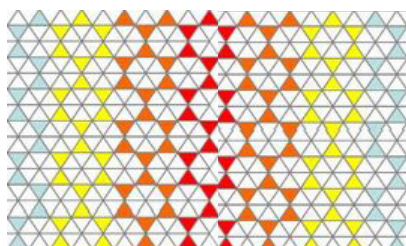
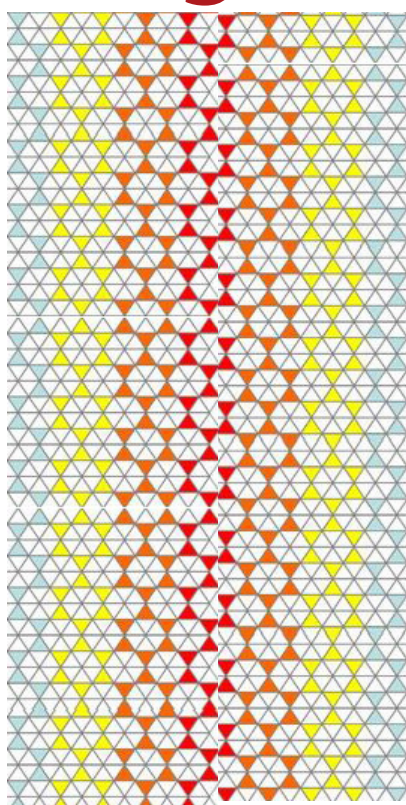


Scientific Report on  
« Novel physics in Kagome Compounds »

An *ESF-HFM* workshop  
held at  
Laboratoire de Physique des Solides, Orsay, France  
18 – 20 january 2010



Novel physics on the  
**Kagome Network**



January 18-20, 2010  
Orsay

[www.lps.u-psud.fr/kagome](http://www.lps.u-psud.fr/kagome)

**Scientific committee**

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# Summary

## Focus

The unprecedented recent burst in materials synthesis of quantum and classical kagome networks, which has served for long, as the ideal playground for geometrically frustrated antiferromagnets, has been rapidly followed, since 2006, by a vast panel of experimental investigations and has triggered novel theoretical scenarios and a critical reinvestigation of older ones. Although the gap between theories and experiments is presently closing, more than 30 years after the seminal ideas of P.W. Anderson, there are still many open issues about the nature of the possible ground states and the ways to probe them.

In this context, the focused and timely ESF-HFM workshop aimed at gathering most of the actors in the field from inside Europe, ie all ESF-HFM members working in this field, as well as from outside Europe, mainly from USA and Japan, and at covering the various facets of the recent work from bulk and artificial materials to theory.

## Organization :

The workshop ran over 2.5 days starting on the morning of monday 18th january and ending after lunch time on wednesday 20th january 2010. It was co-sponsored by CNRS through its special grant supporting the Highly Frustrated Magnetism activity, the RTRA “Triangle de la physique” and the French Physical Society, SFP.

A broad diffusion of the workshop was ensured through the mailing list of the ESF-HFM network and a website was set-up and updated until the workshop started: <http://www.lps.u-psud.fr/kagome>. All potential participants have been reached.

The workshop took place at Laboratoire de Physique des Solides, located on the campus of Paris-Sud University, Orsay, approximately 25 km in the south-west of Paris. Participants were lodged in two nearby hotels, Hôtel d’Orsay (Orsay) and Resid’Home (Bures/Yvette).

Out of the 54 Delegates, 46 came from institutes within our network (France, UK, Germany, Switzerland, Israel, Slovenia) and 8 colleagues from overseas (USA and Japan) were invited. Their travel expenses were covered through the supporting grant from “Triangle de la Physique”. As many as 19 nationalities were represented!

24 participants were experimental physicists, 24 were theorists and 6 were materials scientists/chemists. All participants had accommodations and meals paid for, Israeli participants had their travel expenses partly supported. In addition to overseas speakers, there were 5 invited contributions from our network.

The workshop was organized along several topics, novel kagome spin  $\frac{1}{2}$  physics: models and materials; anisotropic rare earth and cobalt based kagome systems; perturbations to the ideal case; new materials. Theory and experimental/materials talks were intermixed and a buffet poster session was organized on the first evening in order to favour deep exchanges between all participants. This quite focused workshop occurred at the right time where the community working in that field needed to sort out and discuss in details experiments and various proposed models. The special focus on kagome physics, which represents a particular sub-field of highly frustrated magnetism, has been warmly welcome by the participants who were quite prompt and numerous to reply to the first call. In general there was much lively discussion and debate about pending issues. Covering the state of the art in the field of kagome physics, the opportunity for participants to start new collaborations and discussion among various generations of researchers worldwide represent the main outcomes.

### **Scientific Committee :**

- F. Bert, LPS (Orsay, France)
- Ph. Mendels, LPS (Orsay, France)
- G. Misguich, IPhT (CEA Saclay, France)
- A. S. Wills, University College London (London, UK)

### **Local organization:**

F. Bert, B. Daly, M.F. Mariotto and P. Mendels

### **Invited speakers from overseas**

- J. Cumings (University of Maryland): artificial lattices
- J. Gardner (NIST and Indiana University): langasites
- J. Helton (NIST, formerly at MIT): herbertsmithite
- M.A. Hermele (Univ. of Colorado): novel models
- Z. Hiroi (Tokyo University): materials synthesis and volborthite
- R.R.P. Singh (Univ. California, Davis): novel models
- O. Tchernyshyov (John Hopkins University): fermionic excitations

# Scientific Content and Discussion

The physics of frustrated quantum magnetism has recently been boosted on the experimental side by the discovery of novel antiferromagnetic kagome materials which offer alternatives to the well celebrated Néel ground state. Hence the presentation sorted through most representative materials and corresponding models.

## **Herbertsmithite and Kagome Heisenberg Antiferromagnet (KHAF models)**

Overall, ca 40% of the workshop was devoted to the novel structurally perfect kagome spin  $\frac{1}{2}$  material named Herbertsmithite which opened a new avenue in the kagome and more generally novel highly frustrated magnetic states worlds. The session allowed to review the various models for the perfect kagome Heisenberg antiferromagnet and to draw the contours of the real Hamiltonian. This material was first synthesized at MIT and reproduced by various groups of the ESF-HFM network. J. Helton (MIT) opened the workshop discussing his seminal PhD work and a more recent attempt to unravel quantum critical behaviour by tentative scalings of the dynamical susceptibility and low temperature magnetization measurements. Next, in a review spirit, C. Lhuillier (Paris) sketched a state of the art picture of the gap issue and correlations in the KHAF. A. Olariu (Orsay and Lausanne) and M.A. de Vries (Edinburgh and Lausanne), presented their set of PhD seminal results which initially gave a strong support to the absence of a gap and the existence of inter-site mixing which complicates much the ideal Hamiltonian. P. Lemmens (Braunschweig) presented his innovative recent Raman scattering results, paving the way to a direct probe of models through the Raman response. A. Keren (Haifa) made a point which was heavily discussed about a strong anisotropy in Herbertsmithite obtained through magnetization measurements on oriented powders. M. Hermele (U. Colorado) and R.R.P. Singh (U. California) proposed respectively algebraic spin liquid and valence bound crystal ground states for the KHAF. Through a Quantum Dimer Model approach, D. Poilblanc and M. Mambrini (Toulouse), pointed out that the KHAF might be at a quantum critical point between two 36-site unit cell Valence Bond Crystal. Turning to the defects, O. Cepas (Grenoble) examined in detail the consequence of Dzyaloshinskii-Moriya anisotropy on the theoretical phase diagram, with the appearance of a purely quantum critical point and discussed the analysis of ESR lineshapes. The effect of spin vacancies evidenced through NMR measurements (see talk by A. Olariu) was discussed by F. Mila (Lausanne) using exact diagonalization methods. Finally, O. Tchernyshyov (Baltimore) presented his work on exotic elementary spinon excitations for the KHAF.

The **Volborthite** compound is known as the purest spin  $\frac{1}{2}$  kagome-based compound but with a distorted lattice. Z. Hiroi, in his invited talk, gave an overview of the quite original H-T phase diagram recently found out through High Field Magnetization measurements which still lacks some theoretical interpretation. The essence of the low-T H=0 phase starts being better captured with a short range order which requires two propagation vectors to be described and a substantial DM anisotropy as shown by G.J. Nilsen (Edinburgh, Lausanne) in his talk based on his neutron scattering PhD work. In this context of a behaviour at odds with Herbertsmithite, the theoretical discussion of the Hamiltonian through exact diagonalizations by P. Sindzingre (Paris) and ab-initio calculations by O. Janson (Dresden) point at a set of interactions, the complexity of which is related to the distorted character of the structure.

The progress in **Novel and artificial materials** has attracted a lot of attention. R. Colman (London) presented his successful synthetic work on a polymorph of Herbertsmithite, kapellasite and Mg derived  $\text{Cu}^{2+}$  kagome lattices. B. Fak (Grenoble) demonstrated that kapellasite has a specific spin liquid inelastic response in neutron scattering. J. Cumings was given the opportunity to review in his invited talk the recent developments on the growth of artificial spin ice lattices. Spin ice represent a solid playground to test the relevance of such a synthetic approach to the field of Highly Frustrated Magnetism. Finally, L. Fenner presented her results on the effects of frustration on the itinerant ferromagnet  $\text{Fe}_3\text{Sn}_2$ .

One session was devoted to **kagome lattices** decorated with **classical** spins. In the emerging field of easy-axis anisotropic kagome lattices, J. Gardner (NIST) and V. Simonet (Grenoble) reviewed their results on rare-earth langasites which point at the major influence of single-ion anisotropy on the low-T behaviour. When one refers to the richness of rare-earth pyrochlores, this is a vast field which seems to open. A. Sen (Mumbai, Boston) explained theoretically why an original semi-classical spin liquid phase should appear. Recent results on Co-based kagome lattices were then presented. O. Petrenko (Warwick) attracted our attention on the poor description of the magnetic phases under an applied field in the staircase compound  $\text{Co}_3\text{V}_2\text{O}_8$ . P. Manuel (ISIS) and H. Mutka (Grenoble) presented theoretical and experimental neutron work on the mixed valence cobaltate  $\text{YBa}(\text{Ca})\text{Co}_4\text{O}_7$ , a stacking of Co triangular and kagome layers. In the Heisenberg case, I. Rousochatzakis (Lausanne) showed through ED and symmetry arguments that the excitations can be associated with the spatial degeneracy of the semi-classical three sublattice coplanar ground states.

Finally, some recent theoretical developments on hard dimer models on **kagome stripes** were presented, J. Richter (Magdeburg) within a Hubbard model and A. Läuchli (Dresden) by looking at dimer correlations through exact diagonalization studies.

# Assessment of the results and impact of the event on the future direction of the field

This workshop met its primary objective: gather the three “kagome” communities to draw the state of the art in this field and debate about theories, experimental results and materials synthesis in order to generate new ideas and avenues for future research in the field which could be coined: “Towards a spin liquid state, but of what kind?”. The research in the field is indeed at the crossroads of new fundamental states and new generations of materials. Getting a deep overview in this focused field and a better knowledge between the actors in the field appears as a very positive step in this context.

Postdocs and PhD students represented 40% of the audience. They presented a set of very high level results. This reflects the great renewal of the field not only from the topical aspects which was born 20 years ago but also on the generation side!

The focused character of the meeting enabled the participants to have fruitful and unprecedented exchanges. Some collaborations between members of the ESF network were initiated, e.g. the visit program (Keren-Lemmens) for exploratory Raman measurements on novel systems, materials scientists could discuss about better in depth characterization through advanced physical techniques and some experimental results, e.g. for kapellasite (Wills-Mendels-Fak) attracted the attention of theorists who suggest it to be a model system with a complex non-planar and short-range spin correlations (Lhuillier). Overall, having theorists explaining in depth quite involved concepts to experimentalists proves to be quite rewarding for the future directions of the field and vice-versa. A good example is R.R.P Singh, inspired by A. Olariu’s talk produced a paper about spinless defects in the VBC scenario soon after the workshop.

One major concern is about materials synthesis. Only two or three groups at the European scale are working to produce new kagome systems, new synthesis routes or reproducing recently established synthesis routes. In comparison with the importance of the stake of generating a spin liquid on a perfect kagome lattice, this seems too little effort in comparison with the number of physicists engaged in the field! This is not specific to the Highly Frustrated Magnetism domain but rather signals a general trend of chemists going to more industrial related topics which are best funded.

The success of this workshop should encourage in the future to set-up a new network covering most of the field of Highly Frustrated Magnetism which expansion and structuring can certainly be granted, within Europe to our ESF-HFM network.

# Budget

The details about expenses can be obtained from Laboratoire de Physique des Solides and Institut Néel Grenoble which have been in charge the account of this workshop.

The expenses are sorted along global lines:

• Meals, coffee breaks	6483.70 €
• Accomodation for all participants	6641.78 €
• Travel for overseas invited speakers	4949.79 €
• Banquet + RER transport	2423.70 €
• Local costs	914.00 €
<b>TOTAL</b>	<b>21412.97 €</b>

Total support:

• RTRA Triangle de la Physique	5000.00 €
• Société Française de Physique	1672.24 €
• GDR « Magnétisme Frustré », CNRS	3546.00 €
• ESF	11200.00 €
<b>TOTAL</b>	<b>21418.24 €</b>



# Meeting Programme

The workshop had invited oral contributions (30+10 min.), contributions selected from the abstract list (20 + 5 min.) and posters.

Many slots were open for discussions between the various people active in this field.

## Monday, January 18th

**8:30 - 9:00 Welcome**

**9:00 - 10:30**

9:00 - 9:40 S. Helton  
9:40 - 10:05 C. Lhuillier  
10:05 - 10:30 A. Olariu

**10:30 Coffee break**

**11:00 - 12:30**

11:00 - 11:40 M. Hermele  
11:40 - 12:05 M.A. de Vries  
12:05 - 12:30 P. Lemmens

**12:30 - 14:15 Lunch**

**14:15 - 16:00**

14:15 - 14:55 R.R.P. Singh  
14:55 - 15:20 A. Keren  
15:20 - 16:00 F. Mila

**16:00 Coffee break**

**16:30 - 18:00**

16:30 - 16:55 R. Colman  
16:55 - 17:20 B. Fak  
17:20 - 18:00 O. Cépas

**18:00 - 20:00 Poster  
Session and Dinner buffet**

## Tuesday, January 19th

**9:00 - 10:30**

9:00 - 9:40 J. Cumings  
9:40 - 10:20 J.S. Gardner

**10:20 Coffee break**

**10:50 - 12:20**

10:50 - 11:15 A. Sen  
11:15 - 11:55 V. Simonet  
11:55 - 12:20 A. Zorko

**12:20 - 14:15 Lunch**

**14:15 - 15:45**

14:15 - 14:40 O. A. Petrenko  
14:40 - 15:20 P. Manuel  
15:20 - 15:45 H. Mutka

**15:45 Coffee break**

**16:15 - 18:10**

16:15 - 16:55 O. Tchernyshyov  
16:55 - 17:20 I. Rousochatzakis  
17:20 - 17:45 J. Richter  
17:45 - 18:10 L. Fenner

**20:00 Workshop dinner**

## Wednesday, January 20th

**9:00 - 10:30**

9:00 - 9:40 Z. Hiroi  
9:40 - 10:05 O. Janson  
10:05 - 10:30 G. J. Nilsen

**10:30 Coffee break**

**11:00 - 12:55**

11:00 - 11:25 P. Sindzingre  
11:25 - 11:50 D. Poilblanc  
11:50 - 12:05 M. Mambrini  
12:05 - 12:55 A. Läuchli

**Monday, January 18th**

**8:30 – 9:00 Welcome**

**9:00 – 10:30**

9:00 - 9:40 **Spin dynamics and scaling behavior in the spin -  $\frac{1}{2}$  Kagomé Lattice antiferromagnet herbertsmithite.**

*S. Helton, K. Matan, M. P. Shores, B. M. Bartlet, E. A. Nytko, Y. Qiu, D. G. Nocera, Y. S. Lee*

9:40 - 10:05 **The spin -  $\frac{1}{2}$  Kagomé antiferromagnet: static and dynamical correlations, spin gap or criticality ?**

*C. Lhuillier, L. Messio, P. Sindzingre, A. M. Läuchli, O. Cépas*

10:05 - 10:30 **Local properties of the quantum Kagomé antiferromagnet Herbertsmithite.**

*A. Olariu, A. Zorko, F. Bert, P. Mendels, M. A. de Vries, J. C. Trombe*

**10:30 Coffee break**

**11:00 – 12:30**

11:00 - 11:40 **Algebraic spin liquid on the Kagomé lattice.**

*M. Hermele*

11:40 - 12:05 **Static and dynamic correlations in the Kagomé antiferromagnet herbertsmithite – direct evidence of a quantum spin liquid ground state.**

*M. A. de Vries*

12:05 - 12:30 **Interplay of thermal and quantum fluctuations in the Kagomé lattice compound  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$**

*P. Lemmens, D. Wulferding, P. Scheib, V. Gnezdilov, P. Mendels, R. Colman, M. A. de Vries*

**12:30 – 14:15 Lunch**

**14:15 – 16:00**

14:15 - 14:55 **Possible valence bound crystal order in the Kagomé lattice antiferromagnet.**

*R. R. P. Singh*

14:55 - 15:20 **The symmetry of the spin Hamiltonian for spin -  $\frac{1}{2}$  Kagomé lattices and its implications.**

*A. Keren*

15:20 - 16:00 **Non magnetic impurities in Kagomé antiferromagnets.**

*F. Mila*

**16:00 Coffee break**

**16:30 - 18:00**

16:30 - 16:55  **$S = \frac{1}{2}$  Kagomé systems: synthetic challenges and recent advances.**

*R. Colman, B. Fak, A. S. Wills*

16:55-17:20 **Spin dynamics of classical and quantum Kagomé antiferromagnets studied by inelastic neutron scattering.**

*B. Fak, R. Colman, A. S. Wills, G. Nilsen, A. Harrison, J. Ollivier, D. Vissier, T. Unruh*

17:20 - 18:00 **ESR linewidth and lineshape in spin systems.**

*O. Cépas, S. El Shawish, S. Miyashita*

**18:00 - 20:00 Poster Session – Dinner buffet**

## Tuesday, January 19th

### 9:00 – 10:20

9:00 - 9:40 **Developments in Artificial Spin Ice and Artificial Kagome Ice.**  
J. Cumings

9:40 - 10:20 **Spin Correlations in Langasite Compounds – from spin liquid to spin solid.**  
J. S. Gardner

### 10:20 Coffee break

### 10:50 – 12:20

10:50 - 11:15 **Kagomé antiferromagnets with easy - axis anisotropy.**  
A. Sen

11:15 – 11:55 **Cooperative magnetism and magnetocrystalline anisotropy in rare - earth langasites.**  
V. Simonet, E. Lhotel, R. Ballou, J. Robert, K. Marty, P. Bordet, B. Canals, F. Hippert, P. Fouquet, J. Ollivier

11:55 – 12:20 **Local - Probe Investigation of Rare - Earth Based Langasites.**  
A. Zorko, F. Bert, P. Mendels

### 12:20 – 14:15 Lunch

### 14:15 – 15:45

14:15 – 14:40 **Kagomé staircase compound  $\text{Co}_3\text{V}_2\text{O}_8$  in an applied magnetic field: single crystal neutron diffraction study.**  
O. A. Petrenko, N. R. Wilson, G. Balakrishnan, D. McK Paul and G. J. McIntyre

14:40 - 15:20 **Spin correlations in geometrically frustrated  $\text{R}\text{BaCo}_4\text{O}_7$**   
P. Manuel, D. D. Khalyavin L. C. Chapon and J. F. Mitchell

15:20 - 15:45 **Spin fluctuations and freezing in  $\text{Y}_{0.5}\text{Ca}_{0.5}\text{BaCo}_4\text{O}_7$**   
H. Mutka, J. R. Stewart, G. Ehlers, C. Payen, P. Fouquet, R. Lortz

### 15:45 Coffee break

### 16:15 – 18:10

16:15 – 16:55 **Spin excitations with Fermi statistics in the  $S = 1=2$  Heisenberg model on Kagome.**  
Z. Hao, O. Tchernyshyov

16:55 - 17:20 **Symmetry analysis of low - energy spectra of highly frustrated AFMs with  $s > 1=2$**   
I. Rousochatzakis, A. Läuchli, F. Mila

17:20 – 17:45 **Hard - dimer degrees of freedom for the Hubbard model on Kagomé stripes.**  
J. Richter, O. Derzhko, A. Honecker, M. Maksymenko, R. Moessner

17:45 – 18:10 **Frustration in the Itinerant Ferromagnet  $\text{Fe}_3\text{Sn}_2$**   
L. Fenner, A. S. Wills

### 20:00 Workshop dinner : La Bastide Odéon, Paris

**Wednesday, January 20th**

**9:00 – 10:30**

9:00 – 9:40 **Novel Phenomena in the Spin -  $\frac{1}{2}$  Kagome Compound Volborthite.**

*Y. Okamoto, M. Tokunaga, M. Yoshida, M. Takigawa, Z. Hiroi*

9:40 – 10:05 **Volborthite  $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ : Orbital ordering on a distorted Kagomé geometry.**

*O. Janson, J. Richter, P. Sindzingre, H. Rosner*

10:05 - 10:30 **Neutron Scattering Studies of the  $\text{S} = \frac{1}{2}$  Quasi - Kagome System Volborthite.**

*G. J. Nilsen, F. C. Coomer, J. R. Stewart, P. P. Deen, M. A. DeVries, H. M. Rønnow, and A. Harrison*

**10:30 Coffee break**

**11:00 – 12:55**

11:00 – 11:25 **Which model spin Hamiltonian for Volborthite ?**

*P. Sindzingre*

11:25 – 11:50 **The Kagomé Heisenberg Antiferromagnet: at a First - Order Phase Transition close to a Quantum Critical Point ?**

*D. Poilblanc*

11:50 – 12:15 **A Generalized Quantum Dimer Model for the singlet sector of the Kagomé antiferromagnet.**

*M. Mambrini*

12:15 – 12:55 **Dimerization patterns in Kagome based quantum magnets.**

*A. Läuchli*

# Abstracts

## Spin Dynamics and Scaling Behavior in the Spin-1/2 Kagomé Lattice Antiferromagnet Herbertsmithite

**J. S. Helton**<sup>a,b</sup>, K. Matan<sup>a</sup>, M. P. Shores<sup>c</sup>, B. M. Bartlett<sup>c</sup>, E. A. Nytko<sup>c</sup>, Y. Qiu<sup>b,d</sup>, D. G. Nocera<sup>c</sup>, Y. S. Lee<sup>a</sup>

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<sup>b</sup>*NIST Center for Neutron Research, Gaithersburg, MD 20899*

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The mineral herbertsmithite,  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ , is among the best realizations yet achieved of the spin-1/2 kagomé lattice antiferromagnet. This material remains disordered until temperatures of at least 50 mK, with no evidence of a spin gap. Neutron scattering and magnetization measurements offer insight into the low temperature physics of this material. The low energy dynamic susceptibility displays an anomalous scaling relation over a wide range of temperatures, such that  $\chi T^{2/3}$  can be expressed as a function of  $\omega/T$ . This scaling behavior is similar to that observed in a variety of strongly correlated systems that are either near a quantum critical point or feature random interactions.

Monday 18th January 2010

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### The spin-1/2 kagome antiferromagnet: static and dynamical correlations, spin gap or criticality?

Claire Lhuillier<sup>1</sup>, Laura Messio<sup>1</sup>, Philippe Sindzingre<sup>1</sup>, Andreas M. Läuchli<sup>2</sup>, Olivier Cépas<sup>3</sup>

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Twenty years after the first discussions about the putative ground-state of the spin-1/2 kagome model [1], and the first "large scale" computations [2], the understanding of the model has progressed but many questions remain opened. We present various recent theoretical results on the spin-1/2 kagome antiferromagnet, which question some aspects of the common doxa on the subject: spin gap, nature of the short range correlations.

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[1] V. Elser, Phys. Rev. Lett. **62** 2405 (1989)

[2] P.W. Leung and V. Elser, Phys. Rev. B. **47**, 5459 (1993)

[3] P. Sindzingre and C. Lhuillier, Eur. Phys. Lett. **88**, 27009 (2009), cond-mat/0907.4164.

[4] A. Laeuchli, C. Lhuillier, arXiv:cond-mat/0901.1065.

[5] L. Messio, O. Cepas, C. Lhuillier submitted.

# Local Properties of the Quantum Kagome Antiferromagnet Herbertsmithite $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

**A. Olariu**<sup>a</sup>, A. Zorko<sup>b</sup>, E. Kermarrec<sup>c</sup>, F. Bert<sup>c</sup>, P. Mendels<sup>c</sup>, M.A. de Vries<sup>d</sup>, J.-C. Trombe<sup>e</sup>

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<sup>b</sup>*Jožef Stefan Institute, Jamova 39, 1000 Ljubljana, Slovenia*

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The frustration of antiferromagnetic interactions on the loosely connected kagome lattice associated to the enhancement of quantum fluctuations for  $S=1/2$  spins was acknowledged long ago as a keypoint to stabilize novel ground states of magnetic matter. Only very recently, the model compound Herbertsmithite,  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ , a structurally perfect kagome antiferromagnet could be synthesized [1] and allows close comparison to theories. Muon spin resonance investigation [2] has demonstrated the absence of any spin freezing at least down to 50 mK, an energy scale 4000 times smaller than the main antiferromagnetic interaction ( $J \simeq 190$  K). The kagome plane susceptibility could be accurately measured from the  $^{17}\text{O}$  NMR lineshift. It goes through a broad maximum at  $T \simeq J/2$  at variance with the monotonic SQUID susceptibility which can be consistently interpreted as being dominated by a paramagnetic-like defect contribution arising from Zn/Cu intersite mixing [3,4]. At lower temperatures ( $T \simeq J/100$ ), the kagome susceptibility is found to saturate to a non zero value in agreement with the absence of a gap in relaxation measurements[3].

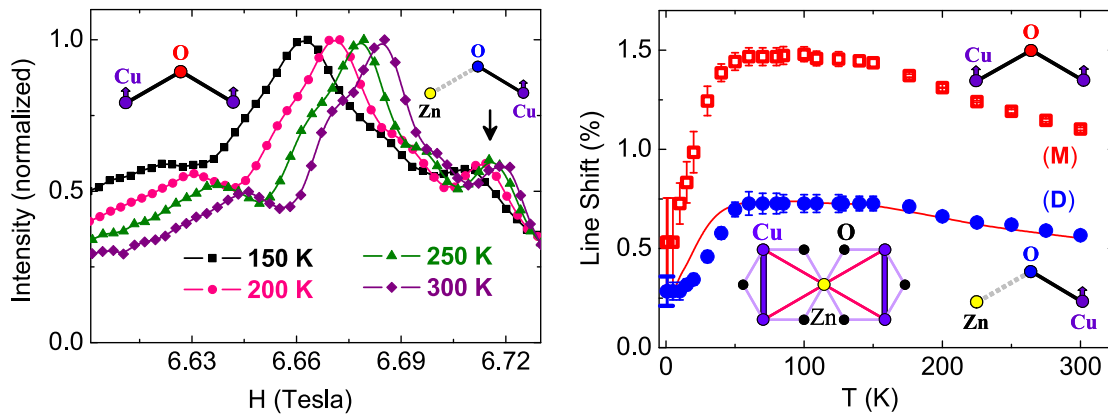


Figure 1  $\text{O}^{17}$  NMR central lines (left) and lineshifts (right) for the two detected O site, in between two Cu (open) and next to a Zn defect in the kagome plane (full).

## References

- [1] P. Shores *et al*, J. Am. Chem. Soc. 127, 13 462 (2005)
- [2] P. Mendels *et al*, Phys. Rev. Lett. 98, 077204 (2007)
- [3] A. Olariu *et al*, Phys. Rev. Lett. , 1 (2008).
- [4] F. Bert *et al* Phys. Rev. B (2007).

# Algebraic Spin Liquid on the Kagome Lattice

**Michael Hermele**

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The nature of the ground state of the  $S = 1/2$  kagome Heisenberg antiferromagnet is one of the most challenging and intriguing open problems of quantum magnetism. Experiments on herbertsmithite showing the absence of both magnetic order and a spin gap led Ran, Lee, Wen and myself to propose that the ground state may be an algebraic spin liquid [1, 2]. This state has a host of remarkable properties – perhaps most striking is the presence of a Dirac-fermion-like spectrum combined with a lack of good quasiparticle excitations, even at the lowest energies. In this talk, I will review our work, and give an update on the status of these ideas in light of recent experiments and numerical results.

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ESF-HFM Kagome-Workshop, Paris, Jan 2010

## Interplay of thermal and quantum fluctuations in the kagomé lattice compound $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

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We present a Raman spectroscopic investigation on the Herbertsmithite  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$  and Kapellasite, which are realizations of the spin  $1/2$  Heisenberg antiferromagnet on a kagomé lattice. In our detailed study we have compared crystals from mineral repositories with hydrothermally prepared powder materials (different antisite disorder) and single crystals. As function of temperature spin correlations evolve in Raman scattering as a quasielastic line of thermally induced fluctuations and a broad, feature less continuum at finite energies. These two components have markedly different temperature dependence and symmetry selection rules. Therefore we attribute them to different physics. Phonon excitations are used to evaluate the significance of symmetry selection rules in the light scattering experiments. Antisite disorder shows up in the linewidth of certain phonon modes. Our results will be compared with recent calculations by O. Cepas, et al, W.H. Ko, et al., and A.M. Läuchli, et al.

Monday 18th January 2010

# Static and dynamic correlations in the kagome antiferromagnet herbertsmithite - direct evidence of a quantum spin liquid ground state.

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Neutron-spectroscopy data on the  $s = 1/2$  kagome antiferromagnet herbertsmithite ( $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ , Fig. 1) reveal a continuum of short-ranged antiferromagnetic dynamic correlations at all measured temperatures[1]. The weak temperature and energy dependence of the inelastic magnetic neutron scattering suggest that dynamic correlations up to THz frequencies persist even as  $T \rightarrow 0$ . In this light the previously reported[2] and some new  $\mu\text{SR}$  results on an extended range of zinc paratacamite compounds ( $\text{Zn}_x\text{Cu}_{4-x}(\text{OH})_6\text{Cl}_2$  with  $0 \leq x \leq 1.2$ ) are discussed, making a case for a highly degenerate groundstate with fast zero-temperature dynamics caused by quantum fluctuations. These fluctuations are suppressed when neighboring kagome layers become coupled as  $x$  is reduced, leading to glassy behavior for  $x \sim 0.5$ . A comparison is made with other materials showing strong dynamic correlations, providing an intuitive picture of the quantum spin liquid state observed in herbertsmithite.

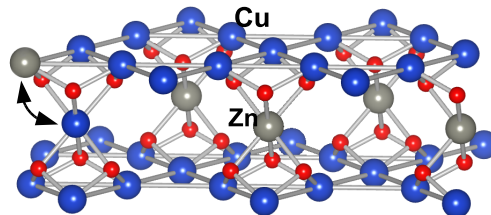


Figure 1. Two kagome layers of O (red) -linked  $\text{Cu}^{2+}$  ions (blue) in zinc paratacamite with  $x = 1$ , also known as herbertsmithite. Replacement of the  $\text{Zn}^{2+}$  ions (shown in grey) with  $\text{Cu}^{2+}$  creates weak exchange pathways between the kagome layers.

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# Possible Valence Bond Crystal Order in the Kagome Lattice Antiferromagnet

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We have used<sup>2,3</sup> series expansion methods to explore Valence Bond Crystal (VBC) order in the ground state of the Kagome Lattice Heisenberg Antiferromagnet. We found that the Honeycomb Valence Bond pattern shown in figure below with dimerized hexagons (H), empty triangles (E) and pinwheels (P), with a minimum 36-site unit cell, has the lowest energy of all Valence Bond configurations. Excitations of this state were studied in the thermodynamic limit and on a 36-site cluster with periodic boundary conditions. We find that this state has weakly dispersive triplet excitations and a large number of singlet excitations, several of which fall below the lowest triplet. The case for this VBC state to be the ground state of the Kagome Lattice Heisenberg Model will be discussed.

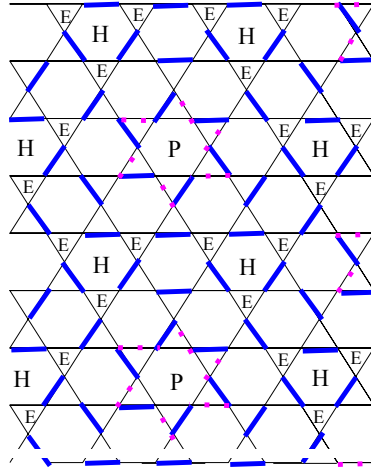


Figure 1. Proposed Valence Bond Crystal order on the kagome lattice.

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# The symmetry of the spin Hamiltonian for spin- $\frac{1}{2}$ kagomé lattices and its implications

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We present magnetization measurements on oriented powder and single crystal of  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ , and single crystals of a new organometallic kagomé lattice  $\text{Cu}(1,3\text{-bdc})$  along and perpendicular to the  $\hat{c}$  axis. Both systems have perfect spin-1/2 kagomé structure but in  $\text{Cu}(1,3\text{-bdc})$  there is no risk of Zn substitution on the kagomé plane. We find a dramatic difference in the magnetization between the two directions at high temperatures in both systems. We show that this difference must emerge from Ising-like exchange anisotropy. The relevance of these findings to the expected ground state and other experiments will be discussed. Some of the results are published in Ref. [1].

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# Non-magnetic impurities in kagome antiferromagnets

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In this talk I will discuss the effect of non-magnetic impurities in  $S=1/2$  and  $S=3/2$  kagome antiferromagnets. For  $S=1/2$ , a non-magnetic impurity does not lead to the formation of a local moment but freezes neighboring bonds into almost perfect singlets [1], in agreement with the singlet-dimer description [2] of the low-energy singlet sector [3]. This freezing resists the presence of Dzyaloshinskii-Moriya interactions far beyond the value where this type of interaction is believed to lead to the  $q=0$  ordered state [4]. A comparison with the  $S=3/2$  case will also be included [5], as well as a discussion of the implications for local probes such as NMR experiments.

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# $S = \frac{1}{2}$ Kagome Systems: synthetic challenges and recent advances

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The introduction of Herbertsmithite to the field of  $S = \frac{1}{2}$  kagome antiferromagnets in 2005 [1] catalysed a burst of theoretical and experimental research into the area. One important realisation from this is the possibility of anti-symmetric exchange, the DM interaction. The DM interaction mixes a ferromagnetic component into the exchange Hamiltonian that, if large enough, will destabilise a dynamic singlet groundstate and promote classical order [2]. These exact diagonalisation studies performed by C epas *et.al.* have shown that for  $|D| < 0.1J$  quantum fluctuations will be strong enough to destabilise a N eel state, explaining the observation of a dynamical groundstate in Herbertsmithite with an experimentally determined  $D$  component of  $0.08J$  [3]. Given these observations and predictions, the need for a library of structures and materials to probe the properties of this  $D/J$  phase diagram is clear.

In this talk we will present three new  $S = \frac{1}{2}$  kagome materials: Kapellasite,  $\alpha\text{-Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$  [4]; Haydeeite,  $\alpha\text{-Cu}_3\text{Mg}(\text{OH})_6\text{Cl}_2$ ; and Mg-Herbertsmithite,  $\gamma\text{-Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$ . Magnetisation measurements show Kapellasite to have no magnetic order down to the lowest temperatures whilst Haydeeite and Mg-Herbertsmithite have magnetic transitions at  $T = 4.2\text{K}$  and  $5.8\text{K}$ , respectively. Low temperature inelastic neutron scattering spectra show Kapellasite to have a liquid-like magnetic excitation spectrum. The ability to probe a range of materials will be key to separating the intrinsic signature of a spin-liquid groundstate from the inherent properties of the individual materials.

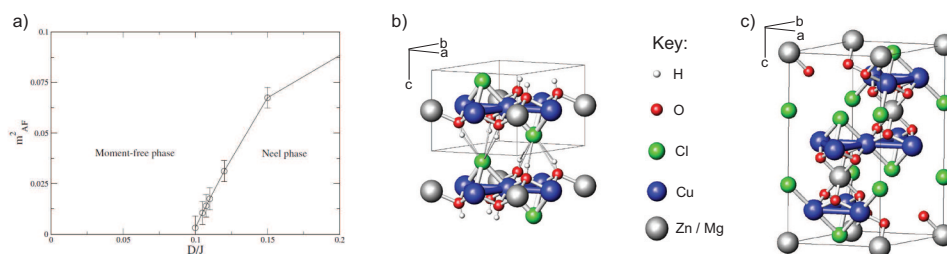


Figure 1a) The  $D/J$  phase diagram calculated by C epas *et.al.* [2], showing a critical boundary between ordered and dynamic singlet groundstates. Herbertsmithite lies to the left of this boundary with  $D = 0.08J$ . b) The structure of Kapellasite (Zn) and Haydeeite (Mg) showing weakly bonded O–H–Cl  $\text{Cu}^{2+}$  and highly 2-dimensional kagome planes. c) Mg-Herbertsmithite, a pyrochlore-like lattice of metal sites, doped with  $\text{Mg}^{2+}$  ions to form  $\text{Cu}^{2+}$  kagome planes.

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# Spin dynamics of classical and quantum kagomé antiferromagnets studied by inelastic neutron scattering

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We have used inelastic neutron scattering to probe the dynamic magnetic susceptibility of the spin-liquid state of two kagomé antiferromagnets, one with classical spin ( $S=5/2$ ) and one with quantum spin ( $S=1/2$ ). Both systems are almost ideal Heisenberg antiferromagnets with weak (or absent) Dzyaloshinsky-Moriya (DM) interaction, as indicated by the absence of long-range magnetic order down to the lowest temperatures. The magnetic ions,  $\text{Fe}^{3+}$  and  $\text{Cu}^{2+}$ , respectively, form perfect undistorted corner-sharing triangles with essentially full occupation ( $>98\%$ ) of the resulting kagomé lattice.

Both systems have strong inelastic magnetic scattering whose inelastic structure factor  $S(Q)$  shows correlations characteristic of a spin liquid. In the  $S=5/2$  system, deuterium jarosite with chemical formula  $(\text{D}_3\text{O})\text{Fe}_3(\text{SO}_4)_2(\text{OD})_6$  [1], the measured  $S(Q)$  is well reproduced by Monte-Carlo simulations where the short-range correlations have staggered vector chirality. In the  $S=1/2$  system,  $\text{Cu}_3\text{Zn}(\text{OD})_6\text{Cl}_2$  alias Kapellasite [2], the measured  $S(Q)$  is very different.

The dynamic magnetic susceptibility is in both cases gapless with typical relaxation behavior, but the temperature dependence is different. The measurements on the  $S=5/2$  system have very recently been extended to lower energies and lower temperatures, down to 0.45 K, using cold neutrons. The temperature dependence of the observed low-energy spin-liquid response will be compared to the dynamic scaling scenario, in which the characteristic energy scale of the spin fluctuations should vanish in the limit  $T \rightarrow 0$ , as well as to recent simulations, where rather well-defined excitations are predicted to emerge at sufficiently low temperatures.

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# ESR Linewidth and lineshape in spin systems

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In the kagome antiferromagnet  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_3$ , recent ESR measurements have suggested that the system has sizeable Dzyaloshinskii-Moriya interactions [1] which possibly places the compound close to a quantum critical point [2]. However relating the ESR linewidth to the anisotropy strength is a long standing issue. Experimentalists have been using the Kubo-Tomita theory [3] for decades. We show by using exact diagonalizations of small 1d spin systems that the Kubo-Tomita theory should underestimate the linewidth by at least a factor 2 in thermodynamic systems. In addition, in finite-size systems, we find strong departures from the Lorentzian lineshape, a prediction which could be observed in molecular magnets, would the mechanism for the relaxation be purely magnetic in origin.[4]

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# Developments in Artificial Spin Ice and Artificial Kagome Ice

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Recently, a model system has been demonstrated, dubbed artificial spin ice, that allows the frustrated magnetic material spin ice to be modeled using rationally-designed and artificially-fabricated nanomagnetic NiFe lattices[1]. This new approach allows spin interactions to be rationally designed and the resulting configurations to be imaged directly in real space. Unfortunately, its initial embodiment on the square lattice did not strictly obey the ice rule for the spins, instead showing only a statistical tendency toward the ice rule. More recently, we have shown that an analogous system can be fabricated on the kagome lattice and the resulting kagome ice presents more promise for studying frustration in nanomagnetic lattices[2]. The artificial kagome ice show rigorous adherence to the ice rule, and even shows a statistical influence of long-range dipolar forces, which may be expected to reduce the frustration, producing a long-range ordered ground state in spin ice.

Our studies of artificial ice have been carried out using Lorentz electron microscopy. This technique allows real-time video rate imaging. Using this, we can capture the magnetic reversal dynamics of this system, including the motion of magnetic monopoles through the lattice. I will present results that show this reversal process in detail and that may lead to a comprehensive understanding of this intriguing new frustrated system.

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## Kagome antiferromagnets with easy-axis anisotropy

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We consider the effect of strong easy-axis anisotropy  $D$  on  $S > 3/2$  spins interacting with antiferromagnetic exchange  $J$  on the Kagome lattice, which make the spins Ising-like. However, the low temperature limit is quite different from the cooperative Ising paramagnet that obtains classically for  $T \ll JS^2$ . We find that  $O(J^3S/D^2)$  multi-spin interactions arising from quantum fluctuations result in a crossover from an intermediate temperature classical cooperative Ising paramagnet to a semiclassical spin liquid with distinct short-ranged correlations for  $T \ll J^3S/D^2$  [1]. We also study the problem in the presence of a magnetic field along the easy-axis and show that leading quantum fluctuations lead to an interesting collinear state that breaks sublattice rotation symmetry, while leaving the translational symmetry of the lattice intact, on the one-third magnetization plateau [2].

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# Spin Correlations in Langasite Compounds – from spin liquid to spin solid

Jason S Gardner (Indiana University and the NIST Centre for Neutron Research)

In the 1980's a concerted effort was made to grow large single crystals of several langasite compounds for industrial applications including laser and piezoelectric engineering. This resulted in huge, high quality, crystals for scientific research but langasites have only recently been studied by those interested in frustration.

Langasite compounds contain planes of magnetic rare-earth (RE) ions on a lattice topologically equivalent to a kagomé net. Their general formula is  $RE_3Ga_5SiO_{14}$  allows chemists to vary the structural properties considerably without destroying the magnetic sublattice of corner sharing triangles. Neutron scattering, NMR, low temperature specific heat and magnetization, and photoluminescence have been used to investigate this inherently two-dimensional structure.

In my talk I will concentrate on recent results from Pr and Nd Langasite. These can have liquid like characteristics to the lowest temperatures but by varying the composition, one is able to keep the kagomé net and tuning the magnetic interactions allowing one to freeze the spin dynamics.

This work is done in collaboration with the quantum materials group at Florida State (C. R. Wiebe et al.) and L. Balicas et al. at the National High Magnetic Field Laboratory.

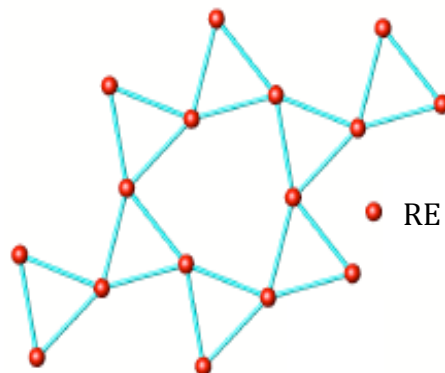


Figure 1: The ab-plane of a RE langasite showing the network of corner sharing magnetic ions.



# Cooperative magnetism and magnetocrystalline anisotropy in rare-earth langasites

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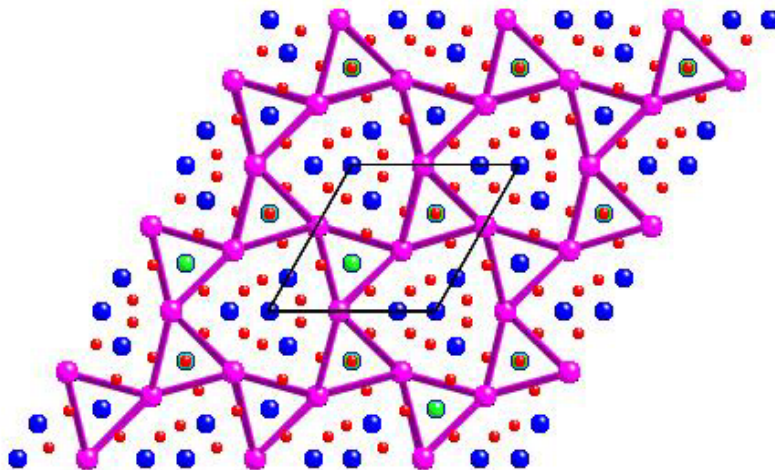
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The  $\text{Nd}_3\text{Ga}_5\text{SiO}_{14}$  and  $\text{Pr}_3\text{Ga}_5\text{SiO}_{14}$  langasites are the first realization of a kagomé lattice of highly anisotropic magnetic moment. The compounds crystallize in the  $P321$  space group. The rare-earth magnetic cations are at the vertices of a lattice, which is topologically equivalent to a kagomé lattice if only nearest-neighbours exchange interactions are relevant (see figure) [1]. They experience a strong magnetocrystalline anisotropy, either uniaxial or multiaxial, the interplay of which with the geometrical frustration is expected to produce unconventional magnetic properties. We have investigated the static and dynamic magnetic properties of these compounds by magnetization, specific heat, and neutron scattering, which revealed puzzling spin dynamics [2]. As to get more insights, magnetization and dc/ac susceptibility measurements down to very low temperature were recently also performed. These were extended to compounds where the non magnetic  $\text{La}^{3+}$  is substituted for the magnetic cations  $\text{Nd}^{3+}$  and  $\text{Pr}^{3+}$ , which helped furthering the understanding of the respective effects of exchange interactions and single-ion magnetism. All the experiments were quantitatively analyzed, by fully taking account of the crystal electric field effects on the  $\text{Nd}^{3+}$  or  $\text{Pr}^{3+}$  ions. The results, and in particular the possible onset of cooperative magnetic behavior emerging only at very low temperature, will be presented.



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# Local-Probe Investigation of Rare-Earth Based Langasites

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The discovery of the Langasite family  $\text{RE}_3\text{Ga}_5\text{SiO}_{14}$  (RE = rare earth) opened a new field of experimental research on the kagomé lattice [1]. Here magnetism is due to rare earths rather than transition metals. Consequently, the classical limit of large spins may often be approached and, as a rule, magnetocrystalline anisotropy is expected to have a strong impact on the magnetic ground state.

Various different compositions with the same crystal structure could be synthesized [1,2]. In our local-probe NMR, NQR and  $\mu^+\text{SR}$  studies we have focussed on two representatives,  $\text{Nd}_3\text{Ga}_5\text{SiO}_{14}$  and  $\text{Pr}_3\text{Ga}_5\text{SiO}_{14}$ , the first being a Kramers' ( $J = 9/2$ ) ion and the second a non-Kramers' ( $J = 4$ ) ion. In the former system a fluctuating ground state, which remains to be fully understood [3], is present down to at least 40 mK [4,5]. The latter similarly does not show any signs of magnetic instabilities. However, it lies on the verge of spin freezing, which can be induced by applying chemical pressure [2]. We shall compare the ground state properties of both materials and discuss the influence of external magnetic field on spin dynamics.

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# Kagomé staircase compound $\text{Co}_3\text{V}_2\text{O}_8$ in an applied magnetic field: single crystal neutron diffraction study

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In the  $\text{M}_3\text{V}_2\text{O}_8$  family ( $\text{M} = \text{Co}, \text{Ni}, \text{Cu}, \text{Mn}$ ), the magnetically coupled ions form a network, which is similar in topology to the Kagomé lattice. The bonding between the  $\text{M}^{2+}$  ions creates layers buckled into a staircase formation, the Kagomé staircase. Although the magnetic properties of different family members vary significantly, a unifying feature of them all is the extreme richness of their magnetic phase diagrams, attributed to a close proximity in energy of several competing states.

In  $\text{Co}_3\text{V}_2\text{O}_8$  (CVO) a magnetic ordering at 11 K is from a paramagnetic to an AFM incommensurate state, while the second transition at 6 K is to a FM state [1]. A number of lock-in transitions associated with incommensurate to commensurate AFM phases have also been observed at intermediate temperatures [1]. In an applied magnetic field, CVO demonstrates a highly anisotropic behaviour in terms of the values of susceptibility and the sequence of field-induced phase transitions [2]. Reports based on different experimental methods agree in general, but disagree in the detailed description of CVO's behaviour in an applied field, particularly in the interpretation of the magnetic phases.

We have performed single crystal neutron diffraction measurements for  $H \parallel a$  and  $H \parallel c$ . The measurements have revealed an additional, previously unreported transition for  $H \parallel a$  in the AFM phase (see Fig. 1). They have also allowed us to unequivocally identify and label all the observed phases for both field directions [3].

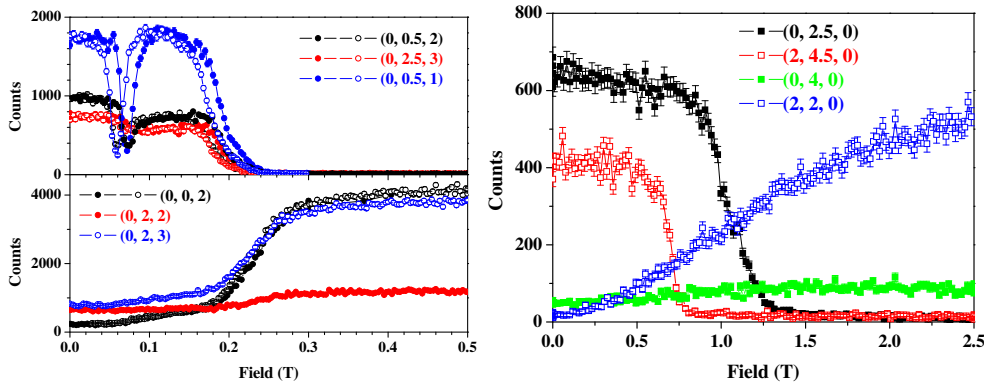


Figure 1. Field dependence of the integrated intensity of several AFM and FM peaks in  $\text{Co}_3\text{V}_2\text{O}_8$  at  $T = 7.5$  K. Solid/open symbols mark increasing/decreasing magnetic field ramps. Left panel:  $H \parallel a$ , AFM (top) and FM (bottom) peaks. Right panel:  $H \parallel c$ .

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# Spin correlations in geometrically frustrated $\text{RBaCo}_4\text{O}_7$

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For several decades, frustrated magnetic systems where a degenerate manifold of ground states is expected owing to the impossibility of satisfying simultaneously all energy terms in the Hamiltonian, have generated huge interest both theoretically and experimentally. In particular, kagome networks have been at the forefront of such studies. Recently, a new class of materials,  $\text{RBaCo}_4\text{O}_7$  whose magnetic lattice is made of triangular and kagome layers of Co ions that are alternatively stacked along the  $c$ -axis of the trigonal crystal structure has been synthesised. Our recent analysis of single crystal neutron diffraction on  $\text{YBaCo}_4\text{O}_7$  through Monte Carlo simulations [1] has shown that the magnetic lattice is best described as a 3D network of corner-sharing triangles and bipyramids. Magnetic correlations are found to be short range, preserving a  $120^\circ$  configuration, in the  $ab$ -plane but long-range between apical Co ions along the  $c$ -axis. This points towards the degrees of freedom in the triangular bipyramid units dictating the configurational degeneracy : a rather different picture than that of pure kagome systems. In this workshop, we will review this work and extend it by mean-field calculations and extensive Monte Carlo modelling [2]. In particular, we will present magnetic phase diagrams as function of  $J_{\text{out}}/J_{\text{in}}$ , relevant for real systems where chemical substitution can tune the ratio between exchange integrals inside the kagome layers,  $J_{\text{in}}$ , and between kagome and triangular layers,  $J_{\text{out}}$ .

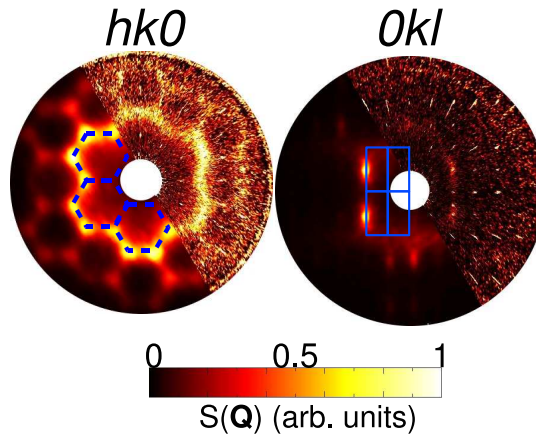


Figure 1. Magnetic diffuse scattering for  $\text{YBaCo}_4\text{O}_7$  in the  $hk0$  (left panel) and  $0kl$  (right panel) reciprocal planes. For each panel, experimental data (right hand side) and MC simulations (left hand side) are presented.

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# Spin fluctuations and freezing in $\text{Y}_{0.5}\text{Ca}_{0.5}\text{BaCo}_4\text{O}_7$

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The  $\text{Y}_{0.5}\text{Ca}_{0.5}\text{BaCo}_4\text{O}_7$  compound investigated by Valldor [1] was subsequently examined with polarized neutron diffraction and proposed as a candidate for a model classical kagomé system by Schweika et al. [2], based on their interpretation relying on low-spin ( $S=0$ ) state on the Co sites that are not in the kagomé planes. This motivated our comprehensive study on the spin dynamics of the compound. We have used SQUID magnetometry and specific heat for bulk characterization of our powder sample and neutron powder diffraction for structural characterization. The temperature dependent diffuse magnetic scattering cross-section measured with cold neutron diffraction with polarization analysis (D7 instrument at ILL) is consistent with the results published earlier. However, best agreement with the experimental data requires that all Co sites carry moments. This situation and the dynamic response  $S(Q, \omega, T)$  propose a revision of the basic ingredients for the physics of this compound. We have seen a broad, temperature dependent quasi-elastic response (IN4 spectrometer at ILL) reaching up to tens of meV at room temperature and persisting down to  $T = 1.5$  K. This feature was not seen in the earlier work due to the experimental conditions. In fact the full magnetic response is not within the kinematic range of cold neutron diffraction leading to a serious shortcoming for obtaining a quantitatively correct powder averaged static structure factor  $S(Q, T)$  in such experiments. In addition, in our sample we see a clear indication of spin freezing at  $T \approx 50$  K, observed with SQUID measurements and studied in detail using neutron spin echo spectroscopy and high-resolution backscattering elastic scans (IN11 and IN16 spectrometers at ILL). On cooling, associated with the freezing we have observed a gradual narrowing of the dynamic response (IN4 and IN5 spectrometers at ILL), leading to a complicated line-shape of the quasi-elastic response. An important conclusion of our work is that a large fraction of the magnetic moment is rapidly fluctuating even in the low- $T$  frozen state. We further conclude that comparison of the magnetic response of  $\text{Y}_{0.5}\text{Ca}_{0.5}\text{BaCo}_4\text{O}_7$  with the ideal models of classical kagomé ground-state [3,4] is not a valid approach.

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# Spin excitations with Fermi statistics in the $S = 1/2$ Heisenberg model on kagome

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Spin excitations in a magnet with ordered moments are magnons, quantized spin waves that behave like particles with integer spin and Bose-Einstein statistics. A magnet lacking long-range order cannot support spin waves and may have unusual magnetic excitations with noninteger spin and different statistics. Heisenberg antiferromagnets with spins of length  $S = 1/2$  on kagome and hyperkagome lattices have emerged as a leading candidate for novel quantum physics. We have recently shown [1] that magnetic excitations in this system are spinons, quasiparticles with spin  $1/2$  and Fermi-Dirac statistics. The ground state of the system is a quantum liquid or solid of small, heavy  $S = 0$  pairs of spinons with a large density of nonmagnetic ( $\Delta S = 0$ ) excitations at low energies. A magnetic ( $\Delta S = 1$ ) excitation amounts to breaking up a pair into two spinons, a process with a threshold energy of  $0.06$  exchange constants. The problem can be mapped onto a compact  $U(1)$  gauge theory on the honeycomb lattice with background “electric” charges, Fig. 1.

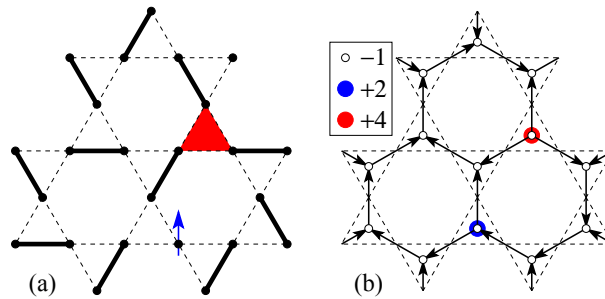


Figure 1. (a) Kagome lattice and the major players: quantum dimers (thick bonds), a defect triangle containing no dimer (red), and an antikink spinon (blue arrow). (b) The mapping to a compact  $U(1)$  gauge theory on a honeycomb lattice. Electric flux on links has strength  $\pm 1$  and is depicted as arrows in the manner of Elser and Zeng [2,3]. The inset shows the  $U(1)$  charges of the lattice sites and quasiparticles.

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# Symmetry analysis of low-energy spectra of highly frustrated AFMs with $s > 1/2$

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We explore the low-energy physics of the  $s > 1/2$  Heisenberg antiferromagnetic model on the kagomé and on related highly frustrated clusters by exploiting the characteristic symmetry pattern of exact diagonalization (ED) spectra. We find that the low-energy excitations embody the spatial and spin symmetry properties of the semiclassical three-sublattice coplanar ground states. The dense excitation features which are manifested in the whole magnetization range can thus be accounted for by the large spatial degeneracy of these semiclassical configurations: Each of these states gives rise to a distinct tower of states and they all appear at low energies albeit split by quantum fluctuations.

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# Hard-dimer degrees of freedom for the Hubbard model on kagomé stripes

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We consider the repulsive Hubbard model on two kagomé stripes of  $N$  sites (see Fig. 1). On both lattices the lowest single-electron band is completely dispersionless (flat). We construct the complete manifold of *exact many-electron* ground states at low electron numbers  $n$  up to  $n = \mathcal{N} + 1$ , where  $\mathcal{N} = N/3$  for the kagomé stripe I and  $\mathcal{N} = N/5$  for the kagomé stripe II. By mapping the electronic degrees of freedom on a classical hard-dimer model we can calculate the degeneracy of these states [1]. As a result, we obtain closed-form expressions for low-temperature thermodynamic quantities around a particular value of the chemical potential  $\mu_0$ . Moreover, we can calculate the zero-temperature magnetization for electron numbers  $0 < n \leq \mathcal{N} + 1$ . We discuss specific features of thermodynamic quantities of these ground-state ensembles such as residual entropy, an extra low-temperature peak in the specific heat, and the existence of ground-state ferromagnetism and paramagnetism. We confirm our analytical results by comparison with exact diagonalization data for finite systems.

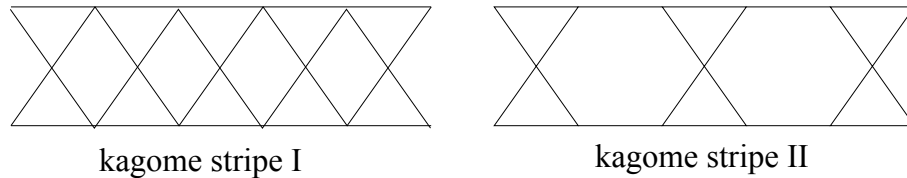


Figure 1. The two kagomé stripes.

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# Frustration in the Itinerant Ferromagnet $\text{Fe}_3\text{Sn}_2$

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$\text{Fe}_3\text{Sn}_2$  is an itinerant, metallic ferromagnet below 640 K, and has its magnetic Fe atoms arranged in an offset kagome bilayer structure [1]. We have prepared powder samples of  $\text{Fe}_3\text{Sn}_2$  by standard solid state synthesis techniques, and single crystals by chemical vapour transport. Evidence for magnetic frustration is provided by the onset of a re-entrant spin glass phase at around 80 K [2], which is confirmed by measurements of the thermoremanent magnetisation. We present refinements of the magnetic structure of  $\text{Fe}_3\text{Sn}_2$  using data from two different neutron diffraction instruments. The first data indicated that  $\text{Fe}_3\text{Sn}_2$  has a temperature dependent, frustrated, non-collinear magnetic structure; however, the more recent data suggests that the magnetic structure is actually almost collinear at all temperatures. The ferromagnetic transition is to a state with the moments lying approximately along the  $c$  axis, and as the temperature is decreased the moments gradually rotate towards the  $ab$  plane, to lie at an angle of  $54^\circ$  with the  $c$  axis at 2 K. SQUID magnetometry measurements of powder samples show that this change in magnetic structure with temperature is accompanied by a gradual increase in magnetic susceptibility below the ferromagnetic transition, down to around 50 K.

We also present recent anomalous Hall effect (AHE) measurements.  $\text{Fe}_3\text{Sn}_2$  has a large saturated Hall resistivity of  $3.2 \mu\Omega\text{cm}$  in a field of 1 T at room temperature [3], making it a notable candidate for spintronics applications based on the AHE. The anomalous Hall coefficient,  $R_S = 6.7 \times 10^{-9} \Omega\text{cmG}^{-1}$ , is three orders of magnitude larger than that of pure Fe, and cannot be understood in terms of the conventional extrinsic AHE mechanisms.

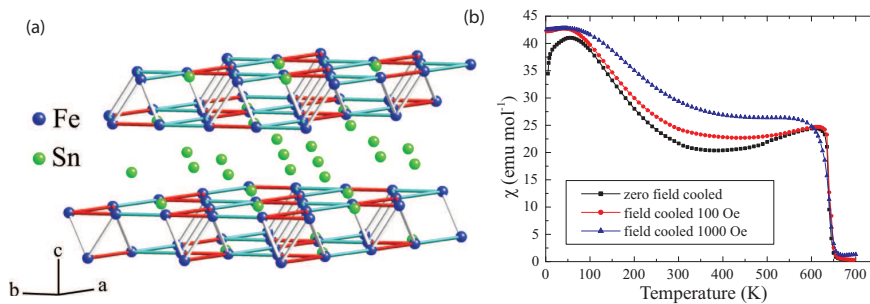


Figure 1. (a) The crystal structure of  $\text{Fe}_3\text{Sn}_2$ . The Fe ions form bilayers of offset kagome networks, and the Sn ions lie in the centre of the kagome hexagons and between the bilayers. (b) Magnetic susceptibility of powder  $\text{Fe}_3\text{Sn}_2$  in applied fields of 100 and 1000 Oe, between 5 and 700 K. The zero-field cooled curve was measured in 100 Oe. The susceptibility continues to increase below  $T_C = 640$  K, and a re-entrant spin glass transition occurs at around 80 K.

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# Novel Phenomena in the Spin-1/2 Kagome Compound Volborthite

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We report on the properties of two spin-1/2 kagome compounds, volborthite [1,2] and vesignieite [3], that possess a slightly distorted and nearly perfect kagome lattices made up of Cu<sup>2+</sup> ions, respectively.

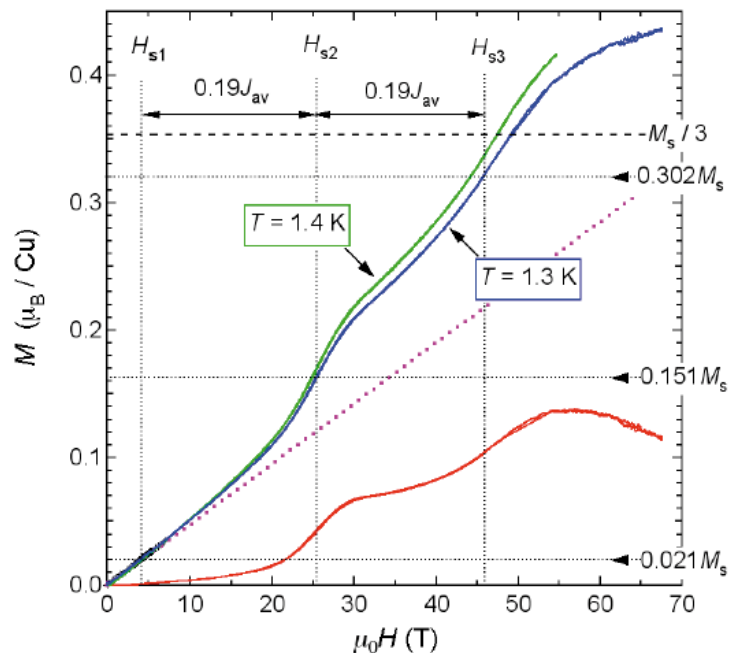
For volborthite, we found in low magnetic fields below 4 T spin-liquid like behavior with no spin gap but with a peculiar transition at 1 K, where V NMR spin-lattice relaxation rate  $1/T_1$  exhibits a sharp peak, and specific heat exhibits not a peak but a kink. There must be a certain phase transition of spins. However, it is not at all a simple long range antiferromagnetic order because of the following facts: both  $1/T_1$  and specific heat are proportional to  $T$ , and NMR spin-spin relaxation rate  $1/T_2$  is much larger than  $1/T_1$ . The former evidences a gapless excitation that may be different from spin-wave excitations, and the latter indicates a very slow spin dynamics surviving down to the lowest temperature. The origin of this 1 K transition is not known.

On the other hand, at higher magnetic fields, three magnetization steps are observed at 4.3, 25.5 and 46 T, followed by a plateau or a vicinal slope at a magnetization  $\sim 0.42M_s$  ( $M_s$  is a saturation magnetization) in higher magnetic fields above 60 T. The large deviation of the plateau magnetization from theoretically predicted  $1/3M_s$  is not due to the distortion of the kagome lattice in volborthite, because a very similar value has been found also for vesignieite having an almost perfect kagome lattice. It seems that the excess magnetization of  $\sim 0.1M_s$  from  $1/3M_s$  is successively gained at the three steps with increasing magnetic field. Such magnetization steps have not been observed in vesignieite, but it is possibly due to a larger amount of impurity spins contained in vesignieite samples thus far obtained, compared with the case of volborthite.

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Figure 1. Magnetization of volborthite. The lowest curve represents the data taken at 1.3 K after the subtraction of the initial linear component indicated by a broken line.



# Volborthite $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$ : Orbital ordering on a distorted kagomé geometry

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Spin-1/2 Heisenberg magnets with a kagomé geometry attract considerable interest after the spin liquid ground state has been proposed for herbertsmithite  $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$ . Unfortunately, the substitutional Cu/Zn disorder, spin anisotropy and a considerable inter-layer coupling in this system intricate further investigations, but inspire a search for new materials. One of them — the mineral volborthite  $\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}$  has been recently claimed<sup>a</sup> as a candidate for a real material realization of a spin-1/2 kagomé lattice. Despite the monoclinic distortion which allows for a non-frustrated magnetic coupling scenario, experiments reveal the presence of frustration and show no clear evidence for long-range order down to 50 mK. Nevertheless, further interpretation of experimental data reveals that the magnetic properties of volborthite cannot be consistently described by a pure kagomé model.

Here, we present a theoretical investigation of this interesting material. Basing on the results of DFT calculations, we estimate the relevant orbitals and couplings to approach the ground state puzzle. Quite unusual for cuprates, we find different magnetically active orbitals for structurally different Cu(I) and Cu(II) atoms: while the latter has a hole in a  $x^2 - y^2$  orbital (like in most cuprates), for Cu(I) the  $3z^2 - r^2$  orbital is half-filled (like in  $\text{CuSb}_2\text{O}_6$ <sup>b</sup>). This has a dramatic influence on exchange integrals, which have been estimated using the LSDA+ $U$  approach. The results of our quantitative microscopic analysis are mapped onto a Heisenberg model. Thermodynamical behaviour and spin correlations are estimated based on exact diagonalization of clusters up to  $N=36$  sites.

We show that the orbital order leads to a picture of coupled frustrated chains rather than a kagomé model. The developed model yields a considerable improvement of the understanding of experimental data.

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# Neutron Scattering Studies of the $S = 1/2$ Quasi-Kagome System Volborthite

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An experimental realisation of the  $S = 1/2$  kagome Heisenberg model remains one of the holy grails of solid state physics. While structurally distorted, volborthite  $[\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}]$  is one of the cleanest ( $\sim 99\%$  coverage) known candidate materials, and recent studies by NMR and  $\mu\text{SR}$  have revealed rich low temperature physics [1]. We have performed inelastic time of flight and polarised diffuse neutron scattering experiments on volborthite, which show an excitation spectrum resembling that of the classical kagome antiferromagnet, and short range order (SRO) at low temperatures ( $< 10$  K), respectively. The low temperature SRO appears to be coplanar and is describable by at least two propagation vectors. The origin of this behaviour is ascribed to anisotropies in the Hamiltonian, evidence for which furthermore exists in the data. These results stand in stark contrast with recent measurements on herbertsmithite [2], where the inelastic spectrum cannot be described semi-classically.

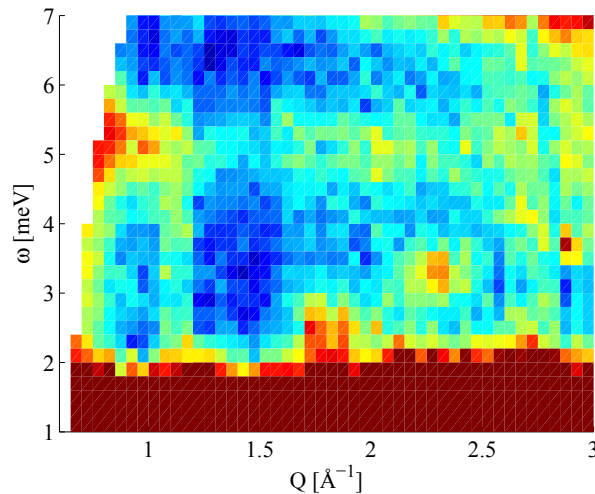


Figure 1. The powder integrated scattering function  $S(Q, \omega)$  at 0.05K.

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# Which model spin Hamiltonian for Volborthite ?

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The thermodynamic properties and magnetization curve of the Heisenberg Antiferromagnet with spatially anisotropic exchanges on the kagomé lattice computed from exact diagonalization are compared with experimental data for the distorted kagomé compound Volborthite, obtained by Hiroi *et al* [1-2]. Exact diagonalization results shows that if one consider a nearest-neighbor model, Volborthite would be then described with a quasi isotropic model [3]. Such a model, however, does not allow to fit fully satisfactorily the experimental data. This points to the presence of additional interactions. It is shown that the fit may be improved by models with more distant exchanges which can strongly differ from the isotropic nearest-neighbor model.

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## The Kagome Heisenberg Antiferromagnet: at a First-Order Phase Transition close to a Quantum Critical Point ?

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Using an effective Quantum Dimer Model approach, exact diagonalisations of periodic clusters with up to 108 sites ( $3 \times 6 \times 6$ ) reveal that the Quantum Heisenberg Antiferromagnet on the Kagome lattice lies at a first-order phase transition between two 36-site unit cell Valence Bond Crystals (VBC) preserving lattice point group symmetries but with opposite parities. Evidence for a nearby Quantum Critical Point characterized by the melting of the even VBC into a  $Z_2$  dimer liquid are given. Implications regarding numerical analysis and experiments are discussed [1].

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# A Generalized Quantum Dimer Model for the singlet sector of the kagome antiferromagnet

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Hardcore dimer models are widely used to explore exotic non magnetic ground states emerging in frustrated bidimensionnal antiferromagnets. However since pionnering work by Rokhsar and Kivelson, many issues are still open in understanding the connection between such models and the parent Heisenberg spin models. We propose an alternative and systematic route to derive generalized Quantum Dimer Models that are suitable for a quantitative description of the low energy singlet sector of the corresponding antiferromagnets. After outlining the main characteristics of the method (details available in D. Schwandt's poster), we will focus on establishing a Generalized Quantum Dimer Model for the low energy singlet sector of the kagome antiferromagnet. Large scale numerical computation (presented by D. Poilblanc) of this model demonstrate its ability to capture the peculiar physics of the heisenberg hamiltonian.

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Wednesday 20th January 2010

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## Neutron scattering and $\mu$ SR investigations of the spin liquid state with quenched disorder in $\text{LuCuGaO}_4$ .

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$\text{LuCuGaO}_4$  has magnetic  $\text{Cu}^{2+}$  and diamagnetic  $\text{Ga}^{3+}$  ions distributed on a triangular bilayer. Susceptibility,  $\mu$ SR and neutron scattering measurements show that at low temperature the spins form a short range correlated state with a gapless continuum of excitations. The development of this spin liquid state is strongly reminiscent of that in other two dimensional frustrated magnets, in particular Herbertsmithite, and we suggest that it is a generic property of such systems, even surviving in the presence of strong quenched disorder.

# Dimerization patterns in kagome based quantum magnets

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We present extensive numerical DMRG and ED results focussing on emergent dimerization patterns in two prototypical kagome systems: i) The kagome strip introduced by Waldtmann *et al.* has a rich phase diagram including gapless and gapped dimerized phases with several distinct spatial patterns ii) We uncovered remarkable dimerization patterns hidden among the low energy singlet states of the 36 sites 2D kagome lattice. We interpret their structure and dynamics and discuss the implications for dimerization in the thermodynamic limit.

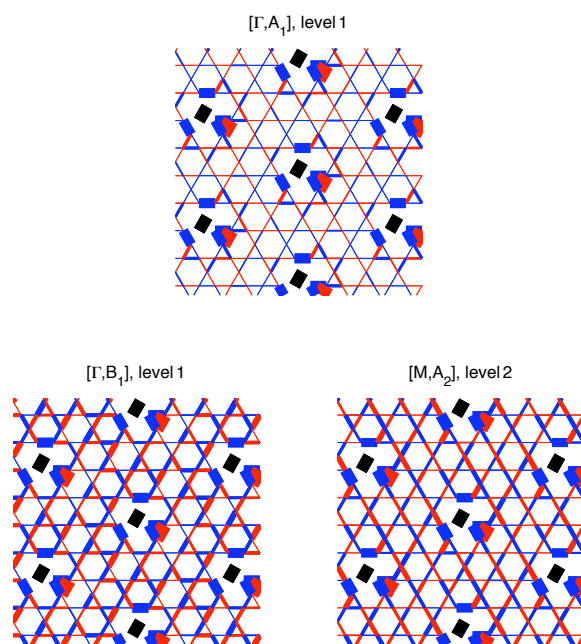


Figure 1: Dimer-dimer correlation patterns in low lying eigenstates of the kagome Heisenberg model on a 36 sites sample. While the ground state (top panel) has quite weak dimer correlations, many low-lying levels show very pronounced dimer patterns. Two such states are shown in the lower panels.

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# Magnetism and Frustration in Hexagonal $\text{RMnO}_3$ with Triangular Lattice

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We present results from high resolution neutron diffraction and inelastic neutron scattering experiments in the frustrated multiferroic hexagonal compounds  $\text{RMnO}_3$  ( $R=\text{Ho, Yb, Sc, Y}$ ). These results clearly highlight a strong magneto-elastic coupling in the whole family. We can correlate the atomic positions, the type of magnetic structure and the nature of the spin waves whatever the R ion and temperature. We show that magnetic frustration is at play not only in the Mn magnetic planes, but also along the c-axis due to the competition between two exchange paths. The key parameter to stabilize a given type of magnetic order is the position of the Mn ions in the unit cell with respect to a critical threshold of  $1/3$ , which determines the sign of the coupling between Mn triangular planes.

A Mn displacement around this  $1/3$  value tends to release the interplane frustration as understood by looking at the exchanges paths (cf. Fig. 1). For this specific position a 2D magnetic order could be expected. This situation may be realized in  $\text{InMnO}_3$  where coupling between adjacent planes seems to vanish.

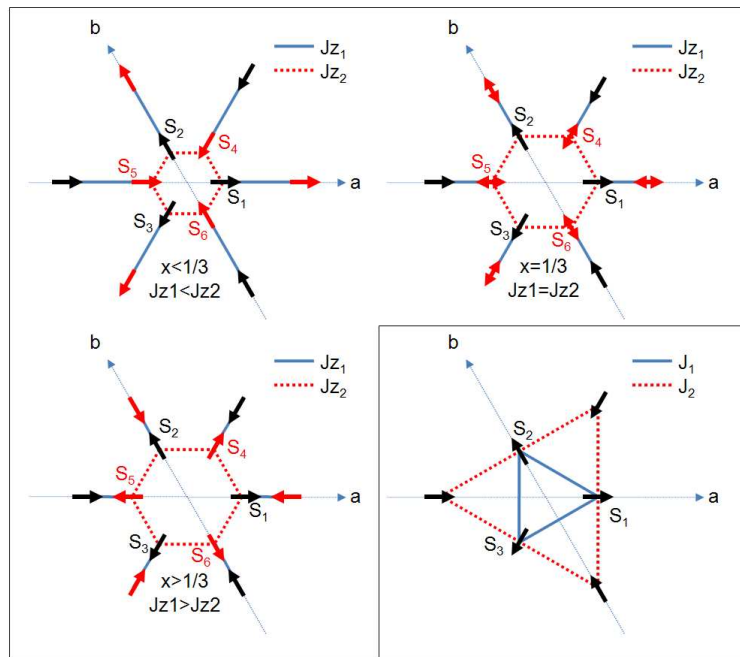


Figure 1. Scheme of the exchange paths in hexagonal  $\text{RMnO}_3$ . Effective coupling between planes is driven by the Mn shift from the frustrated  $1/3$  position.

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# Towards the Kasteleyn Transition in Kagomé Ice

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There has been much interest recently in spin ice materials on account of the discovery of ‘magnetic monopoles’ [1] and the associated phenomenon, ‘magnetricity’ [2]. Behind these results there is a deeper and even more fascinating physics: the physics of topological constraint. Kagomé Ice is a topologically constrained phase formed by the application of a magnetic field along an easy axis of the spin ice model. It has been shown to map onto a hardcore dimer model on a hexagonal lattice [3] with the associated implication that, under the influence of an appropriate perturbation, the critical phase will undergo a Kasteleyn phase transition [4]; a transition that, until observed in spin ice by members of this group [5,6], had previously only been seen in biological systems.

We explain the unusual characteristics of this transition in Kagomé Ice and present recent experimental and theoretical work on this system which clearly shows some of these signatures.

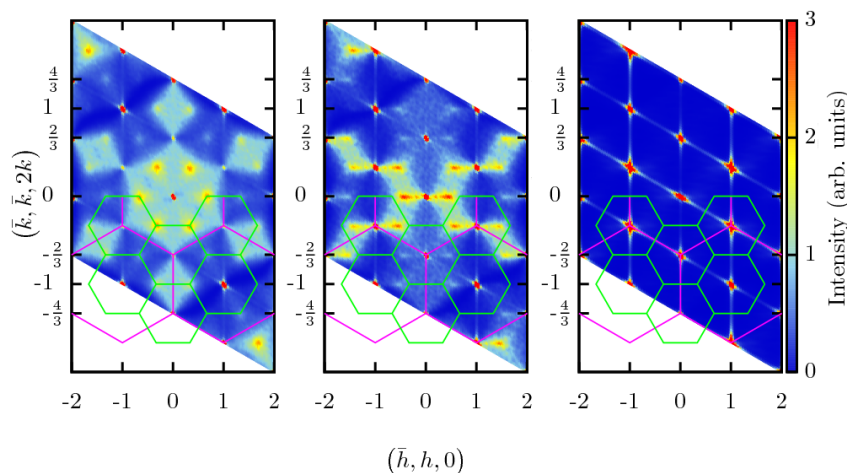


Figure 1: The effect of a small tilt in the magnetic field away from the [111] direction on the Kagomé plane unpolarised neutron scattering map. The left figure has no field component on the Kagomé plane, the right figure has a strong enough component to have driven the system through the Kasteleyn transition and the central figure shows an intermediate field value.

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# Algebraic Spin Liquid on the Kagome Lattice

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The nature of the ground state of the  $S = 1/2$  kagome Heisenberg antiferromagnet is one of the most challenging and intriguing open problems of quantum magnetism. Experiments on herbertsmithite showing the absence of both magnetic order and a spin gap led Ran, Lee, Wen and myself to propose that the ground state may be an algebraic spin liquid [1, 2]. This state has a host of remarkable properties – perhaps most striking is the presence of a Dirac-fermion-like spectrum combined with a lack of good quasiparticle excitations, even at the lowest energies. In this talk, I will review our work, and give an update on the status of these ideas in light of recent experiments and numerical results.

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## <sup>35</sup>Cl NMR Study of the S=1/2 Kagome Antiferromagnet $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

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We report <sup>35</sup>Cl NMR measurements on the S=1/2 Kagome system, Herbertsmithite  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ . Using a well-oriented powder sample, the NMR frequency shift can be traced for two different orientations with the applied field parallel and perpendicular to the *c*-axis. We find a large broadening of the spectrum together with several features emerging only at low temperature below 5 K. The local spin susceptibility probed by <sup>35</sup>Cl is compared to the ones obtained by different nuclei (<sup>17</sup>O and <sup>1</sup>H) as well as the bulk susceptibility.

We will present a frustrated spin system based on pentagonal rings that form a loosely connected network. This structure is perfectly ordered, being a subtiling of the five-fold symmetric Penrose tiling, and is interesting to study, since it may lead to disordered ground states, as the Kagome lattice does. In addition, experimental groups in Liverpool have shown that it may be possible to realize such spin systems by adsorption of magnetic atoms on a good quasiperiodic substrate. We studied the minimal energy configurations of classical spins on a structure which is based on a finite size approximant of the perfect infinite quasiperiodic structure with periodic boundary conditions. This choice allows to eliminate boundaries but instead introduces a small number of defects (dangling bonds). The results obtained by a Monte Carlo calculation are shown in the figures below. fig.1 represents a snapshot of the spin angles for spins in a small area of the structure. The angles between pairs of nearest neighbor spins can be seen to fall into two groups: those corresponding to  $4\pi/5$  ( angle that minimizes the bond energies for spins placed around an isolated pentagon) and  $\pi$  for spins on either side of an unfrustrated bond. This is shown more clearly in fig.2, where the two types of bonds are shown by color coding the bonds : red for the frustrated and blue for the unfrustrated bonds. Other novel frustrated pentagonal systems will be briefly discussed.

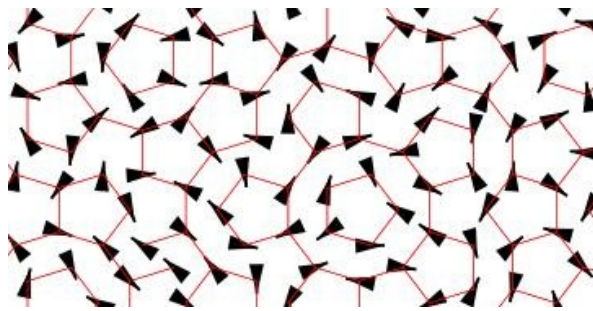


Fig.1 A low energy spin configuration

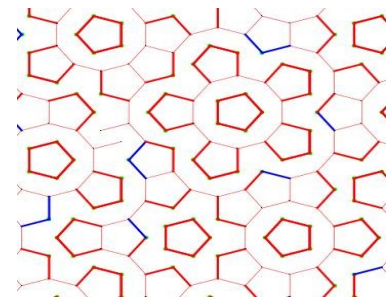


Fig.2 The bond energies (red: low, blue:high)

# Kapellasite, a new candidate for $S=1/2$ frustrated magnetism on the kagome lattice ?

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The recent discovery of the first structurally perfect 2D kagome compound, Herbertsmithite  $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$ , has generated much excitement in the community as the very first representative of a  $S=1/2$  kagome antiferromagnet (KAFM), which stabilizes a quantum spin liquid state. More recently, a new candidate for such a state has been synthesised [1]: the Kapellasite, a polymorph of Herbertsmithite, i.e. a compound with the same chemical formula but with a different structure. Its crystal structure may be a better approximation to the KAFM than Herbertsmithite, as the kagome layers are only weakly coupled by O-H...Cl hydrogen bonds, giving rise to a highly 2D system with well defined cleavage planes along (001). Our experiments involve local techniques such as  $^{35}\text{Cl}$  NMR and  $\mu\text{SR}$  down to respectively 1.2 K and 60 mK. We performed NMR measurement on magnetically aligned powders of Kapellasite, which allow the determination of the local susceptibility of the Cu kagome planes.

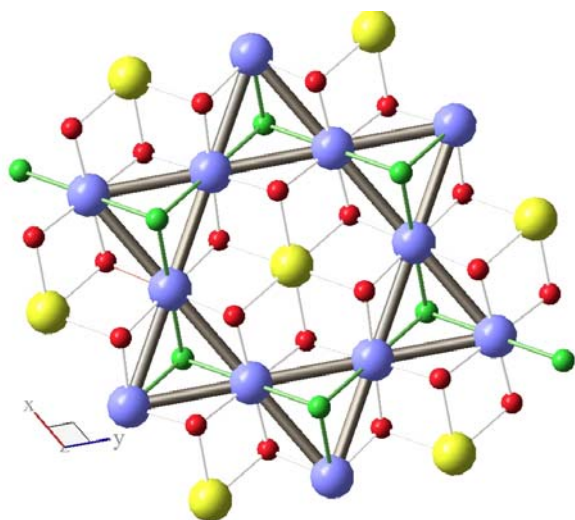


Figure 1. The kagome lattice of Kapellasite is constituted by copper atoms ( $S=1/2$ , blue).

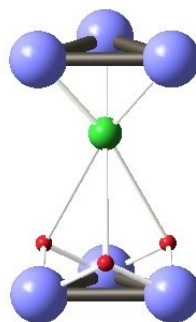


Figure 2. Local environnement of chlorine atom (green).

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# FIELD-INDUCED TRIPLET BEC IN FRUSTRATED BILAYERS

## Application to the Han Purple pigment $\text{BaCuSi}_2\text{O}_6$

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Building on recent neutron scattering<sup>a</sup> and high-field NMR experiments<sup>b</sup>, we investigate the field-induced Bose-Einstein condensation (BEC) of the triplet excitations observed with an exotic criticality<sup>c</sup> in the frustrated spin- $\frac{1}{2}$  bilayers compound  $\text{BaCuSi}_2\text{O}_6$ . Using a realistic frustrated model with two types of bilayers<sup>d</sup>, we are able to solve several puzzles, and make various predictions. We show that the perfect interlayer frustration leads to a 2D-like critical exponent  $\phi = 1$  for the field-temperature phase boundary  $T_c \sim (H - H_c)^\phi$  while the BEC phase displays 3D coherence.

<sup>a</sup> Ch. Rüegg *et al.*, Phys. Rev. Lett. **98**, 017202 (2007).

<sup>b</sup> S. Krämer *et al.*, Phys. Rev. B **76**, 100406(R) (2007).

<sup>c</sup> S. Sebastian *et al.*, Nature **441**, 617 (2006).

<sup>d</sup> N. Laflorencie and F. Mila, Phys. Rev. Lett. **102**, 060602 (2009).

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## Muon-Spin Spectroscopy of the organometallic spin 1/2 kagomé-lattice compound $\text{Cu}(1,3\text{-benzenedicarboxylate})$

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Using muon spin resonance we examine the organometallic hybrid compound  $\text{Cu}(1,3\text{-benzenedicarboxylate})$  [ $\text{Cu}(1,3\text{-bdc})$ ], which has structurally perfect spin 1/2 copper kagomé planes separated by pure organic linkers. This compound has antiferromagnetic interactions with Curie-Weiss temperature of  $-33$  K. We found slowing down of spin fluctuations starting at  $T = 1.8$  K, and that the state at  $T \rightarrow 0$  is quasi-static with no long-range order and extremely slow spin fluctuations at a rate of  $3.6 \mu\text{sec}^{-1}$ . This indicates that  $\text{Cu}(1,3\text{-bdc})$  behaves as expected from a kagomé magnet and could serve as a model compound. More can be found in Ref. [1].

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[1] L. Marcipar, O. Ofer, A. Keren, E. A. Nytko, D. G. Nocera, Y. S. Lee, J. S. Helton, and C. Baines, Phys. Rev. B **80**, 132402 (2009)

# Schwinger Boson Mean-Field Theory on the Kagomé lattice with Dzyaloshinskii-Moriya interaction

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On experimental realisations of spin 1/2 Kagomé antiferromagnets, the Dzyaloshinskii-Moriya (DM) interaction ( $\sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$ ) is present. This interaction can induce a transition from a spin liquid to a magnetically ordered phase [1]. The goal of our study is to find experimental signatures of this interaction. Few theoretical tools are able to describe strong quantum fluctuations. Among them, the Schwinger boson mean-field theory (SBMFT) [2] is known to possibly describe topological spin liquids as well as long-range ordered states. After a presentation of the SBMFT method, we present the zero temperature phase diagram of the Heisenberg antiferromagnet on the Kagomé lattice perturbed by a DM interaction (Fig. 1). Different phases labelled both by their fluxes and by their order (spin liquid or Néel phase) appear in this phase diagram. The influence of the Schwinger boson density will be analysed. Dynamical structure factors in different regions of the phase diagram will be presented.

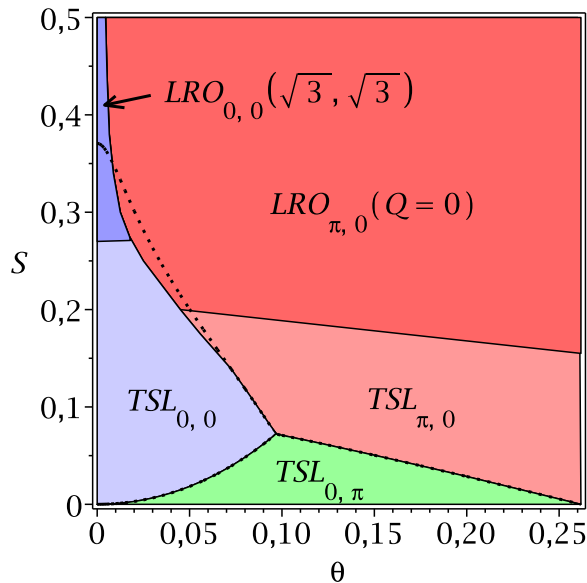


Figure 1. The zero temperature phase diagram of the Kagomé antiferromagnet ( $\theta = D/2J$ ). Long range ordered phases (LRO) and topological spin liquids (TSL) are found. Each phase is labelled by the fluxes ( $\Phi_{Hex}, \Phi_{Rho}$ ) around hexagons and rhombus.

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# Magnetic order in YbMnO<sub>3</sub> studied by Neutron Diffraction and Mössbauer Spectroscopy

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The magnetic ordering of the hexagonal multiferroic compound YbMnO<sub>3</sub> with triangular Mn lattice has been studied between 100 K and 1.5 K by combining neutron powder diffraction, <sup>170</sup>Yb Mössbauer spectroscopy and magnetization measurements. The Yb moments of the two crystallographic sites order at two different temperatures, the 4*b* site together with the Mn moments (at  $T_N \simeq 85$  K) and the 2*a* site well below (at 3.5 K). The temperature dependences of the Mn and Yb moments are explained within a molecular field model, showing that the 4*b* and 2*a* sites order via Yb-Mn and Yb-Yb interactions respectively. A simple picture taking into account the local Mn environment of the Rare earth R (4*b*) ion is proposed to couple R and Mn orders in hexagonal RMnO<sub>3</sub> manganites. The nature and symmetry of the R-Mn interactions yielding the R order are discussed.

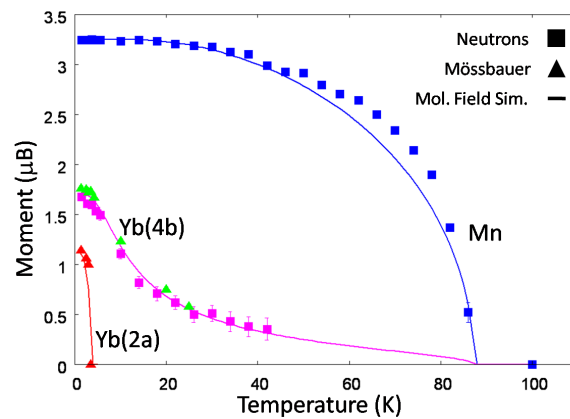


Figure 1. Observed Mn, Yb(4*b*) and Yb(2*a*) moments from neutron powder diffraction and Mössbauer measurements (dots) versus temperature. The solid lines correspond to the molecular field calculation.

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# Electrical and magnetic properties of the organic kagome system $(\text{EDT-TTF-CONH}_2)_6 [\text{Re}_6\text{Se}_8(\text{CN})_6]$ : a transport and Electron Spin Resonance study

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Frustrated magnetism on the kagome lattice was intensively investigated in the past years, in particular after the discovery of the structurally perfect Herbertsmithite with spin 1/2. While most studies focus on localized spins, the itinerant case is less addressed and very few experimental realizations are available. From the theoretical point of view, the metal-insulator transition was studied for the system at half filling, and it was shown that the frustration stabilizes the metallic state up to relatively high values of the coulomb repulsion [1].

Here we present an experimental study of the hybrid organic-inorganic system  $(\text{EDT-TTF-CONH}_2)_6 [\text{Re}_6\text{Se}_8(\text{CN})_6]$ , for which organic dimers organize on a kagome lattice at high temperature, above 200 K. Four holes with spin 1/2 are shared by the three dimers of the unit cell. We present resistivity measurements that reveal a metallic behavior of the kagome plane. On the other side, the spin susceptibility extracted from Electron Spin Resonance (ESR) spectra has a Curie-Weiss temperature dependence, indicating the presence of localized moments with antiferromagnetic correlations and  $\theta_{CW} \sim 170$  K. This system is therefore a rare, if not unique, example of an itinerant kagome antiferromagnet. The ESR signal, dominated by the magnetic contribution, provides evidence for Dzyaloshinsky-Moriya interaction with  $D/\theta_{CW} \sim 2\%$ . This value is much smaller than that obtained for the inorganic Herbertsmithite  $D/\theta_{CW} \sim 8\%$ .

At lower temperature, a smooth structural transition drives the system into a triclinic state. The sliding motion of molecules within one of the dimers leads to the formation of antiferromagnetic chains with spin 1/2. By means of ESR, we track the evolution of the structure under hydrostatic pressure and we show that a net increase of the transition temperature occurs above 0.7 GPa.

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Motivated by the recent NMR experiments on  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ , we study the effect of nonmagnetic defects on the antiferromagnetic spin-1/2 kagome lattice [1,2]. We use exact diagonalization methods to study the effect of two such defects on finite-size systems. Our results, obtained without adjustable parameters, are in good quantitative agreement with recent 17O NMR data. They provide support for the experimental interpretation of the presence of defects within the kagome layers due to Zn/Cu substitutions. Our results also show that disorder effects become relevant at lower temperatures, raising questions about the experimental evidence for the absence of an intrinsic spin gap in the kagome two-dimensional layers.

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## Derivation of an effective Quantum Dimer Model

**D. Schwandt**<sup>a,b</sup>, M. Mambrini<sup>a,b</sup>, D. Poilblanc<sup>a,b</sup>

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The understanding of frustrated bidimensional antiferromagnets with exotic, non-magnetic ground-states is often difficult because of the absence of efficient analytical and numerical techniques. Effective Hamiltonians can be a powerful tool to simplify the underlying physical model without losing important physical aspects. In order to find effective models it is important to understand how they can arise. One prominent example is the Rokhsar Kivelson model and its underlying Heisenberg spin model.

We propose a way to systematically derive general quantum dimer models (GQDM) for Heisenberg antiferromagnets. It turns out that there is a relation to the linked cluster theorem, a prerequisite for a local effective model (presented by M. Mambrini). We manage to understand and extend the emergence of Rokhsar Kivelson models and successfully apply this technique to the kagome antiferromagnet (presented by D.Poilblanc).

### References

[1] D. Schwandt, M. Mambrini and D. Poilblanc, ArXiv.

[2] D. Poilblanc, M. Mambrini and D. Schwandt, ArXiv.

# Synthesis of atacamites $\text{Cu}_3\text{M}(\text{OH})_6\text{Cl}_2$ ( $\text{M} = \text{Zn}, \text{Mg}, \text{Cd}$ )

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The *atacamite* structural family, of which the *herbertsmithite* is the most famous member, is a series of lamellar hydroxychlorides with a perfect Kagome network of  $\text{Cu}^{2+}$  cations with generic formula  $\text{Cu}_3\text{M}(\text{OH})_6\text{Cl}_2$  (see Fig.1). The structure can be rhombohedral, space group R-3m, orthorhombic Pmcn or monoclinic  $\text{P2}_1/\text{n}$  depending on composition [1].

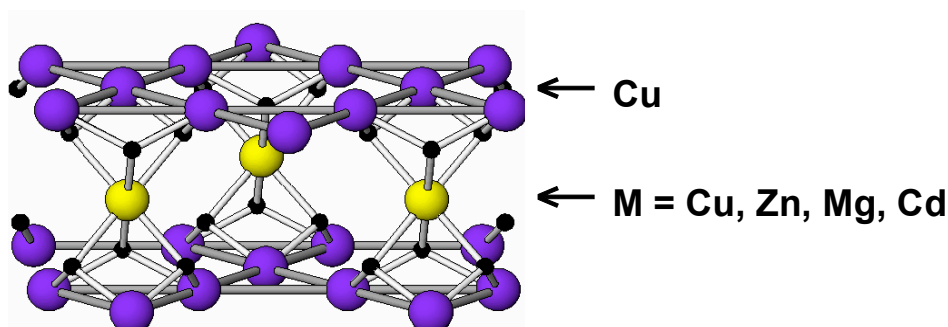


Figure 1. Atacamite structure, showing Kagome planes of  $\text{Cu}^{2+}$ .

The classical synthesis route of atacamites consists in a hydrothermal treatment of copper hydroxycarbonate in water [2]. It seems to be unsuitable to  $\text{M} \neq \text{Zn}$ . We also investigated a procedure first proposed by Feitknecht et al. [3], using metallic copper as a starting reagent and a gentle oxidation by air or gaseous oxygen in a solution of  $\text{MCl}_2$  ( $\text{M} = \text{Cu}, \text{Zn}, \text{Mg}, \text{Cd}$ ).

We will describe in this communication the results of synthesis of various compounds of this family. The hydrothermal route allowed us to obtain well-formed crystals of  $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$ , although the size remains sub-millimetric (Fig.2). The reaction with metallic Cu is well suited to the synthesis of pure Cd and Mg atacamites. Main results are summarized in the table:

<i>M</i>	<i>method</i>	<i>product</i>	<i>Symmetry</i>
Cu	Cu + $\text{CuCl}_2$	Pure atacamite	$\text{P2}_1/\text{n}$ ,
Zn	hydrothermal	Pure atacamite	R-3m
Zn	Cu + $\text{ZnCl}_2$	(no atacamite formed)	
Cd	hydrothermal	(no atacamite formed)	
Cd	Cu + $\text{CdCl}_2$	Pure atacamite	R-3m
Mg	Cu + $\text{MgCl}_2$	Pure atacamite	R-3m

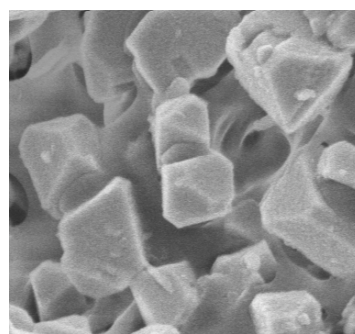


Figure 2.  $\text{Cu}_3\text{Zn}(\text{OH})_6\text{Cl}_2$ , crystals (full scale 2 mm)

## References

- [1] RS.W. Braithwaite et al., *Mineral. Mag.* 68, 527 (2004)
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# The Correlation Density Matrix tool for extracting dominant correlations in strongly interacting systems

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Dominant correlations between two disjoint blocks in a strongly interacting spin system can be revealed in an unbiased way by computing the Correlation Density Matrix between them, and singular value decompose it. After an introduction and a discussion about consequences of symmetries on it, this method will be applied to the spin-1/2 antiferromagnet on the Kagome lattice for illustration. Specific low-lying eigenstates as well as the general finite temperature behavior of this model will be investigated.

## References

[1] J. Sudan and A.M. Läuchli, *Correlation Density Matrix study of the  $S = 1/2$  kagomé antiferromagnet: relevance of the valence bond crystal scenario*, in preparation.

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## Raman scattering study of the kagomé lattice compound $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

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We present a Raman spectroscopic investigation on the Herbertsmithite  $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$ , which is a realization of the spin  $\frac{1}{2}$  Heisenberg antiferromagnet on a kagomé lattice – a highly frustrated compound with no evidence of magnetic order down to 50 mK.

Our measurements show a strong low energy (“quasi-elastic”) scattering contribution, which is depleted algebraically to low temperatures, revealing a weak polarization dependent scattering continuum. Our observations give reason to believe that the ground state of this system is gapless and of algebraic spin-liquid nature, as already predicted theoretically.

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