An introduction to quantum spin liquids
Part I

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1. Introduction and definitions
   - Which spin models are we taking about?
   - The classical limit
   - "Moderate" quantum fluctuations
   - Absence of magnetic order
   - Mechanisms to destroy the long-range order

2. Quantum spin liquids: general definitions and properties
   - A first definition for spin liquids
   - Valence-bond crystals
   - A second definition for spin liquids
   - Quantum paramagnets
   - The Lieb-Schultz-Mattis et al. theorem
   - The short-range RVB picture
   - A third definition for spin liquids
   - Fractionalization

From Hubbard to Heisenberg

- Zero temperature $T = 0$
- Correlated electrons on the lattice

The starting point is the Hubbard model:

$$\mathcal{H} = -\sum_{i,j,\sigma} t_{i,j} c^{\dagger}_{i,\sigma} c_{j,\sigma} + h.c. + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow}$$

At half-filling (i.e., $N_e = N_s$) for $U \gg t$, an insulating state exists
For $U/t \to \infty$, by perturbation theory, we obtain the Heisenberg model:

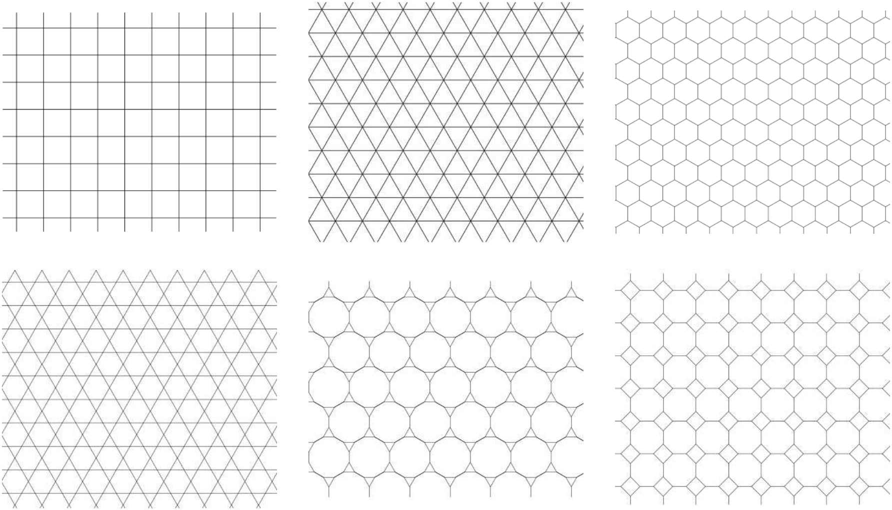
$$\mathcal{H} = \sum_{i,j} J_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j,k,l} (P_{i,j,k,l} + h.c.) + \dots$$

- Spin SU(2) symmetric models

> Here, I will discuss **spin models** (frozen charge degrees of freedom)
> Spin liquids in the Hubbard model (with also charge fluctuations)
> are possible, but much harder to detect

Some example for the lattice structure

Two-dimensional lattices



Simple considerations for classical spins

We want to find the lowest-energy spin configuration for classical spins
Consider the case of Bravais lattices (i.e., one site per unit cell)

$$E[\{\mathbf{S}_i\}] = \frac{1}{2} \sum_i \sum_r J(r) \mathbf{S}_i \cdot \mathbf{S}_{i+r}$$

with the *local* constraint $\mathbf{S}_i^2 = 1$
By Fourier transform:

$$E = \frac{1}{2} \sum_k J(k) \mathbf{S}_k \cdot \mathbf{S}_{-k}$$

Look for solutions with the *global* constraint: $\sum_i \mathbf{S}_i^2 = N \longrightarrow \sum_k \mathbf{S}_k \cdot \mathbf{S}_{-k} = N$

Assume $J(k)$ minimized for $k = k_0$

Take $\mathbf{S}_k = 0$ for all k's except for $k = \pm k_0$

$$\mathbf{S}_{k_0} = \frac{\sqrt{N}}{2} \begin{pmatrix} 1 \\ i \\ 0 \end{pmatrix} \qquad \mathbf{S}_{-k_0} = \mathbf{S}_{k_0}^* = \frac{\sqrt{N}}{2} \begin{pmatrix} 1 \\ -i \\ 0 \end{pmatrix}$$

$$\mathbf{S}_i = \frac{1}{\sqrt{N}} \left(\mathbf{S}_{k_0} e^{i k_0 r_i} + h.c. \right) = \{\cos(k_0 r_i), \sin(k_0 r_i), 0\}$$

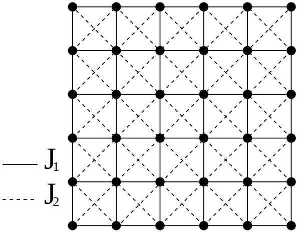
The *local* constraint is automatically satisfied!

Spiral configuration (in general non-collinear – coplanar)

Example: Classical $J_1 - J_2$ model on the square lattice

$J(k) = 2J_1 (\cos k_x + \cos k_y) + 4J_2 \cos k_x \cos k_y$

- For $J_2/J_1 < 1/2$, $k_0 = (\pi, \pi)$
- For $J_2/J_1 > 1/2$, $k_0 = (\pi, 0)$ or $(0, \pi)$
  The two sublattices are decoupled
  (free angle between spins in A and B sublattices)
- For $J_2/J_1 = 1/2$, $k_0 = (\pi, k_y)$ or (k_x, π)
  highly-degenerate ground state:
  $\mathcal{H} = \text{const.} + \sum_{\text{plaquettes}} (\mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4)^2$



Quantum fluctuations

In order to include the quantum fluctuations, perform a $1/S$ expansion

$$\mathcal{H} = \sum_{i,j} J_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j$$

- Let us denote by $\theta_j = k_0 \cdot r_j$
- Make a rotation around the z axis

$$\begin{cases} \tilde{S}_j^x = \cos\theta_j S_j^x + \sin\theta_j S_j^y \\ \tilde{S}_j^y = -\sin\theta_j S_j^x + \cos\theta_j S_j^y \\ \tilde{S}_j^z = S_j^z \end{cases}$$

- Perform the Holstein-Primakoff transformations:

$$\begin{cases} \tilde{S}_j^x = S - a_j^\dagger a_j \\ \tilde{S}_j^y \simeq \sqrt{\frac{S}{2}} \left(a_j^\dagger + a_j \right) \\ \tilde{S}_j^z \simeq i \sqrt{\frac{S}{2}} \left(a_j^\dagger - a_j \right) \end{cases}$$

Quantum fluctuations

At the leading order in $1/S$, we obtain:

$$\mathcal{H}_{\rm sw} = \mathrm{E}_{\rm cl} + \frac{S}{2} \sum_k \left\{ A_k a_k^\dagger a_k + \frac{B_k}{2} \left(a_k^\dagger a_{-k}^\dagger + a_{-k} a_k \right) \right\}$$

Where:

$$\mathrm{E}_{\rm cl} = \frac{1}{2} N S^2 J_{k_0}$$

$$\begin{cases} A_k = J_k + \frac{1}{2}(J_{k+k_0} + J_{k-k_0}) - 2J_{k_0} \\ B_k = \frac{1}{2}(J_{k+k_0} + J_{k-k_0}) - J_k \end{cases}$$

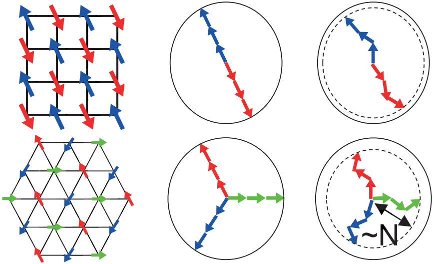
By performing a Bogoliubov transformation:

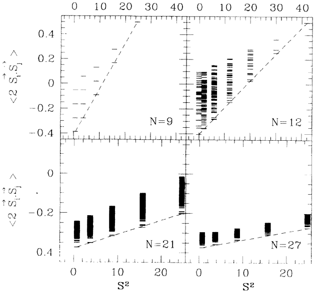
$$\boxed{\mathcal{H}_{sw} = \mathrm{E}_{\rm cl} + \sum_k \omega_k (\alpha_k^\dagger \alpha_k + \tfrac{1}{2})}$$

- Zero-point quantum fluctuations
- Leading-order corrections to the magnetization $\langle \tilde{S}_j^x \rangle = S - \langle a_j^\dagger a_j \rangle$

"Renormalization" of the classical state

The classical ground state is "dressed" by quantum fluctuations





- The lattice breaks up into sublattices
- Each sublattice keeps an extensive magnetization

- Spontaneously broken SU(2) symmetry
  Goldstone theorem
  Gapless spin waves ($S = 1$)

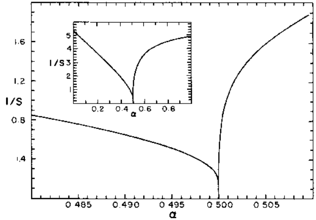
Anderson, Phys. Rev. **86**, 694 (1952)

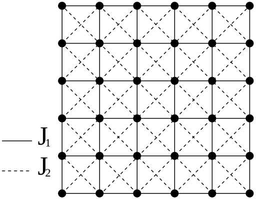
Bernu, Lhuillier, and Pierre, Phys. Rev. Lett. **69**, 2590 (1992)

Bernu, Lecheminant, Lhuillier, and Pierre, Phys. Rev. B **50**, 10048 (1994)

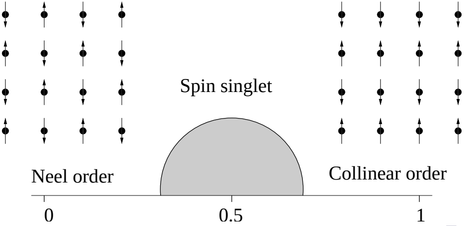
Absence of magnetic order in the strongly frustrated regime

$$\mathcal{H} = \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \alpha \sum_{\langle\langle i,j \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$



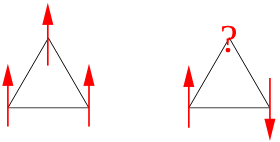


Chandra and Doucot, Phys. Rev. B **38**, 9335 (1988)



We have to stay away from the classical limit

- Small value of the spin S, e.g., $S = 1/2$ or $S = 1$
- Frustration of the super-exchange interactions
  (not all terms of the energy can be optimized simultaneously)



- Low spatial dimensionality
  In $D = 1$ there is no magnetic order, given the Mermin-Wagner theorem
  (not possible to break a continuous symmetry in D=1, even at $T = 0$)
  $D = 2$ is the "best" choice
- [Large continuous rotation symmetry group, e.g., SU(2), SU(N) or Sp(2N)]

Arovas and Auerbach, Phys. Rev. B **38**, 316 (1988); Arovas and Auerbach, Phys. Rev. Lett. **61**, 617 (1988)

Read and Sachdev, Phys. Rev. Lett. **66**, 1773 (1991); Read and Sachdev, Nucl. Phys. **B316**, 609 (1989)

A SL is a state without long-range magnetic order

A spin liquid is a state without magnetic order
the structure factor $S(q)$ does not diverge, whatever the q is

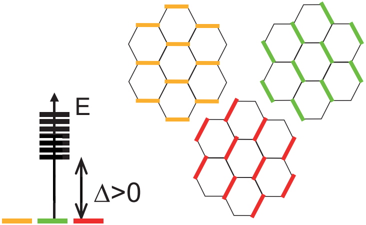
$$S(q) = \frac{1}{N} \langle \Psi_0 | \left| \sum_j \mathbf{S}_j e^{iqr_j} \right|^2 | \Psi_0 \rangle = \frac{1}{N} \sum_{j,k} \langle \Psi_0 | \mathbf{S}_j \cdot \mathbf{S}_k | \Psi_0 \rangle e^{iq(r_j - r_k)}$$

$$S(q) = \begin{cases} O(1) & \text{for all q's} \quad \longrightarrow \text{short-range correlations} \\ S(q_0) \propto N & \text{for} q = q_0 \quad \longrightarrow \text{long-range order} \end{cases}$$

- Can be checked by using Neutron scattering

- Mermin-Wagner theorem implies that *any* 2D Heisenberg model at $T > 0$ is a SL according to this definition

A SL is a state without long-range magnetic order

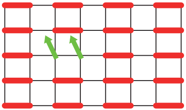


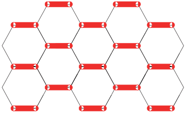
$J_1 - J_2$ Heisenberg model on the hexagonal lattice

Fouet, Sindzingre, and Lhuillier, Eur. Phys. J. B **20**, 241 (2001)

Properties:

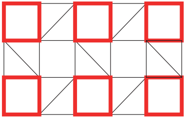
- Short-range spin-spin correlations
- Spontaneous breakdown of some lattice symmetries → ground-state degeneracy
- Gapped $S = 1$ excitations ("magnons" or "triplons")





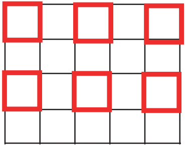
$J_1\!-\!J_2$ model

Fouet, Sindzingre, and Lhuillier, EPJB (2001)



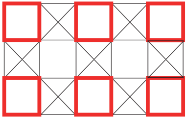
Shastry-Sutherland lattice

Koga and Kawakami, PRL (2000)



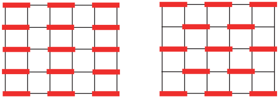
$J_1\!-\!J_2\!-\!J_3$ model

Mambrini, Lauchli, Poilblanc, and Mila, PRB (2006)



Heisenberg model on the Checkerboard lattice

Fouet, Mambrini, Sindzingre, and Lhuillier, PRB (2003)



Heisenberg model with a 4-spin ring exchange

Lauchli, Domenge, Lhuillier, Sindzingre, and Troyer, PRL (2005)

+ others...

Spin liquid: a second definition

A spin liquid is a state without any spontaneously broken (local) symmetry

- This definition rules out magnetically ordered states that break spin SU(2) symmetry (also NEMATIC states)

- This definition rules out valence-bond crystals that break some lattice symmetries

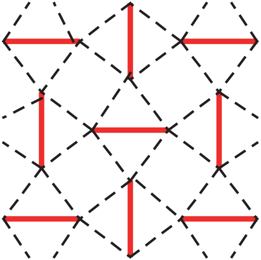
Remark I: "local" means that there is a *physical* order parameter
that can be measured by some local probe

Remark II: within this definition we also rule out CHIRAL SLs
that break time-reversal symmetries

Wen, Wilczek, and Zee, Phys. Rev. B **39**, 11413 (1989)

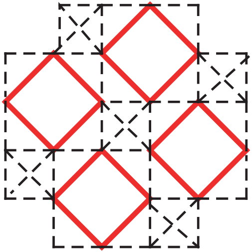
Quantum paramagnets

There are few examples of magnetic insulators without any broken symmetry



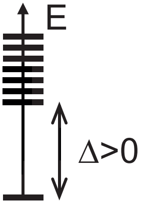
$SrCu_2(BO_3)_2$

Kageyama et al., Phys. Rev. Lett. **82**, 3168 (1999)



CaV_4O_9

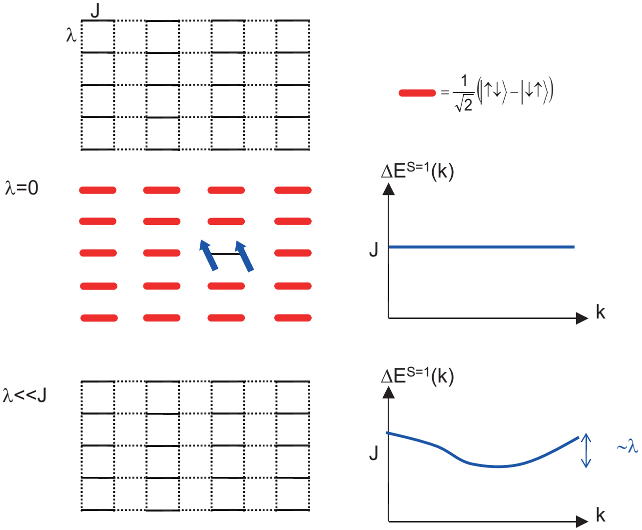
Taniguchi et al., J. Phys. Soc. Jpn. **64**, 2758 (1995)



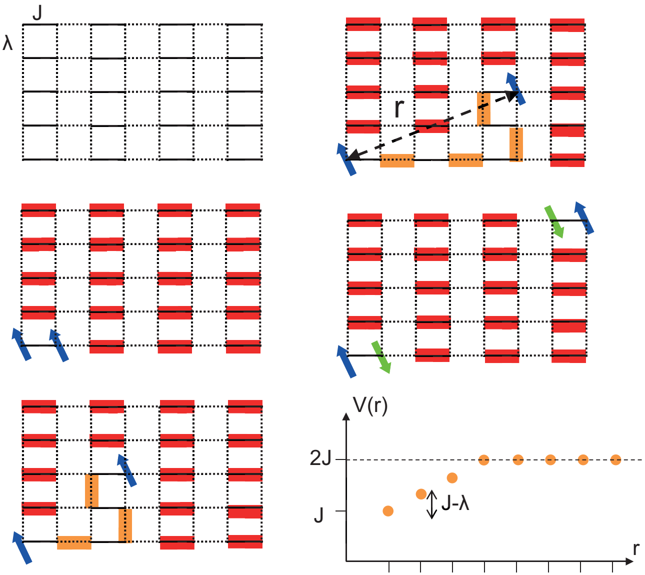
Non-degenerate ground state

Properties:

- No broken symmetries
- Even number of spin-1/2 in the unit cell
- Adiabatically connected to the (trivial) limit of decoupled blocks
- No phase transition between $T = 0$ and $T = \infty$
  $\rightarrow$ "simple" quantum paramagnet at $T = 0$


J
λ
$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$
λ=0
$\Delta E^{S=1}(k)$
J
k
λ<<J
$\Delta E^{S=1}(k)$
J
~λ
k

Quantum paramagnets and VBCs are not fractionalized



A system with half-odd-integer spin in the unit cell cannot have a gap and a unique ground state

Valid in the thermodynamic limit for periodic boundary conditions and

$$L_1 \times L_2 \times \cdots L_D = odd$$

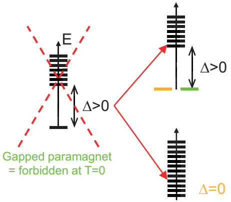
- The original theorem by Lieb, Schultz, and Mattis refers to 1D

Lieb, Schultz, Mattis, Ann. Phys. (N.Y.) **16**, 407 (1961); see also, Affleck and Lieb, Lett. Math. Phys. **12**, 57 (1986)

- Since then, several attempts to generalize it in 2D

Affleck, Phys. Rev. B **37**, 5186 (1988); Bonesteel, Phys. Rev. B **40**, 8954 (1989);

Oshikawa, Phys. Rev. Lett. **84**, 1535 (2000); Hastings, Phys. Rev. B **69**, 104431 (2004)



Case 1) Ground-state degeneracy
a) Valence-bond crystal
b) Resonating-valence bond SL
(gapped but with a topological degeneracy)
Case 2) Gapless spectrum
a) Continuous broken symmetry (magnetic order)
b) Resonating-valence bond SL
(gapless, i.e., critical state)

Proof of the Lieb-Shultz-Mattis theorem for the Heisenberg chain

- Consider the Heisenberg model on a chain:

$$\mathcal{H} = \sum_{i=1}^{N} \mathbf{S}_i \cdot \mathbf{S}_{i+1}$$

with periodic boundary conditions ($\mathbf{S}_{N+1} \equiv \mathbf{S}_1$), even N, and half-odd integer spins

Theorem:

There exists an excited state with an energy that vanishes as $N \to \infty$

- $|\Psi_0\rangle$ is the ground state of $\mathcal{H}$ with energy E_0.
- Assume that $|\Psi_0\rangle$ is a singlet ("almost" always the case)
- Consider the twist operator $\mathcal{O} = \exp\{\frac{2\pi i}{N} \sum_{j=1}^{N} j S_j^z\}$
- Denote $|\Psi_1\rangle = \mathcal{O}|\Psi_0\rangle$

Then:

(1) $\langle \Psi_1 | \Psi_0 \rangle = 0$
(2) $\lim_{N\to\infty} [\langle \Psi_1 | \mathcal{H} | \Psi_1 \rangle - E_0] = 0$

Consider the translation operator $\mathcal{T}$:

$$\mathcal{T}\mathbf{S}_j\mathcal{T}^{-1} = \mathbf{S}_{j+1} \qquad \mathcal{T}\mathbf{S}_N\mathcal{T}^{-1} = \mathbf{S}_1$$

$$[\mathcal{H},\mathcal{T}] = 0 \qquad \mathcal{T}|\Psi_0\rangle = e^{ik_0}|\Psi_0\rangle$$

$\langle\Psi_0|\Psi_1\rangle = \langle\Psi_0|\mathcal{O}|\Psi_0\rangle = \langle\Psi_0|\mathcal{T}\mathcal{O}\mathcal{T}^{-1}|\Psi_0\rangle$

$\mathcal{T}\mathcal{O}\mathcal{T}^{-1} = \mathcal{O}\exp\left(2\pi i S_1^z\right)\exp\left(-\frac{2\pi i}{N}S^z_{\rm tot}\right)$

Then, $\exp\left(-\frac{2\pi i}{N}S^z_{\rm tot}\right)|\Psi_0\rangle = |\Psi_0\rangle$, since $|\Psi_0\rangle$ is a singlet.

$$\exp\left(2\pi i S_1^z\right) = \begin{cases} +1 & S = 0,1,2,\cdots \\ -1 & S = 1/2, 3/2, 5/2, \cdots \end{cases}$$

- Therefore, for half-odd integer spin: $\langle\Psi_0|\Psi_1\rangle = -\langle\Psi_0|\Psi_1\rangle$

$\langle\Psi_1|\mathcal{H}|\Psi_1\rangle = E_0 + \langle\Psi_0|\{\cos(\frac{2\pi}{N}) - 1\}\sum_{j=1}^N (S_j^x S_{j+1}^x + S_j^y S_{j+1}^y)|\Psi_0\rangle$

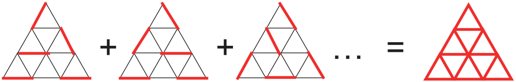
$\langle\Psi_0|(S_j^x S_{j+1}^x + S_j^y S_{j+1}^y)|\Psi_0\rangle \leq S^2$

- We obtain an upper-bound for the energy: $\langle\Psi_1|\mathcal{H}|\Psi_1\rangle - E_0 \leq \frac{2\pi^2 J S^2}{N} + O(N^{-3})$

- Anderson's idea: the short-range resonating-valence bond (RVB) state:

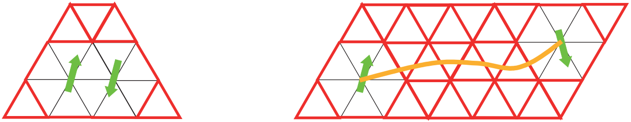
  Anderson, Mater. Res. Bull. **8**, 153 (1973)

  Linear superposition of many (an exponential number) of valence-bond configurations

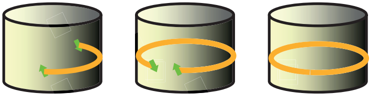


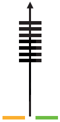
Spatially uniform state

- Spin excitations? No dimer order → we may have deconfined spinons



- Spinon fractionalization and topological degeneracy





Distinct ground states that are not connected by any local operator

Wen, Phys. Rev. B **44**, 2664 (1991); Oshikawa and Senthil, Phys. Rev. Lett. **96**, 060601 (2006)

Spin liquid: a third definition

A spin liquid is a state without any spontaneously broken (local) symmetry, with a half-odd-integer spin in the unit cell

- This definition rules out magnetically ordered states that break spin SU(2) symmetry (also NEMATIC states)

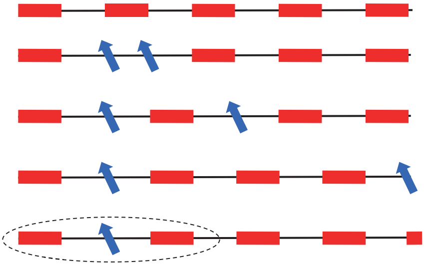
- This definition rules out valence-bond crystals that break some lattice symmetries

- This definition rules out quantum paramagnets that have an even number of spin-half per unit cell

A spin liquid sustains fractional (spin-1/2) excitations

What is fractionalization?

- Majumdar-Ghosh chain (1D): $\mathcal{H} = J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} + \frac{J}{2} \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2}$
- The exact ground state is known (two-fold degenerate), perfect dimerization



The "initial" $S=1$ excitation can decay into two spatially separated spin-1/2 excitations (spinons)

Finite-energy state with an isolated spinon (the other is far apart) domain wall between two dimerization patterns

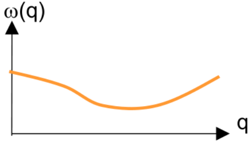
- A spinon is a neutral spin-1/2 excitation, "one-half" of a $S=1$ spin flip. (it has the same spin as the electron, but no charge)
- Spinons can only be created by pairs in finite systems
  The question is to understand whether they can propagate at large distances, as two elementary particles

Inelastic neutron scattering: spinon continuum

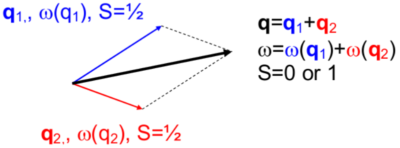
The inelastic neutron scattering is a probe for the dynamical structure factor

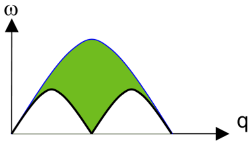
$$S(q,\omega) = \int dt \langle \Psi_0 | S^-_{-q}(t) S^+_q(0) | \Psi_0 \rangle e^{-i\omega t}$$

- The elementary excitations are spin-1 magnons: $S(q,\omega)$ has a single-particle pole at $\omega = \omega(q)$



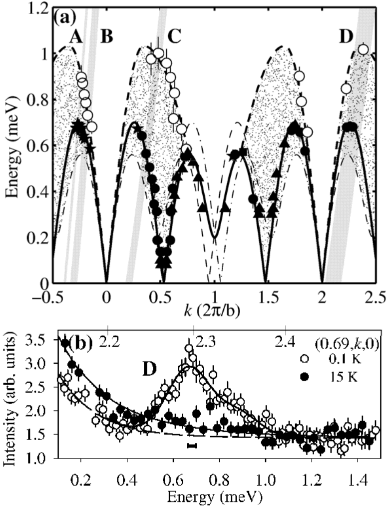
- The spin-flip decays into two spin-1/2 excitations $S(q,\omega)$ exhibits a two-particle continuum





Neutron scattering on Cs_2CuCl_4

Coldea, Tennant, Tsvelik, and Tylczynski, Phys. Rev. Lett. **86**, 1335 (2001)



Almost decoupled layers

Strongly-anisotropic triangular lattice

$J' \simeq 0.33J$: **quasi-1D**