



SrCu₂(BO₃)₂ - Dirac dispersion and other cool stuff

Laboratory for Quantum Magnetism (LQM)

Switzerland

Ecole Polytechnique Federale de Lausanne

Florida beach style







The Stars of the Show

Diane Lancon, $LQM \Rightarrow P$ Mohamed Zayed, $LQM \Rightarrow C$ Julio L. Larrea, $LQM \Rightarrow C$ Julio L. Larrea, $LQM \Rightarrow C$ Ellen FoghLQMGaetan GiriatLQMChristian Ruegg, $PSI \Rightarrow PS$ Katja PomjakushinaPSIAndreas LaeuchliU. InnsbFrederic MilaEPFLKazu KakuraiJ-PARC

+ many more: Crystal growers beamline scientists, colleagues, ... LQM \Rightarrow PSI LQM \Rightarrow Carnegie Mellon LQM \Rightarrow U. São Paulo LQM LQM PSI \Rightarrow PSI* PSI U. Innsbruck EPFL





RIKEN Center for Emergent Matter Science



The University of Tokyo The Institute for Solid State Physics Shastry-Sutherland – a special decoration of the square lattice

- Diagonal J > square J' \Rightarrow frustrated orthogonal dimers
- J'/J=0.5 "Shastry-Sutherland point": Ground state is product state of singlets on J-bonds
- Dimer singlet phase stable for range of J' & inter-layer J"





"geometrically broken symmetry" Dimer-covering dictated by model SrCu2(BO3)2 is physical realization of this model



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Slide 4



Exact Dimer Ground State and Quantized Magnetization Plateaus in the Two-Dimensional Spin System SrCu₂(BO₃)₂

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For: Exact dimer ground state and quantized magnetization plateaus in the two-dimemsional spin system SrCu2(BO3)(2). ... More

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"Correlated hopping"

Triplets almost localised only hop at 6th order in perturbation



Figure 2.24: Lowest-order hopping process for a singlet-triplet excitation to the next-nearest neighbour. $|t_m\rangle$ represents an $S^z = m$ triplet state and $|s\rangle$ is a singlet. Figure reformatted from topical review [47].





Direct Evidence for the Localized Single-Triplet Excitations and the Dispersive Multitriplet Excitations in SrCu₂(BO₃)₂

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Higher energies: a series of bound states



Need theory for dynamics structure factor



(a) Normalised LET dataset along the high symmetry directions and $E_i = 12 \text{ meV}$



(b) Calculated spectral weightby perturbative unitary transformation of an effective Hamiltonian, figure from [62]



Dzyaloshinskii-Moriya coupling





(a) Magnetic excitations spectra at 1.6K (b) Frequency-field diagram at 1.6K for $\mathbf{B}//\mathbf{c}$





Triplet wave theory

PHYSICAL REVIEW B 83, 024413 (2011)

Effect of Dzyaloshinskii-Moriya interactions on the phase diagram and magnetic excitations of SrCu₂(BO₃)₂

Judit Romhányi,^{1,2} Keisuke Totsuka,³ and Karlo Penc¹



$$\mathcal{H} = J \sum_{n.n.} \mathbf{S}_{i} \cdot \mathbf{S}_{j} + J' \sum_{n.n.n.} \mathbf{S}_{i} \cdot \mathbf{S}_{j} - h_{z} \sum_{i} S_{i}^{z} \\ + \sum_{n.n.} \mathbf{D}_{ij} \cdot (\mathbf{S}_{i} \times \mathbf{S}_{j}) + \sum_{n.n.n.} \mathbf{D}'_{ij} \cdot (\mathbf{S}_{i} \times \mathbf{S}_{j})$$
Ground state: $|\Psi\rangle = \prod |\tilde{s}\rangle_{A}|\tilde{s}\rangle_{B}$
 $|\tilde{s}\rangle_{A} = |s\rangle_{A} + \lambda |t_{y}\rangle_{A}$
 $|\tilde{s}\rangle_{B} = |s\rangle_{B} - \lambda |t_{x}\rangle_{B}$
 $E_{0} = \frac{\langle \Psi | \mathcal{H} | \Psi \rangle}{\langle \Psi | \Psi \rangle}$ minimization gives $\lambda = -\frac{D}{2J}$

JR et al PRB 83 024413 (2011)

 $|t_z\rangle = -\frac{i}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle)$

the new triplet basis:

$$\begin{split} |\tilde{t}_{x}\rangle_{A} &= |t_{x}\rangle_{A} & |\tilde{t}_{x}\rangle_{B} = \lambda|s\rangle_{B} + |t_{x}\rangle_{B} \\ |\tilde{t}_{y}\rangle_{A} &= -\lambda|s\rangle_{A} + |t_{y}\rangle_{A} & |\tilde{t}_{y}\rangle_{B} = |t_{y}\rangle_{B} \\ |\tilde{t}_{z}\rangle_{A} &= |t_{z}\rangle_{A} & |\tilde{t}_{z}\rangle_{B} = |t_{z}\rangle_{B} \end{split} \qquad \begin{aligned} |t_{x}\rangle &= \frac{i}{\sqrt{2}}(|\uparrow\uparrow\rangle - |\downarrow\downarrow\rangle) \\ |t_{y}\rangle &= \frac{1}{\sqrt{2}}(|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle) \end{aligned}$$

correspond to the 6 different `triplet'-like excitation

Triplet Wave theory: Romhanyi, Penc et al



analytical solution:

geometrical factors:

 $\gamma_1 = \cos\frac{k_x}{2}\cos\frac{k_y}{2}$

 $\gamma_3 = \sin \frac{k_x - k_y}{2}$



with

$$\Omega_{\pm} = \sqrt{(h_z \pm 2D'_{\perp}\gamma_1)^2 + \frac{D^2 J'^2}{4J^2}(\gamma_2^2 + \gamma_3^2)}$$

full bond-wave theory gives the same results, linear in D





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Hall effect of triplons in a dimerized quantum magnet Predict field-tuned Dirac dispersion



Figure 2 | Topological transitions in the triplon bands. (a) A spin-1 Dirac cone with three bands touching. (b) Triplon dispersion for $h^2 = 0$. The basal plane shows the enlarged BZ corresponding to one dimer per unit cell, with $k = (\pi, \pi)$ at the M and $k = (\pi, 0)$ and $(\pi, 0)$ at the X points. The smaller structural BZ is shown in green. The band structure hosts spin-1 Dirac cones at the BZ edge centres X. (c-e) Evolution of triplon bands and Chern numbers upon tuning magnetic field. Bands with non-zero Chern number appear for $0 < h^2 < h_c$ and are shown in colour. At $h^2 = h_c$ (d), the bands touch at a spin-1 Dirac cone at Γ . For $h^2 > h_c$ as in e, the Chern numbers remain zero.



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Predicts protected edge states & thermal Hall effect



Figure 4 | Protected edge states. (a) Band structure of $SrCu_2(BO_3)_2$ on a cylindrical strip periodic in *x*, but with open edges along *y*. The width along *y* is taken to be very large. We recover the bulk states corresponding to three triplon bands. In addition, four edge states appear, connecting the Chern bands. Edge modes shown in blue and cyan (red and brown) are leftmovers (right-movers). (b) Wavefunctions of the four edge states for an arbitrary k_x on a strip of width W = 8 dimers. The colour of the dimer bond represents the triplon weight with black corresponding to zero. Rightmoving edge states are localized on the bottom edge, whereas left-movers are localized on the top edge.



Figure 5 | Thermal Hall effect. (a) Thermal Hall conductivity versus external magnetic field at different temperatures. The threshold fields $h^z = \pm h_c$ are shown as vertical dashed lines. (b) Thermal Hall signal at $h^z = h_c/2$ versus temperature. The circles are direct evaluation of the formula, the line is an approximation applicable to SrCu₂(BO₃)₂ in the limit where bandwidths are much smaller than the gap. (c) The filling fraction (boson occupation number) for the three bands, indexed by Chern numbers. It grows much slower with temperature than κ^{xy} .





Figure 2.48: Calculated dispersion (red lines) superimposed on the IN5 excitation spectra around Γ at $Q = (0 \ 2 \ 0)$. a) no applied field with three modes with lower and upper mode degenerate between $S^z = \pm 1$. b) At 0.7T, the degeneracy is lifted and the $S^z = \pm 1$ modes split. c) 1.4T : Critical field with Dirac crossing at the Brillouin zone center Γ . d) 2.1T : trivial band gap opens up and modes are split further.

Topological triplon modes and bound states in a Shastry-Sutherland magnet

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Summary: Dirac physics in SrCu2(BO3)2

- Predicted Dirac dispersion in SCBO
- Confirmed with neutron scattering (us + Coldea et al)
- Predicted thermal Hall effect
- Experiments ongoing, has been observed in other materials
- Predicted topological surface states how measure

Considerations:

- Triplet are quasi-localized, how interesting is Dirac?
- Dirac is only 2 of 5 modes around 3meV, hard to control population thermal or pumped





Unusual temperature damping







Two-component line-shape







Unusual temperature damping

PRL 113, 067201 (2014) PHYSICAL REVIEW LETTERS

week ending 8 AUGUST 2014



Correlated Decay of Triplet Excitations in the Shastry-Sutherland Compound $SrCu_2(BO_3)_2$



Correlated decay

- Localized triplet radius
- If neutron-triplet touches thermal triplet => decay





• Probability of decaying with an existing singlet:

$$P_s(T) = n_s(T)^{2\pi(2R/a)^2}$$





Spin-dependent correlated decay

LQ



b) $Q = (0.75 \ 0.75), 8T$

• T⁰

Summary: Correlated decay

- Localized triplets at 3meV=35K
- Strong damping already at 6K
- We propose "correlated decay model"
 - Fits temperature and field dependence
- Spin-dependent correlated decay
 - $-T_{-}$ pushed up in energy by hardcore repulsion
 - $-T_{+}$ decay faster because interact with T₋ population
- Our model is unfounded need theoretical work
- Probably need to take multi-triplets into account





What happens in high magnetic fields?



(a) Magnetic excitations spectra at 1.6K (b) Frequency-field diagram at 1.6K for $\mathbf{B}//\mathbf{c}$

- Both triplet and 2-triplet gap closes
- Plateaus in magnetization





Magnetization plateaus

- Many new phases in high magnetic field
- Condensing triplets into ground state



NMR and magnetization in very high fields

Magnetic Superstructure in the Two-Dimensional Quantum Antiferromagnet SrCu₂(BO₃)₂

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HZB Research Proposal

HFM-191-00023

Experiment title: 1/8 plateau phase in Shastry-Sutherland compound SrCu2(BO3)2	Proposal type:	Standard (ST)
	Scientific College:	HFM

Abstract:

We propose to investigate the coveted 1/8 magnetization plateau phase in the unique Shastry-Sutherland compound SrCu2(BO3)2. We have designed a 10kbar pressure cell optimized for this specific experiment, which will bring the 1/8 phase down to accessible 25.5T. We propose a diffraction part to search for the ordering vectors of the 1/8 phase and an inelastic part to investigate the new excitation spectrum in the 1/8 phase. The proposing team bring together all the knowhow needed for this challenging experiment to succeed.

Proposer (All correspondence concerning this proposal will be sent to the proposer)				
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Investigating the 1/8 magnetization plateau in SrCu₂(BO₃)₂ using *high field*, *high pressure* and *low temperature*



Takigawa et al., PRL **110**, 067210 (2013) Corboz and Mila, PRL **112**, 147203 (2014) Schneider et al., PRB **93**, 241107(R) (2016)

Goal: neutron scattering investigations of the 1/8 plateau phase **Required conditions:** 26T, 1GPa, 200mK







Results from the 1/8 phase:

Probably no 1/8 Bragg peaks, but we are still digging in the data





Spectroscopy to 26T @ 100mK (P=0)



- Triplets fade: temperature or "tower of states"?
- Evidence of new ground state?
- Analysis and theory underway





Summary high magnetic fields:

- Neutrons at 25T is challenging, and no longer available !
- I have project for neutron diffraction to 60 T and spectroscopy to 40T (pulsed magnet with dedicated neutron source)
- Just need 25M\$ and about 10 years...
- Meanwhile, what about just pressure?





Intermediate phase ?

$J'>>J \Rightarrow$ Square lattice AFM SCBO close



Table 2. Phase boundary points calculated by several thec intermediate phase transition. The phase transition is alsc_

the Sp(N) Shastry–Sutherland model) and [53] (field theory approach for generalized Shastry–Sutherland models). Since the model is generalized in both methods, we have not included their results in the table.

Main method	$(J'/J)_{c1}$	$(J'/J)_{c2}$	Intermediate phase
Variational method [36]	0.5		_
Schwinger boson mean field theory [49]	0.6	0.9	Helical ordered state
Exact diagonalization (up to 20 sites) [35]	0.70(1)		
Ising expansion [46]	0.691(6)		
Dimer expansion [47]	0.697(2)		
Plaquette expansion [39]	0.677(2)	0.86	Plaquette singlet
Series expansion [54]	0.69	0.83 or —	Columnar or —
Exact diagonalization (up to 32 sites) [52]	0.67	Bigger than 0.71	Plaquette singlet

Miyahara et Ueda, J. Phys.: Condens. Matter 15 (2003) R327–R366





Pressure tuning of interactions?

- Predict how pressure tunes balance of J's ?
- Need to be very lucky !







Many other works on SCBO under pressure

- Haravifard: X-ray (using structure as proxy for singlet population)
- Waki: NMR (disadvantage need symmetry breaking field)
- Sakurai: ESR (also needs finite magnetic field)
- Guo: Specific heat (integrates over excitation spectrum)
- Zheludev: Raman
- [10] S. Haravifard, D. Graf, A. E. Feiguin, C. D. Batista, J. C. Lang, D. M. Silevitch, G. Srajer, B. D. Gaulin, H. A. Dabkowska, and T. F. Rosenbaum, Nat Commun 7, 11956 (2016).
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Susceptibility to 10 kbar

• Fit to ED-calculations + RPA for inter-layer



Extract and extrapolate pressure dependence of J and J' It seems $\alpha = J'/J$ is increasing !





Inelastic Neutron Scattering: gap decrease with pressure !

- New Steel-Aluminum for inelastic neutron scattering
- P max ~17kbar, sample volume 1.5cm³ (cell by Ravil Sadykov)
- Gap decreases with pressure ☺







Linear softening up to 16kbar















However, there is a new lower-E excitation ©







Are we in a new phase?

2-triplet bound state almost catches up with 1-triplet 2Δ - $E_b = \Delta$ Indeed, E_b seems almost pressure independent







Energy match theoretical phase diagram





LAM

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Zero pressure structure factor

- Not simple sum of orthogonal cosines
- Until we have good model for P=0
 S(q) cannot hope for P>P_c
- So lets compare expt P=0 to P>P_c structure factors





- Δ excitation same S(q) as triplet at P=0
- LE excitation peaked near (1,0,0)







Simple 4-spin calculation: Singlet GS

- Full plaquette: T₁ triplet and T₂ quartet excitations
- S(q): T₁ peaks near (1,0,0), T₂ identical P=0 S \rightarrow T



- S(q): T_1 as full plaquette, T_2 different from P=0 S \rightarrow T
 - Data confirm 4-spin plaquette singlet ground state
- Data most consistent with singlets residing on full plaquettes

Empty of full plaquettes?

Competition between intermediate plaquette phases in $SrCu_2(BO_3)_2$ under pressure

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Peierls-like distortion



0.92

0.9

0.88

1

FPP/

0.74

dimer

0.7

 J_1'/J

0.66

Haldane

0.78



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Haldane

iPEPS ----SE ---+---

1.02 1.04 1.06 1.08 1.1 1.12

 J'_{1}/J'_{2}

What about AFM order at higher α ?









PE cell – new (100) peak

Consitent with AFM order above 40 kbar





However, structural transition around 5GPa

• Monoclinic above 4.7GPa at room temperature



Physica B: Condensed Matter Volumes 359–361, 30 April 2005, Pages 980-982



Crystal structure and lattice dynamics of ${\rm SrCu}_2({\rm BO}_3)_2$ at high pressures

I. Loa ª A 🖾, F.X. Zhang ª, K. Syassen ª, P. Lemmens ª, W. Crichton ^b, H. Kageyama ^c, Y. Ueda ^d



So, still Shastry-Sutherland Above 40kbar and T<100K ?





Temperature dependence of the pressure induced monoclinic distortion in the spin S = 1/2 Shastry–Sutherland compound $SrCu_2(BO_3)_2$

M.E. Zayed ^{a,b,c,*}, Ch. Rüegg ^{c,d,e}, E. Pomjakushina ^f, M. Stingaciu ^{f,g}, K. Conder ^f, M. Hanfland ^h, M. Merlini ^h, H.M. Rønnow ^b



Solid State Communications 186 (2014) 13-17

Structural transition comes close but never below 4.0 GPa Hence appear to be above AFM transition





9

Phase diagram:



CAMEA Concept (Continuous Angle Multiple Energy Analysis)

Cover several final energies by a series of vertically scattering analyzers

Cover large range of scattering angles by analyzer arcs



- Data on single scattering plane \Rightarrow Ideal for extreme environments
- But, CAMEA competitive for "all" experiments on small samples, parametric studies, fast mapping etc.
- \Rightarrow Factor 10 100 better if (**q**,E) covered useful

F. Groitl et al., Review of Scientific Instruments, 87, 035109 (2016)





CAMEA at PSI











 MnF_2







$SrCu_2(BO_3)_2$







Extreme sample environments

- Data focused on single scattering plane (3-135° coverage)
- Ideal for extreme environments





High-pressure anvil cells



- Split-coil high field magnets
- But, CAMEA competitive for "all" experiments on small samples, parametric studies, time resolved etc.





Summary:

- ✓ Dirac dispersions in SCBO
- ✓ Correlated decay: unusual thermal lineshapes
- ★ High field 1/8 plateau state
- ✓ QPT to Plaquette singlet state & Neel state
- ✓ CAMEA spectrometer & crazy idea for 40T

Come work with us! (<u>Henrik.Ronnow@epfl.ch</u>)

CAMEA neutron postdoc New project HERO on composite order and excitations: 2 PDOC positions 2 PhD students

Physicist/engineer on innovation project (ski-mountaineering & olympics)



Thank you !



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Slide 59