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DQMP Flat Club – 2D Magnets
Christian Rüegg :: Director :: PSI

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- Introduction: Physics, Materials and Experiments
- Dirac Magnons in CrBr_3 and their Damping S. Nikitin *et al.*, Phys. Rev. Lett. **129**, 127201 (2022).
- Strong Spin-lattice Coupling in LiCrO_2 S. Toth *et al.*, Nature Comm. **7**, 13547 (2016).
- Quantum Criticality and Dynamics in 2D S. Allenspach *et al.*, Phys. Rev. Lett./Res. (2020/21).
- $\text{SrCu}_2(\text{BO}_3)_2$ Out-of-Equilibrium Magnons F. Giorgianni *et al.*, Phys. Rev. B **107**, 184440 (2023).

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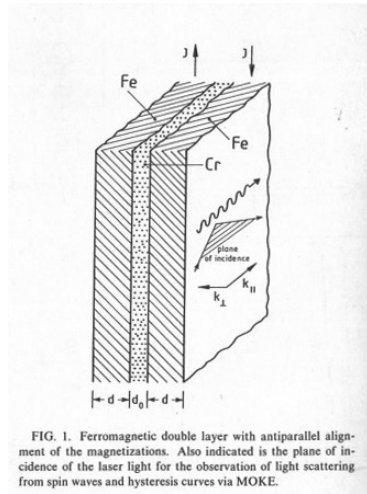
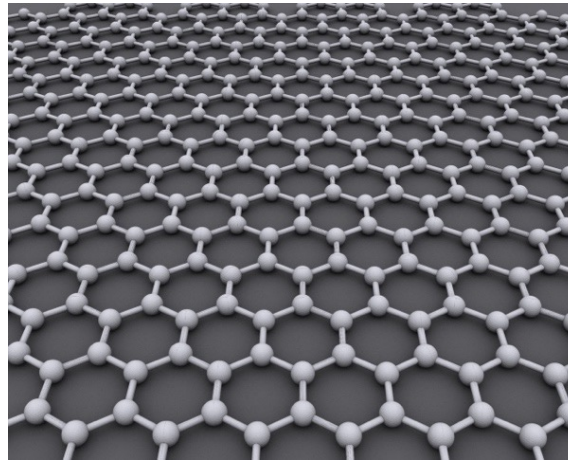
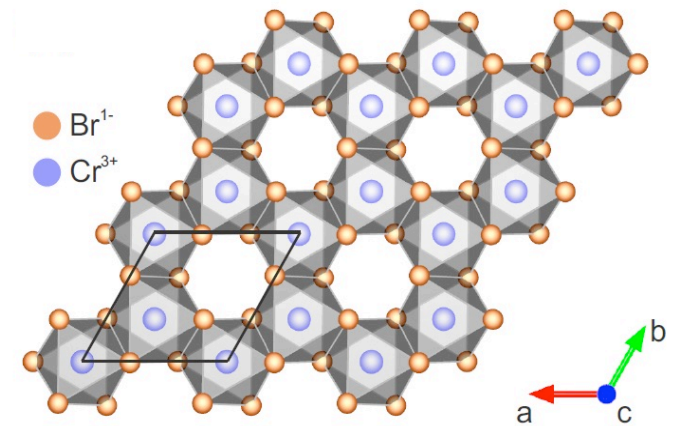


FIG. 1. Ferromagnetic double layer with antiparallel alignment of the magnetizations. Also indicated is the plane of incidence of the laser light for the observation of light scattering from spin waves and hysteresis curves via MOKE.

Nobel Prize 2007



Nobel Prize 2010



2D Metal Films

2D to 3D materials
Atomic control of thickness
Many applications (e.g. GMR)

Graphene

2D material
2D electronic states
Single layer, few layers and devices

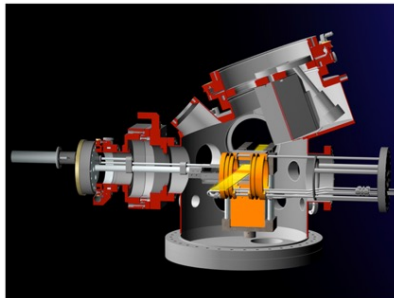
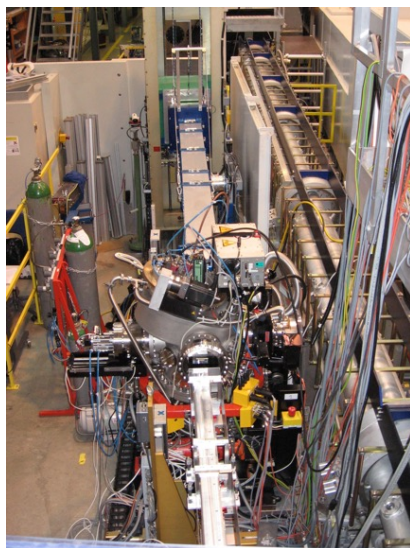
2D Magnets (e.g. square, honeycomb)

2D sub-structure in 3D material
2D magnetic states (3D at $T < T^*$)
Thin films, bulk and some applications

2D Materials – Magnetism of a single layer

In-situ sputtering of metallic thin films on the neutron reflectometer AMOR (SINQ)

- Focusing guide based on elliptic Selene principle, sample size $2 \times 20 \text{ mm}^2$ (now 1 mm^2)
- Polarized neutron beam, sensitivity: 1 layer
- Counting time: 30 m per layer (0.5 m in the future)

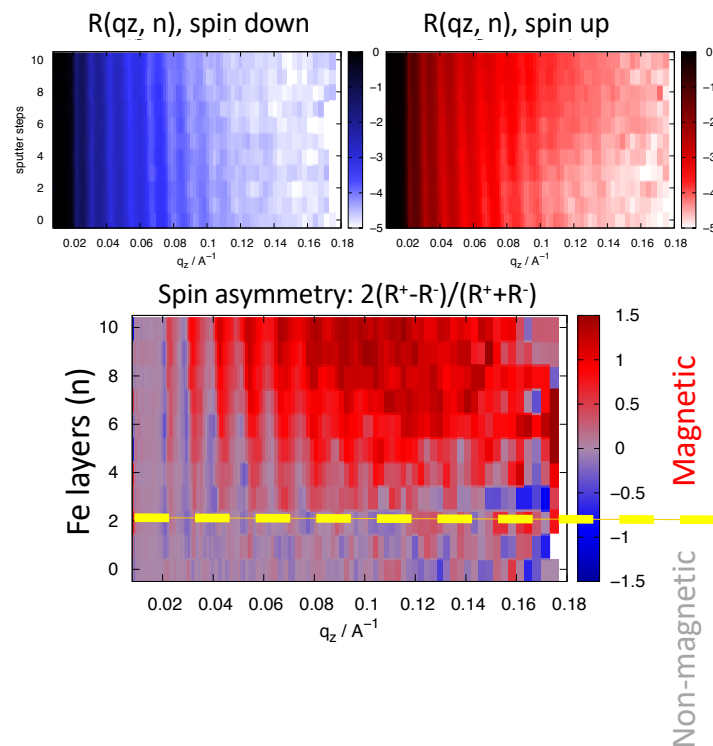


Sputtering chamber:

- Fe and other metals
- rotating gun
- in-situ sample positioning

Collaboration:

P. Böni, TU Munich; J. Mannhart, Univ. Augsburg; J. Stahn, PSI



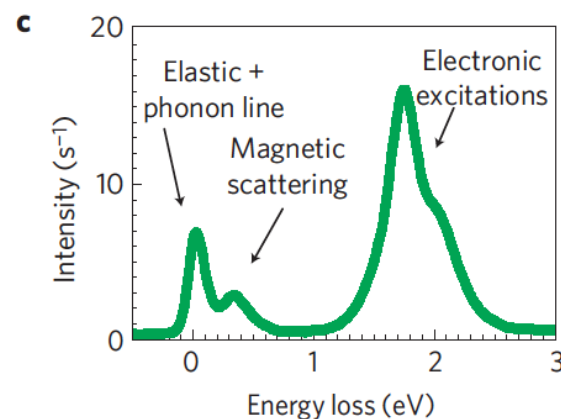
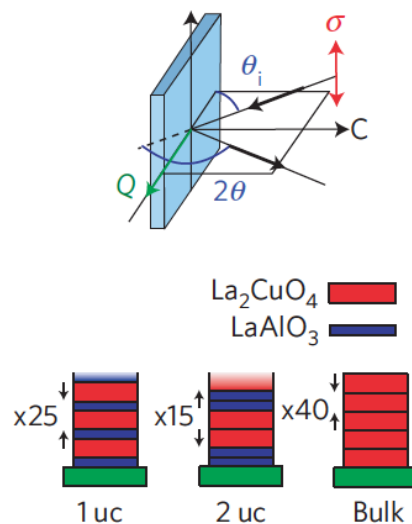
LETTERS

PUBLISHED ONLINE: 2 SEPTEMBER 2012 | DOI:10.1038/NMAT3409

nature
materials

Spin excitations in a single La_2CuO_4 layer

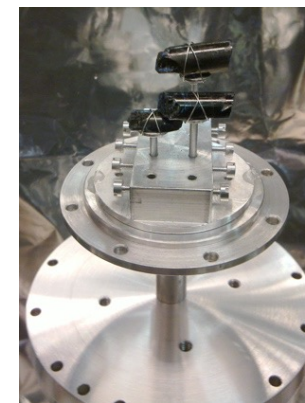
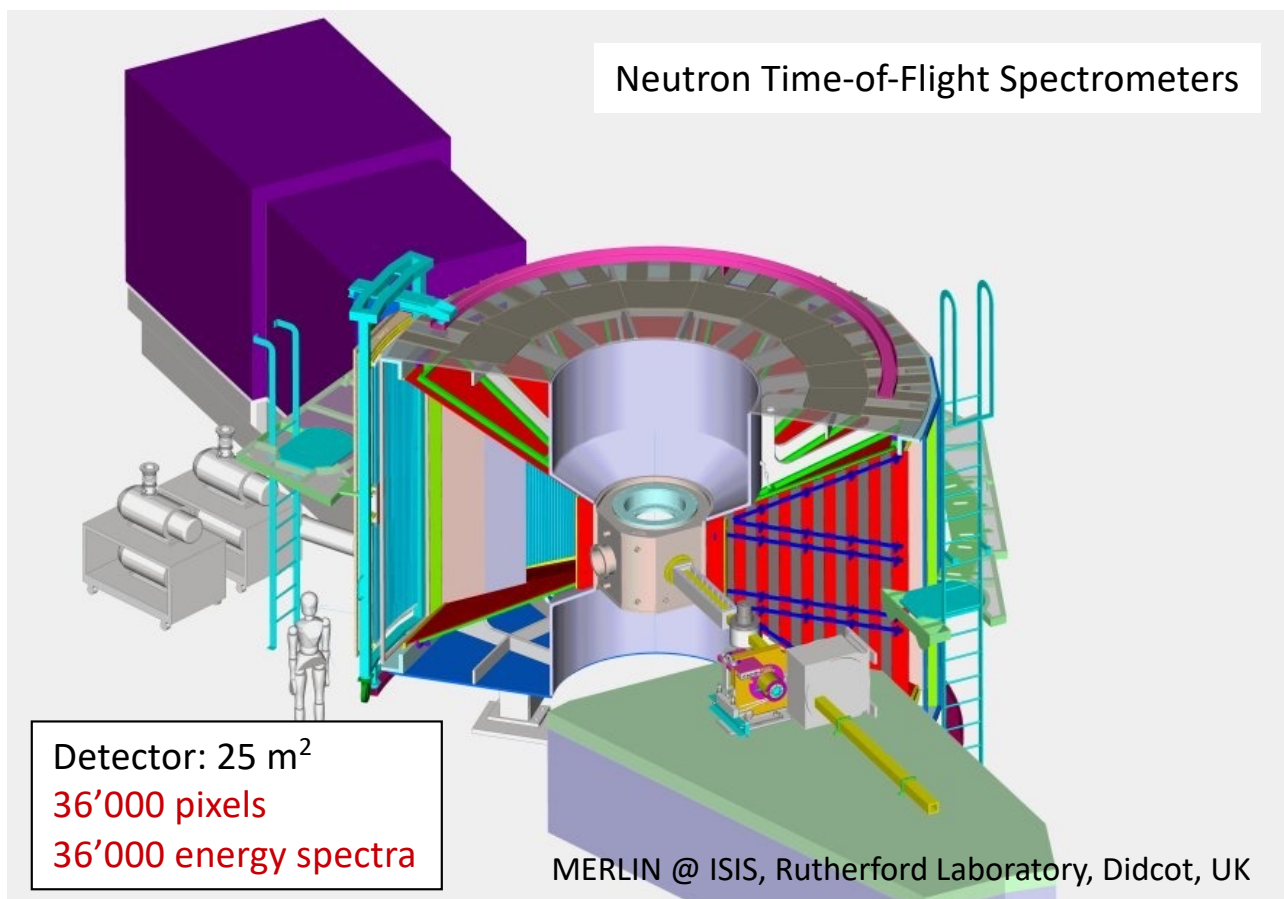
M. P. M. Dean^{1*}, R. S. Springell^{2,3}, C. Monney⁴, K. J. Zhou^{4†}, J. Pereiro^{1†}, I. Božović¹, B. Dalla Piazza⁵,
H. M. Rønnow⁵, E. Morenzoni⁶, J. van den Brink⁷, T. Schmitt⁴ and J. P. Hill^{1*}



RIXS (ADDRESS Beamline at the SLS)

M.P.M. Dean *et al.*, Nature Materials **11**, 850 (2012).

2D Materials – Excitations in bulk samples



M. Mena *et al.*, Phys. Rev. Lett. **124**, 257201 (2020).

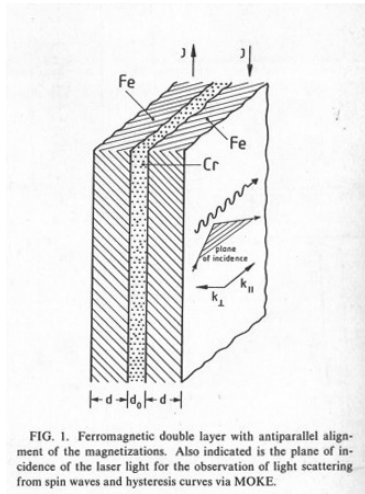
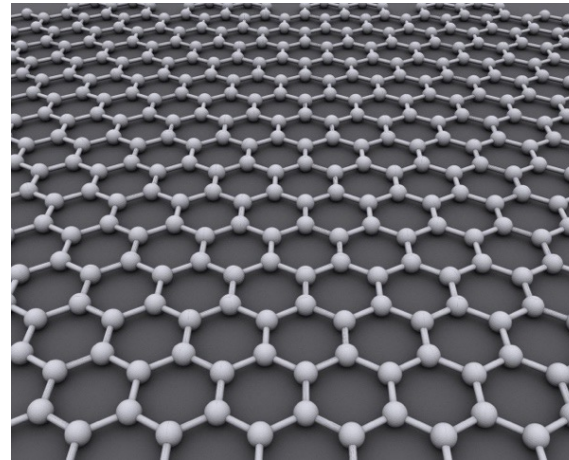
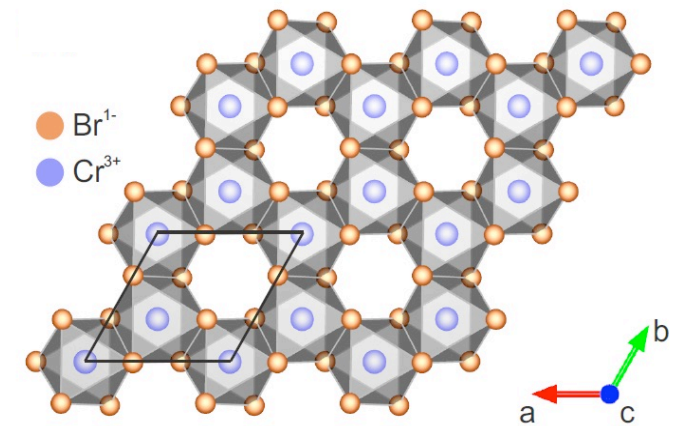


FIG. 1. Ferromagnetic double layer with antiparallel alignment of the magnetizations. Also indicated is the plane of incidence of the laser light for the observation of light scattering from spin waves and hysteresis curves via MOKE.

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Thin films, bulk and some applications

Experimental challenges? What information is missing from experiments?

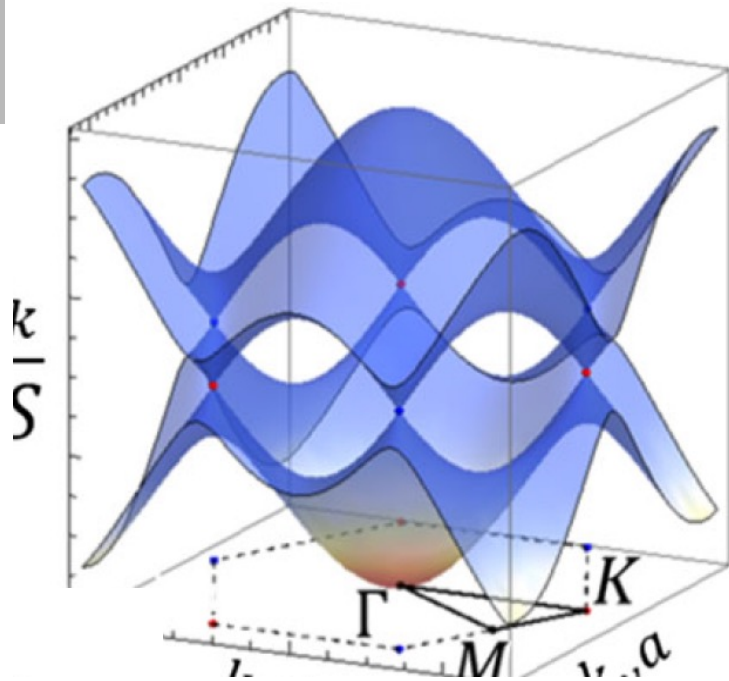
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Dirac Magnons in CrBr_3 and their Damping

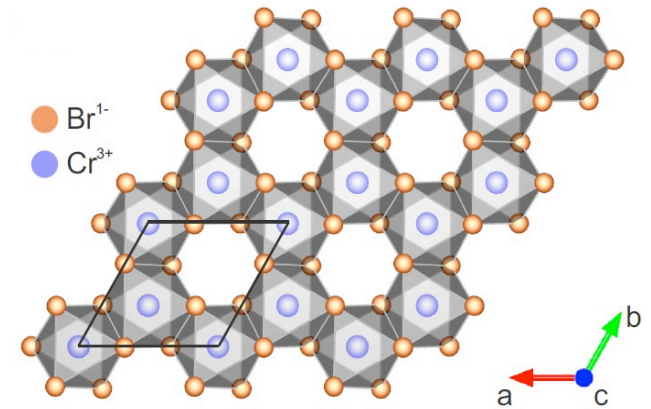
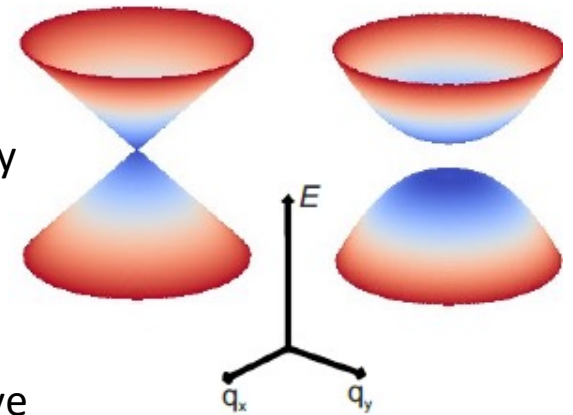
Spin Waves in a Honeycomb Ferromagnet



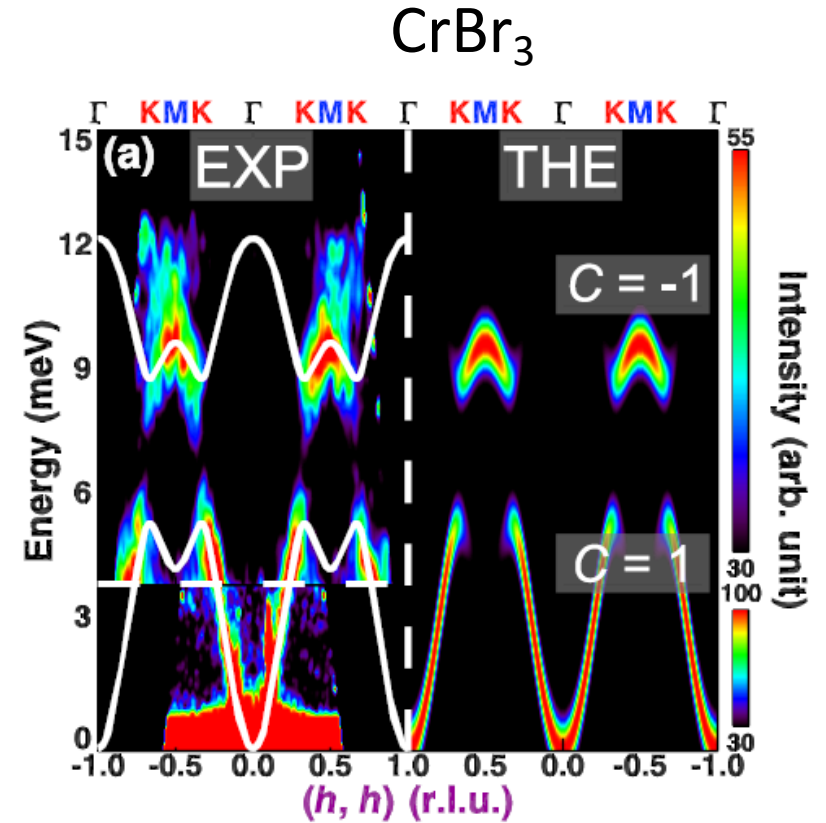
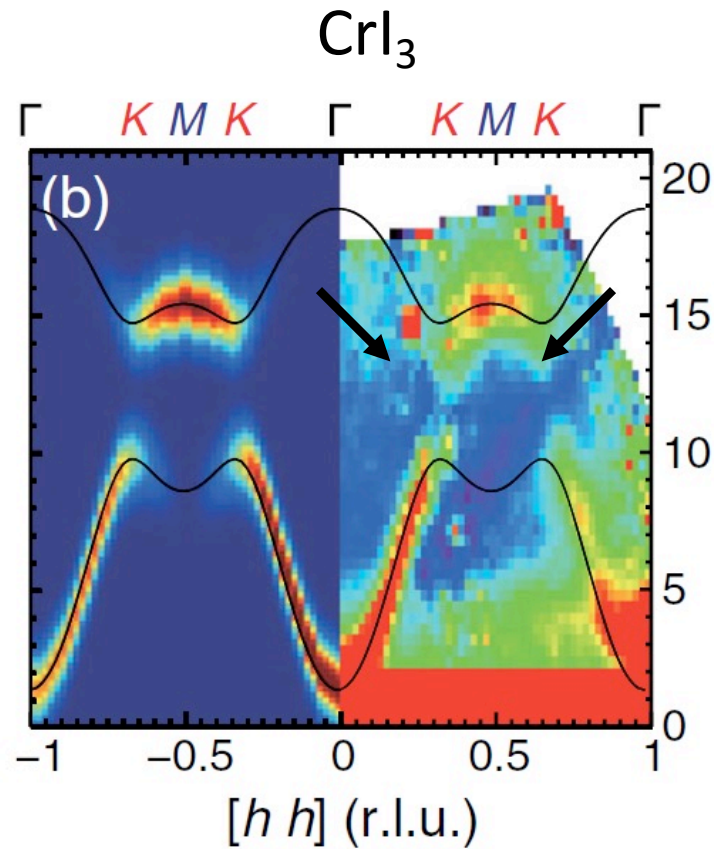
- Dirac cone at finite energy
- Spin gap opens if the inversion symmetry is broken
- Top and bottom band have Chern numbers of ± 1

$$C_{\pm} = \frac{1}{2\pi} \int_{\text{BZ}} d^2k \Omega_{\mathbf{k},\pm} = \mp 1,$$

- S. S. Pershoguba *et al.*, PRX **8**, 011010 (2018).
- L. Chen *et al.*, PRX **8**, 041028 (2018).

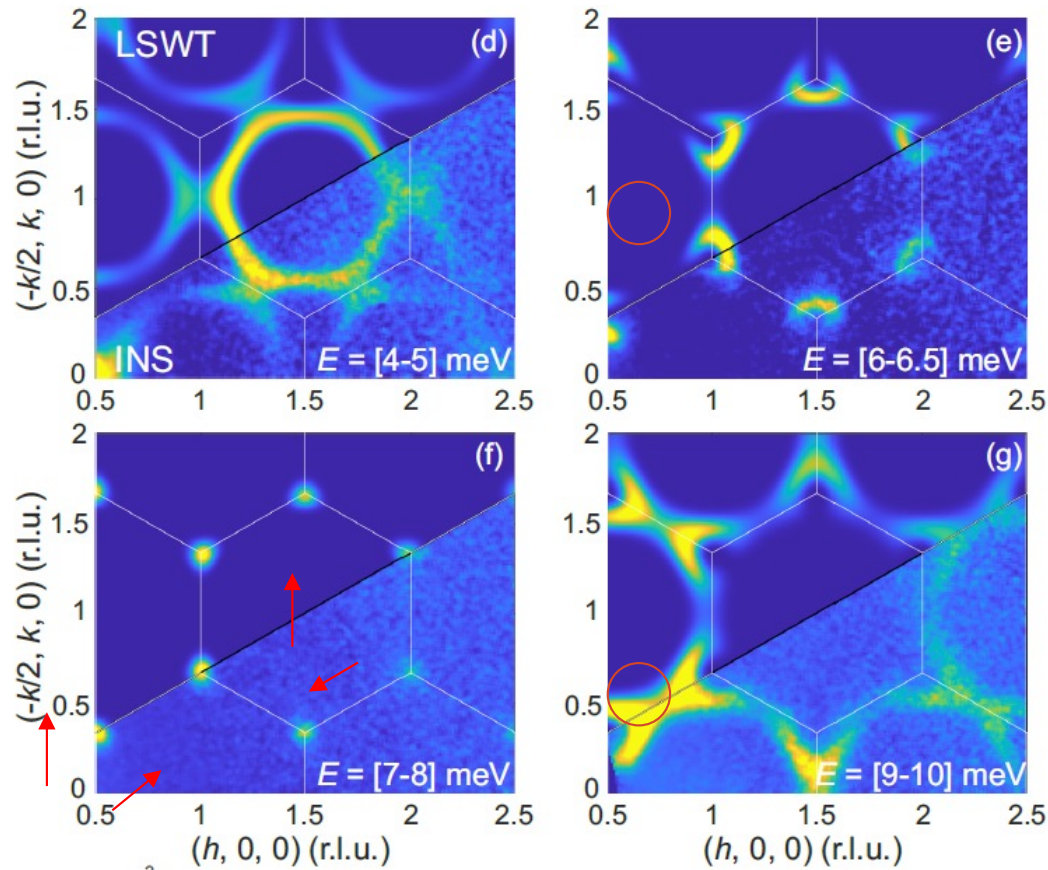


Spin Dynamics in CrX₃: DM Interaction matters?

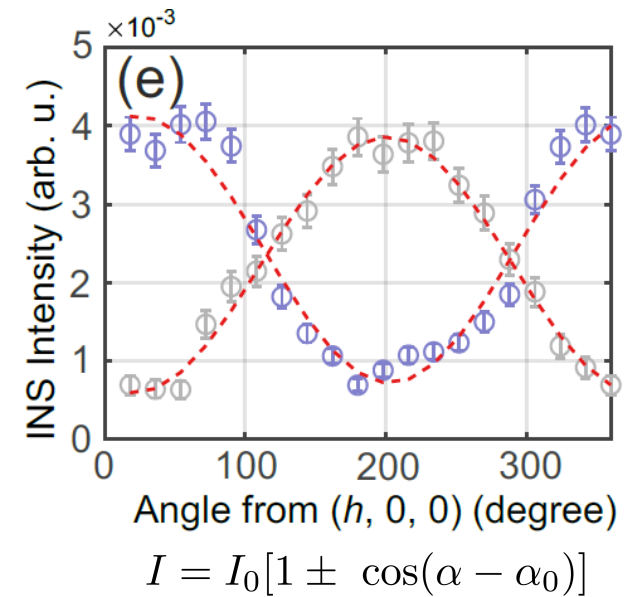


- Z. Cai *et al.*, PRB **104**, L020402 (2021).
- L. Chen *et al.*, PRX **8**, 041028 (2018).
- L. Chen *et al.*, PRX **11**, 031047 (2021).

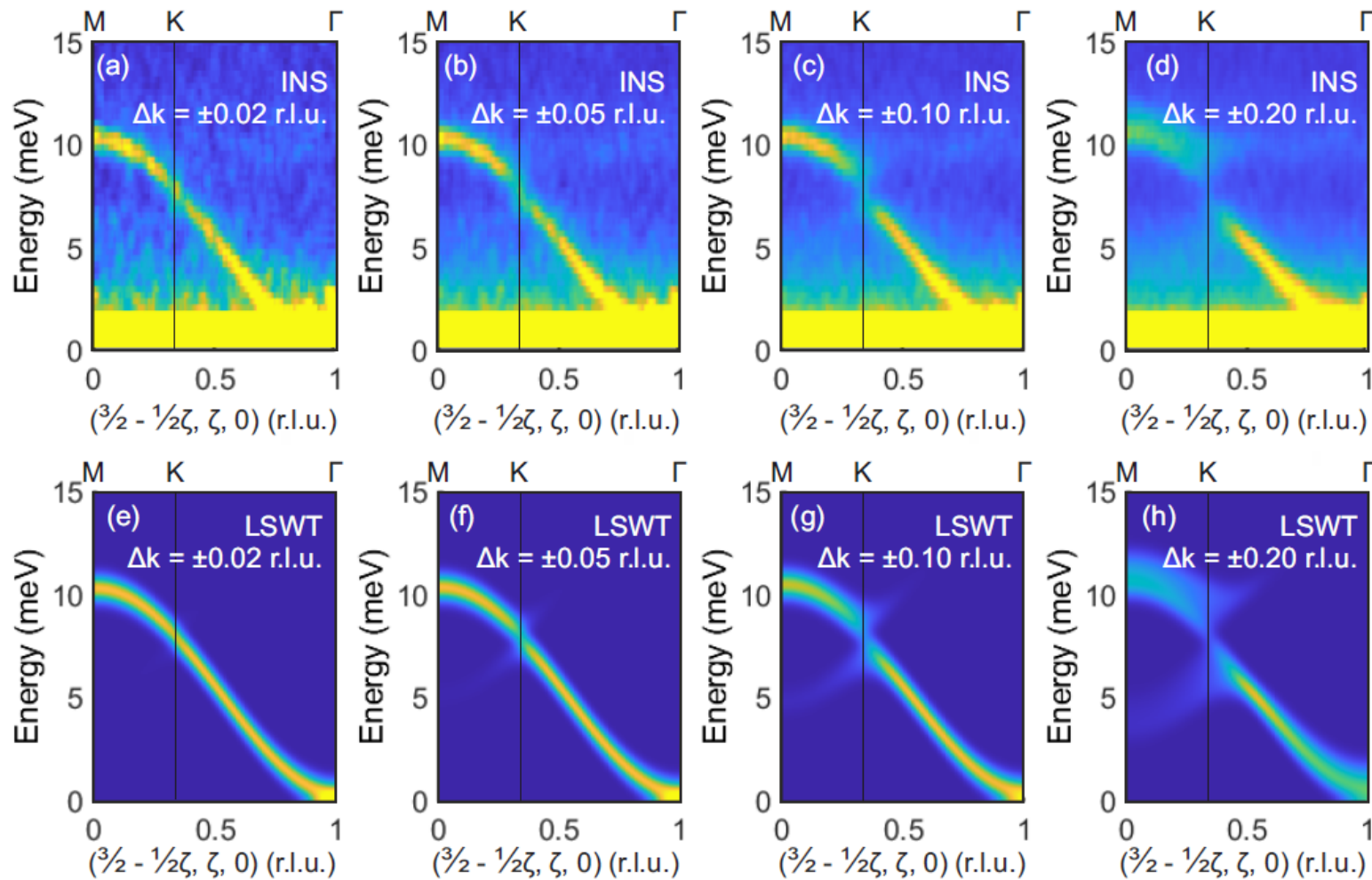
INS-ToF Data: Constant-Energy Slices and LSWT



- Perfect quantitative agreement between INS and LSWT
- Clear intensity at the Dirac point
- Intensity winding around K -point



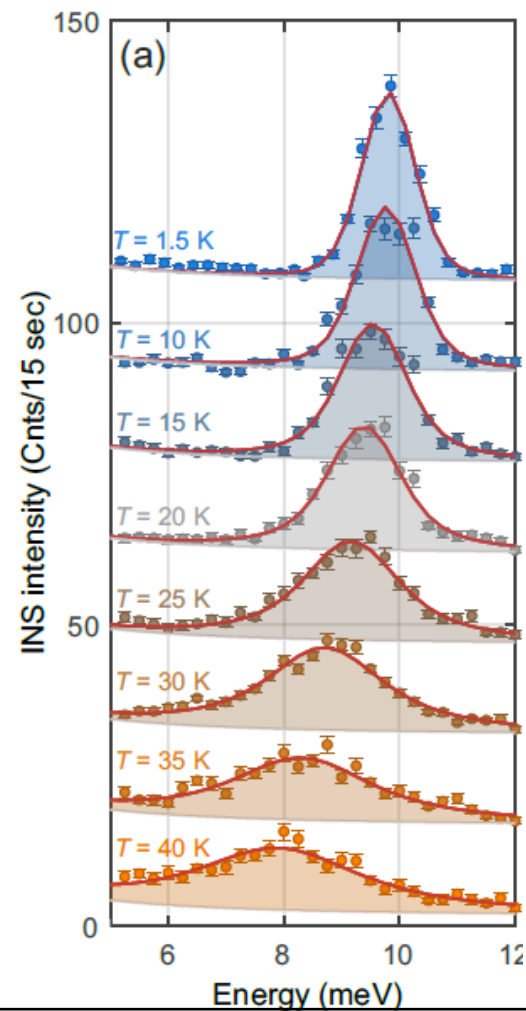
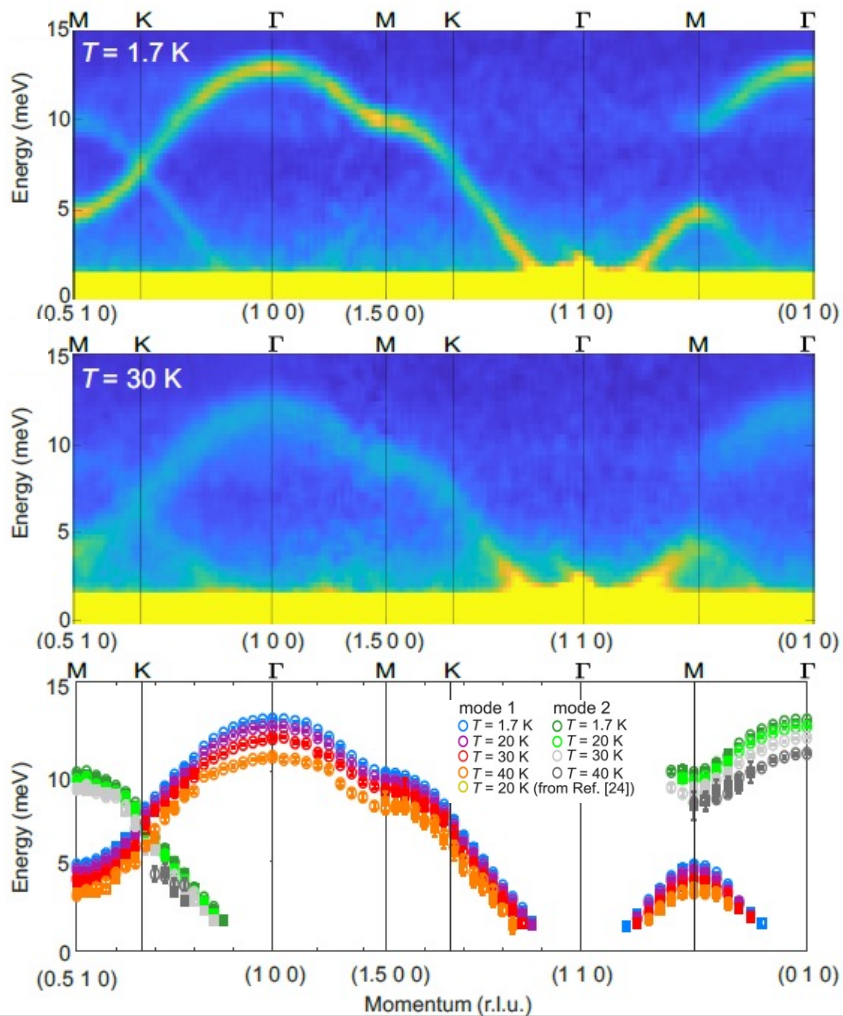
Data Analysis – Momentum Integration Range



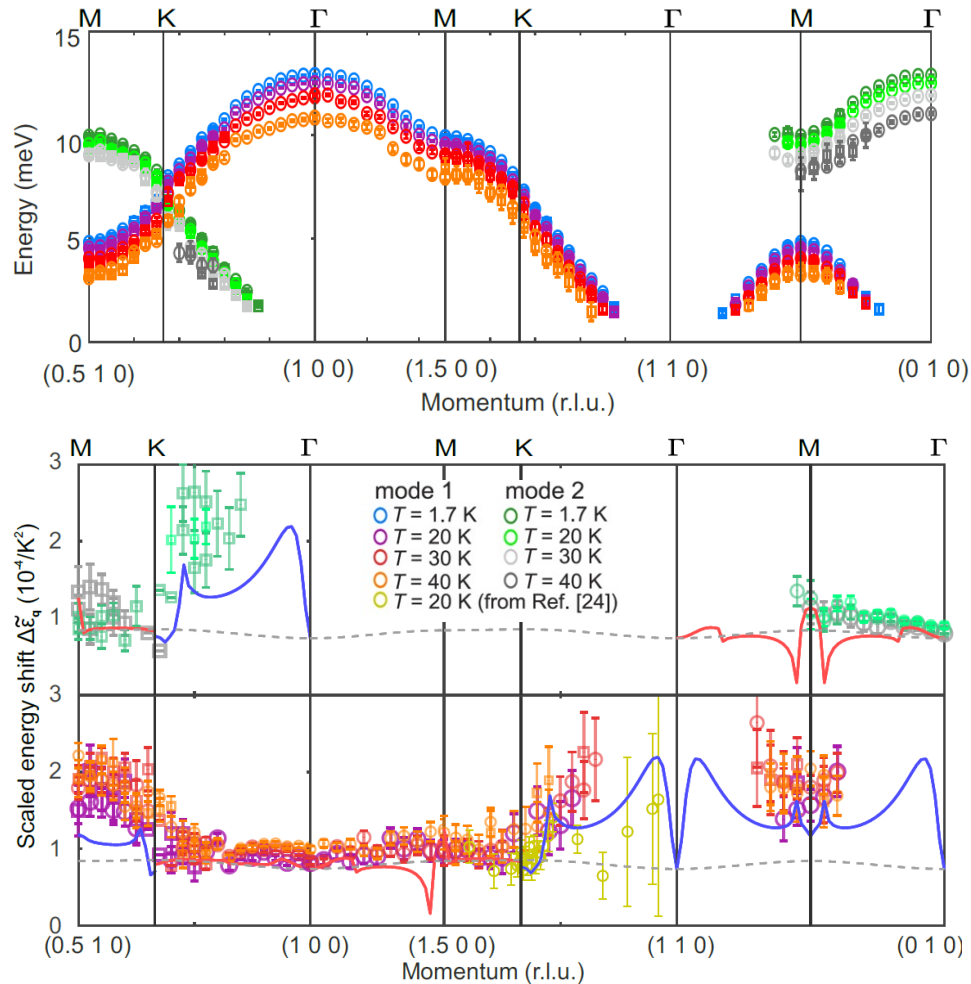
• S. Nikitin *et al.*,
Phys. Rev. Lett. **129**,
127201 (2022).

Agree with results
on CrCl_3 by S-H. Do
et al., Phys. Rev. B
106, 060408 (2022).

High-Temperature INS data



Temperature-induced Magnon Renormalization



- Good collapse for T^2 -scaling
- Renormalization is stronger for the down band (magnons below K-point)
- Renormalization for the up-band can be well described by Hartree term alone.
- **Q**-dependence is monotonic, sharp Van-Hove-like peaks are absent

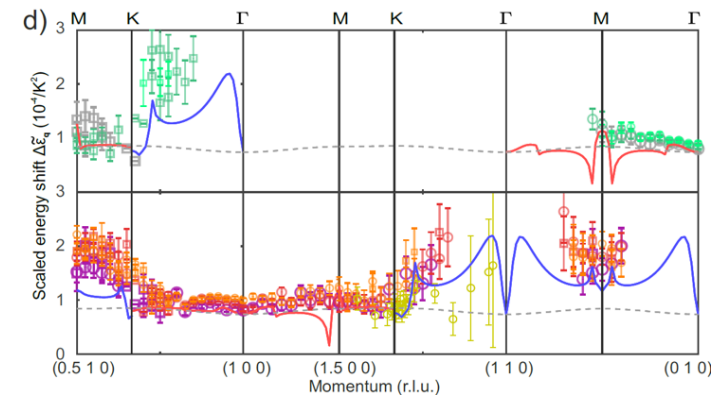
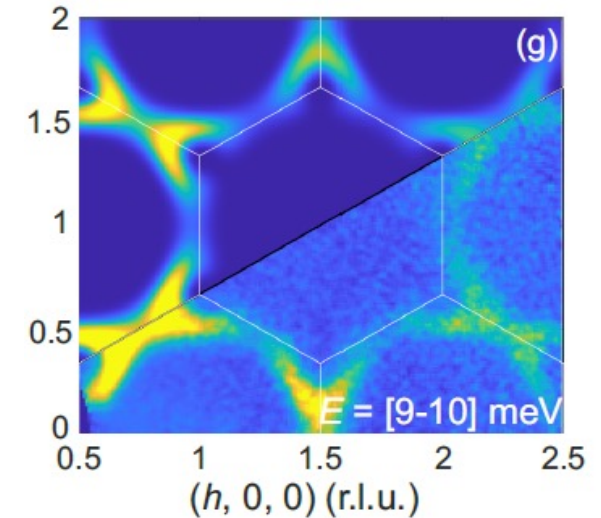
$$\Delta\tilde{\varepsilon}_{\mathbf{q}}(T) = \frac{\varepsilon_{\mathbf{q}}(0) - \varepsilon_{\mathbf{q}}(T)}{\varepsilon_{\mathbf{q}}(0)T^2}$$

cf. S. S. Pershoguba *et al.*, PRX **8**, 011010 (2018).

Dirac Magnons in CrBr₃

- CrBr₃ is a perfect realization of a quasi-two-dimensional honeycomb FM with Heisenberg exchange interactions
- Magnons show no gap at the K-point, and the previous report on the topological gap was most likely based on inaccurate data analysis of the TOF INS data
- The Dirac magnons at the K-point show clear winding of spectral intensity, predicted for nodal quasiparticles
- The linewidth and the dispersion bandwidth scale with T^2 , in agreement with theory
- The measured \mathbf{Q} -dependence of the linewidth and magnon renormalization lacks the predicted Van-Hove-like peaks indicating to the need for more sophisticated theoretical analysis

- S. Nikitin *et al.*, Phys. Rev. Lett. **129**, 127201 (2022).



PHYSICAL REVIEW LETTERS **131**, 246601 (2023)

Phonon Topology and Winding of Spectral Weight in Graphite

N. D. Andriushin¹, A. S. Sukhanov¹, A. N. Korshunov², M. S. Pavlovskii², M. C. Rahn¹ and S. E. Nikitin^{3,4,*}

¹Institut für Festkörper- und Materialphysik, Technische Universität Dresden, D-01069 Dresden, Germany

²Kirensky Institute of Physics, Siberian Branch, Russian Academy of Sciences, Krasnoyarsk 660036, Russian Federation

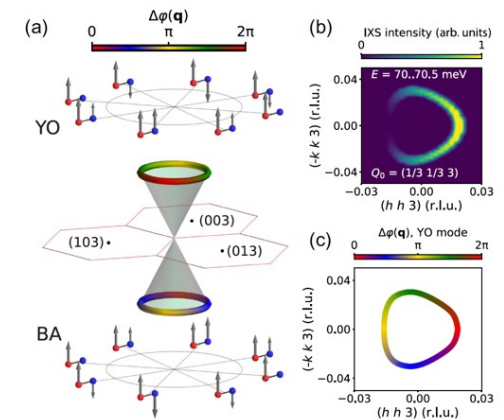
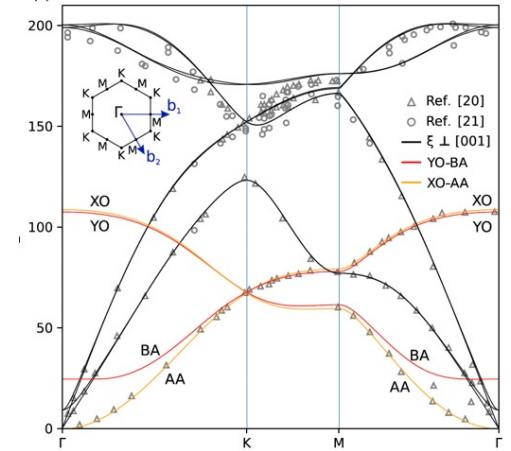
³Quantum Criticality and Dynamics Group, Paul Scherrer Institut, CH-5232 Villigen-PSI, Switzerland

⁴Laboratory for Neutron Scattering and Imaging, Paul Scherrer Institut, CH-5232 Villigen-PSI, Switzerland

(Received 17 March 2023; accepted 19 October 2023; published 14 December 2023)

The topology of electronic and phonon band structures of graphene is well studied and known to exhibit a Dirac cone at the K point of the Brillouin zone. Here, we applied inelastic x-ray scattering (IXS) along with *ab initio* calculations to investigate phonon topology in graphite, the 3D analog of graphene. We identified a pair of modes that form a very weakly gapped linear anticrossing at the K point that can be essentially viewed as a Dirac cone approximant. The IXS intensity in the vicinity of the quasi-Dirac point reveals a harmonic modulation of the phonon spectral weight above and below the Dirac energy, which was previously proposed as an experimental fingerprint of the nontrivial topology. We illustrate how the topological winding of IXS intensity can be understood in terms of atomic displacements and highlight that the intensity winding is not in fact sensitive in telling quasi- and true Dirac points apart.

DOI: 10.1103/PhysRevLett.131.246601



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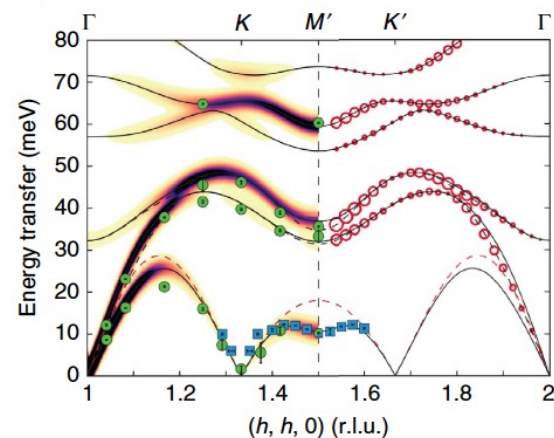
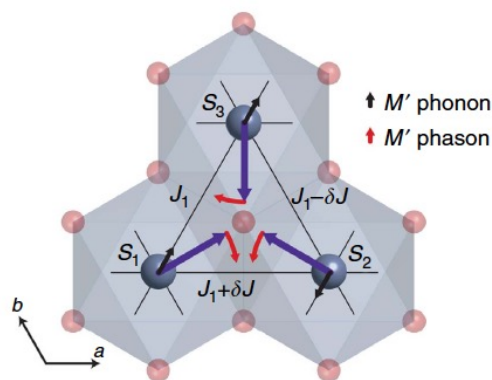
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Strong Spin-Lattice Coupling in LiCrO_2

2D Triangular Lattice - Electromagnons

Strong coupling of magnetism and structure probed with meV resolution by non-resonant inelastic X-ray scattering

Interplay of magnetism and structural properties are believed to be a key ingredient for technological applications of materials e.g. in multiferroics.

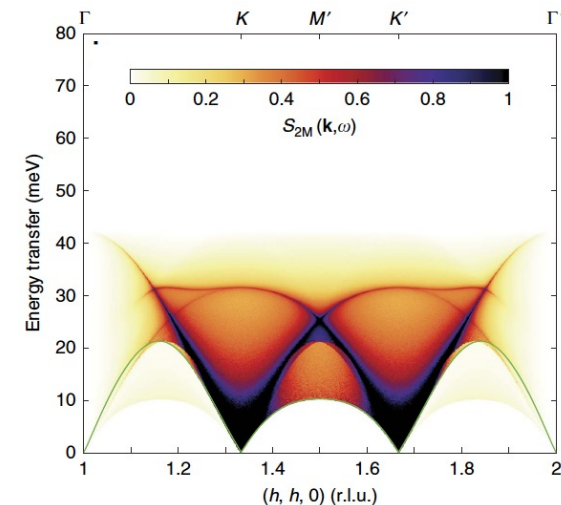


Electromagnon dispersion probed by inelastic X-ray scattering in LiCrO_2 . S. Toth, B. Wehinger, K. Rolfs, U. Stuhr, H. Takatsu, K. Kimura, T. Kimura, H.M. Ronnow, Ch. Rüegg, Nature Comm. **7**, 13547 (2016).

Strong coupling of magnetism and structure probed with meV resolution by non-resonant inelastic X-ray scattering

Interplay of magnetism and structural properties are believed to be a key ingredient for technological applications of materials e.g. in multiferroics.

- Direct exchange in LiCrO_2
- Strong mixing of matrix elements up to 30% possible
- Resolution of 1 meV to probe magnetism on ultra-small samples (hard X-ray IXS)
- Can be applied to many systems to probe both one- and multi-magnon processes, magnetoelastic coupling, etc.



Electromagnon dispersion probed by inelastic X-ray scattering in LiCrO_2 . S. Toth, B. Wehinger, K. Rolfs, U. Stuhr, H. Takatsu, K. Kimura, T. Kimura, H.M. Ronnow, Ch. Rüegg, *Nature Comm.* **7**, 13547 (2016).

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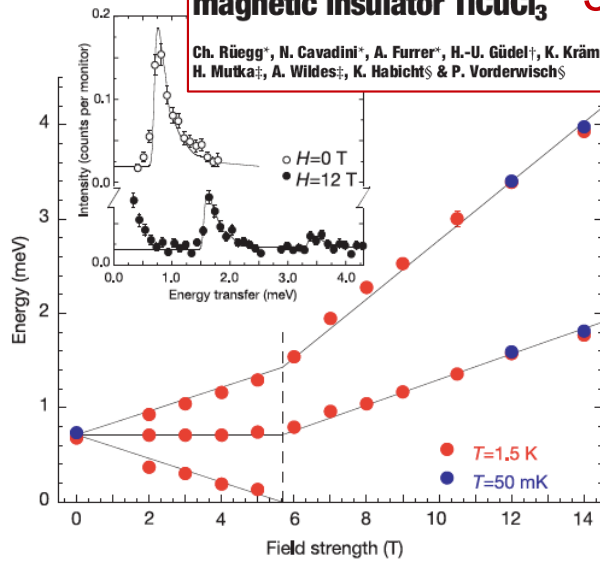
Quantum Criticality and Dynamics in 2D



Bose-Einstein Condensates, 1D, 2D, 3D Quantum Criticality, Spin Luttinger-liquids, Spin Liquids, etc

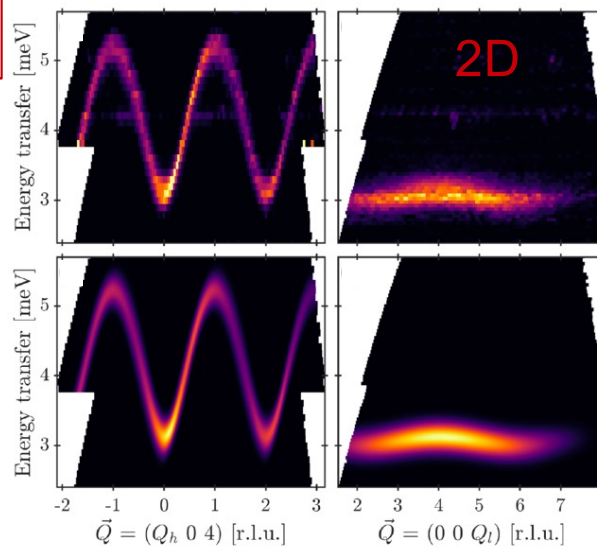
Bose-Einstein condensation of the triplet states in the magnetic insulator TlCuCl_3 3D

Ch. Rüegg*, N. Cavadini*, A. Furrer*, H.-U. Güdel†, K. Krämer†, H. Mutka‡, A. Wildes‡, K. Habicht§ & P. Vordenwisch§

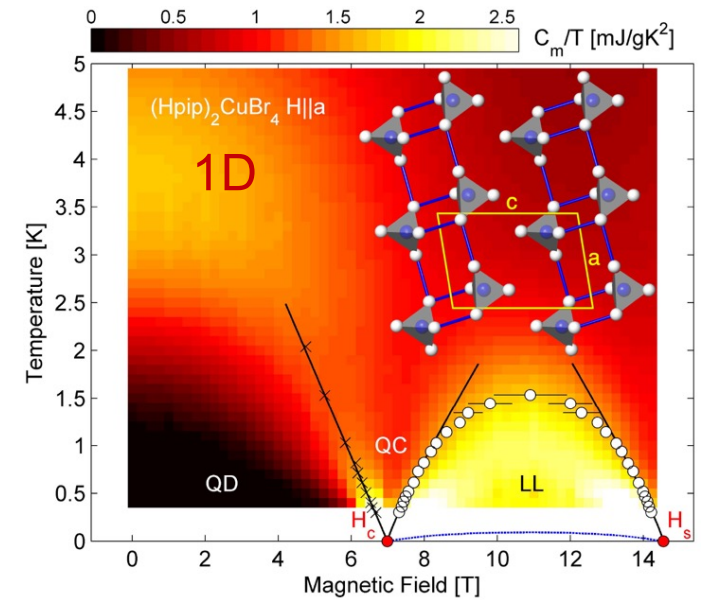


• Ch. Rüegg *et al.*, Nature **423**, 62 (2003).

$\text{BaCuSi}_2\text{O}_6$

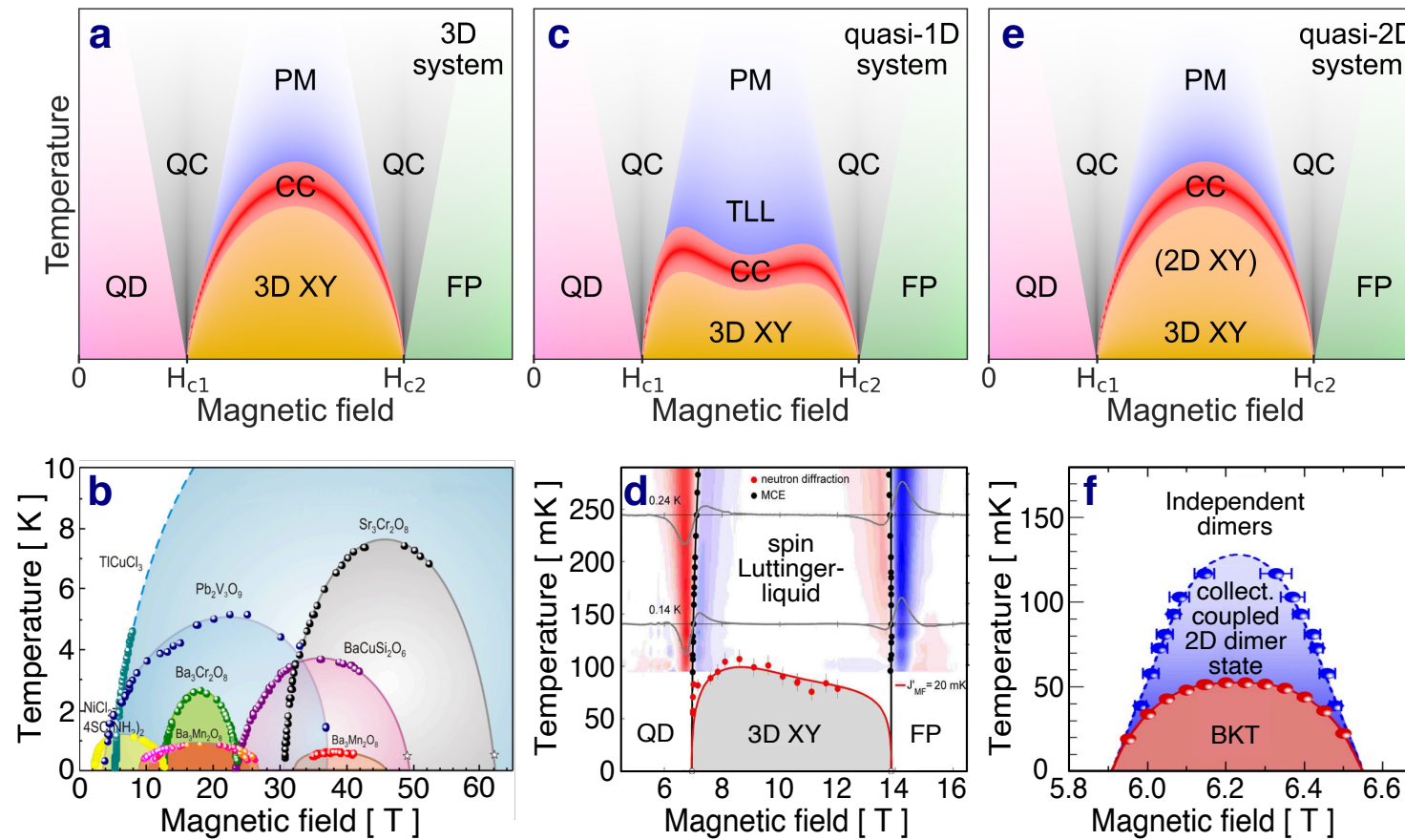


- S. Allenspach *et al.*, Phys. Rev. Lett. **124**, 177205 (2020).
- S. Allenspach *et al.*, Phys. Rev. Research. **3**, 023177 (2021).
- S. Allenspach *et al.*, Phys. Rev. B **106**, 104418 (2022).



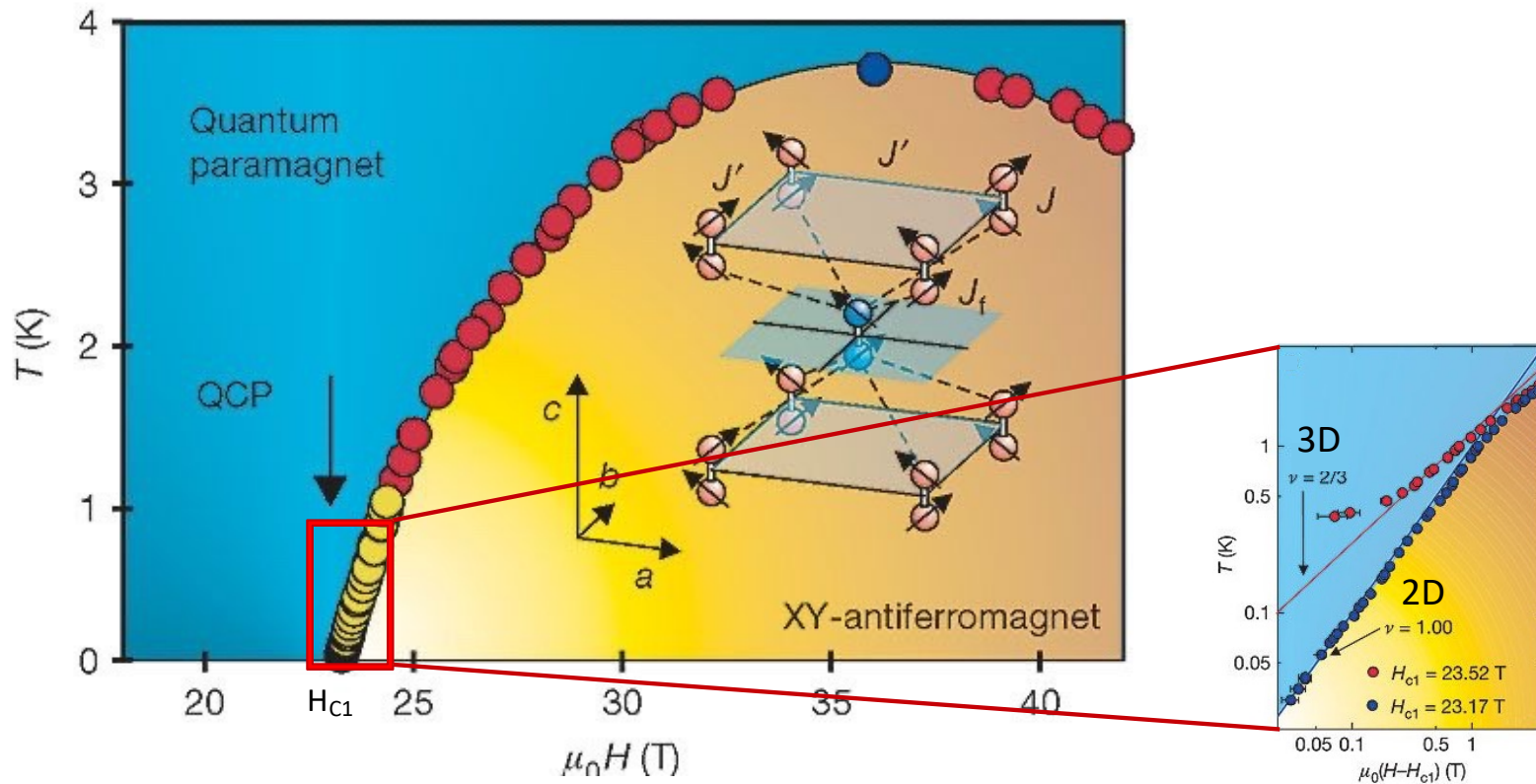
- Ch. Rüegg *et al.*, Phys. Rev. Lett. **101**, 247202 (2008).
- B. Thielemann *et al.*, Phys. Rev. B **79**, 020408(R) (2009).
- H. Ryll *et al.* Phys. Rev. B **89**, 144416 (2014).
- S. Ward *et al.* Phys. Rev. Lett. **118**, 177202 (2017).

Quantum Criticality in Spin-Dimer Systems



- V. Zapf *et al.*, *Rev. Mod. Phys.* **86**, 563 (2014).
- B. Thielemann *et al.*, *Phys. Rev. B* **79**, 020408(R) (2009).
- U. Tutsch *et al.*, *Nat. Commun.* **5**, 5169 (2014).











Dimensional Reduction (?) – BaCuSi₂O₆

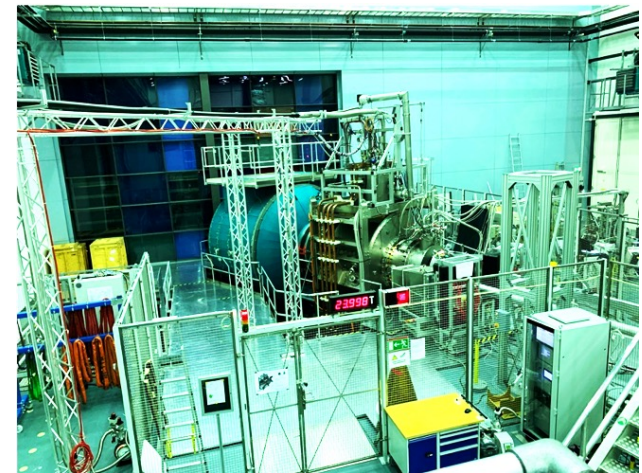
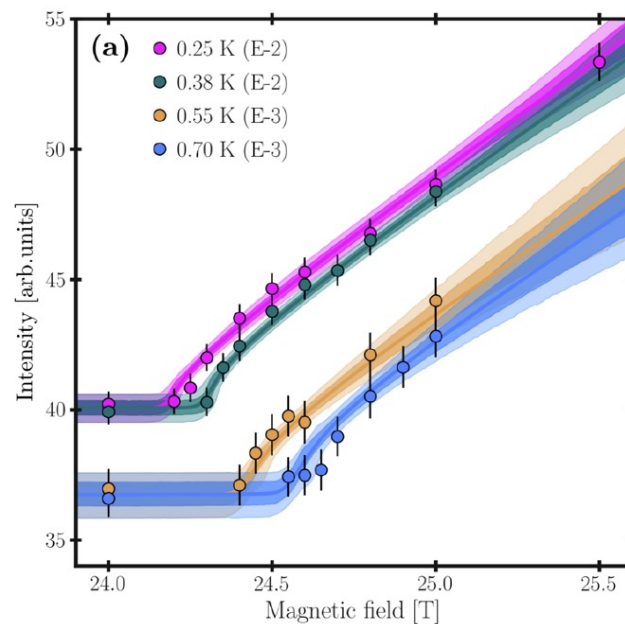
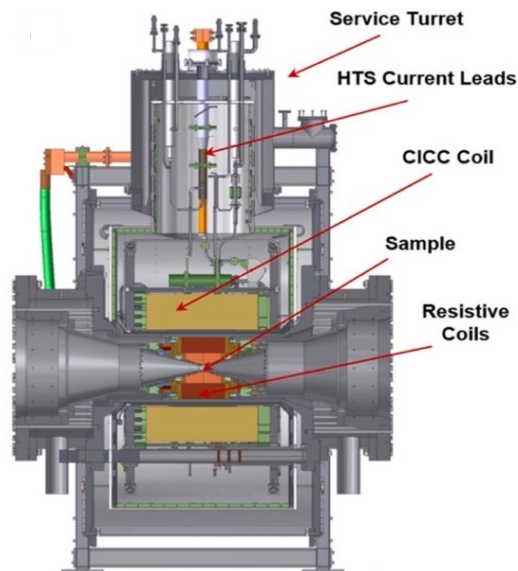


• S.E. Sebastian *et al.*, Nature **411**, 617 (2006).

Magnetic Order Parameter in $\text{BaCuSi}_2\text{O}_6$

Investigating field-induced magnetic order in Han purple by neutron scattering up to 25.9 T

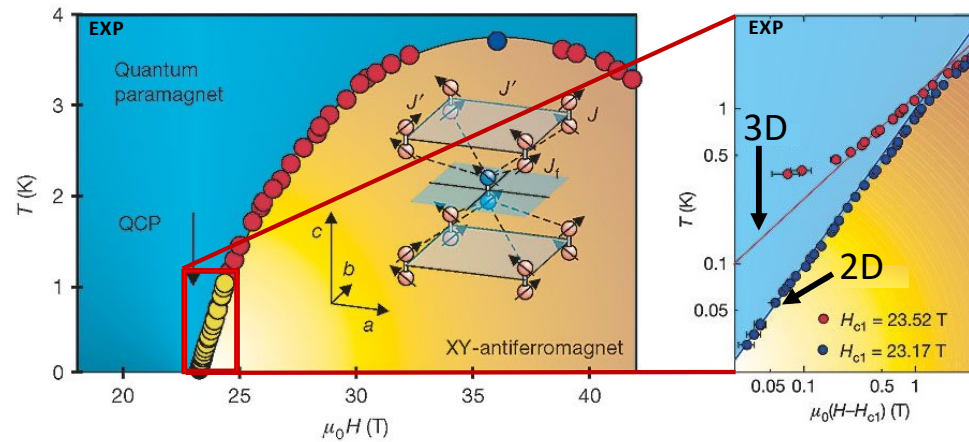
S. Allenspach ^{1,2}, A. Madsen ^{1,3}, A. Biffin,⁴ M. Bartkowiak ⁵, O. Prokhnenko ⁵, A. Gazizulina ⁶, X. Liu,⁷ R. Wahle,⁵
S. Gerischer,⁵ S. Kempfer,⁵ P. Heller,⁵ P. Smeibidl ⁵, A. Mira ^{8,9}, N. Laflorencie,¹⁰ F. Mila ¹¹,
B. Normand ^{1,11} and Ch. Rüegg ^{1,2,11,12}



• S. Allenspach *et al.*, Phys. Rev. B **106**, 104418 (2022).

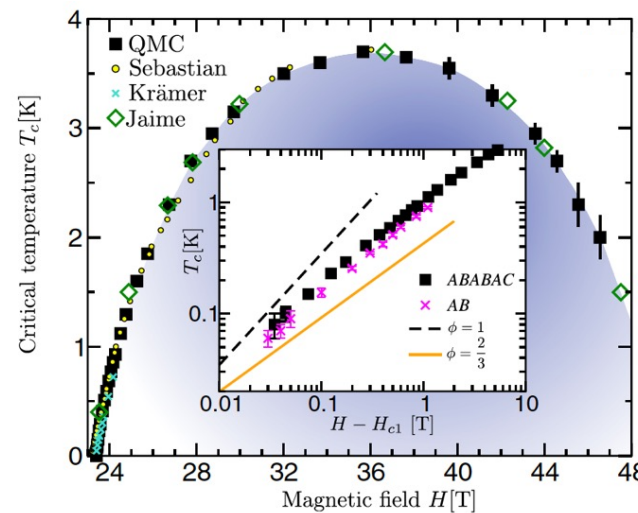
Phase Diagram from Quantum Monte Carlo

Experimental Phase Diagram



Quantum Monte Carlo Phase Diagram

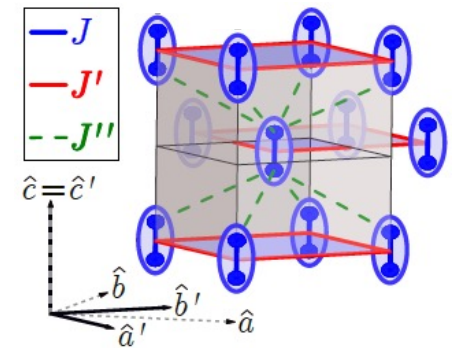
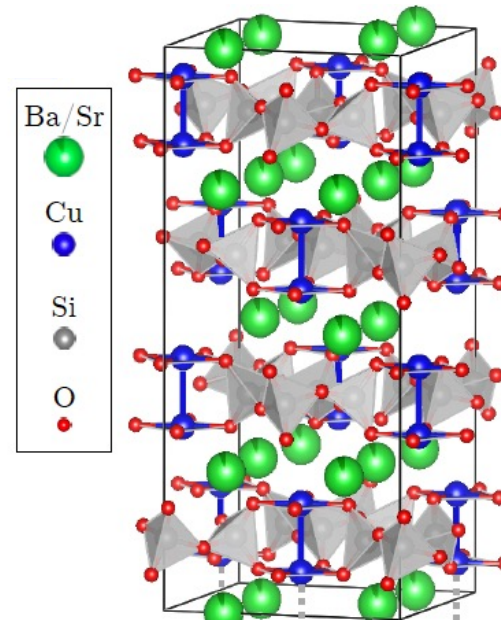
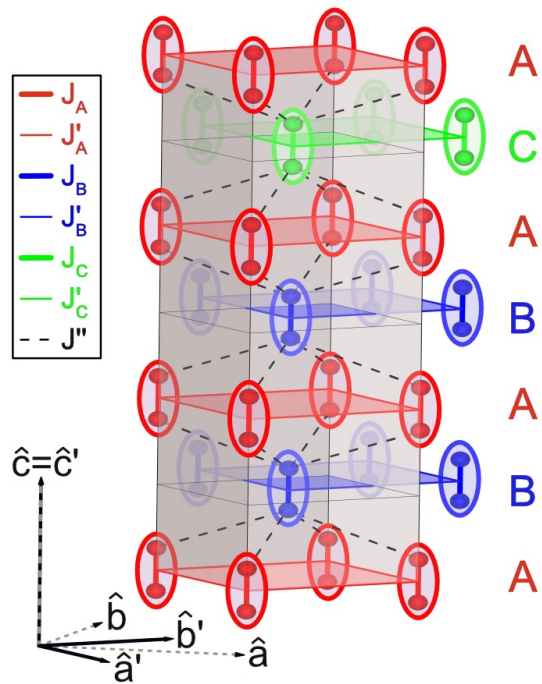
- ⇒ FM $J_{\text{intra-bilayer}}$
- ⇒ FM $J_{\text{inter-bilayer}}$
- ⇒ 3 dimer types with stacking



• S.E. Sebastian *et al.*,
Nature, **411**, 617 (2006).

• S. Allenspach *et al.*,
Phys. Rev. Lett. **124**, 177205 (2020).

Sr/Ba Substitution in BaCuSi₂O₆





















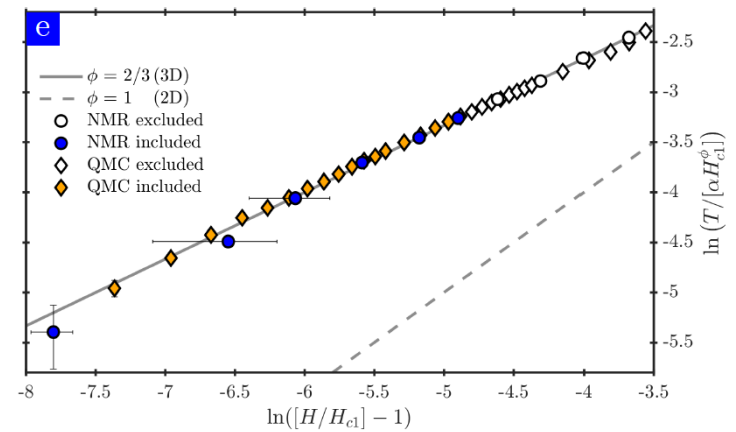
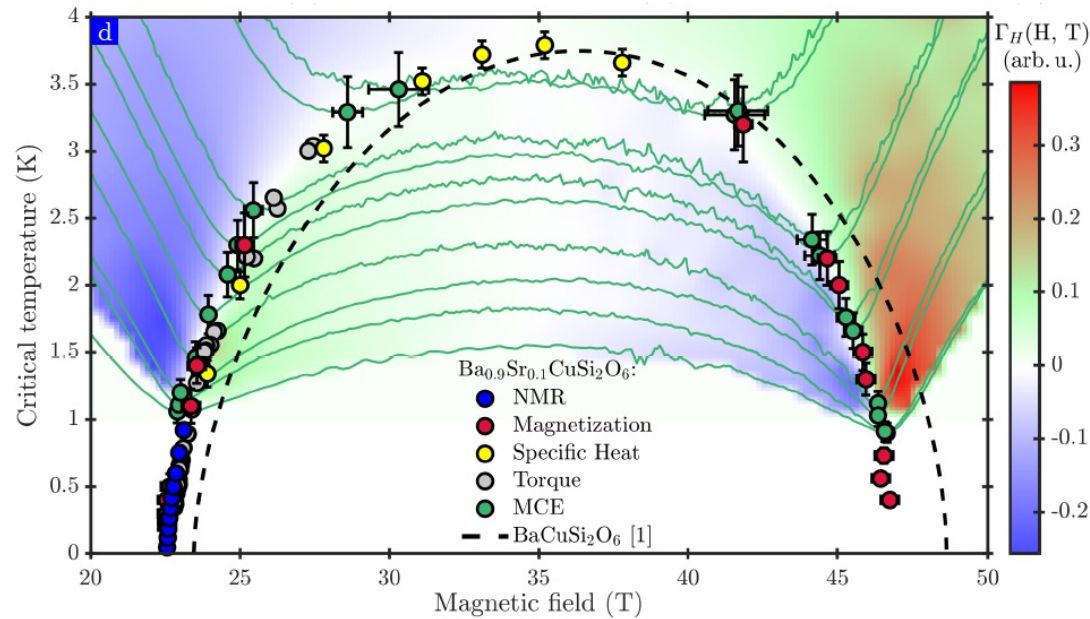
- S. Allenspach *et al.*, Phys. Rev. B **106**, 104418 (2022).
- S. Allenspach *et al.*, Phys. Rev. Lett. **124**, 177205 (2020).
- Ch. Rüegg *et al.*, Phys. Rev. Lett. **98**, 017202 (2007).

- S. Allenspach *et al.*, Phys. Rev. Research **3**, 023177 (2021).

3D Quantum Criticality in $\text{Ba}_{0.9}\text{Sr}_{0.1}\text{CuSi}_2\text{O}_6$

Revealing three-dimensional quantum criticality by Sr substitution in Han purple

Stephan Allenspach ^{1,2} Pascal Pupal,^{3,4,5} Joosep Link ⁶ Ivo Heinmaa ⁶ Ekaterina Pomjakushina ³
 Cornelius Krellner ⁴ Jakob Lass ^{7,8} Gregory S. Tucker,^{7,9} Christof Niedermayer ⁷ Shusaku Imajo ¹⁰
 Yoshimitsu Kohama ¹⁰ Koichi Kindo,¹⁰ Steffen Krämer ¹¹ Mladen Horvatić ¹¹ Marcelo Jaime,¹² Alexander Madsen ^{1,13}
 Antonietta Mira ^{13,14} Nicolas Laflorencie ¹⁵ Frédéric Mila ⁹ Bruce Normand ^{1,9,16} Christian Rüegg ^{1,2,9,17}
 Raivo Stern ⁶ and Franziska Weickert¹⁸



• S. Allenspach *et al.*, Phys. Rev. Research 3, 023177 (2021).

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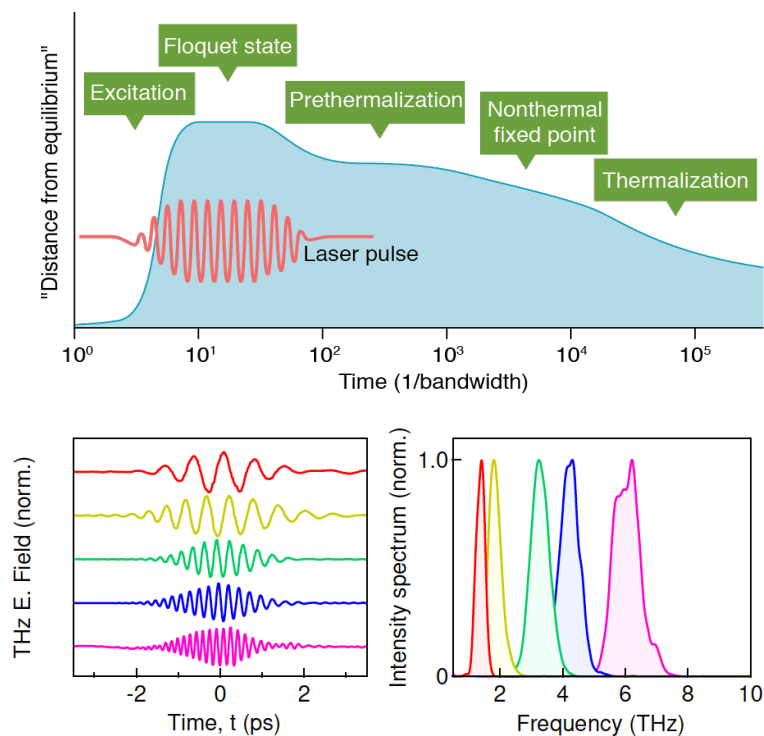
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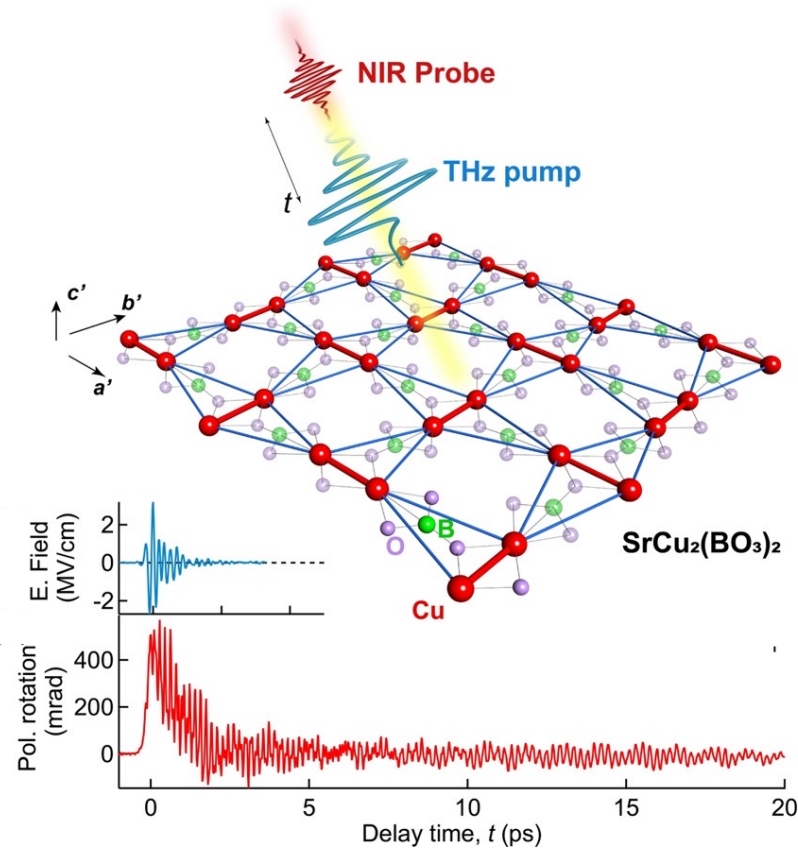


2D Quantum Magnets beyond Equilibrium

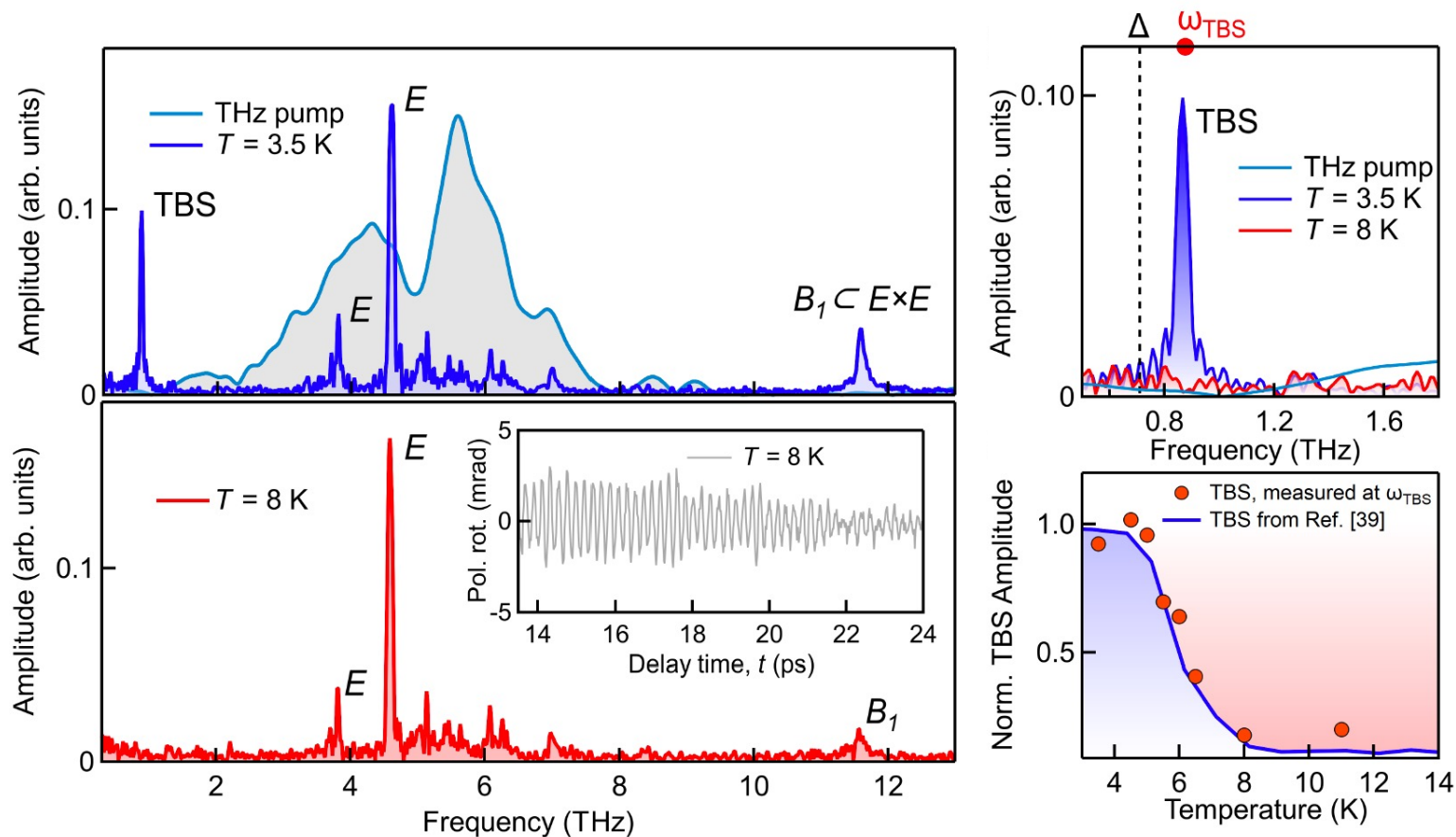
Physics beyond Equilibrium



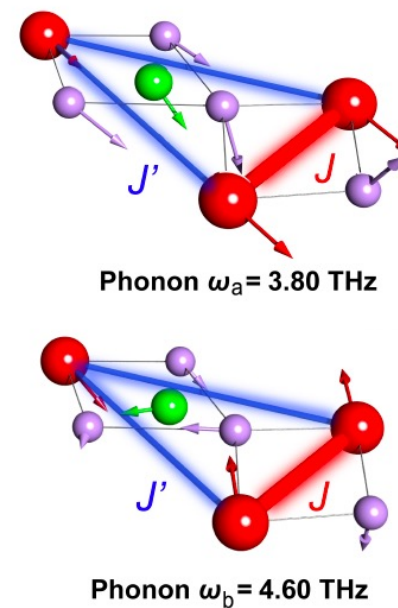
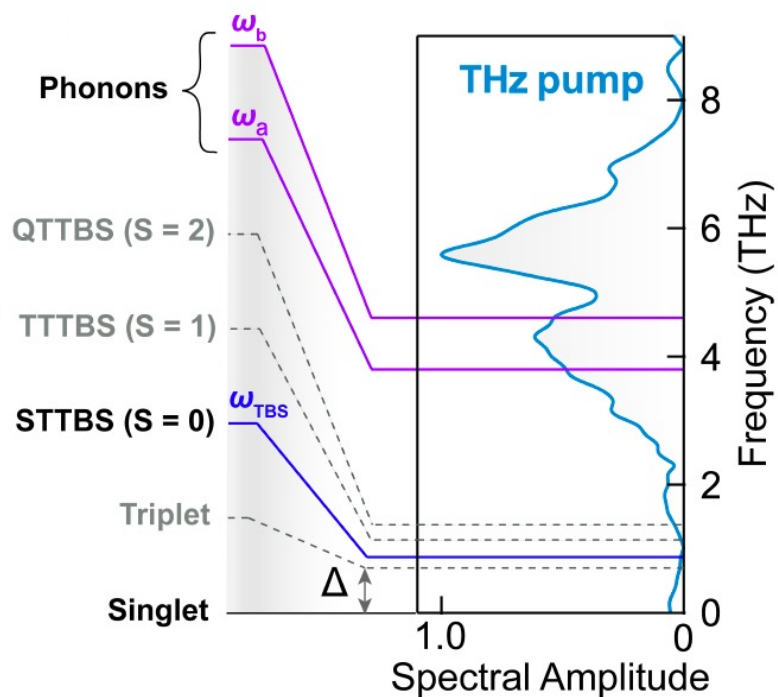
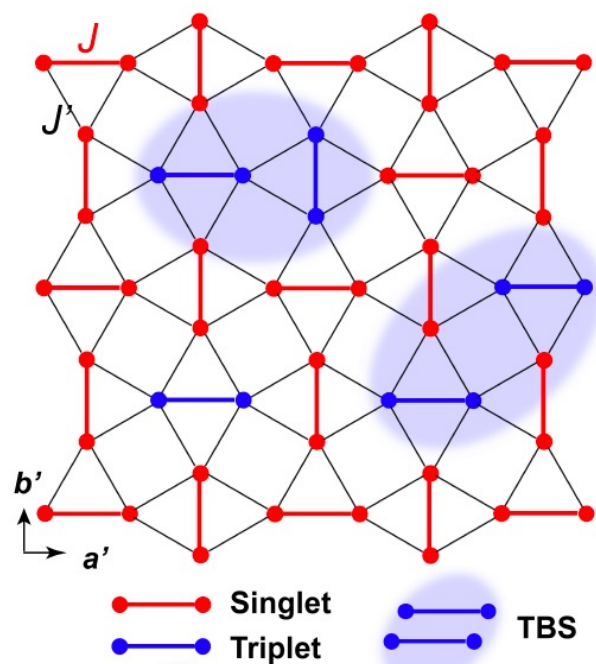
• C. Vicario *et al.*, Appl. Phys. Lett. **117**, 101101 (2020).



• F. Giorgianni *et al.*, Phys. Rev. B **107**, 184440 (2023).



• F. Giorgianni *et al.*, Phys. Rev. B **107**, 184440 (2023).



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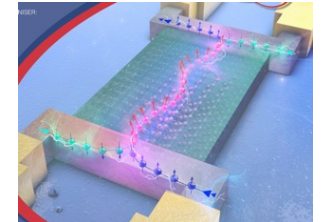
Devices from Quantum Magnets

LETTERS

PUBLISHED ONLINE: 26 SEPTEMBER 2016 | DOI: 10.1038/NPHYS3895

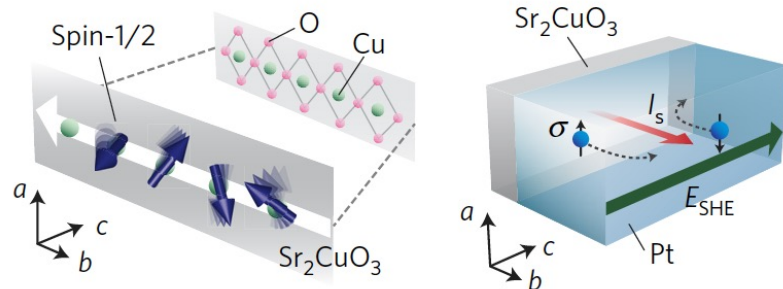
nature
physics

Switches for Quantum Spintronics ?



One-dimensional spinon spin currents

Daichi Hirobe^{1*}, Masahiro Sato^{2,3†}, Takayuki Kawamata⁴, Yuki Shiomi^{1,2}, Ken-ichi Uchida^{1,5}, Ryo Iguchi^{1,2}, Yoji Koike⁴, Sadamichi Maekawa^{2,3} and Eiji Saitoh^{1,2,3,6*}



• D. Hirobe *et al.*, Nature Physics **13**, 30 (2017).

(I)



(II)



(III)



(IV)



For (III) cf. D. Loss, Y. Tserkovnyak *et al.*

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- Introduction: Physics, Materials and Experiments
- Dirac Magnons in CrBr_3 and their Damping S. Nikitin *et al.*, Phys. Rev. Lett. **129**, 127201 (2022).
- Strong Spin-lattice Coupling in LiCrO_2 S. Toth *et al.*, Nature Comm. **7**, 13547 (2016).
- Quantum Criticality and Dynamics in 2D S. Allenspach *et al.*, Phys. Rev. Lett./Res. (2020/21).
- $\text{SrCu}_2(\text{BO}_3)_2$ Out-of-Equilibrium Magnons F. Giorgianni *et al.*, Phys. Rev. B **107**, 184440 (2023).

