# Quantum Spin Liquids



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# Outline

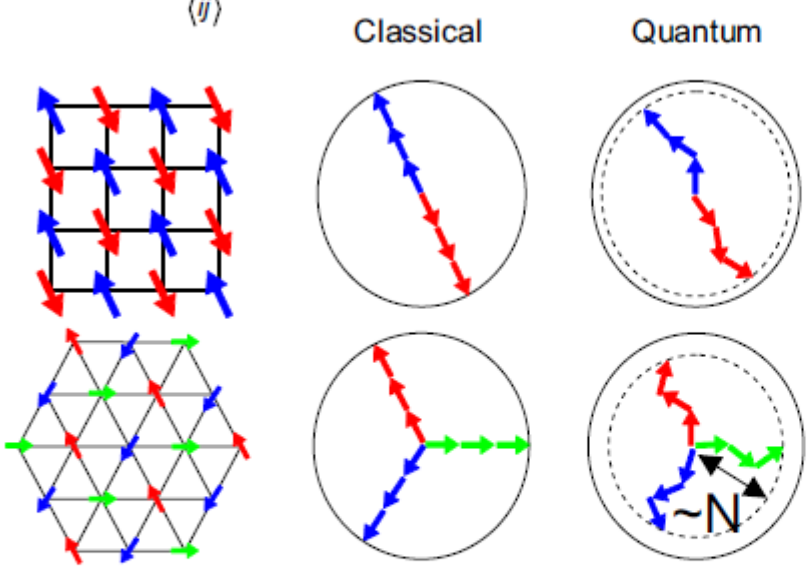


- Neel order and quantum fluctuations.
- 3 definitions for quantum spin liquid
- Fractional excitations
- QSL in real world
- Search for spin liquid on honeycomb lattices.

# Neel order



$$H = J\sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j$$




Anderson, PR 1953
Bernu *et al.*, PRL 1992, PRB 1994
Lhuillier, cond-mat/0502464

- The lattice breaks up in sub-lattices
- Spontaneously broken SU(2) symmetry
  - Goldstone theorem
  - ⇒ Gapless spin waves ($\Delta S^z=1$)
- The classical ground-state is "dressed" by **zero-point fluctuations**. But each sub-lattice keeps an **extensive magnetization**
- Possible description using a "1/S" expansion

What happens if quantum fluctuations are strong enough to destroy the magnetic order ?

# The effect of quantum fluctuations



- For small spins (i.e $S = \frac{1}{2}$), spin fluctuation due to quantum mechanical uncertainty principle are comparable with the size of the spin itself.

- This fluctuations persist down to absolute zero and may melt any order which results in quantum spin liquid .

- Quantum fluctuations are similar to thermal ones, however they can be phase coherent.

# Mechanisms to destroy magnetic long-range order



1-Low spatial dimension

2-Low Hilbert dimension (small spin)

3-Low coordination number (number of nearest neighbors)

4-Frustration

5-Big (continuous) spin symmetry group (i.e SU(2), U(1))

# Three Definitions for QSL



1- QSL is a state with no magnetic long-range order at $T = 0$ .

2- QSL is a state with no spontaneously broken symmetry at $T = 0$.

3-QSL is a state with neutral spin-1/2 excitations (spinons).

- QSL is a superposition state in which the spins simultaneously point in many different directions , so the spins are highly entangled with one another.
- To have QSL ground state, a material must have a large frustration factor ( say $f \rangle 100$) .

# Quantum Spin Liquid: Definition.1



*A spin liquid is a state without magnetic long-range order*

- More precisely, the structure factor S(**q**) never diverges, whatever **q**.

$$S(\mathbf{q}) = \frac{1}{N}\langle 0 \left| \sum_i \vec{S}_i \exp(i\mathbf{q}\cdot\mathbf{r}_i) \right|^2 |0\rangle$$

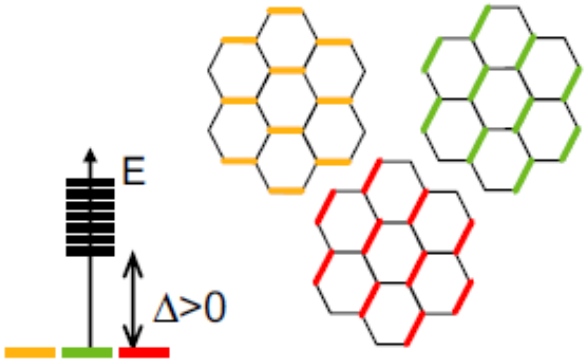
$$= \frac{1}{N}\sum_{ij}\langle 0|\vec{S}_i\cdot\vec{S}_j|0\rangle \exp(i\mathbf{q}\cdot(\mathbf{r}_i-\mathbf{r}_j))$$

$$= \begin{cases} \approx O(1)\ \forall \mathbf{q} \Leftrightarrow \text{short-range mag. order} \\ \exists \mathbf{q}_0 \,/\, S(\mathbf{q}_0) \approx O(N) \Leftrightarrow \text{long-range mag. order} \end{cases}$$

- Can be checked using neutron scattering. But also, μ-SR, NMR, …
- Mermin-Wagner theorem ⇒ *any* 2D Heisenberg model at T>0 is a S.L. according to this def. ☹

# Valence Bond Crystals (VBC)






$J_1$-$J_2$ Heisenberg model
(hexagonal lattice)
Fouet *et al.* EPJB 2001

$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$ Singlet, total spin S=0

Properties:

- Short-ranged spin-spin correlations
- Spontaneous breakdown of some lattice symmetries
  - ⇒ Ground-state degeneracy
- Gapped ΔS=1 excitations ("magnons" or "triplons")

# Quantum Spin Liquid: Definition.2



*A spin liquid is a state without any spontaneously broken symmetry*

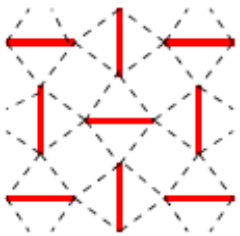
- This def. excludes Néel ordered states, which break the SU(2) sym. (also spin nematics)
- This def. excludes valence-bond crystals, which break some lattice sym.

# Quantum Paramagnets

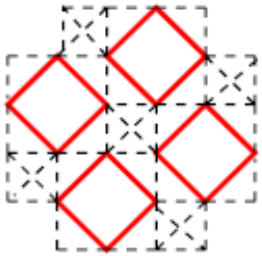


- Some magnetic insulators without any broken sym.

$= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$ S=0 spin singlet, or dimer

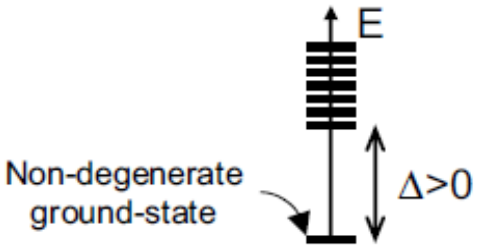


$SrCu_2(BO_3)_2$ Kageyama *et al.* (1999)



$CaV_4O_9$ Taniguchi *et al.* J. Phys. Soc. Jpn (1995)

$\Delta \approx 100$ K - 1st 2D spin-gap system




Other examples: coupled dimer systems: $TlCuCl_3$, **etc.**

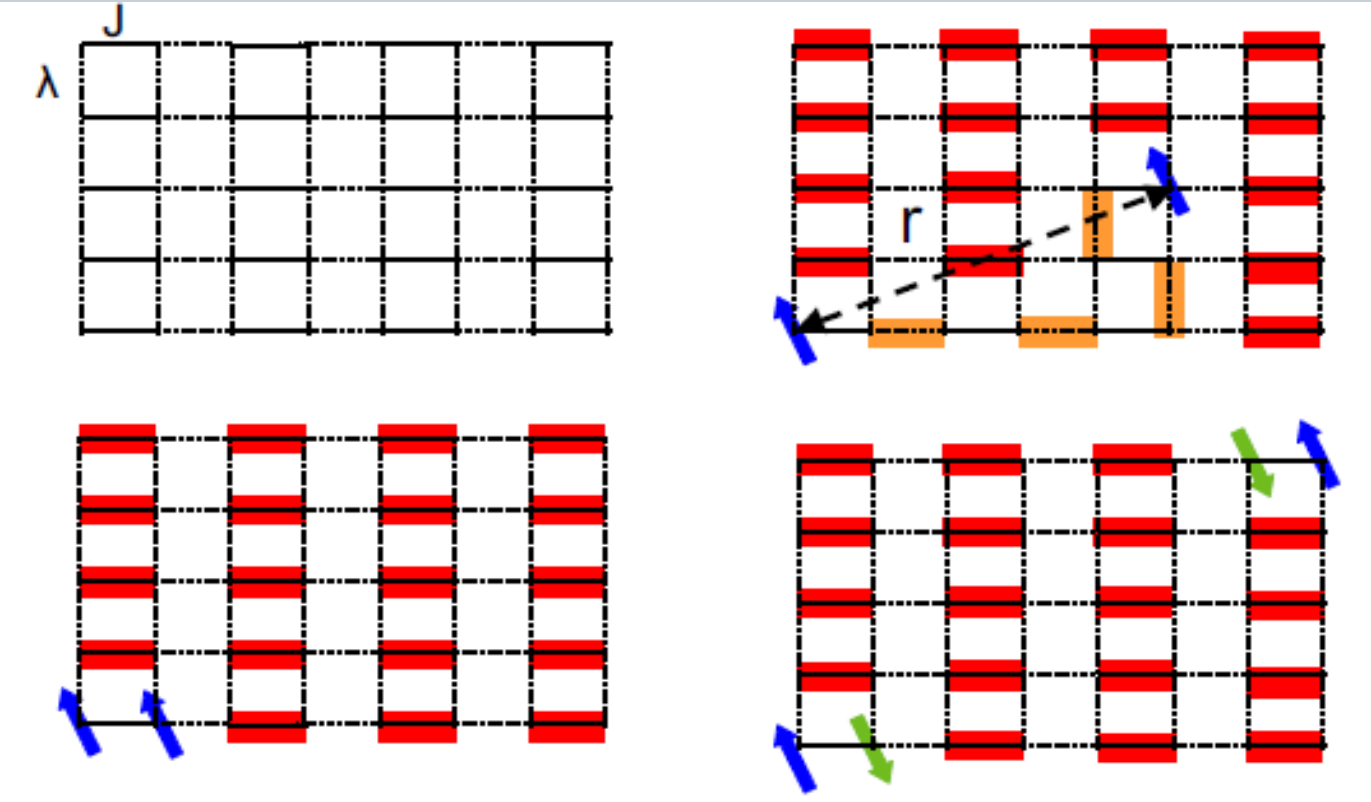
- Properties:
  - **Even number of spin-½ in the crystal unit cell**
  - No broken symmetry
  - Adiabatically connected to the (trivial) limit of *decoupled* blocks
  - No phase transition between T=0 and T=∞

    ⇒ "simple" quantum paramagnet at T=0

# Confinement of Spinons in Quantum Parmagnets and VBC



- Spinons are chargeless S=1/2 excitations. In quantum paramegnets and VBC's spin excitations are not fractionalized.



# What else?



- In a quantum paramagnet, the unit cell contains an ***even*** number of spin-½
- In a VBC, the unit cell is spontaneously enlarged to enclose a ***even*** number of spin-½
- Are there other types of wave-functions with short-range spin-spin correlations ?
(with just *one* spin ½ per unit cell in particular?)

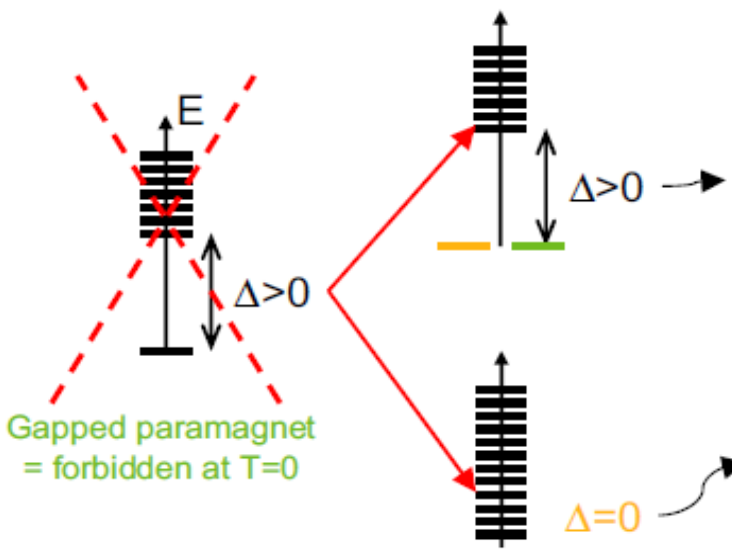
# Lieb-Schultz Mattis-Hasings Theorem



- Lieb-Schultz-Mattis (D=1: 1961) Hastings (D>1: Phys. Rev. B 2004; Europhys. Lett. 2005)

  [See also: Affleck 1988; Bonesteel 1989; Oshikawa 2000; GM *et al. 2002*]

*"A system with a half-odd-integer spin in the unit cell*
*(+ periodic boundary conditions, + dimensions $L_1 \times L_2 \times \ldots \times L_D$ with $L_2 \times \ldots \times L_d$=odd)*
*cannot have a gap and a unique ground-state*
*(in the thermodynamic limit)."*




1) Ground-state degeneracy

a- "Conventional" broken symmetry
(valence bond crystal for instance)

b- Resonating valence bond liquid ($Z_2$)
or more complex topological structure

2) Gapless spectrum

a- Continuous broken sym. (Néel order)

b- Critical state

spin liquids with fractional excitations

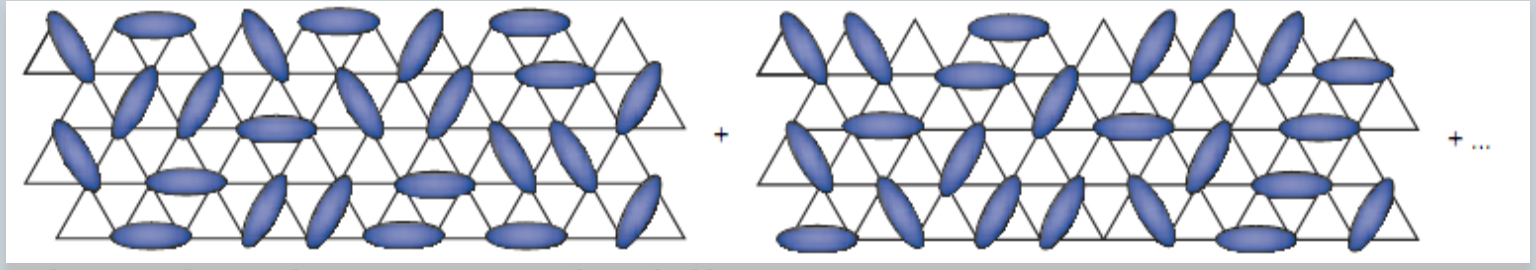
# Resonating Valence Bond (RVB) State



- First proposal : spin-1/2 AF Heisenberg model on a 2D hexagonal lattice.

Fazekas and Anderson (1974) proposed that the ground state wave function can be written as a linear combination of singlet configurations . Each configuration consists of nearest neighbor valence bonds. This state is called "Resonating Valence Bond".

$\Psi_{RVB} =$




Each valence bond represent the following superposition:

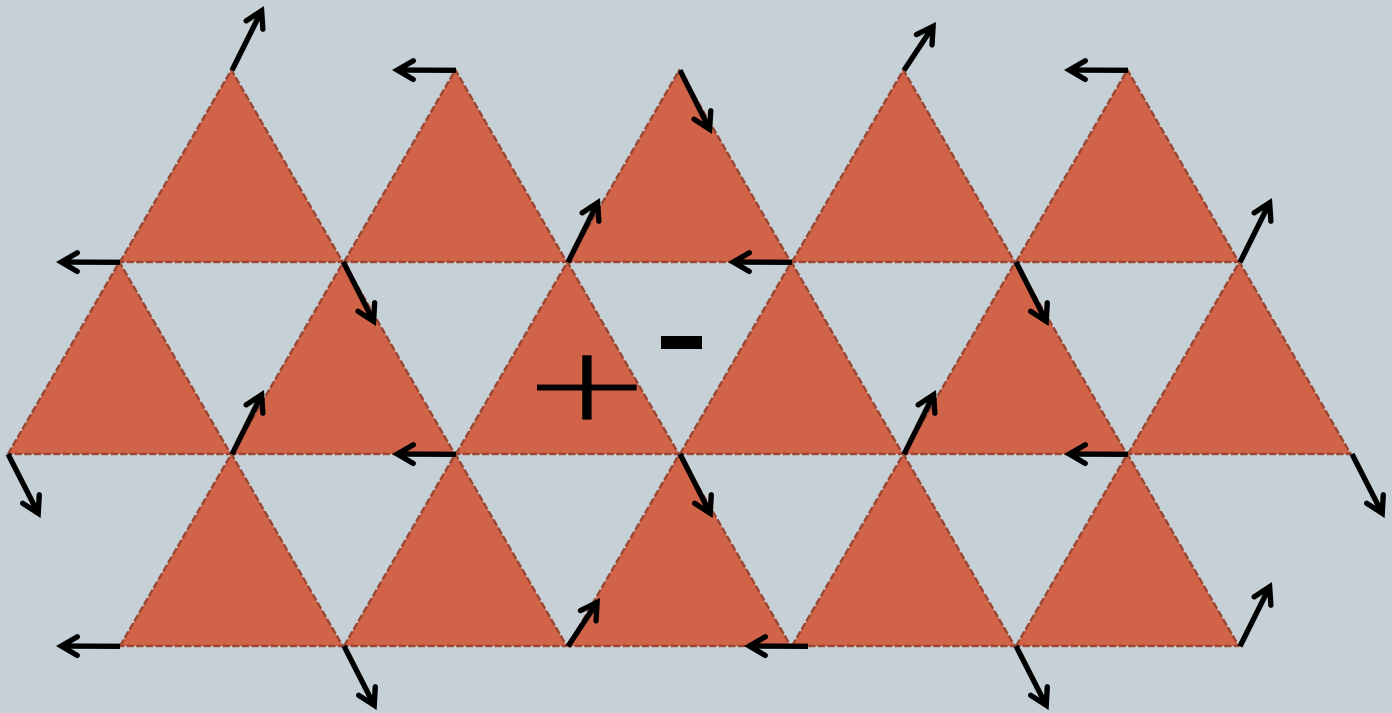
$$(i, j) = \frac{1}{\sqrt{2}}(\alpha_i \beta_j - \beta_i \alpha_j)$$

$$\alpha = |\uparrow\rangle, \beta = |\downarrow\rangle$$

# Ground State



- True ground state is long-range ordered (Chiral Neel order).

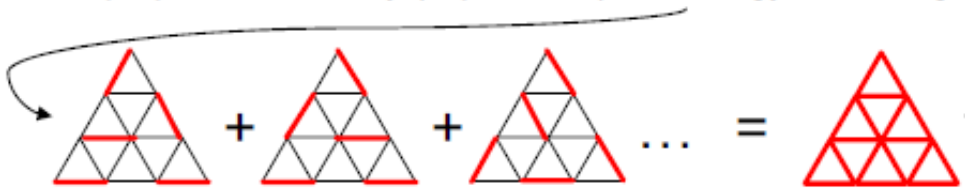



- The quantum fluctuations are not strong enough to destroy ordering.

# Fractionalization in Short-Rnage RVB

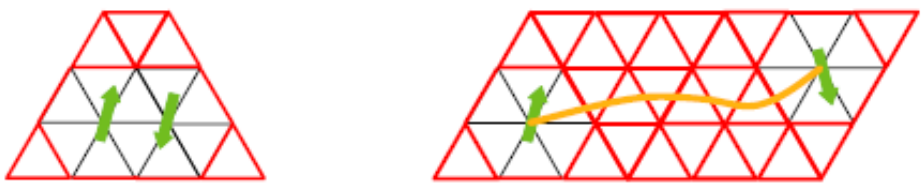


Linear superposition of many (exponential) low-energy short-range valence-bond configurations




Spatially *uniform* state

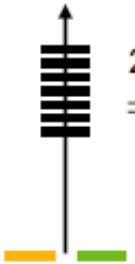
- Spin-½ excitations ?
  VBC ⇒ linear potential between spinons
  no dimer order ⇒ we *may expect* deconfined spinons



- Topological degeneracy & spinon fractionalization

adiabatic process → New ground-state

**Topological degeneracy**
(X. G. Wen 1991) ↔
**fractionalization**
See also Oshikawa & Senthil
PRL 96, 060601 (2006)



2-fold degeneracy
⇒ Satisfies LSMH

# Quantum Spin Liquid: Definition. 3



Definition 3:

*A spin liquid is a state which sustains fractional (spin-½) excitations*

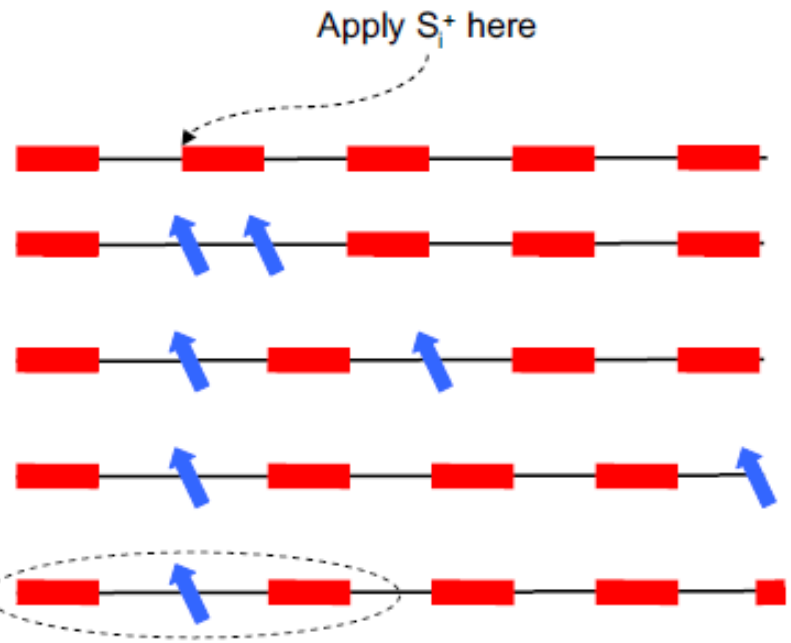
- What is fractionalization ?
  - Existence of (finite energy) excitations with quantum number(s) which are fraction of the elementary degrees of freedom.
    Most famous example: charges q=e/3 in the fractional quantum hall effect.
  - In magnetic systems:
    A spinon is a neutral spin-½ excitation, "one half" of a $\Delta S^z=1$ spin flip.
    (it has the same spin as an electron, but is has no *charge*)
  - Spinons can only be created by pairs in finite systems (combining $S^+$ and $S^-$ operators can only change $S^z$ by some integer) The question is to understand if they then can propagate at large distances from each other, as two elementary particles.

# Fractional Excitation: 1D example



- Majumdar-Gosh chain $H = \sum_i \vec{S}_i \cdot \vec{S}_{i+1} + \frac{1}{2}\sum_j \vec{S}_j \cdot \vec{S}_{j+2}$

The initial $S^z$=+1 excitation can *decay* into *two* spatially separated spin-½ excitations (spinons)




Finite-energy state with an *isolated* spinon (the other is far apart)

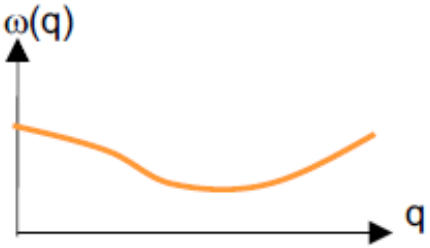
# How to detect spinon excitations?



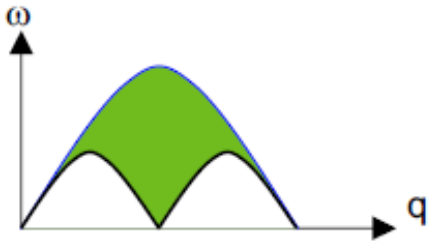
Inelastic neutron scattering : probe for the dynamical structure factor S(**q**,ω).

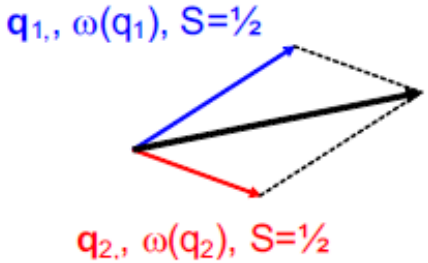
$$S(\mathbf{q},\omega) = \int dt \langle 0 | S^{-}_{-\mathbf{q}}(t) S^{+}_{\mathbf{q}}(0) | 0 \rangle e^{-i\omega t}$$

- If the elementary excitations are spin-1 magnons : S(**q**,ω) has single-particle **pole** at ω= ω(**q**)




- If the spin flip decays into two spin-½ excitations S(**q**,ω) exhibits a two-particle **continuum**

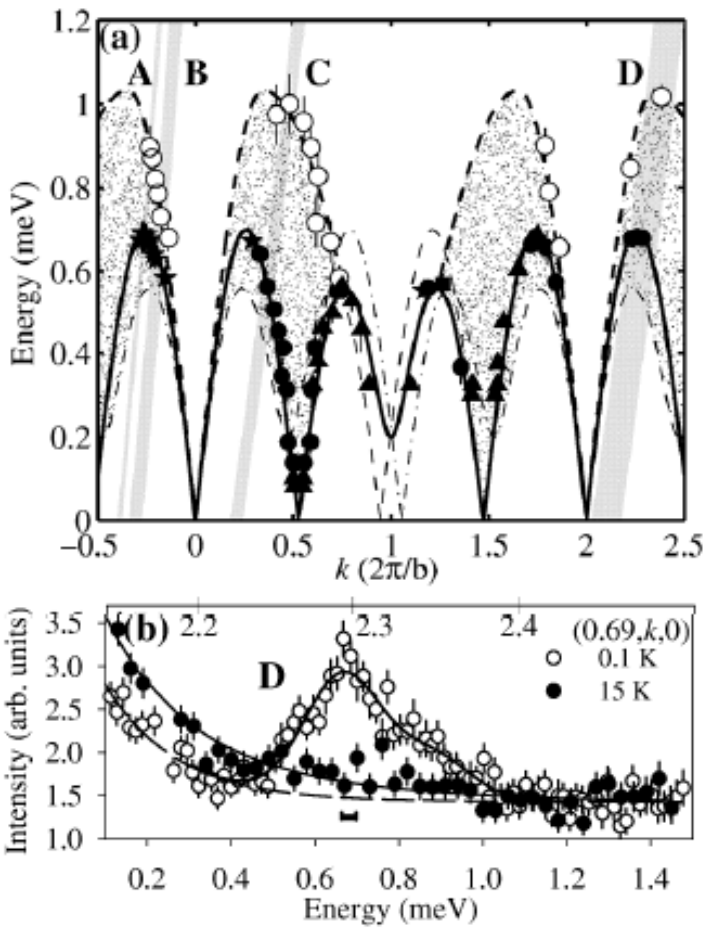


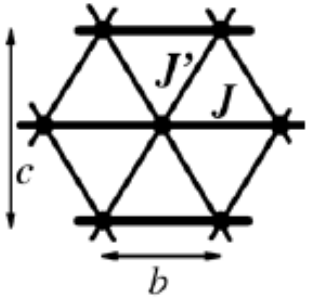

$q = q_1 + q_2$
$\omega = \omega(q_1) + \omega(q_2)$
S=0 or 1

# Real material example






- Neutron scattering on $Cs_2CuCl_4$
  R. Coldea *et al.* (2000)



# Candidates for farctionalized QSL 1n D>1



1)D=2

-Triangular lattice s=1/2 systems,

examples: Organic compounds $(BEDT\text{-}TTF)_2Cu_2(CN)_3$ (ET) $EtMe_3Sb[Pd(dmit)_2]_2$ (dmit)

- Kagome lattice s=1/2 systems,

examples: $ZnCu_3(OH)_6Cl_2$, $BaCu_3V_2O_8(OH)_2$,

$Cu_3V_2O_7(OH)_2.2(H_2O)$

2) D=3 :

Hyper-kagome S=1/2 system

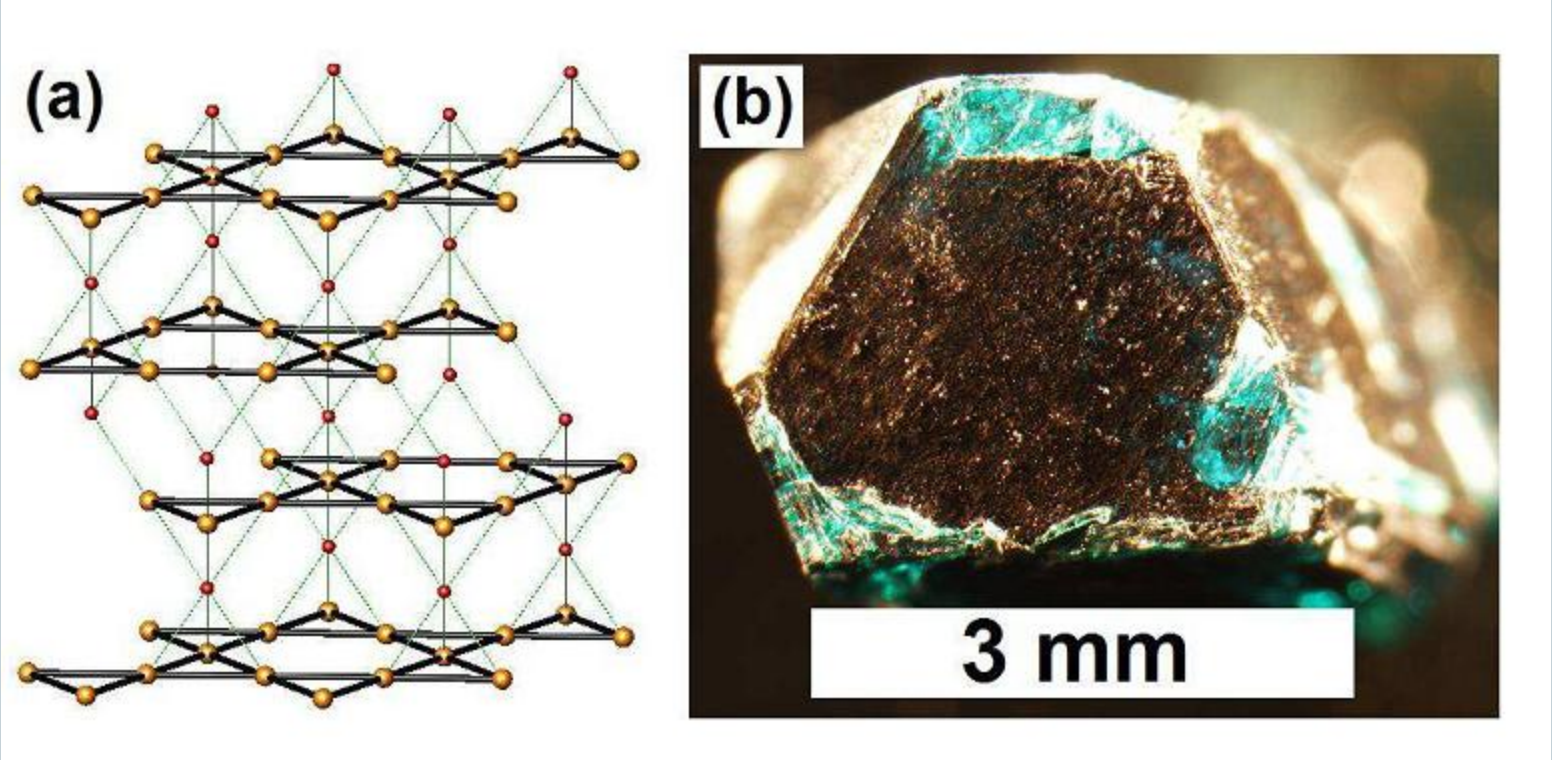
example: $Na_3Ir_4O_8$

# Examples of Real Kagome Magnets



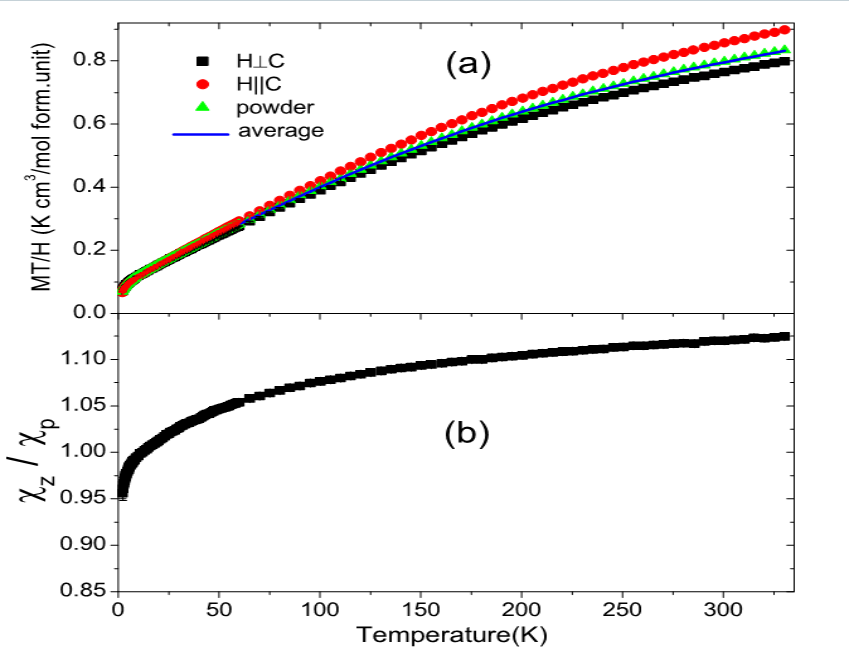
- Herbertsmithite : layers of weakly coupling $s = \frac{1}{2}$ Kagome lattices.

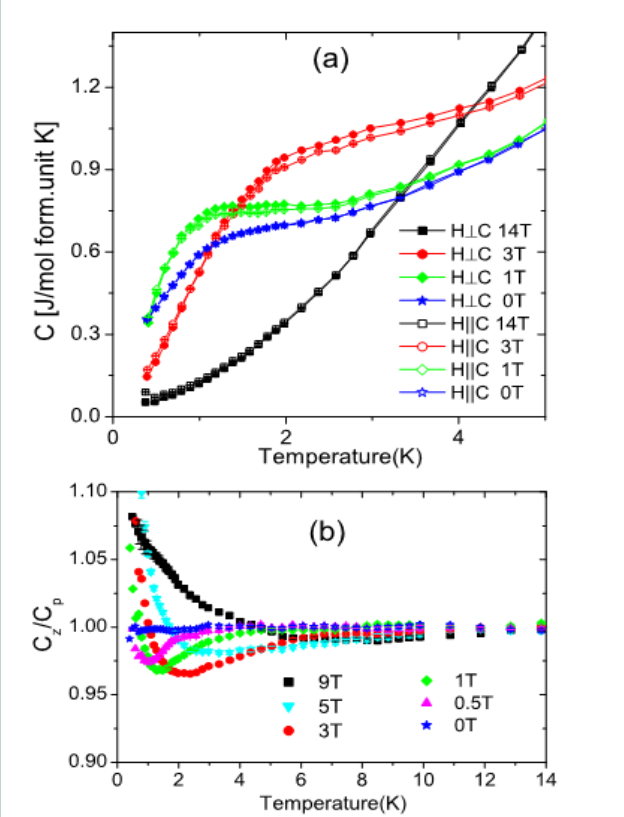
$$ZnCu_3(OH)_6Cl_2$$



# Magnetic susceptibility and heat capacity measurements



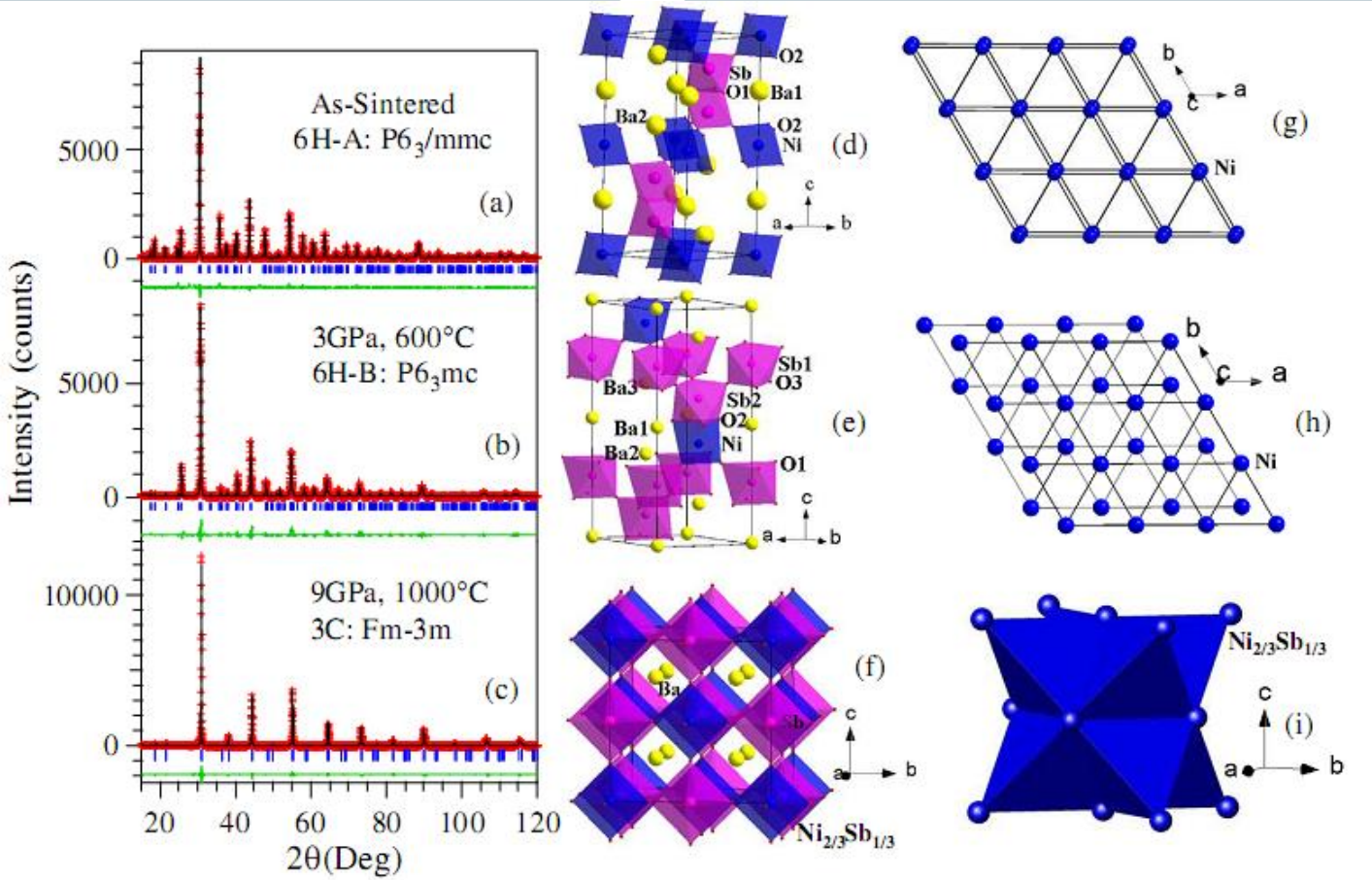



$$H = J\sum_{\langle i,j\rangle} S_i.S_j + \Delta(\sum_{\langle i,j\rangle} S_i^x.S_j^x + S_i^y.S_j^y)$$

# QSL in S=1 system : $Ba_3NiSb_2O_9$

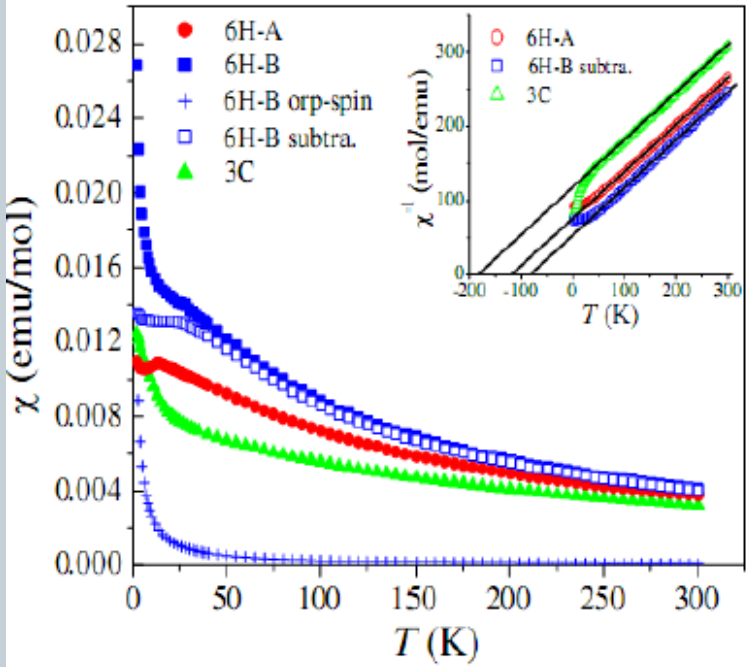





J.G. Cheng, G. Li, L. Balicas, J.S. Zhou, J.B. Goodenough, C. Xu, and H.D. Zhou, Phys. Rev. Lett. **107**, 197204 (2011).

# DC susceptibility



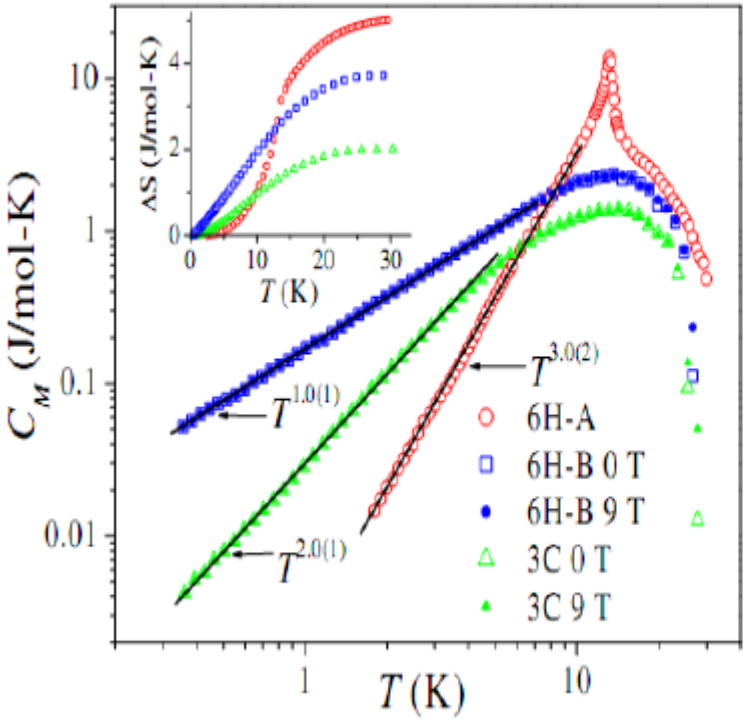



**Results**

- There is no difference between Field-Cooled (FC) and Zero-Field-Cooled (CFC) measurements.
- All three samples are insulating
- $\mu_{eff} \approx 3.54\ \mu_B$
- 6H-A: $\theta_{CW} = -116.9K, \quad T_N = 13.5K$
- 6H-B: $\theta_{CW} = -75.6$
- 3C: $\theta_{CW} = -182.5$
- No ordering down to T=0.35 K for 6H-B and 3C samples.

# Magnetic Heat Capacity






**Results**

- $C_M$ is not affected by applying magnetic field for 6H-B and 3C phases
- $C_M \approx \gamma T^{\alpha}$
- 6H-A: $\alpha = 3.0$
- 6H-B: $\alpha = 1.0$
- 3C: $\alpha = 2.0$

# Results



- 6H-A phase has long-range magnetic orderring with 3D magnons due to interlayer interactions.

- 6H-B phase is a candidate for gapless QSL ground state having possible spinon Fermi surface with S=1.

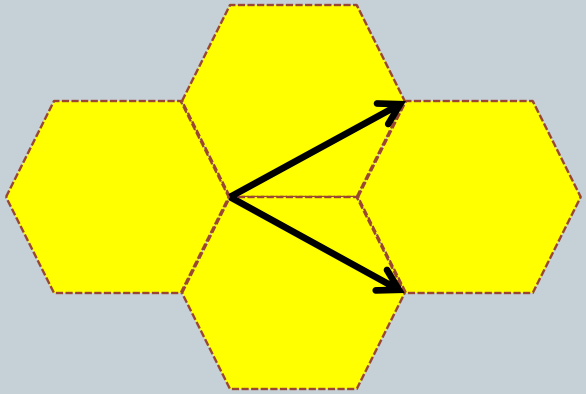
- 3C phase with a edge-shared tetrahedral lattice (fcc) is a candidate for gapless 3D-QSL state with S=1.

# Why Honeycomb Lattice?



- It is two dimensional.

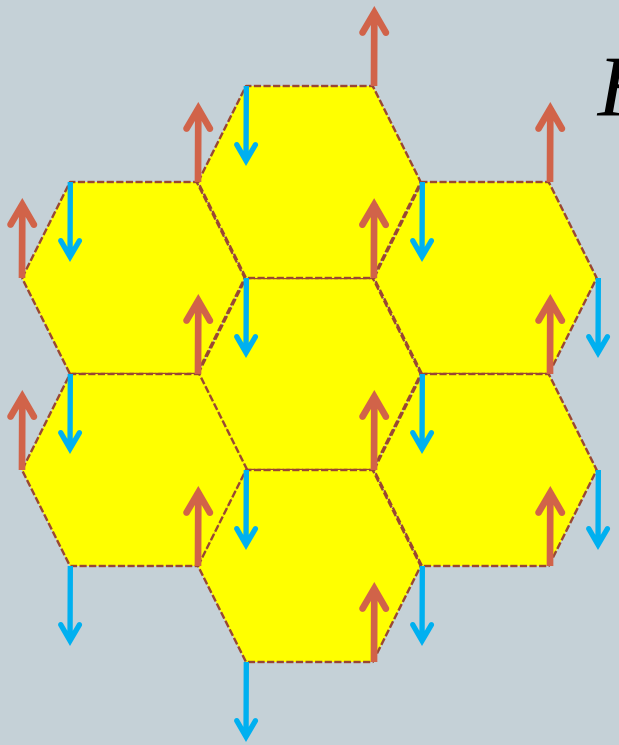
- It has minimum coordination number among 2D lattices. (z=3)
- Therefore spin-1/2 AF Heisenberg model on honeycomb lattice might be a good candidate to have QSL ground state.



# Nearest Neighbor AF Heisenberg Model on Honeycomb Lattice



- Classical state has staggered ordering (Neel order).



$$H = J\sum_{\langle i,j\rangle} S_i . S_j , J > 0$$

- Order parameter (staggered magnetization)

$$m_s = \frac{\left|\sum_{i\in A}\langle S_i\rangle - \sum_{i\in B}\langle S_i\rangle\right|}{N}$$

# Quantum Ground state



- Variational Monte Carlo:

  Trial Wave function

$$|\Psi\rangle = \sum_{\substack{i_\alpha \in A \\ j_\beta \in B}} h(i_1 - j_1) \cdots h(i_n - j_n)(i_1, j_1) \cdots (i_n, j_n)$$

- Short-bond RVB wave functions are not good . (Not a spin liquid)
- Long-bond RVB wave functions give the lowest energy.
- Neel order exist, however because of quantum fluctuations its order parameter reduces to about half of its classical value.

Quantum fluctuations are not again strong enough to melt the Neel order.

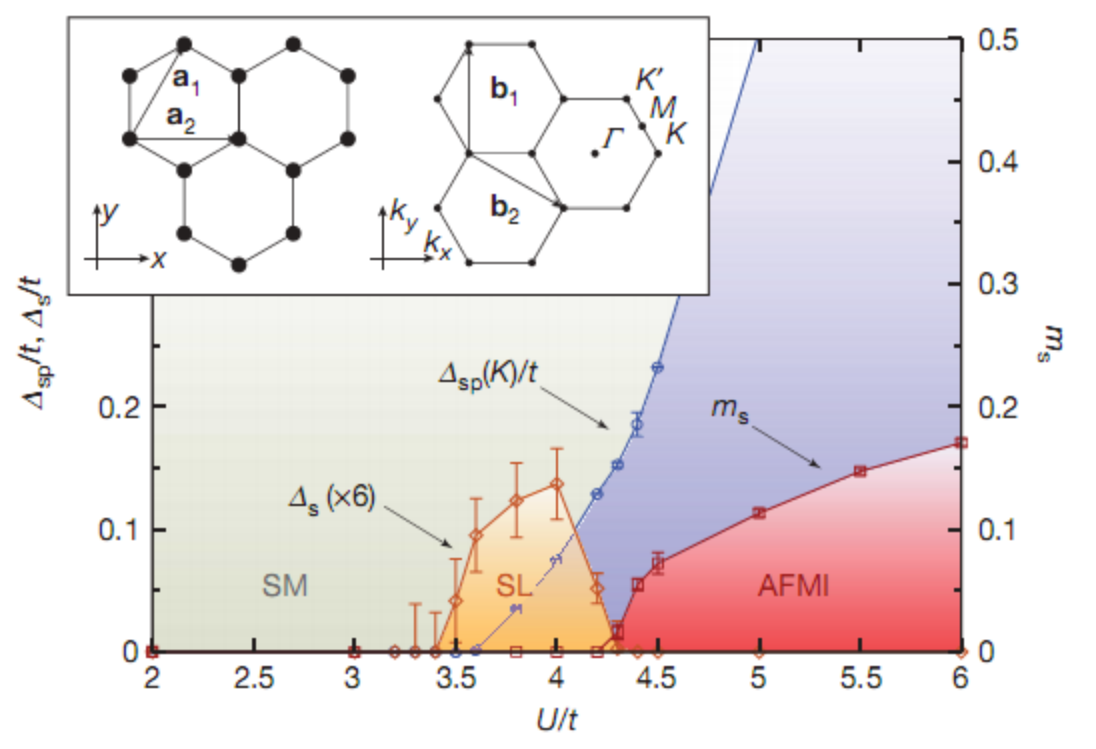
Z. Noorbakhsh, F. Shahbazi, S. A. Jafari, and G. Baskaran, JPSJ 78, 054701 (2009).

# Hubbard Model on Honeycomb Lattice



- Quantum Monte Carlo simulation
- Possible RVB spin liquid for 3.5 < U/t < 4.3

$$H = -t \sum_{\langle i,j \rangle, \alpha} (c_{i\alpha}^{\dagger} c_{j\alpha} + c_{j\alpha}^{\dagger} c_{i\alpha}) + U \sum_{i} n_{i\uparrow} n_{i\downarrow}$$




- Z. Y. Meng et al, Nature 464, 847 (2010).

# Effective Spin Hamiltonian

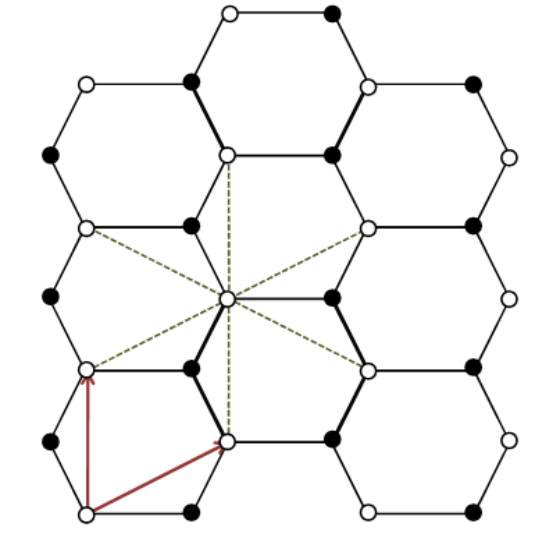


- To fourth order, the perturbation theory results in a J1-J2 Heisenberg model as the effective Hamiltonian of the Hubbard model on the honeycomb lattice :

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

$$J_1 = 4\frac{t^2}{U} - 16\frac{t^4}{U^3}$$

$$J_2 = 4\frac{t^4}{U^3}$$



# Exact Diagonalization



- ED in $S^z$ basis versus ED in nearest neighbor valence bond (NNVB) basis.

Consider the set of NNVB configurations as: $$|c_\alpha\rangle = \prod_{(i,j)\in\alpha} (i_\uparrow j_\downarrow - i_\downarrow j_\uparrow).$$

The GS can be expanded in this babsis: $$|\psi_0\rangle = \sum_\alpha w(c_\alpha)|c_\alpha\rangle,$$

Unlike $S^z$ basis, NNVB states are not orthogonal. Therefore one needs to solve the generalized eigen-value problem: $$\det[\mathcal{H} - E\mathcal{O}] = 0,$$

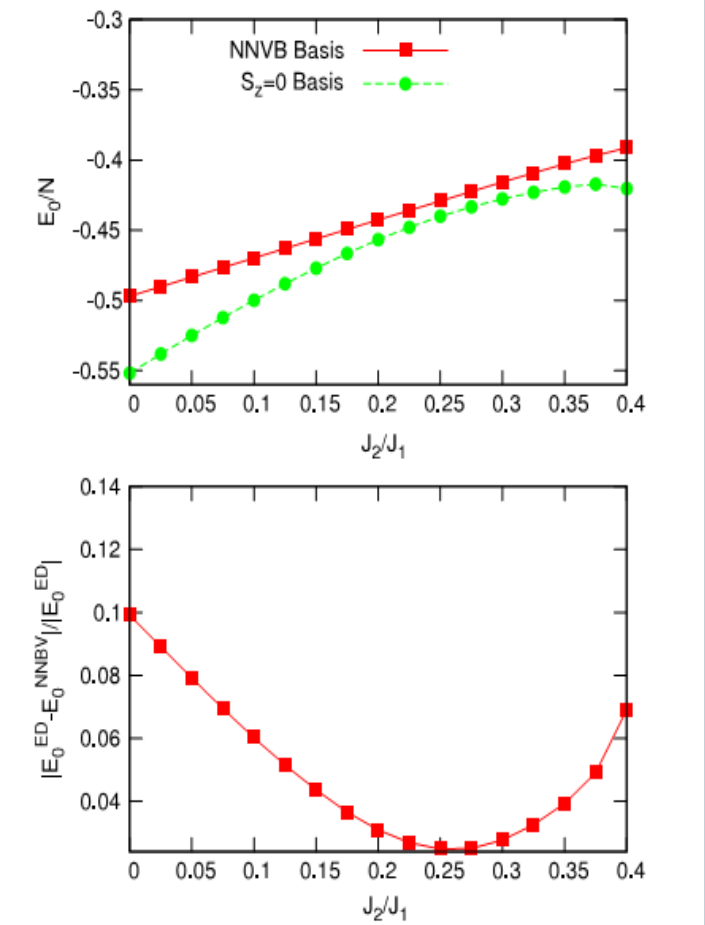
Overlap matrix : $$\mathcal{O} = \langle c_\beta | c_\alpha \rangle$$

# Ground State Energy



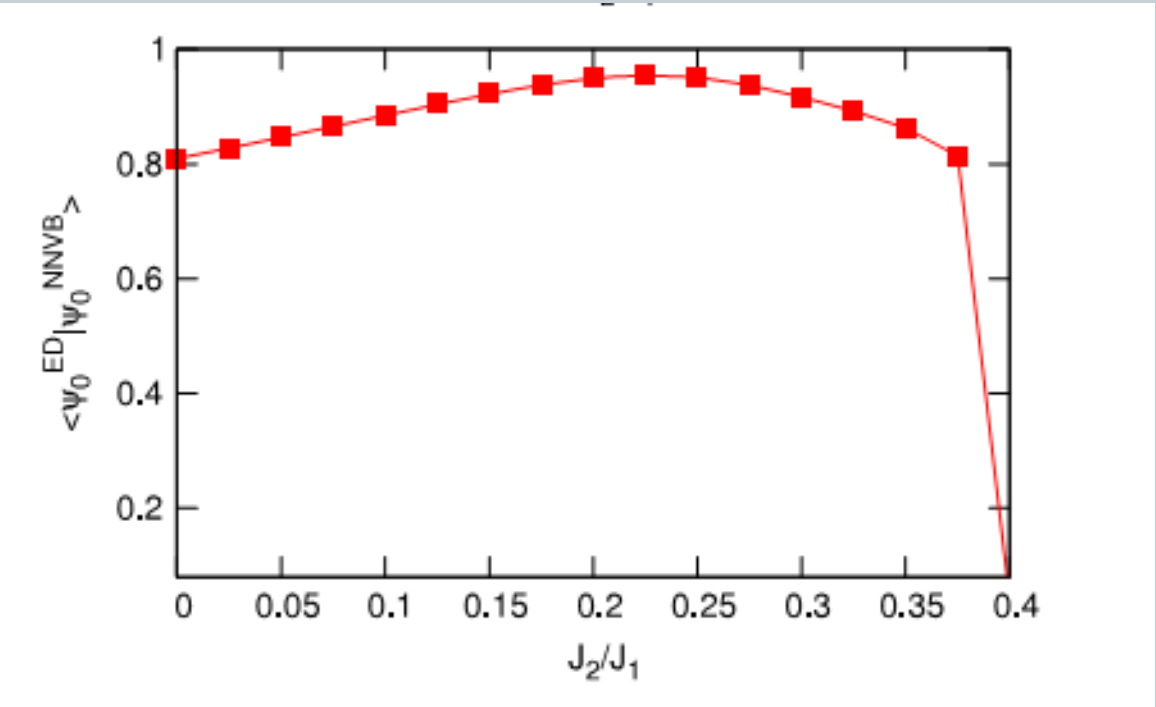
- GS energy agrees well in both basis for $0.15 < \frac{J_2}{J_1} < 0.35$

H. Mosadeq, F. Shahbazi, and S. A. Jafari, JPCM 23, 226006 (2011),.



# Overlap of The Two GS Wavefunctions





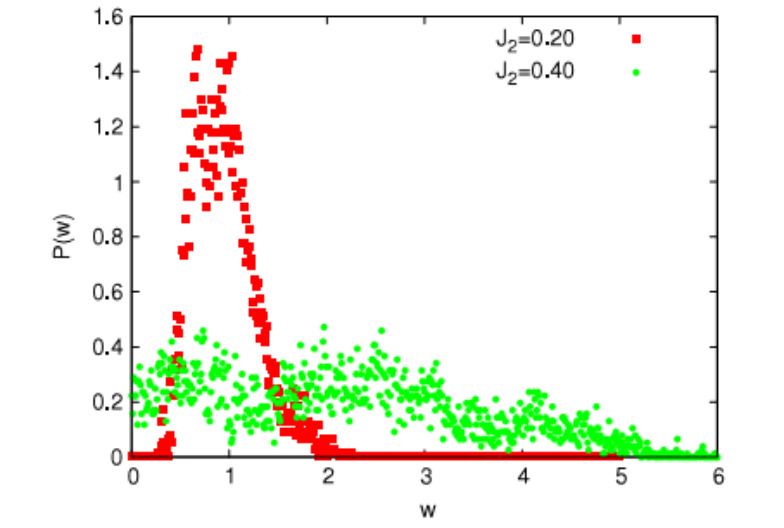
# Distribution of NNVB Configurations

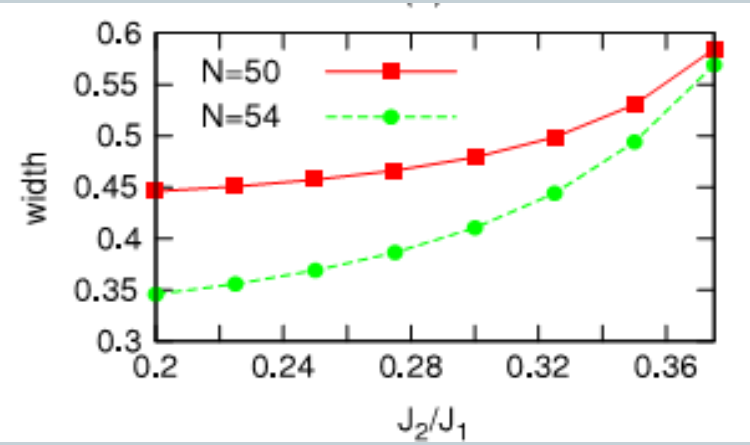


The width of Probability distribution function of the weights of NNVB configurations is finite at large lattice sizes.

RVB liquid Should have zero width.

Possibility of long-range dimer-dimer correlation.



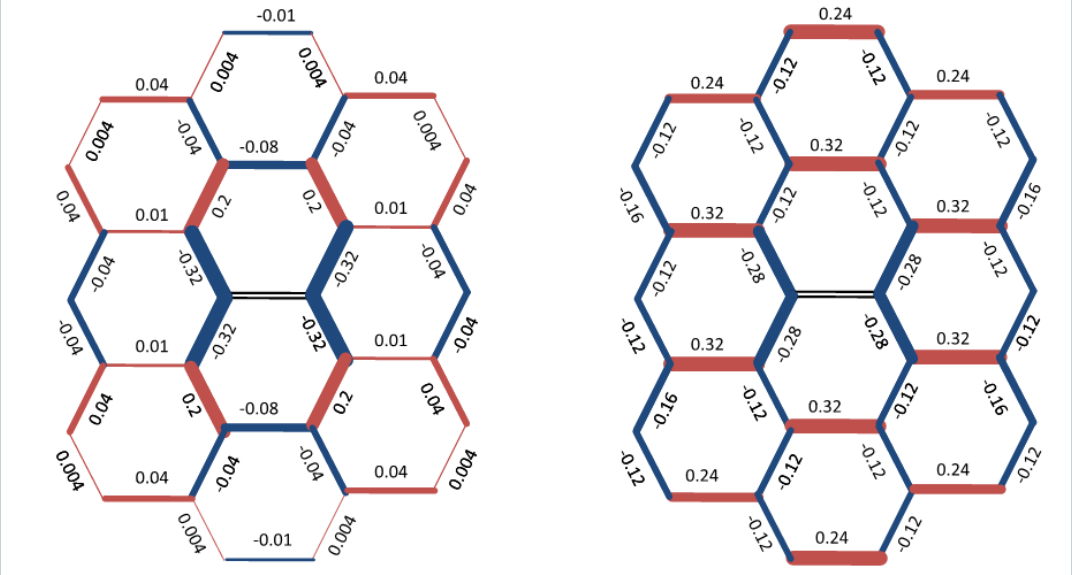



# Dimer-Dimer Correlation



$$C(\alpha, \alpha') = \langle P_\alpha P_{\alpha'} \rangle - \langle P_\alpha \rangle \langle P_{\alpha'} \rangle$$

$$P_{kl} = 2(\mathbf{S}_k \cdot \mathbf{S}_l) + \frac{1}{2}$$




$$\frac{J_2}{J_1} = 0.3$$

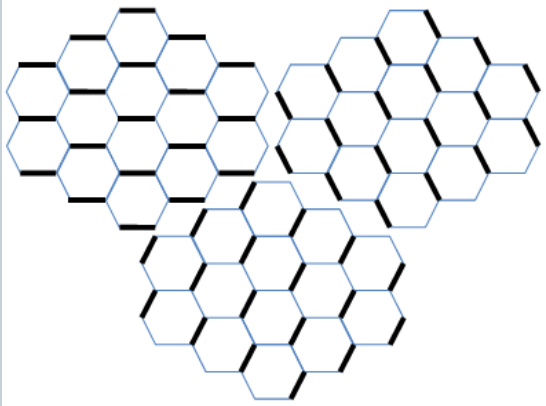
$$\frac{J_2}{J_1} = 0.4$$

# Proposed GS's

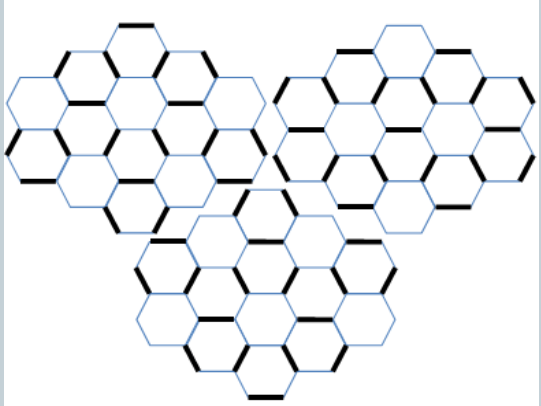


There GS consistent with symmetries of honeycomb lattice have been proposed,

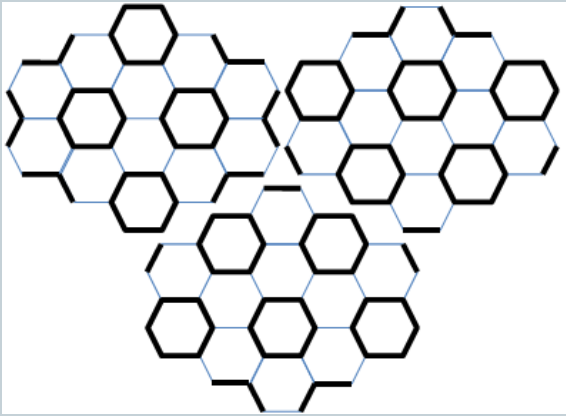
Staggered dimerized (SD), Columnar (proposed by Read and Sachdev ) and Plaquette Valence bond (PL ) state.



Staggered Dimerized



Columnar Dimerized



Pleaquette VB

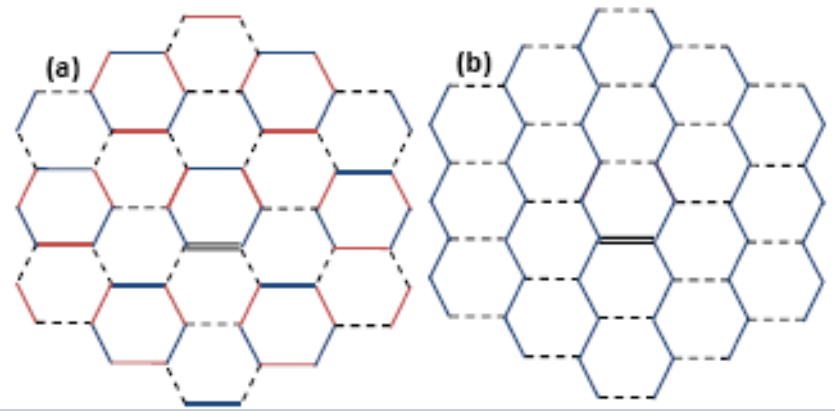
# Structure Factors



- Structure factor which plays the role of order parameter is defined as:

$$S_\lambda = \sum_{\alpha'} \varepsilon_\lambda(\alpha')\, C(\alpha, \alpha'),$$

$C(\alpha, \alpha')$ is dimer-dimer correlation and $\varepsilon_\lambda(\alpha)$ is the phase factor, appropriately defined for each of trial state:




Red and blue links correspond to +1, -1 phase, respectively, while the dashed links stand for zero.
(a) CD and PL, (b) SD.

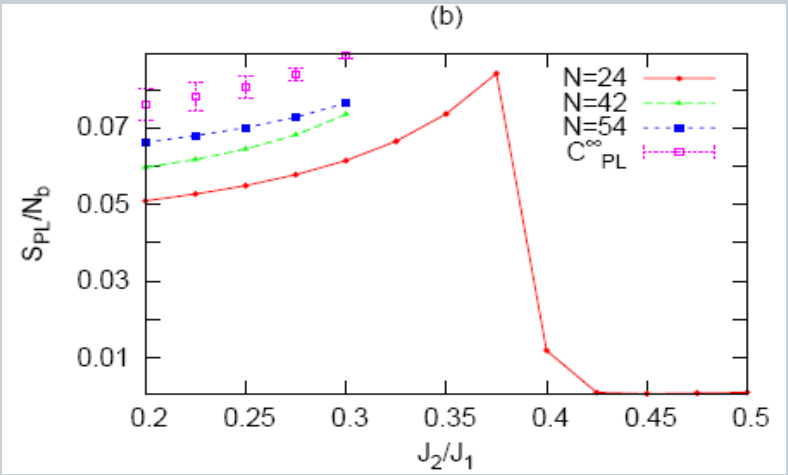
# Results

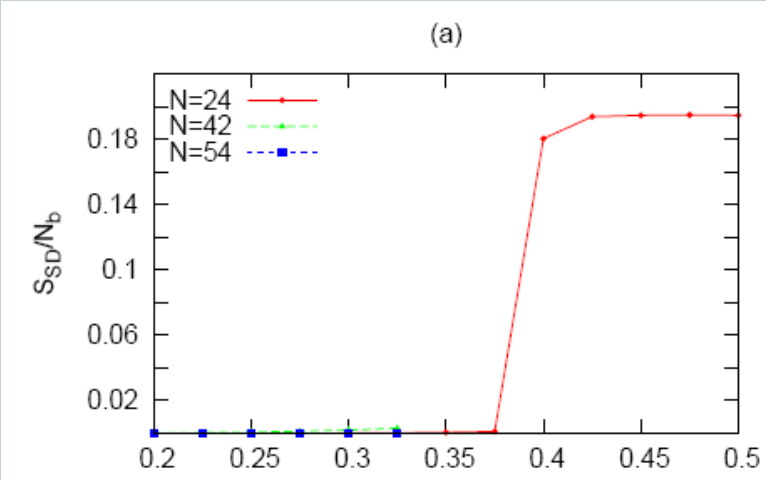


- Scaling of structure factors is given by:

$$\frac{S_\lambda}{N_b} = C_\lambda^\infty + \frac{A}{N}$$

| Trial state | $\psi_{SD}$ | $\psi_{PL}$ | $\psi_{RS}$ |
|---|---|---|---|
| $C^\infty{}_{SD}$ | 1/4 | 0 | 0 |
| $C^\infty{}_{PL}$ | 0 | 0.125(5) | 3/8 |



# Plaquette-Plaquette Correlation



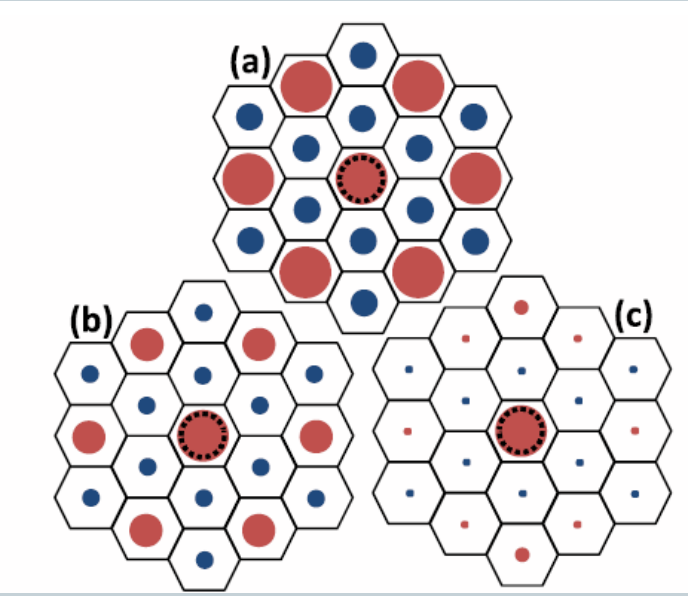
- Defining PL-PL correlation as:

$$C(p,q) = \langle Q_p Q_q \rangle - \langle Q_p \rangle^2,$$
$$Q_p = \frac{1}{2}(\Pi_p + \Pi^{-1}{}_p),$$

Where p, q stand for different plaquette and $\Pi_p$ is the cyclic exchange operator which permutes six spins around a hexagon in clockwise direction.

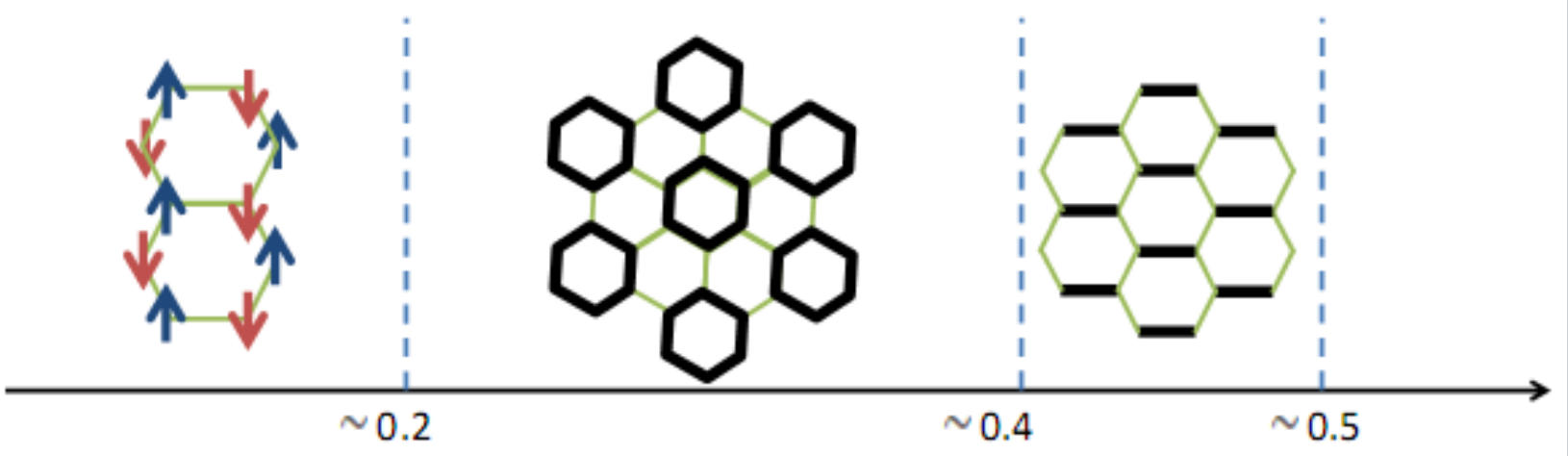
Red (blue) circles denote positive (negative) correlations. The radius of circles are proportional to the value of plaquette-plaquette correlation.
(a) The plaquette-plaquette correlation map calculated for PL trial state.
(b), (c) Exact plaquette-plaquette correlation at

$$\frac{J_2}{J_1} = 0.3, 0.5$$



# Phase Diagram

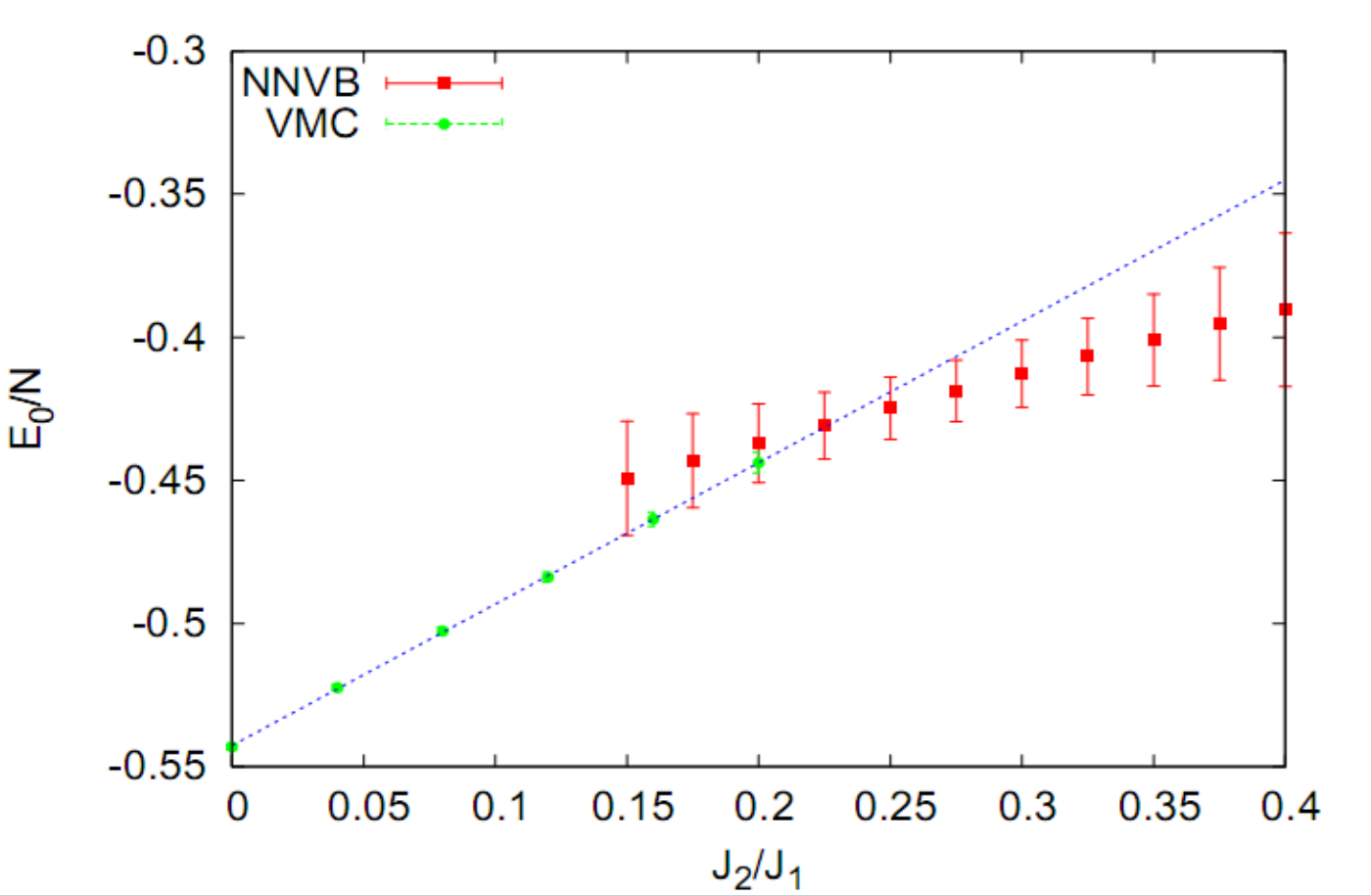





Neel order Plaquette order Stagger dimerized

# Order of transition?





# Conclusion



1- ED results show that the ground state of J1-J2 AF Heisenberg model on honeycomb lattice for $0.15 < \frac{J_2}{J_1} < 0.35$ can be well described within Hilbert space spanned by NNVB basis set.

2- In this region the GS is not a RVB spin liquid but a state with long-range plaquette ordering.

# THANKS FOR YOUR ATTENTION