Ferromagnets, antiferromagnets and magnets in-between

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Heisenberg model $\mathcal{H} = J \sum \boldsymbol{S}_i \boldsymbol{S}_j$

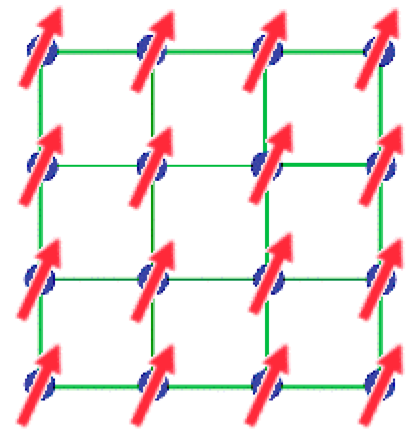
$J < 0$ ferromagnet: Macroscopic magnetization

$\boldsymbol{S}^{\mathrm{tot}} = \sum \boldsymbol{S} = N \boldsymbol{S}$

$[S_x^{\mathrm{tot}}, S_y^{\mathrm{tot}}] = i \hbar S_z^{\mathrm{tot}} \rightarrow [S_x, S_y] = i (\hbar/N) S_z$

Quantum fluctuations are suppressed

$[\mathcal{H}, S_z^{\mathrm{tot}}] = 0 \ ; \ [\mathcal{H}, \boldsymbol{S}^{\mathrm{tot}\,2}] = 0$

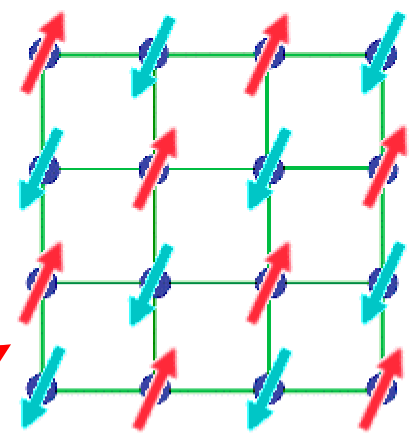


Macroscopic $\leftrightarrow$ no quantum effects

$J > 0$ antiferromagnets: Two sublattices

$\boldsymbol{S}^{\mathrm{tot}}{}_A = \sum_A \boldsymbol{S} = (N/2)\, \boldsymbol{S} \quad \boldsymbol{S}^{\mathrm{tot}}{}_B = \sum_B \boldsymbol{S} = (N/2)\, \boldsymbol{S}$

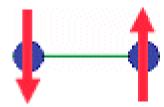
$[\mathcal{H}, \boldsymbol{S}^{\mathrm{tot}}] \neq 0$



Macroscopic $\leftrightarrow$ quantum state

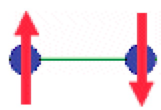
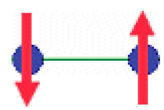
Cannot be the ground state!

Heisenberg "antiferromagnet": two spins

Two electrons  $E = -\frac{1}{4} J$

This is not rotation invariant, although $S^{tot}=0$

We can do better! Coherent superposition:

$1/\sqrt{2}($  -  $)$ $E = -\frac{3}{4} J$

How can we do this for more than two spins? Not trivial...

Valence bond solid (Sachdev,Read 1990)

Resonating valence bond (Anderson, Fazekas, 1974, Anderson, 1987)

Stability of Néel state

Magnon excitation spectrum:

$\omega(q) = JS\sqrt{(1-\cos 2(qa))} \sim q$ for small q

Quantum fluctuations suppress sublattice magnetization (Anderson, 1952)

$S_A^{tot}/N = ½(S - \delta S)$

$$\delta S \approx \frac{1}{2S}\frac{1}{N}\sum_{\mathbf{R}}\left\langle S_{\mathbf{R}}^{-}S_{\mathbf{R}}^{+}\right\rangle = \frac{1}{2S}\int\frac{d^3\mathbf{Q}}{v_{BZ}}\frac{g^{-+}(\mathbf{Q})}{\hbar\omega(\mathbf{Q})} \sim \int g(q)/\omega(q)\,\mathrm{d}^D q$$

Quantum correction depends on dimensionality D, will diverge in one dimension if there is no gap in magnon spectrum

ESR measures the q=0 magnon

Stabilize the Néel state: anisotropy

$$\mathcal{H} = \sum J_x(\boldsymbol{r}) S_{\boldsymbol{m}}{}^x S_{\boldsymbol{m+r}}{}^x + J_y(\boldsymbol{r}) S_{\boldsymbol{m}}{}^y S_{\boldsymbol{m+r}}{}^y + J_z(\boldsymbol{r}) S_{\boldsymbol{m}}{}^z S_{\boldsymbol{m+r}}{}^z$$

Single ion anisotropy:

$$\mathcal{H}_{\mathrm{A}} = \sum D_x (S_m{}^x)^2 + D_y (S_m{}^y)^2 + D_z (S_m{}^z)^2$$

Exchange anisotropy:

$$\mathcal{H}_{\mathrm{EA}} = \sum J(S_j{}^x S_{j+1}{}^x + S_j{}^x S_{j-1}{}^x + S_j{}^y S_{j+1}{}^y + S_j{}^y S_{j-1}{}^y + S_j{}^z S_{j+1}{}^z + S_j{}^z S_{j-1}{}^z)$$

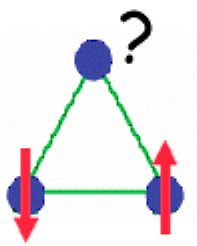
Dzyaloshinski-Moriya $\boldsymbol{D}(\boldsymbol{S}_1 \mathrm{x} \boldsymbol{S}_2)$ term:

$$\mathcal{H}_{\mathrm{D}} = \sum D(S_j{}^x S_{j+1}{}^z - S_j{}^z S_{j+1}{}^x + S_j{}^x S_{j-1}{}^z - S_j{}^z S_{j-1}{}^x)$$

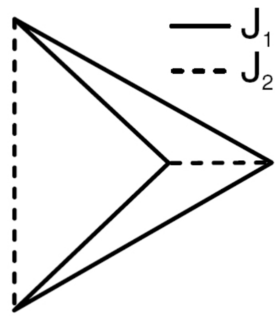
All are due to spin orbit coupling
Leads to gap in magnon spectrum

Kill the Néel state: Frustration

Geometric: Third spin unhappy

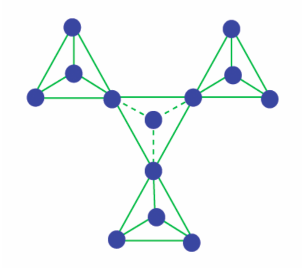


$Cu_2Te_2O_5Cl_2$

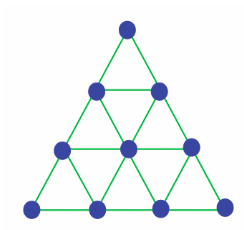


Pyrochlore, spinel

$Ho_2Ti_2O_7$ $ZnZr_2O_7$

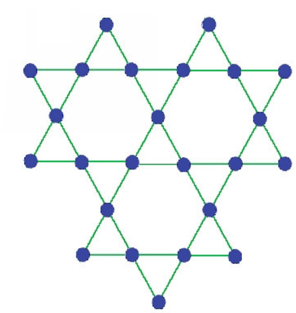


$CsCoCl_3$, $NaNiO_2$



Kagomé

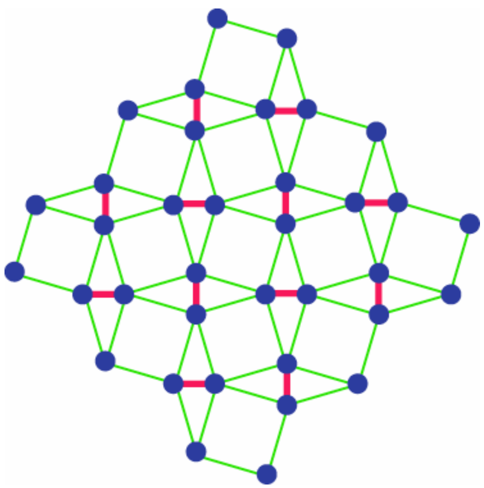
$KFe_3(OH)_6(SO_4)_2$



Kill the Néel state: Interaction beyond first neighbor

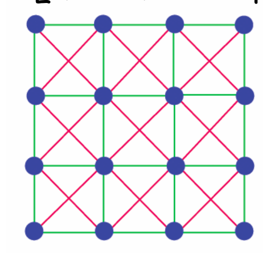
Often leads to helical order, incommensurate wavelength

$SrCu_2(BO_3)_2$



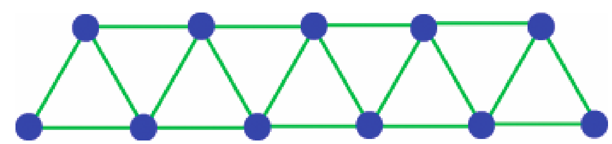
J_1-J_2

$Li_2(VO)SiO_4$,



cikk-cakk lánc

$LiCu_2O_4$



Search for the RVB ground state

Excitations in Neel state: spin 1 magnons

$$\psi_l = S_l^+|0>$$

$$\psi_{\mathbf{k}} = \frac{1}{\sqrt{2S}}\frac{1}{N}\sum_l \mathrm{e}^{\mathbf{ikR}_l} Sl^+|0>$$

Excitations in RVB state: spin ½ spinons

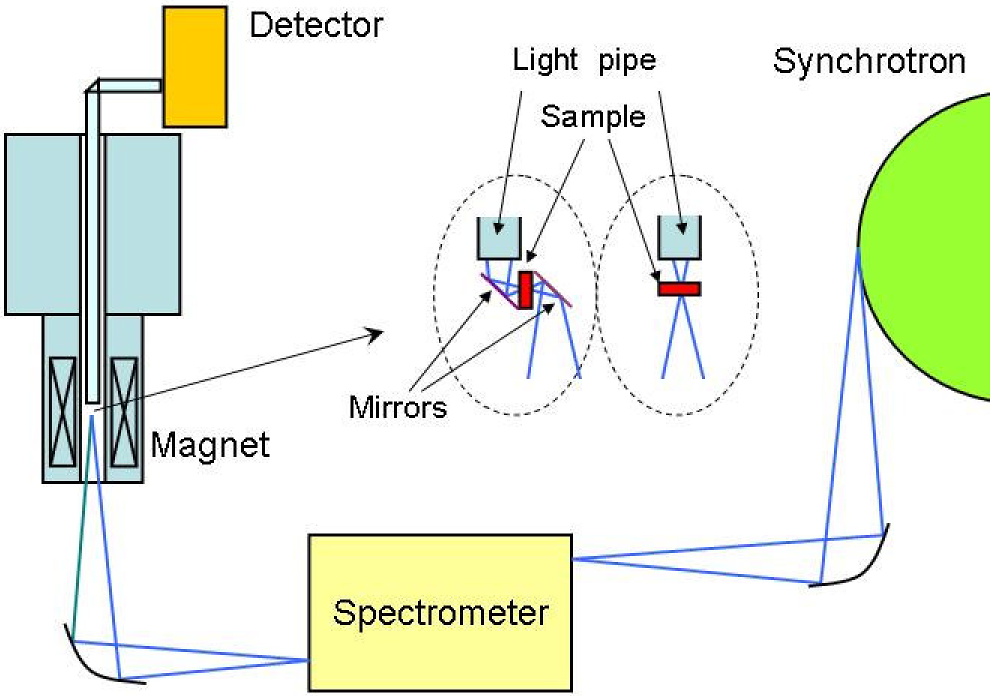
Soliton (domain wall) in one dimension

Examples:
Cs_2CuCl_4 Triangular plane J, J'
$LiCu_2O_2$ Triangular chain

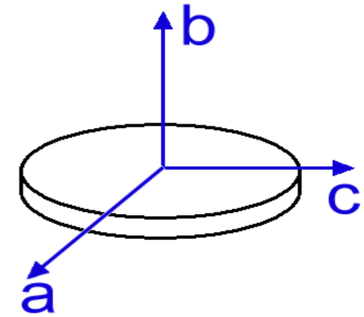
Also:

$NaNiO_2$	Triangular, ferromagnet/antiferromagnet
$Ni_5(TeO_3)_4Cl_2$	Complex unit cell

Instrument: Magnet & Spectrometer



Sample:



Incident light always along b

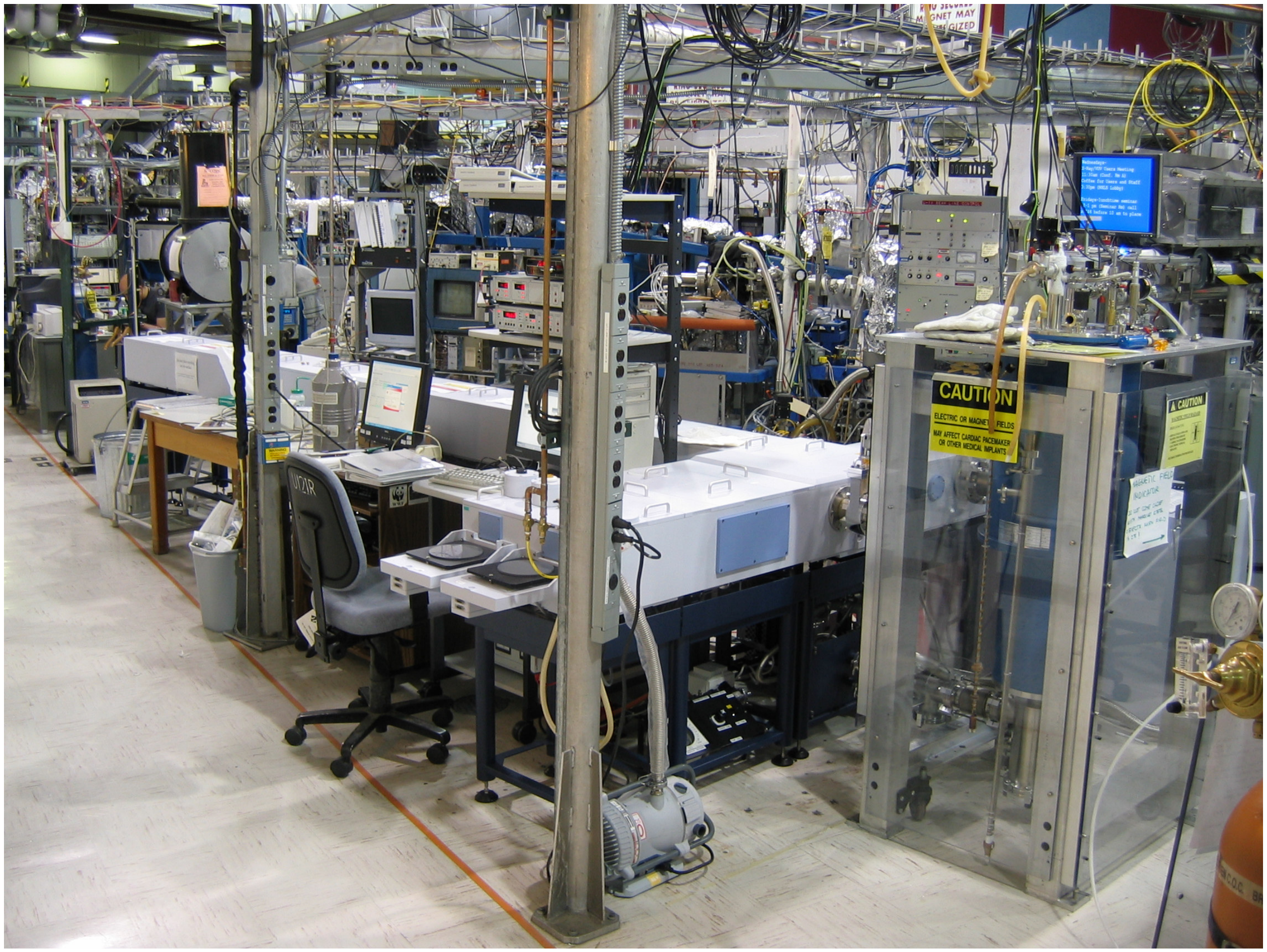
External field can point parallel or perpendicular to plane: along a, or b or c

Polarization: along a or c

Measure spectrum at many fixed fields.

Map the ESR absorption over the whole range of fields/frequencies

Brookhaven Lab, National Synchrotron Light Source, U12IR



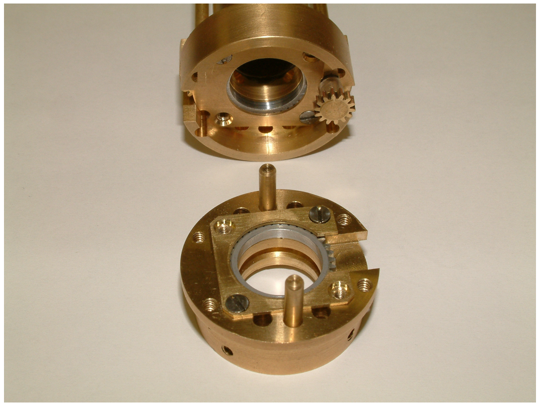
14/16 Tesla

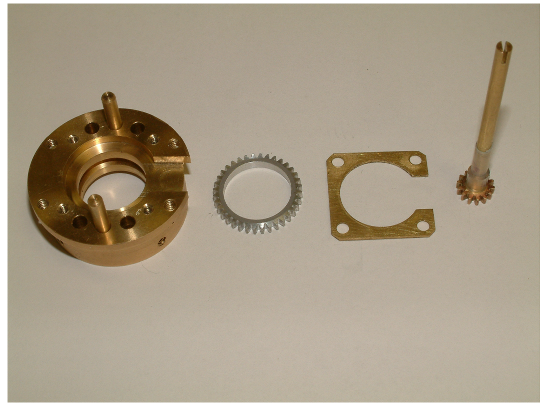
8 - 200 cm-1
(Up to visible)

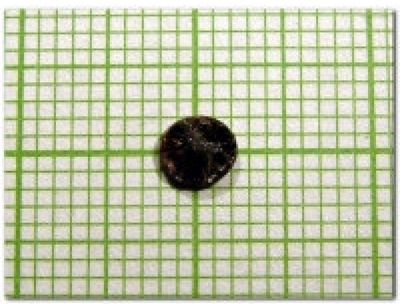
1.8K-300K

Transmission

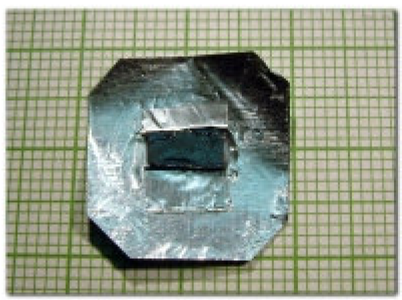
Samples and sample holders







$LaMnO_3$

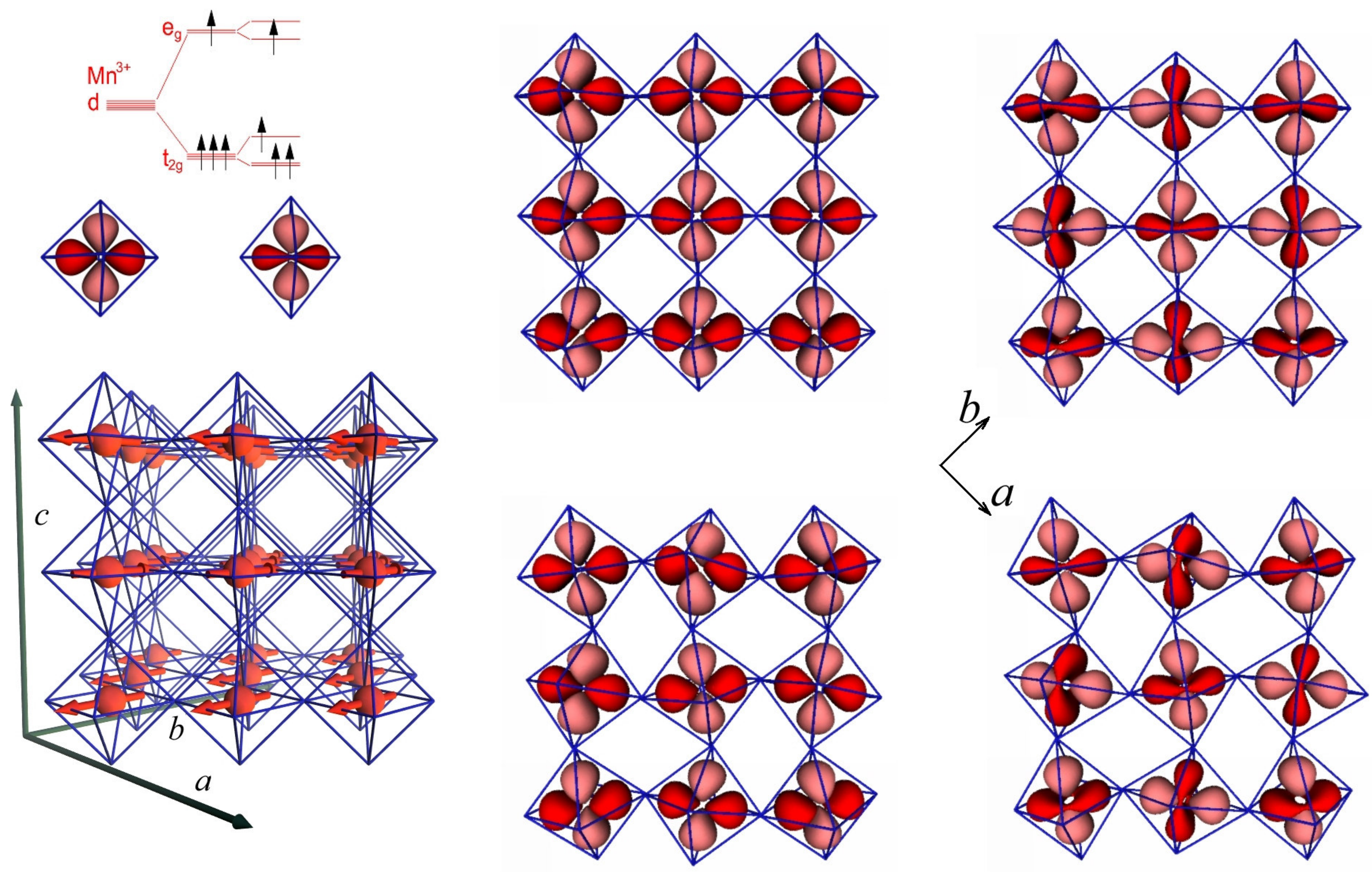


$LiCu_2O_2$

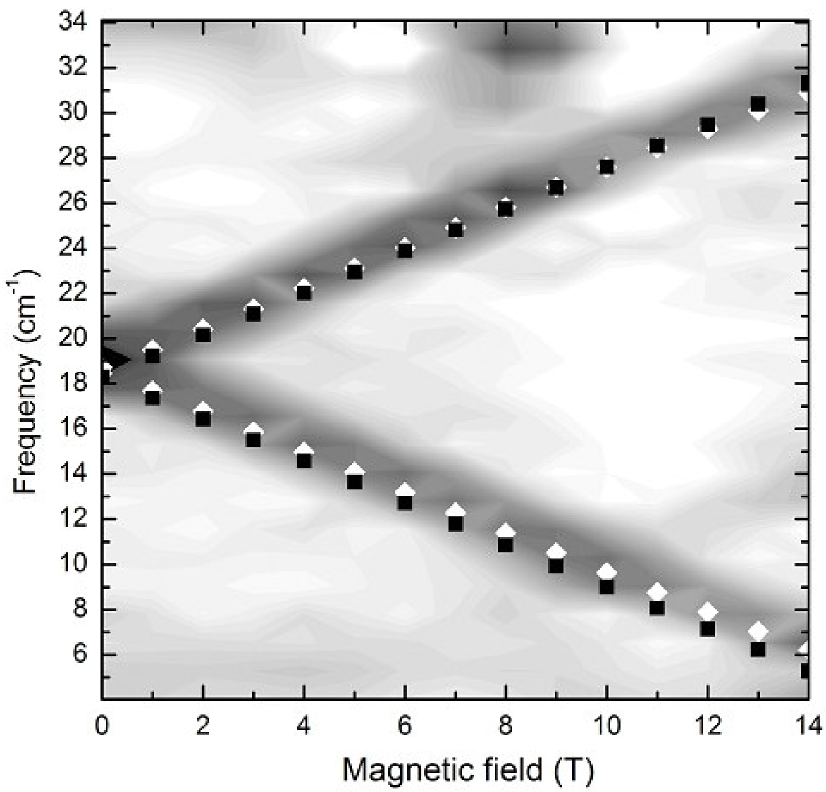
$Ni_5(TeO_3)_4Cl_2$

$Cu_2Te_2O_5Cl_2$

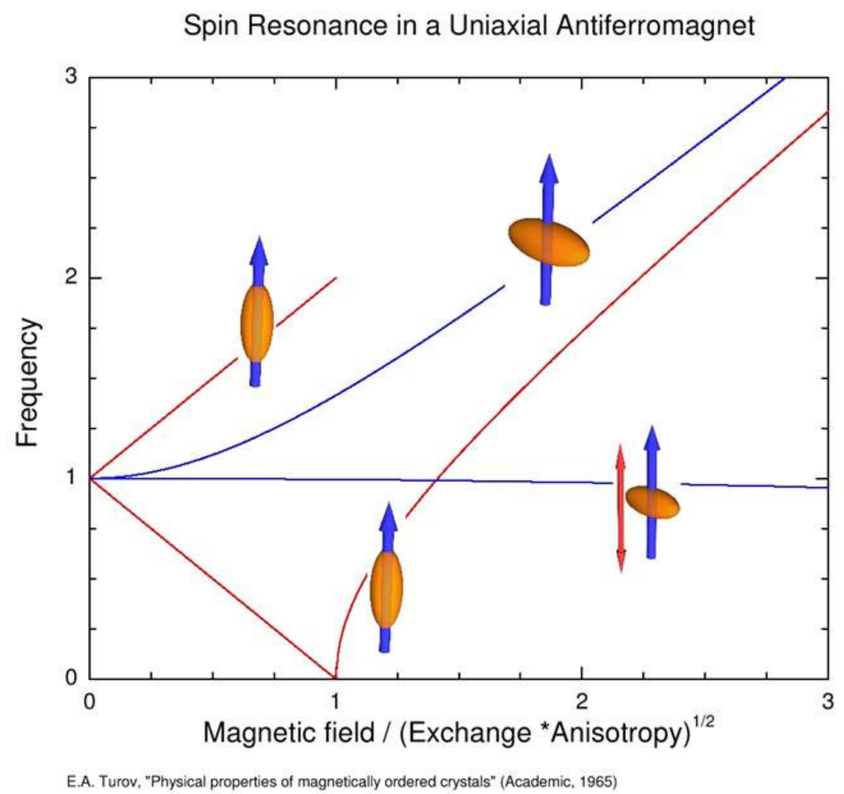
$LaMnO_3$: “simple” antiferromagnet



$LaMnO_3$: ESR

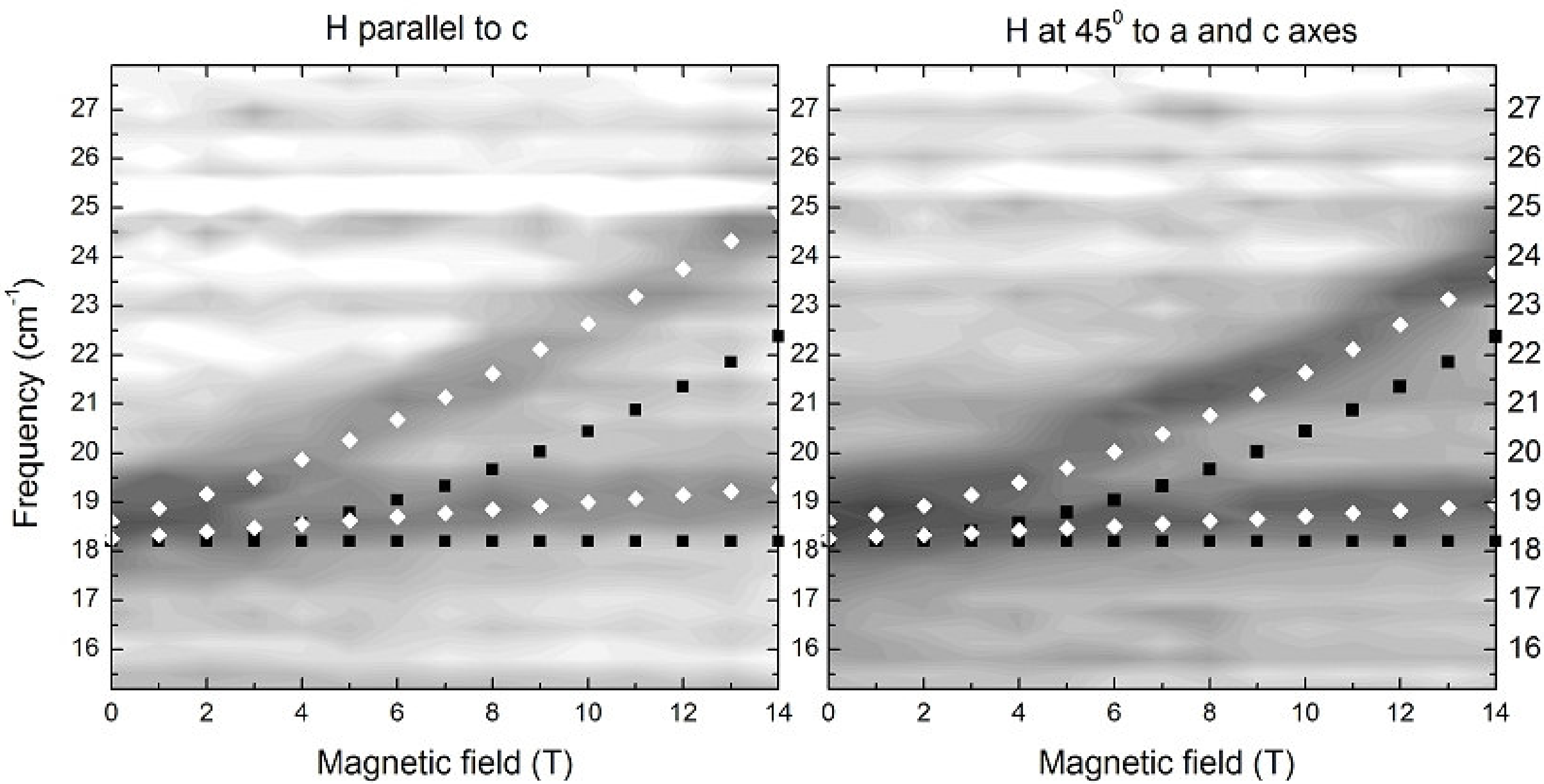


Measurement: H along easy axis
Black: Kittel's theory
White: D – M interaction



Kittel, 1952

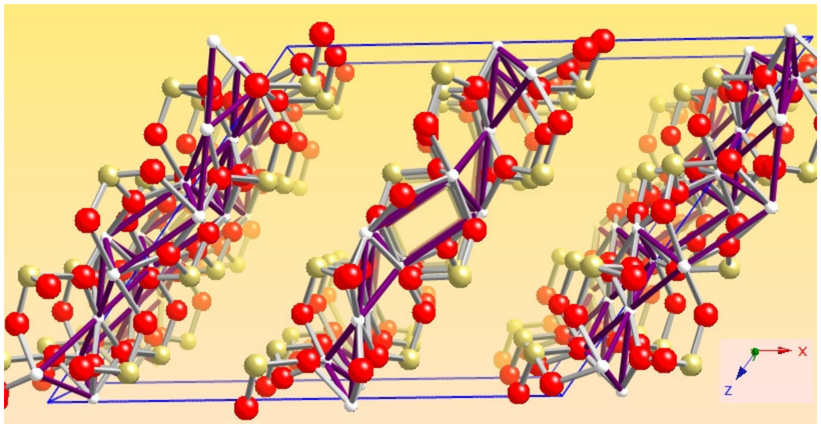
$LaMnO_3$: ESR

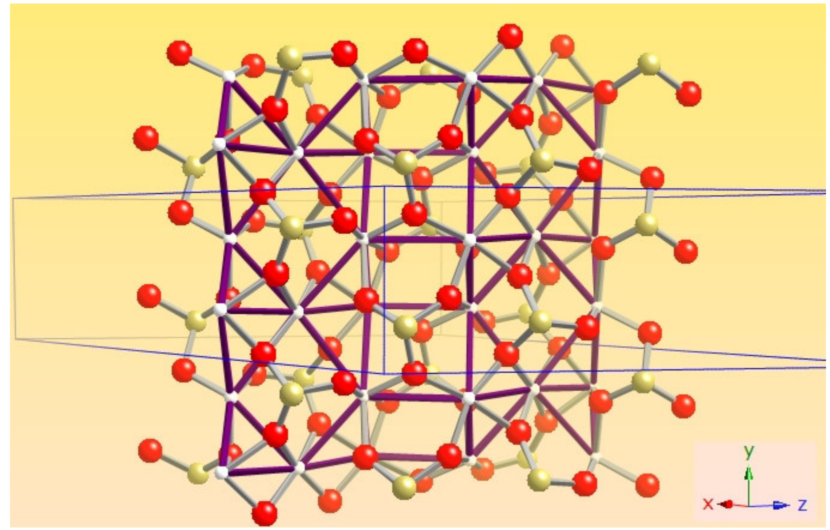


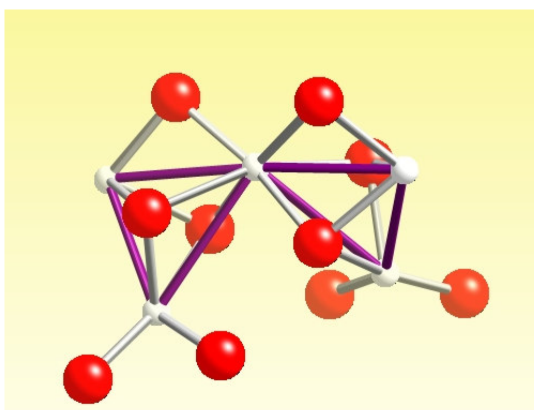
H perpendicular to easy axis -- Kittel result does not work

L. Mihály, D. Talbayev, L.F. Kiss, J. Zhou, T. Fehér and A. Jánossy Phys. Rev. B **69** 024414, (2004)
D.Talbayev L. Mihaly J. Zhou, Phys.Rev. Letters, **93** 017202 (2004)

$Ni_5(TeO_3)_4Cl_2$: "optical" magnons



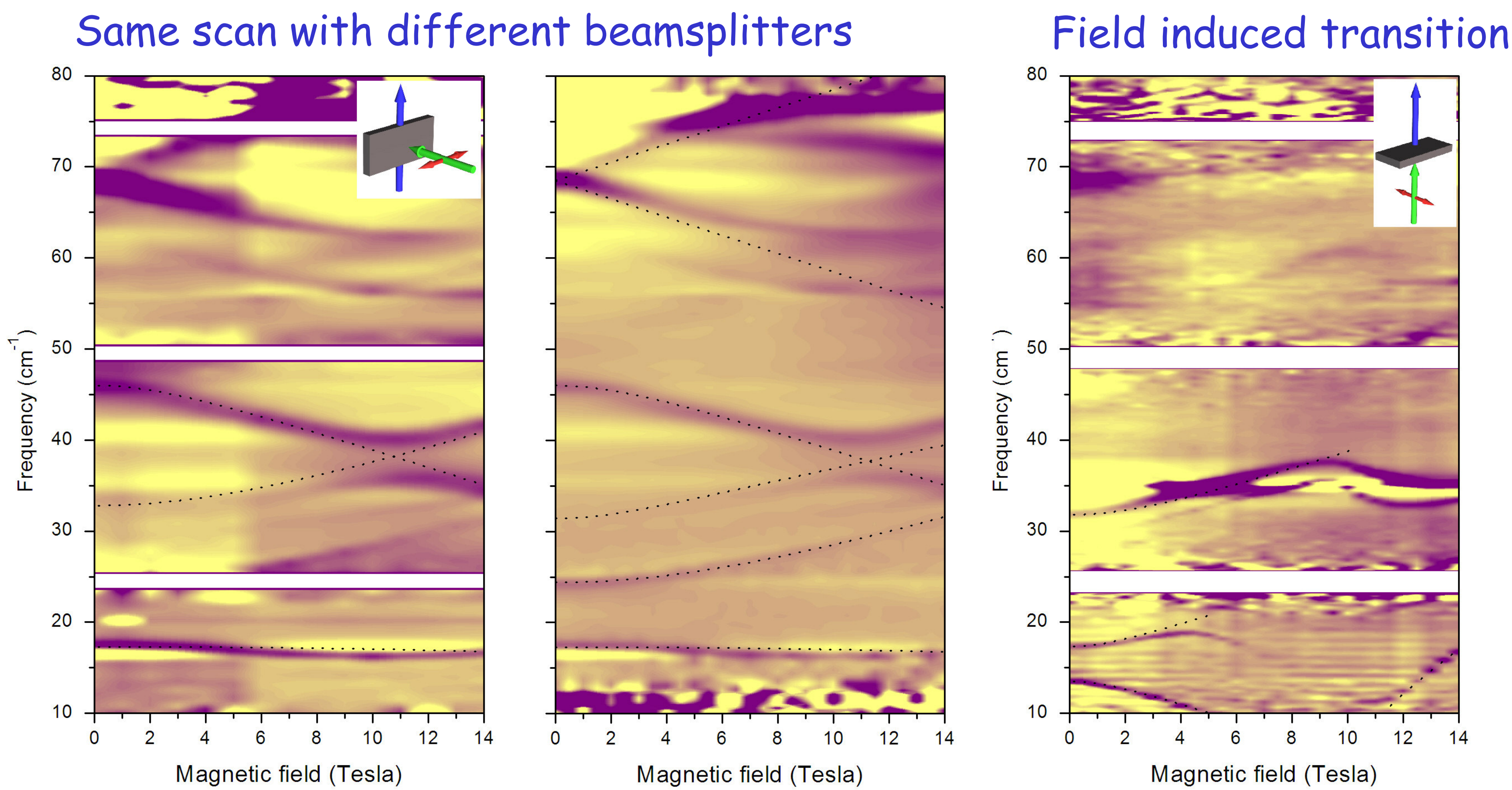




Building block has 5 Ni sites:
More magnons at q=0 - similar to optical phonons

$Ni_5(TeO_3)_4Cl_2$

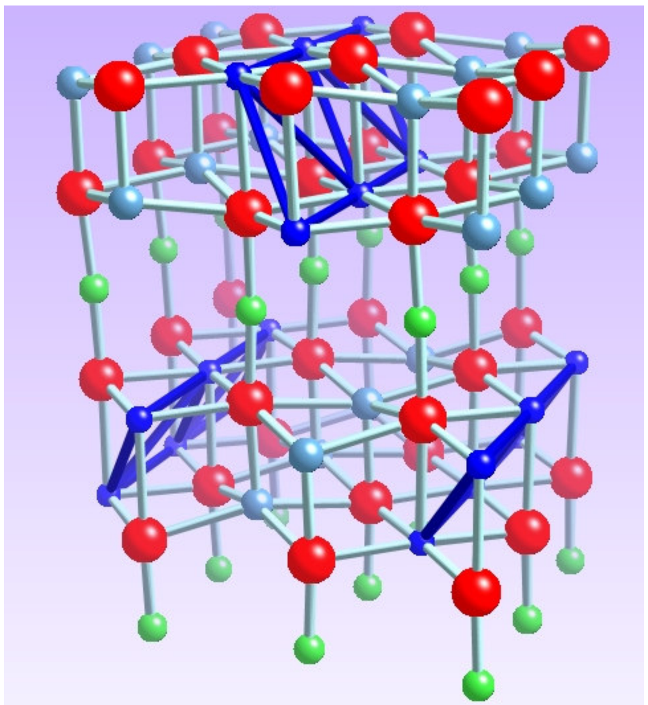
At least 8 (possibly 10) modes. Intensity depends on polarization. Field induced transition for field applied perpendicular to sample plane.

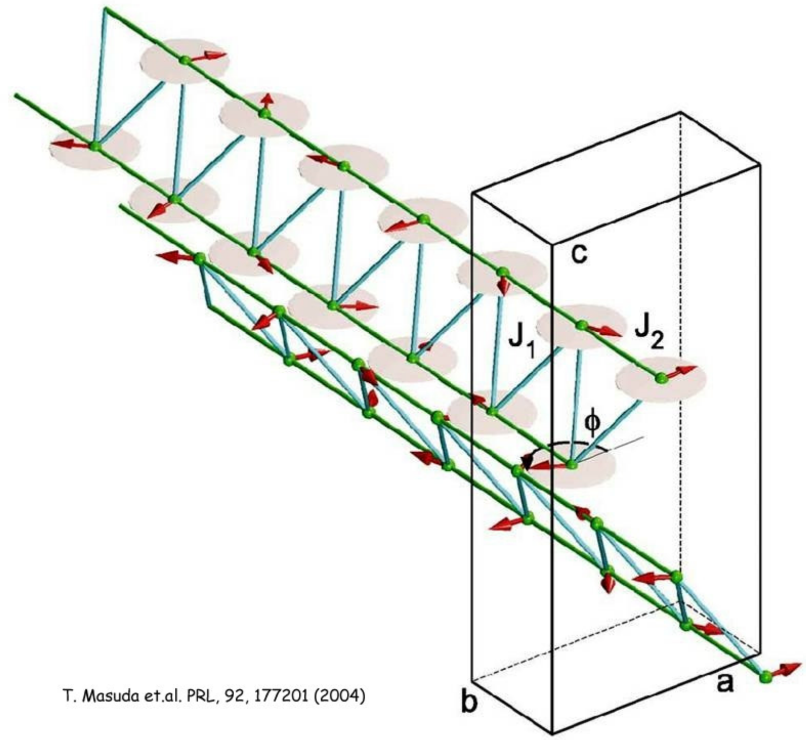


L. Mihaly, T. Feher, B. Nafradi, H. Berger, L. Forro, to be published

$LiCu_2O_2$: Helical order

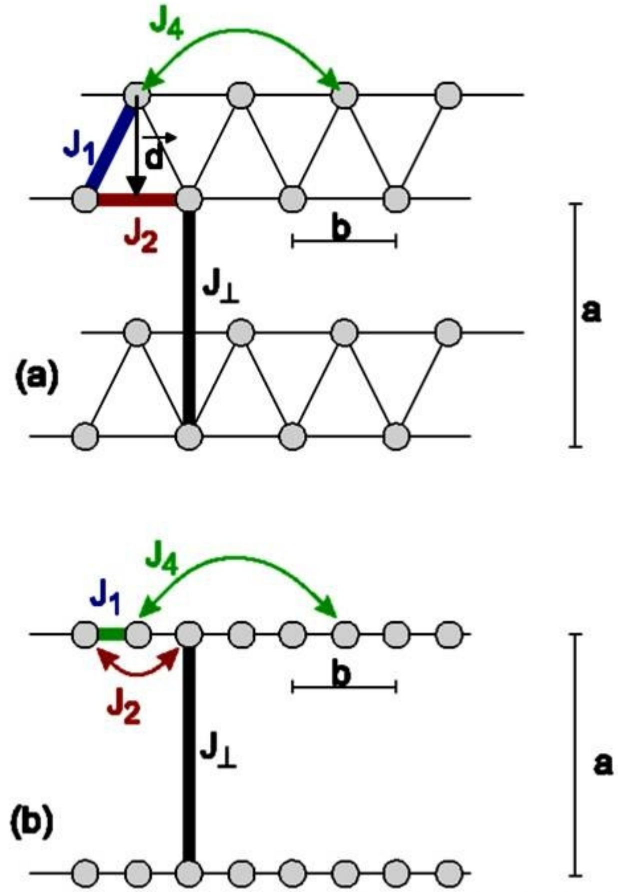
One dimensional chains, „triangular ladder",
spin order determined from neutron scattering





T. Masuda et.al. PRL, 92, 177201 (2004)

Interactions in $LiCu_2O_2$



T. Masuda et.al. cond-mat 0412625

Frustrated exchange coupling

$$\mathcal{H} = \sum_{i,j} J_1 S_{i,j} S_{i+1,j} + J_2 S_{i,j} S_{i+2,j} + J_4 S_{i,j} S_{i+4,j} + J_\perp S_{i,j} S_{i,j+1} - g\mu_B H S^y_{i,j} + \mathcal{H}'$$

J_1=6.4meV
J_2=-11.9meV (ferromagnetic.)
J_4= 7.4meV
J_{perp}=1.8meV

From neutron scattering

T. Masuda et al. cond-mat/0412625,
Phys. Rev. B **72**, 014405 (2005)

Frustration --> helical spin order.

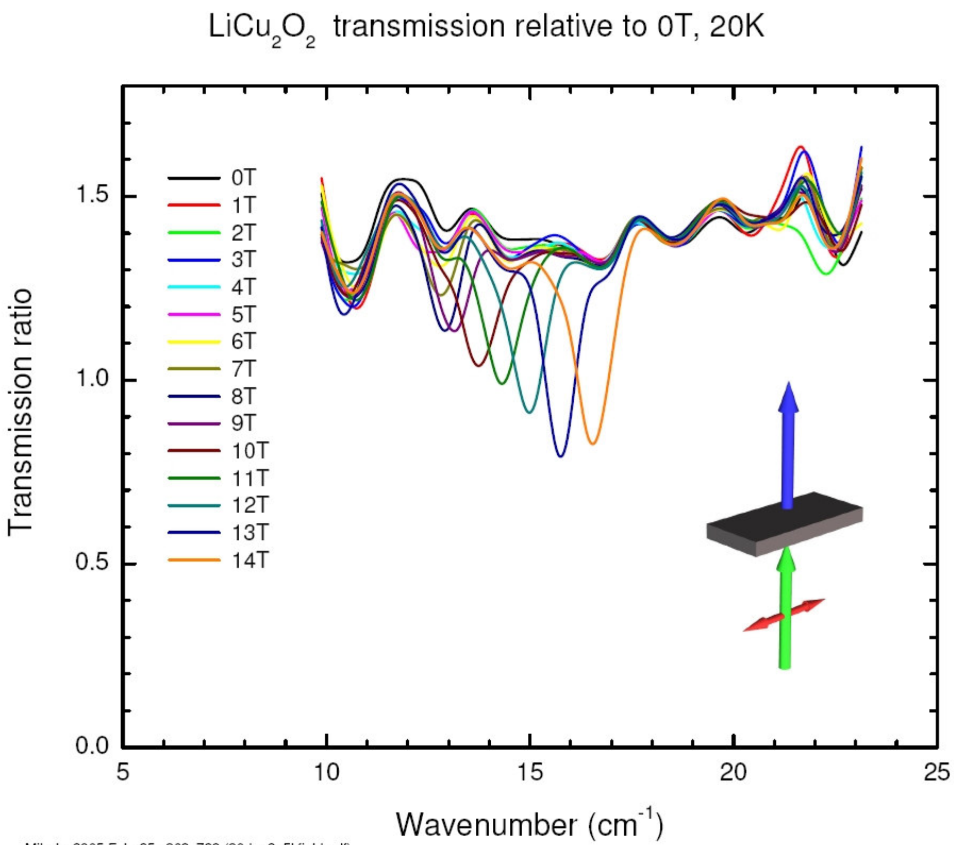
J-s are not enough! What selects the plane of the helix? How large is the gap at $\mathbf{q}=0$ in the magnon spectrum?

ESR provides the answer

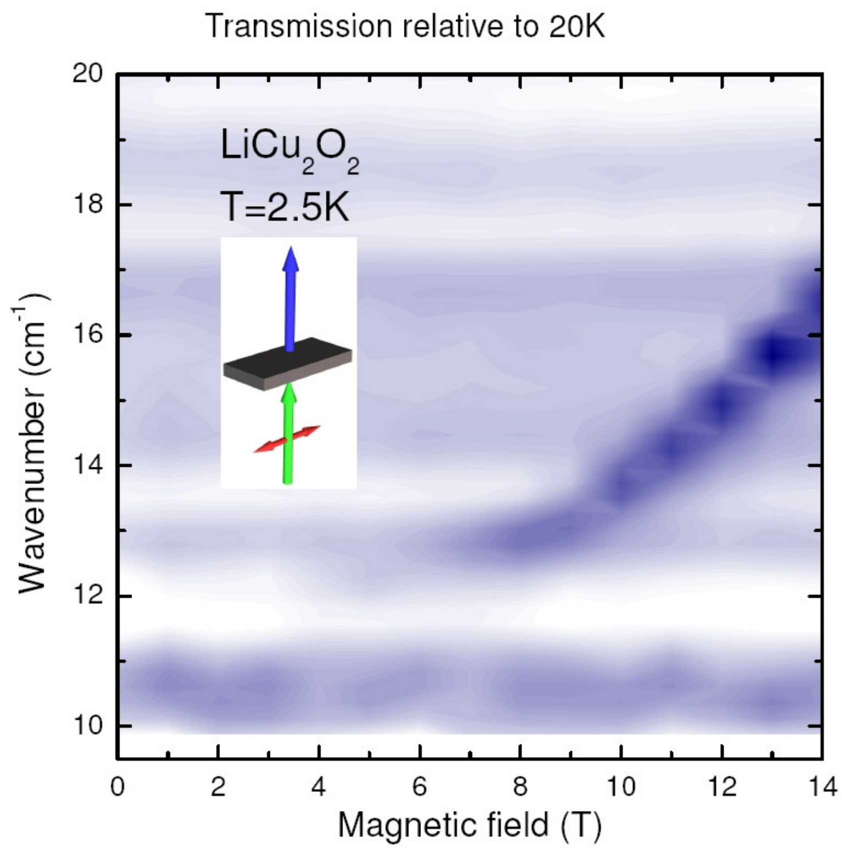
Exchange anisotropy

$$\mathcal{H}' = D_{ex} S^y_{i,j} S^y_{i+1,j}$$

ESR on $LiCu_2O_2$

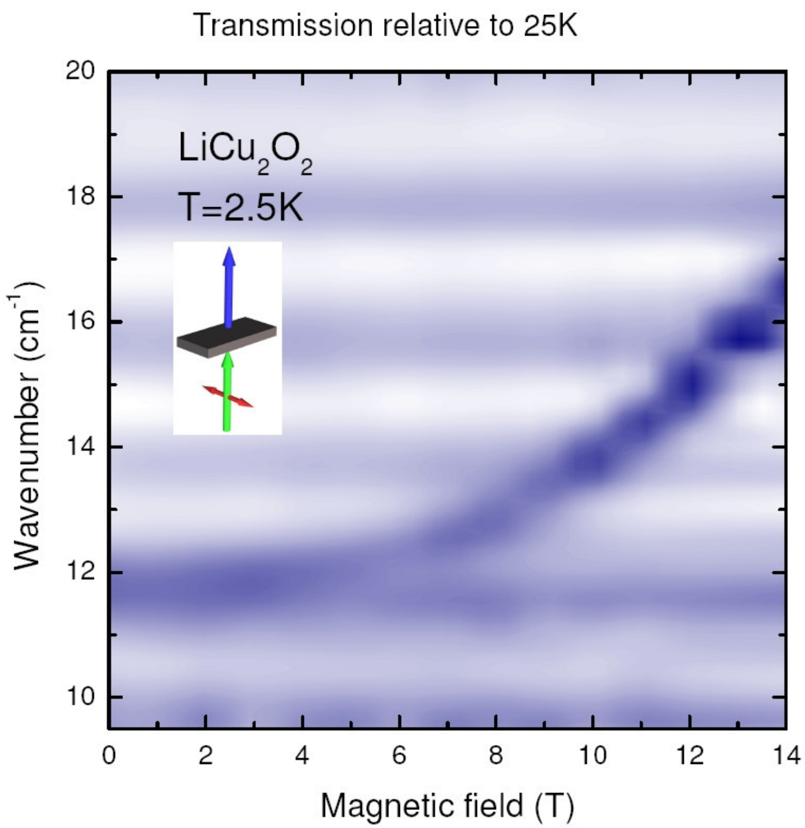


Mihaly, 2005 Feb. 25 v963..728 (90deg2p5kfield.pdf)

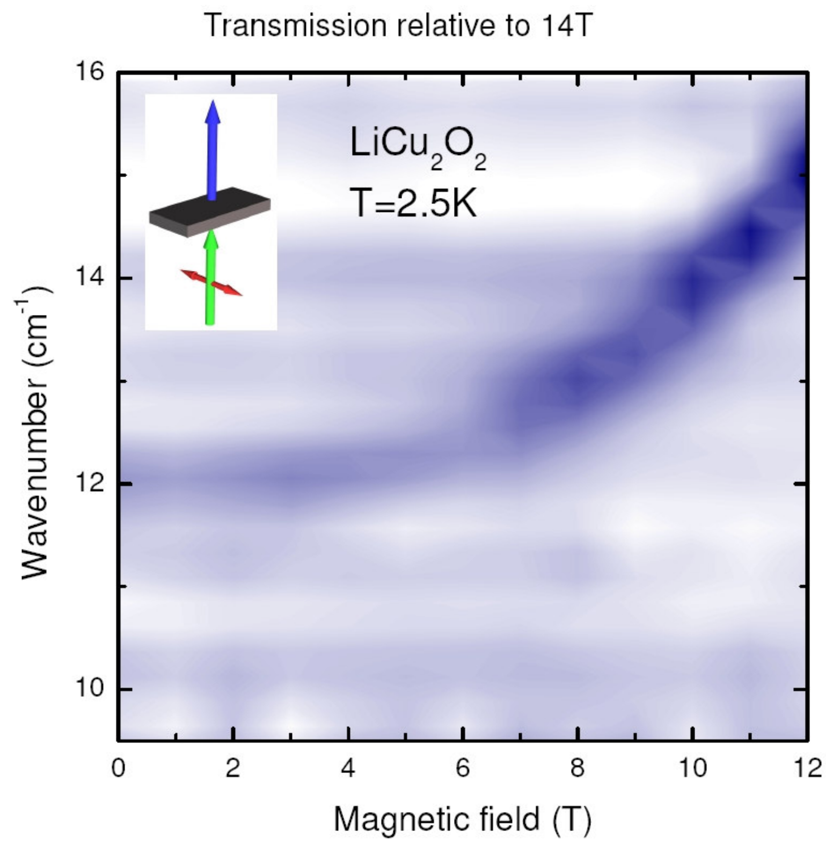


Mihaly 2005 Feb. 25 v963..728 (90deg2p5kfield_2d)

Two representations – same data

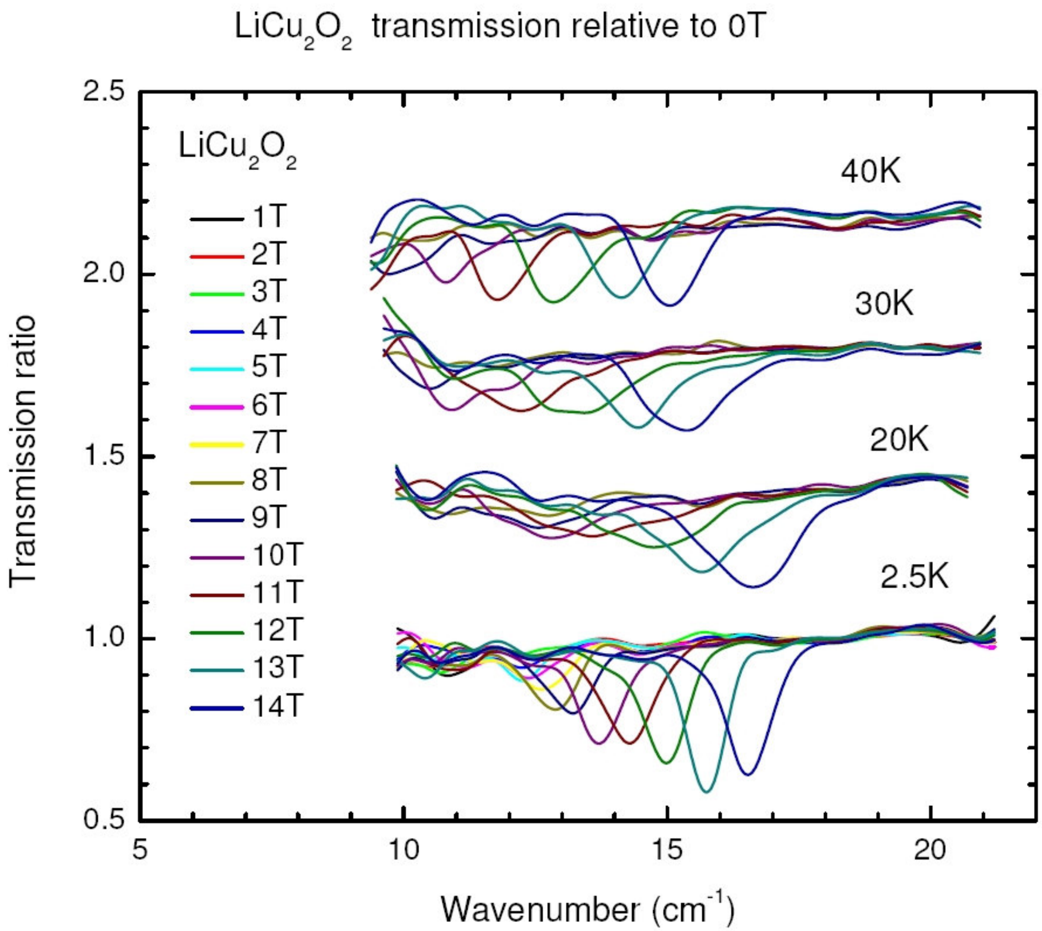


Mihaly, 2005 Feb. 26 t941..881 (0deg2p5Kfield)

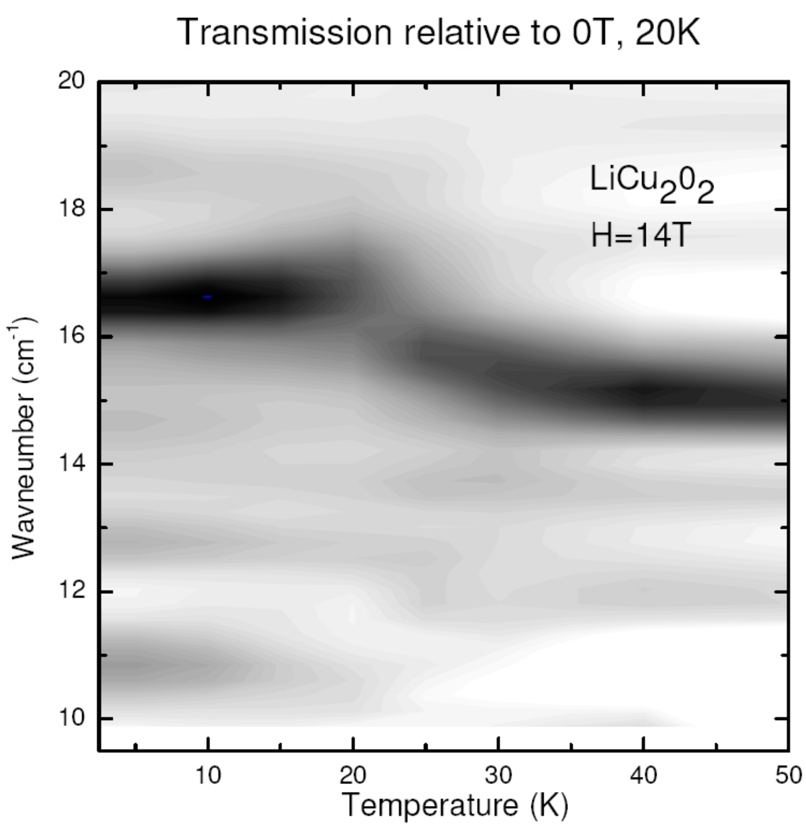


Mihaly, 2005 Feb. 26 t941..881 (0deg2p5Kfield_2d_14T)

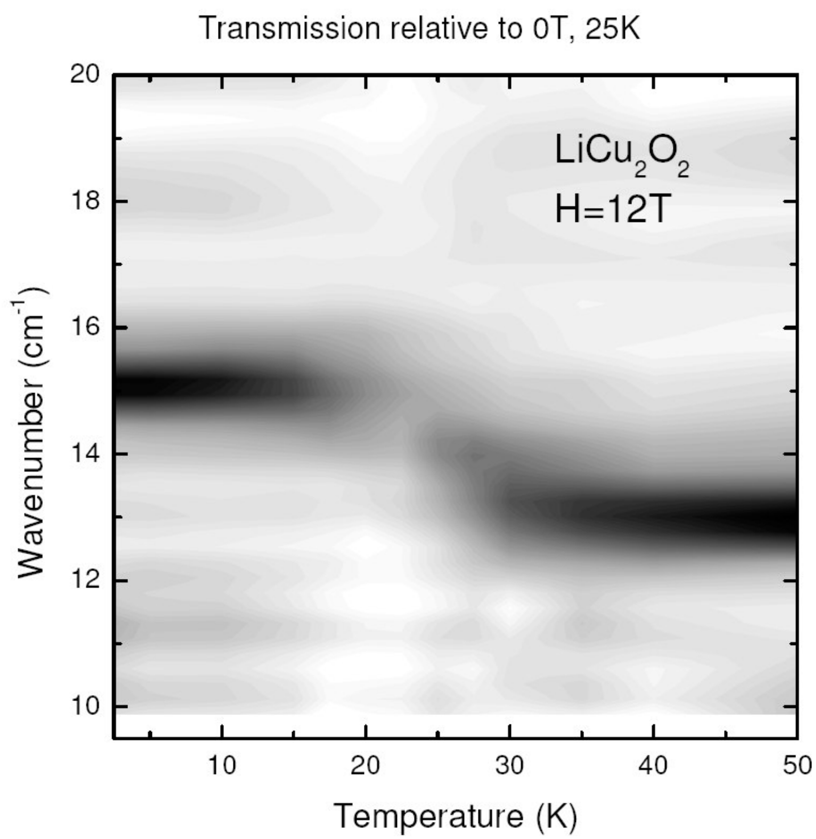
Temperature dependence



Temperature dependence at fixed fields

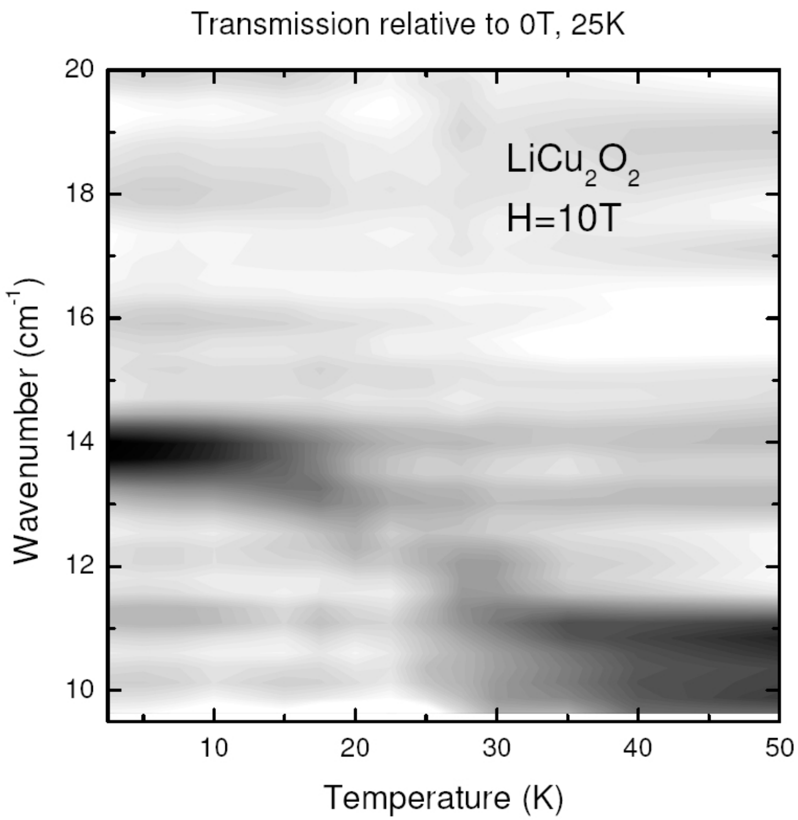


Mihaly, 2005 Feb. 26 u527..u421 (90deg14Ttemp_2d)

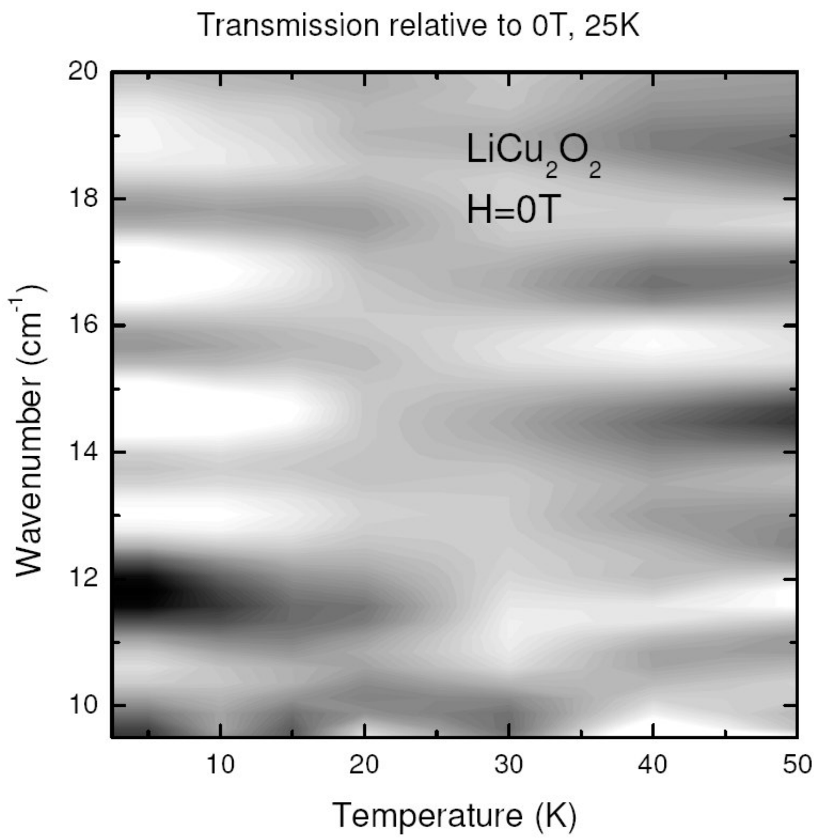


Mihaly, 2005 March 6 x728..638 (0deg12Ttemp_2d)

Temperature dependence at fixed fields



Mihaly, 2005 March 6 x983..786 (0deg10Ttemp)



Mihaly, 2005 March 6 x728..638 (0deg0Ttemp)

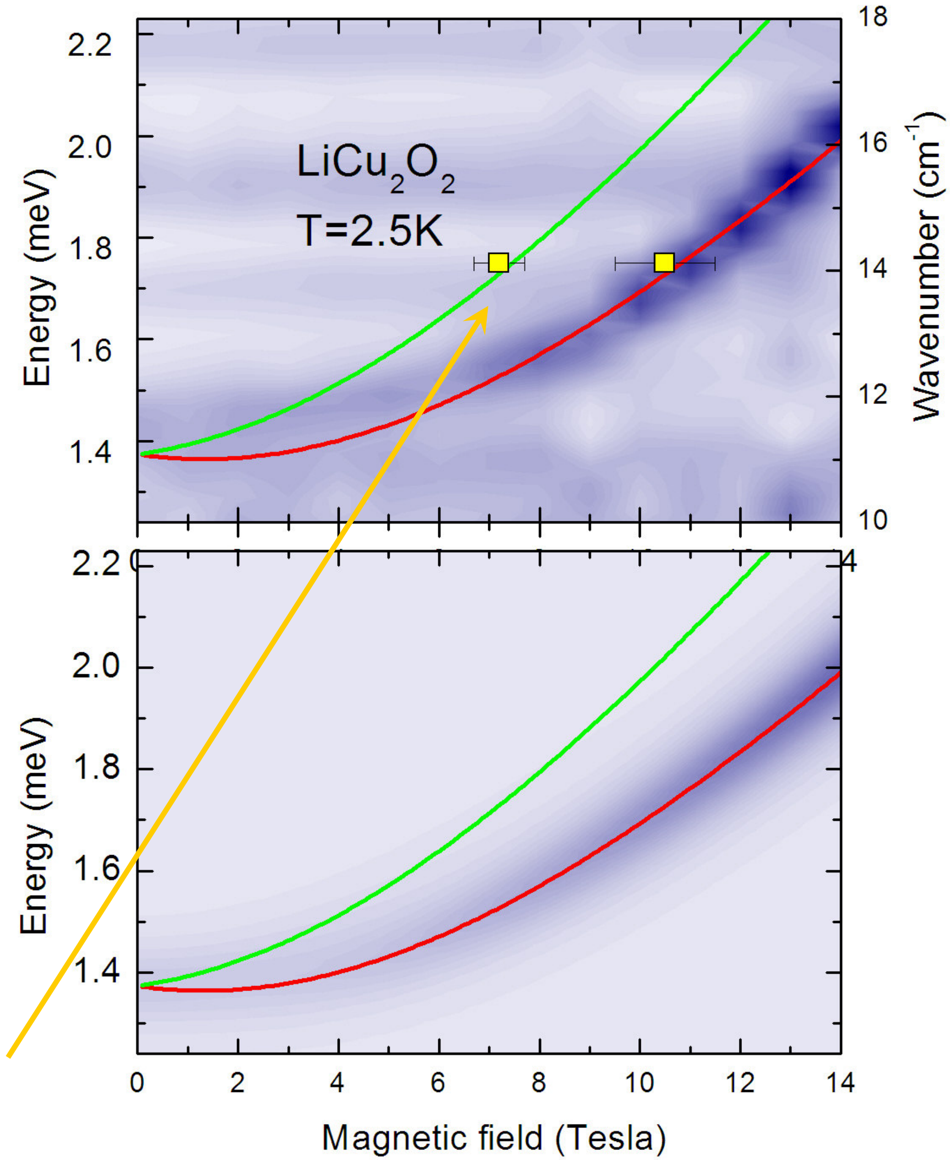
$LiCu_2O_2$: ESR

Helical order with field induced canting

$$J' = \frac{J(2\mathbf{Q}) + J(\mathbf{0})}{4} - \frac{J(\mathbf{Q})}{2}$$

Zero field gap:

$$\Delta = 2S\sqrt{J' \cos(\phi/2) D_{ex}}$$

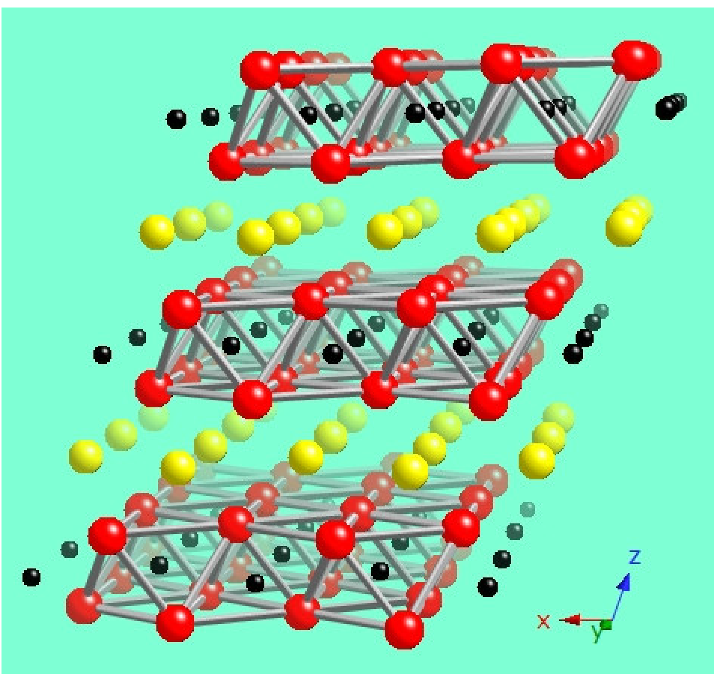


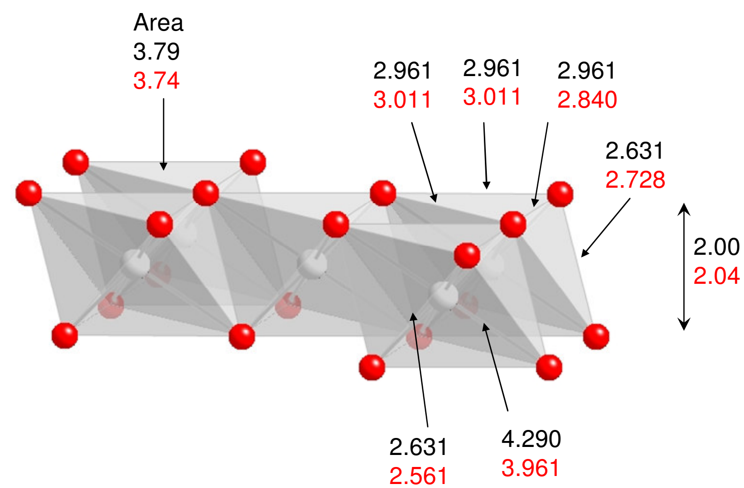
Two branches.
ESR susceptibility is strongly field dependent, weak signal for upper branch.
Weak signal observed at 420GHz AFTER theory predicted it

L. Mihály, B. Dóra, A Ványolos, H. Berger, L. Forró, cond-mat 0601701 (2006)
+ B. Náfrádi

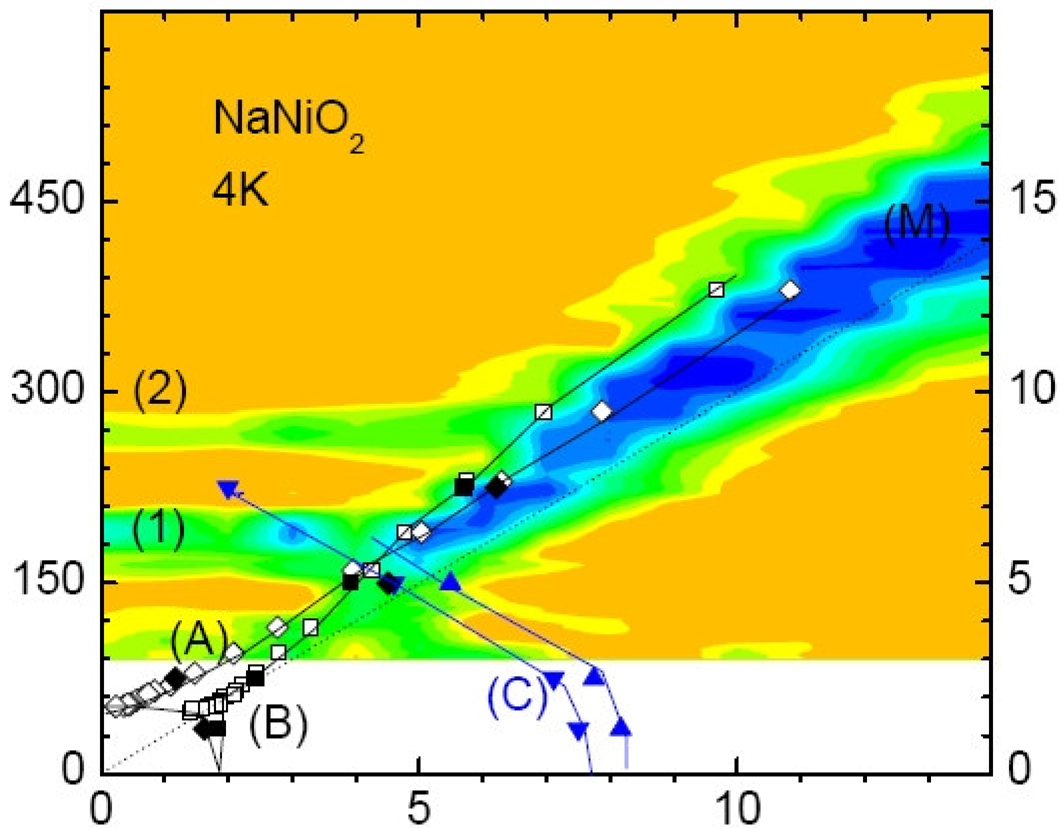
$NaNiO_2$: triangular layered

- Ni^{3+} ions in $t^6_{2g}e^1_g$ configuration
- High temperature structure: rhombohedral, with triangular Ni lattice
- Cooperative JT distortion at 475K; becomes monoclinic
- Magnetic order at 20K
- powder sample





$NaNiO_2$ (DeBrion)



Color map: NSLS
Circles: ESR in Grenoble
Triangles: ESR in Budapest
Dotted lines: calculated from g-factors
Solid lines: guide to the eye

$NaNiO_2$ spin Hamiltonian

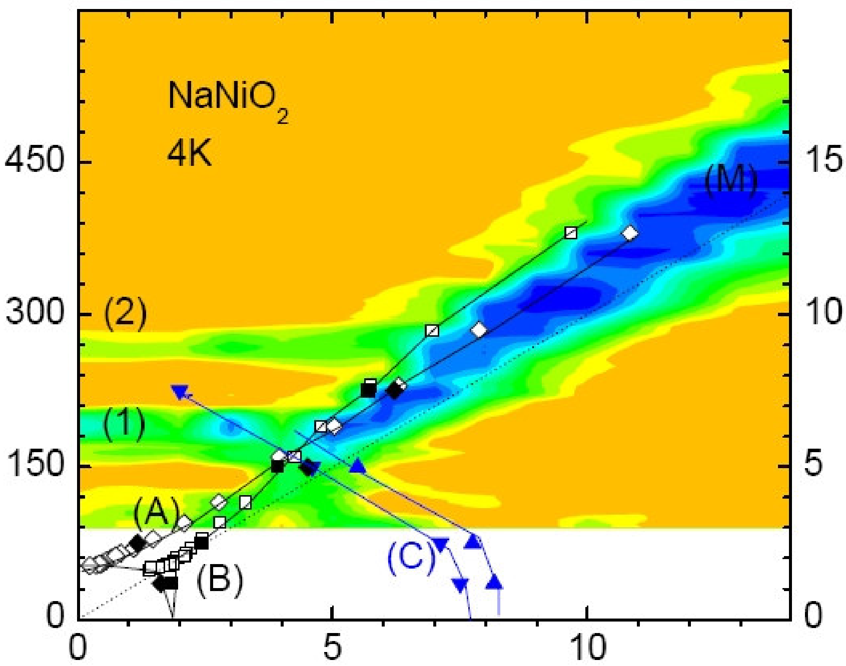
$$\mathcal{H}_0 = J_F \sum_{ab} \mathbf{S}_i \mathbf{S}_j + J_{AF} \sum_{c} \mathbf{S}_i \mathbf{S}_j + g\mu_B \mathbf{S}\mathbf{H}$$
$$+ J_F^z \sum_{ab} S_i^z S_j^z + J_{AF}^z \sum_{c} S_i^z S_j^z$$
$$+ J_F^x \sum_{ab} S_i^x S_j^x + J_{AF}^x \sum_{ab} S_i^x S_j^x$$

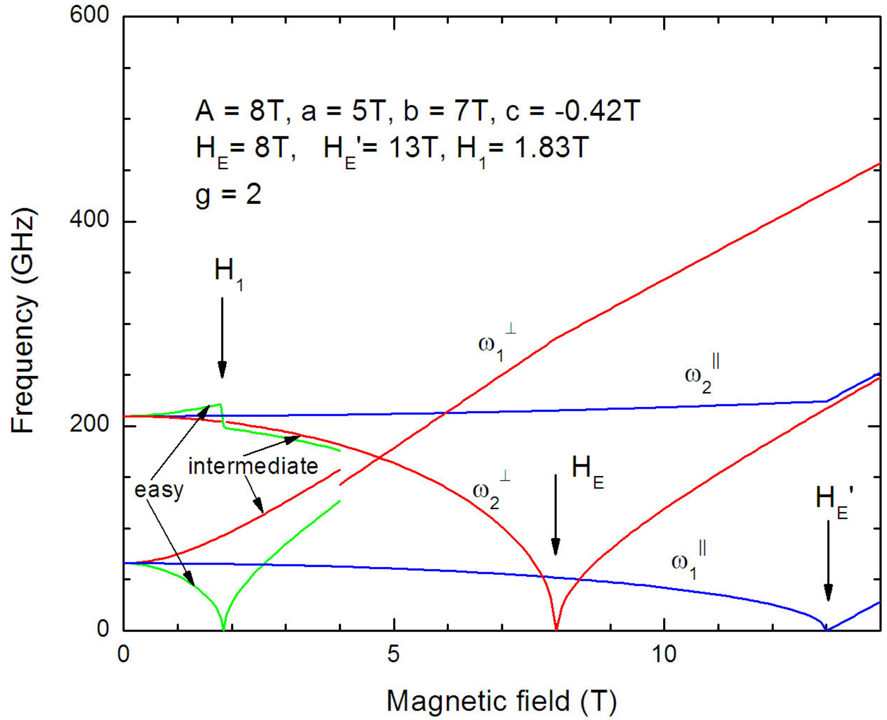
$$\mathcal{H}_{MF} = \frac{N}{2}(2S)^2 \{6[J_F(\mathbf{m}_1^2 + \mathbf{m}_2^2) + J_{AF}(2\mathbf{m}_1\mathbf{m}_2) +$$
$$J_F(m_{1z}^2 + m_{2z}^2) + J_{AF}(2m_{1z}m_{2z}) +$$
$$+ J_F(m_{1x}^2 + m_{2x}^2) + J_{AF}(2m_{1x}m_{2x})] +$$
$$g\mu_B \frac{1}{2S}(\mathbf{m}_1 + \mathbf{m}_2)\mathbf{H}\}$$

$$\mathcal{H}_{MF} = N\{\frac{A}{2}\mathbf{m}^2 + \frac{a}{2}m_z^2 + \frac{b}{2}l_z^2 + \frac{c}{2}m_x^2 + \frac{d}{2}l_x^2 - \mathbf{m}\mathbf{h}\}$$

NaNiO$_2$ bi-axial anisotropy

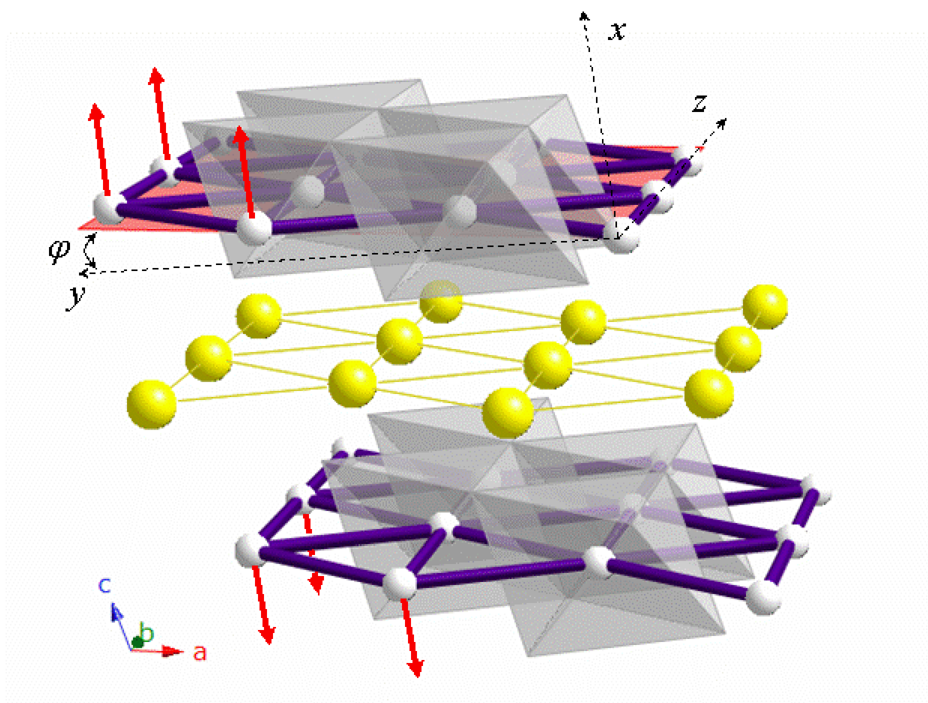
Easy axis within easy plane





$NaNiO_2$ spin orientation

In agreement with neutrons



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