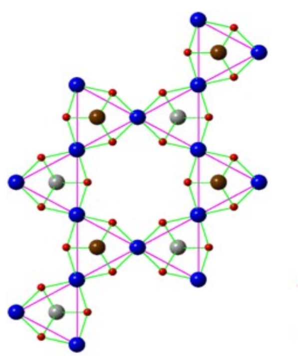
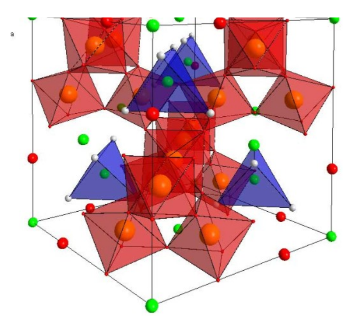
Introduction to Magnetism and Magnetic Materials – part I

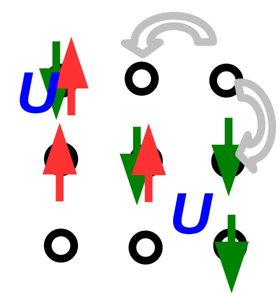
DMR - 1644779
DMR - 2046570

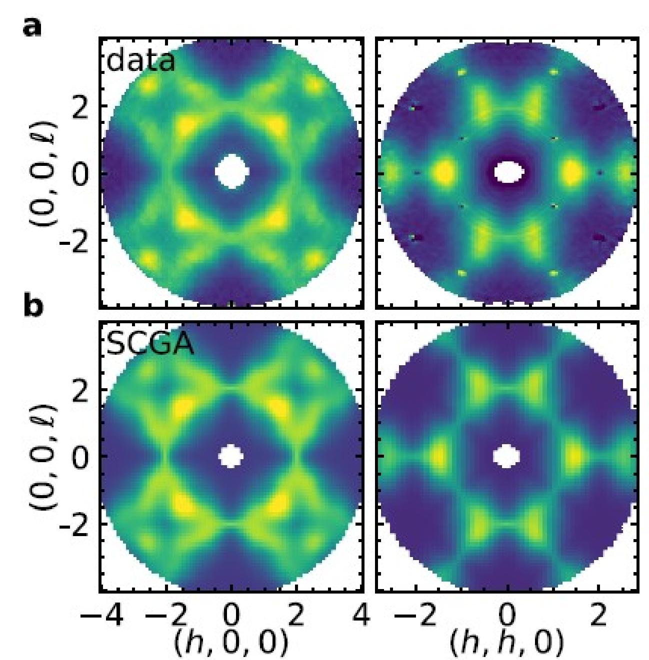
Hitesh J. Changlani
Florida State University and MagLab

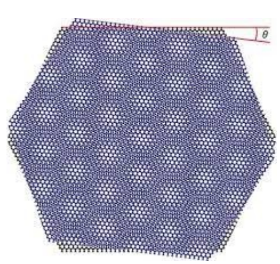
Planck computing + RCC @ FSU

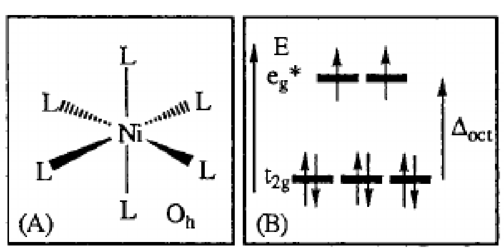


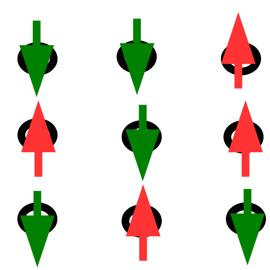








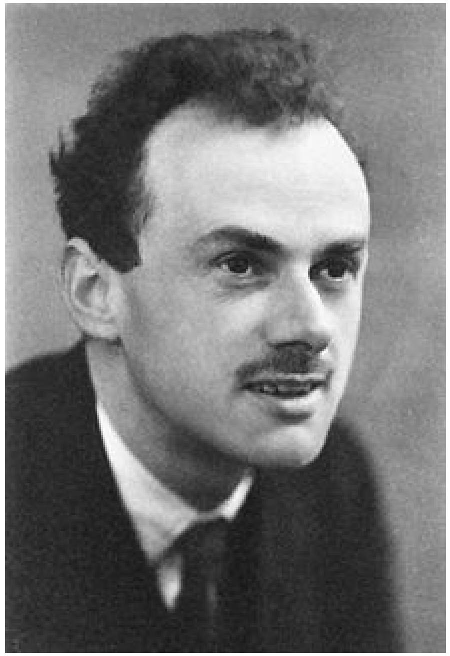




Magnetic Properties from First Principles School, Turkey, Nov. 24, 2021

Inspiration from Dirac

(1933)



Dirac as a young man

Dirac, FSU professor (1972-84), Lucasian Professor (1932-70)

“The underlying physical laws necessary for the mathematical theory of a **large part of physics** and the **whole of chemistry** are thus completely known, and the difficulty is only that the exact application of these laws leads to equations much **too complicated to be soluble**. It therefore **becomes desirable that approximate practical methods of applying quantum mechanics** should be developed, which can lead to an explanation of the main features of **complex atomic systems without too much computation.”**

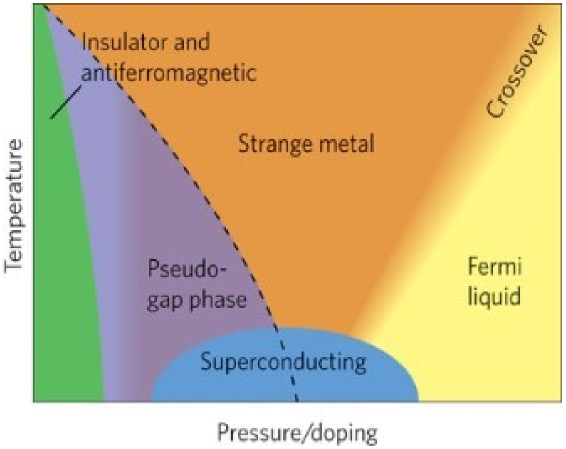
- *P.A.M. Dirac, Proceedings of the Royal Society of London. Series A, 6 April 1929*

$$H\,\psi = E\,\psi$$

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?

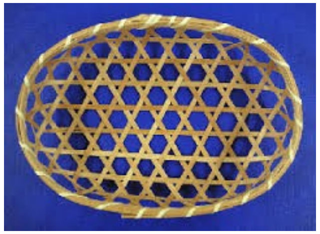
- What are some (new and old) **problems in magnetism** that we care about solving and what is the status of their solution?



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

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

"Kagome" Japanese basket



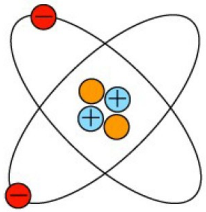
Herbertsmithite

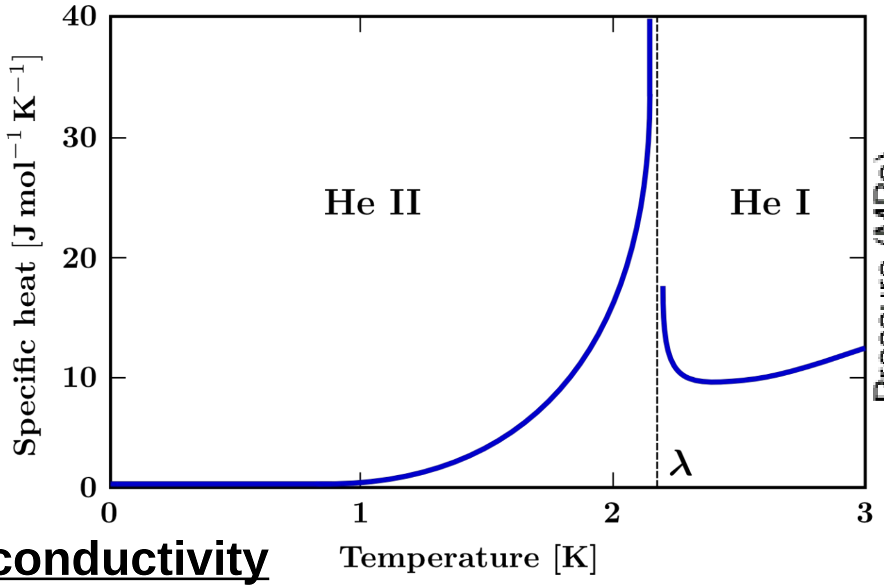
- What are some **analytic + numerical techniques** that have been useful?

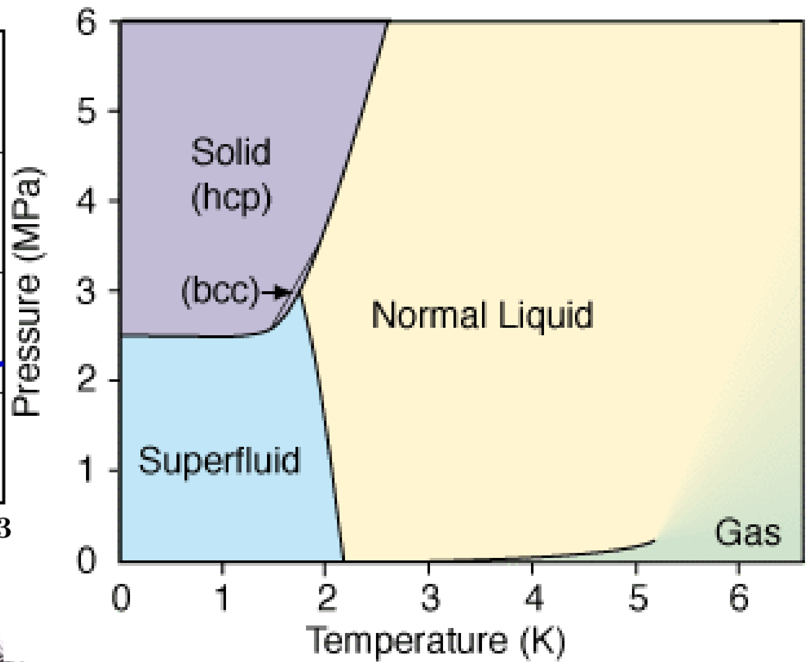
- **Where are we headed to next**? Important inputs from numerical techniques (such as density functional theory) are crucial for making connections to what is seen in experiments. The grand challenge is to develop **predictive tools** to study these systems.

H.J. Changlani, FSU

Condensed matter physics is the study of "complex atomic systems" or materials

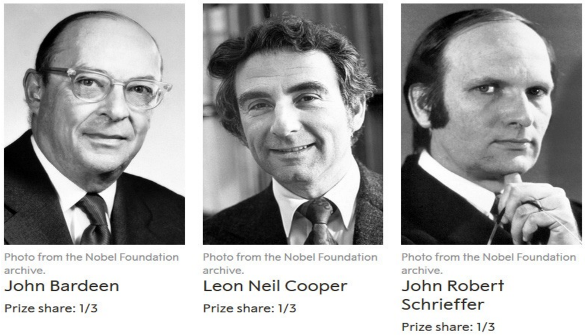


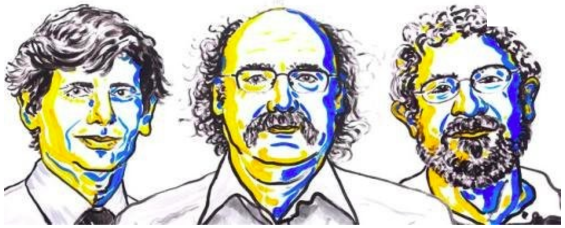




http://ltl.tkk.fi/research/theory/helium.html

<u>Superconductivity</u>





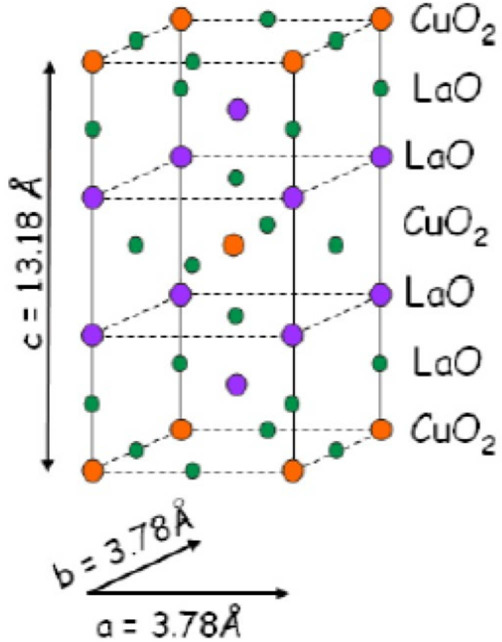
<u>Topological phase transitions</u>

Study of phases of matter and critical phenomena. Solids, liquid, gases (classical) → Superfluids, metals, insulators, superconductors…. (quantum mechanics important!)

Landau, Ginzburg, Wilson, Fisher, Widom, Domb, Bardeen, Cooper, Schrieffer, Gor'kov, Leggett. Bogoliubov, de Gennes, Anderson, Kohn, Neel, Kosterlitz, Thouless and many more...

Most phases can be characterized by some local order parameter and are now "well understood"

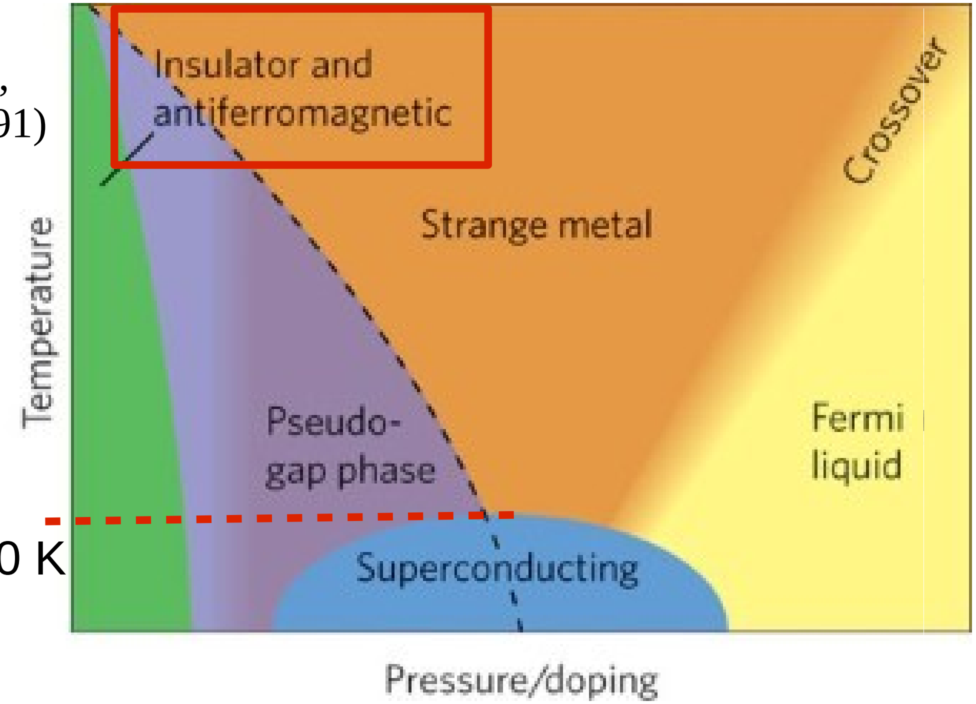
Condensed matter physics is the study of "complex atomic systems" or materials



J. Hoffman's website

Rev. Mod. Phys., Manousakis (1991)

30 – 170 K



P. Canfield, *Nature Materials* (2011)

Discovered by Bednorz, Mueller (1986) , Nobel prize 1987
Followed closely by discovery of YBCO by P. Chu in 1987
Also see work by R. Cava + coworkers (1987)

Technological applications

- Magnetic levitation (Meissner effect)
- Large scale power transmission (less heating)
- Quantum interference devices (SQUIDs)
- Quantum spin liquids for quantum computing

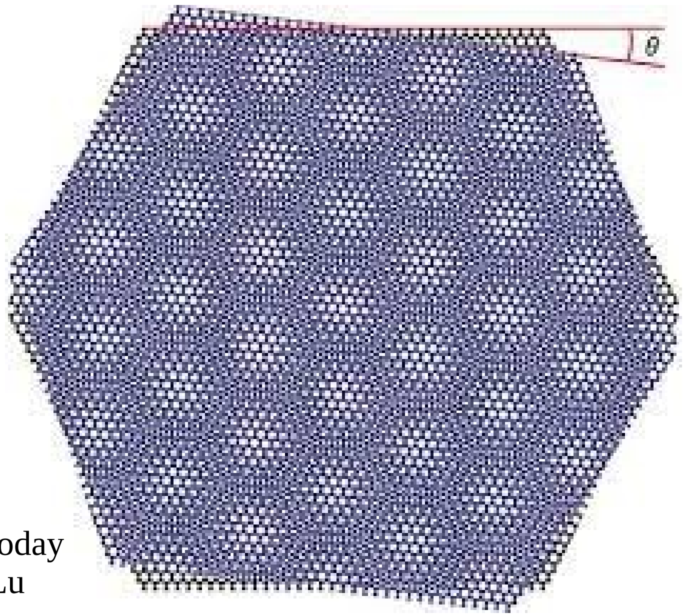
Our research: Theory of strong correlations

- Many-body treatment – band theory inadequate
- Questions in magnetism and disordered systems
- "Topological" phases (no local order parameter)
- Connections to real materials
- Numerical algorithms and development

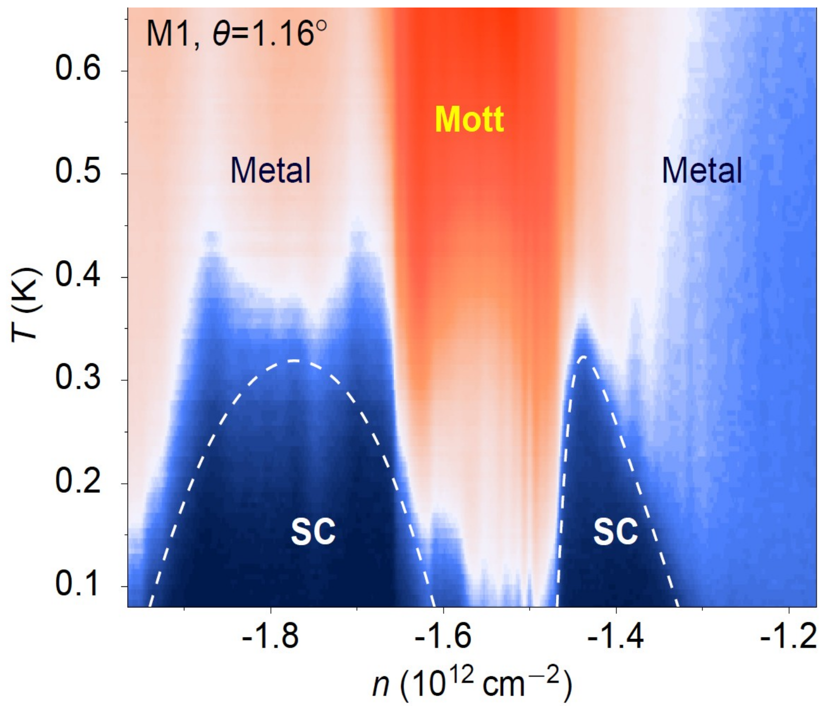
More recent 2D materials

Why do some materials (complex atomic systems) show rich phase diagrams?

Twisted graphene



H. Hill, Physics Today
Credit: ICFO/X. Lu



Y. Cao et al, Nature (2018)

Superconductivity discovered by Y. Cao et al, Nature (2018) – P. Jarillo-Herrero at MIT
Suspicions of superconductivity in this flat band system by R. Bistrizer and A. MacDonald, PNAS (2011)

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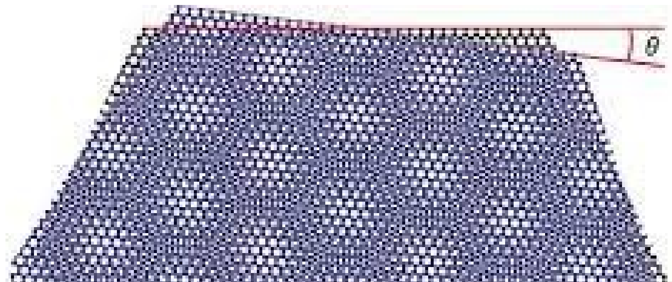
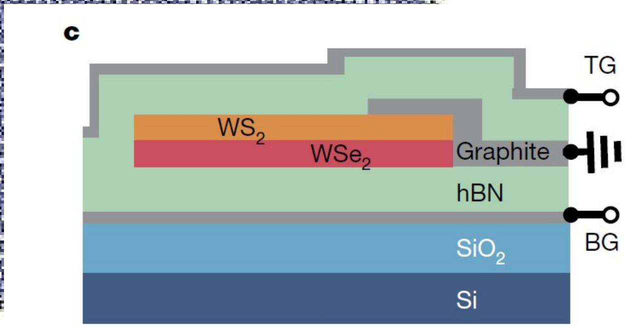
Twisted WS_2/WSe_2



H. Hill, Physics Today
Credit: ICFO/X. Lu



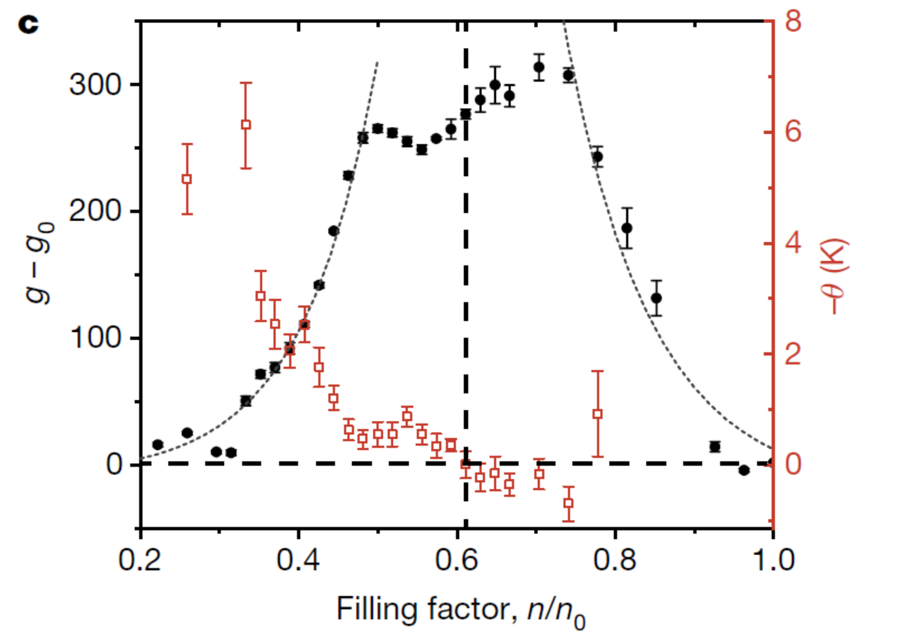


Fig. Y. Tang et al, Nature (2020)

Curie Weiss temperatures vs filling

Superconductivity discovered by Y. Cao et al, Nature (2018) – P. Jarillo-Herrero at MIT
Suspicions of superconductivity in this flat band system by R. Bistrizer and A. MacDonald, PNAS (2011)

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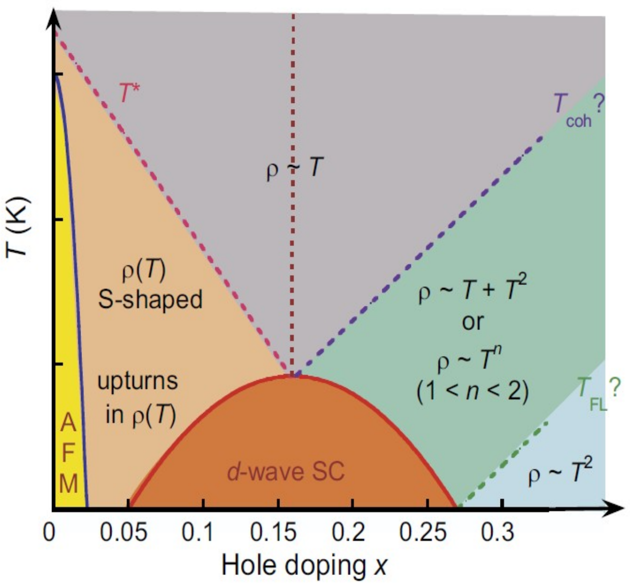
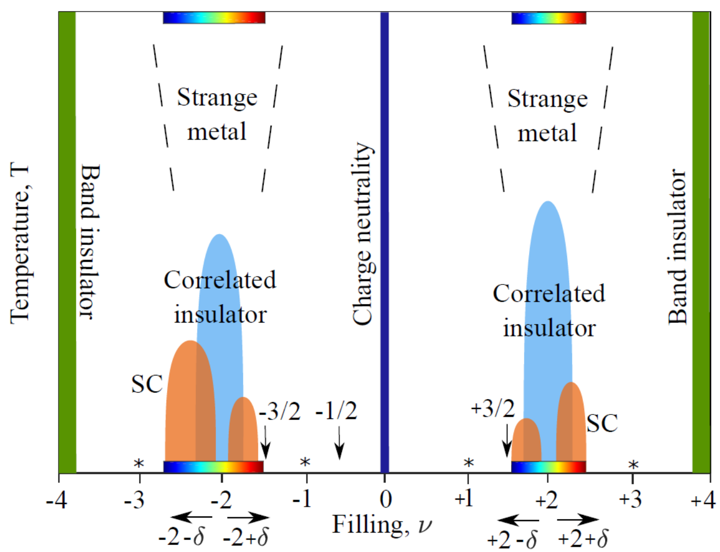
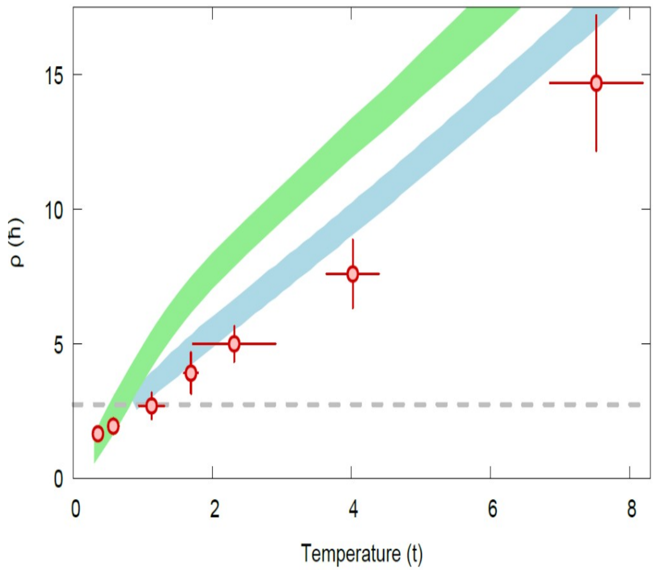
There are multiple phases that we still do not understand...

For example, linear with temperature resistivity appears to be ubiquitous in condensed matter systems (also recent cold atom systems) - "strange metal", "non Fermi liquid", "bad metal", "extremely correlated metal"….







High Tc Cuprates

fig: N. Hussey, J. Phys: Cond. Matt. 20 (2008) 12301 + many other experiments eg. MacKenzie (Dresden), Shekhter, Boebinger (NHMFL), Analytis (Berkeley), Ramshaw (Cornell)...

Twisted bilayer graphene

fig: Cao, Chowdhury, et al.,
Phys. Rev. Lett. 124, 076801 (2020)

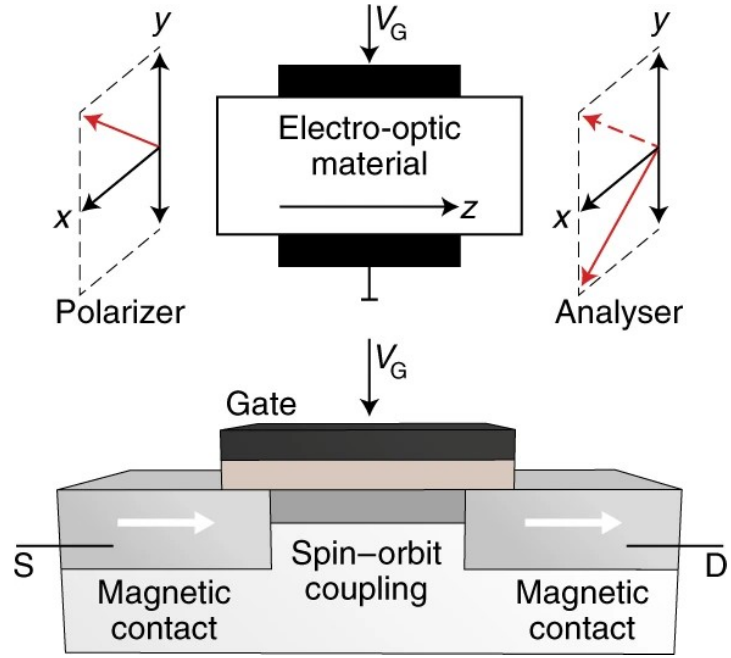
Cold atom Hubbard model

fig: P.T. Brown et al.,
Science, 363, 379-382 (2019)

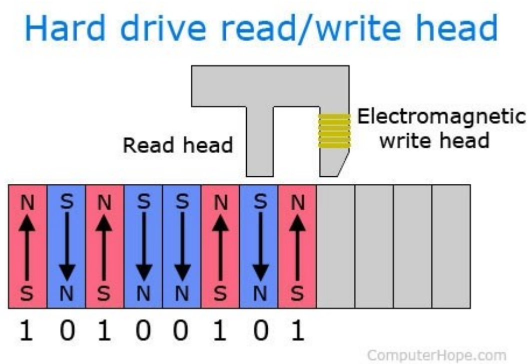
Some of our recent work: A. Patel, **HJC,** arXiv:2106.01381 (2021)

Magnetism is ubiquitous and useful

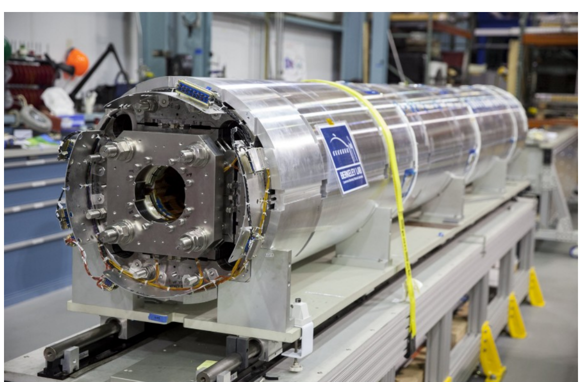
Refrigerator magnet
(amazon.com)

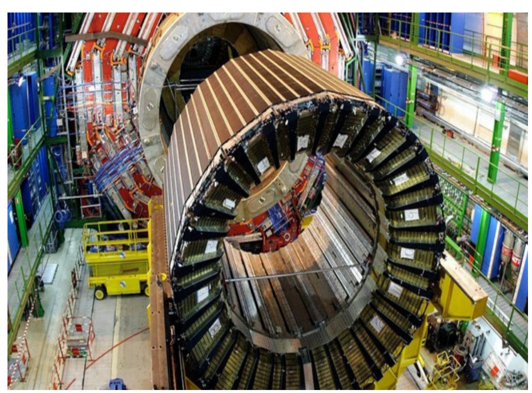


Computers!



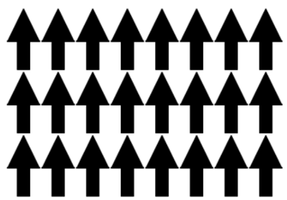
Spintronics: Datta-Das “spin transistor”
Fig: S. Datta, Nat. Electronics (2018)



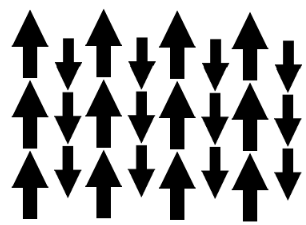


Niobium tin magnets used in the
Large Hadron Collider
(Fig: lbl.gov)

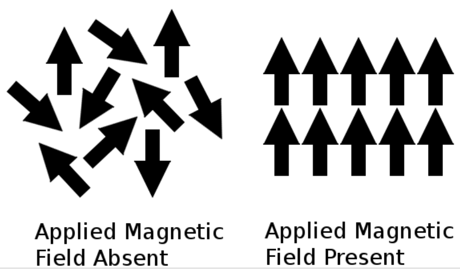
Some common forms of magnetism



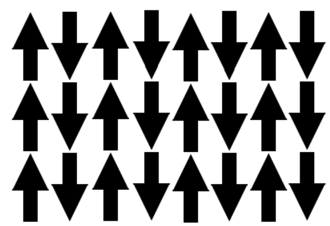
Ferromagnetism



Ferrimagnetism

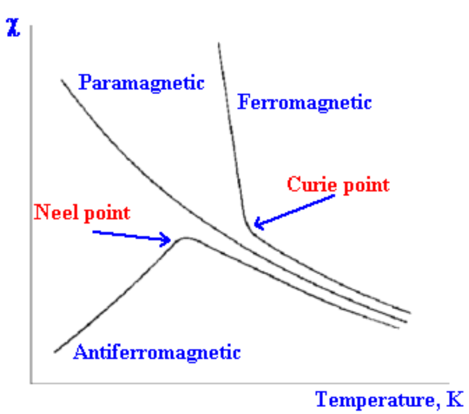


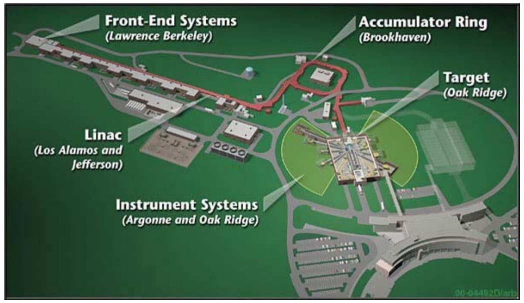
Paramagnetism

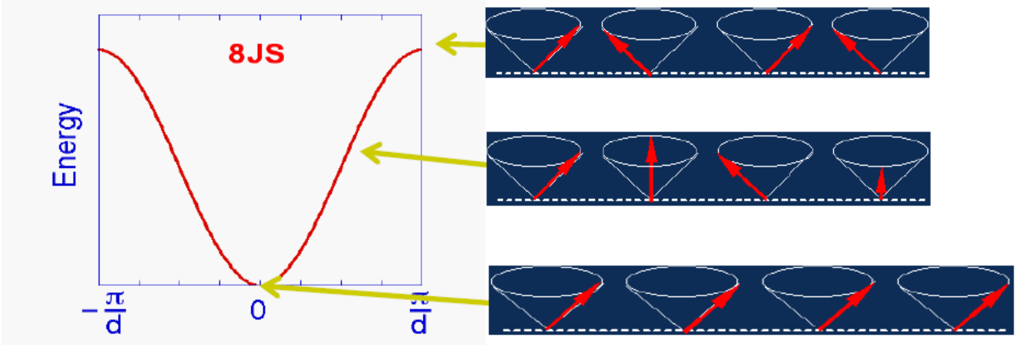


Anti Ferromagnetism

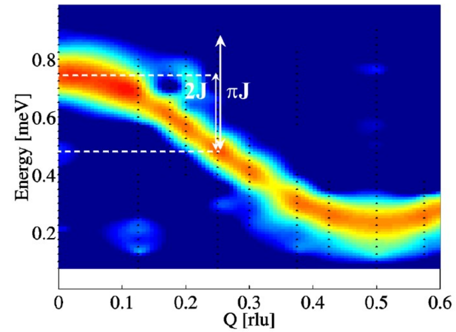
Wiki + http://electrons.wikidot.com/magnetism-iron-oxide-magnetite







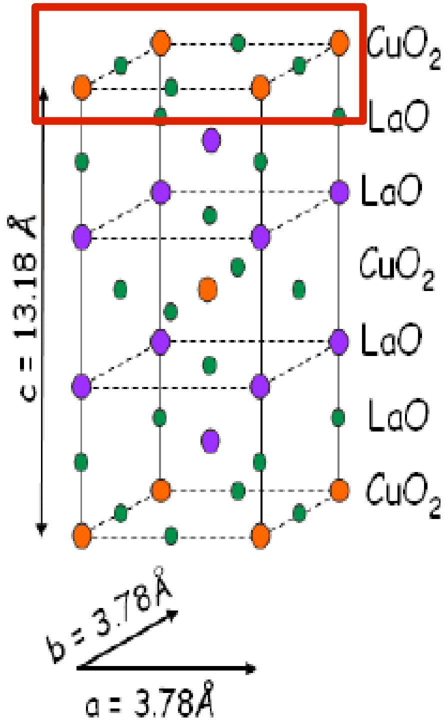
$CuSO_4 \cdot 5D_2O$



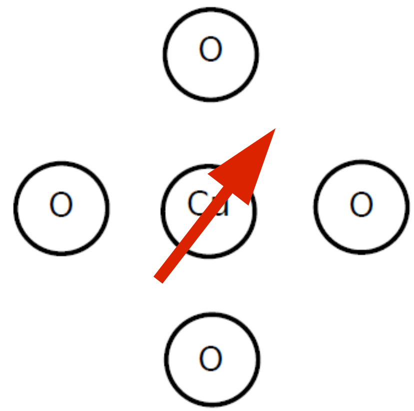
dispersion = 2SJ (1-cos(kd))

Fig: Images from H. Ronnow (EPFL)

This problem demands an unconventional approach

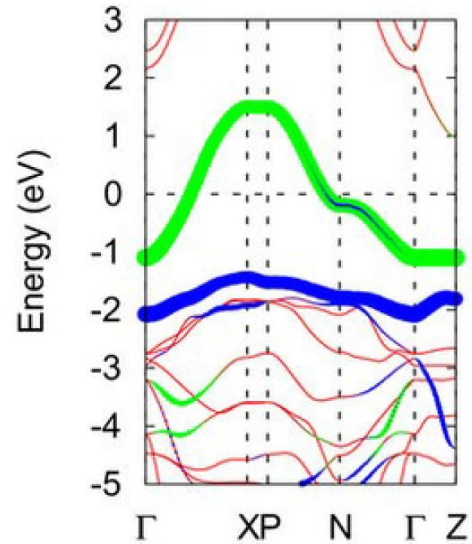


$Cu^{2+}: 4s^0 3d^9$



d electron, quite localized, not itinerant

No band gap



Jang et al, Sci. Rep (2015)

Text book band theory predicts a metallic state (1 electron per unit cell)

Yet, it is a known insulator. <u>Interactions</u> important! (N. Mott)

Note: Band theory is <u>interesting</u> in its own right, basis of field of topological insulators

Trying to simulate such systems has led to “DFT+U”, “DFT+DMFT” etc.

W. Kohn

(inventor of DFT)

$H\,\psi = E\,\psi$

P.W. Anderson

(pioneer of "lattice model approach")

Full H, approximate ψ

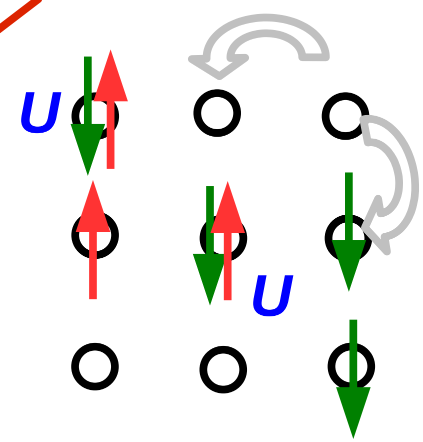
$$H = -\frac{1}{2}\sum_i \nabla_i^2 - \sum_{I,i}\frac{Z_I}{|r_I - r_i|} + \sum_{i,j,i<j}\frac{1}{|r_i - r_j|}$$

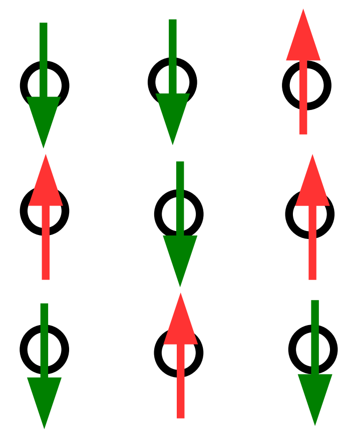
??

Approximate "effective" H

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

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





The real thing, deals with **all electrons.**

Hilbert space large, **smaller systems**.

Important physics is often not apparent

Connecting the two worlds is **not** trivial
(we invented "density matrix downfolding")

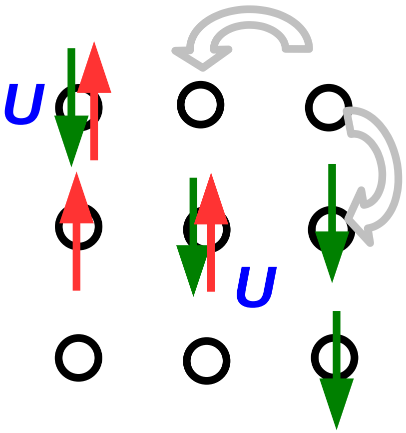
HJC, H. Zheng, L. Wagner, *J. Chem Phys* (2015)
H.Zheng*, **HJC*** et al. *Front. Phys.* (2018)

Also see work by O. Andersen, Dasgupta-Saha et al

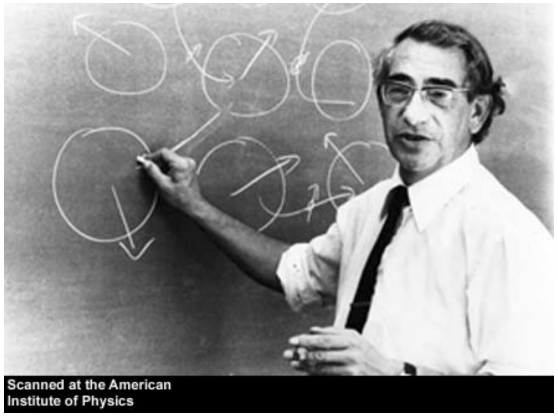
Deals with **valence electrons** only (physically insightful), eg. models of high Tc superconductivity

Smaller Hilbert space locally, more conducive for **larger scale simulations**

Hubbard model on a square lattice: a 50+ year old problem



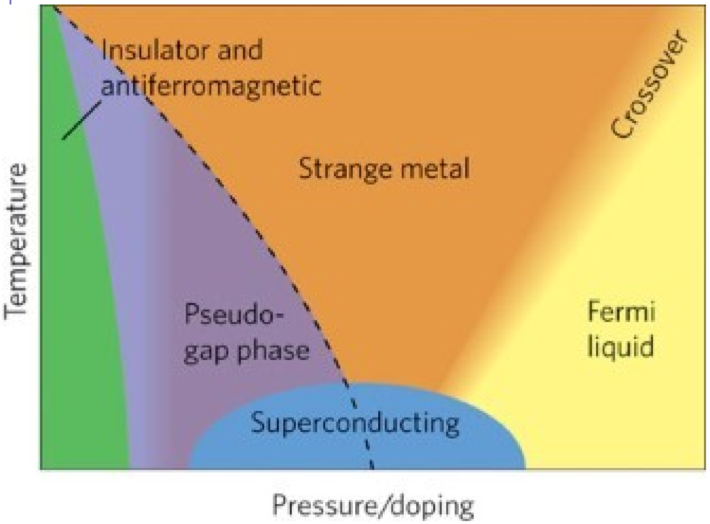
$$H_{\text{Hubbard}} = -t \sum_{\langle i,j \rangle,\sigma} c^{\dagger}_{i,\sigma} c_{j,\sigma} + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$



editorial

The Hubbard model at half a century

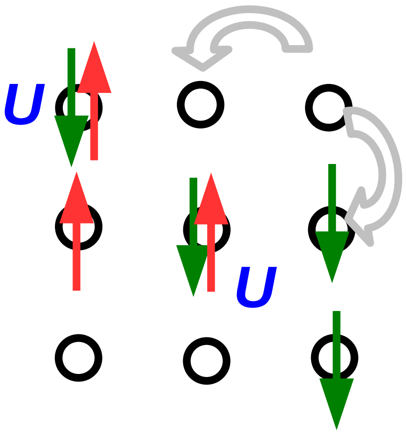
Models are abundant in virtually all branches of physics, with some achieving iconic status. The Hubbarc model, celebrating its golden jubilee this year, continues to be one of the most popular contrivances of theoretical condensed-matter physics.



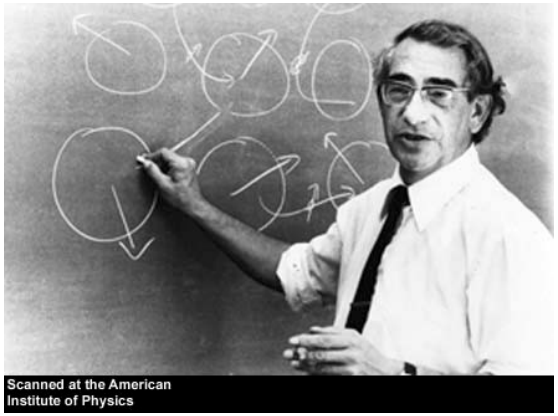
Can it describe this??

The simplicity of the Hubbard model, when written down, is deceptive.

Hubbard model on a square lattice: a 50+ year old problem



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

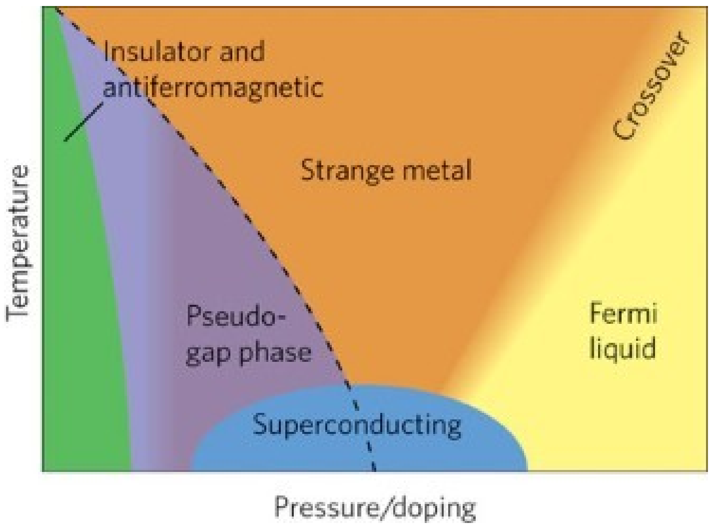


PHYSICAL REVIEW X 5, 041041 (2015)

Solutions of the Two-Dimensional Hubbard Model: Benchmarks and Results from a Wide Range of Numerical Algorithms

J. P. F. LeBlanc,[1] Andrey E. Antipov,[1] Federico Becca,[2] Ireneusz W. Bulik,[3] Garnet Kin-Lic Chan,[4] Chia-Min Chu
Youjin Deng,[6] Michel Ferrero,[7] Thomas M. Henderson,[3,8] Carlos A. Jiménez-Hoyos,[3] E. Kozik,[9] Xuan-Wen Liu
Andrew J. Millis,[10] N. V. Prokof'ev,[11,12] Mingpu Qin,[13] Gustavo E. Scuseria,[3,8] Hao Shi,[13] B. V. Svistunov,[11,1]
Luca F. Tocchio,[2] I. S. Tupitsyn,[11] Steven R. White,[5] Shiwei Zhang,[13] Bo-Xiao Zheng,[4]
Zhenyue Zhu,[5] and Emanuel Gull[1,*]

(Simons Collaboration on the Many-Electron Problem)



Recent work:
no superconductivity in the nearest neighbor Hubbard model at optimal doping!

Can it describe this??

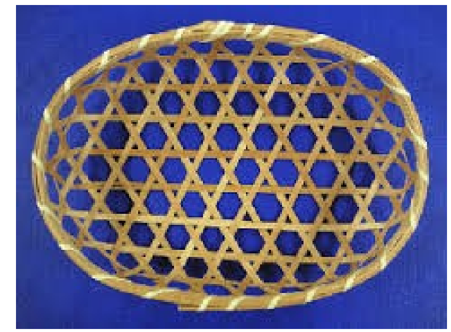
Q: So what is the minimal model which has a superconducting ground state??

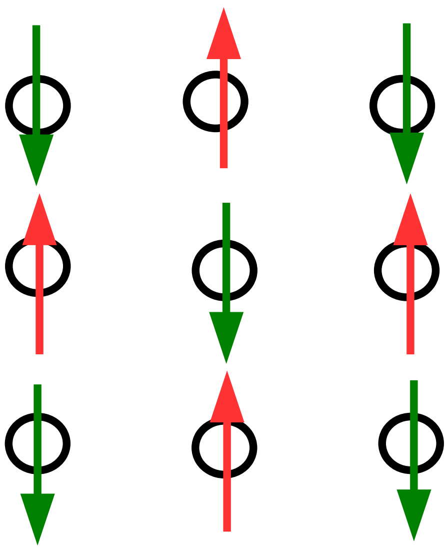
Posing one problem gives rise to many other problems

"Frustrated" Antiferromagnets which are "Mott" insulators

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

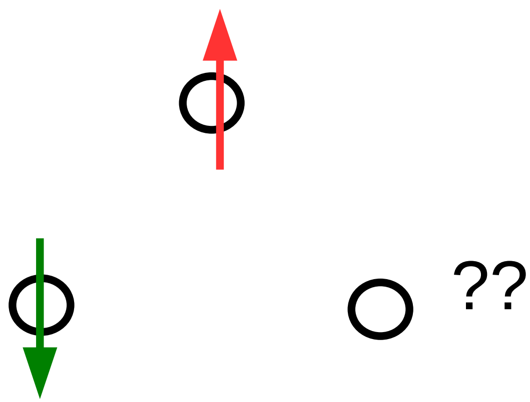
"Kagome" Japanese basket





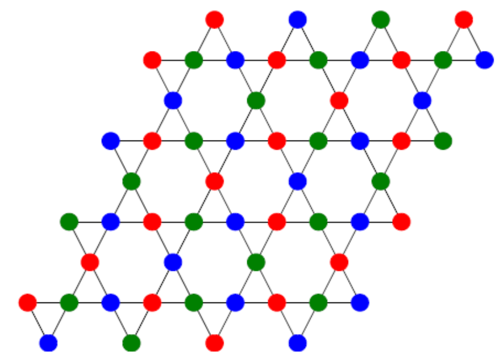
Square lattice

Un - frustrated



Triangular

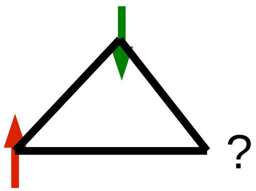
Frustrated



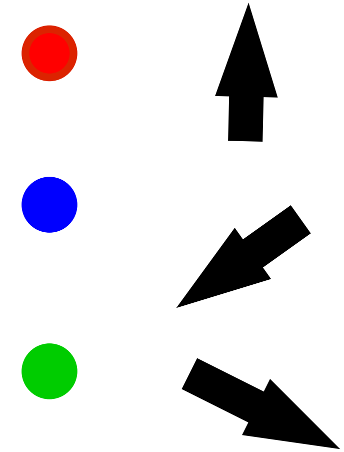
Kagome

A central question: Which state wins in energy?

Often no clear (single) winner, so one needs a **full many-body treatment**

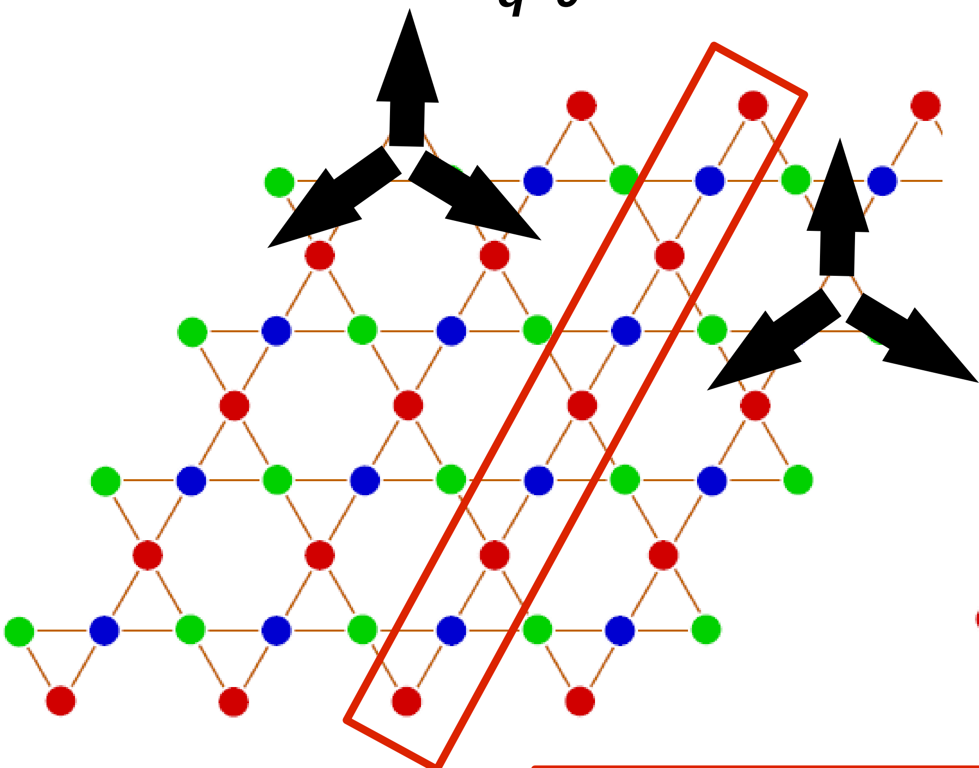


Physics of “frustrated” magnetic systems

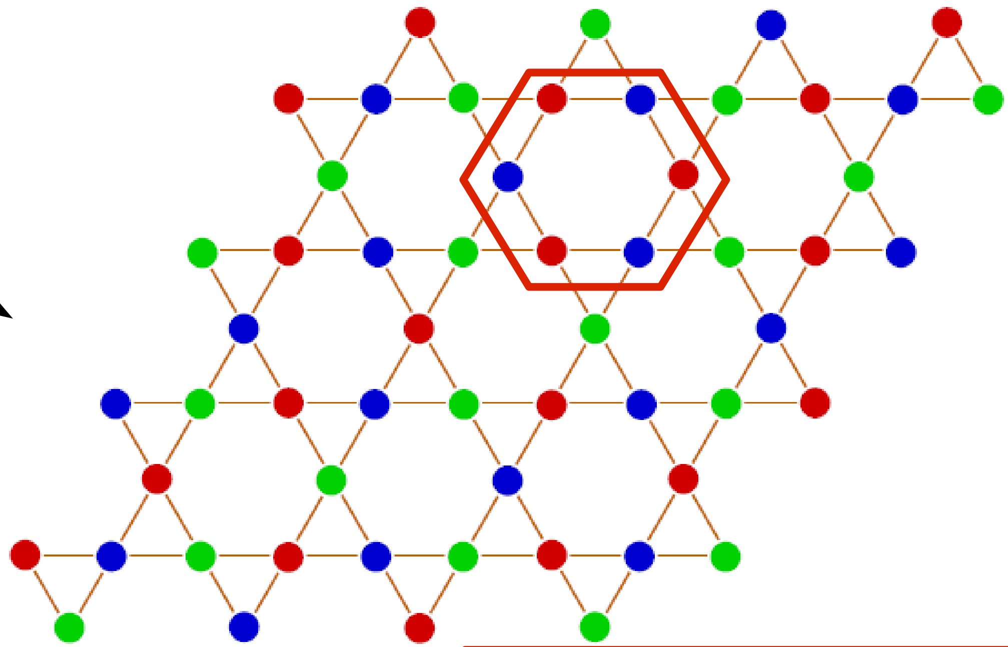


Several (exponential) degeneracies at semi classical level
Each triangle satisfies the minimum energy condition

q=0



√3 x √3

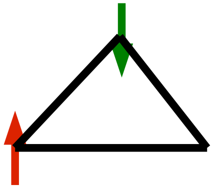


Rotate “two color loops” to get more solutions

Baxter (1970) calculated exact exponent for large N

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

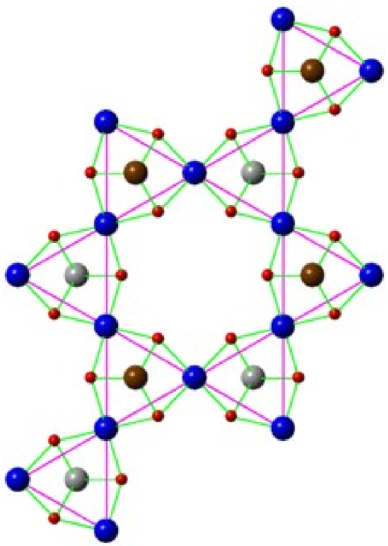
quantum “order by disorder” effects can generally lift this degeneracy (Villain 80, Henley, 87), but not always Also see: Chubukov (PRL 1992)

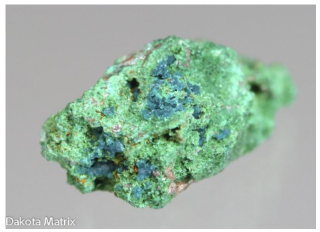


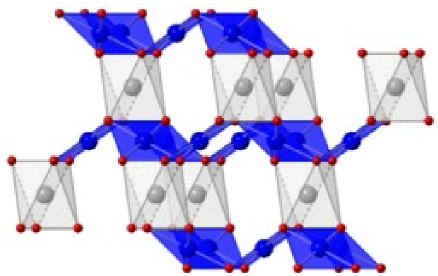
Physics of “frustrated” magnetic systems

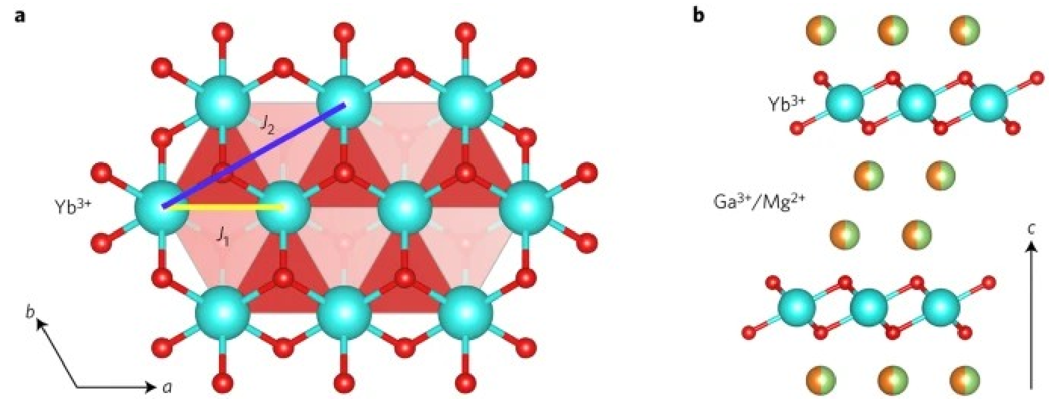
Material realizations – harbor “valence bond solids” and “spin liquids”

Kagome





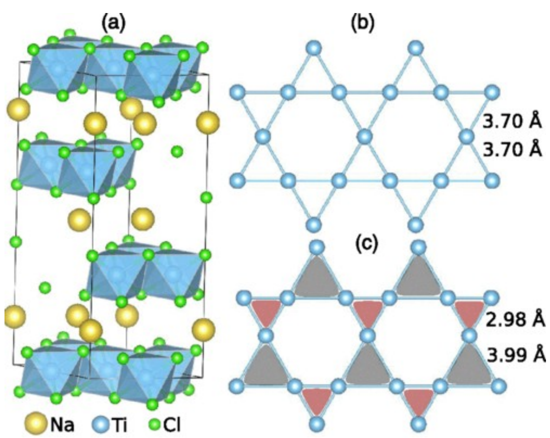


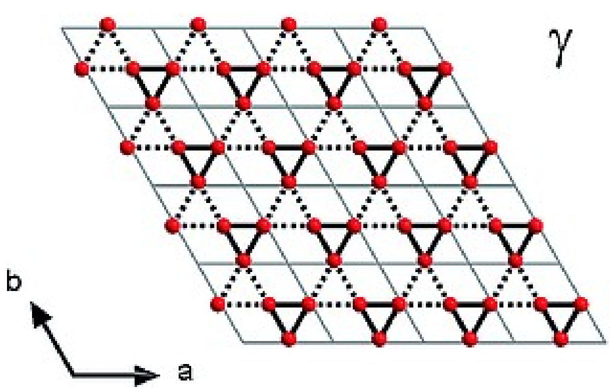


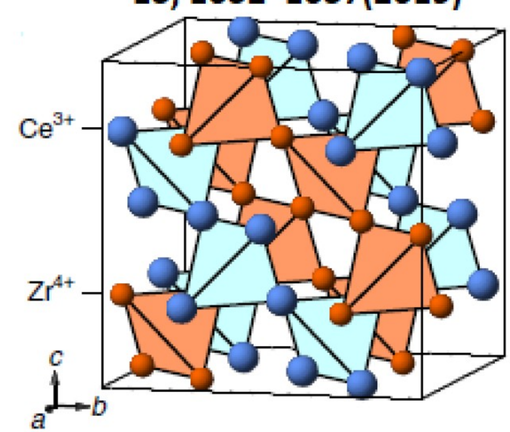
Triangular lattice system: $YbMgGaO_4$
Fig: J. Paddison et al, Nat. Physics (2017)

S=1/2 Herbertsmithite (barlowite, kapellasite…)
Nice review: M. Norman, Rev. Mod. Phys. (2018)

B Gao et al. Nat. Phys. 15, 1052–1057(2019)







S=1 $Na_2Ti_3Cl_8$, Fig: A. Paul et al (PRL 2020)

Pyrochlores
$Ce_2Zr_2O_7$, $Ho_2Ti_2O_7$, $Yb_2Ti_2O_7$,….

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

$$H\,\psi = E\,\psi$$

"Solve" the Schrodinger equation

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

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

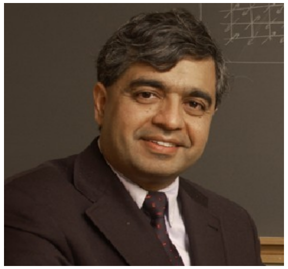
Summit (Oak Ridge) has largest storage array = 250 PetaBytes

Some approximations: next lecture

Lessons learnt from spin ½ kagome (30 years of work on one slide!)

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

Problem has long history: Elser, Harris, Kallin, Berlinsky, Sachdev, Henley, Huse, Singh, Senthil, Nikolic, Marston, Zeng, Lhuillier, Bernu, Mila, Chubukov, Yang, Girvin ....



S. Sachdev, V. Elser 1990's Spin liquid

T. Senthil 2003: valence bond solid + Marston-Zeng (1991)+Singh(2007)

S. White Liquid: 2011

Recent progress (<u>not</u> a complete list)

Y. Ran, M. Hermele, P. Lee, X. Wen *(PRL 2007)*
G. Evenbly, G. Vidal (*PRL 2010*)
S. Yan, D. A. Huse, S.R. White (*Science 2011*)
S. Depenbrock, et al (*PRL* 2012)
H.C. Jiang, Z. Wang, L. Balents (*Nature 2012*)
Wan, Tchernyshyov (*PRB 2013*)
Punk, Chowdhury, Sachdev (*Nature Physics* 2014)
Hwang, Huh, Kim (*PRB* 2015)
He, Bhattacharjee, Pollmann, Moessner (*PRL* 2015)
He, Zaletel, Oshikawa, Pollmann (*PRX* 2017)
Ralko, Mila, Rousochatzakis (*PRB* 2018)

<u>Summary of what is going on</u>

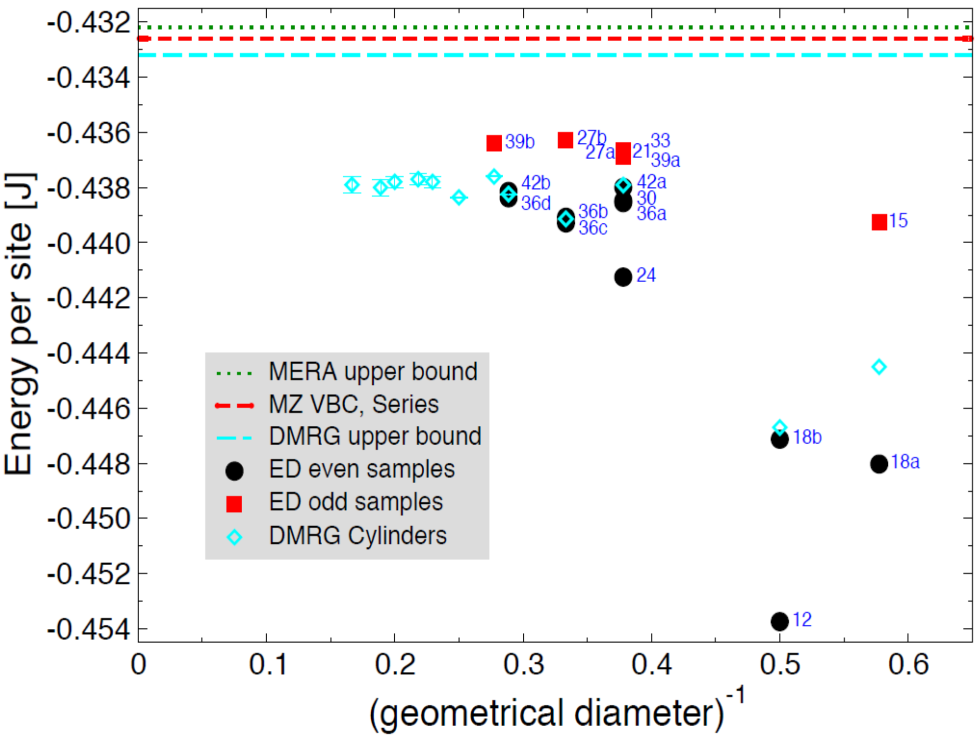


Fig. Lauchli, Sudan, Sorensen, PRB (2011)

Small energy scales and wildly different physics! ("spin liquid" vs. ordered states)

From a more <u>materials perspective</u>
0.002*200 K = 0.4 K = 0.04 meV/atom

Tiny scales (1-10 meV is the best we have)

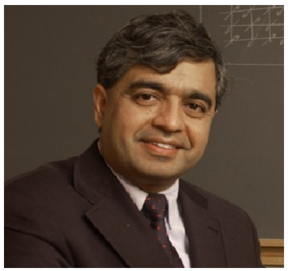
Phase diagrams can be complex

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

$$H_{XXZ}[J_z] = \sum_{\langle i,j \rangle} S_i^x S_j^x + S_i^y S_j^y + J_z \sum_{\langle i,j \rangle} S_i^z S_j^z$$

$$H[J_z, J_2] = H_{XXZ}^{\text{nn}}[J_z] + J_2 H_{XXZ}^{\text{nnn}}[J_z]$$

Problem has long history: Elser, Harris, Kallin, Berlinsky, Sachdev, Henley, Huse, Singh, Senthil, Nikolic, Marston, Zeng, Lhuillier, Bernu, Mila, Chubukov, Yang, Girvin ....



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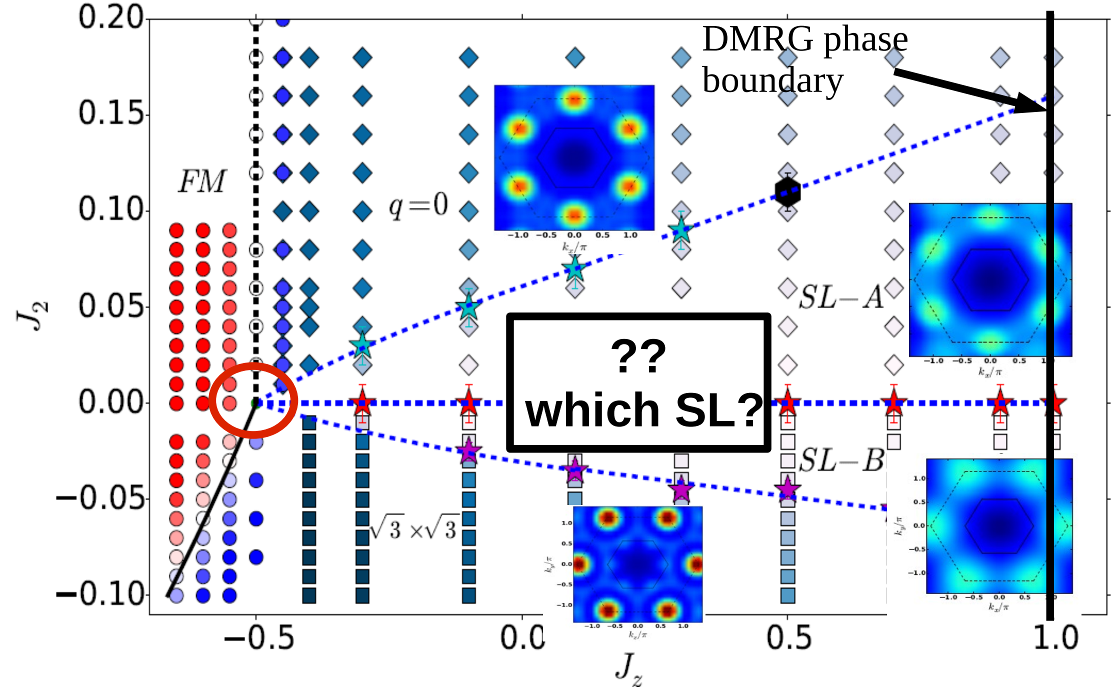
S. White Liquid: 2011



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He, Zaletel, Oshikawa, Pollmann (*PRX* 2017)
Ralko, Mila, Rousochatzakis (*PRB* 2018)

<u>Our ground state phase diagram (2018)</u>

Motivated by a (ground state) exactly solvable point in one of the simplest models known in the literature (XXZ) (**HJC** et al PRL 2018, **HJC** et al, PRB 2019)

<u>Natural next question</u>:

<u>Where</u> is the real material on these types of diagrams?

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	–	–			
6	Suzuki and Suga [54, 55]	Magnetic specific heat	−1.53	−24.4	+5.25	−0.95	–	–			
7	**Yadav et al.** [49][b]	Quantum chemistry (MRCI)	+1.2	−5.6	+1.2	−0.7	+0.25	–			
8	Ran et al. [27]	Spin wave fit to INS gap	–	−6.8	+9.5	–	–	–			
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	–	–	–	–	–	

[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.

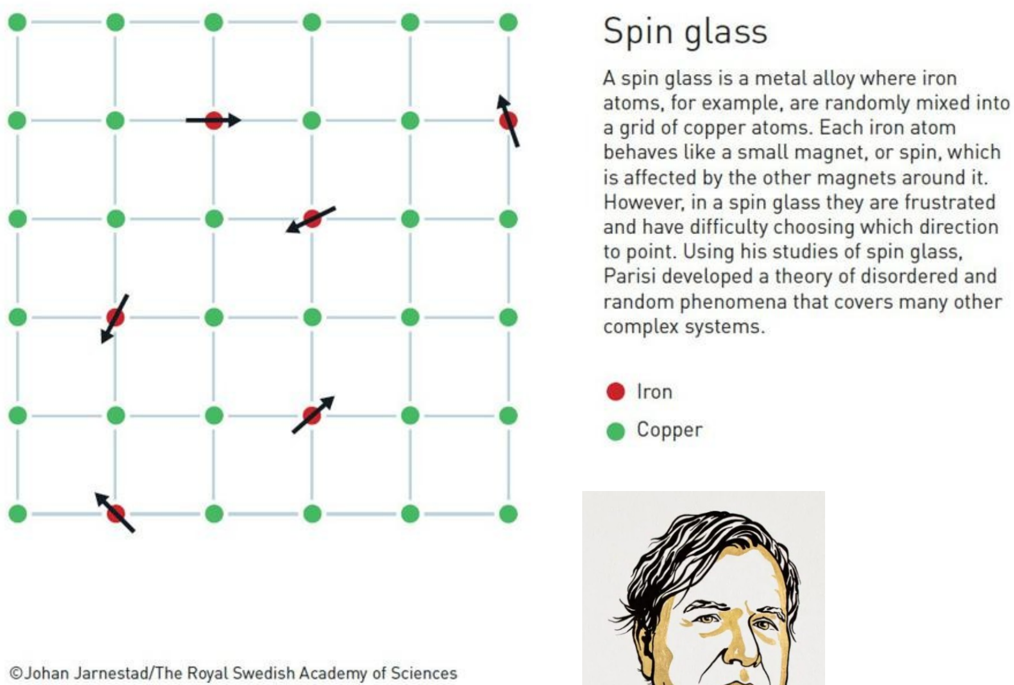
$$\mathcal{H}_1 = \sum_{\langle ij\rangle_\gamma} [J\mathbf{S}_i \cdot \mathbf{S}_j + K S_i^\gamma S_j^\gamma + \Gamma(S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) + \Gamma'(S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta + S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma)],$$

<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)

Many kinds of magnetic materials, with rich physics

(Disordered) spin glasses

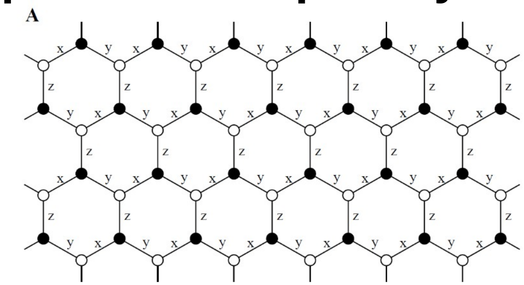


(2021)

C. Orzel, forbes.com

G. Parisi

Spin-orbit coupled systems



A. Kitaev

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Annals of Physics 321 (2006) 2–111

ANNALS of PHYSICS

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"Topological quantum computation"

Anyons in an exactly solved model and beyond

Alexei Kitaev *

California Institute of Technology, Pasadena, CA 91125, USA

Received 21 October 2005; accepted 25 October 2005

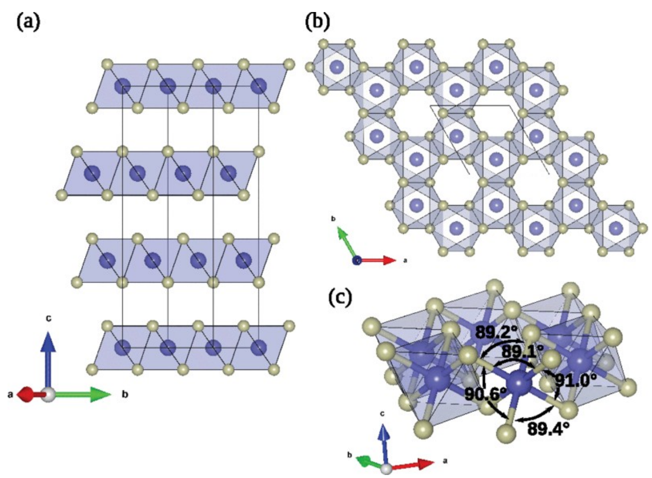
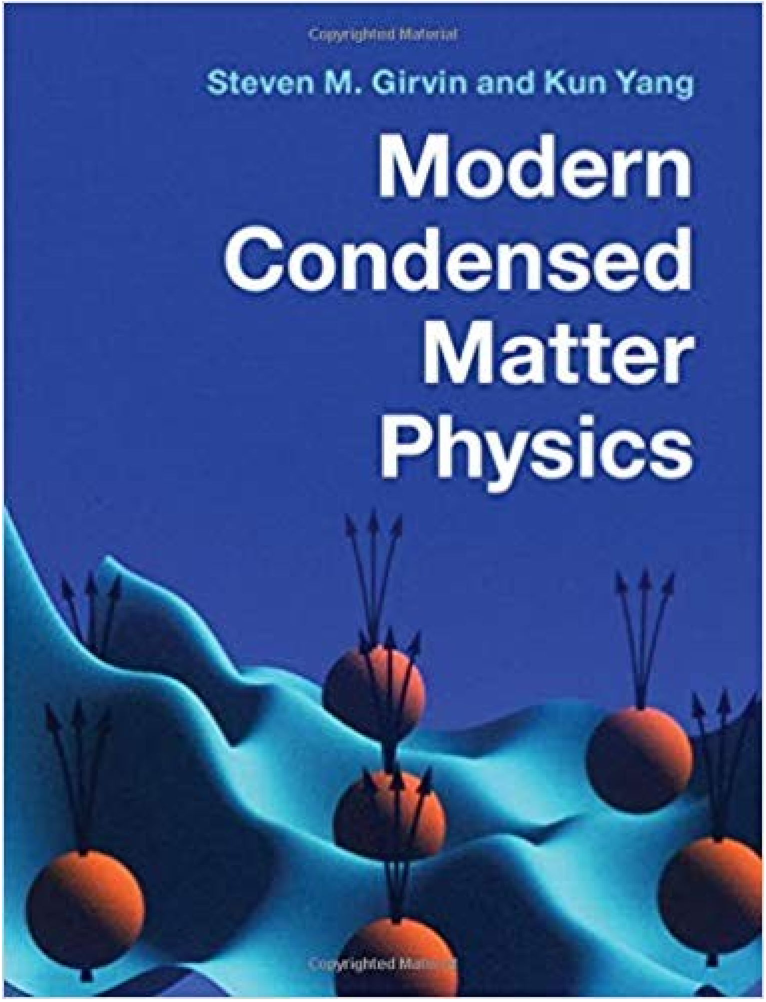
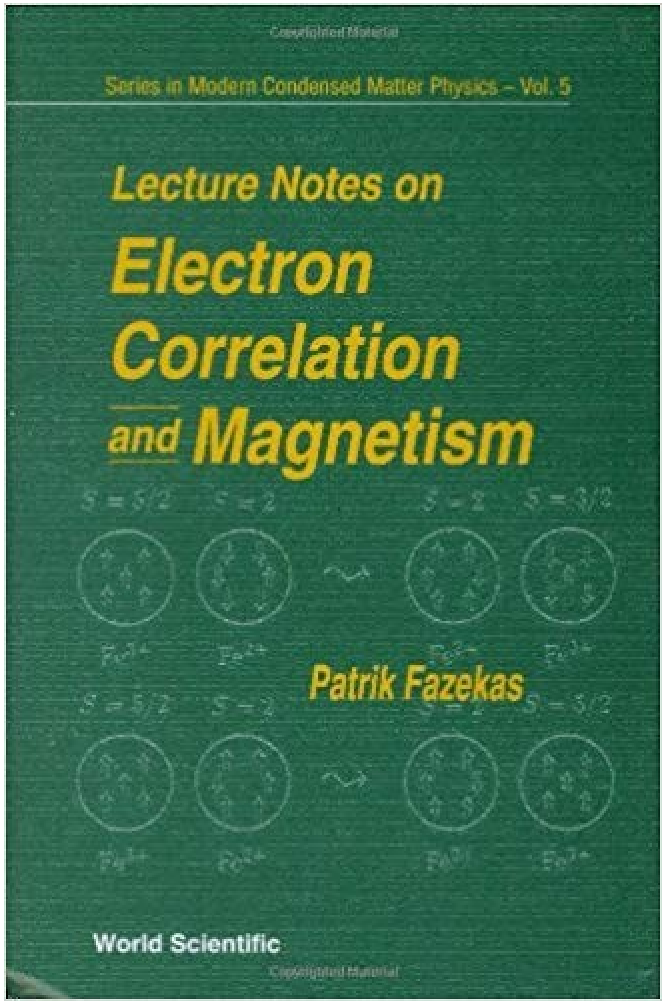


Fig: Ruthenium chloride
Plumb et al, PRB (2014)

Summary: study of magnetism may be useful… but what is its origin?

Some nice books to learn the basics





I have posted some notes on **https://sites.google.com/site/hiteshchanglani/teaching**