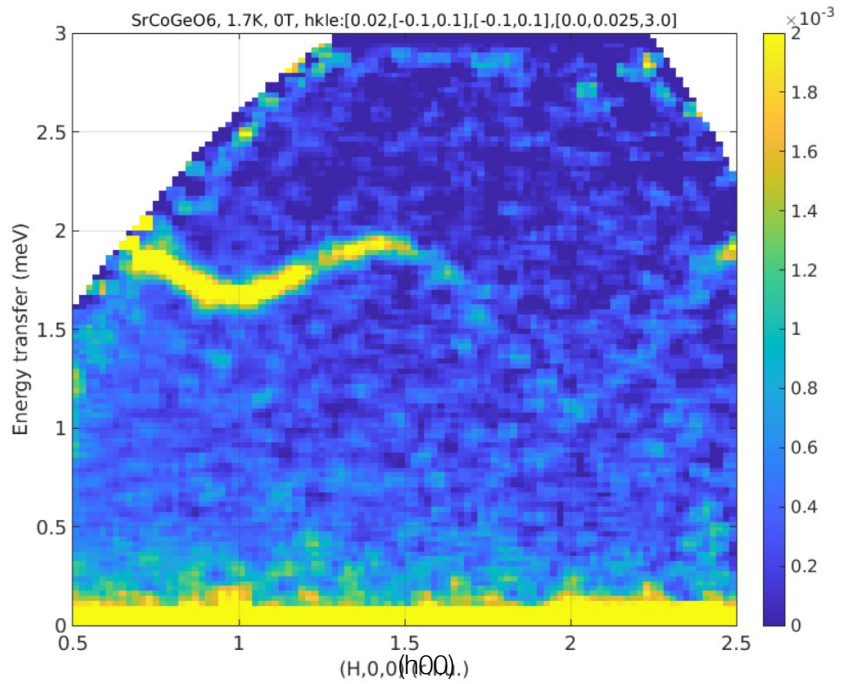
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$$\tan 2\theta = \frac{2J^{xz}}{J^{xx} - J^{zz}}$$

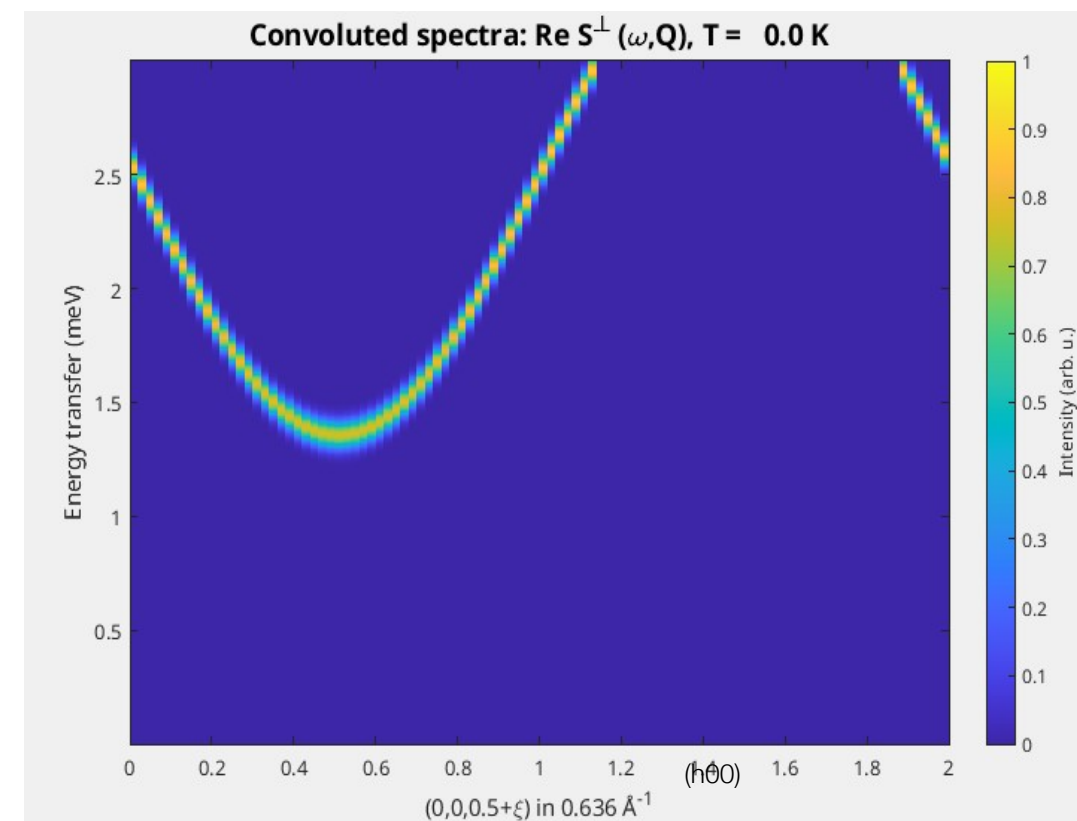
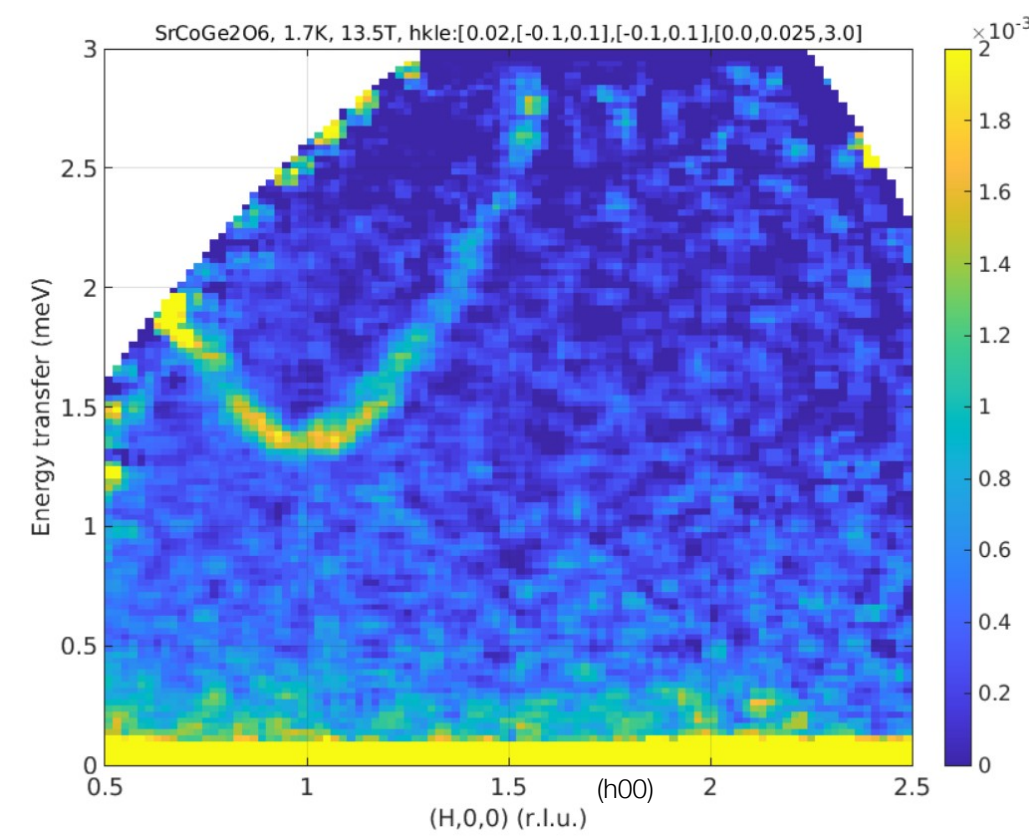


$$\hat{J}_{\text{chain}} = \begin{pmatrix} -1.12 & 0 & 0.13 \\ 0 & -0.1 & 0 \\ 0.13 & 0 & -0.85 \end{pmatrix}$$

J1=0.2 meV, J2=0.8 meV



H=13.5T, H||c



H=8T, H||b

