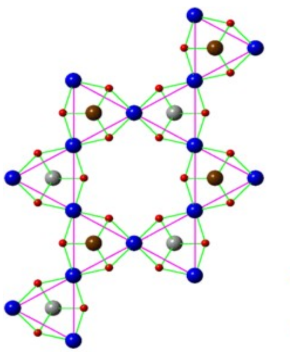
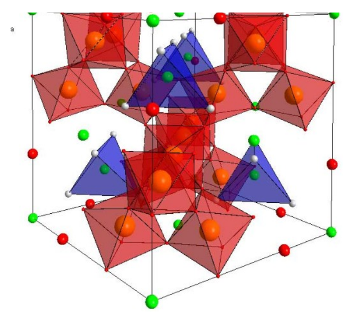
Introduction to Magnetism and Magnetic Materials – part II

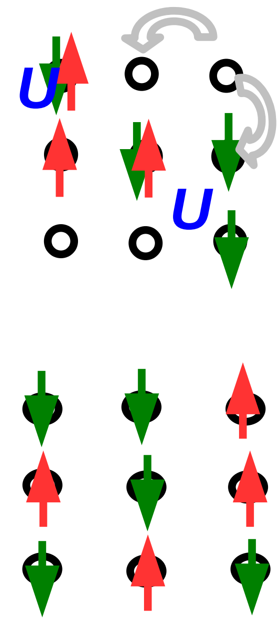
DMR - 1644779
DMR - 2046570

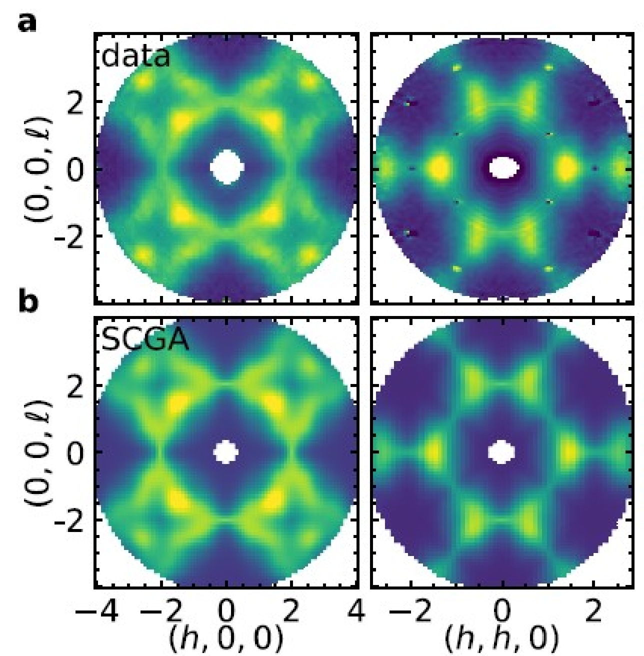
Hitesh J. Changlani
Florida State University and MagLab

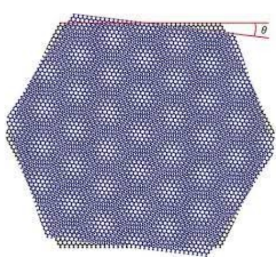
Planck computing + RCC @ FSU

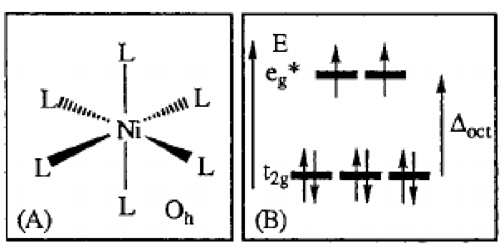










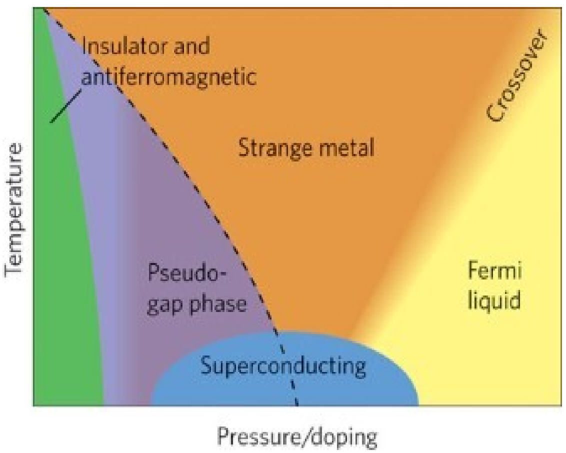


Magnetic Properties from First Principles School, Turkey, Nov. 24, 2021

Outline of the lectures

- ***What, why* and *how* of quantum magnetism**: How does a spin model arise from a fermionic one? What new phases of matter can emerge in these materials?

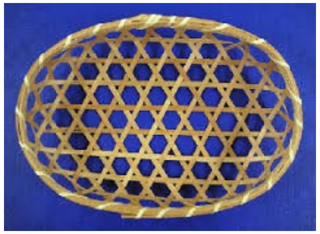
- What are some (new and old) **problems in magnetism** that we care about solving and what is the status of their solution?



$$H_{\text{Hubbard}} = -t \sum_{\langle i,j \rangle,\sigma} c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_i n_{i,\uparrow} n_{i,\downarrow}$$

$$H_{\text{Heisenberg}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j ;$$

“Kagome” Japanese basket

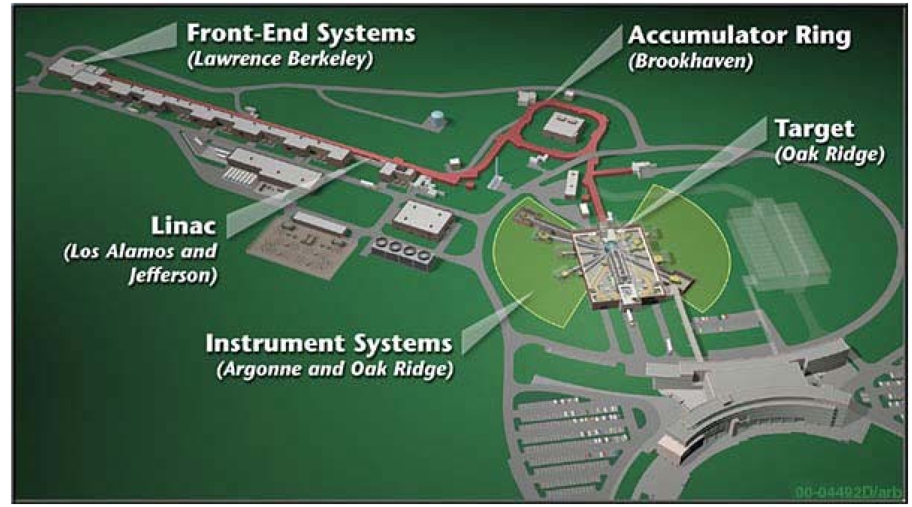


Herbertsmithite

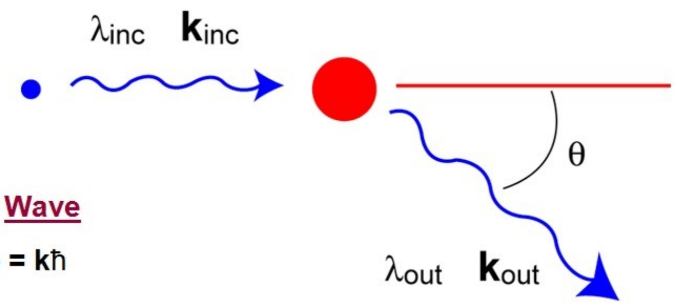
- What are some **analytic + numerical techniques** that have been useful?

- **Where are we headed to next**? Important inputs from numerical techniques (such as density functional theory) are crucial for making connections to what is seen in experiments. The grand challenge is to develop **predictive tools** to study these systems.

Neutron scattering: an important probe for magnetism



Principles of neutron scattering



Particle

$E_{kin} = \frac{mV^2}{2}$

Wave

$\mathbf{p} = \mathbf{k}\hbar$

Scattering proces

$\hbar\omega = E_{out} - E_{inc} = \frac{m}{2}(V_{out}^2 - V_{inc}^2)$ information about microscopic motion

$\mathbf{Q} = \mathbf{k}_{out} - \mathbf{k}_{inc}$ information about microscopic structure

Fig. from Helmholtz-Zentrum Berlin

$$\mathcal{S}^{\mu\nu}(\mathbf{q},\omega) = \frac{1}{2\pi N}\sum_{i,j=1}^{N}\int_{-\infty}^{\infty} dt\, e^{-i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j)+i\omega t} \times \langle s_i^{\mu}(t) s_j^{\nu}(0)\rangle.$$

- Neutron has spin (and no charge), which interacts with electron spin
- Neutrons produced are typically in the few meV to few 100 meV range, energy transfer is also roughly that order

How should we calculate spin dynamics*?

see: S. Zhang, H.J.C, K. Plumb, O. Tchernyshyov, R. Moessner, *Phys. Rev. Lett.* (2019)

Monte Carlo + Landau-Lifshitz spin dynamics (MD/LL)

A. Keren, PRL (1994)
P. Conlon, J. Chalker, PRL (2009)

Effective magnetic field experienced by spin, where H = Hamiltonian

$$\frac{d}{dt}\mathbf{s}_i = -\mathbf{s}_i \times \frac{\partial H}{\partial \mathbf{s}_i}$$

Average dynamics over many initial configurations (IC) chosen from a thermal ensemble generated in Monte Carlo (MC)

$$\langle s_i^\mu(t) s_j^\nu(0)\rangle = \sum_{\text{IC from MC}} s_i^\mu(t) s_j^\nu(0)\big|_{\text{IC}}$$

In practice, time evolution with fourth order Runge Kutta method.

Approx. 10,000 MC samples were used, and up to 8192 spins

Stochastic "Large N" theory (SLN)

P. Conlon, J. Chalker, PRL (2009)
D. Garanin, B. Canals, PRB (1999)

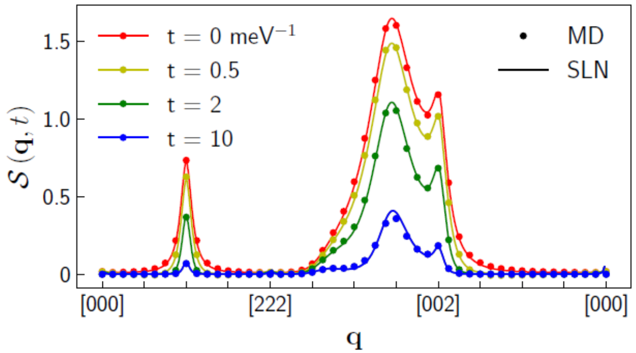
Model for hydrodynamics of spins (Langevin equation) in the presence of a random "noise" term which mimics the role of temperature

$$\frac{d}{dt}s_i^\mu = \gamma \sum_j \Delta_{ij} \frac{\partial E}{\partial s_j^\mu} + \xi_i^\mu(t)$$

Gaussian noise term

1) Assume spin is a "soft" one – i.e. its length is not fixed
2) Assume spin has N components, N large

Under these conditions, an exact analytical expression can be obtained, but γ needs to be fit to MD.



Linear spin wave theory (LSWT)

P. W. Anderson, PR (1952)
R. Kubo, PR (1952)

The only semi-quantum approach among the three techniques

LSWT requires a unique ground state to perturb around

We use Monte Carlo to "sample" many local low energy minima (these include ordered and disordered configs)

LSWT in real space (8192 sites)

(parallely also realized by X. Bai et al PRL 2019 – Mourigal and Chalker groups)

(*Detailed math. expressions for dynamical structure factor in the supplement of our paper)

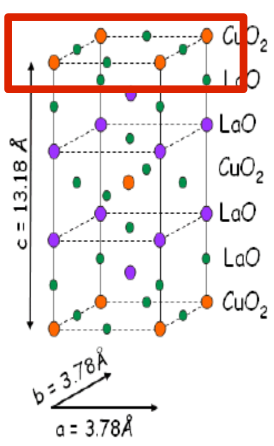
The undoped cuprate – effectively a 2D square lattice AFM

$$H_{\text{Heisenberg}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$

Neutron Scattering Studies of Antiferromagnetic Correlations in Cuprates

John M. Tranquada

Neutron scattering studies have provided important information about the momentum and energy dependence of magnetic excitations in cuprate superconductors. Of particular interest are the recent indications of a universal magnetic excitation spectrum in hole-doped cuprates. That starting point provides motivation for reviewing the antiferromagnetic state of the parent insulators, and the destruction of the ordered state by hole doping. The nature of spin correlations in stripe-ordered phases is discussed, followed by a description of the doping and temperature dependence of magnetic correlations in superconducting cuprates. After describing the impact on the magnetic correlations of perturbations such as an applied magnetic field or impurity substitution, a brief summary of work on electron-doped cuprates is given. The chapter concludes with a summary of experimental trends and a discussion of theoretical perspectives.



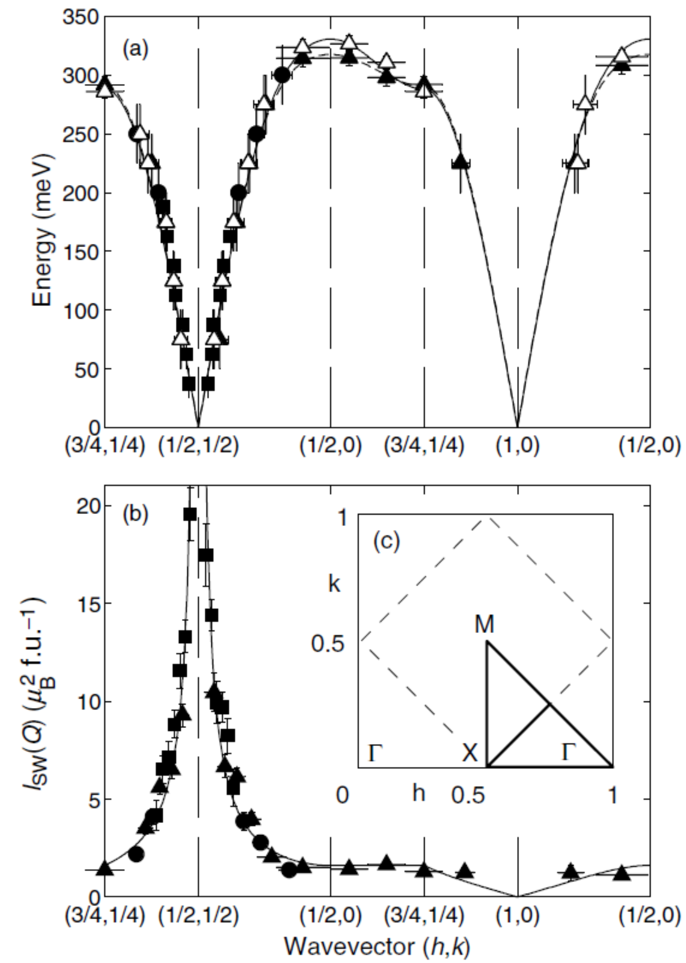
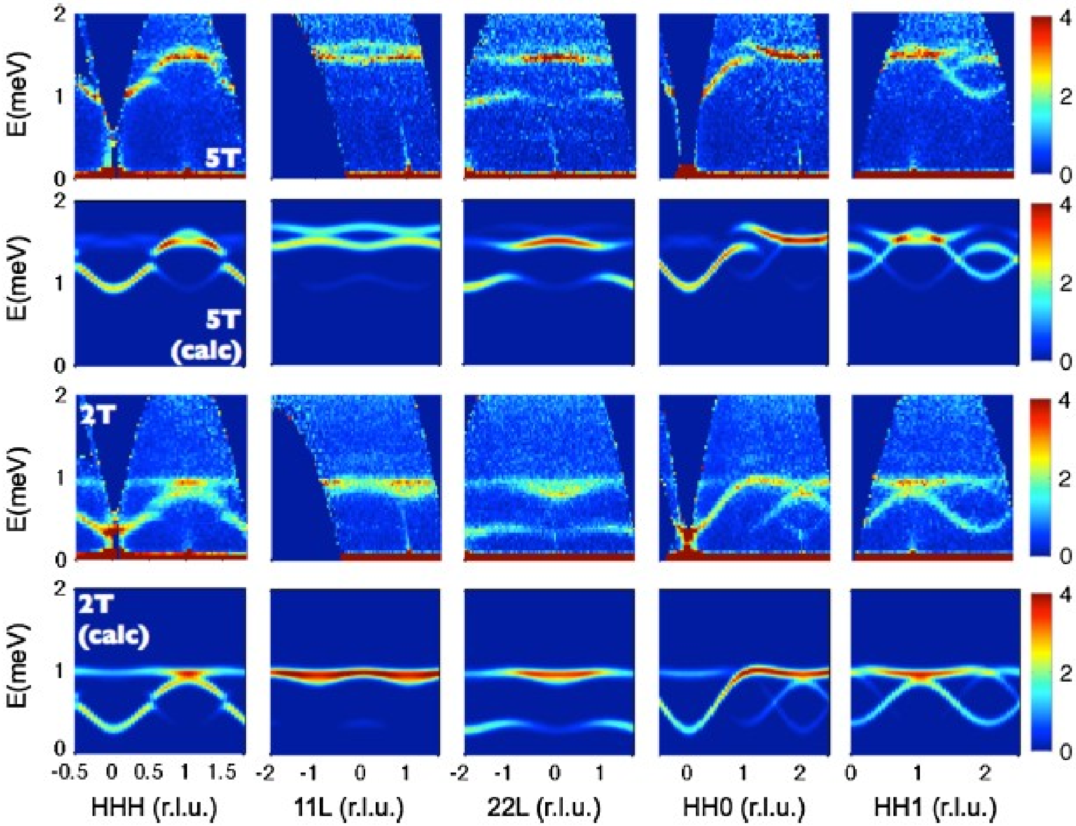


Figure 6.9. (a) Spin-wave dispersion in La_2CuO_4 along high-symmetry directions in the 2D Brillouin zone, as indicated in (c); $T = 10$ K (295 K): open (filled) symbols. Solid (dashed) line is a fit to the 10-K (295-K) data. (b) Spin-wave intensity vs. wave vector. Line is prediction of linear spin-wave theory. From Coldea et al. [68].

Spin wave theory in practice – account for general spin directions



$$
\mathbf{s}_i = \sqrt{S^2 - S(x_i^2 + y_i^2)}\,\mathbf{u}_i + \sqrt{S}(x_i \mathbf{v}_i + y_i \mathbf{w}_i)
$$

$$
\approx \left(S - \frac{x_i^2 + y_i^2}{2}\right)\mathbf{u}_i + \sqrt{S}(x_i \mathbf{v}_i + y_i \mathbf{w}_i).
$$

$$
\mathcal{S}^{\mu\nu}_{\text{quantum}}(\mathbf{q},\omega) = \frac{S}{N}\sum_{i,j=1}^{N} e^{-i\mathbf{q}\cdot(\mathbf{r}_i - \mathbf{r}_j)} \times \sum_{\alpha} (\eta_i^\mu \cdot \psi_\alpha^i)\,(\eta_j^\mu \cdot \psi_\alpha^j)^\star\,\delta(\omega - \omega_\alpha)\mathrm{sgn}(\omega_\alpha)
$$

$$
\psi_\alpha^i \equiv \left([\psi_\alpha]_{2i}, [\psi_\alpha]_{2i+1}\right)^T
$$

$$
\eta_i^\mu \equiv (v_i^\mu, w_i^\mu)
$$

PHYSICAL REVIEW X **1**, 021002 (2011)

Quantum Excitations in Quantum Spin Ice

Kate A. Ross,[1] Lucile Savary,[2] Bruce D. Gaulin,[1,3,4] and Leon Balents[5,*]
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(Received 22 July 2011; published 3 October 2011)

PHYSICAL REVIEW LETTERS **122**, 167203 (2019)

Dynamical Structure Factor of the Three-Dimensional Quantum Spin Liquid Candidate $NaCaNi_2F_7$

Shu Zhang,[1,2] Hitesh J. Changlani,[3,4,1,2] Kemp W. Plumb,[5] Oleg Tchernyshyov,[1,2] and Roderich Moessner[6]
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[6]*Max-Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany*

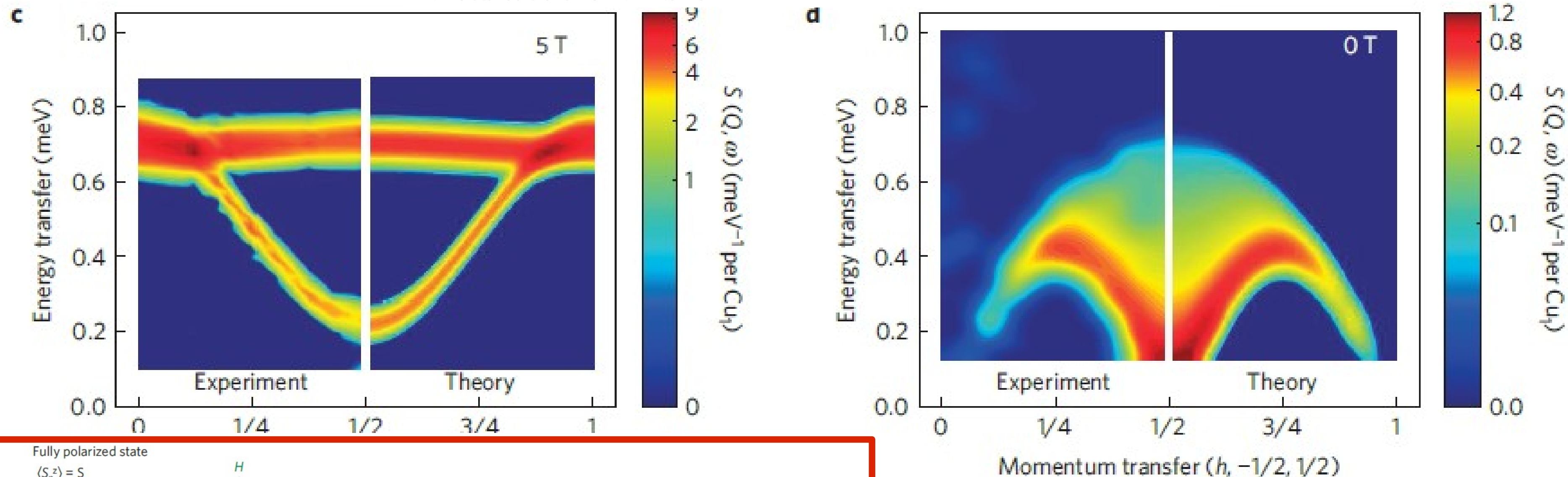
How should we deal with problems with no ordered state? Continuum of excitations due to "fractionalization"

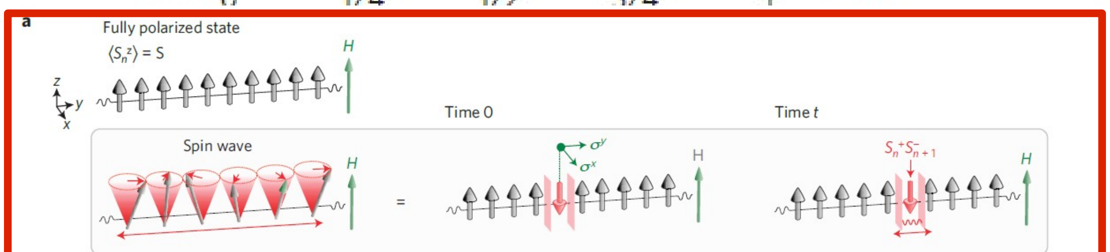
$CuSO_4 \cdot 5D_2O$

nature physics | ARTICLES
PUBLISHED ONLINE: 16 JUNE 2013 | DOI: 10.1038/NPHYS2652

Fractional spinon excitations in the quantum Heisenberg antiferromagnetic chain

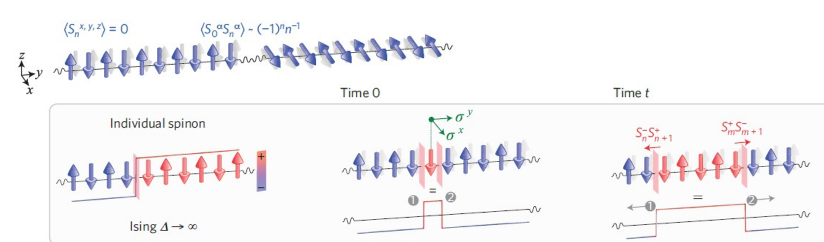
Martin Mourigal[1,2,3]*, Mechthild Enderle[1], Axel Klöpperpieper[4], Jean-Sébastien Caux[5], Anne Stunault[1] and Henrik M. Rønnow[2]





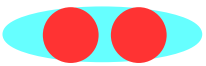
Spin wave theory works!

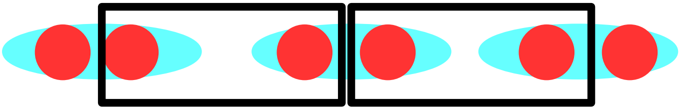
Beyond spin wave!



Low dimension + integer spin is even more unusual!

$$H_{\text{Heisenberg}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$

Spin-1 = 



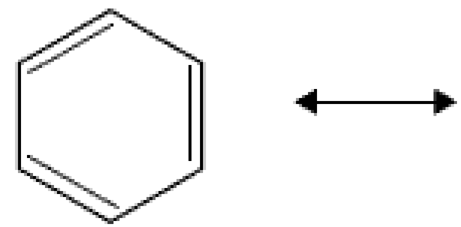
Thouless **Haldane** **Kosterlitz**

Consequence of “topology” and “fractionalization”

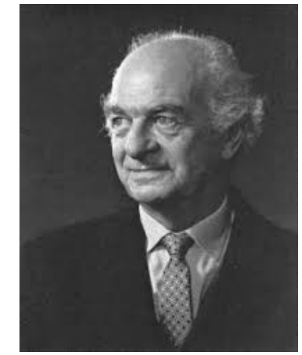
(2016)

H.J. Changlani, FSU

“Quantum spin liquids”: ground state is disordered
(States outside conventional Landau-Ginzburg-Wilson paradigm)

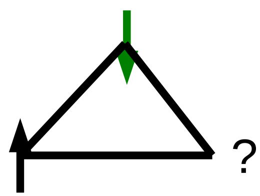


Two resonance Kekule forms of benzene

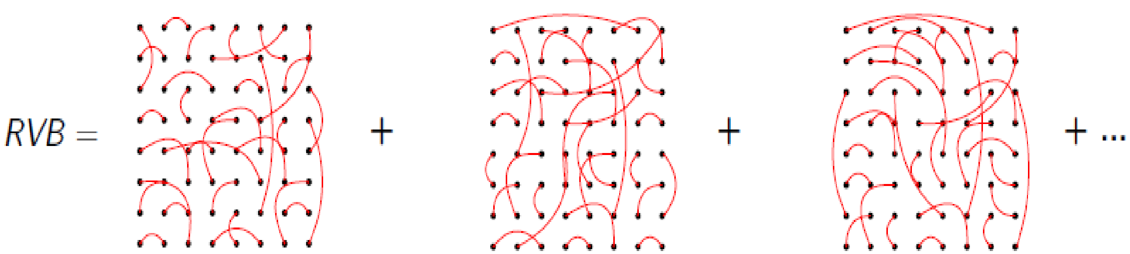


Linus Pauling

PW Anderson



Original proposal of triangular lattice spin liquid



F. Becca's talk

Spin liquid is a phase of matter with

(1) no symmetry breaking, no local order parameter to characterize phase

(2) yet have long range “entanglement” (property important for collective/fractional excitations)

(3) More modern definitions emphasize role of topology and also relax (1)

Possibly for quantum computing (Kitaev)

Current effort: look for it in real materials!

Review: L. Savary, L. Balents, Rep. Prog. Phys. (2017)

OK.. let us just try to solve this on a (big) computer!

$$H\,\psi = E\,\psi$$ “Solve” the Schrodinger equation

$$|\psi\rangle = \sum_{q_1 q_2 \ldots . q_N} \Psi^{q_1 q_2 \ldots . q_N} \, |q_1 q_2 \ldots . q_N\rangle$$

q on a site i is ○ ↑ ↓ ↓↑ or ↑ ↓ (depends on model)

Problem?

The Hilbert space is **HUGE!**

N=100 spins ⇒ $2^{100} \sim 10^{30}$ states ⇒ 10^{16} PetaBytes

Summit (Oak Ridge) has largest storage array = 250 PetaBytes

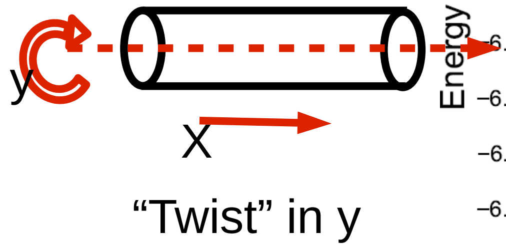
Exact diagonalization (30-50 sites/spins)

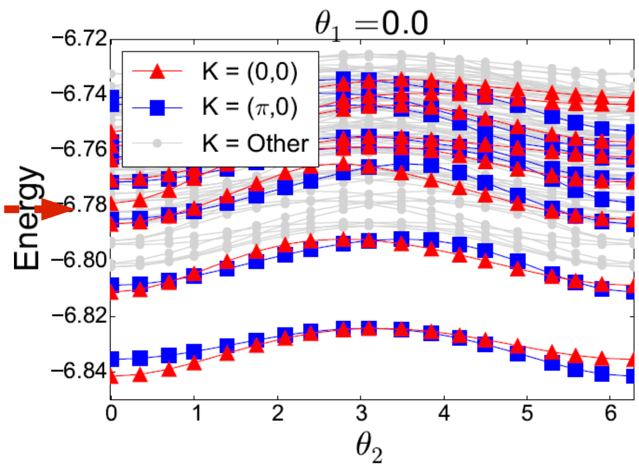
Conceptually simple, but tricks to make it work efficiently

Toolkit for theoretical toy "experiments" for discovering exotic phases

Laughlin's flux pumping experiment to detect "topological order" (chiral spin liquid).
K. Kumar, **HJC**, B. Clark, E. Fradkin, *PRB* (2016)

Simplex solid phase in a spin 1 system.
HJC, A. Lauchli, *PRB* (R) (2015)



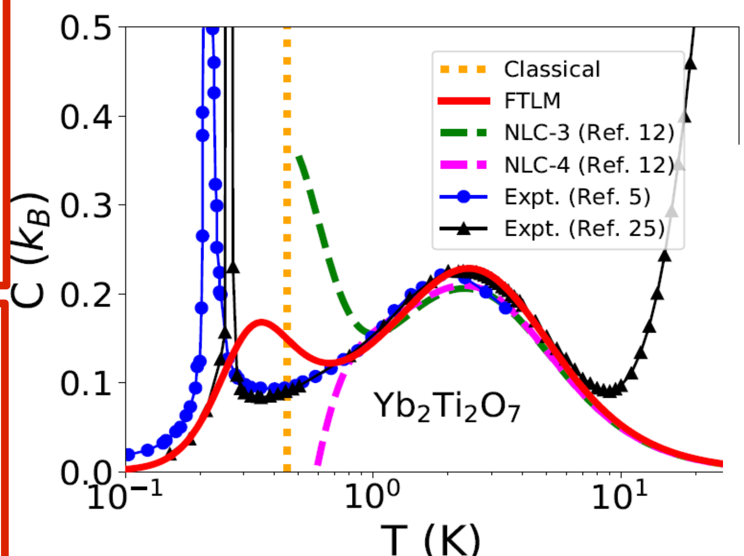


Finite temperature treatments

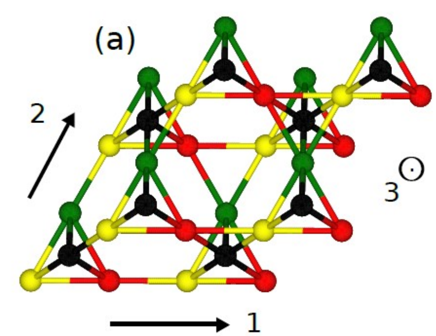
"quantum spin ice" candidate material (32 site cluster, 536 million dimensional space, 1 million core hours for phase diagram)

HJC (2017), A. Scheie et al (**HJC** lead theorist) *PRL* (2017), *PNAS* (2020)

Recent finite temperature Lanczos (FTLM) calculations on experimentally relevant pyrochlores (frustrated systems in 3d)
HJC (2017)



pyrochlore

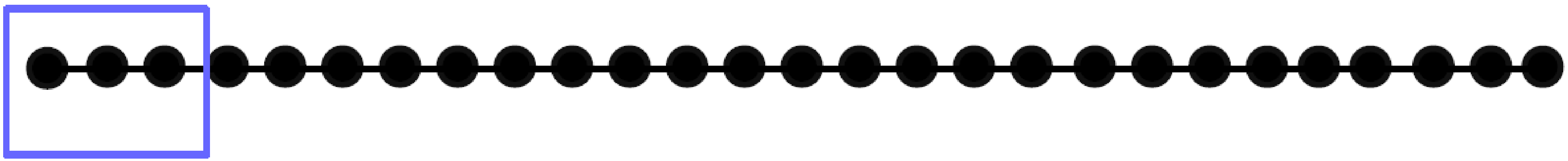


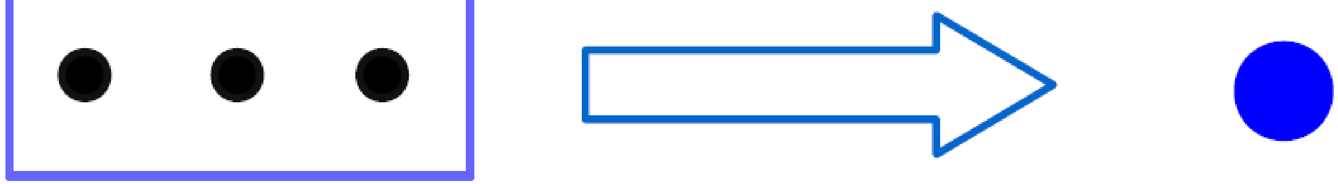
Fingerprints of order and **entanglement** from reduced density matrices

Li+Haldane, C. L. Henley + **HJC...** *Vishvanath, Bernevig, Grover, K. Yang, Melko*

Divide and conquer strategy: Renormalization group

Key advances due to Ken Wilson ,  **L. Kadanoff, M. Fisher, B. Widom …**



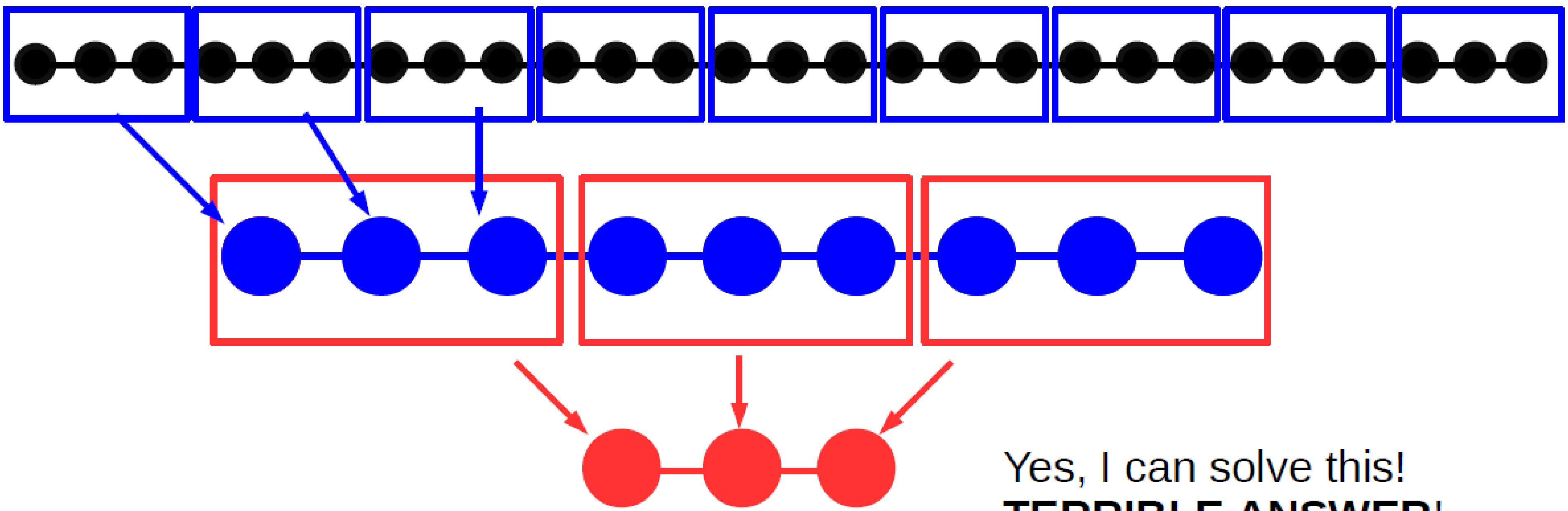


"Truncate" could mean any set of rules that reduce 2x2x2 =8 states to 2 states.

Divide and conquer strategy: Renormalization group

Key advances due to Ken Wilson ,  L. Kadanoff, M. Fisher, B. Widom …

$$H_{\text{Heisenberg}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j ,$$



Yes, I can solve this!
TERRIBLE ANSWER!

Problem???

Does not work very well.
Inter block interactions
/block boundaries are important!

Solution

Incorporate **correct boundary conditions** when truncating!

Steve White
(PRL, 1992)

H.J. Changlani, FSU

Efficient quantum wavefunctions: an information compression problem

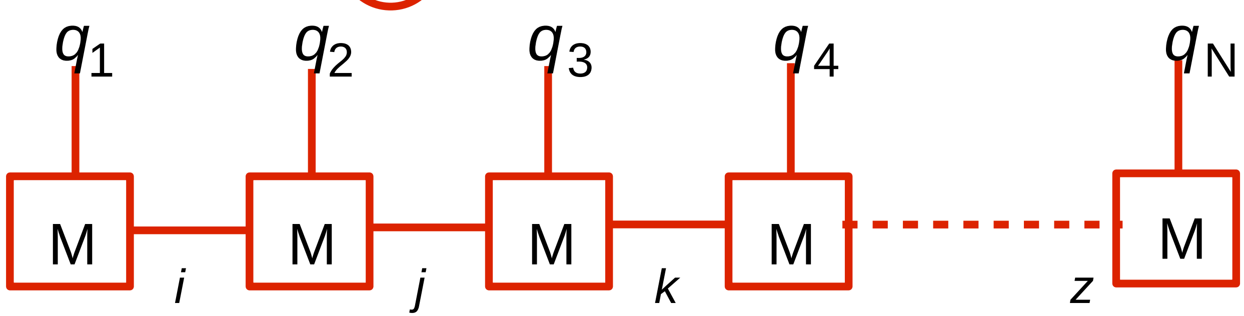
matrix product state (MPS): basis of density matrix renormalization group
(S. White 1992, building on Wilson's work Later: Ostlund-Rommer+Vidal, Verstraete + others...)

$$|\Psi\rangle = \sum_{q_1 q_2 \dots q_N} \Psi^{q_1 q_2 \dots q_N} |q_1 q_2 \dots q_N\rangle$$

Physical indices
Spins/electron
occupation number

$$\Psi^{q_1 q_2 q_3 \cdots q_N} = \sum_{i,j,k,\dots z} M_i^{q_1} M_{ij}^{q_2} M_{jk}^{q_3} \dots M_z^{q_N}$$

Auxiliary indices
(summed over)
have dimension D



Matrix
Product State
(MPS)

Amount of information ≈ N sites x (dimension of matrix=D)^2

For a 100 spin problem with D=200 need about 60 MB (vs 10^{16} PB)

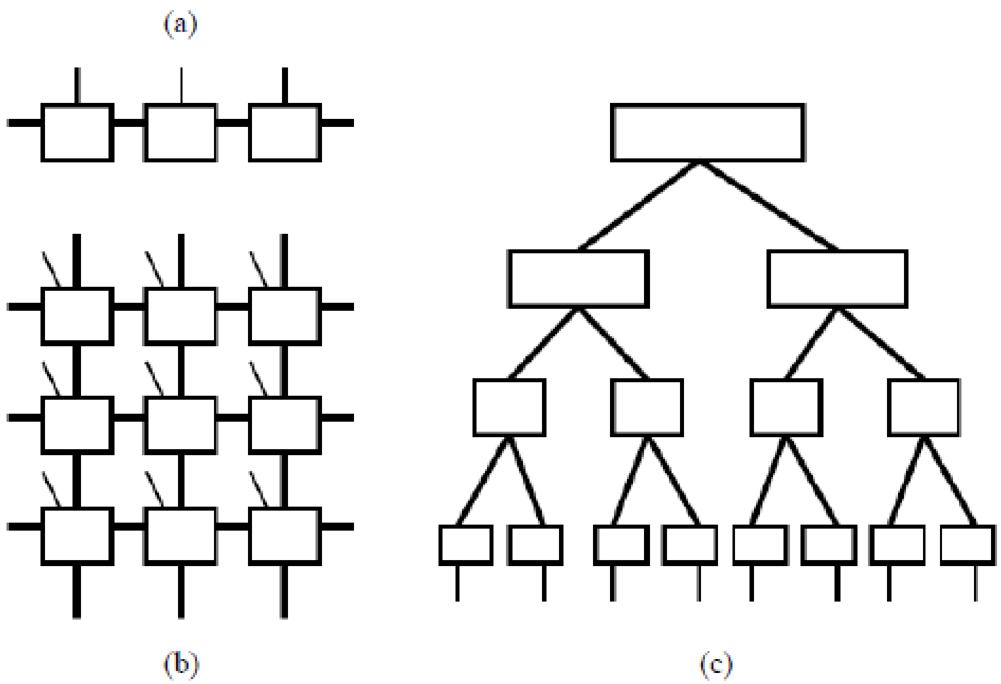
Information compression can be done in several ways

Tensor networks: very much an ongoing endeavor

Miles Stoudenmire (Flatiron Inst.)

ITENSOR

MPS/ DMRG



TPS /PEPS

TTN

Other networks/related methods

MERA: Vidal PRL' 08

GTNS: Z. Gu, X.G. Wen (2010)

CPS: HJC, Kinder, Umrigar, Chan, PRB (2009); Marti et al, (2010) Mezzacapo et al (2009)

PESS: T. Xiang et al. (2013)

For a nice review:

Verstraete, Cirac, Murg (2009) arXiv 0907.2796

Impact of this field has been far reaching (S. White, PRL milestone paper 1992)

- Ads-CFT and MERA
- Quantum entanglement
- Quantum chemistry
- Quantum information
- Neural networks/machine learning
- Many-body localization

Machine learning/artificial neural networks and quantum many-body physics

RESEARCH ARTICLE

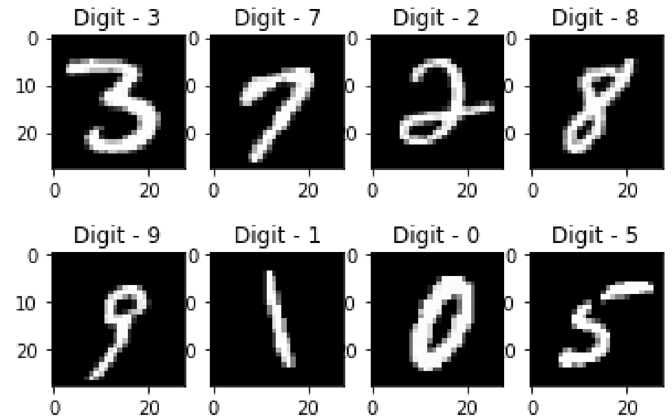
MANY-BODY PHYSICS

Solving the quantum many-body problem with artificial neural networks

Giuseppe Carleo[1]* and Matthias Troyer[1,2]

The challenge posed by the many-body problem in quantum physics originates from the difficulty of describing the nontrivial correlations encoded in the exponential complexity of the many-body wave function. Here we demonstrate that systematic machine learning of the wave function can reduce this complexity to a tractable computational form for some notable cases of physical interest. We introduce a variational representation of quantum states based on artificial neural networks with a variable number of hidden neurons. A reinforcement-learning scheme we demonstrate is capable of both finding the ground state and describing the unitary time evolution of complex interacting quantum systems. Our approach achieves high accuracy in describing prototypical interacting spins models in one and two dimensions.

$$\Psi_M(\mathcal{S};\mathcal{W}) = \sum_{\{h_i\}} e^{\sum_j a_j \sigma_j^z + \sum_i b_i h_i + \sum_{ij} W_{ij} h_i \sigma_j^z}$$



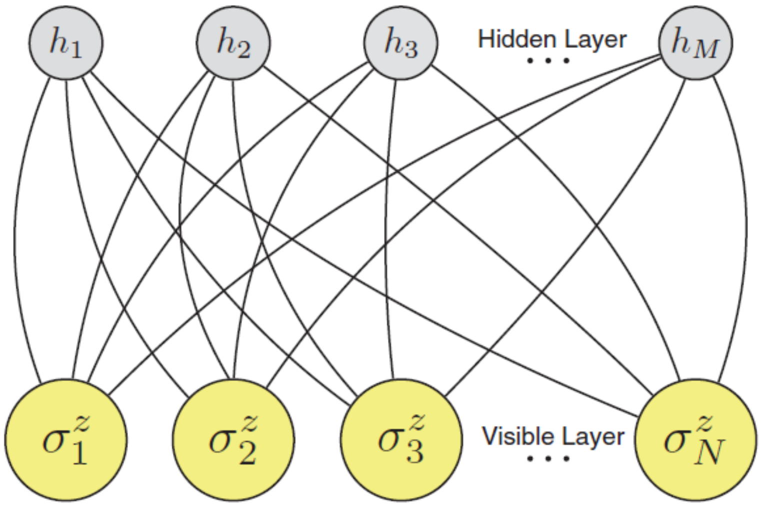
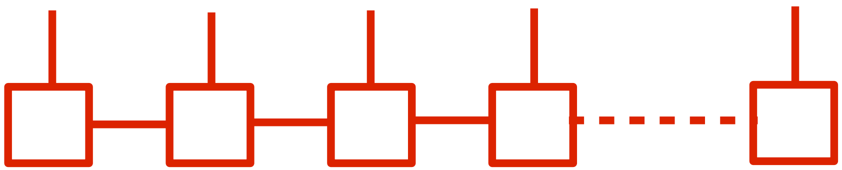


Fig. 1. Artificial neural network encoding a many-body quantum state of *N* spins. A restricted Boltzmann machine architecture that features a set of *N* visible artificial neurons (yellow dots) and a set of *M* hidden neurons (gray dots) is shown. For each value of the many-body spin configuration $\mathcal{S} = (\sigma_1^z, \sigma_2^z, \ldots, \sigma_N^z)$, the artificial neural network computes the value of the wave function $\Psi(\mathcal{S})$.

Closely related to "correlator product states" proposed by us in 2009 (**HJC**, J. Kinder, C. Umrigar, G. K. Chan, PRB 2009)

G. Chan (Caltech)

Artificial neural networks vs matrix product states (MPS)
(cartoon not to be taken too literally...)



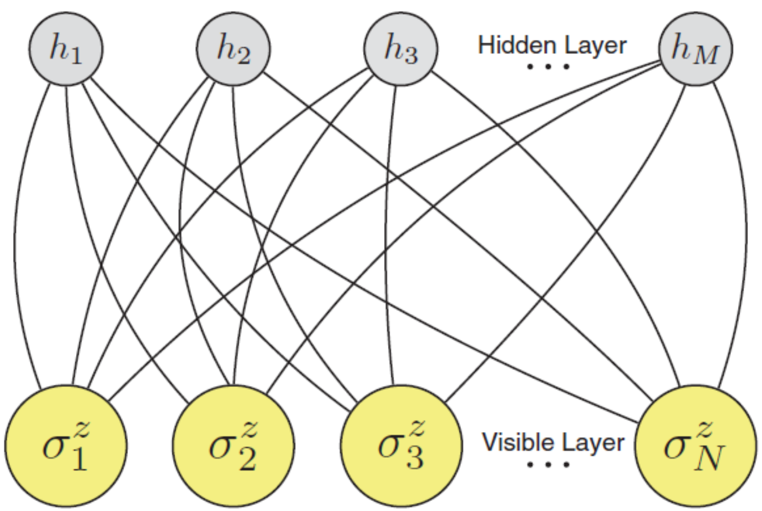
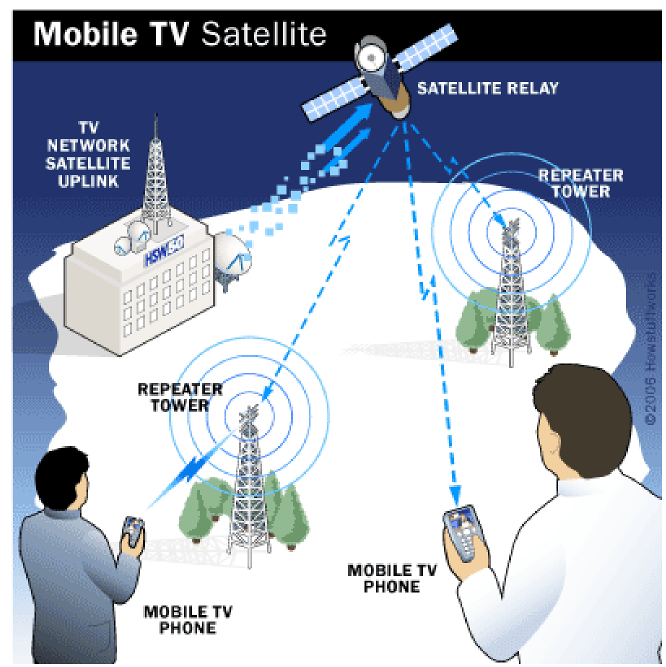


Fig. 1. Artificial neural network encoding a many-body quantum state of *N* spins. A restricted Boltzmann machine architecture that features a set of *N* visible artificial neurons (yellow dots) and a set of *M* hidden neurons (gray dots) is shown. For each value of the many-body spin configuration $\mathcal{S} = (\sigma_1^z, \sigma_2^z, \ldots, \sigma_N^z)$, the artificial neural network computes the value of the wave function $\Psi(\mathcal{S})$.

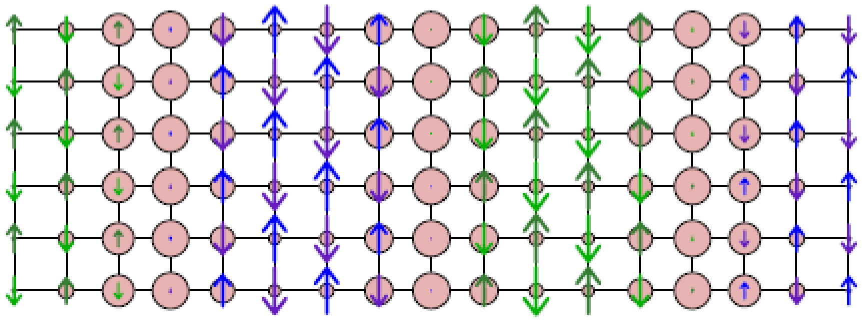


MPS/DMRG (Density Matrix Renormalization Group)

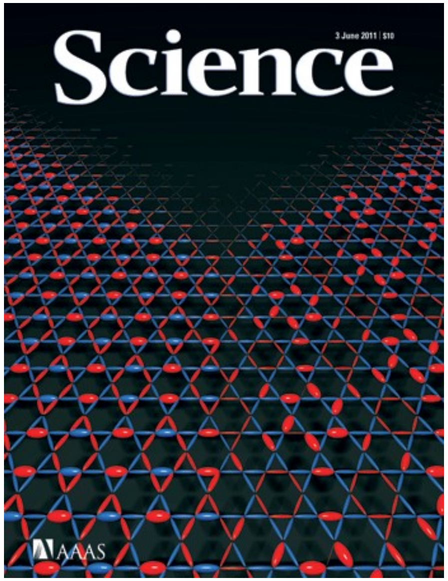
Many success stories

Few 100 – few 1000 sites/particles

Spin and charge stripes in t-J model of cuprates (still somewhat controversial)



Numerics: White, Scalapino (2003)
Expt: Tranquada Theory: Kivelson, Fradkin
Other viewpoints: Hellberg, Manousakis, Sorella



S. Yan, D. Huse, S. White (2011)

Several applications

Ground and excited states of low dimensional systems, conclusive numerical proof of Haldane gap

Time evolution of quantum systems

Quantum chemical systems (Especially polymers)

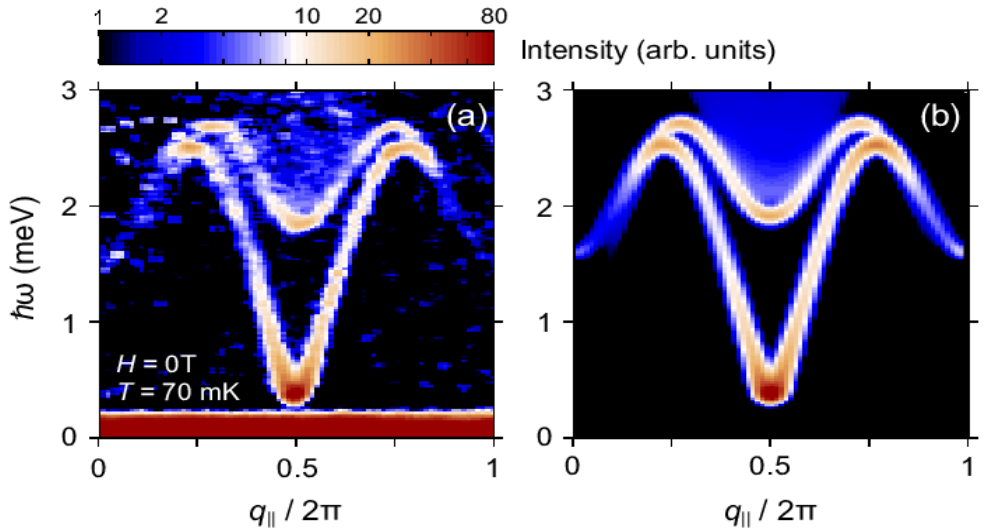
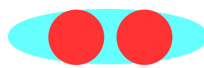
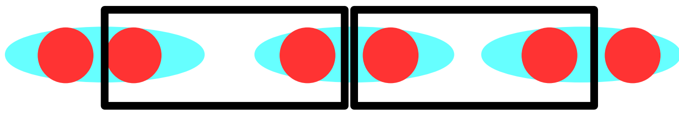


Fig. From Giamarchi, Kollath

S=1 kagome physics with DMRG

$$H_{\mathrm{Heisenberg}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$

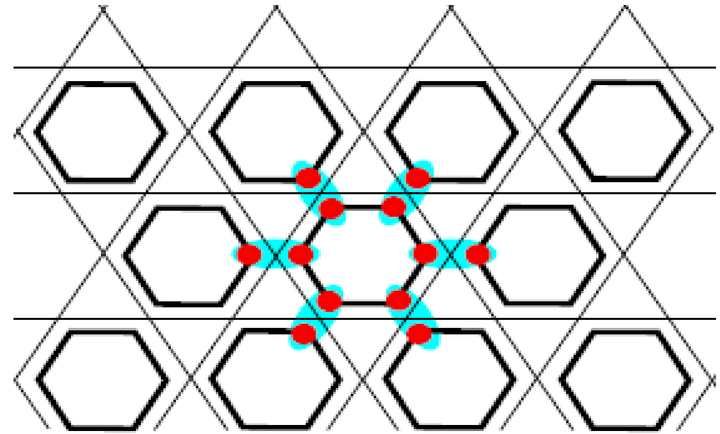
Spin-1 = 



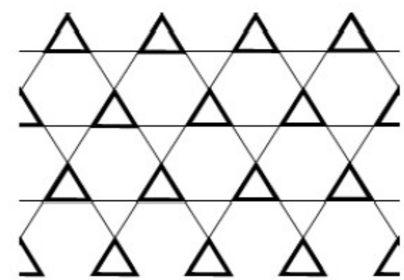
Consequence of "fractionalization"

Haldane

See Affleck, Kennedy, Lieb, Tasaki model



K. Hida, JPSJ (2000) – variationally superior



Wins!

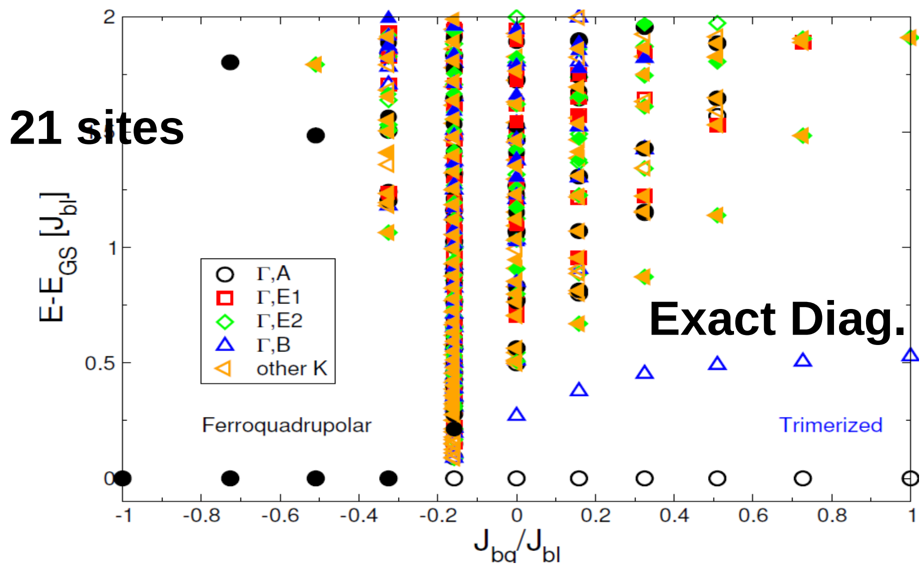
+ many other competitors
(eg. √3 x √3 within 1/S)

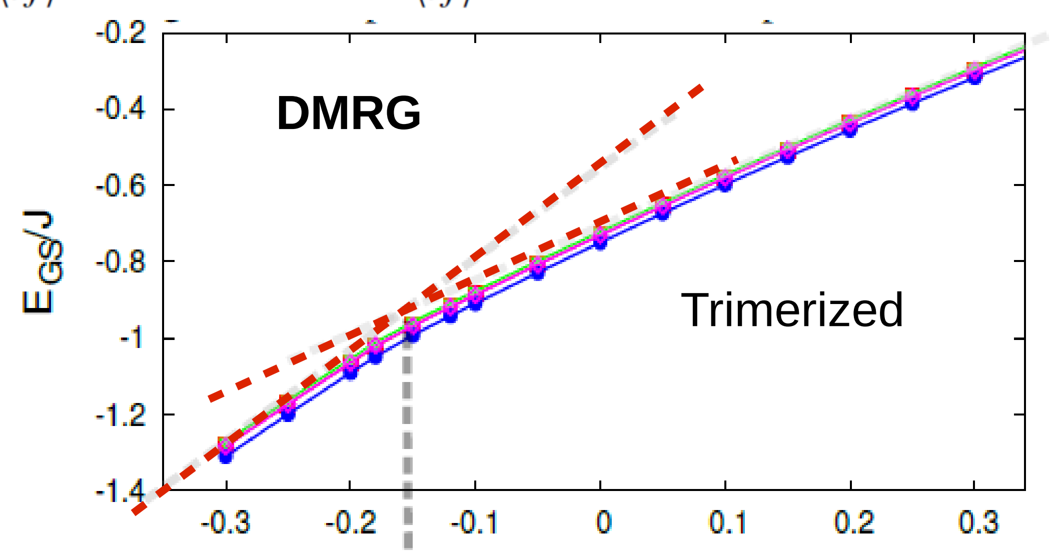
DMRG+ED: **HJC**, A. Lauchli, PRB(R) (2015)
Tensor nets: T. Liu et al. (von Delft), PRB (2015)
Series expansion: Oitmaa, Singh (2016)
Earlier related: Arovas, PRB (2008), Corboz et al., PRB (2012)
More recently for $Na_2Ti_3Cl_8$: Paul, Chung, Birol, **HJC**, PRL (2020)

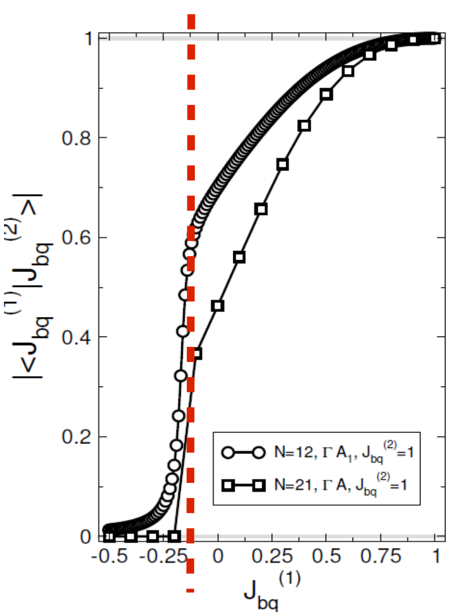
S=1 kagome: Tune biquadratic term

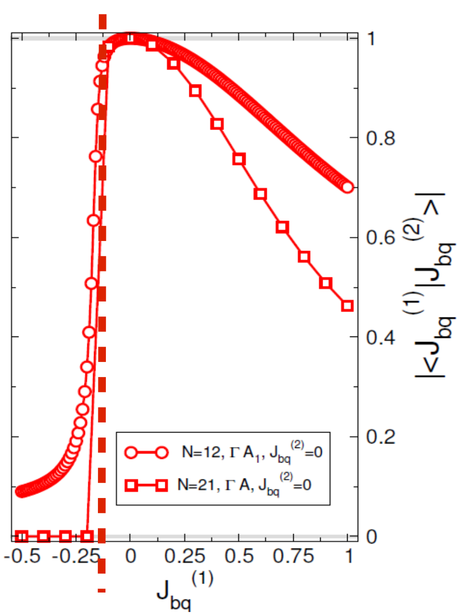
HJC, A.M. Lauchli, *PRB(R)*, 2015

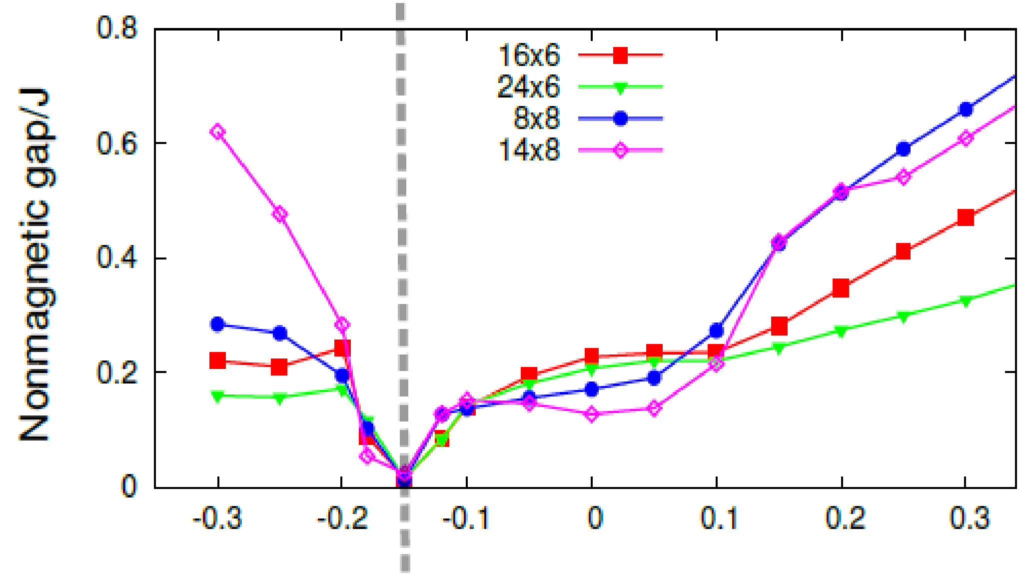
$$\mathcal{H} = J_{bl} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_{bq} \sum_{\langle ij \rangle} (\mathbf{S}_i \cdot \mathbf{S}_j)^2$$



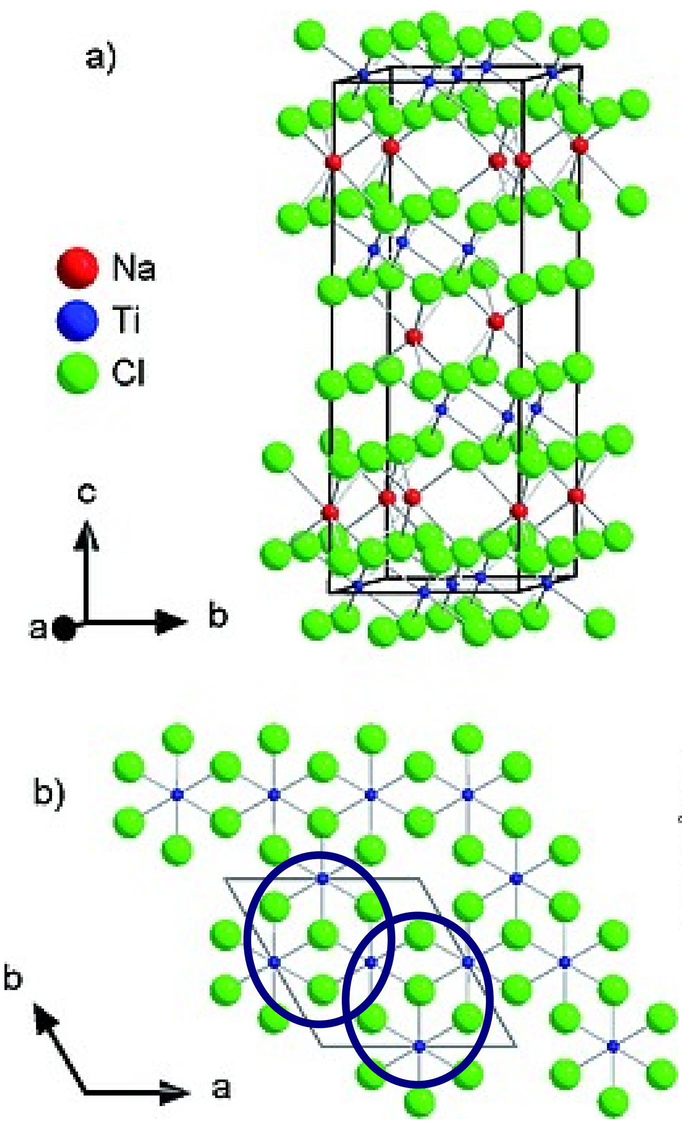






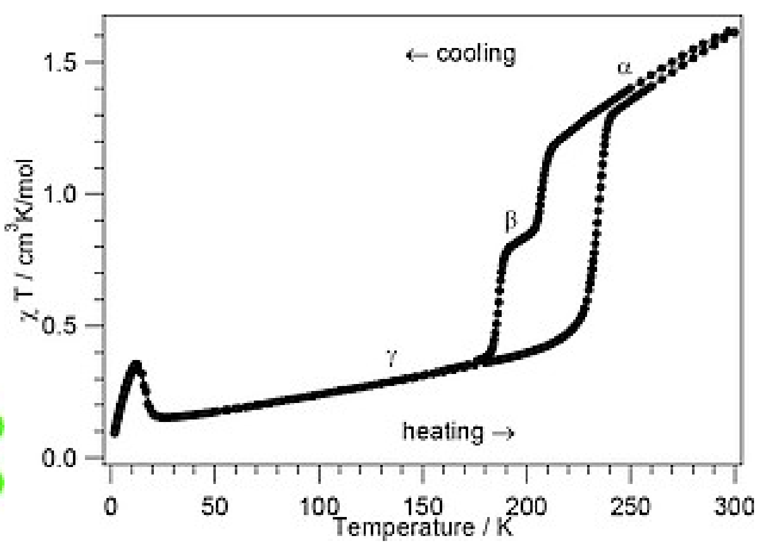


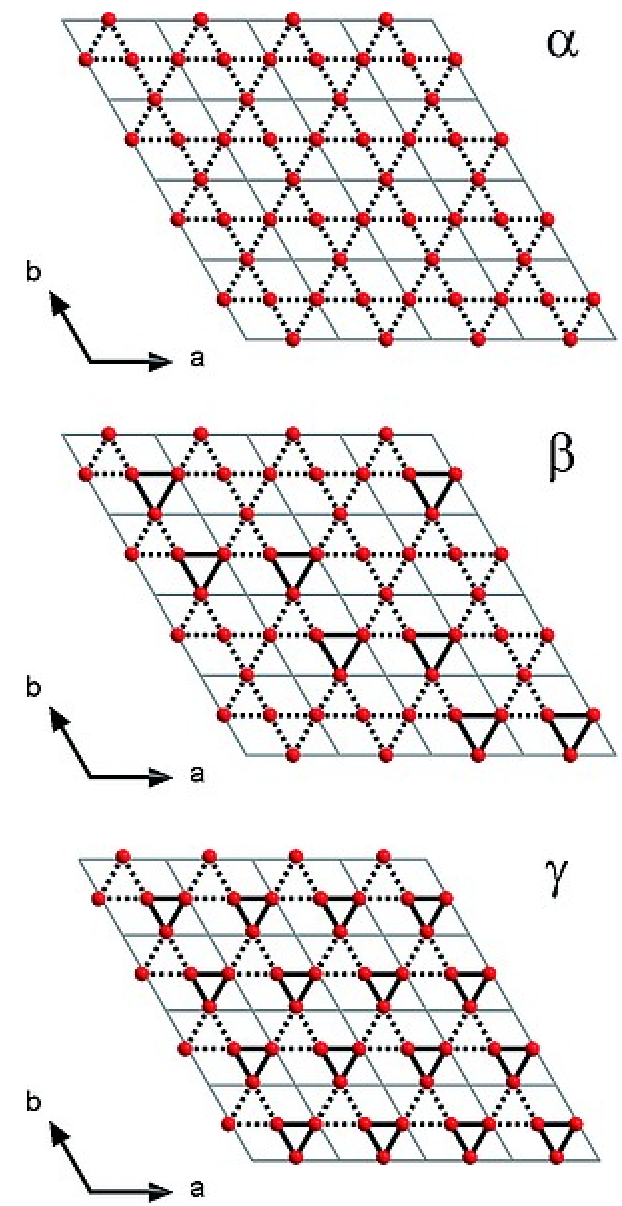
What do experiments say? (*S=1* realization $Na_2Ti_3Cl_8$)



$Ti^{2+}: 4s^0\ 3d^2$

Titanium has strong Hunds coupling, hence its two spin ½ electrons are strongly ferromagnetically coupled





Hinz et al (1995), Hanni et al (2017),
Now grown and characterized at Hopkins by McQueen group (JHU) reports spontaneous inversion symmetry breaking + **our recent work with T. Birol's group** (PRL 2020)

DMRG applications

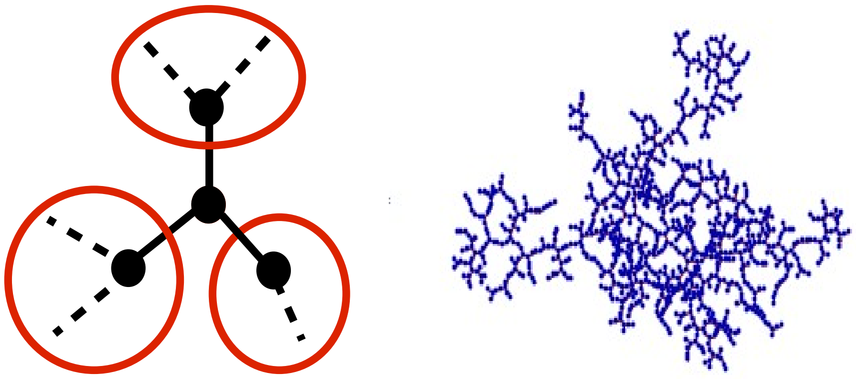
+ Some of our contributions in this rapidly growing field

Historically, many successes: Haldane gap in spin 1 chain, time evolution of 1d quantum systems (Haldane, Nobel Prize 2016)

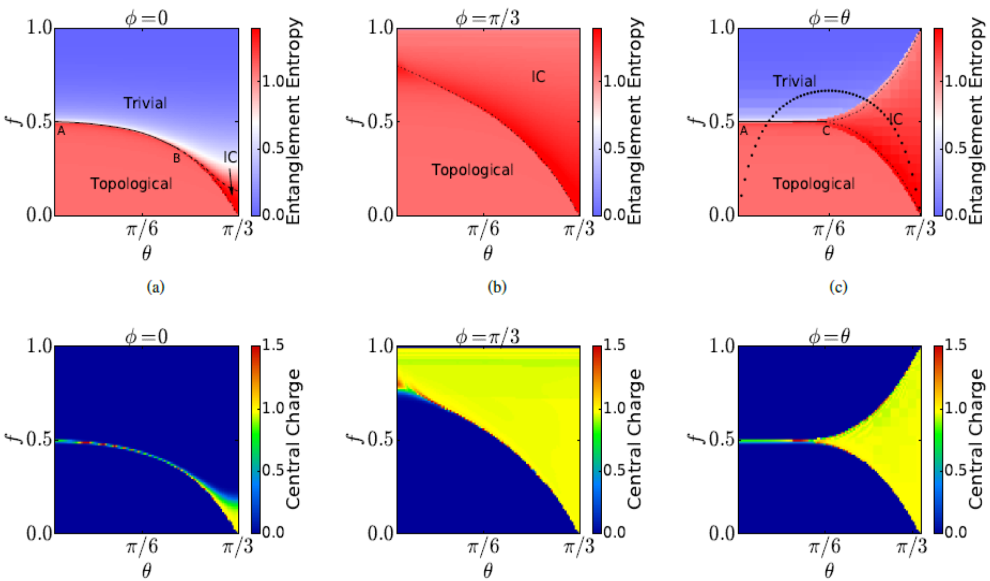
Applications to quasi 2D systems relatively recent Yan, Huse, White, Science (2011)

Practical and conceptual issues being researched: choice of geometries, excited states, thermal properties, computational expense

Dilution disordered magnets (DMRG on trees)

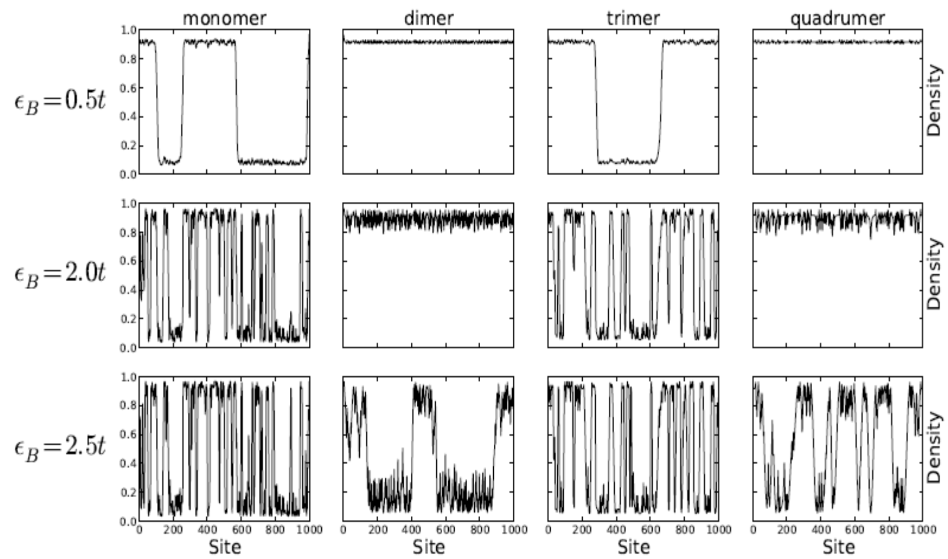


HJC, Ghosh, Pujari, Henley, *PRL* (2013)
HJC, Ghosh, Henley, Lauchli, *PRB* (2013)



Phase diagrams in 1d

Y. Zhuang, **HJC,** Tubman, Hughes *Phys. Rev. B* 2015



Interplay of disorder and interactions

HJC, Tubman, Hughes, *Scientific Reports 2016*

No theory of everything: Tremendous progress but unique limitations

	Exact	DMRG/MPS	Tensor nets	Q Monte Carlo	Variational MC	DMFT/ DMET
Lattice	any	1D, trees, 2D cylinders (HJC)	2D	any	any	any
Size (model dependent)	**Approx 40**	1000+ in 1D, 100 + in 2D **Width limited**	10 x 10 or "infinite"	Large (few 100 – few 1000)	Large	**Limited by size of local cluster**
Finite temp./excitations?	Yes finite temp experiments 3d pyrochlore HJC (2017)	**Ongoing (eg. METTS, White et al), topological degeneracies elusive**	**Formally yes, not done yet**	**Excited states difficult**	**No, but ongoing (eg. Clark, VAFT)**	Yes
Major error	None	Low entanglement, but error goes away, gapless systems difficult	Low entanglement, but error goes away, gapless systems difficult	**Sign problem or fixed node bias, error remains**	**Variational bias, Error remains but can be made accurate for certain problems**	**Form of Greens' function (DMFT), choice of mean field bath (DMET)**
Real materials ? (Hubbard model enough?)	No	Molecules White,Chan	Not yet	Yes Ceperley, Umrigar, Zhang	Yes	Yes DFT+DMFT Kotliar,Imada

Combine these
Clark, HJC (2014)

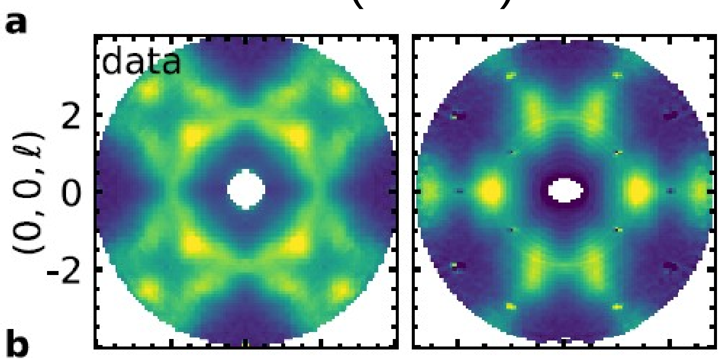
Recent efforts: theoretical formulation of map from solids to models
HJC, Zheng, Wagner et al (2015, 2017)

“Predicting” dynamics of strongly correlated magnets

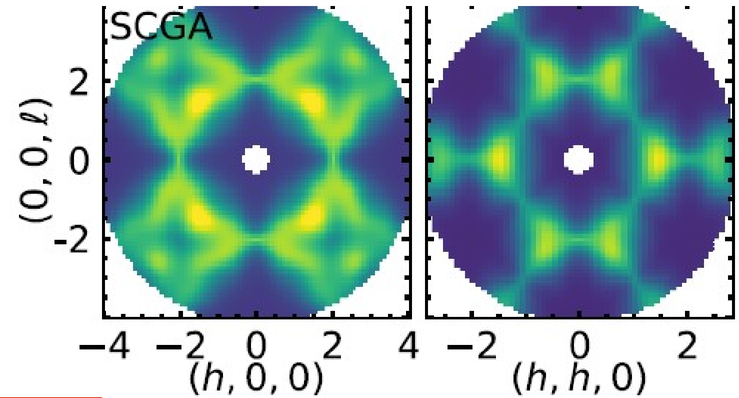
Look at lots of experiments and learn Hamiltonians: some “success” stories

Frustrated *S=1* pyrochlore $NaCaNi_2F_7$

K. Plumb (Brown)



Fit magnetic couplings to STATIC structure factor (typically 0.1 meV to 10 meV)

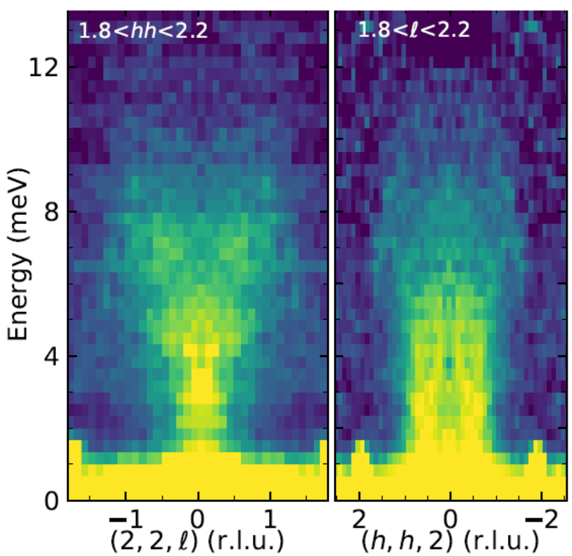


Plumb, **HJC,** et al, Nat. Phys (2019)
Zhang, **HJC**, et al, PRL (2019)

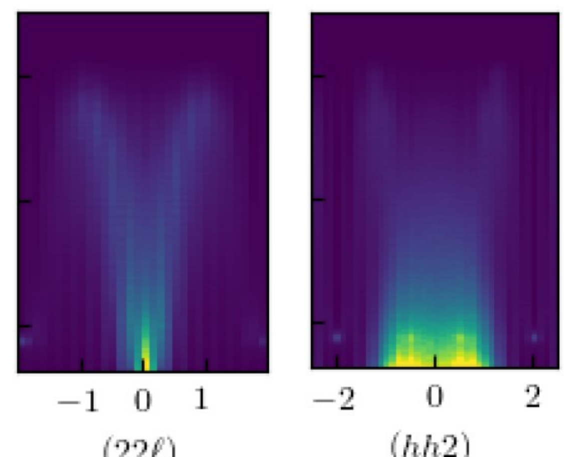
Other work by us in this area:
Scheie et al, PRL (2017), PNAS (2020)

Lattice model only of magnetic ions

Calculations with classical and semiclassical model methods (pyrochlore – 3d version of kagome)



make prediction for DYNAMIC properties (classical Monte Carlo + molecular dynamics or semiclassical spin wave theory)



Indications for a quantum spin liquid

S. Zhang, **HJC,** Plumb, Moessner, Tchernyshyov, Phys Rev. Lett. (2019)

"Predicting" dynamics of strongly correlated magnets

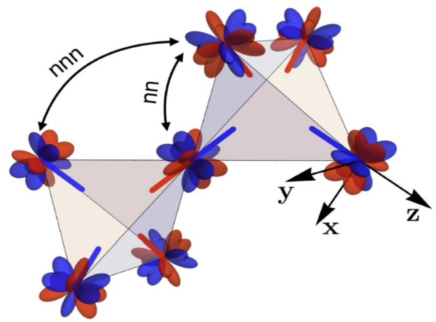
Look at lots of experiments and learn Hamiltonians: some "success" stories

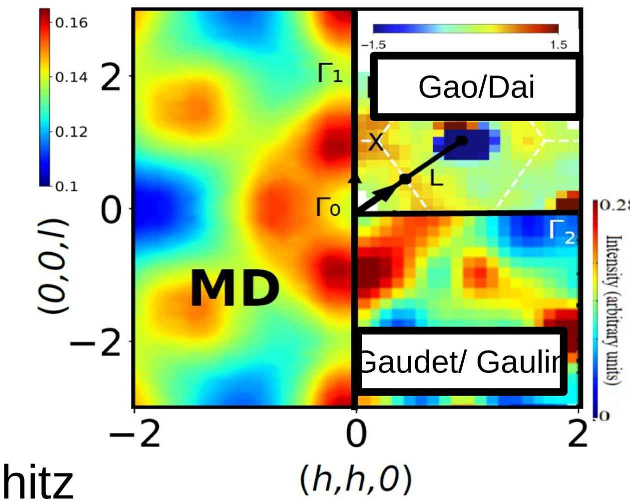
Frustrated *S=1/2* pyrochlore $Ce_2Zr_2O_7$



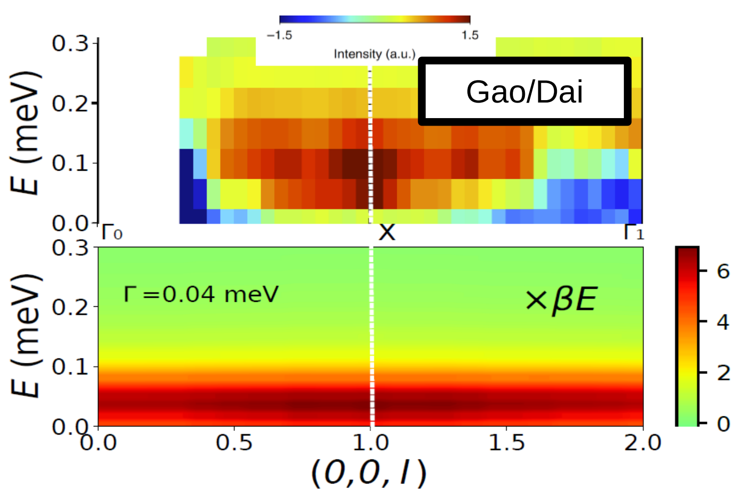


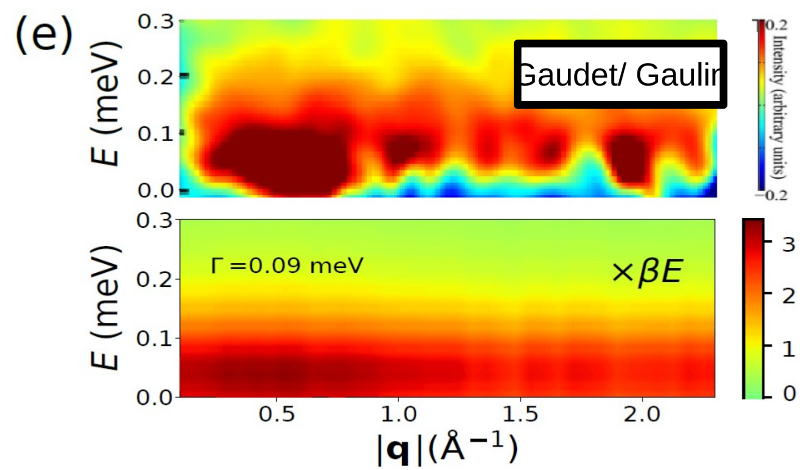
Anish Bhardwaj (FSU/NHMFL)





Using rescaled Molecular dynamics/ Landau Lifshitz





A. Bhardwaj, S. Zhang, H. Yan, R. Moessner, A. Nevidomskyy, **H.J.C.,** arXiv:2108.01096 (2021), under review

What makes such problems in magnetism challenging?

We need DFT + other methods to reliably predict properties of magnetic materials.

Data shown here is for alpha ruthenium chloride which is a Kitaev spin liquid material.

TABLE I. The spin Hamiltonians for α-$RuCl_3$ considered in this work. Dashes (–) indicate that the value is unavailable or negligible. The bolded models are considered in the main text, and results for the other models are given in the Supplemental Information. Asterisks in the 'BA' column signify that the full Hamiltonian has different values for the X/Y bonds compared with the Z bonds, and that the parameter values given in the row have been bond averaged.

	Reference	Method	J_1	K_1	Γ_1	Γ'_1	J_2	K_2	J_3	K_3	BA
1	**Winter et al. PRB** [48][a]	Ab initio (DFT + exact diag.)	−1.7	−6.7	+6.6	−0.9	–	–	+2.7	–	⋆
2	**Winter et al. NC** [28]	Ab initio-inspired (INS fit)	−0.5	−5.0	+2.5	–	–	–	+0.5	–	
3	**Wu et al.** [41]	THz spectroscopy fit	−0.35	−2.8	+2.4	–	–	–	+0.34	–	
4	Cookmeyer and Moore [53]	Magnon thermal Hall (sign)	−0.5	−5.0	+2.5	–	–	–	+0.1125	–	
5	**Kim and Kee** [47]	DFT + t/U expansion	−1.53	−6.55	+5.25	−0.95	–	–	[illegible]	[illegible]	[illegible]
6	Suzuki and Suga [54, 55]	Magnetic specific heat	−1.53	−24.4	+5.25	−0.95	–	–	[illegible]	[illegible]	[illegible]
7	**Yadav et al.** [49][b]	Quantum chemistry (MRCI)	+1.2	−5.6	+1.2	−0.7	+0.25	–	[illegible]	[illegible]	[illegible]
8	Ran et al. [27]	Spin wave fit to INS gap	–	−6.8	+9.5	–	–	–	[illegible]	[illegible]	[illegible]
9	Hou et al. [50][c]	Constrained DFT +U	−1.87	−10.7	+3.8	–	–	–	+1.27	+0.63	⋆
10	Wang et al. [51][d]	DFT + t/U expansion	−0.3	−10.9	+6.1	–	–	–	+0.03	–	
11	**Eichstaedt et al.** [45, 57][e]	Fully ab initio (DFT + cRPA + t/U)	−1.4	−14.3	+9.8	−2.23	–	−0.63	+1.0	+0.03	⋆
12	Eichstaedt et al. [45, 57][e]	Neglecting non-local Coulomb	−0.2	−4.5	+3.0	−0.73	–	−0.33	+0.7	+0.1	⋆
13	Eichstaedt et al. [45, 57][e]	Neglecting non-local SOC	−1.3	−13.3	9.4	−2.3	–	−0.67	+1.0	+0.1	⋆
14	Banerjee et al. [22]	Spin wave fit	−4.6	+7.0	–	–	–	–	–	–	
15	Kim et al. [46, 56]	DFT + t/U expansion	−12	+17	+12	–	–	–	–	–	
16	Kim and Kee[47][f]	DFT + t/U expansion	−3.5	+4.6	+6.42	−0.04	–	–	–	–	
17	Winter et al. PRB [48][g]	Ab initio (DFT + exact diag.)	−5.5	+7.6	+8.4	+0.2	–	–	+2.3	–	
18	Ozel et al. PRB [58]	Spin wave fit / THz spectroscopy	−0.95	+1.15	+3.8	–	–	–	–	–	
19	Ozel et al. PRB [58]	Spin wave fit / THz spectroscopy	+0.46	−3.50	+2.35	–	–	–	–	–	

$$\mathcal{H}_1 = \sum_{\langle ij \rangle_\gamma} \left[J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma\left(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha\right) + \Gamma'\left(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma\right)\right],$$

[a] Using the proposed minimal model, which is bond averaged and neglects small $\Gamma'_1 = -0.9$ meV. Values for the monoclinic ($C2/m$) crystal structure.
[b] We use the sign convention in Refs. [54, 56].
[c] This work gives values for several values of U. Here we use the $U = 3.5$eV parameters.
[d] Values for the C2 structure.
[e] These are the parameters from the preprint version in Ref. [57]. They were revised in the published version, Ref. [45]. In the Supplemental Information we show that this slight modification does not affect our conclusions.
[f] Case 0, corresponding to P3 structure and weaker Hund's coupling than in Model 15.
[g] Values for P3 structure.

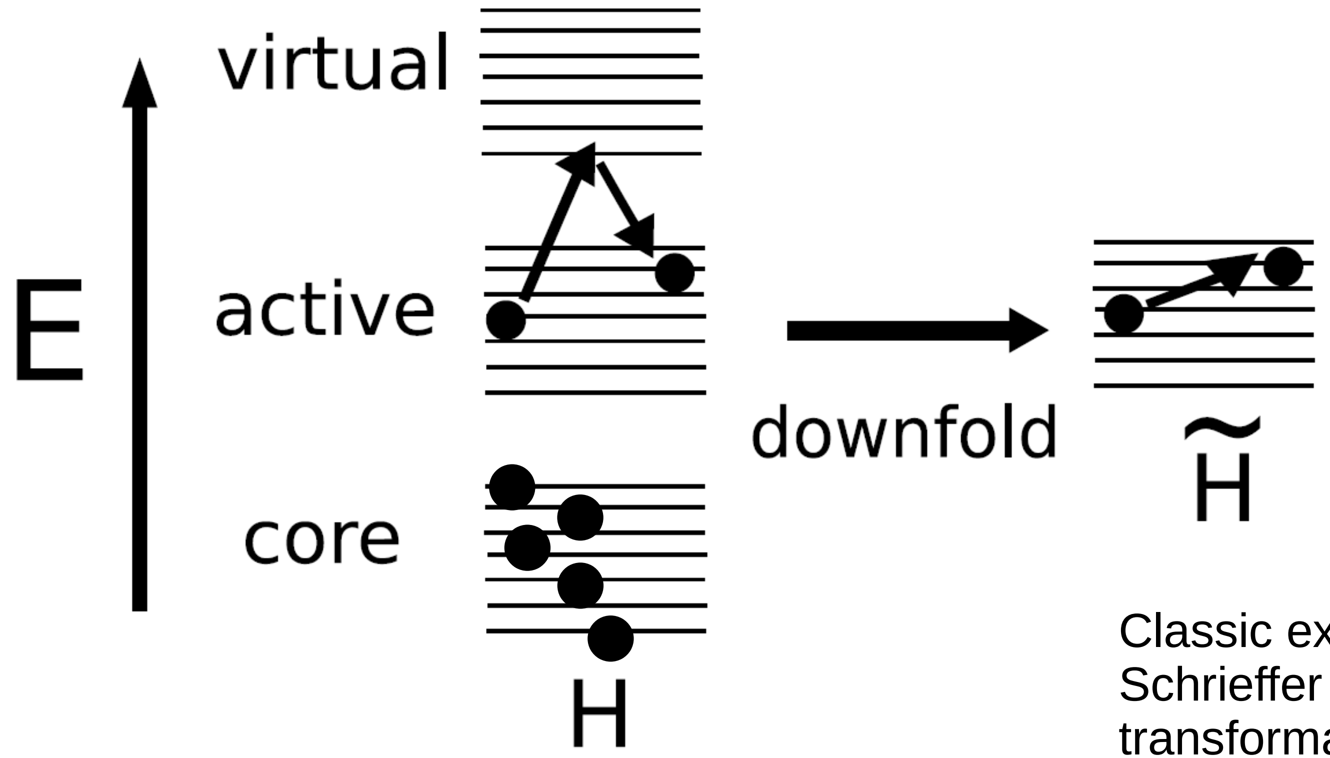
<u>No single Hamiltonian</u> describes <u>all</u> experiments (neutron scattering, specific heat etc.)

Ref. P. Laurell, S. Okamoto, NPJ (2020)
also P.Maksimov, A. Chernyshev, PRR (2020)

What exactly do we mean by "effective Hamiltonian"?

posing and offering a solution to the problem = "density matrix downfolding"

HJC, H. Zheng, L. Wagner, *J. Chem Phys* (2015)
H.Zheng*, **HJC*** et al. *Front. Phys.* (2018)



L. Wagner (UIUC)

Classic example is the Schrieffer Wolff transformation

A real material has many bands (orbitals) – s,p,d,… but the models we like to work with have fewer bands

How does one compress this information to get an effective Hamiltonian?

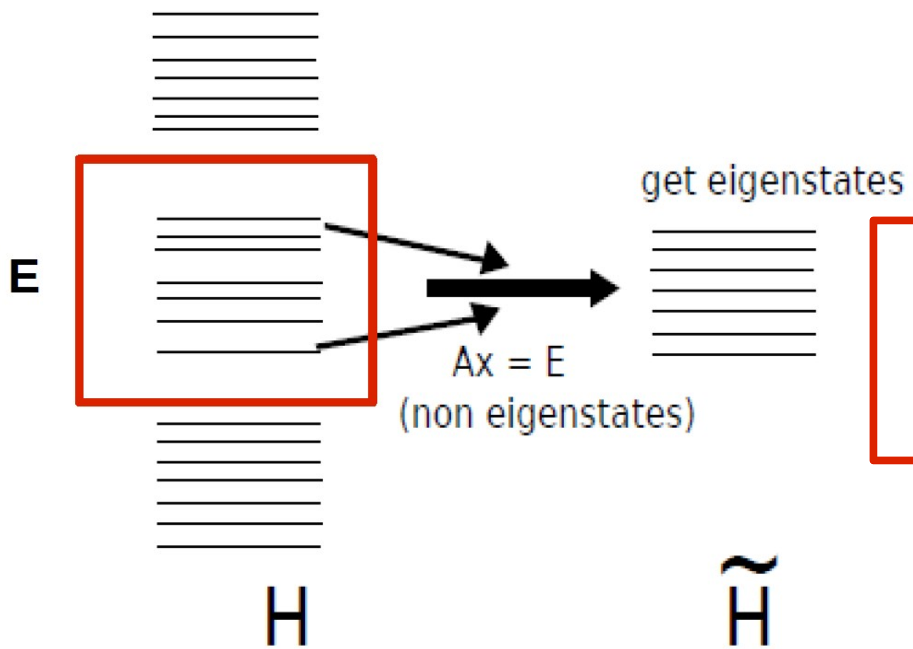
H.J.Changlani, FSU

Effective Hamiltonian is an operator compression problem

"density matrix downfolding"

HJC, H. Zheng, L. Wagner, *J. Chem Phys* (2015)
H.Zheng*, **HJC*** et al. *Front. Phys.* (2018)

Given a set of low energy wavefunctions **(not necessarily eigenstates)** how does one "learn" the effective Hamiltonian parameters



Reconstruction problem

$$\tilde{H} = C + \sum_{ij} t_{ij} c_i^\dagger c_j + \sum_{ijkl} V_{ijkl} c_i^\dagger c_j^\dagger c_l c_k$$

Finally solve

$$\tilde{H}\psi = E\psi$$

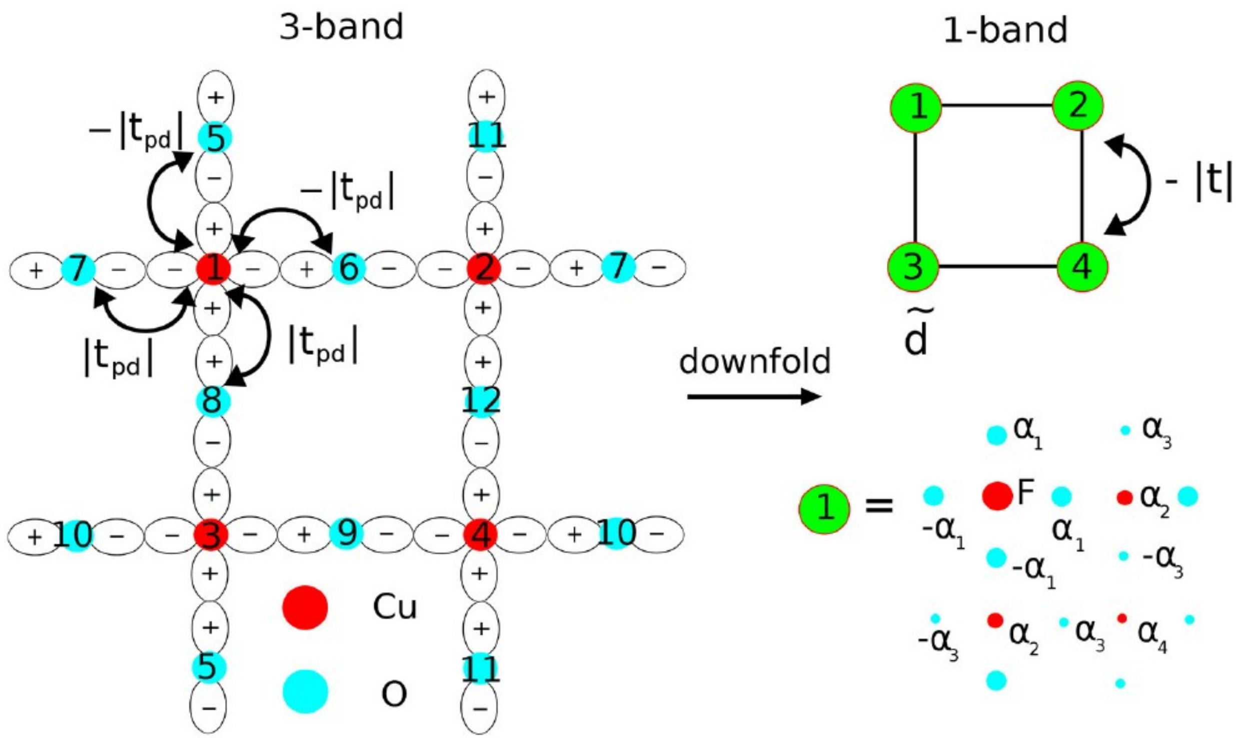
Our criterion :

$$\langle c_i^\dagger c_j \rangle_{\text{Model}} = \langle c_i^\dagger c_j \rangle_{\text{Ab-initio}}$$

$$\langle c_i^\dagger c_j^\dagger c_l c_k \rangle_{\text{Model}} = \langle c_i^\dagger c_j^\dagger c_l c_k \rangle_{\text{Ab-initio}}$$

Toy example: Three to one band model at half filling

H.Zheng*, HJC*, K. Williams, B. Busemeyer, L. K. Wagner, Front. Phys (2018)



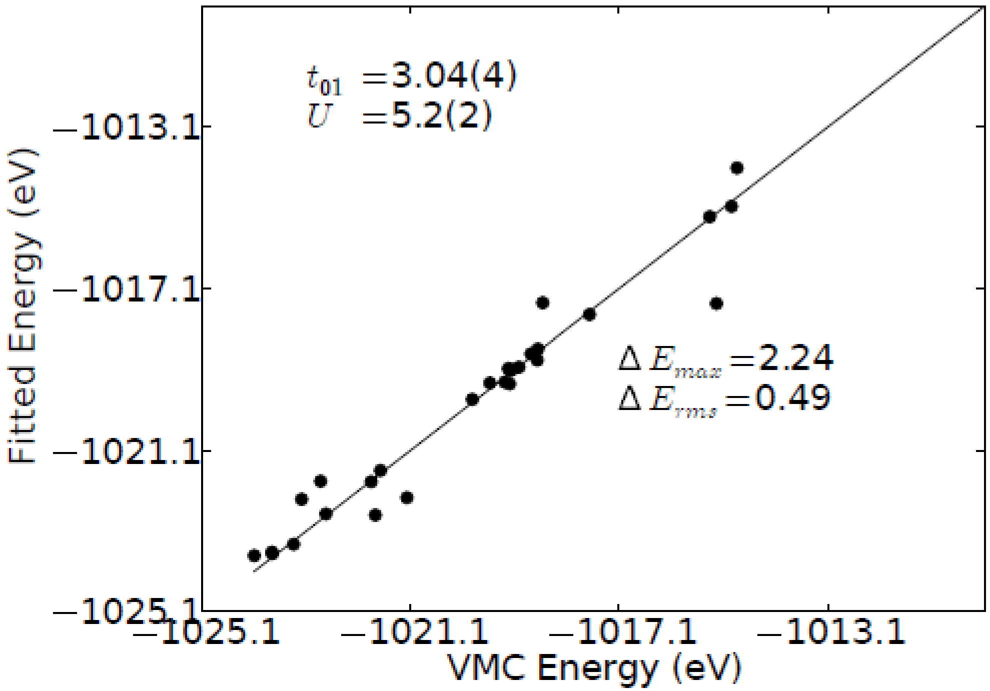
$$H = \epsilon_p \sum_{j\in p,\eta} n_{j,\eta} + \epsilon_d \sum_{i\in d,\eta} n_{i,\eta} + t_{pd} \sum_{\langle i\in d, j\in p\rangle,\eta} \mathrm{sgn}(p_i, d_j)\left(c^\dagger_{i,\eta} c_{j,\eta} + \mathrm{h.c.}\right)$$
$$+U_p \sum_{j\in p} n_{j,\uparrow} n_{j,\downarrow} + U_d \sum_{i\in d} n_{i,\uparrow} n_{i,\downarrow} + V_{pd} \sum_{\langle i\in p, j\in d\rangle} n_j n_i \, ,$$

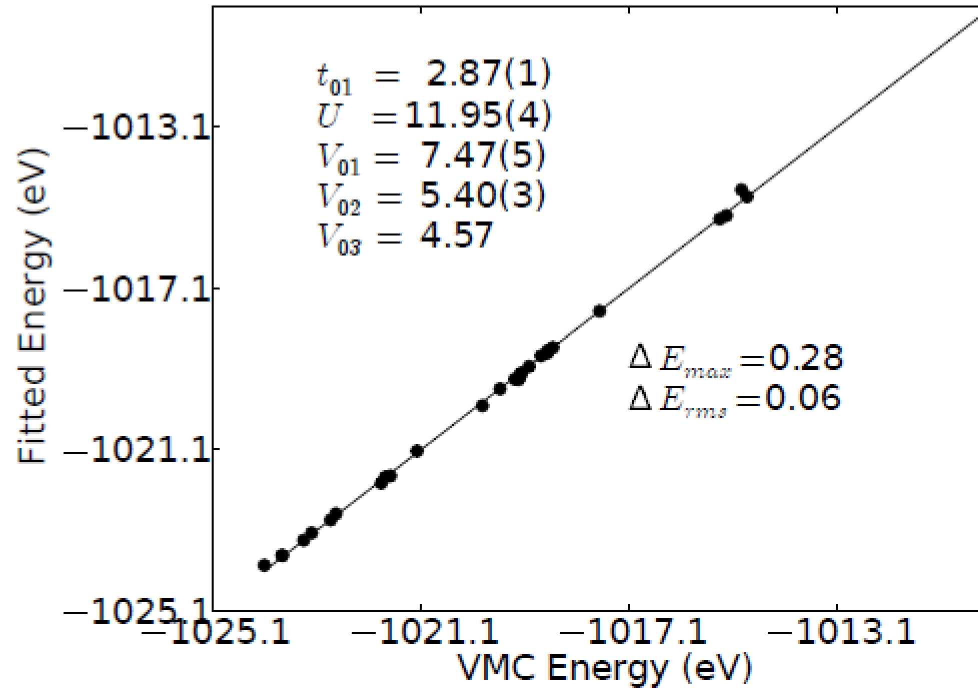
$$\Delta \equiv \epsilon_p - \epsilon_d$$

$$H = E_0 - t \sum_{\langle i,j\rangle,\eta} \tilde{d}^\dagger_{i,\eta} \tilde{d}_{j,\eta} + U \sum_i \tilde{n}^i_\uparrow \tilde{n}^i_\downarrow$$

Benzene molecule

Hubbard and extended Hubbard (PPP) model





Hubbard only

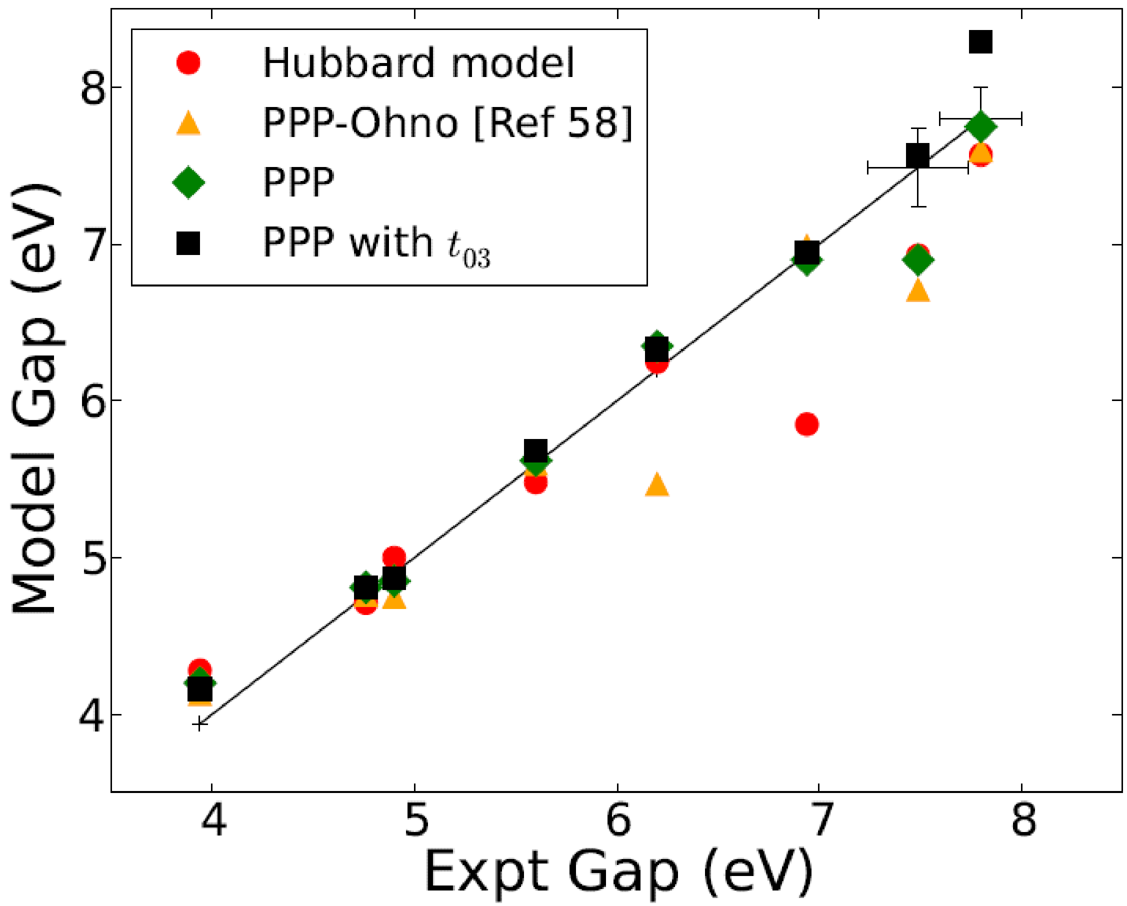
$$H = -t \sum_{ij} c^\dagger_{i,\sigma} c_{j,\sigma} + \text{h.c.} + U \sum_i n_{i,\uparrow} n_{i,\downarrow}$$

Extended Hubbard

$$H = -\sum_{ij} t_{ij} c^\dagger_{i,\sigma} c_{j,\sigma} + \text{h.c.} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} + \sum_{ij} V_{ij} n_i n_j$$

Benzene molecule

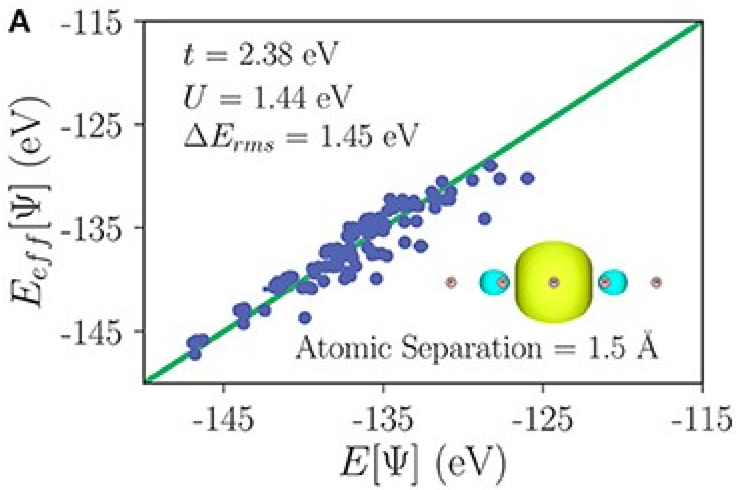
Reconstructing eigenstates by solving lattice model of 6 electrons
+ comparison to experiment



Some subtleties:
Need to calculate parameters from variational and diffusion MC, and extrapolate

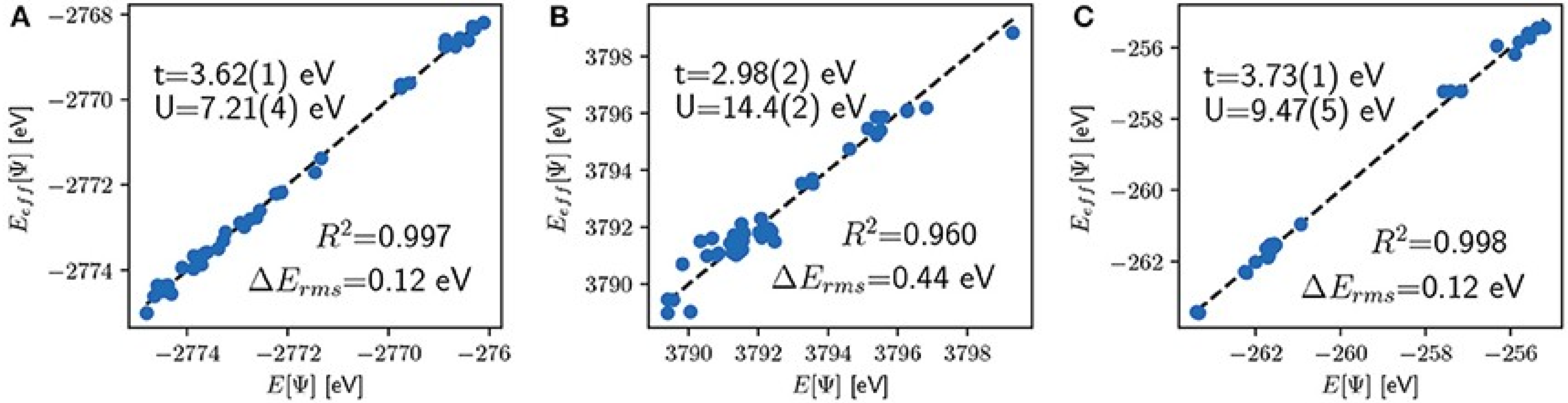
Comparable to previous semi-empirical fits + We DO NOT use experimental data!

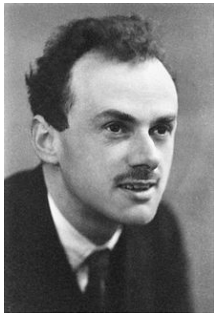
Other problems of interest



Hydrogen chain
(Kiel Williams)

Graphene
(Huihuo Zheng)





Summary

- Quantum magnetism offers a wide variety of physical phenomena especially in low dimensions and/or problems with frustration (eg. spin liquidity)

- As a community, we are closer to realizing Dirac’s vision of intelligent approximations for understanding all the beautiful effects of quantum mechanics in a many-body setting

- New improved numerical methodologies are being researched to have more predictive power. This is a challenge, and schools such as this one are much needed!

- Those studying materials and models need to talk more with one another! There may not be a “theory of everything”