Strongly Correlated Phases in Layered Materials

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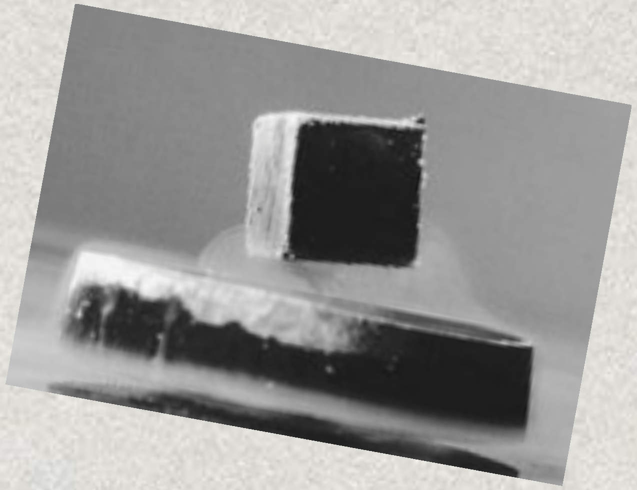
SIMPLE RULES

Computational methods are an important tool to allow us to learn how simple rules form complicated behavior.

Emergent Phenomena



COMPLICATED BEHAVIOR

Strongly Correlated Systems!!

$$L_{\mathrm{QCD}} = \bar{\psi}_i (i\gamma^\mu (D_\mu)_{ij} - m\delta_{ij})\psi_j - \frac{1}{4} G^a_{\mu\nu} G^{\mu\nu}_a$$

$$\sout{L_{\rm QCD} = \bar{\psi}_i(i\gamma^\mu(D_\mu)_{ij} - m\delta_{ij})\psi_j - \frac{1}{4}G^a_{\mu\nu}G^{\mu\nu}_a}$$

SIMPLE RULES

COMPLICATED BEHAVIOR

$$\hat{G} \equiv e^{-\tau H} \qquad |\Psi_0\rangle \propto \lim_{N\to\infty} \hat{G}^N |\Psi_T\rangle$$

$$H = -\frac{\hbar^2}{2m}\sum_i \nabla_i^2 + \sum_{ij}\frac{1}{|r_i - r_j|} + V_{\rm ext}(R)$$

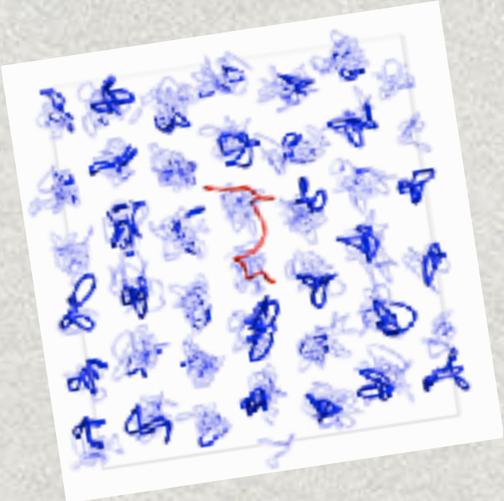
(for electronic systems)

Diffusion Monte Carlo

$$H = -\frac{\hbar^2}{2m}\sum_i \nabla_i^2 + \sum_{ij} V_{\rm aziz}(r_i - r_j)$$

(for bosonic systems)

Path Integral Monte Carlo



TODAY: INTERESTING MAGNETIC PHASES

Clark, et. al JCP (2011)

Clark and Ceperley, PRL (2006)

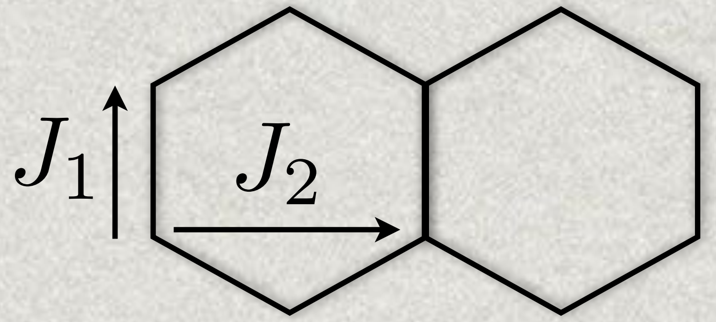
Biroli, Clark,et. al, PRB (2011)

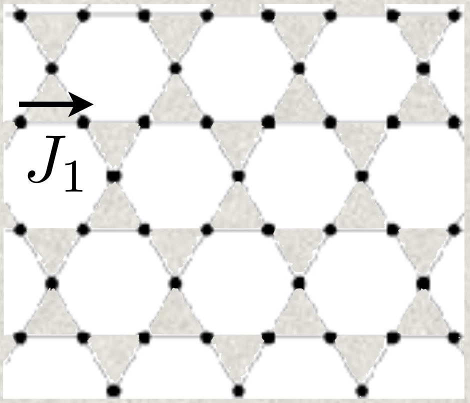
What are the right simple rules for systems with interesting magnetic phases?

You could start here: $$H = -\frac{\hbar^2}{2m}\sum_i \nabla_i^2 + \sum_{ij}\frac{1}{|r_i - r_j|} + V_{\text{ext}}(R)$$

Integrate out orbital degrees of freedom

But it's easier to be here: $$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j + ...$$

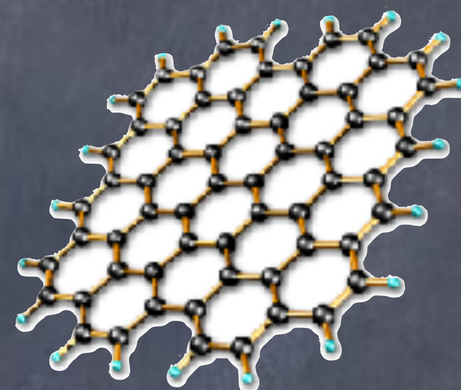




One of the biggest stories in cosmology in the last decades has been that we didn't know about most of the "stuff" that makes up the universe.

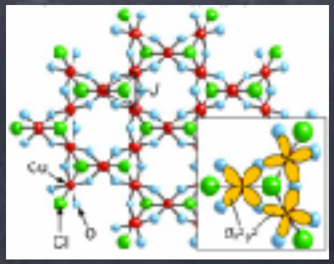
Today we will see that we probably don't know about most the phases in the universe.

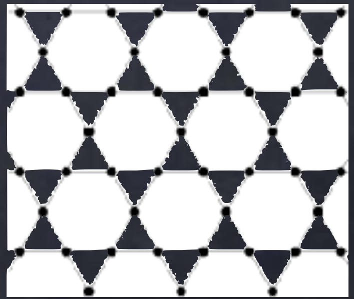
and that these phases show up in materials like these!



Modified Graphene

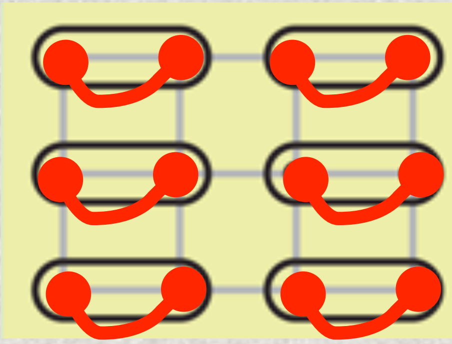
Herbertsmithite:
$ZnCu_3(OH)_6Cl_2$





Outline:

- What are these phases?
- How does QMC work?
- Identifying these phases in these materials.

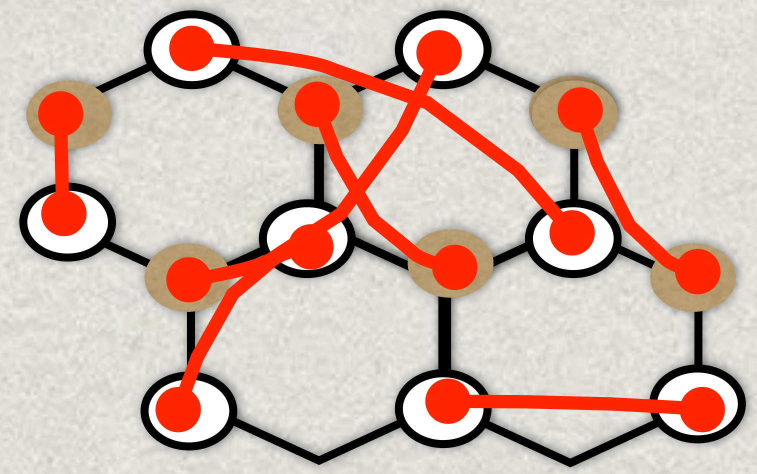


VALENCE BOND CRYSTAL

$$|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle$$

2 lattice sites pair up and are anti-correlated!

Monogamous pairs!



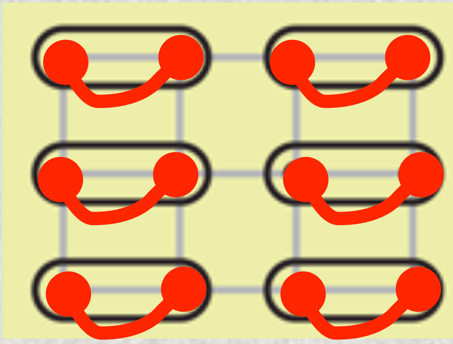
SPIN LIQUID

This is misleading. There are hundreds of different spin liquid phases. These are real different phases just as different as liquid and solid.

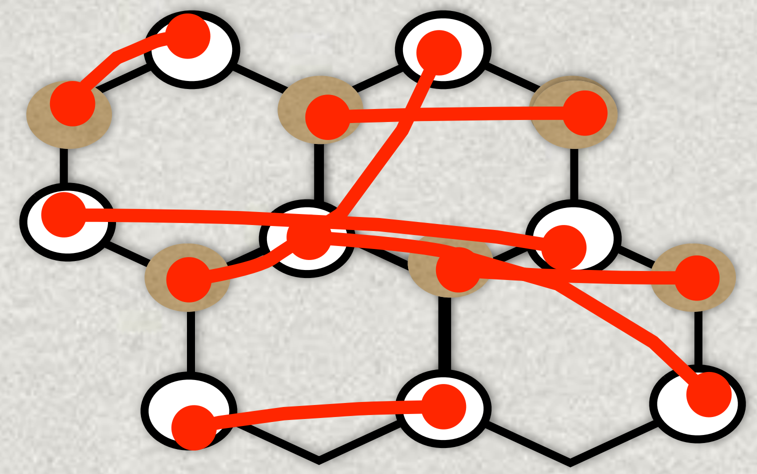
Anderson originally wrote down for superconductivity in 1980's.

Full circle: Today, thinking about superconductors as instability of spin liquids.

Anderson, Science (1987)



VALENCE BOND CRYSTAL



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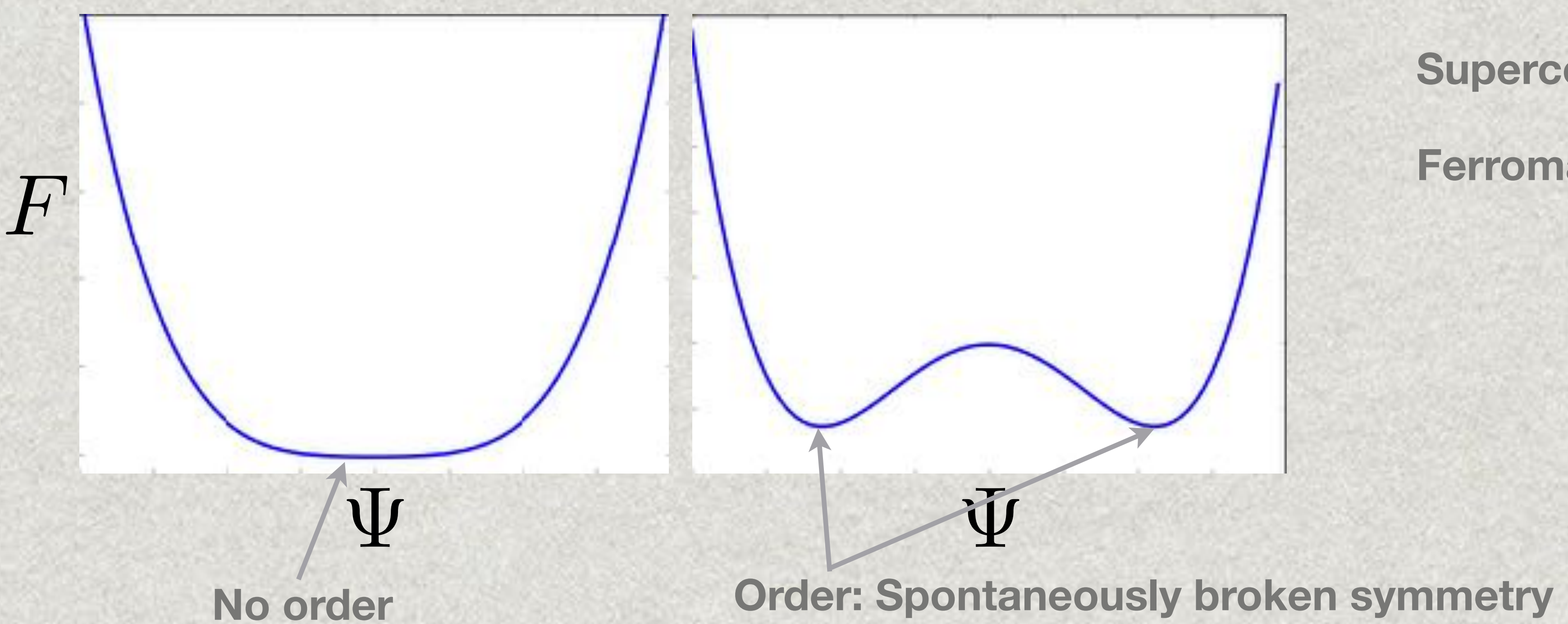
Landau Theory of Phase Transitions

$$F = r\Psi^2 + s\Psi^4 + (\nabla\Psi)^2$$

Order Parameter: Local probe of the system. Non-zero in phase you care about and zero otherwise.

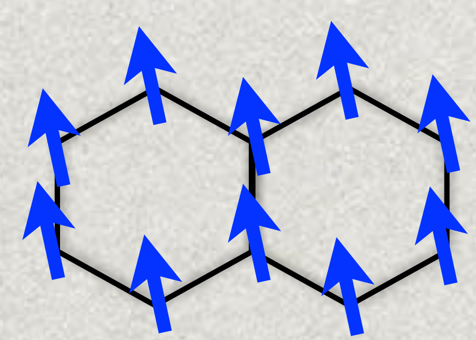
$$r \sim (g_c - g)$$

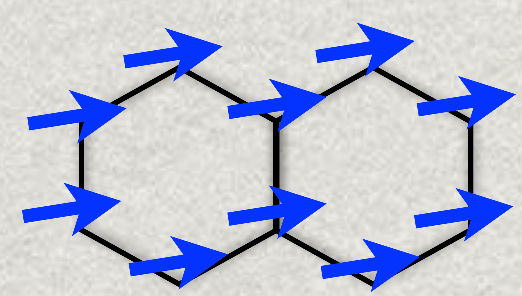


Order Parameters

Superconductor: $\Psi = c_\uparrow c_\downarrow$

Ferromagnet: $\Psi = c_\alpha \sigma_{\alpha\beta} c_\beta$



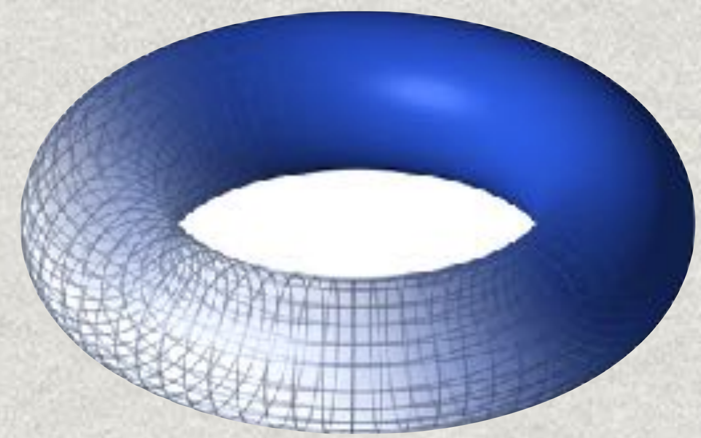


Topological Phases

Some phases can't be probed locally!

No local order parameter and no symmetry breaking.

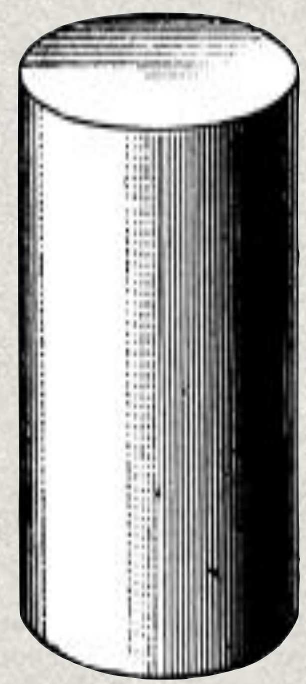
This state is featureless!



Landau's Theory doesn't work!

- **Hard to identify**

- **Very Stable**
- **Local Perturbations can't break**
- **Useful for quantum computation**
- **Interesting Theoretically**



Topological?

Knows the manifold on which it lives.

Cylinder: 2 ground states

Torus: 4 ground states

PHASES AN UNDERGRAD MIGHT NOW:

liquid
gas
solid
plasma

PHASES A FIRST YEAR GRADUATE STUDENT MIGHT NOW:

liquid
gas
solid
plasma
antiferromagnet
superfluidity
superconductivity

ACTUAL PHASES

liquid
gas
solid
plasma
antiferromagnet
superfluidity
superconductivity
spin liquid A
spin liquid B
spin liquid C
spin liquid D
spin liquid E
spin liquid F
spin liquid G
spin liquid H
spin liquid I
spin liquid J
spin liquid K
...

The fundamental laws necessary for the mathematical treatment of a large part of physics and the whole of chemistry are thus completely known, and the difficulty lies only in the fact that the application of these laws leads to equations that are too complex to solve.

- Paul Dirac

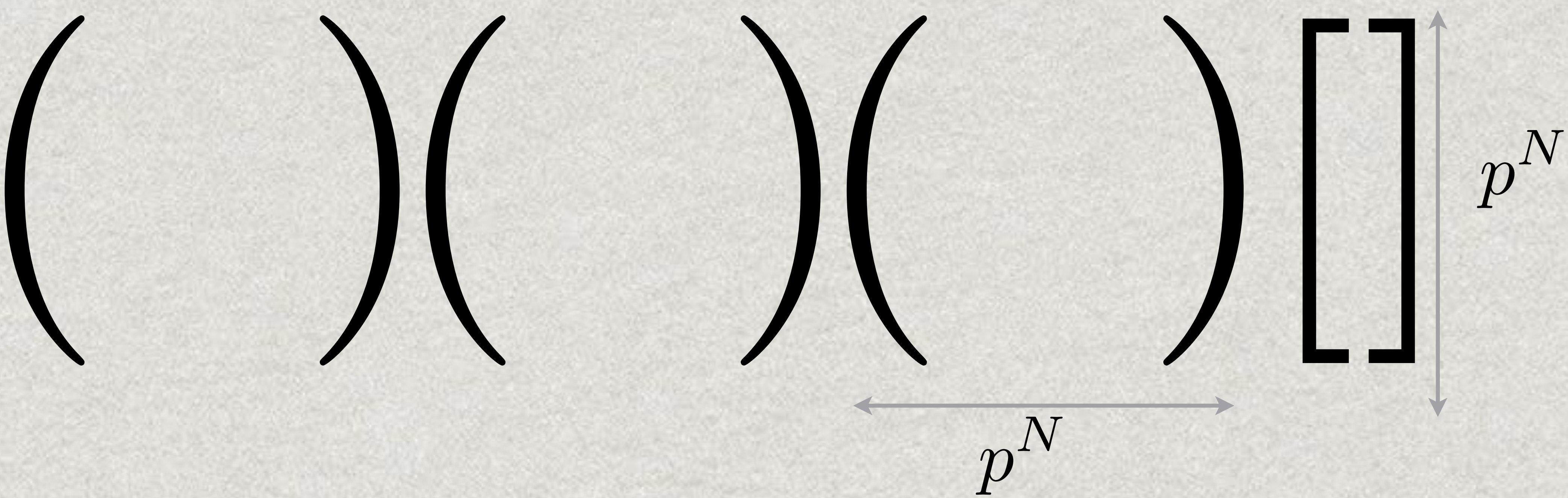
“let’s solve the equations”

$\hat{G} \equiv e^{-\tau H}$ ← This is a matrix

$|\Psi_T\rangle$ ← This is a vector

$$|\Psi_0\rangle \propto \lim_{N \to \infty} \hat{G}^N |\Psi_T\rangle$$

Too big and too slow!



Maybe we can do this stochastically?

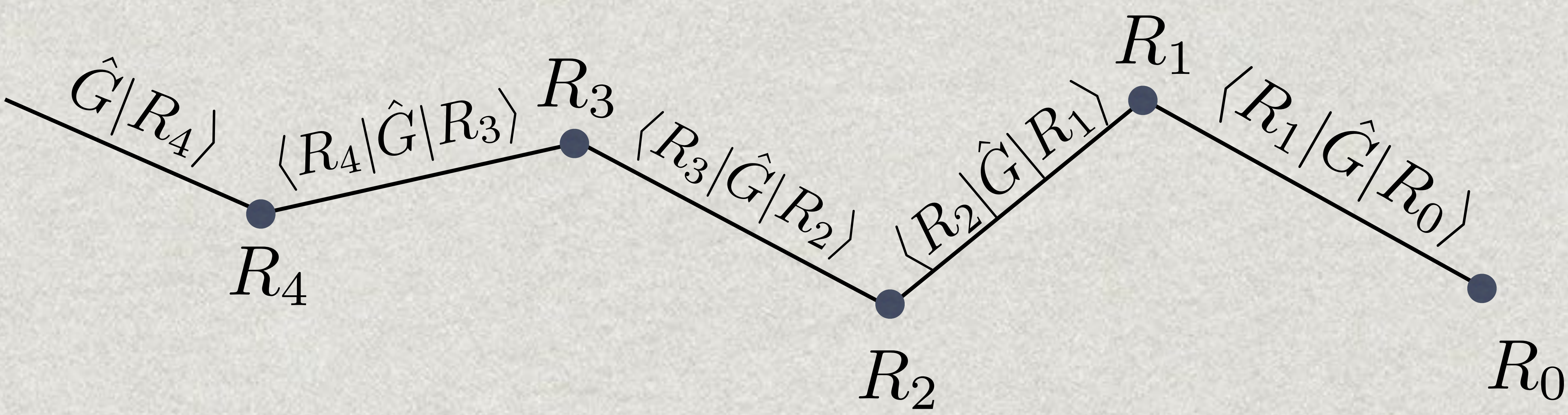
Quantum Monte Carlo

$$|\Psi_0\rangle \propto \lim_{N\to\infty} \hat{G}^N |\Psi_T\rangle$$

$$\begin{pmatrix} a & b & c \\ d & e & f \\ g & h & i \end{pmatrix}$$

$$[\hat{G}]^N = \hat{G}\hat{G}\hat{G}\hat{G}\hat{G}|\Psi_T\rangle$$

$$\sum_{R_0,R_1,R_2,R_3,R_4} \hat{G}|R_4\rangle\langle R_4|\hat{G}|R_3\rangle\langle R_3|\hat{G}|R_2\rangle\langle R_2|\hat{G}|R_1\rangle\langle R_1|\hat{G}|R_0\rangle\langle R_0|\Psi_T\rangle$$



1. Choose R_0 with probability $\langle R_0|\Psi_T\rangle$.

2. Choose R_1 with probability $\langle R_2|\hat{G}|R_1\rangle$.

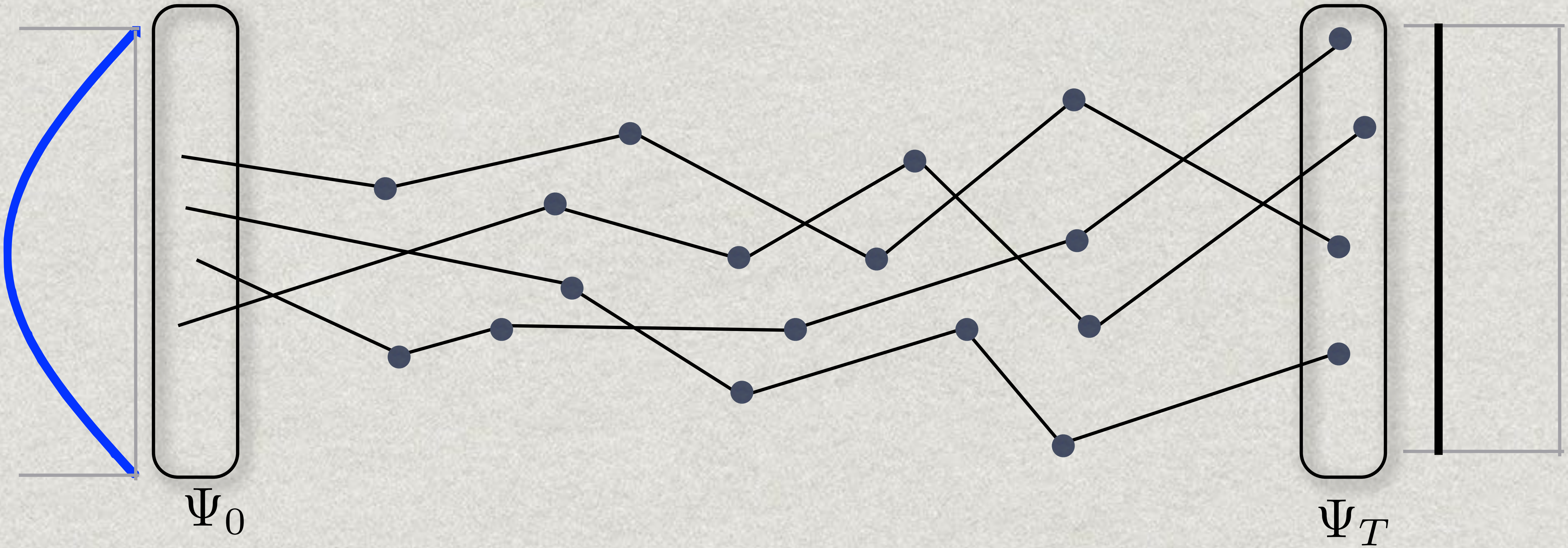
3. ...

Quantum Monte Carlo

$$|\Psi_0\rangle \propto \lim_{N\to\infty} \hat{G}^N|\Psi_T\rangle$$

$$[\hat{G}]^N = \hat{G}\hat{G}\hat{G}\hat{G}\hat{G}|\Psi_T\rangle$$

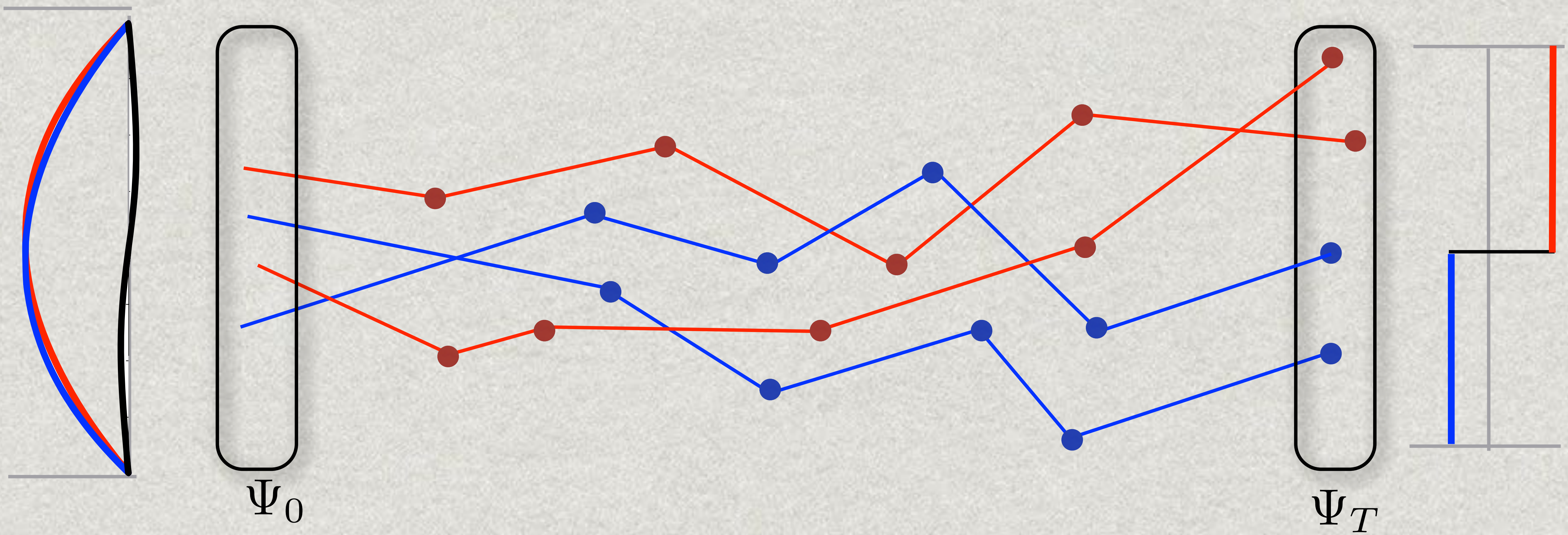
$$\sum_{R_0,R_1,R_2,R_3,R_4} \hat{G}|R_4\rangle\langle R_4|\hat{G}|R_3\rangle\langle R_3|\hat{G}|R_2\rangle\langle R_2|\hat{G}|R_1\rangle\langle R_1|\hat{G}|R_0\rangle\langle R_0|\Psi_T\rangle$$



Swept under the rug

We've assumed Ψ_T is a probability distribution.

"THE FERMION SIGN PROBLEM"



Dirac was right :(

(unless you happen to be a boson)

Exact answers are hard. Maybe we can get an approximate answer by guessing the wave-function.

Time Honored Approach

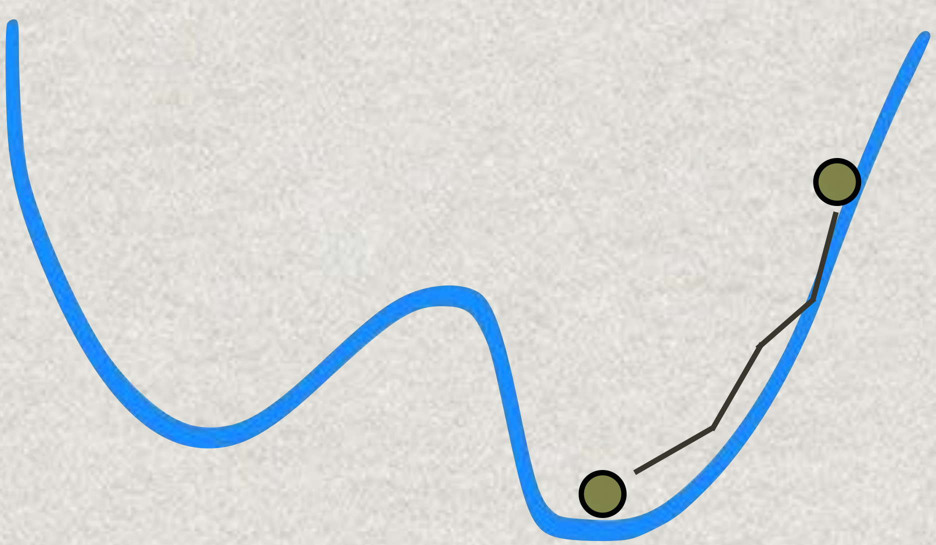
- BCS Superconductivity
- Fractional Quantum Hall

Hilbert Space is a big place

$\Psi[\alpha_1, \alpha_2, \alpha_3, \ldots]$

How do we decide amongst these wave-functions?

Variational Principle: $E_0 = \langle \Psi_0 | H | \Psi_0 \rangle \leq \langle \Psi_T | H | \Psi_T \rangle$



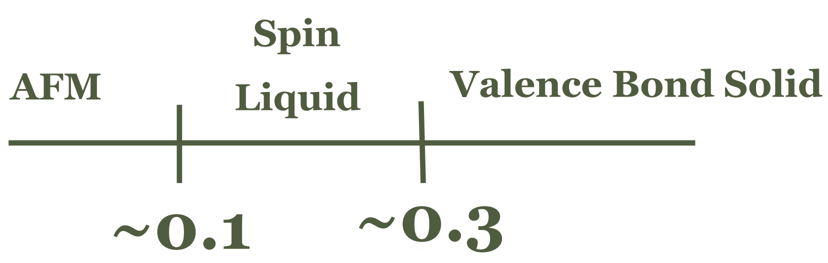
$$\frac{\partial \langle E \rangle}{\partial \alpha}$$

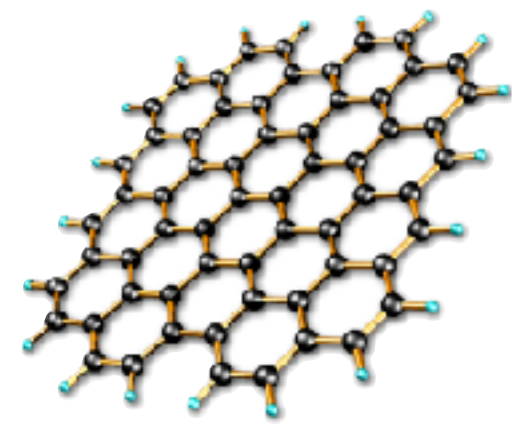
Variational Monte Carlo!!

Guessing a wave function: from an art to a science.

Honeycomb Lattices

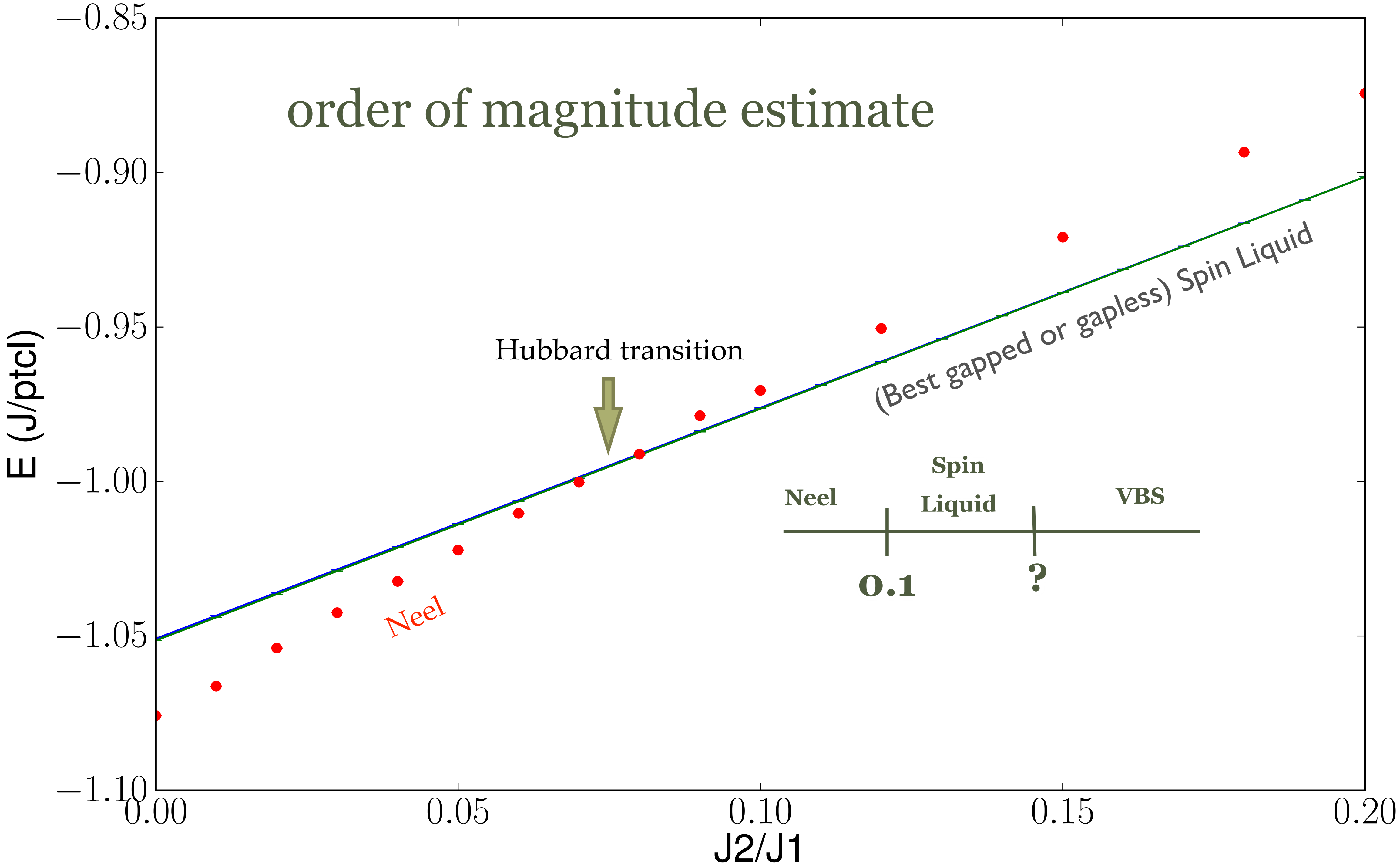
$$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j + ...$$





Which spin liquid phase is it?

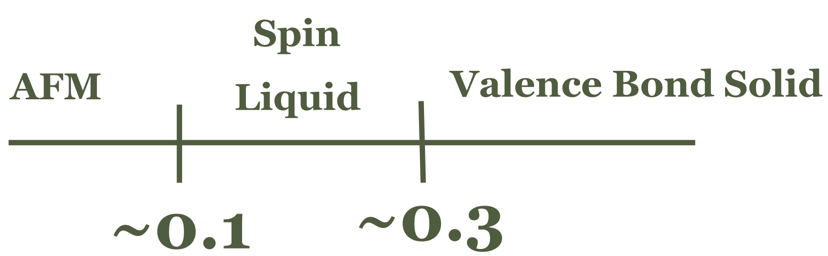
AFM State

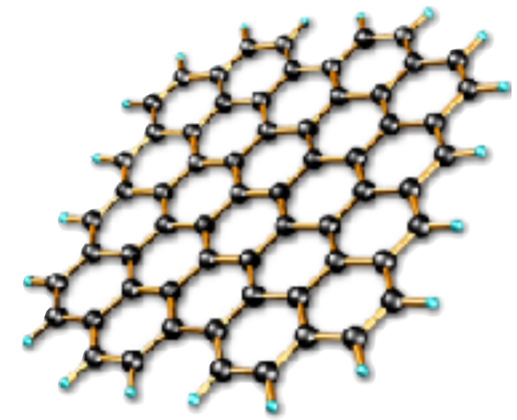


order of magnitude estimate
Hubbard transition
(Best gapped or gapless) Spin Liquid
Neel
Neel
Spin
Liquid
VBS
0.1
?
E (J/ptcl)
J2/J1
−0.85
−0.90
−0.95
−1.00
−1.05
−1.10
0.00
0.05
0.10
0.15
0.20

Honeycomb Lattices

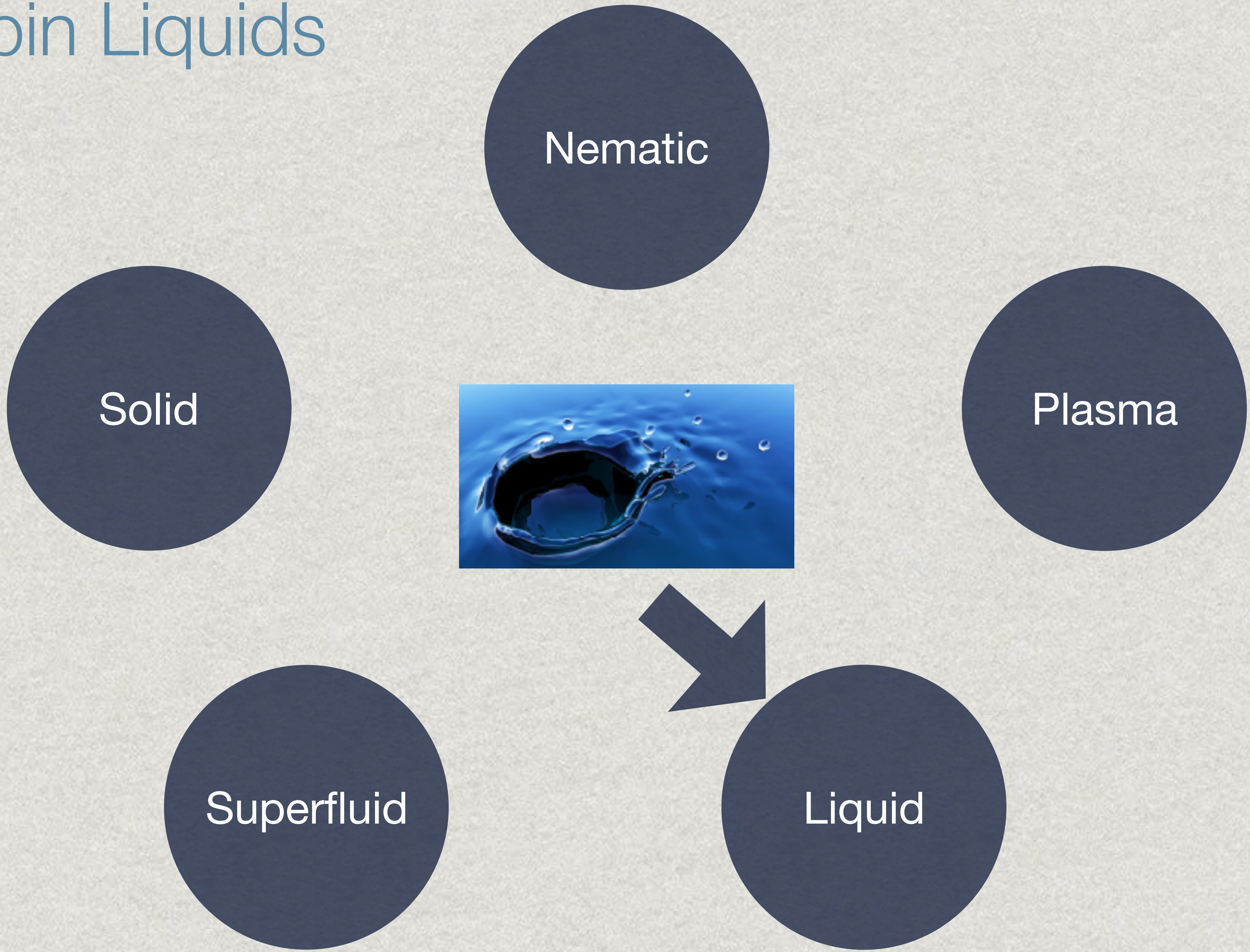
$$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j + \ldots$$



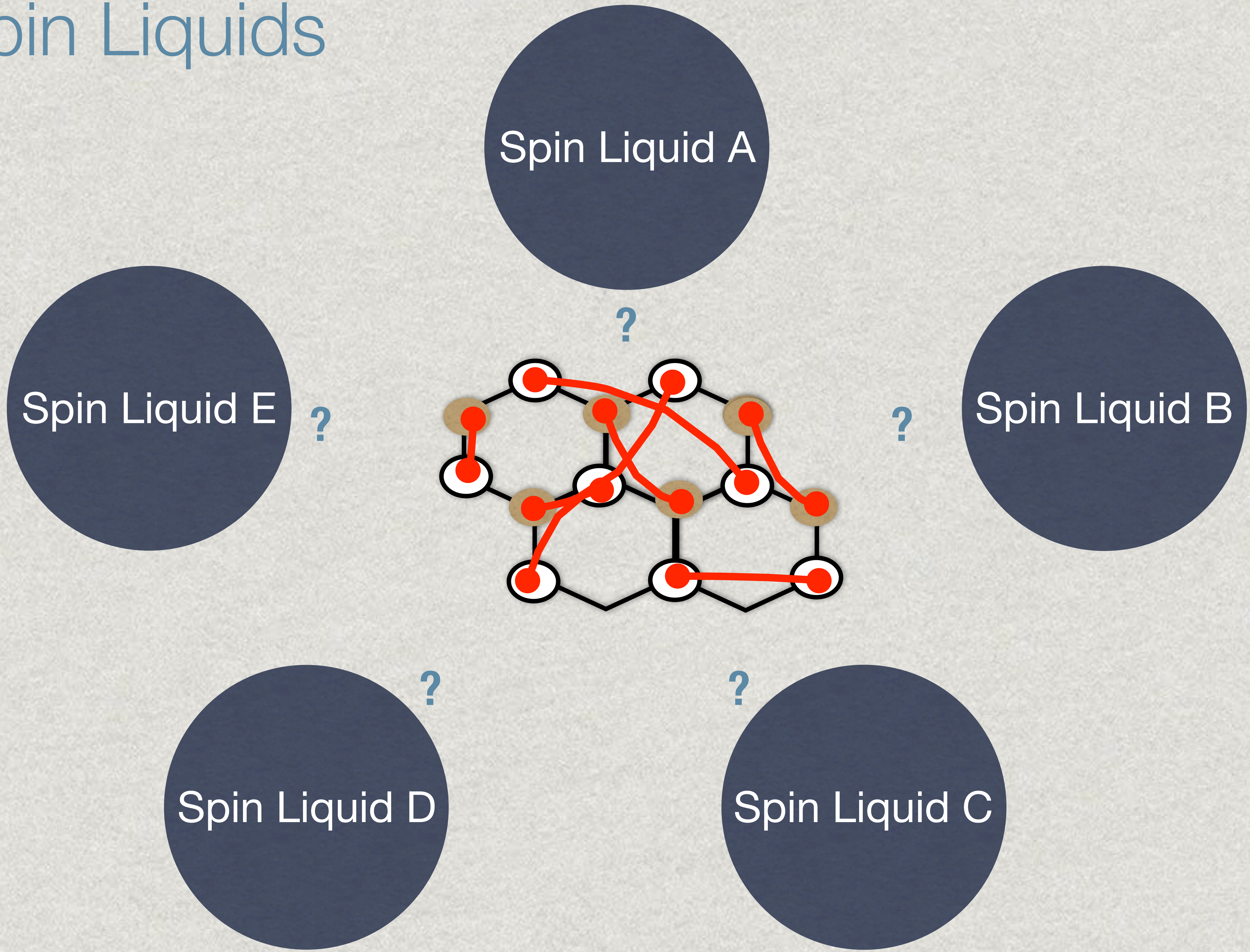


Which spin liquid phase is it?

Spin Liquids



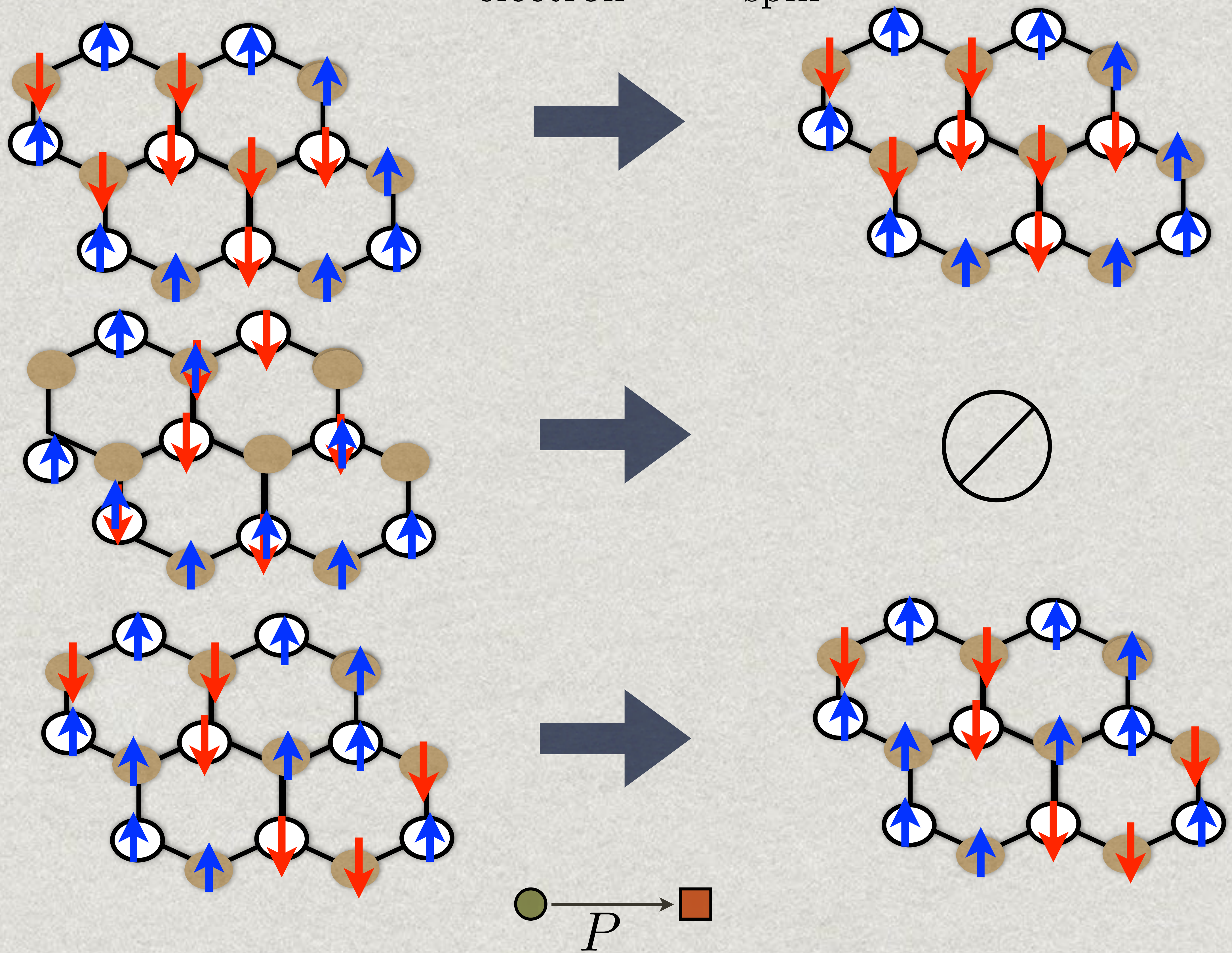
Spin Liquids



An important trick

ELECTRONS **SPINS**

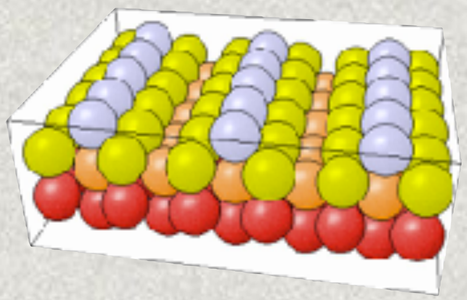
$$P\Psi_{\text{electron}} = \Psi_{\text{spin}}$$

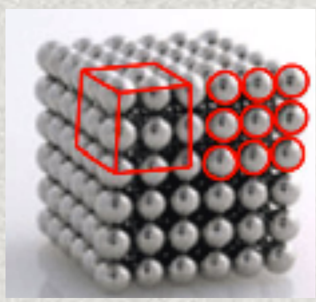


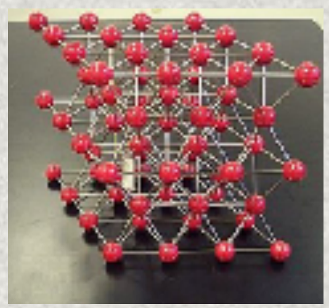
Which phases?

Let's recall how we classify "normal" phases:

SOLIDS

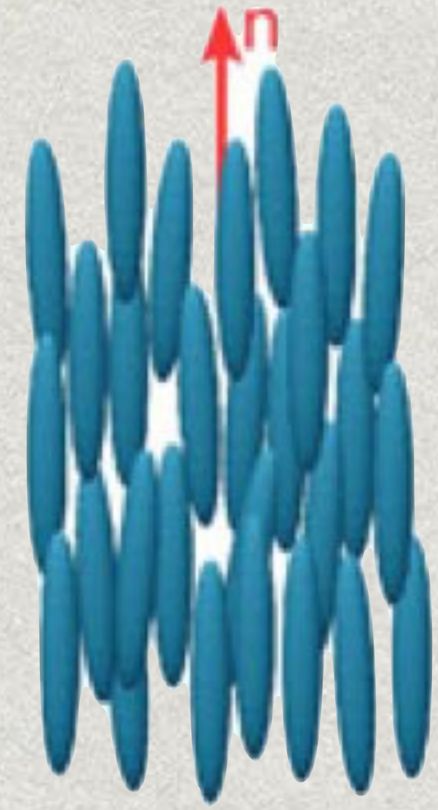






These are solids because they come back to themselves on discrete T and R.

NEMATIC



These are nematic because they come back to themselves under all T and R around axis.

This is liquid because it comes back to itself under all T and R.

LIQUID

Which phases?

- FCC Solid: Comes back to itself under T(1), R(90)
- Nematic: Comes back to itself under T,R(axis)
- Liquid: Comes back to itself under T,R

Does this work with spin liquids?

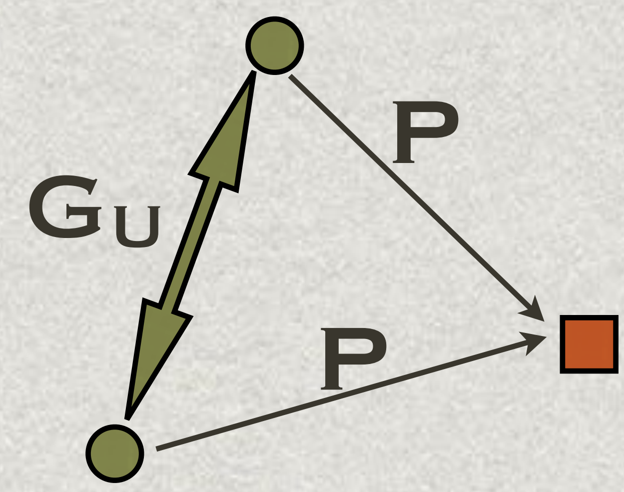
Spin liquids don’t break any symmetries. They are essentially featureless. Either there is one spin liquid phase or another approach is needed.

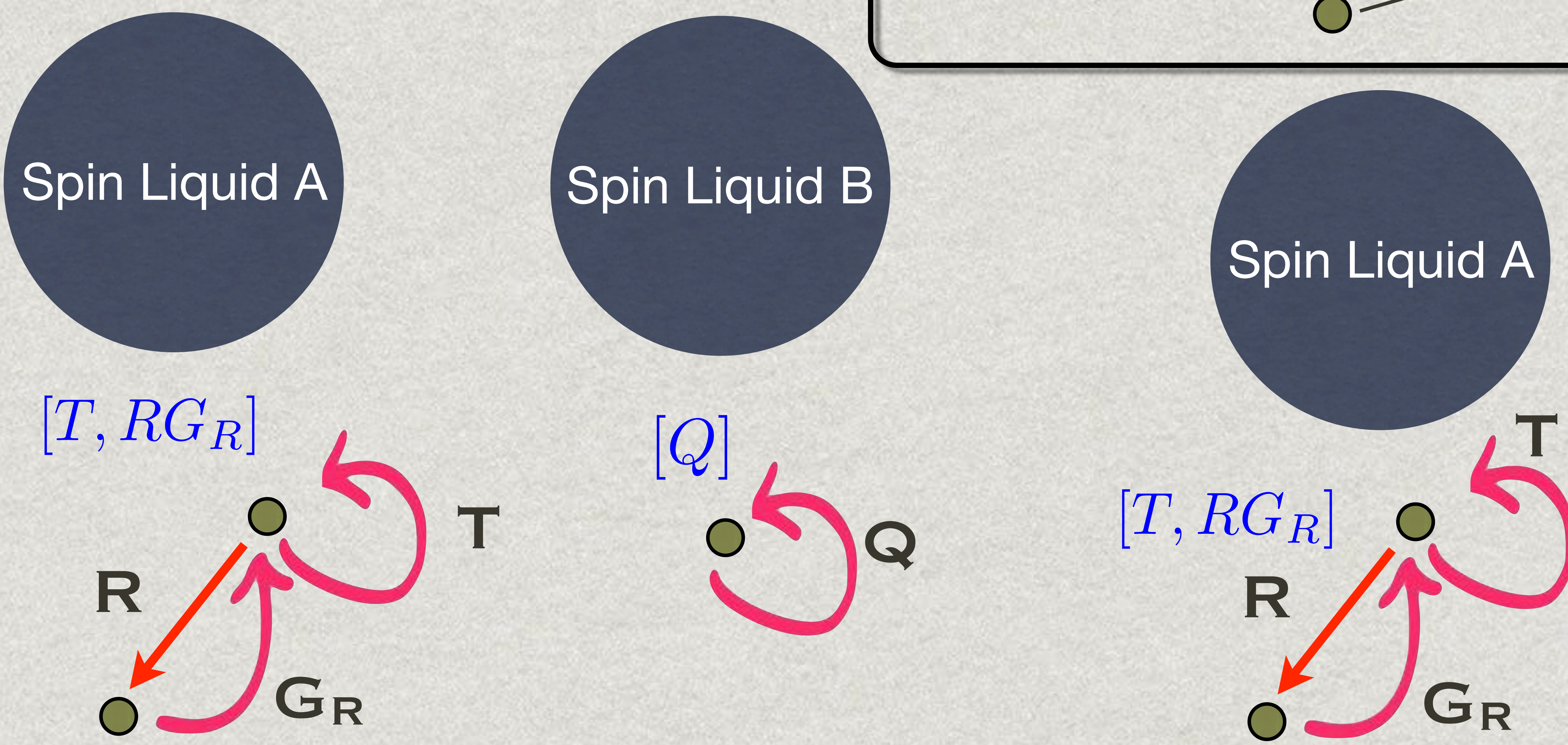
Experimentally: Hard! Have ■, not ●

Numerically: We can optimize over ●

2 electron wf → 1 spin wavefunction

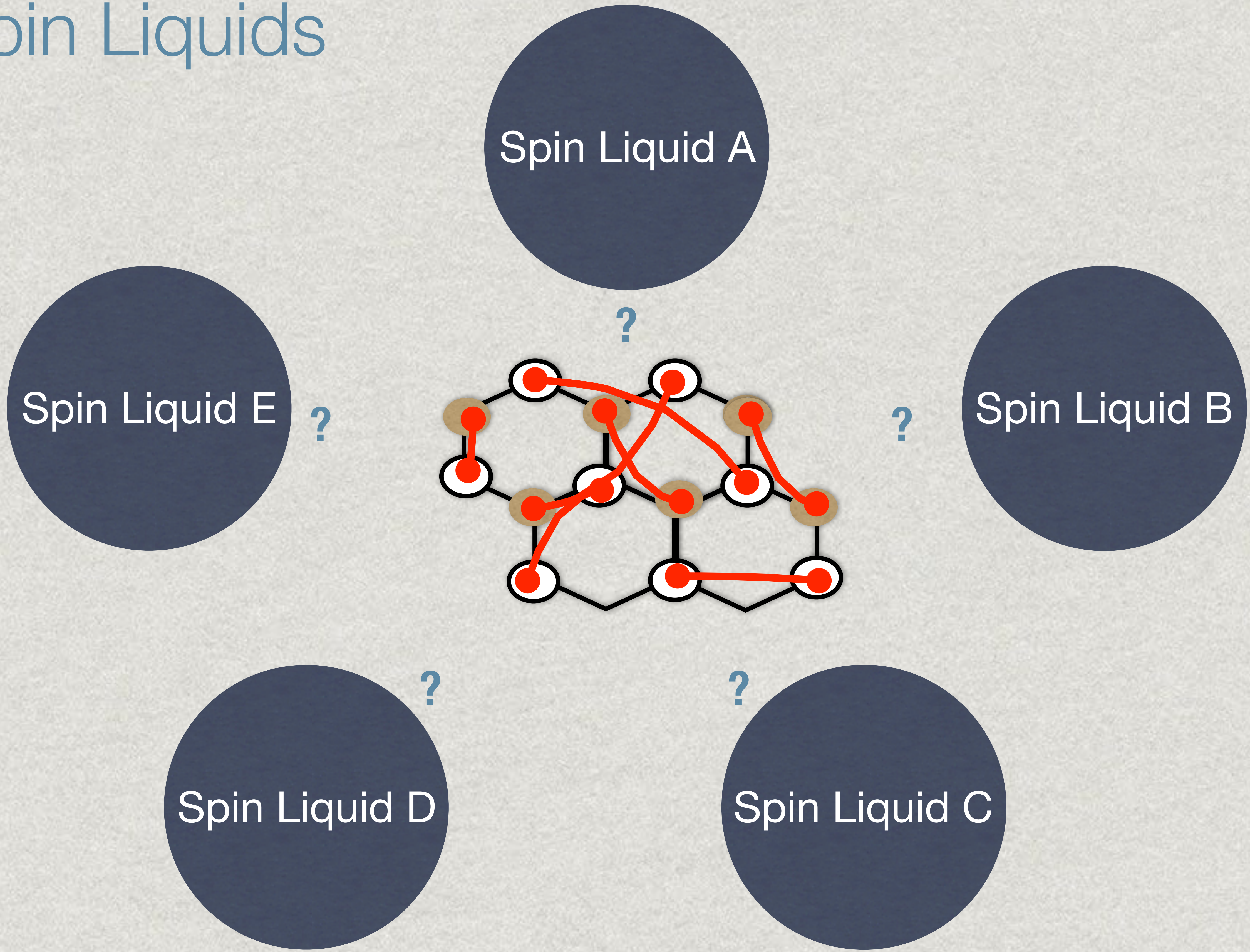
These 2 states are related by a local SU(2) gauge transformation G_U





Wen, PRL (2002)

Spin Liquids



Spin Liquids

Spin Liquid A

Spin Liquid B

Spin Liquid C

Spin Liquid D

Spin Liquid E

Spin Liquids

Spin Liquid A

Spin Liquid B

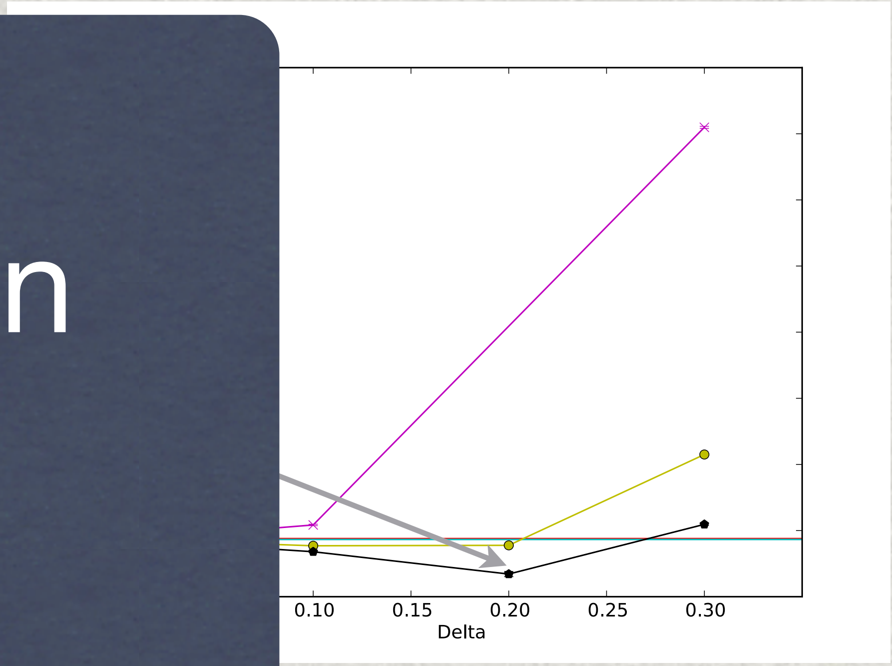
Spin Liquid C

Spin Liquid D

Spin Liquid E

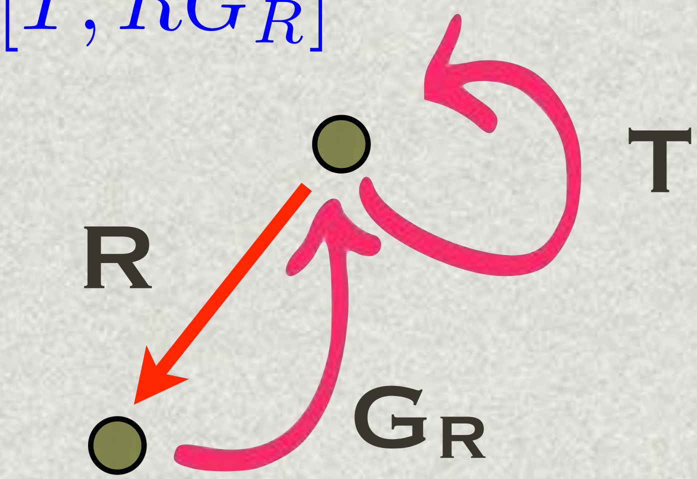
I. Find the "electron wave-function"

Why this spin liquid?



II. Figure how it transforms.

$[T, RG_R]$



Clark, Abanin, Sondhi PRL (2011)

What properties must a good spin liquid wave function have?

$$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j + \ldots$$

To minimize the J1 energy you want to obey the Marshall sign rule.

Positive		Negative
$\Psi(\uparrow\downarrow\uparrow\downarrow\uparrow)$	$\longrightarrow$	$\Psi(\downarrow\uparrow\uparrow\downarrow\uparrow)$
$\Psi(\downarrow\uparrow\uparrow\uparrow\downarrow)$	$\longrightarrow$	$\Psi(\uparrow\downarrow\uparrow\uparrow\downarrow)$
$\Psi(\uparrow\uparrow\downarrow\uparrow\downarrow)$	$\longrightarrow$	$\Psi(\uparrow\uparrow\uparrow\downarrow\downarrow)$

Quantify by

$$\sum_{\langle ij \rangle \in s} 1/2 \frac{\langle s'_{ij} | \Psi \rangle}{\langle s \, | \Psi \rangle} \text{ if } s_i \neq s_j$$

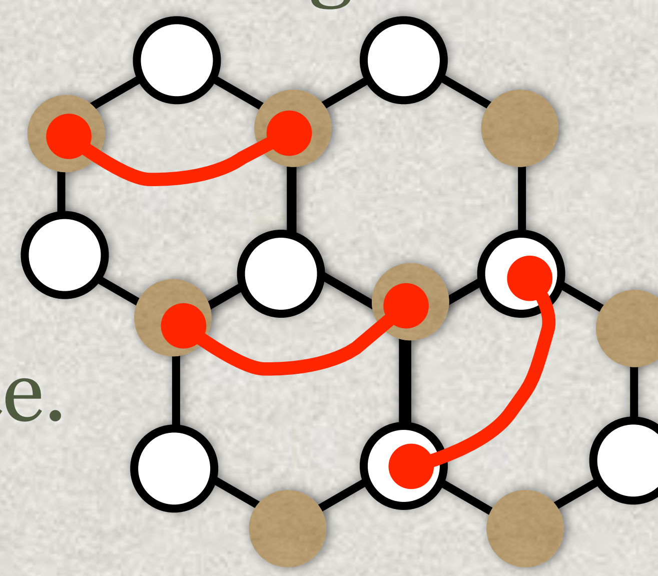
To minimize the J2 energy AA (BB) singlets must exist.

i.e. $\phi(\vec{r}_{AA}) \neq 0$

Why?

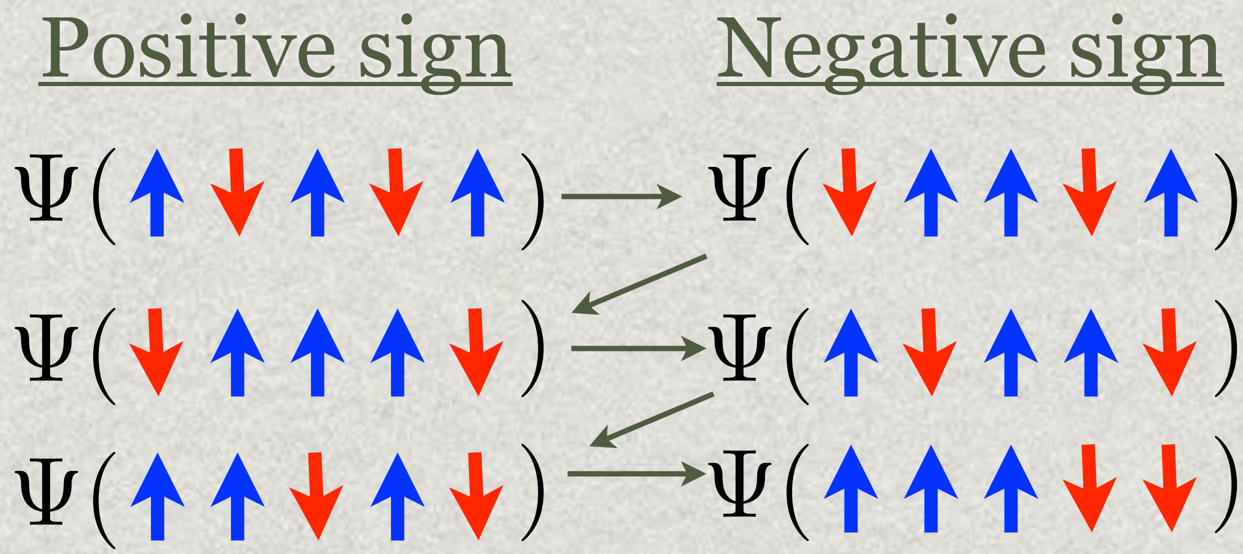
Decouples to triangle sublattice.

No sign rule, but must connect lattice.



Clark, Abanin, Sondhi, In preparation

Marshall Sign



$$H = J_1 \sum_{\langle ij \rangle} S_i \cdot S_j$$

Any wf that minimizes the ground state energy of H (on a bipartite lattice), has the sign structure as above.

$$\langle E \rangle = \langle \Psi | H | \Psi \rangle = \sum_s \langle \Psi | s \rangle \langle s | H | \Psi \rangle = \sum_s |\langle \Psi | s \rangle|^2 \frac{\langle s | H | \Psi \rangle}{\langle s | \Psi \rangle}$$

Let s'_{ij} be s with spins i and j flipped.

$$\frac{\langle s | H | \Psi \rangle}{\langle s | \Psi \rangle} = \sum_{\langle ij \rangle \in s} \begin{cases} 1/4 & \text{if } s_i = s_j \\ -1/4 + 1/2 \frac{\langle s'_{ij} | \Psi \rangle}{\langle s | \Psi \rangle} & \text{if } s_i \neq s_j \end{cases}$$

If we can choose the sign of Ψ so this is always negative, this is the best we can do.

Even with J2, this minimizes the J1 energy.

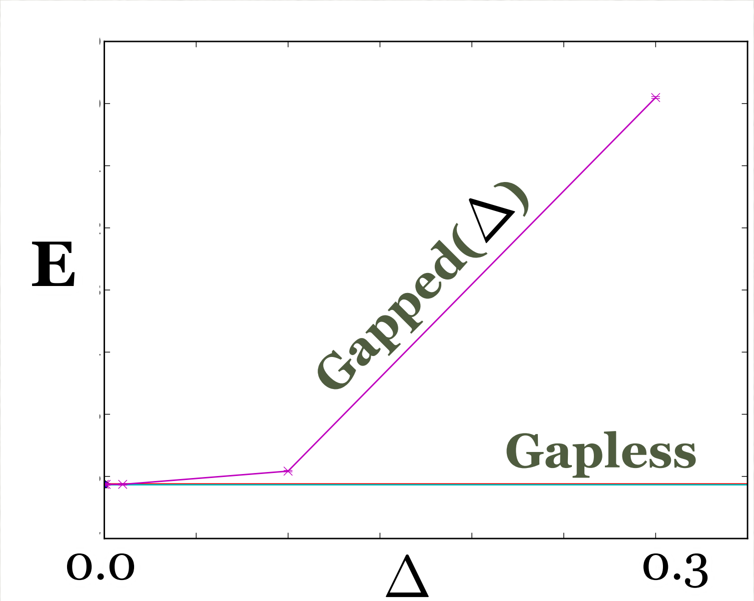
How do some spin liquids stack up?

<u>Spin Liquid C:</u>

Obeys the Marshall sign rule: J1

Zero weight on AA singlets: J2

$$\phi(r_{iA}, r_{jA}) = 0$$

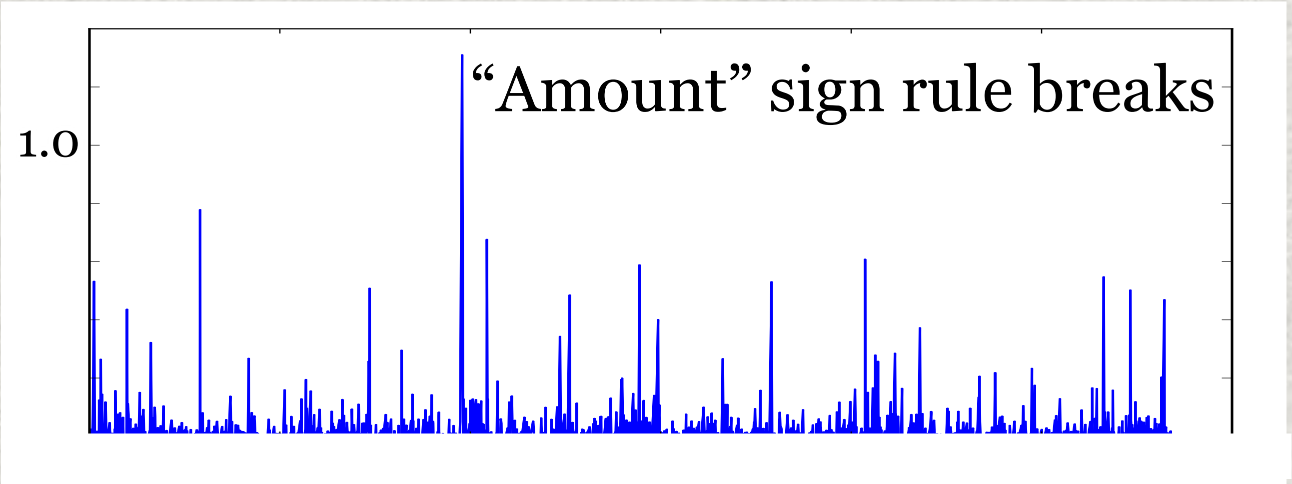


<u>Spin Liquid B</u>:

Breaks Marshall sign rule: J1

Weight on AA singlets: J2

$$\phi(r_{iA}, r_{jA}) \neq 0$$



MC Time

Can we get a win/win?

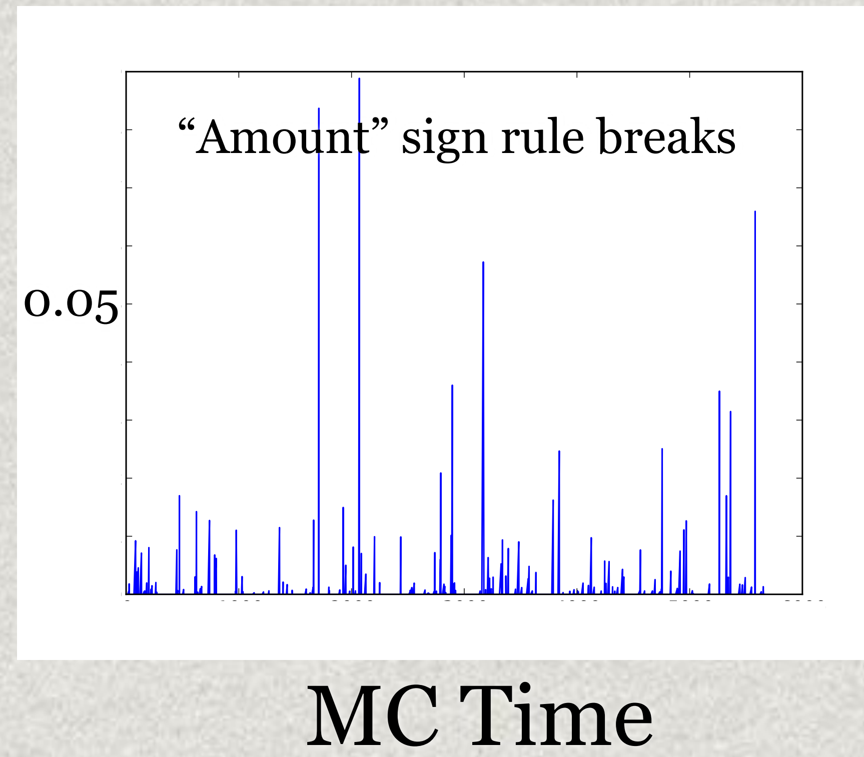
J2/J1=0.1
N=10

Spin Liquid A:

For the right parameter choice:

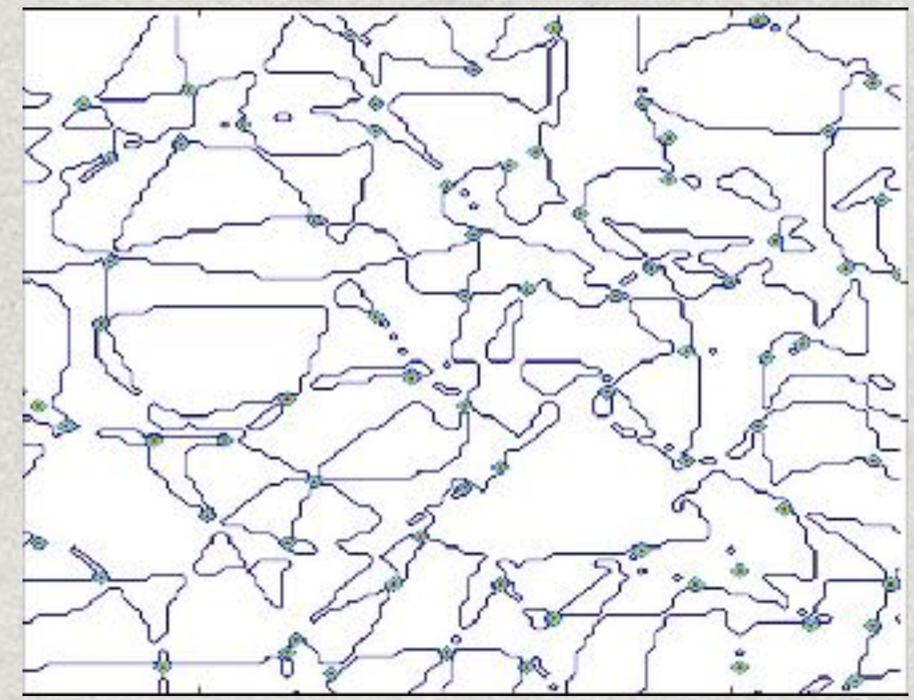
"Obeys" Marshall sign rule: J1 ☺

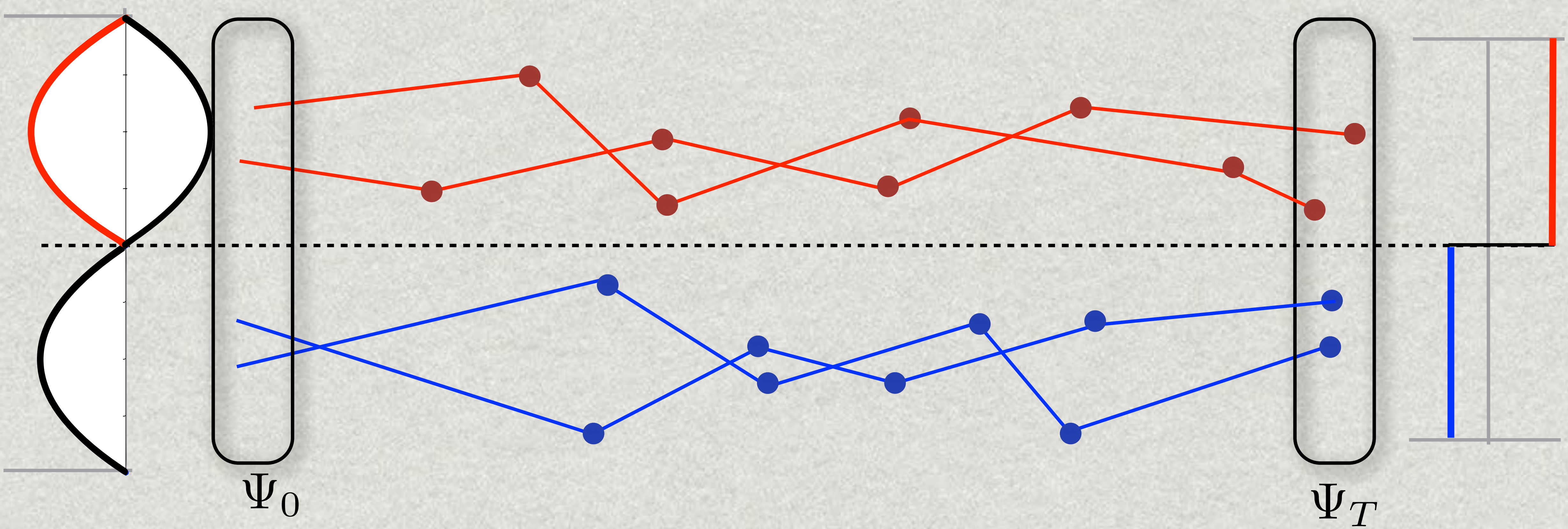
Weight on AA singlets: J2 ☺



The winning spin liquid is good both because of energetics and for qualitative reasons!!

Why “nothing” matters.





Herbertsmithite

Which of the many spin liquid phases is it in?

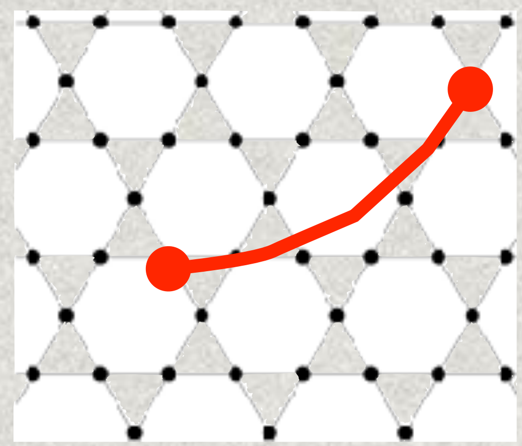
Spin Liquids on the kagome.

Spin Liquid A

?

Spin Liquid E

?



?

Spin Liquid B

?

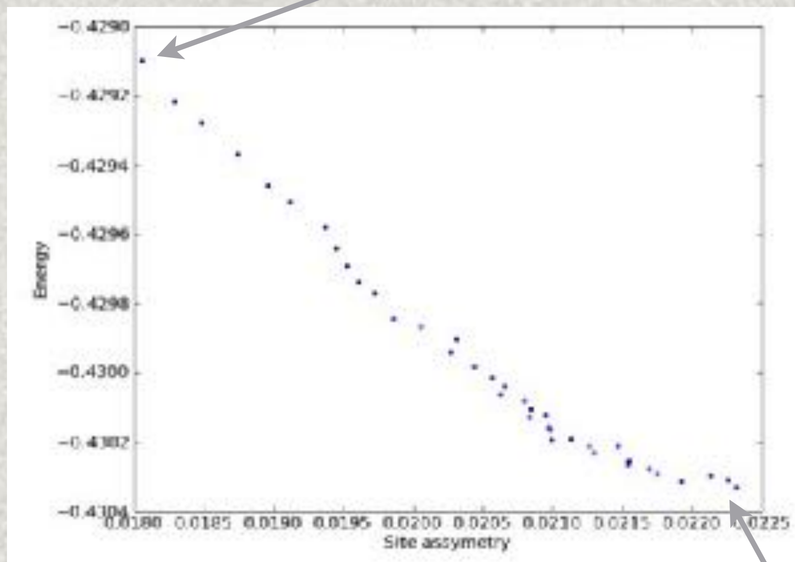
?

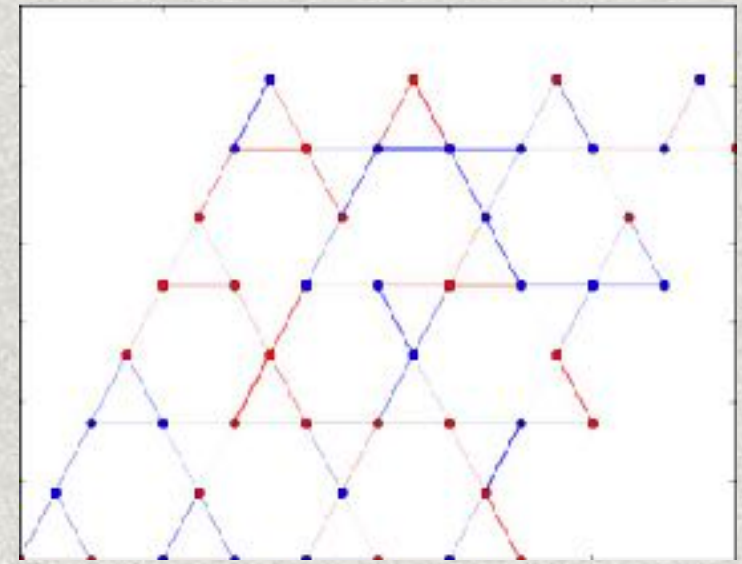
Spin Liquid D

Spin Liquid C

but there's a problem ...

spin liquid C





our state.

- we've found an instability to this spin liquid.[**]

  (which isn't even a spin liquid).

- and DMRG[*] finds a spin liquid which isn't spin liquid C.

Do any of the spin liquids look like the one found in DMRG?

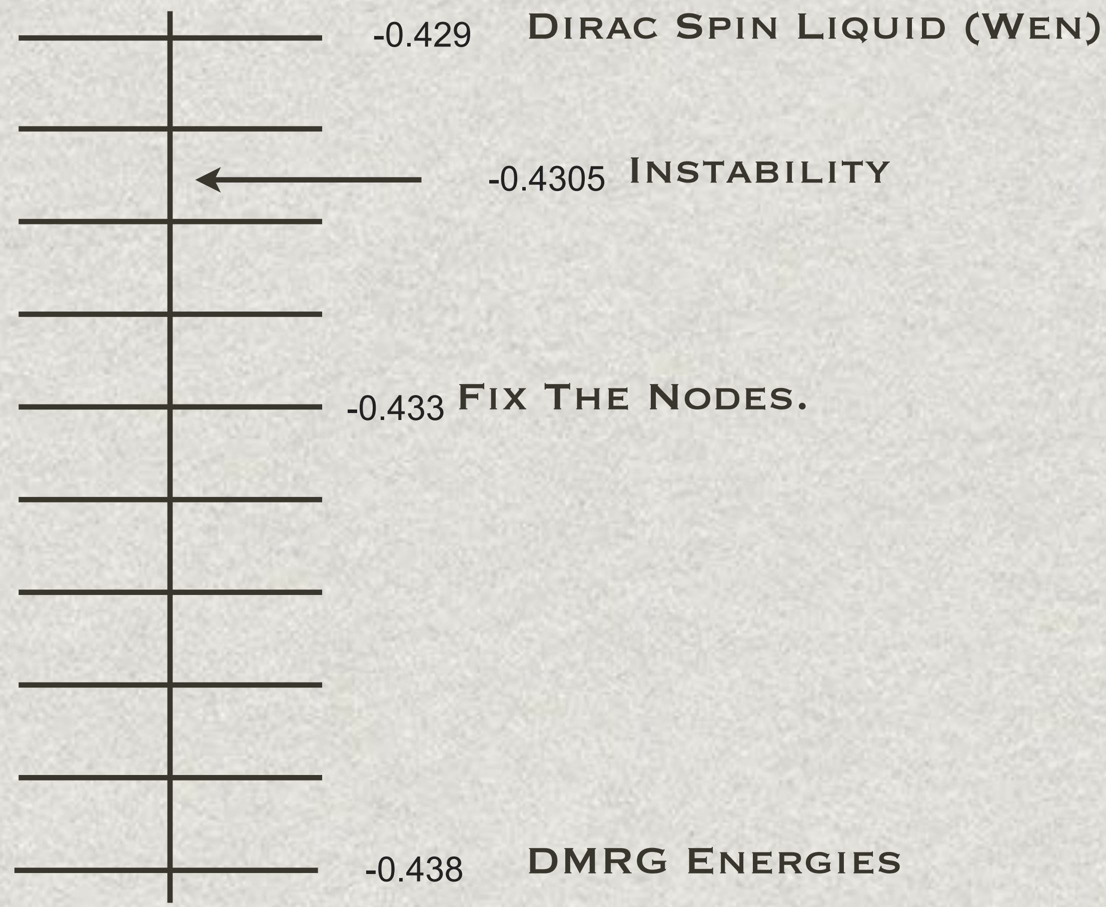
[*] Yan, White, Huse: Science, Jun (2012)

[**] Clark, Kinder, Lawler, Neuscamann, Chan, In Preparation

A comparison

Allowed kagome spin liquids

~~Spin Liquid A~~
~~Spin Liquid B~~
~~Spin Liquid C~~
~~Spin Liquid D~~
~~Spin Liquid E~~
~~Spin Liquid F~~
~~Spin Liquid G~~
~~Spin Liquid H~~
~~Spin Liquid I~~
~~Spin Liquid J~~
~~Spin Liquid K~~
~~Spin Liquid L~~
~~Spin Liquid M~~
~~Spin Liquid N~~
~~Spin Liquid O~~
~~Spin Liquid P~~
~~Spin Liquid Q~~



Maybe it's just the nodes.

Numerics has shown that 20 years of theory on the spin liquids appear to be broken. This is very exciting!

and we are one step closer to understanding ....

Conclusions

There are hundreds of different spin liquid phases. They probably make up most the phases in the world!

Using quantum Monte Carlo techniques, we are just beginning to find them in real materials! We've gone from a theorist playground to something real.

Something about the theory of these systems is broken! Room for new theoretical approaches that combine numerics and analytics.

Exciting time to be doing condensed matter!