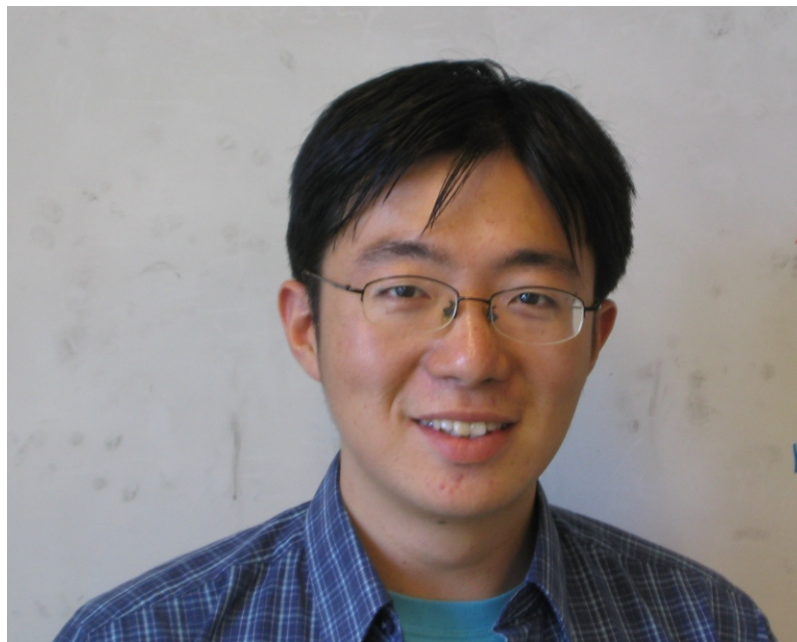


Global phase diagrams of two-dimensional quantum antiferromagnets



Cenke Xu



Yang Qi

Subir Sachdev
Harvard University



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1. Review of experiments

Phases of the $S=1/2$ antiferromagnet on the anisotropic triangular lattice

2. The Z_2 spin liquid and its excitations

Spinons and visons

3. Field theory for spinons and visons

Berry phases and the doubled Chern-Simons theory

4. Is κ -(ET) $_2$ Cu $_2$ (CN) $_3$ a Z_2 spin liquid ?

Thermal conductivity of κ -(ET) $_2$ Cu $_2$ (CN) $_3$

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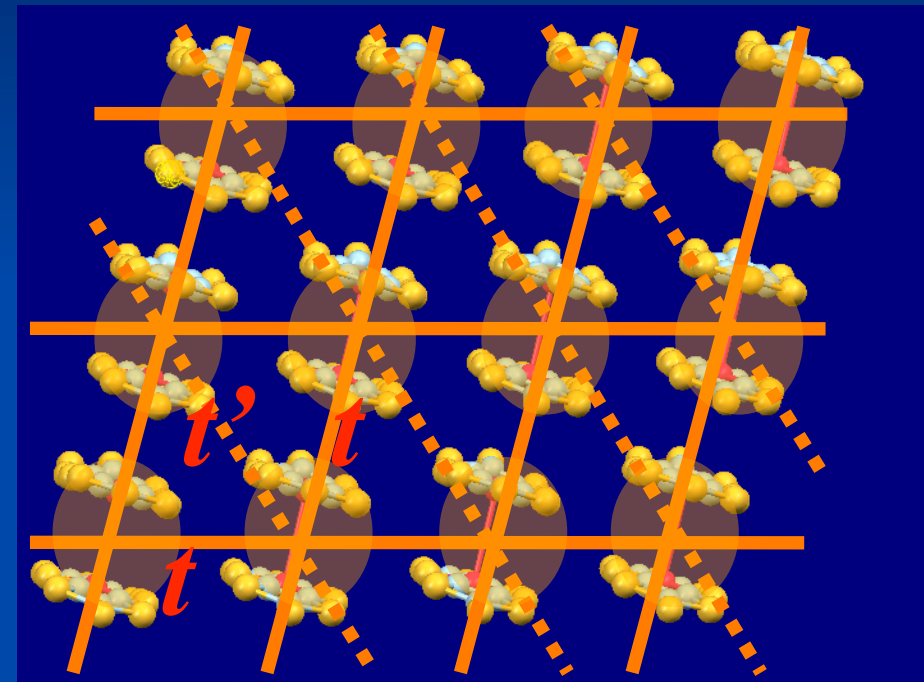
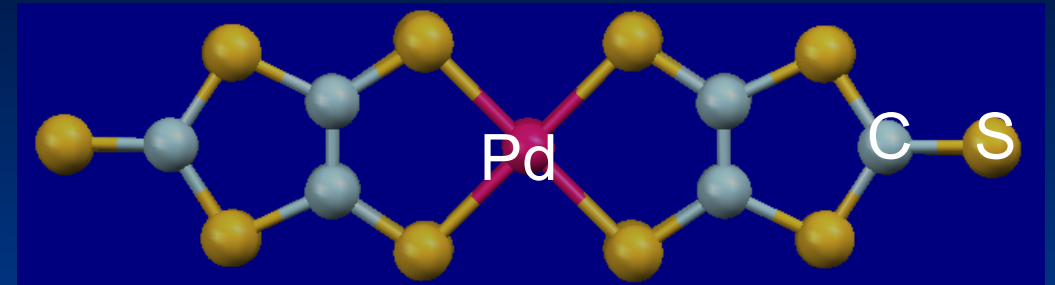
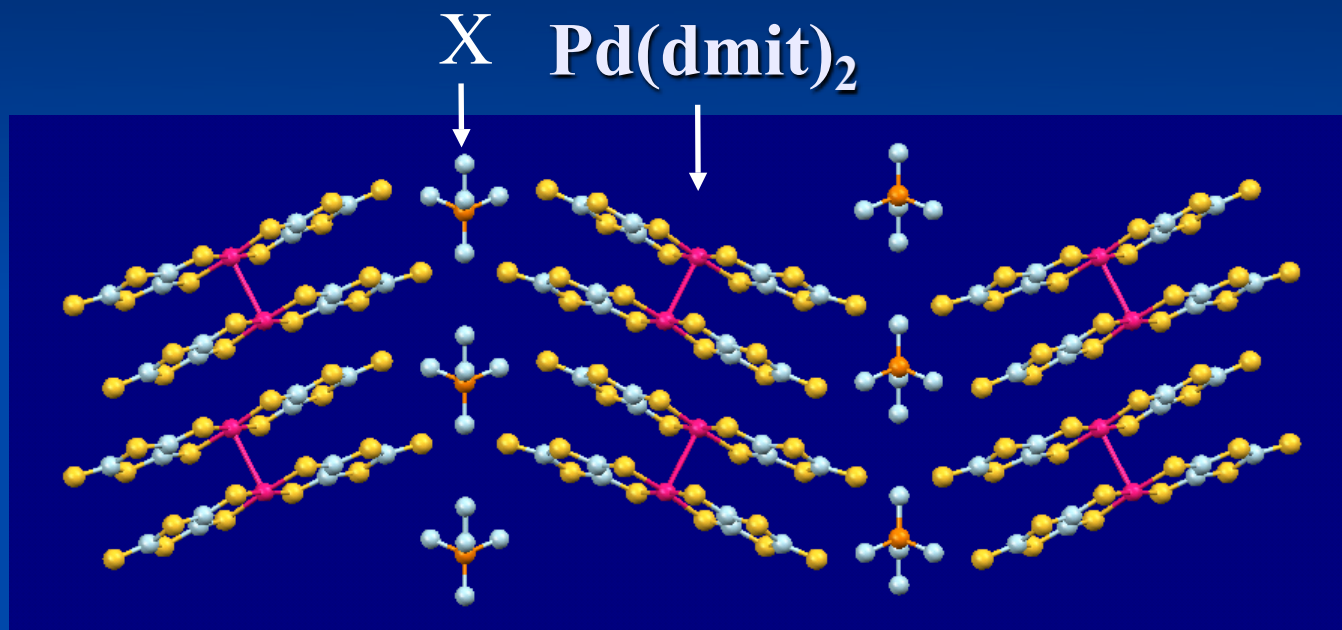
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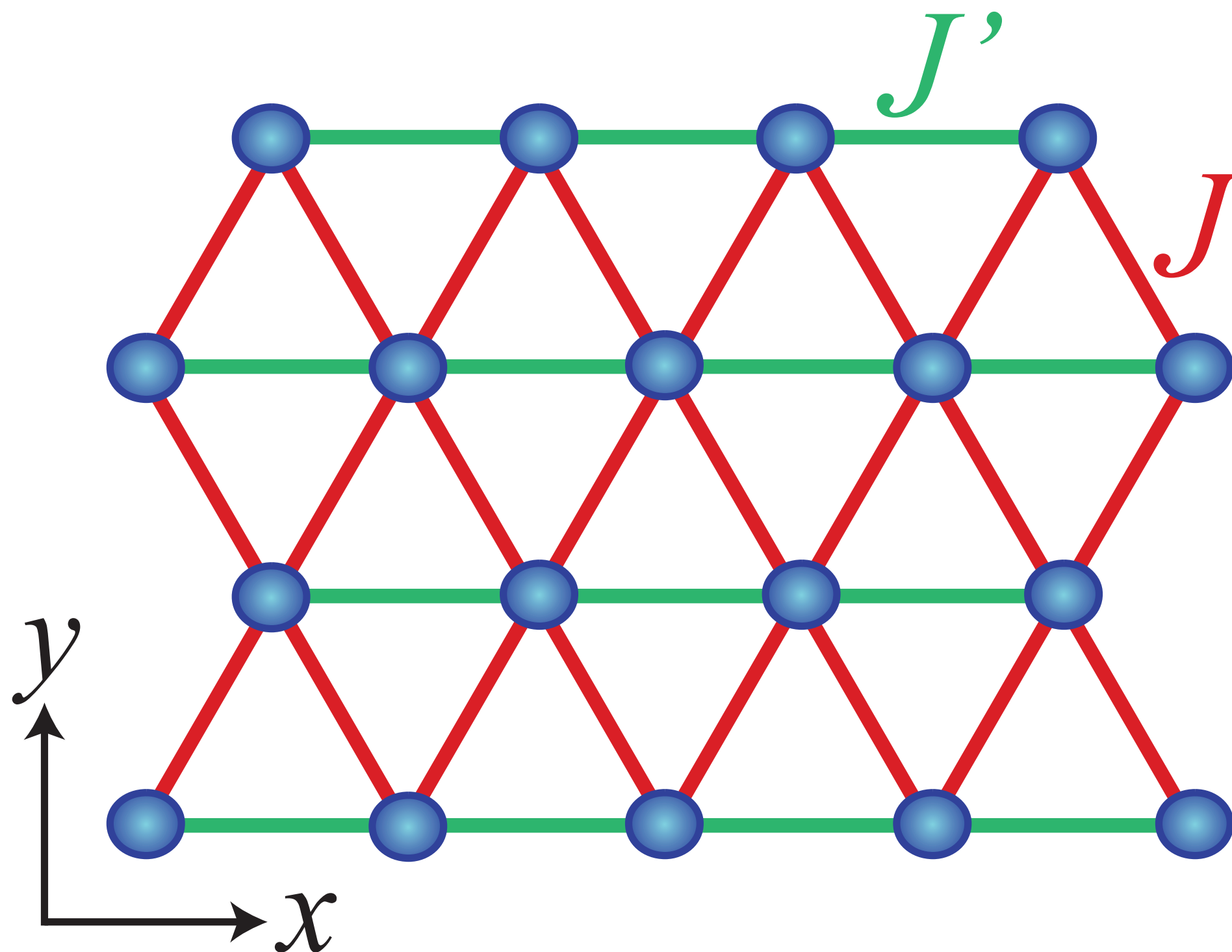
Thermal conductivity of κ -(ET)₂Cu₂(CN)₃



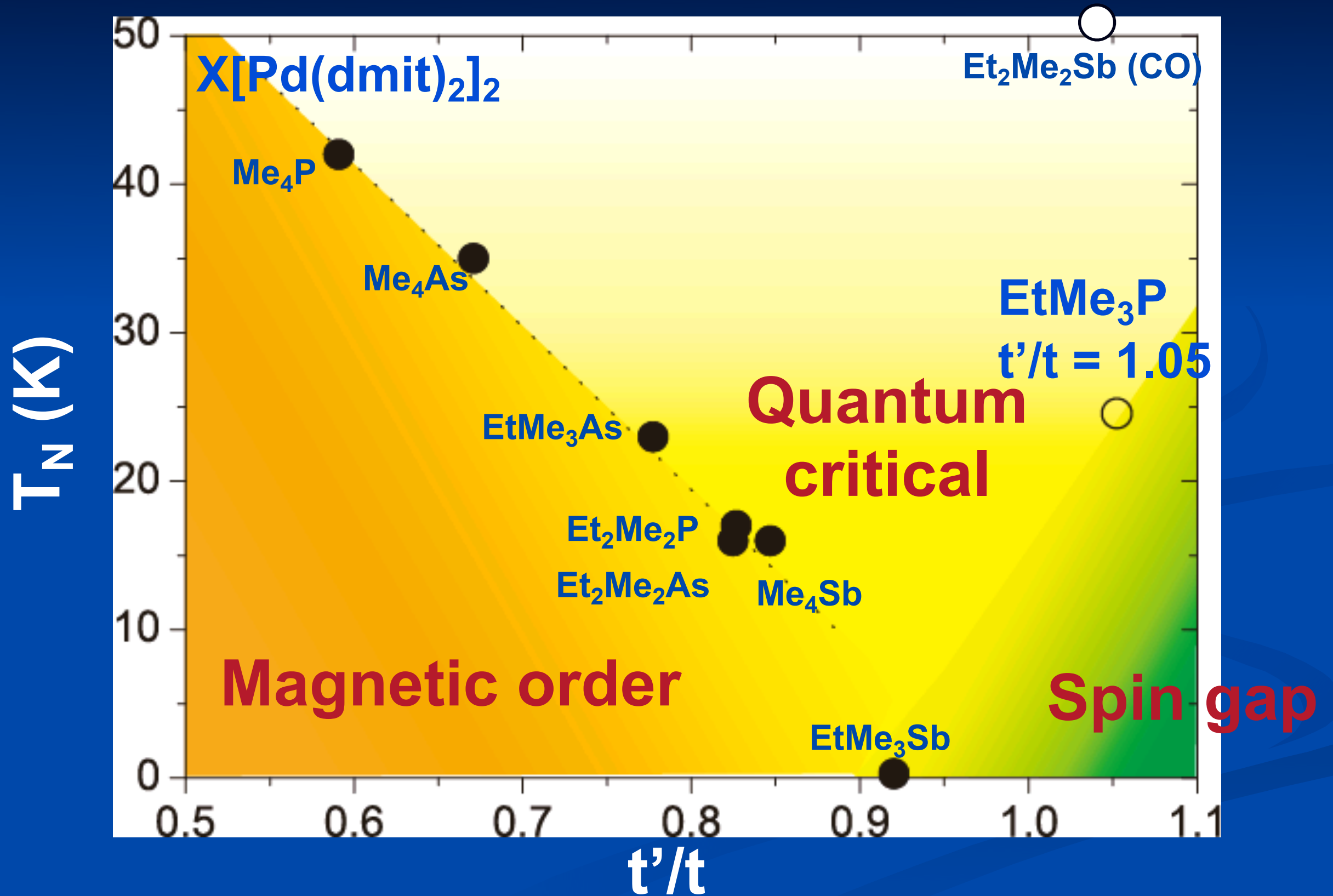
Half-filled band $\rightarrow$ Mott insulator with spin $S = 1/2$

Triangular lattice of $[\text{Pd}(\text{dmit})_2]_2$
 $\rightarrow$ frustrated quantum spin system

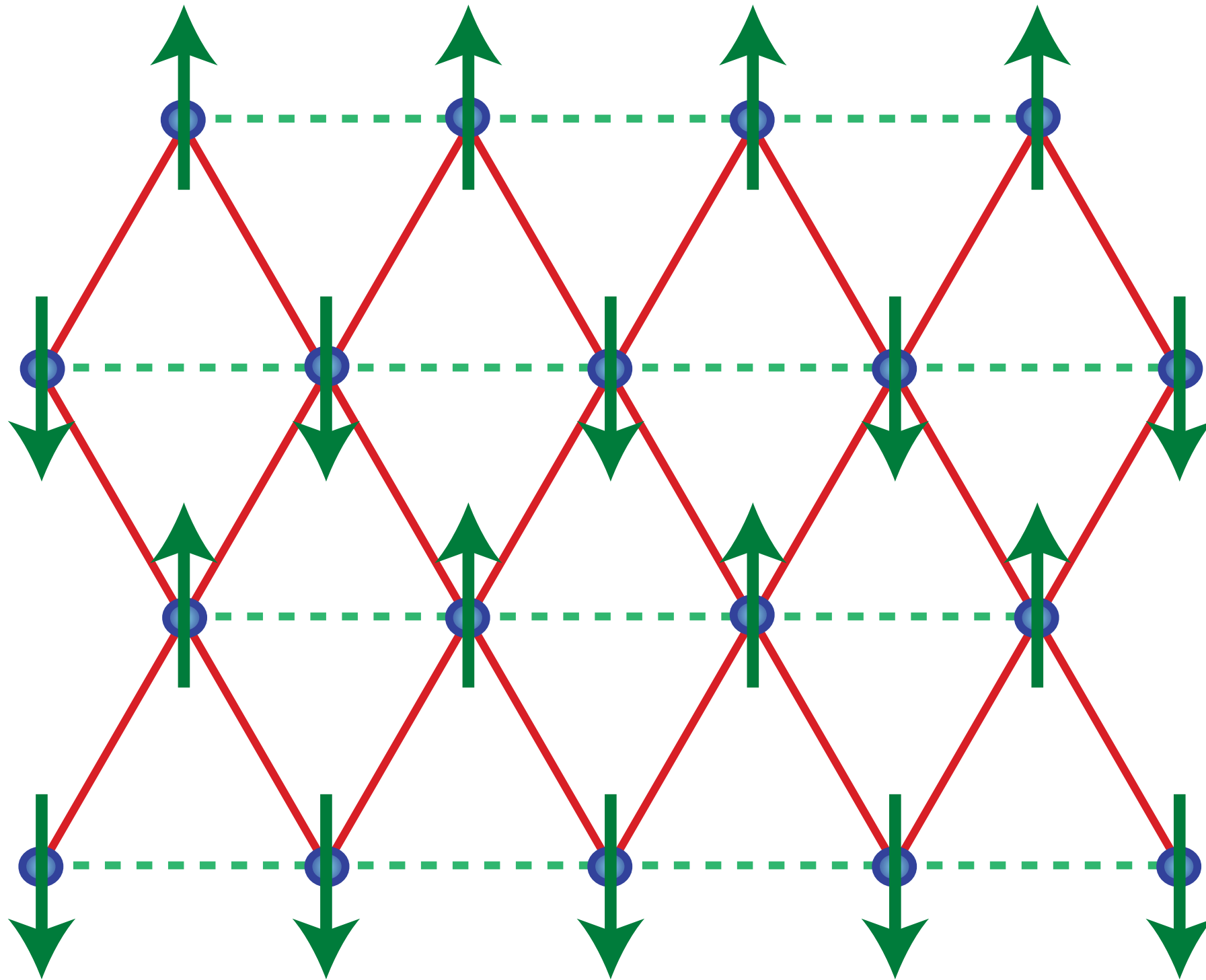
$$H = \sum_{\langle ij \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j + \dots$$



Magnetic Criticality

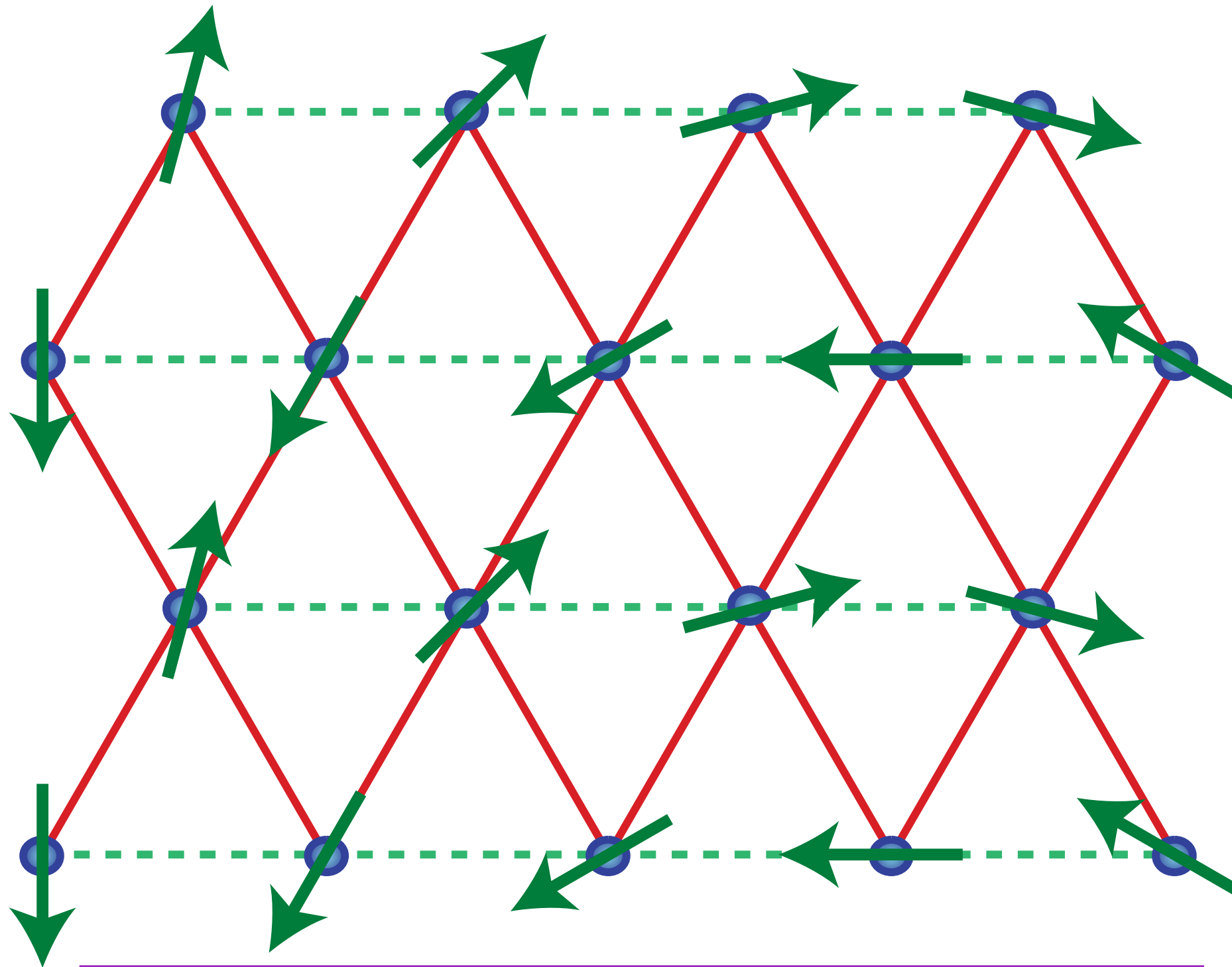


Anisotropic triangular lattice antiferromagnet



Classical ground state for small J'/J

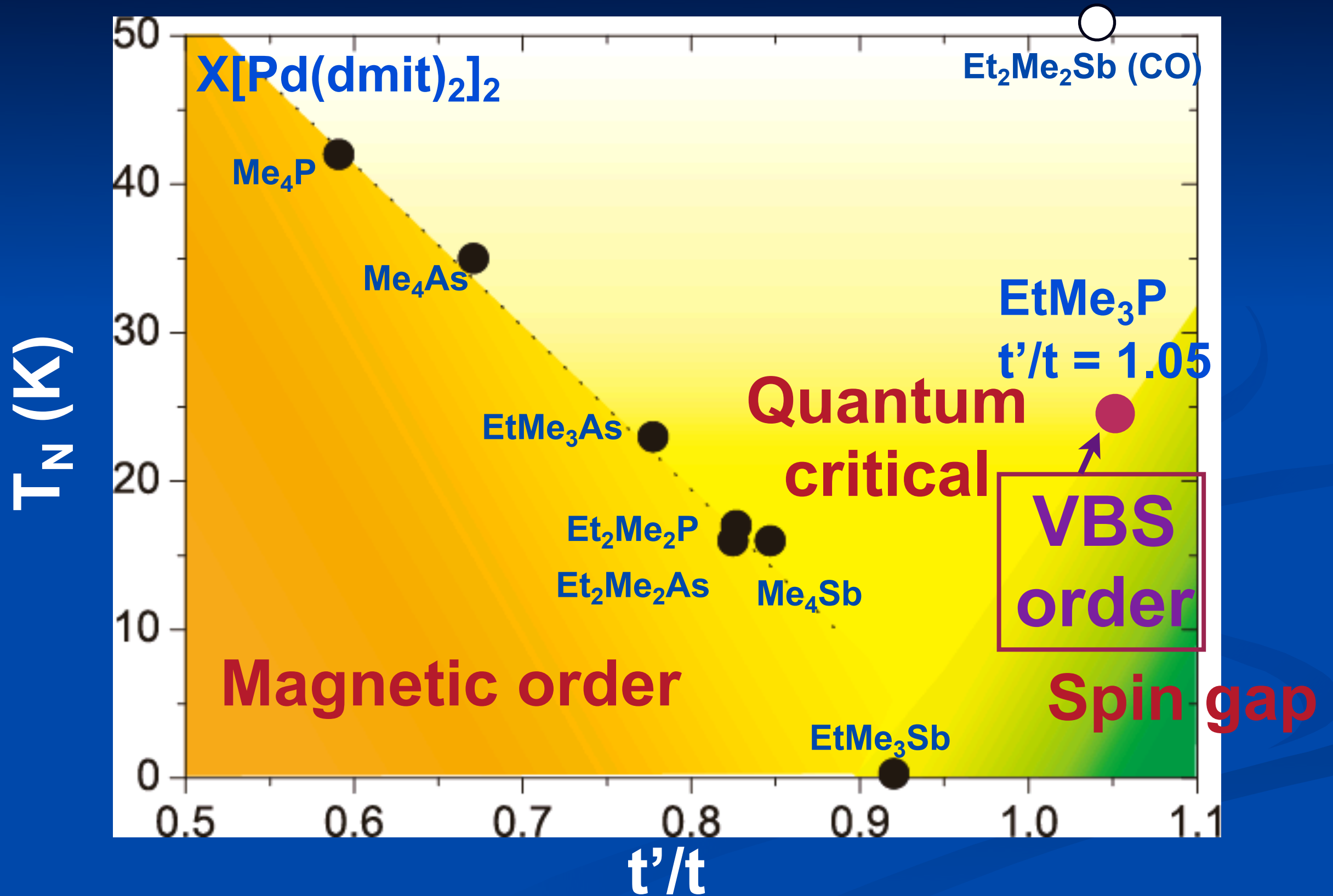
Anisotropic triangular lattice antiferromagnet



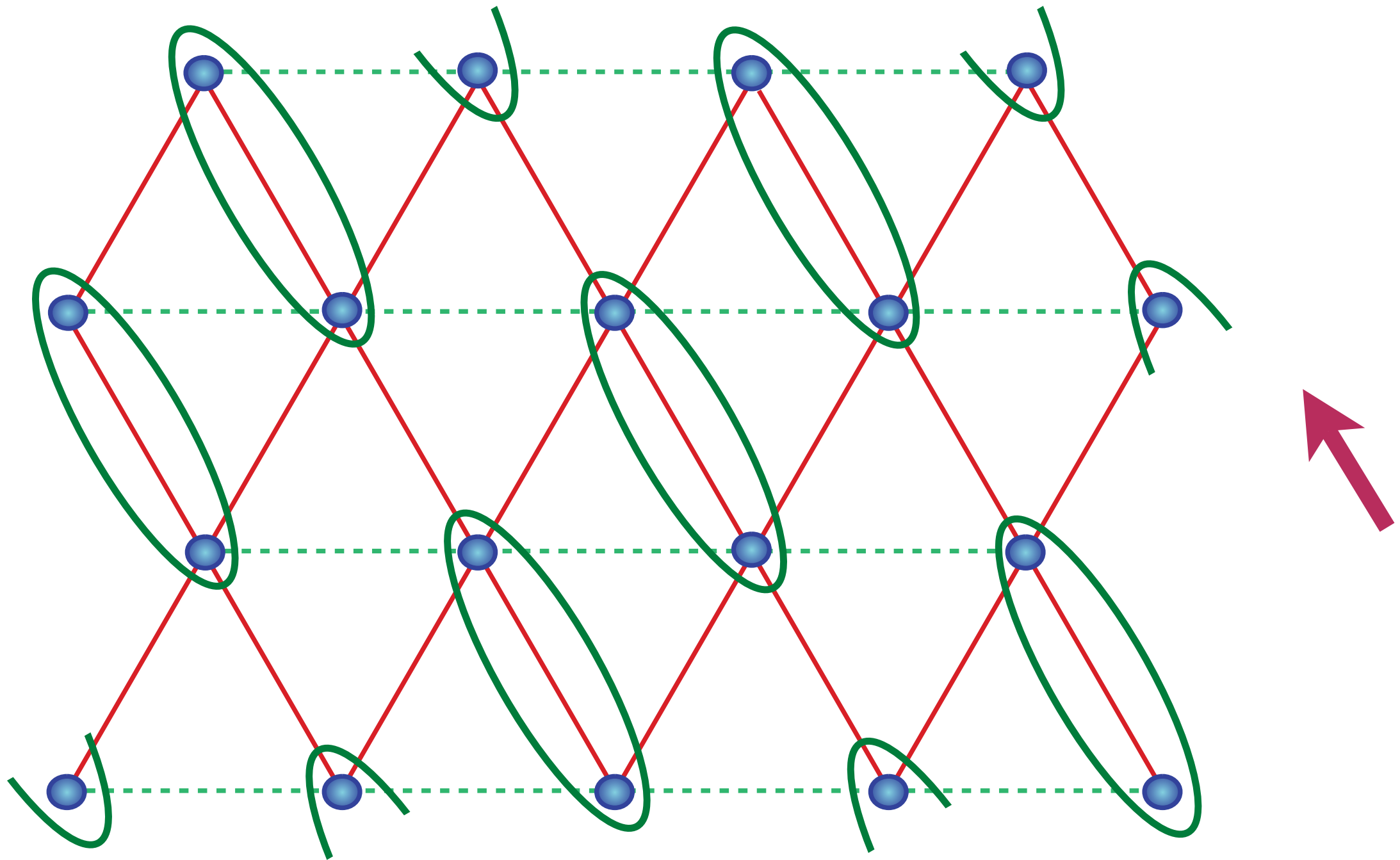
Classical ground state for large J'/J

Found in Cs_2CuCl_4

Magnetic Criticality

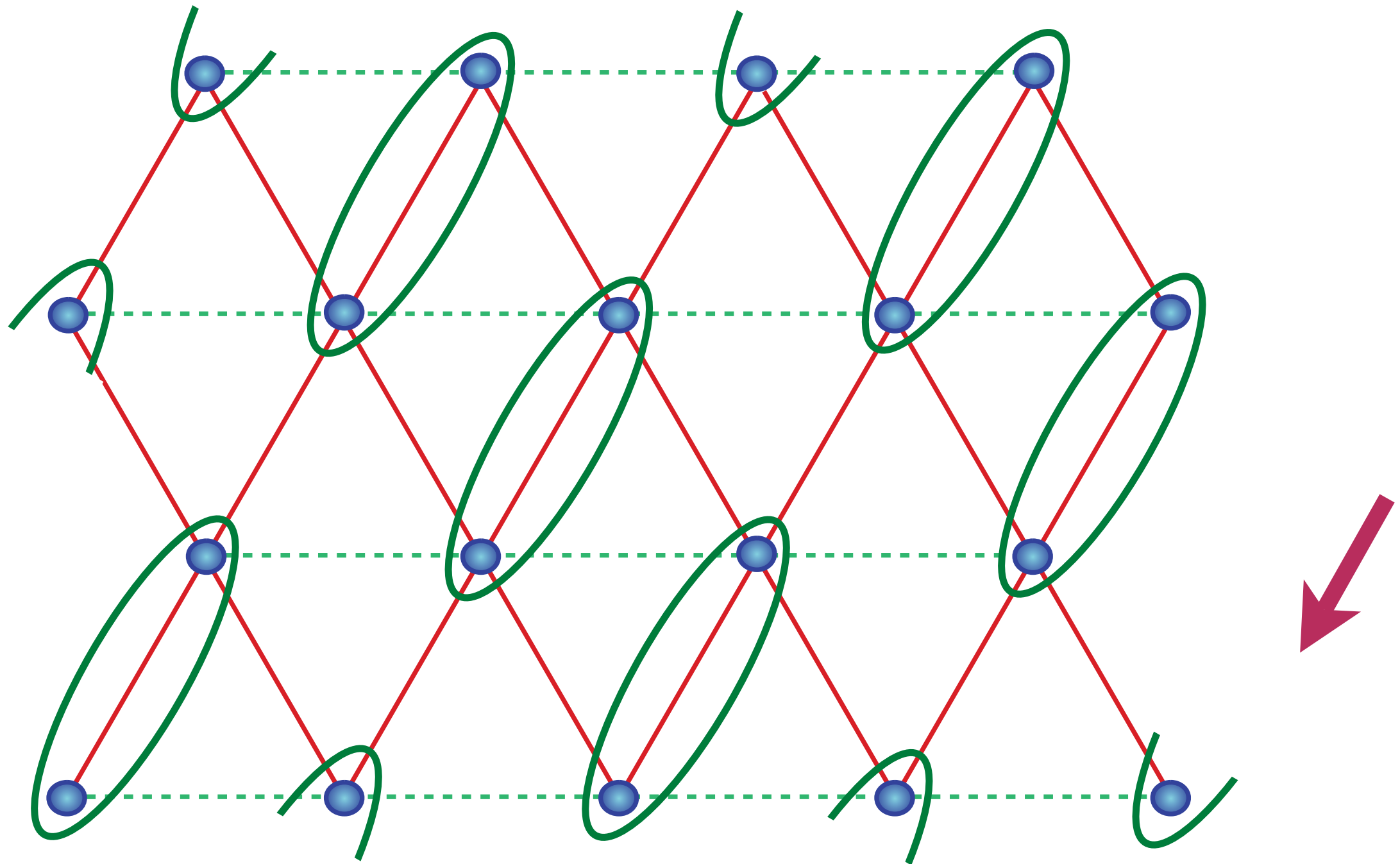


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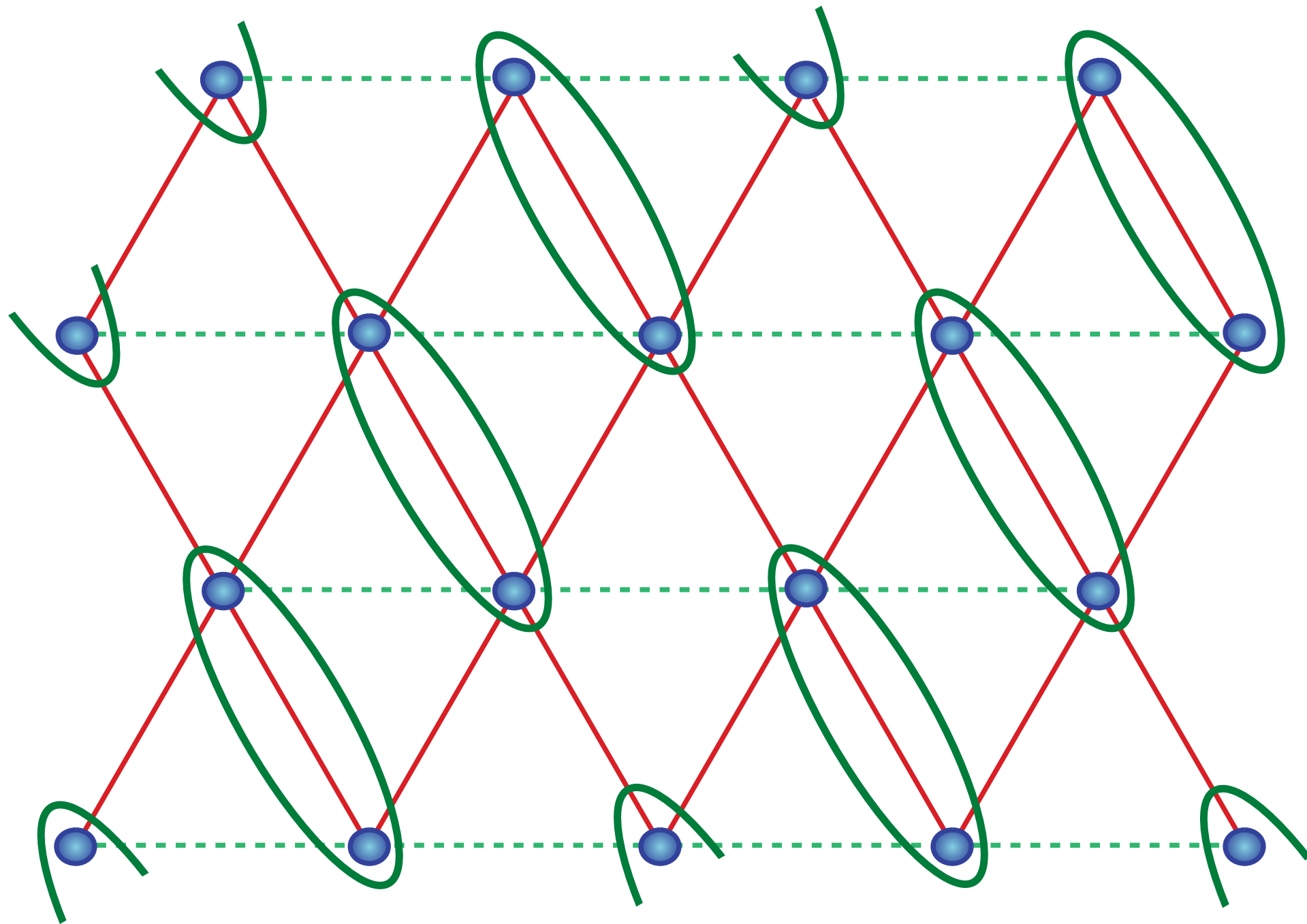
Valence bond solid

Anisotropic triangular lattice antiferromagnet



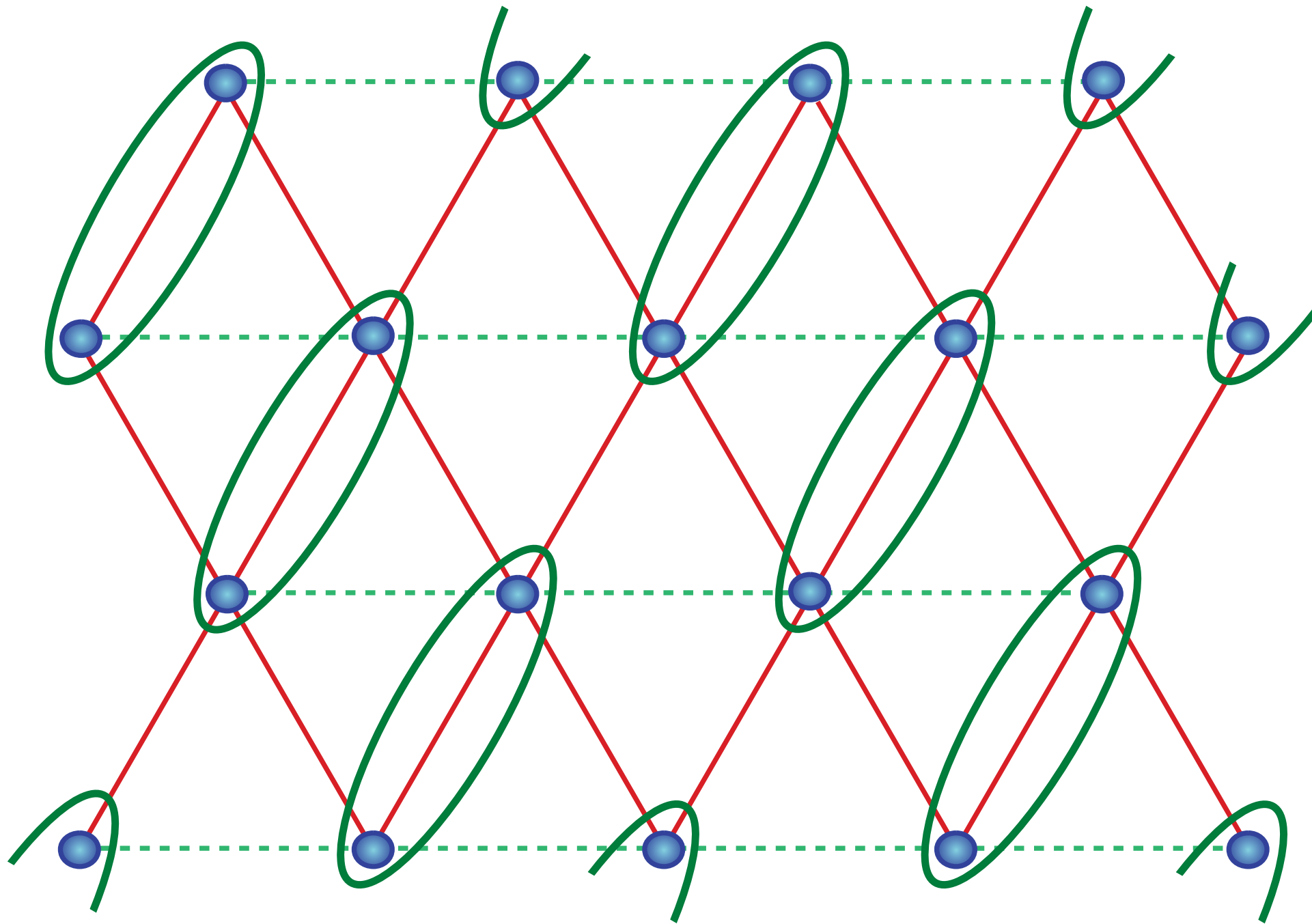
Valence bond solid

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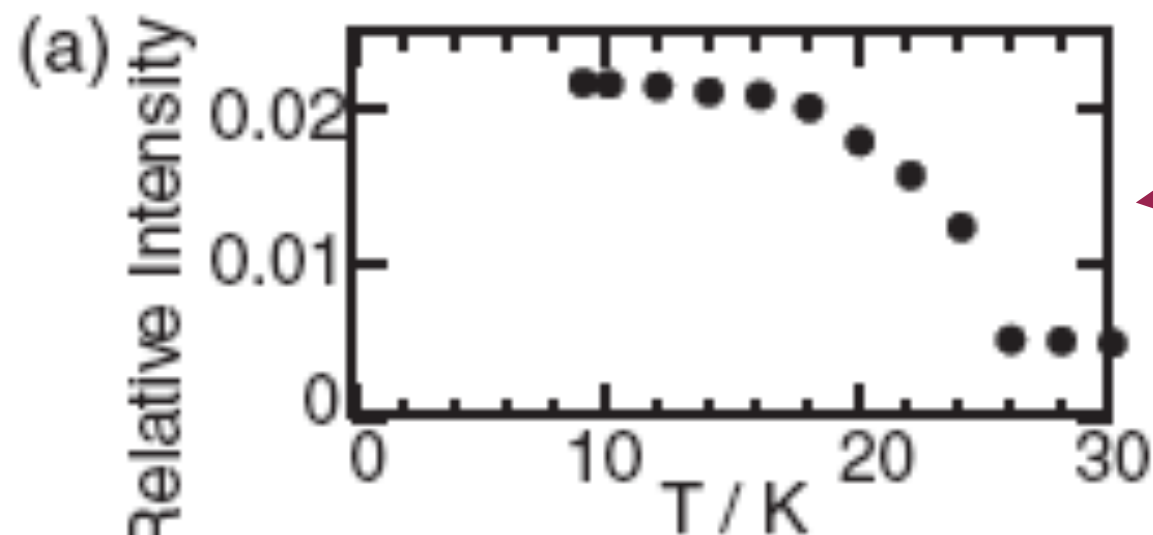
Valence bond solid

Anisotropic triangular lattice antiferromagnet

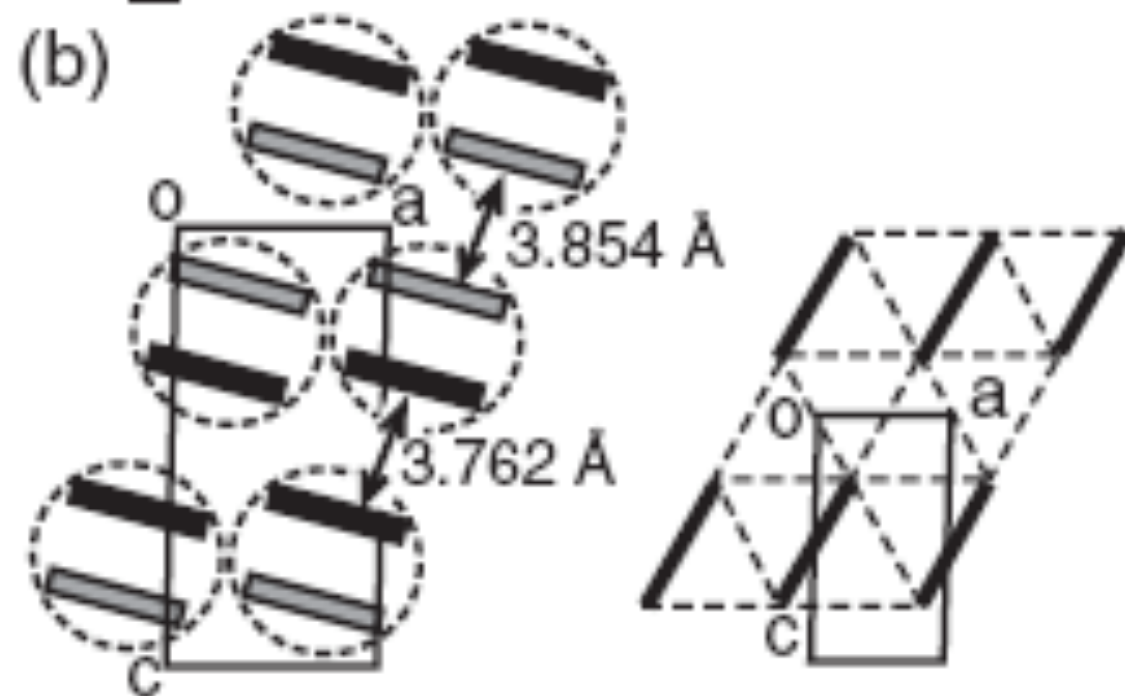


Valence bond solid

Observation of a valence bond solid (VBS) in $\text{ETMe}_3\text{P}[\text{Pd}(\text{dmit})_2]_2$



X-ray scattering

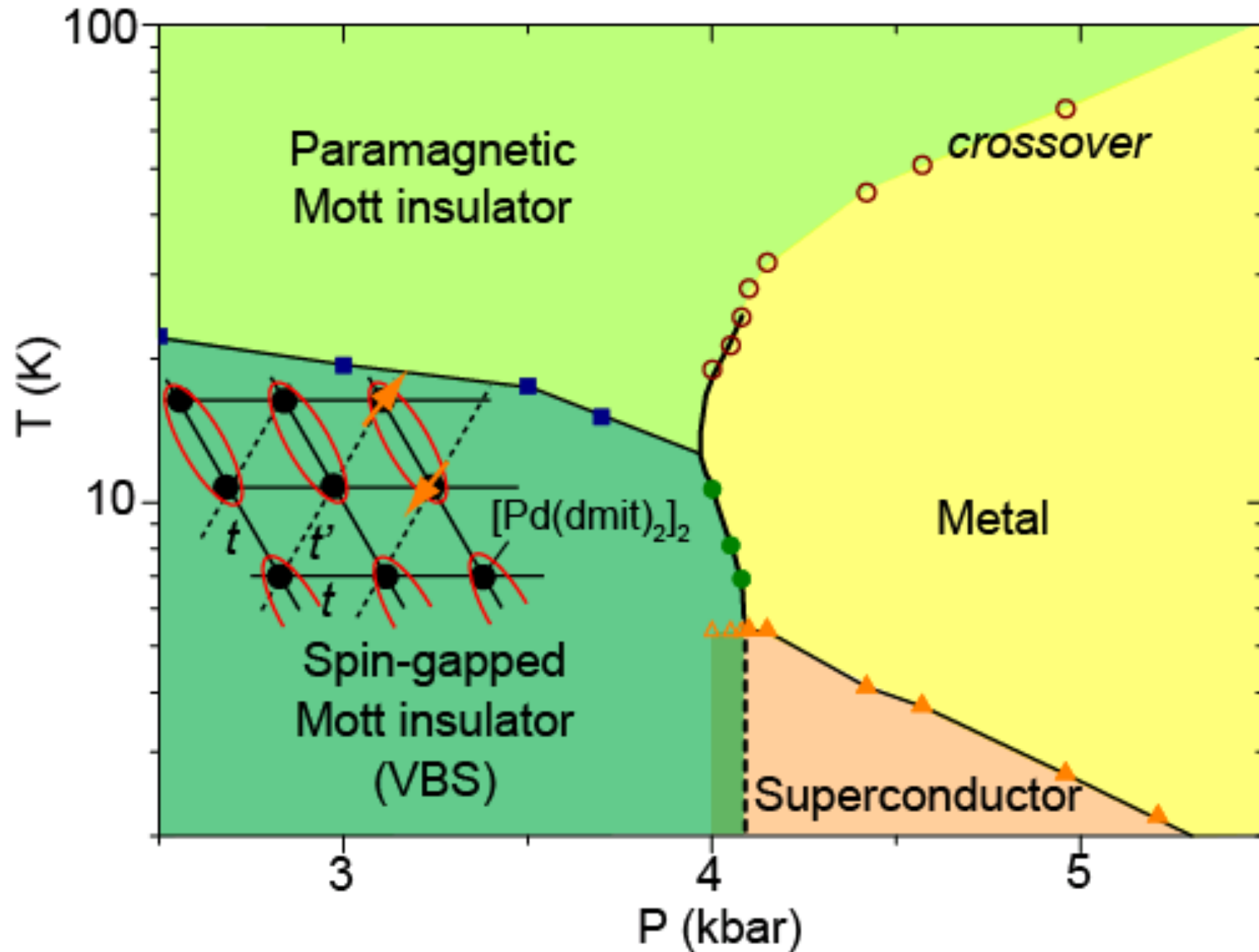


Spin gap ~ 40 K
 $J \sim 250$ K

M. Tamura, A. Nakao and R. Kato, *J. Phys. Soc. Japan* **75**, 093701 (2006)

Y. Shimizu, H. Akimoto, H. Tsujii, A. Tajima, and R. Kato, *Phys. Rev. Lett.* **99**, 256403 (2007)

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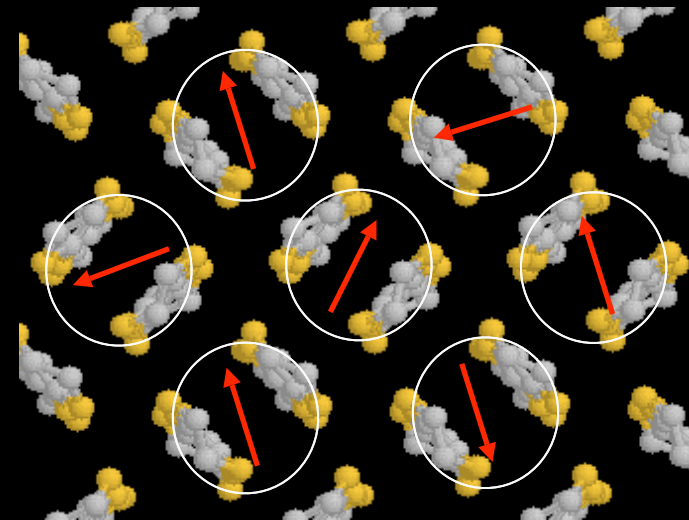
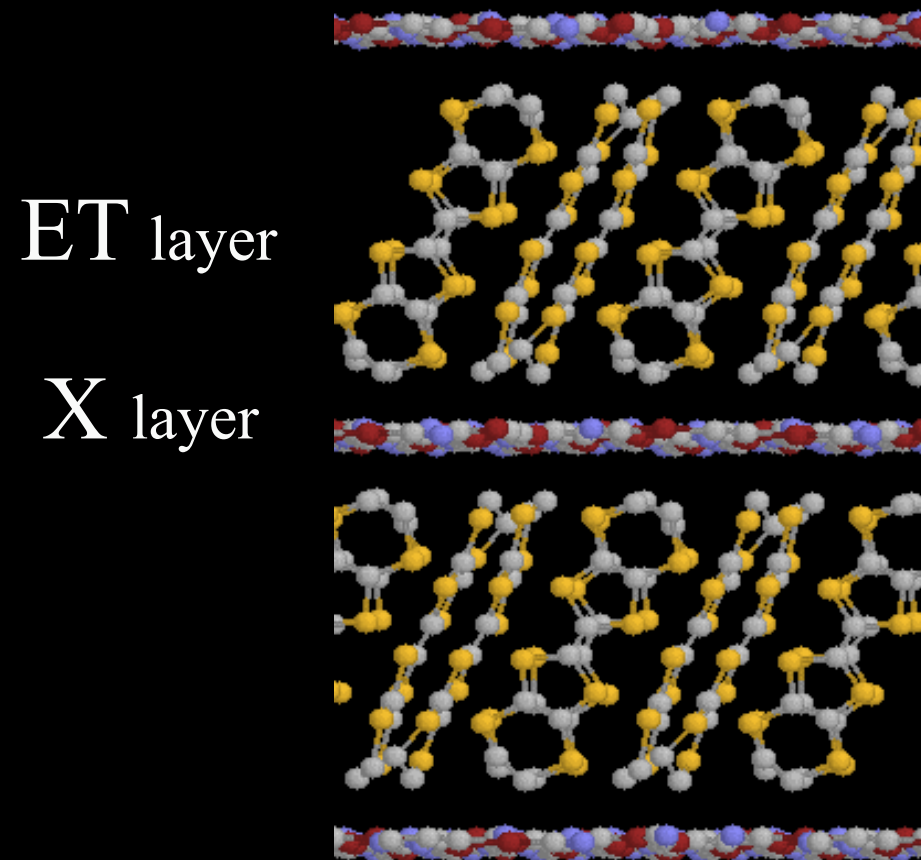


Pressure-
temperature
phase
diagram

M. Tamura, A. Nakao and R. Kato, *J. Phys. Soc. Japan* **75**, 093701 (2006)

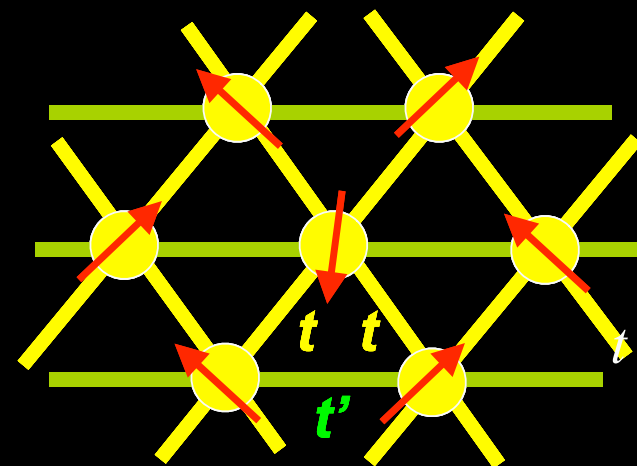
Y. Shimizu, H. Akimoto, H. Tsujii, A. Tajima, and R. Kato, *Phys. Rev. Lett.* **99**, 256403 (2007)

Q2D organics κ -(ET)₂X; spin-1/2 on triangular lattice



Kino & Fukuyama

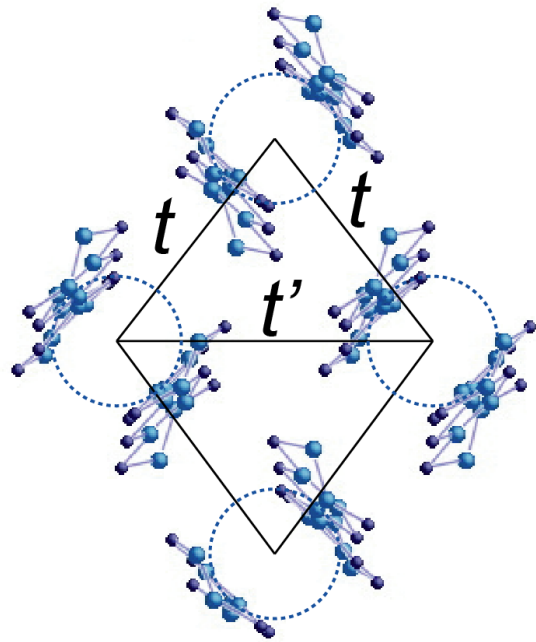
dimer model



Triangular lattice
Half-filled band

X ⁻	Ground State	t'/t
Cu ₂ (CN) ₃	Mott insulator	1.06
Cu[N(CN) ₂]Cl	Mott insulator	0.75
Cu[N(CN) ₂]Br	SC	0.68
Cu(NCS) ₂	SC	0.84

$\kappa\text{-(BEDT-TTF)}_2\text{Cu}_2(\text{CN})_3$

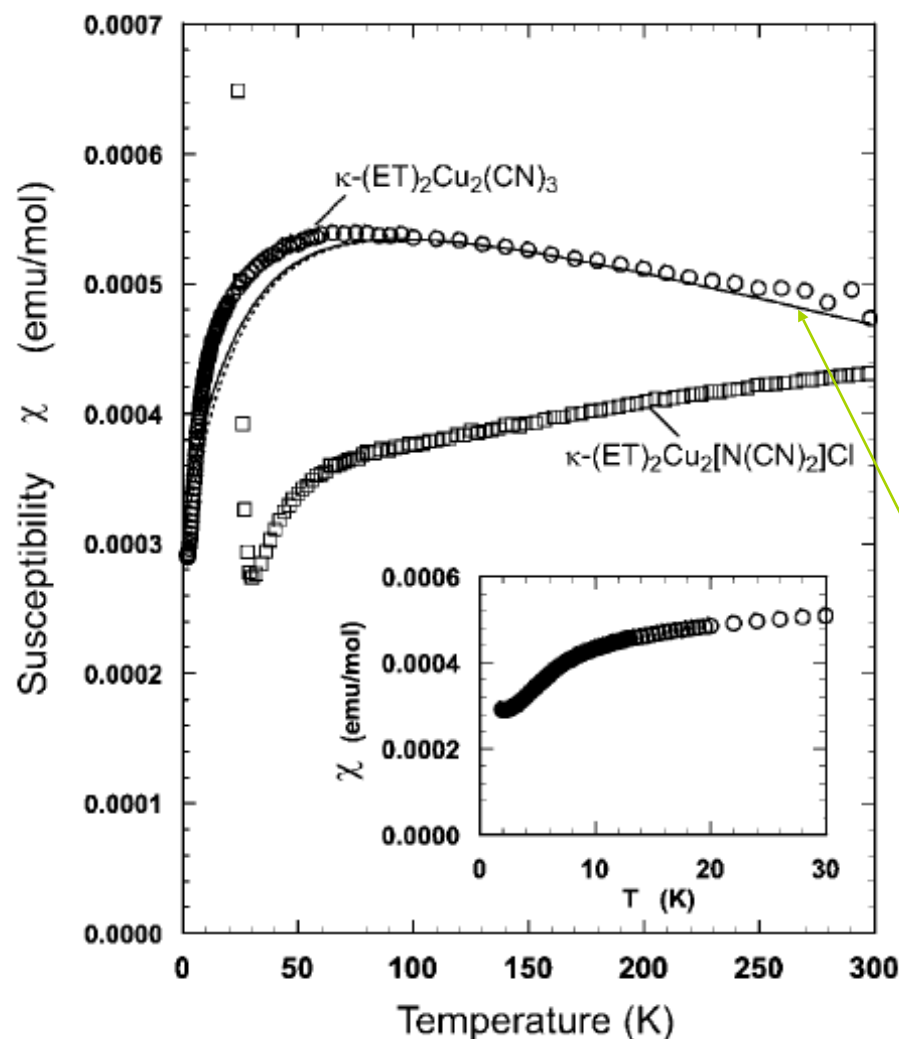


- face-to-face pairs of BEDT-TTF molecules form dimers by strong coupling.

- Dimers locate on a vertex of triangular lattice and ratio of the transfer integral is ~ 1 .

$$\frac{t'}{t} = 1.06, \frac{U}{t} = 8.2$$

- Charge +1 for each ET dimer; Half-filling Mott insulator.



- $\text{Cu}_2[\text{N}(\text{CN})_2]\text{Cl}$ ($t'/t = 0.75$, $U/t = 7.8$)

Néel order at $T_N = 27$ K

- $\text{Cu}_2(\text{CN})_3$ ($t'/t = 1.06$, $U/t = 8.2$)

No sign of magnetic order down to 1.9 K.

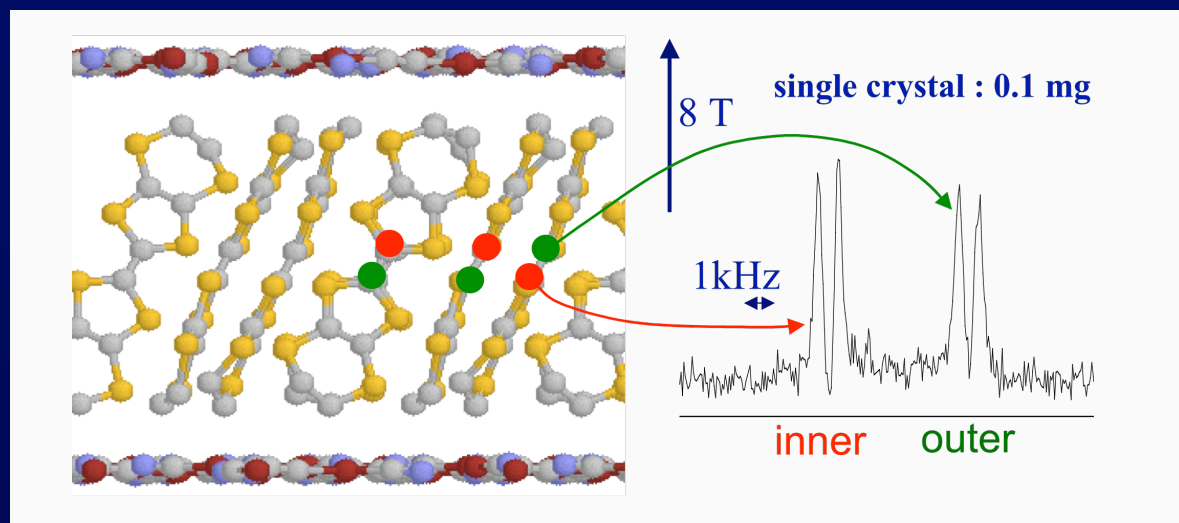
Heisenberg High-T Expansion
(PRL, 71 1629 (1993))

$J \sim 250$ K

Spin excitation in $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

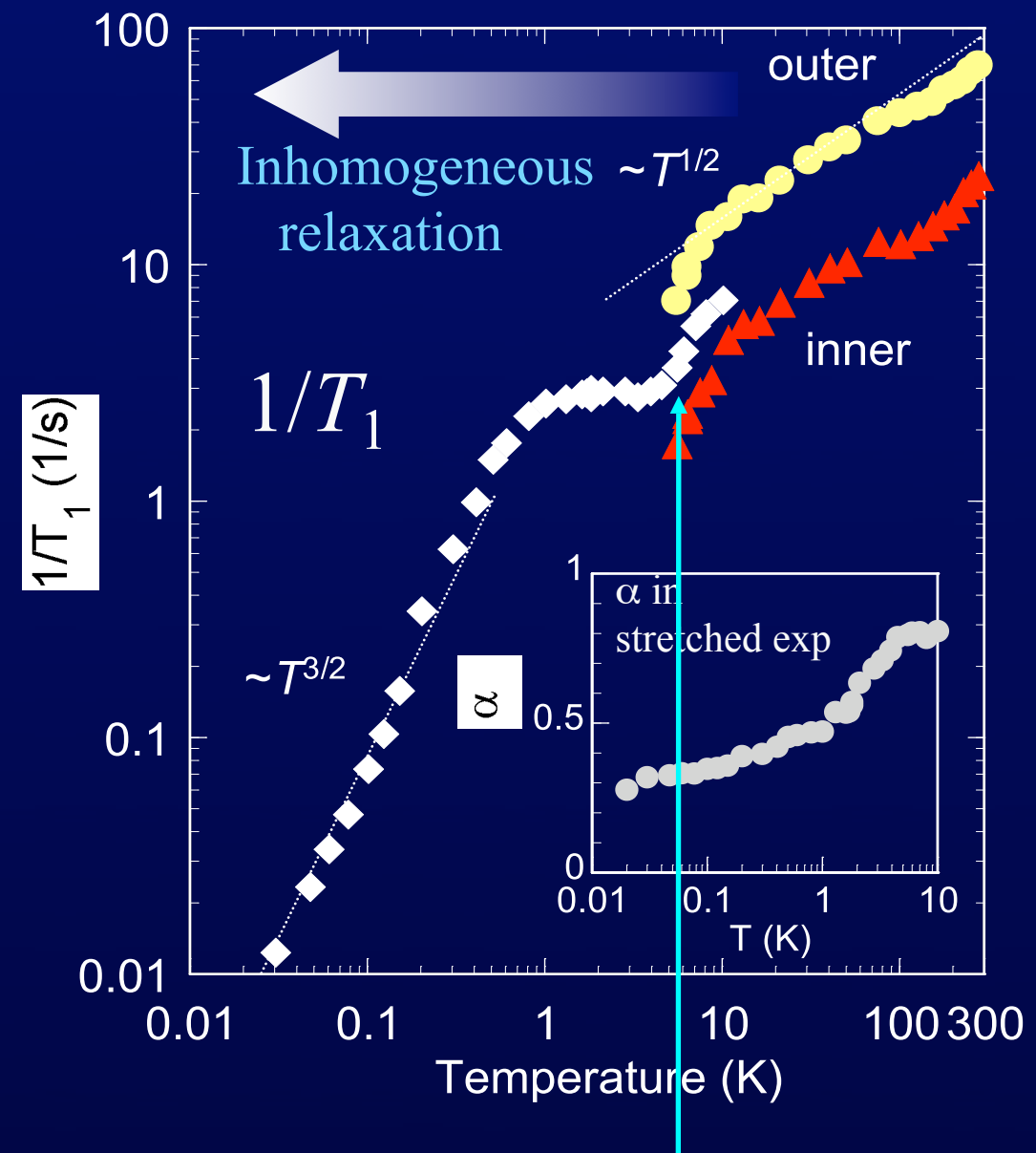
^{13}C NMR relaxation rate

Shimizu *et al.*, PRB 70 (2006) 060510



$1/T_1 \sim$ power law of T

Low-lying spin excitation at low- T



Anomaly at 5-6 K

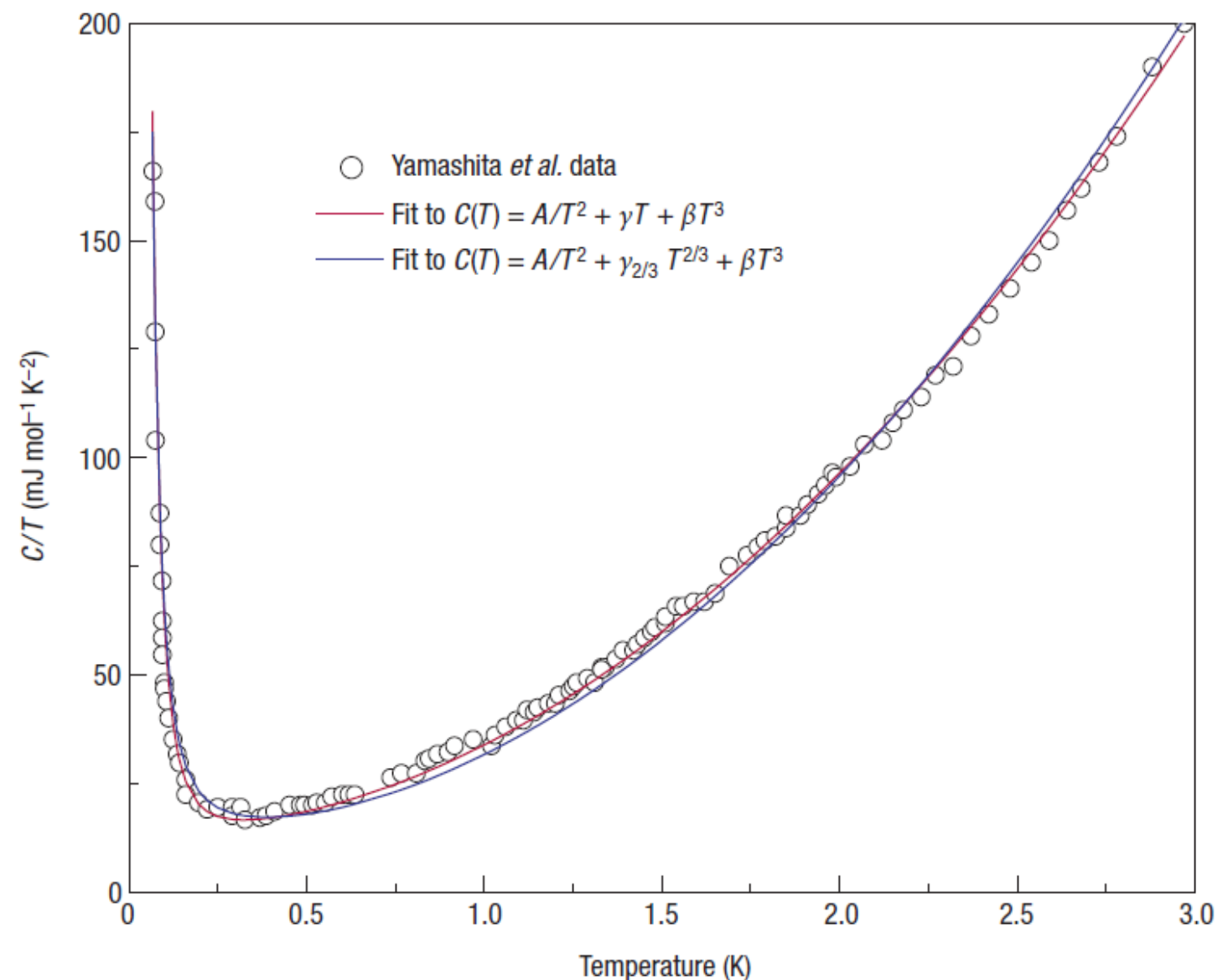
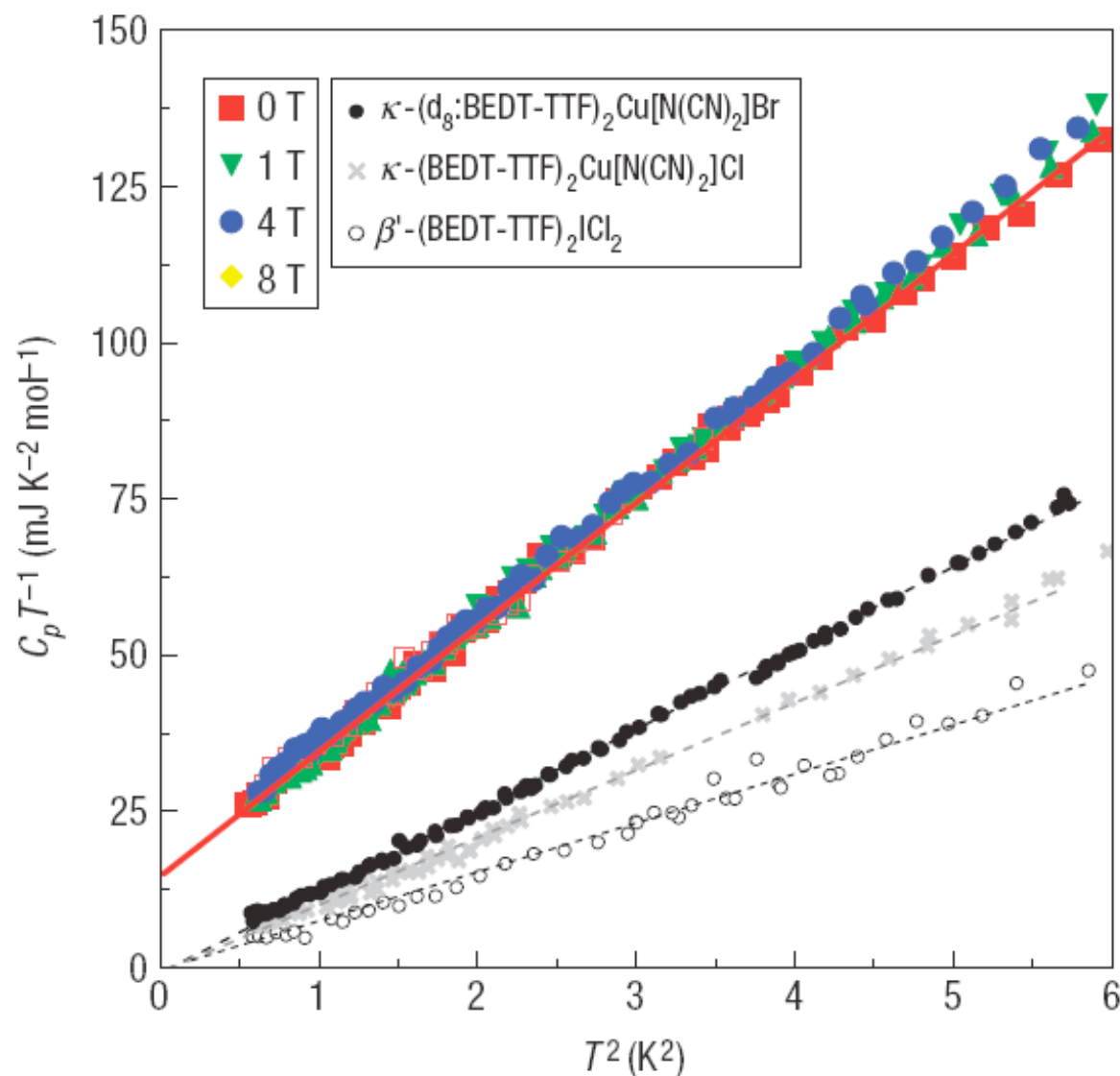
Heat capacity measurements

Thermodynamic properties of a spin-1/2
spin-liquid state in a κ -type organic salt

SATOSHI YAMASHITA¹, YASUHIRO NAKAZAWA^{1,2*}, MASAHARU OGUNI³, YUGO OSHIMA^{2,4},
HIROYUKI NOJIRI^{2,4}, YASUHIRO SHIMIZU⁵, KAZUYA MIYAGAWA^{2,6} AND KAZUSHI KANODA^{2,6}

$$\gamma = 15 \text{ mJ} / \text{K}^2 \text{mol}$$

Evidence for Gapless spinon?

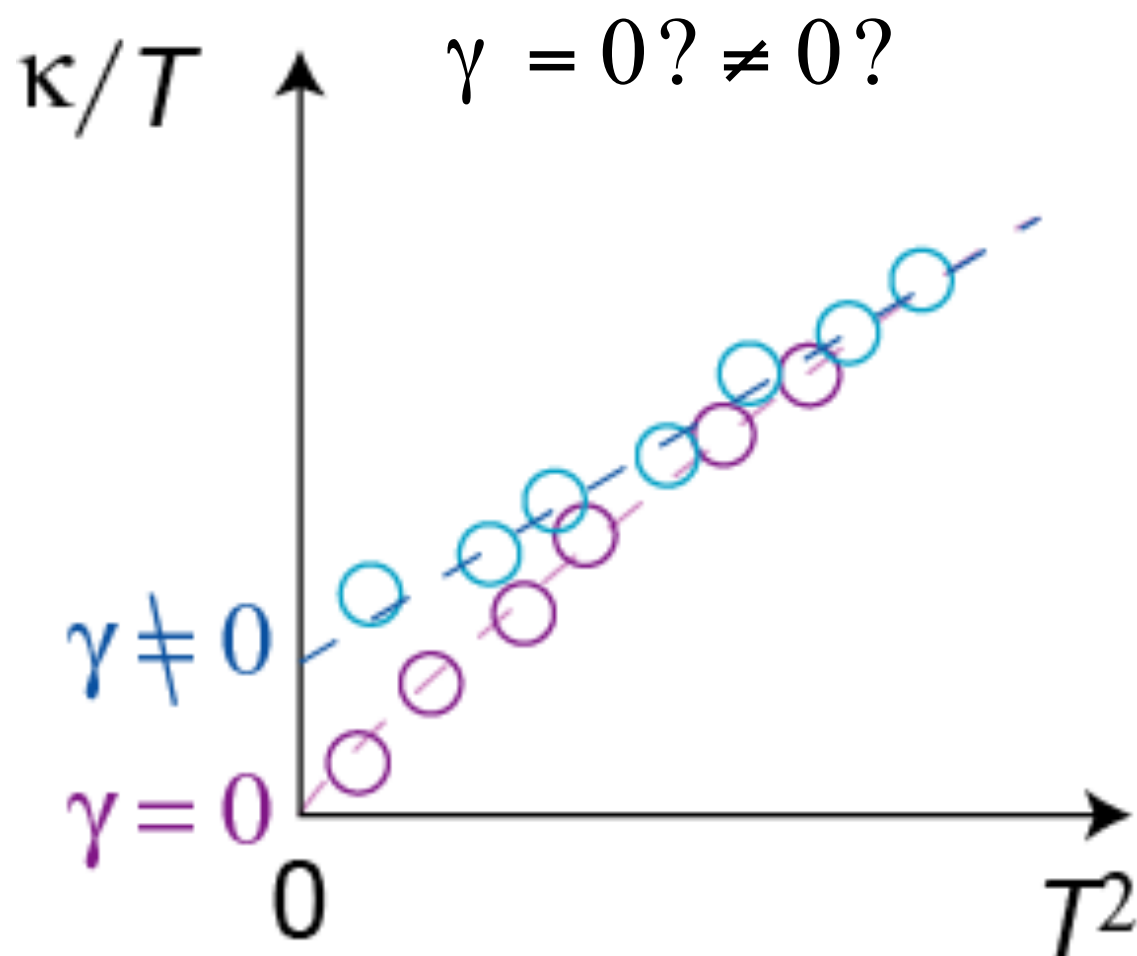


Thermal-Transport Measurements

Only itinerant excitations carrying entropy can be measured without localized ones

- no impurity contamination
 - $1/T_1$, χ measurement $\leftarrow$ free spins
 - Heat capacity $\leftarrow$ Schottky contamination

Best probe to reveal the low-lying excitation at low temperatures.

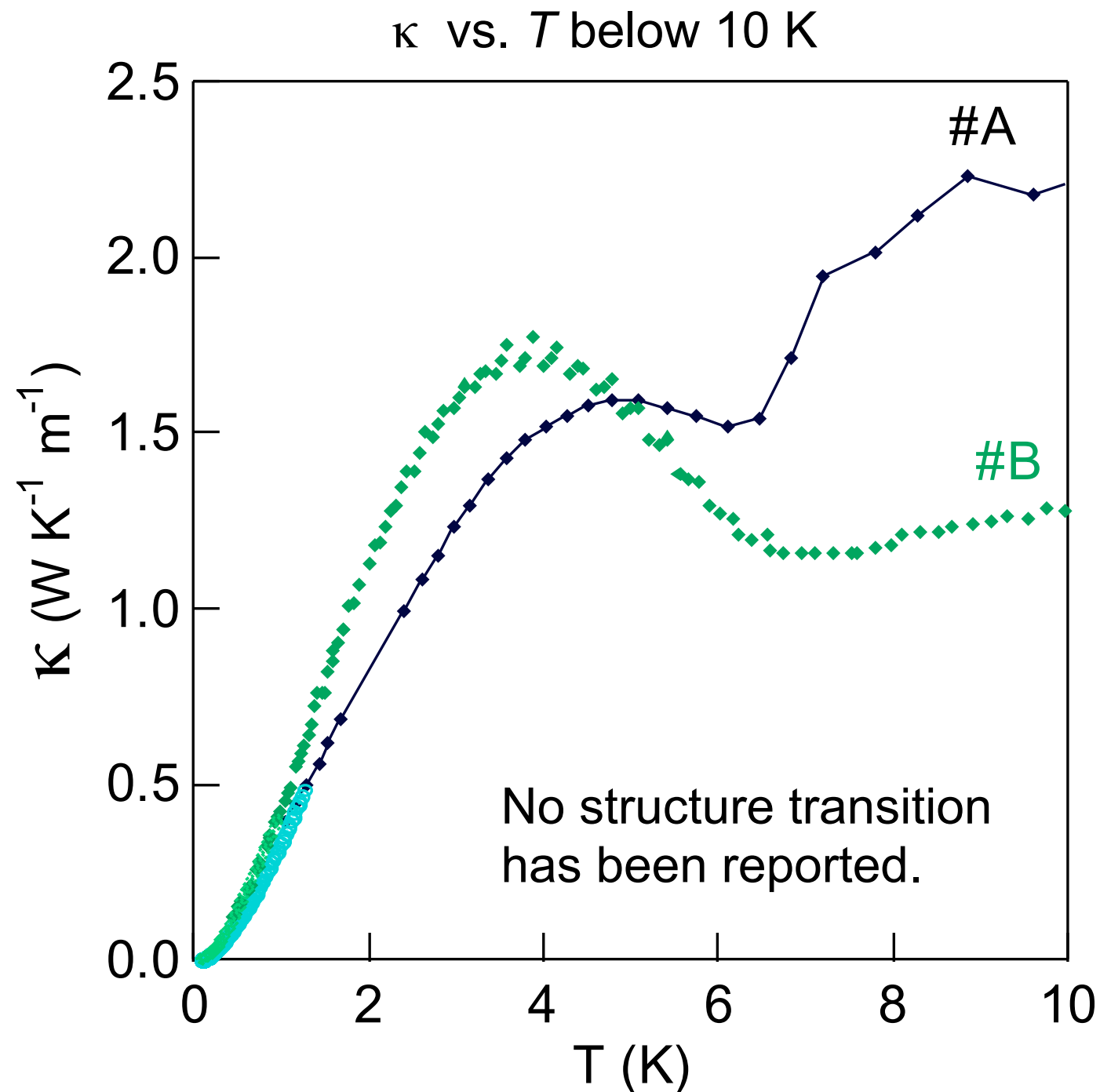


$$\frac{\kappa}{T} \approx \frac{1}{T} (C \propto \gamma + \beta T^2) \propto \gamma + \beta T$$

$C \propto \gamma T + \beta T^3$
 γ : Gapless spin liquid
 (Spinon)
 β : Phonon

- What is the low-lying excitation of the quantum spin liquid found in κ -(BEDT-TTF)₂Cu₂(CN)₃.
- Gapped or Gapless spin liquid? Spinon with a Fermi surface?

Thermal Conductivity below 10K

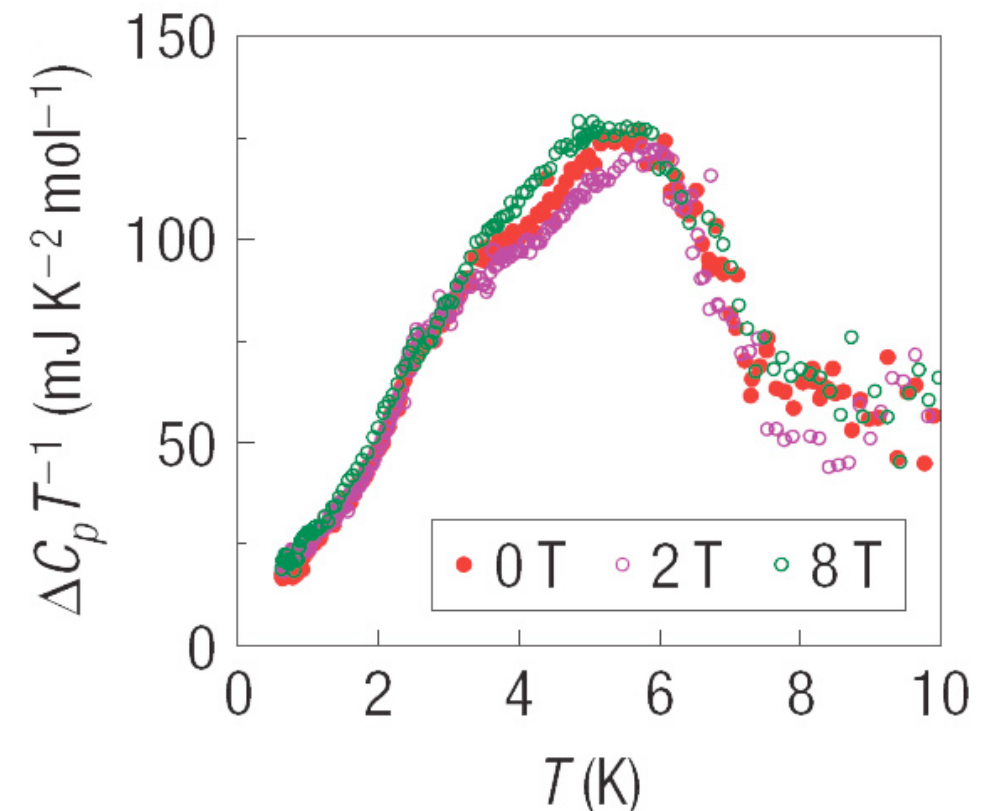


Similar to

- $1/T_1$ by ^1H NMR
- Heat capacity



Difference of heat capacity between
 $X = \text{Cu}_2(\text{CN})_3$ and $\text{Cu}(\text{NCS})_2$
(superconductor).



S. Yamashita, et al., Nature Physics 4, 459 - 462 (2008)

- Magnetic contribution to κ
- Phase transition or crossover?
 - Chiral order transition?
 - Instability of spinon Fermi surface?

M. Yamashita *et al.*, Nature Physics online, doi:10.1038/nphys1134

Thermal Conductivity below 300 mK

- Convex, non- T^3 dependence in κ
- Magnetic fields enhance κ

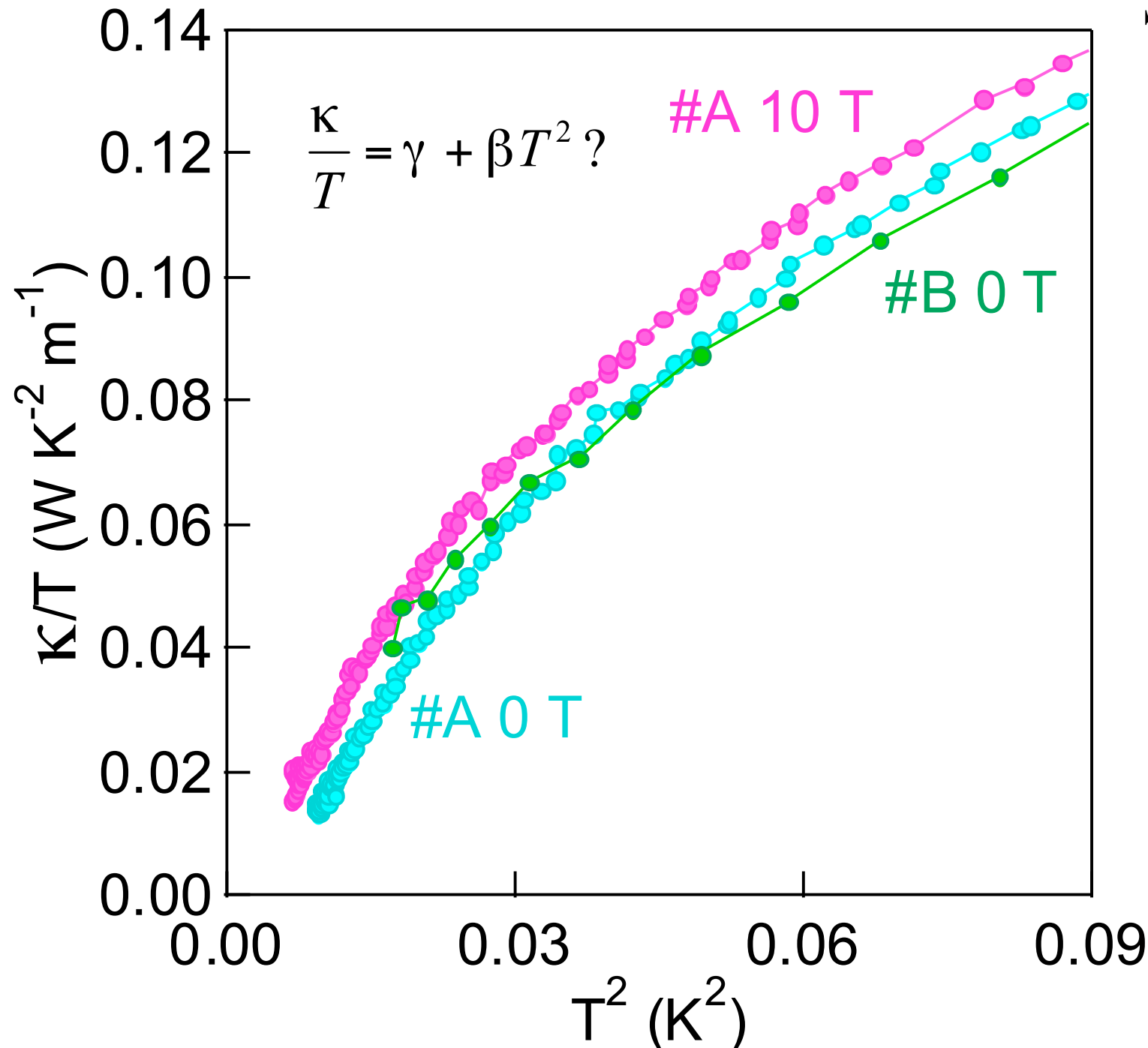


$$\kappa = \kappa_{phonon} + \kappa_{mag}$$

$$(\kappa_{phonon} \propto T^3 \text{ in low } T)$$

Substantial portion of κ_{mag} in κ

κ/T vs T^2 Plot

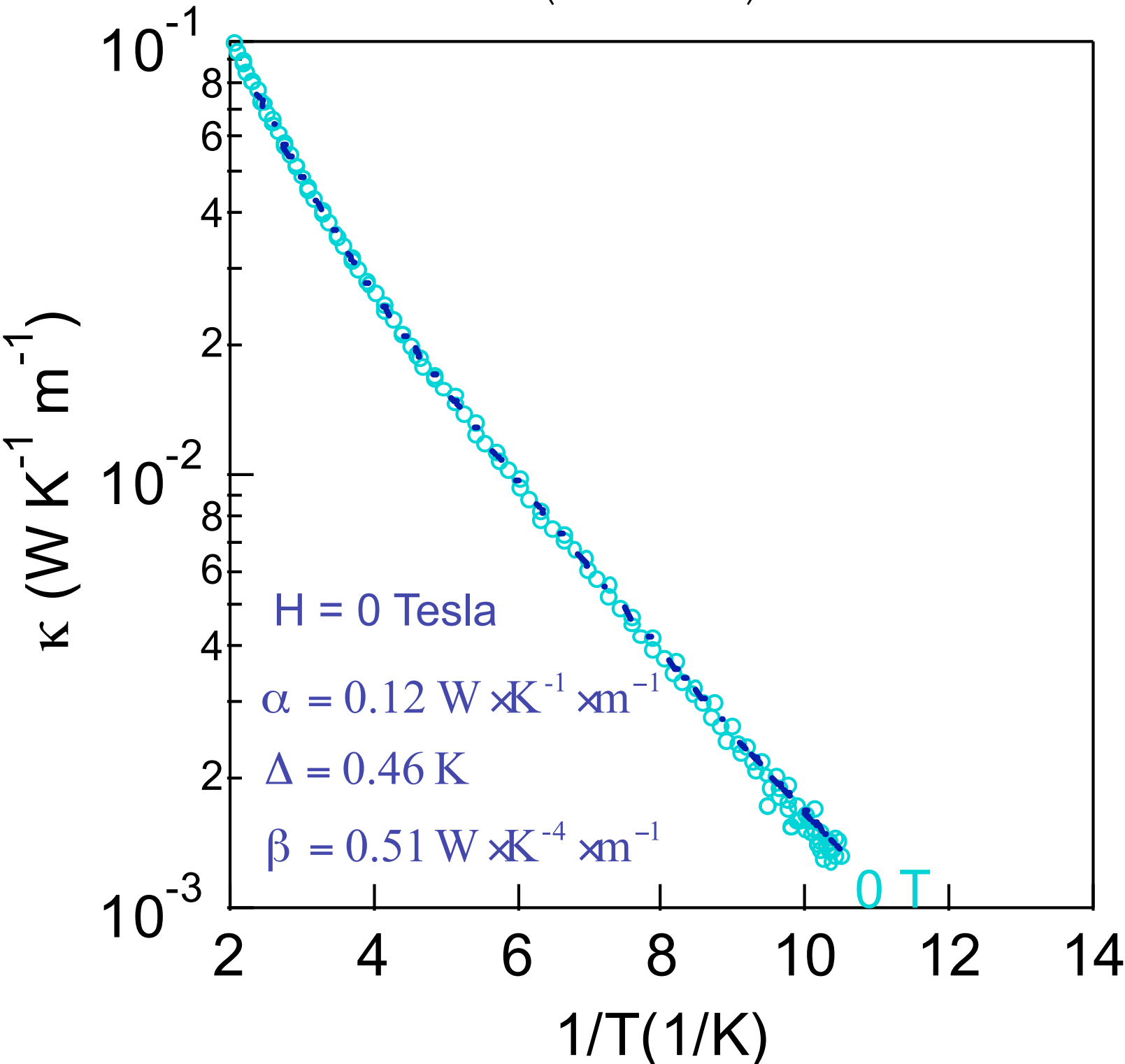


$$\underline{\gamma = 0}$$

Note: phonon contribution has no effect on this conclusion.

Arrhenius plot

$$\kappa = \alpha \exp\left(-\frac{\Delta}{k_B T}\right) + \beta T^3$$



• Arrhenius behavior for $T < \Delta$!

• Tiny gap

➤ $\Delta = 0.46 \text{ K} \sim J/500$

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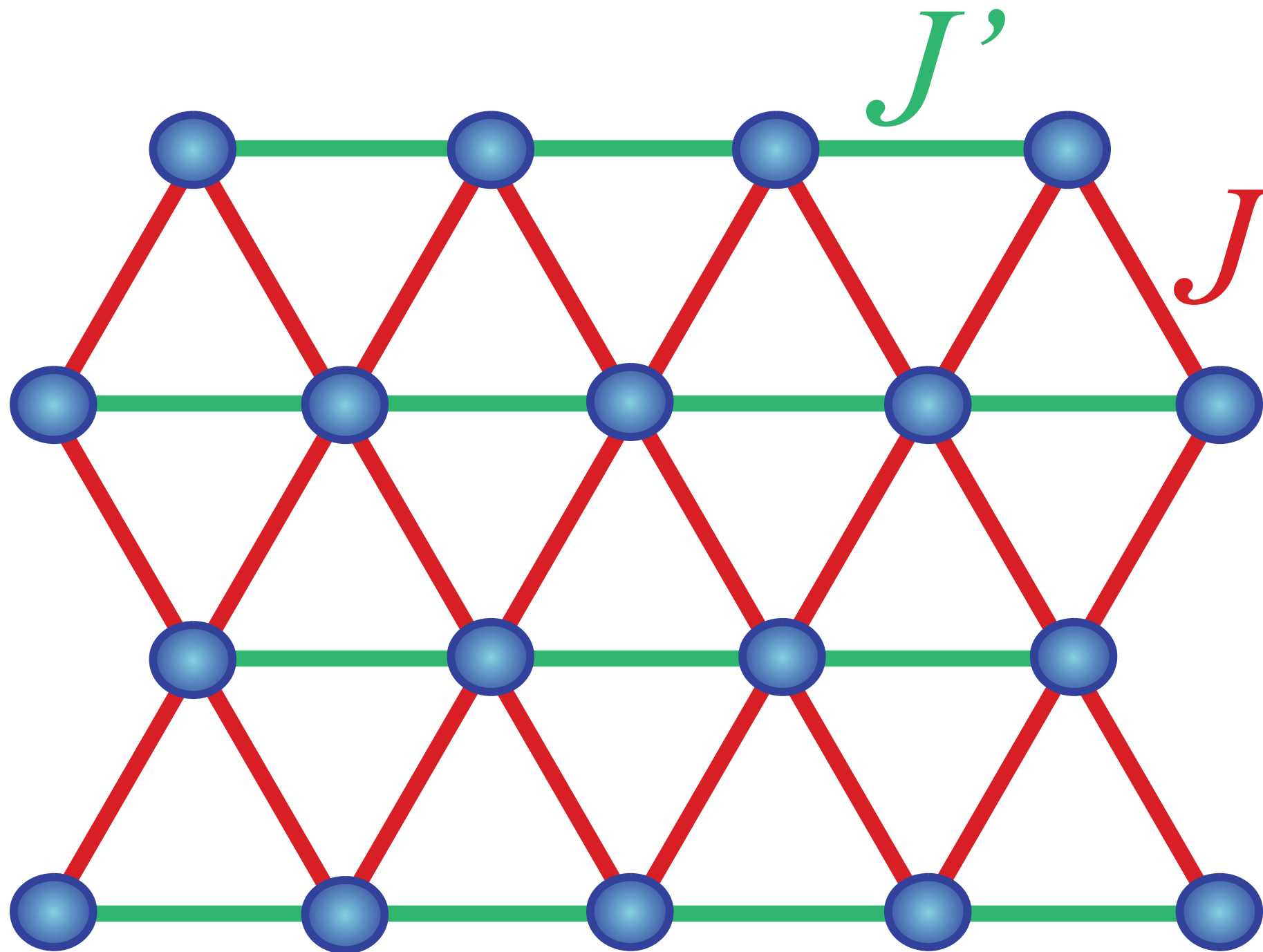
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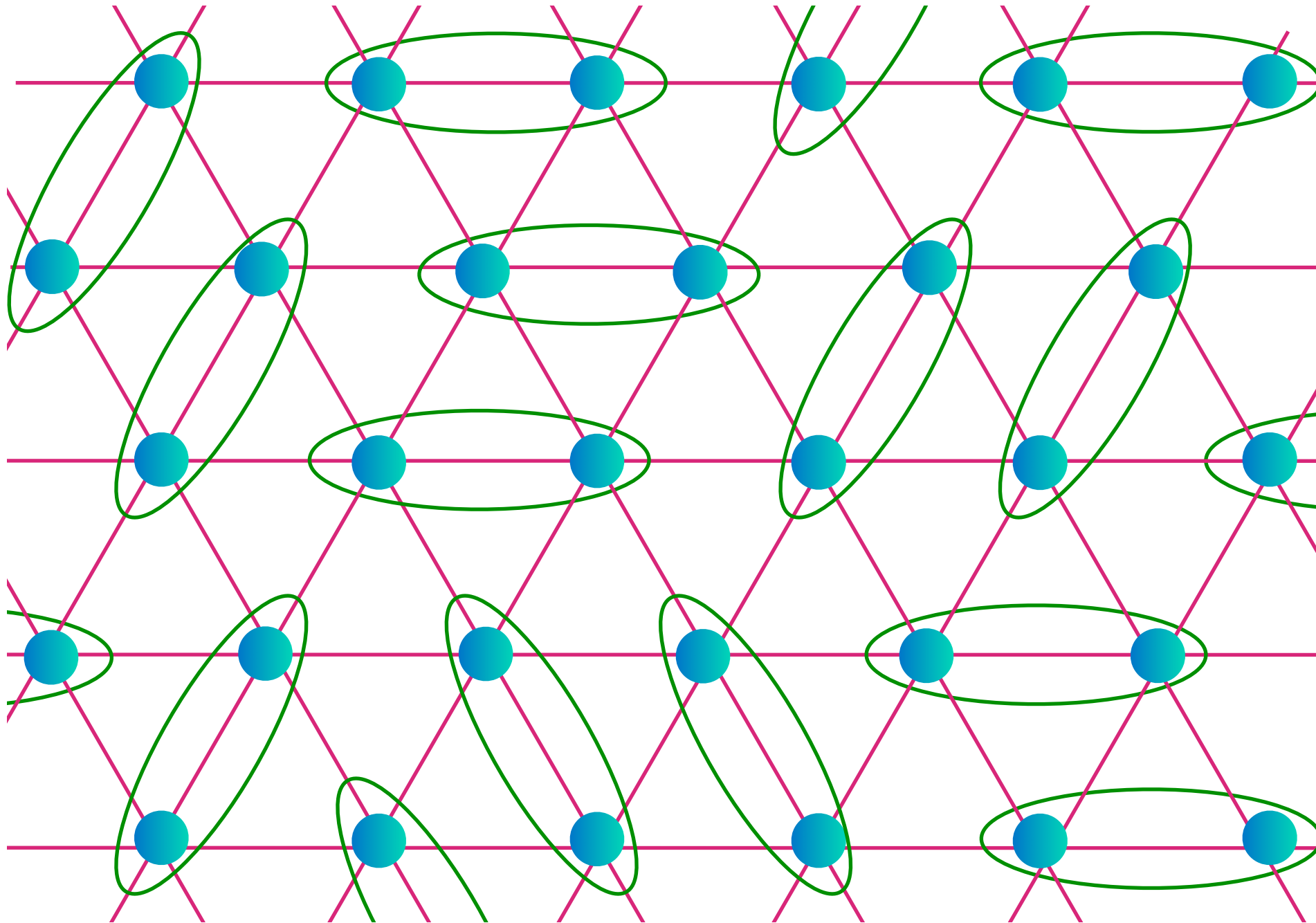
Anisotropic triangular lattice antiferromagnet



Triangular lattice antiferromagnet

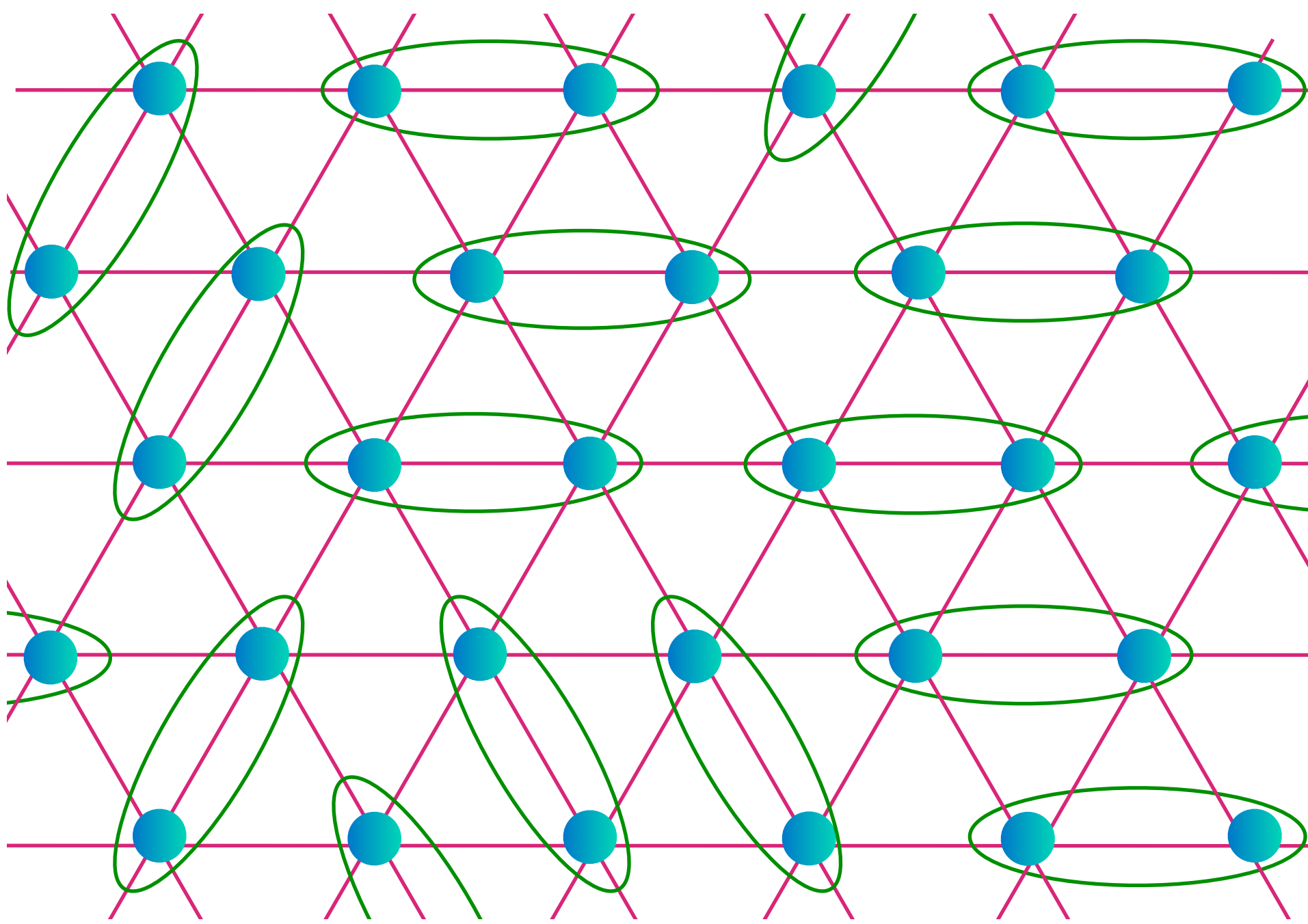
Spin liquid obtained in a generalized spin model with $S=1/2$ per unit cell


$$= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$



Triangular lattice antiferromagnet

Spin liquid obtained in a generalized spin model with $S=1/2$ per unit cell



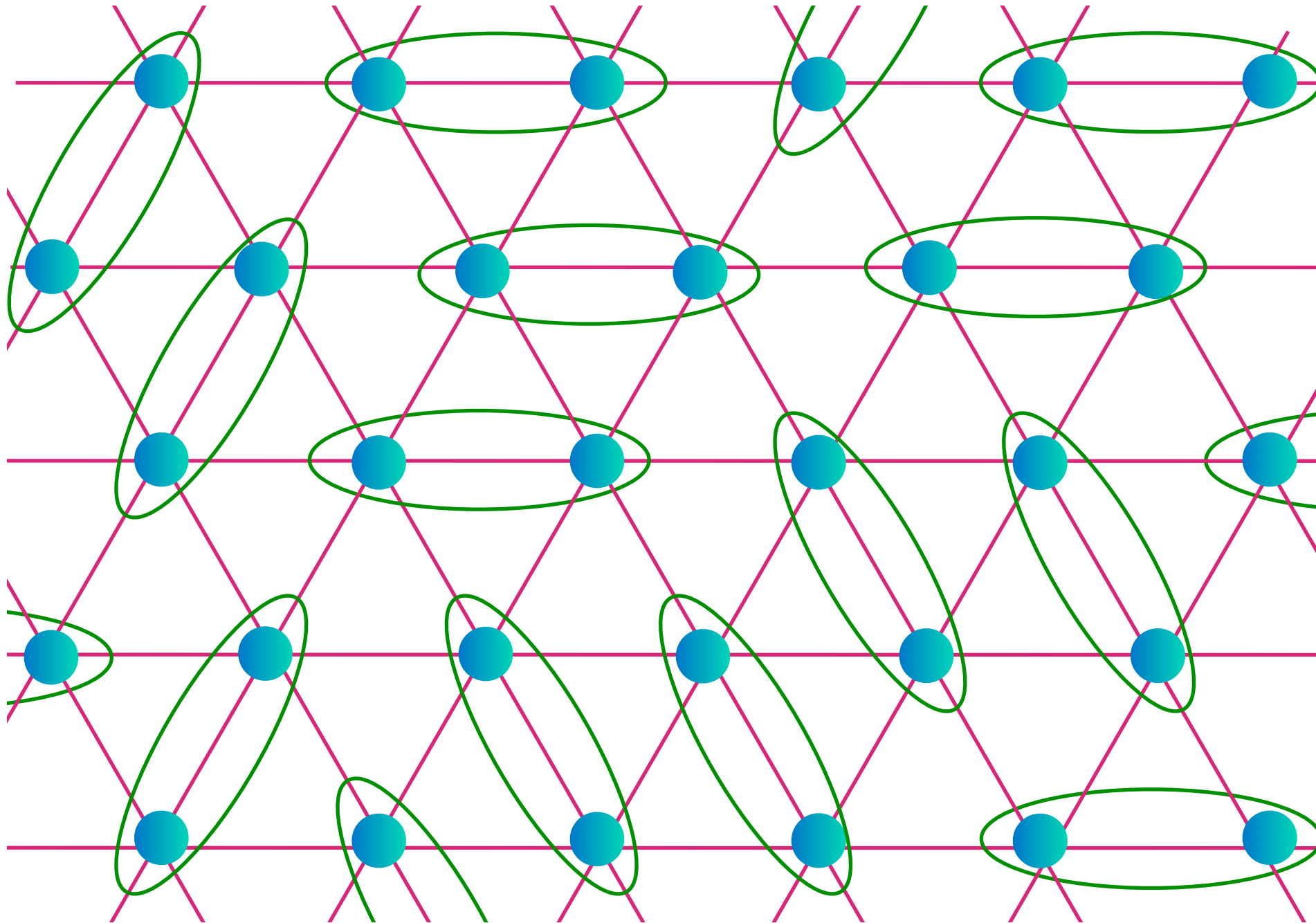
The diagram shows a triangular lattice of blue spheres (spins) connected by magenta lines. Green ellipses are drawn around pairs of sites, indicating a dimerized state. The ellipses are oriented in various directions, some horizontally, some vertically, and some diagonally, representing different dimer configurations.

$$= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

Triangular lattice antiferromagnet

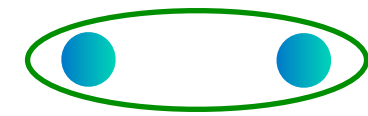
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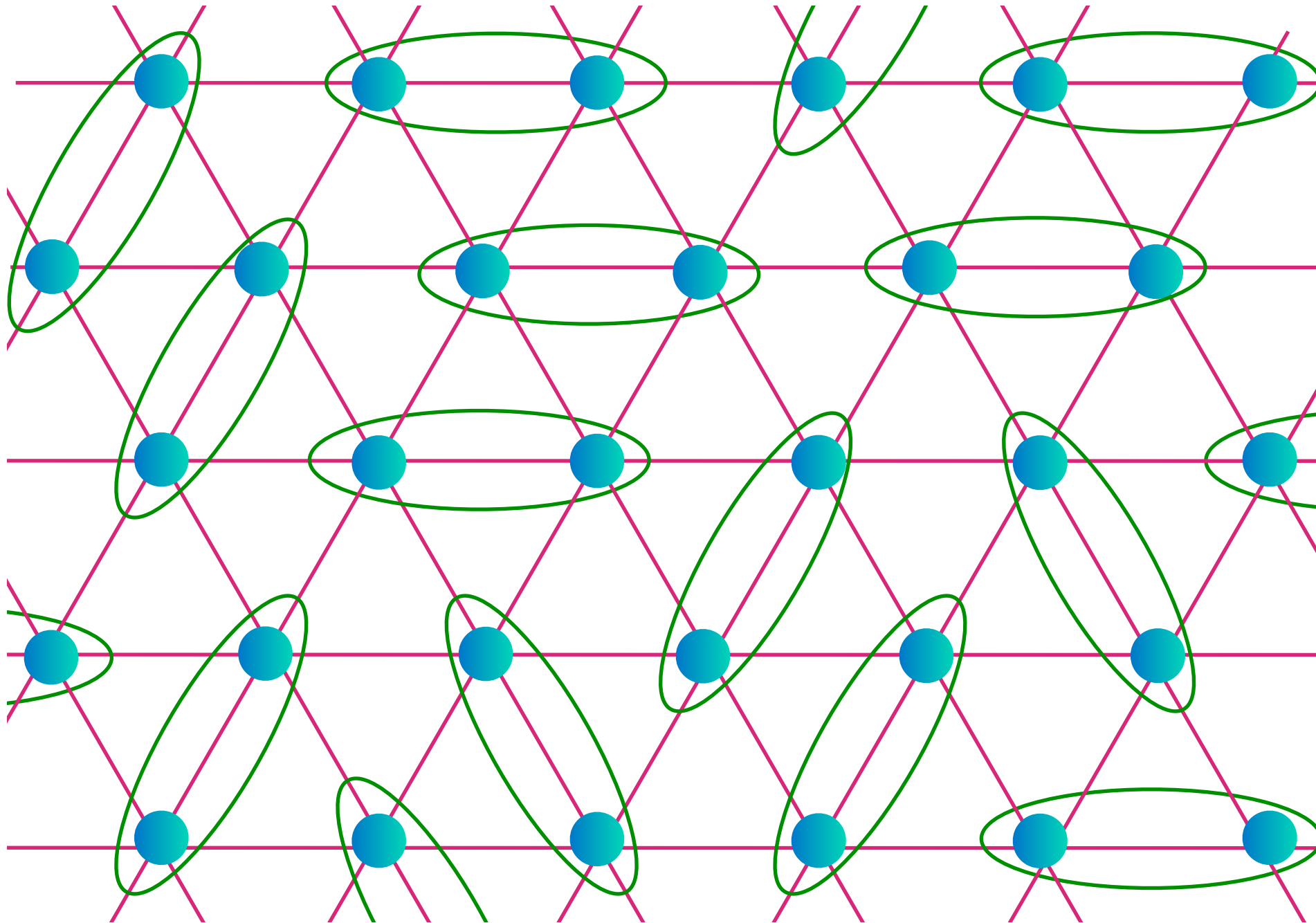

$$= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$



Triangular lattice antiferromagnet

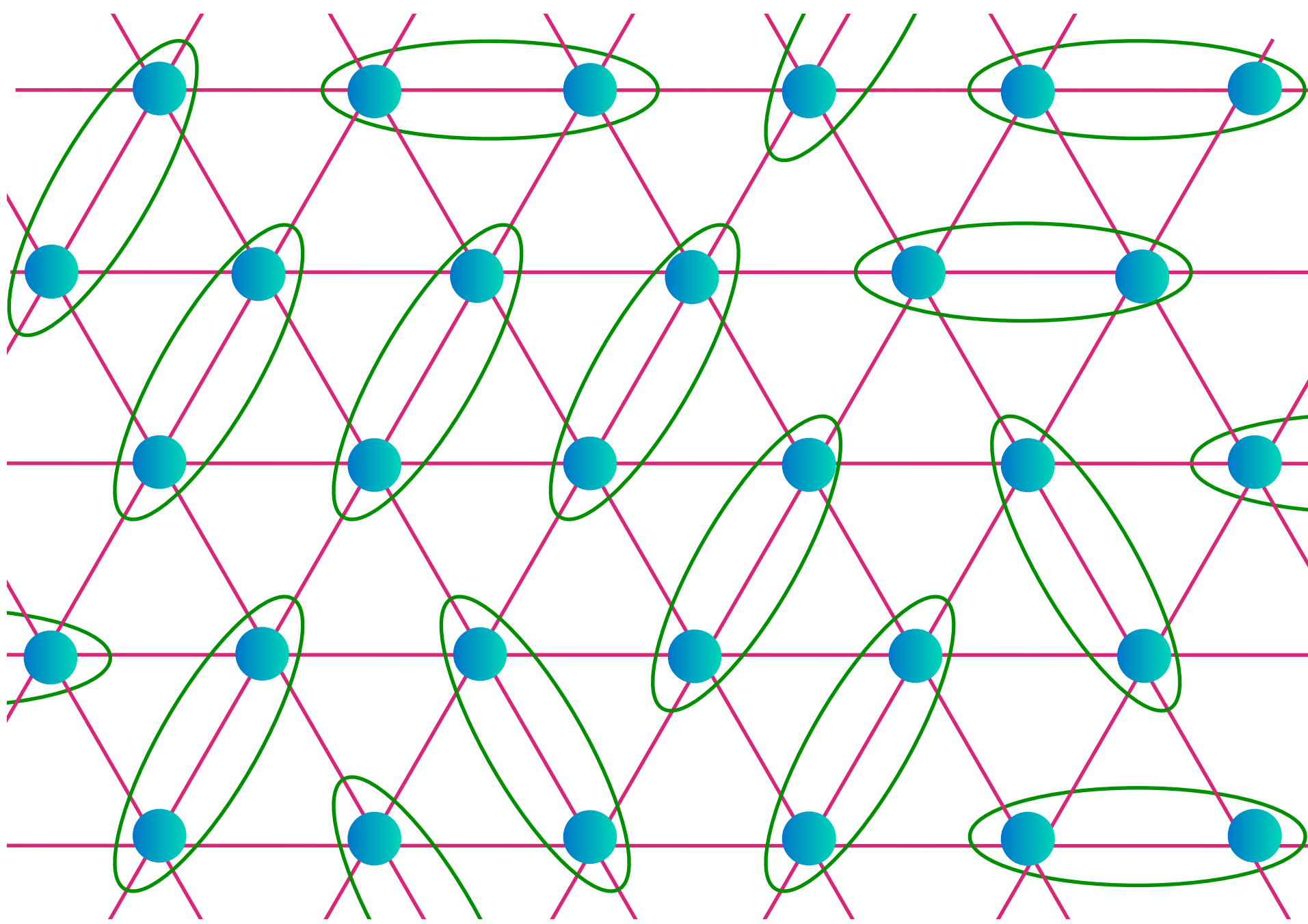
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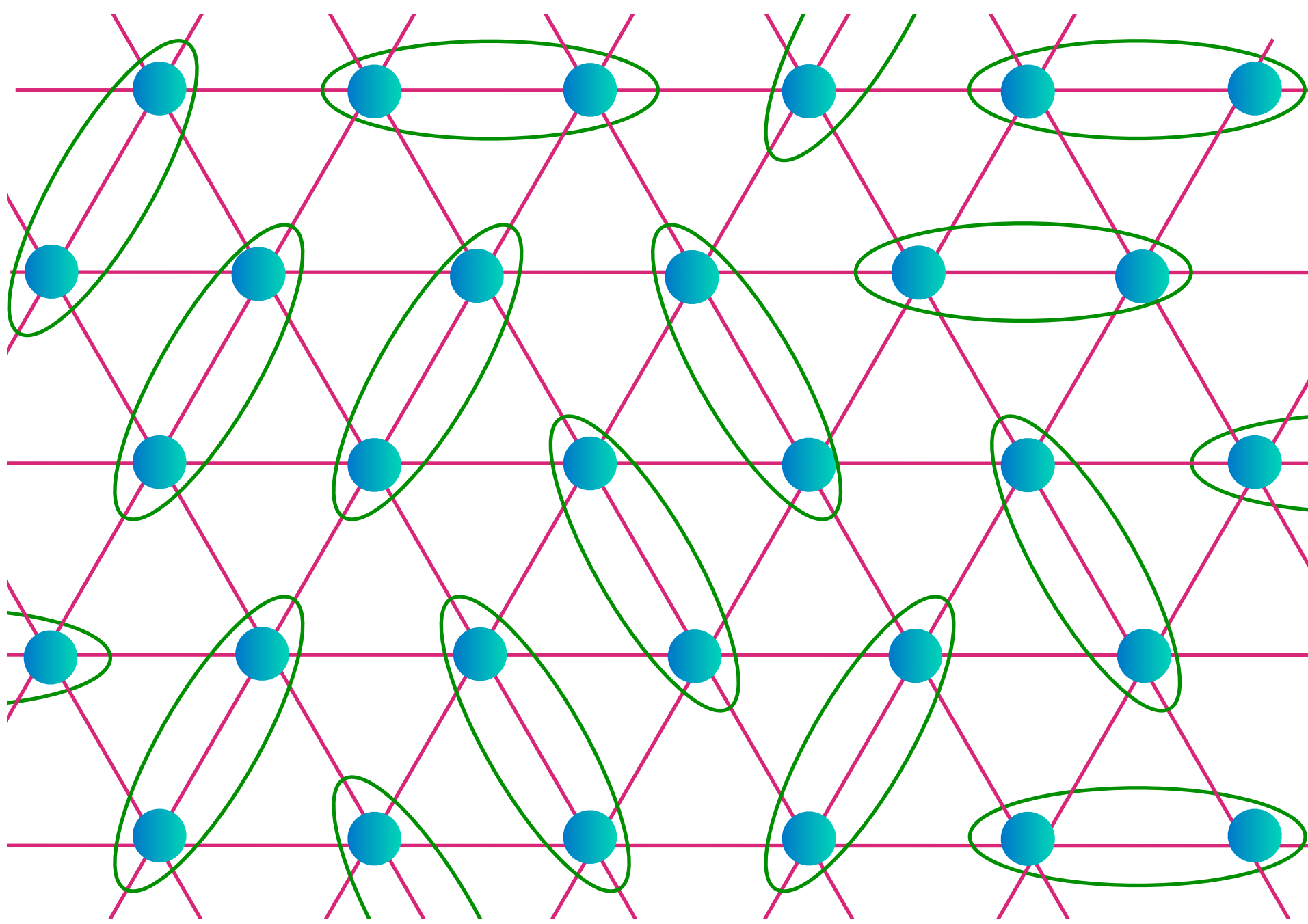


The diagram shows a triangular lattice of blue spheres (sites) connected by magenta lines. Green ellipses are drawn around pairs of sites, representing the spin liquid state. The ellipses are oriented in various directions, indicating a disordered or fluctuating state.

$$= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

Triangular lattice antiferromagnet

Spin liquid obtained in a generalized spin model with $S=1/2$ per unit cell

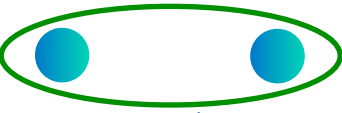


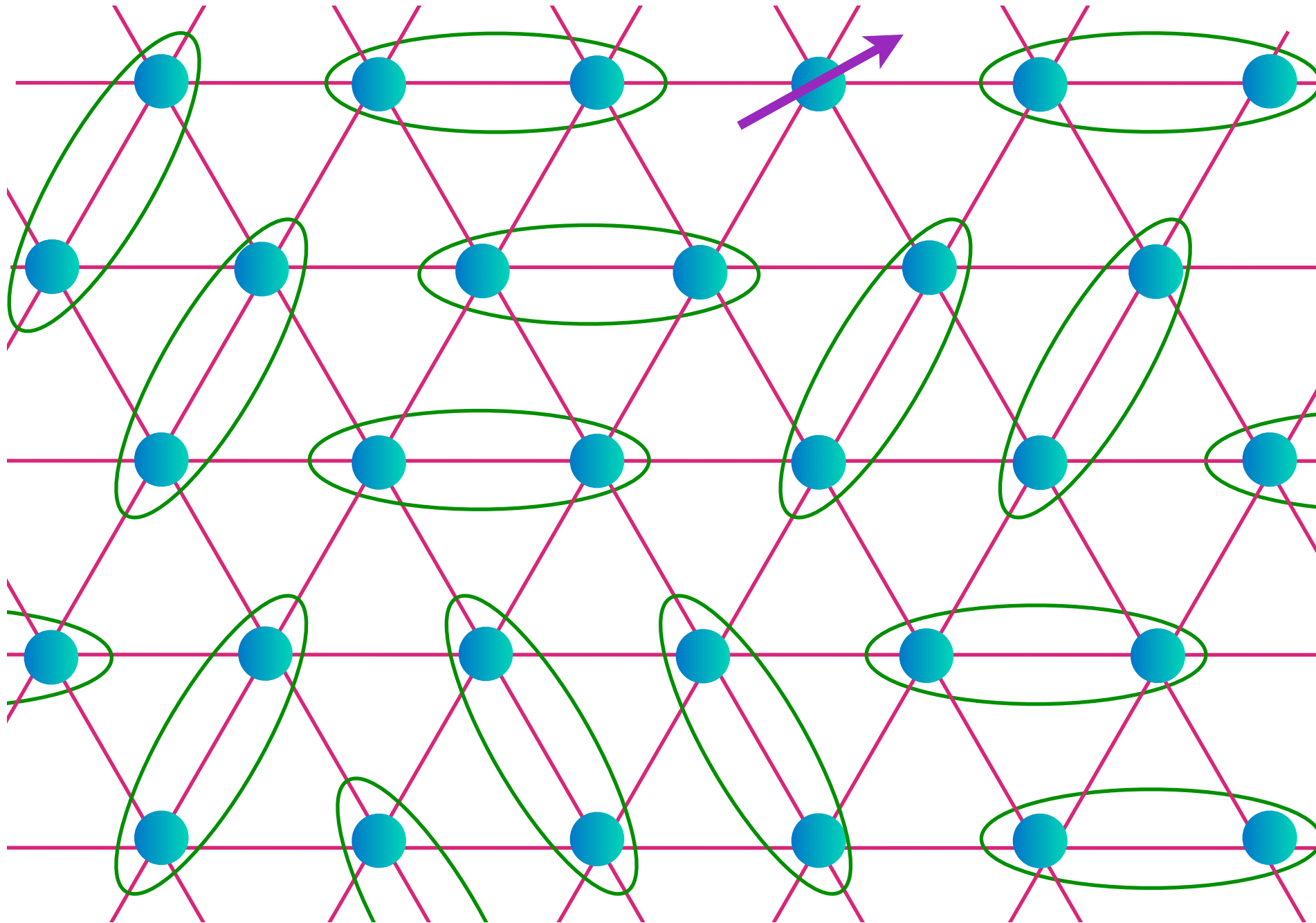
The diagram shows a triangular lattice of blue spheres (sites) connected by magenta lines. Green ellipses are drawn around pairs of sites, representing the unit cells of the spin liquid. The ellipses are oriented in various directions, illustrating the lack of long-range order.

$$= \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$

Excitations of the Z_2 Spin liquid

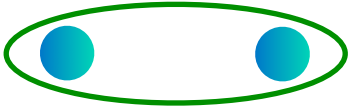
A spinon

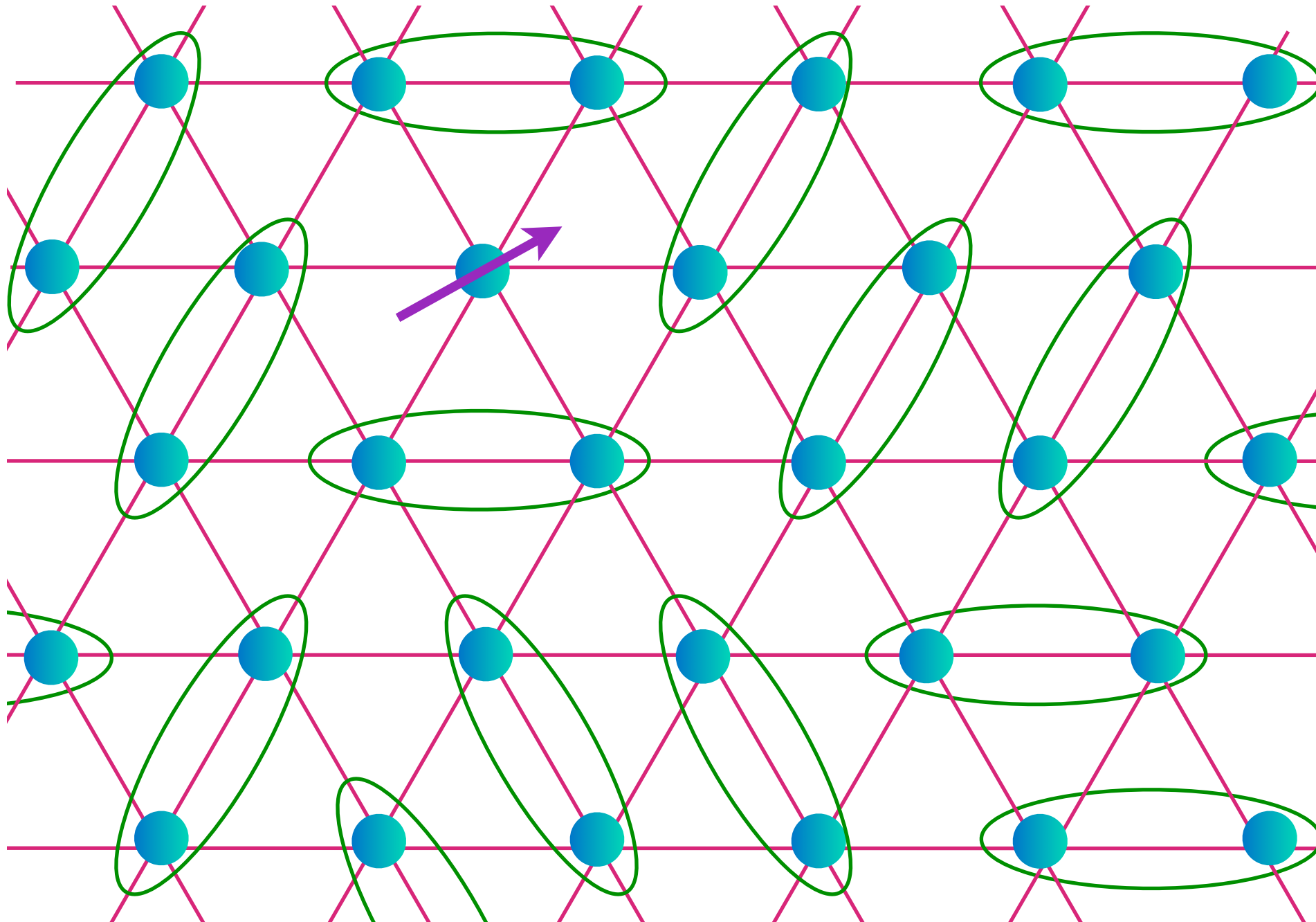

$$= \frac{1}{\sqrt{2}} \left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$



Excitations of the Z_2 Spin liquid

A spinon

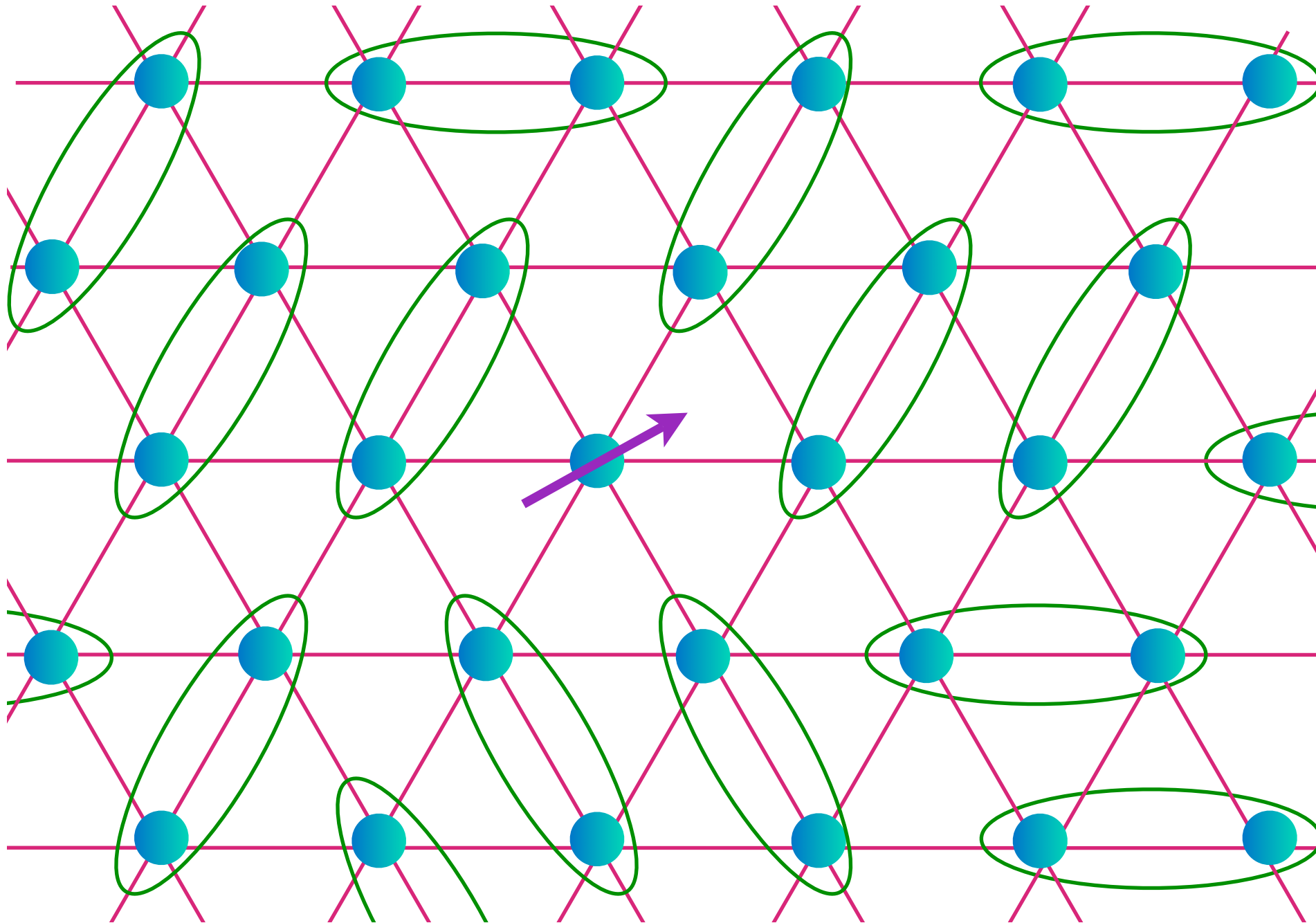

$$= \frac{1}{\sqrt{2}} \left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$



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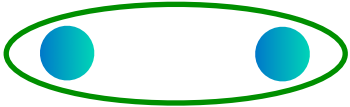
A spinon

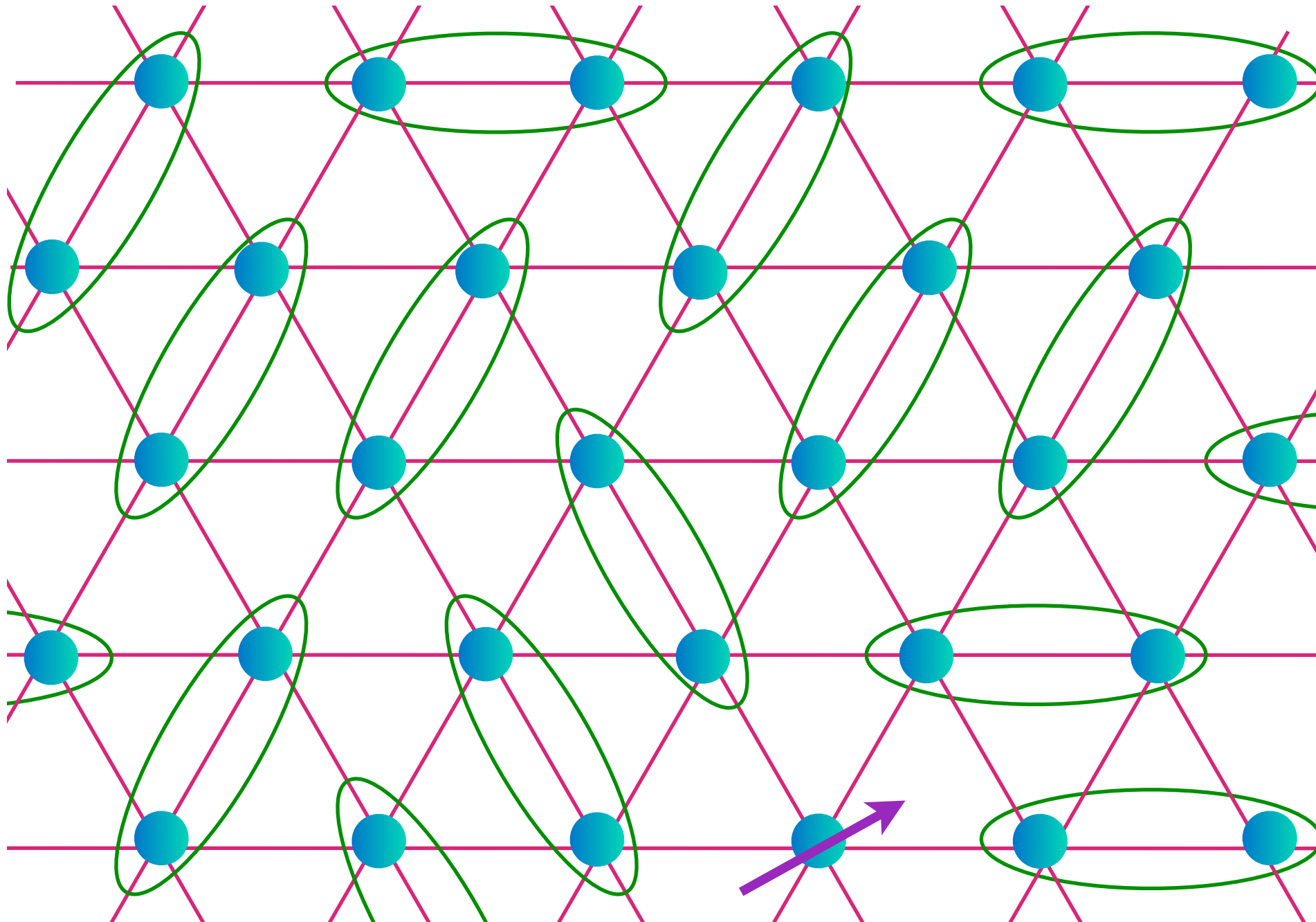

$$= \frac{1}{\sqrt{2}} \left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$



Excitations of the Z_2 Spin liquid

A spinon


$$= \frac{1}{\sqrt{2}} \left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle \right)$$



Excitations of the Z_2 Spin liquid

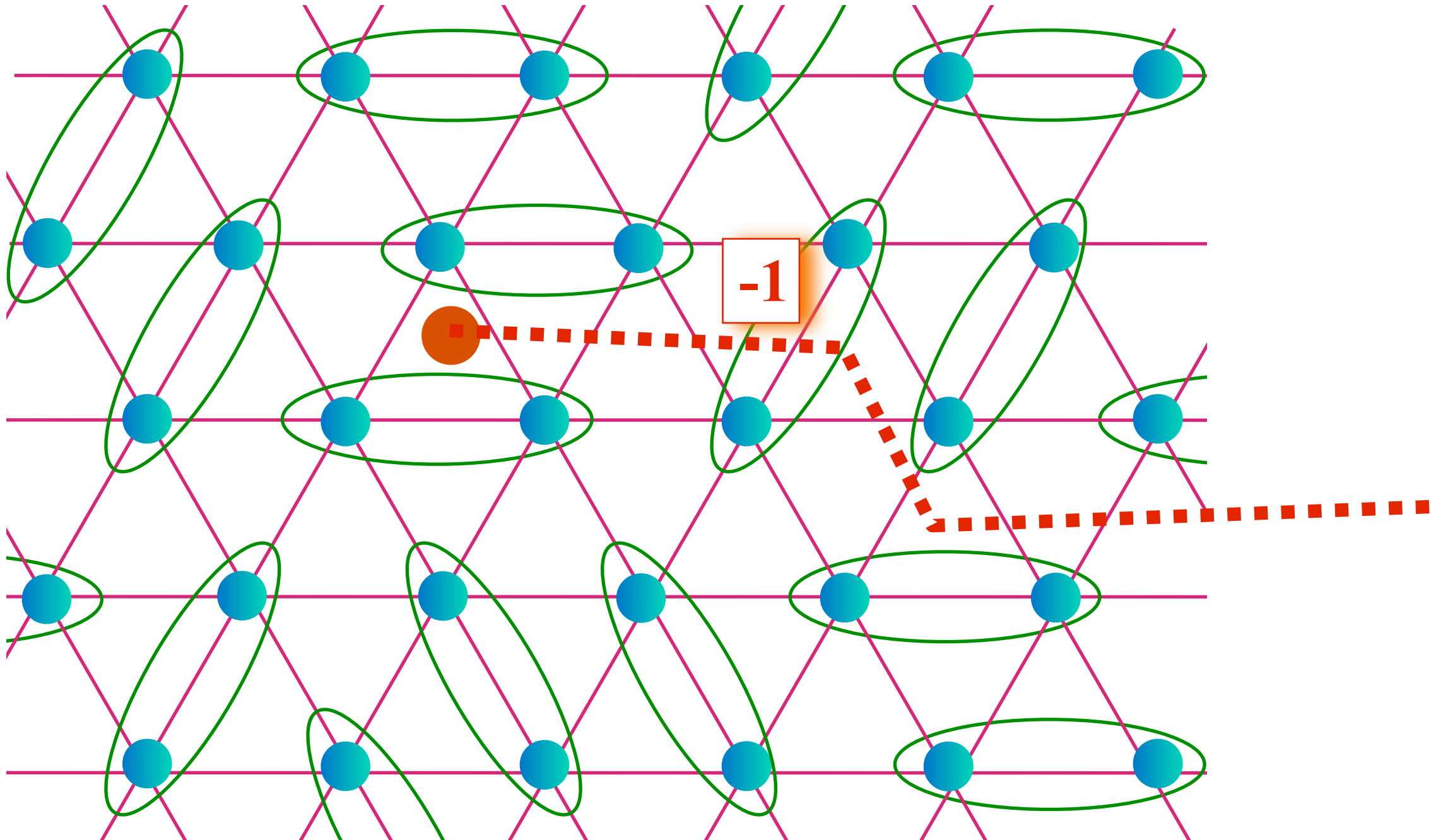
A vison

- A characteristic property of a Z_2 spin liquid is the presence of a spinon pair condensate
- A vison is an Abrikosov vortex in the pair condensate of spinons
- Visions are the dark matter of spin liquids: they likely carry most of the energy, but are very hard to detect because they do not carry charge or spin.

Excitations of the Z_2 Spin liquid

A vison

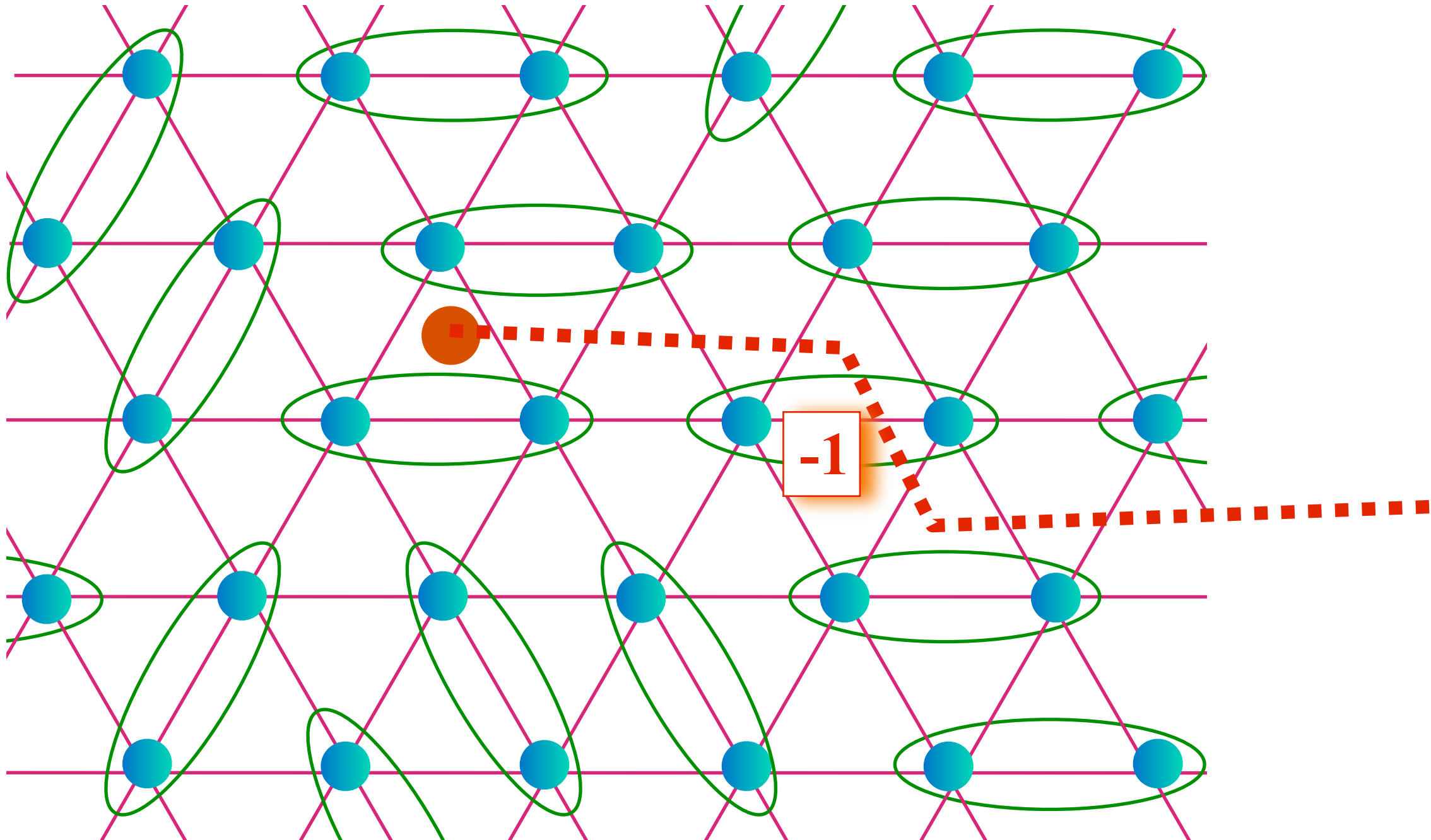
$$\begin{array}{c} \text{---} \bullet \text{---} \bullet \text{---} \\ \text{---} \end{array} = \frac{1}{\sqrt{2}} (|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$$



Excitations of the Z_2 Spin liquid

A vison

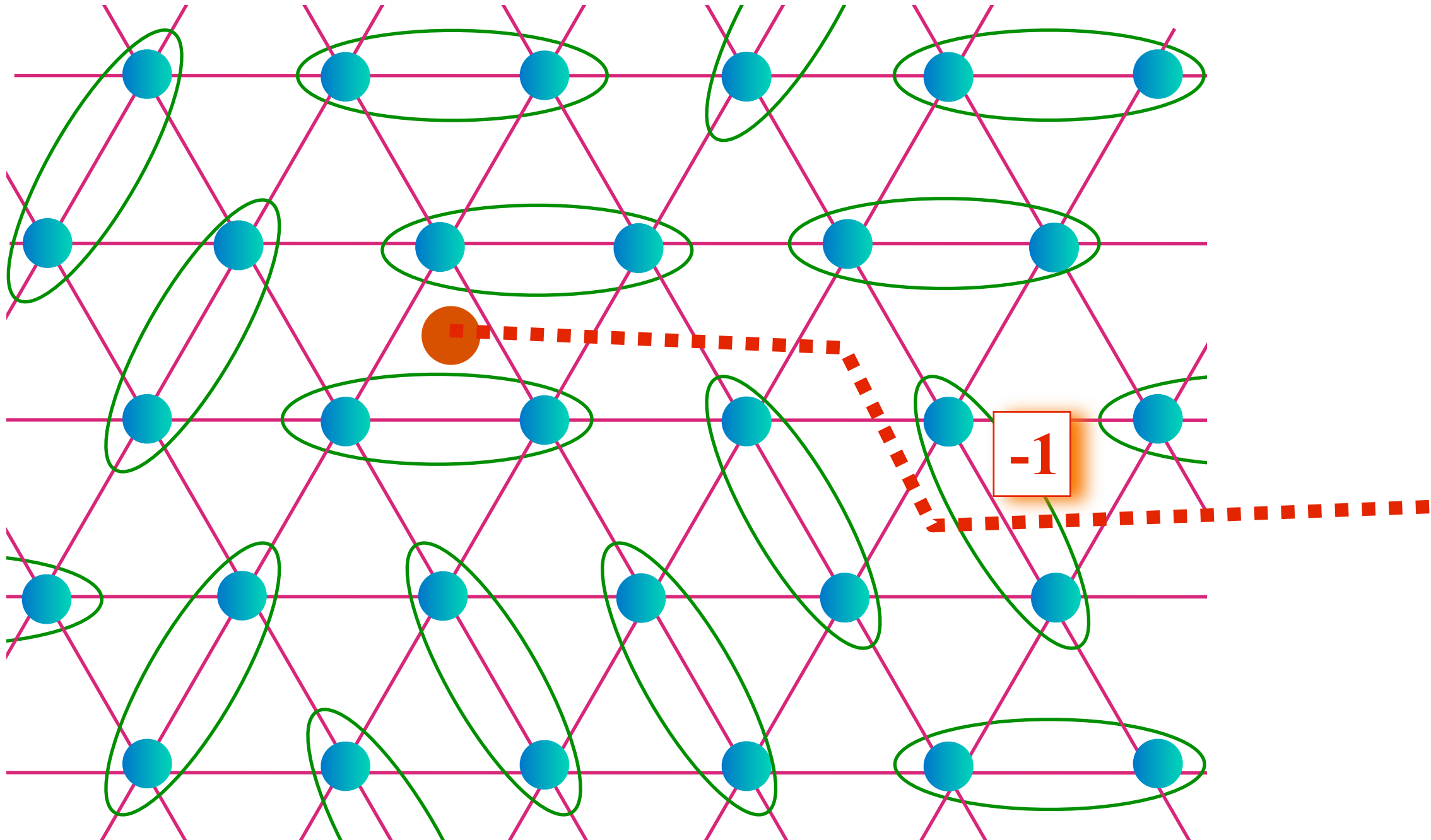
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A vison

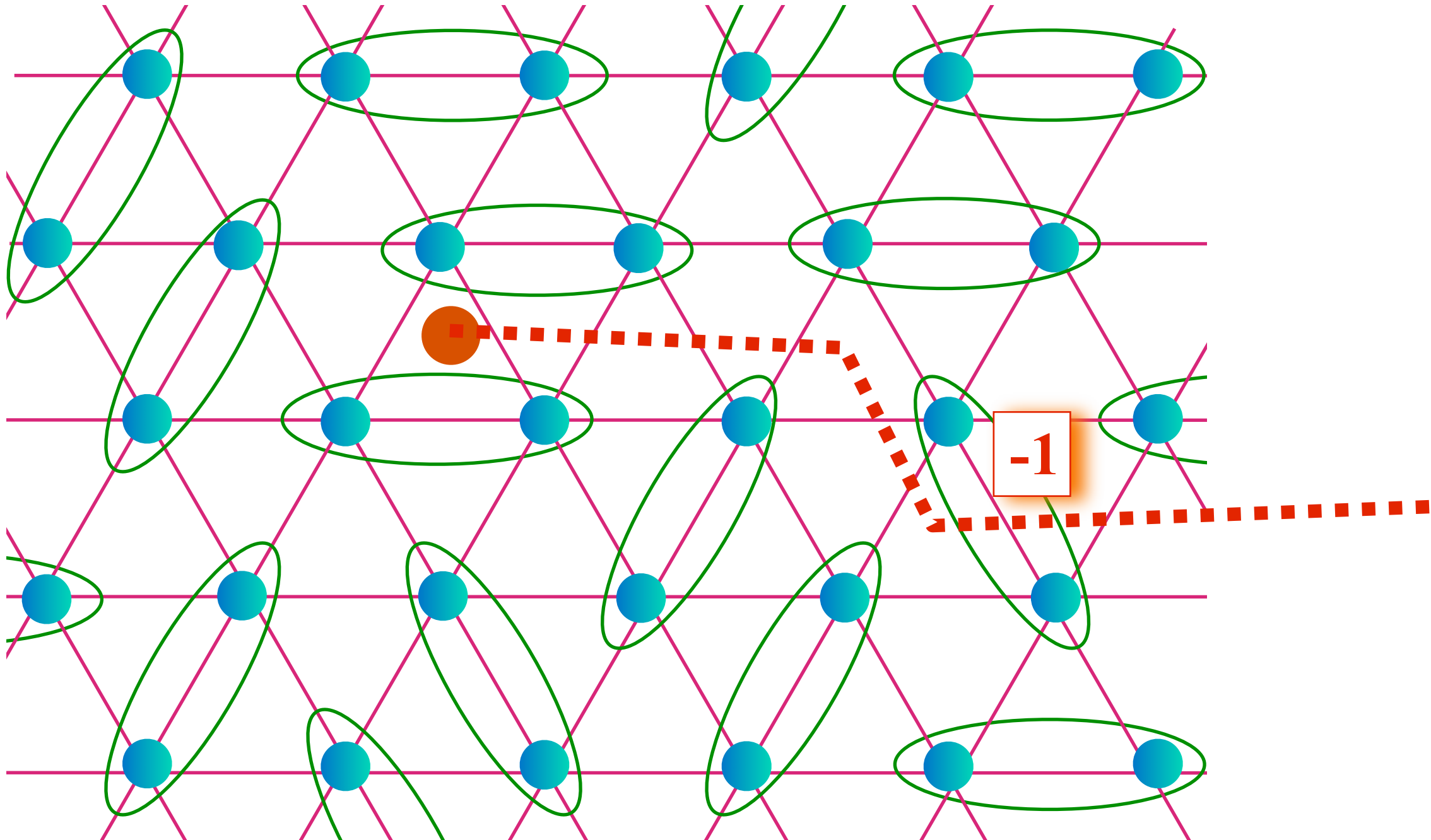
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Excitations of the Z_2 Spin liquid

A vison

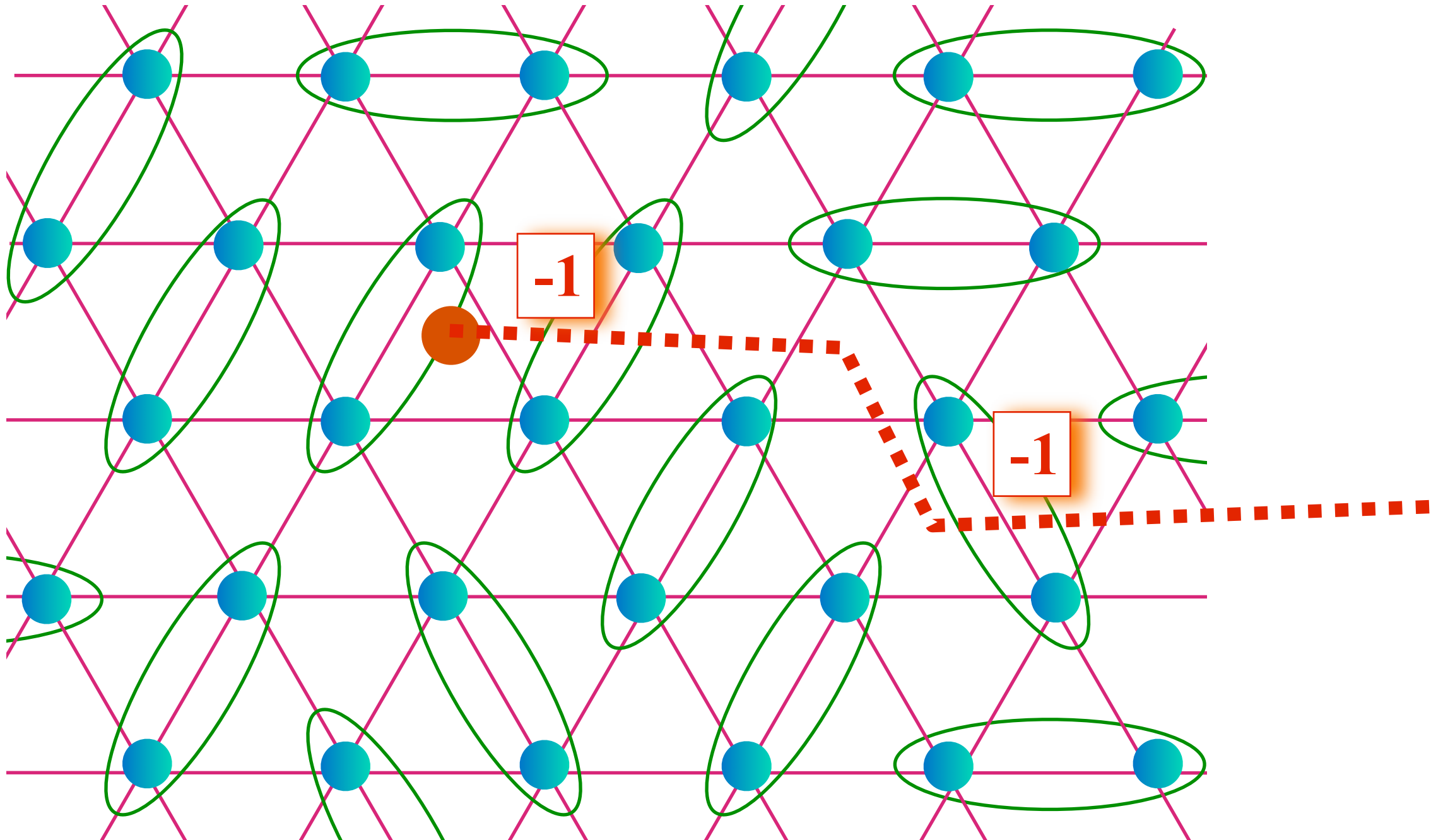
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Excitations of the Z_2 Spin liquid

A vison

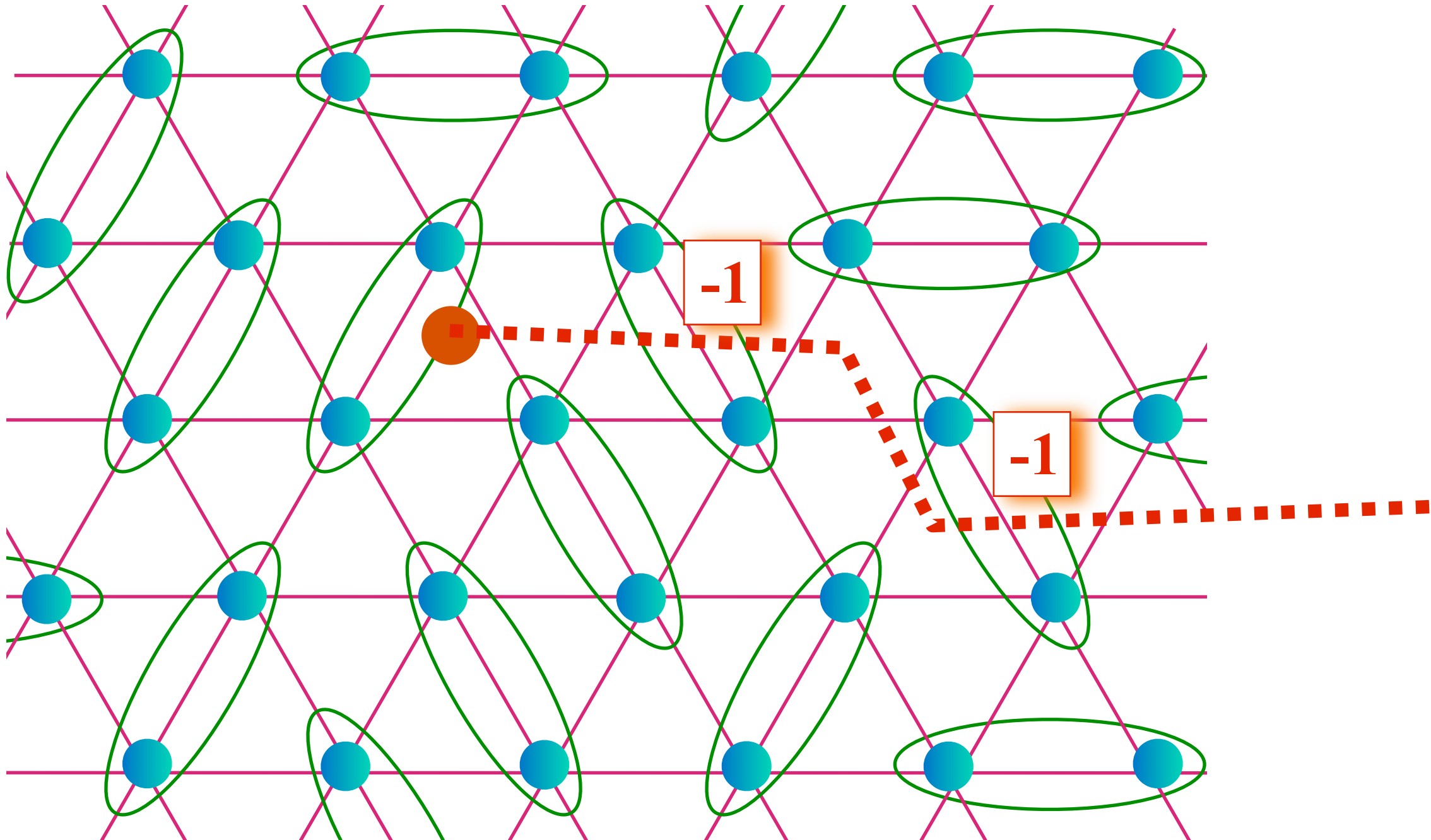
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Excitations of the Z_2 Spin liquid

A vison

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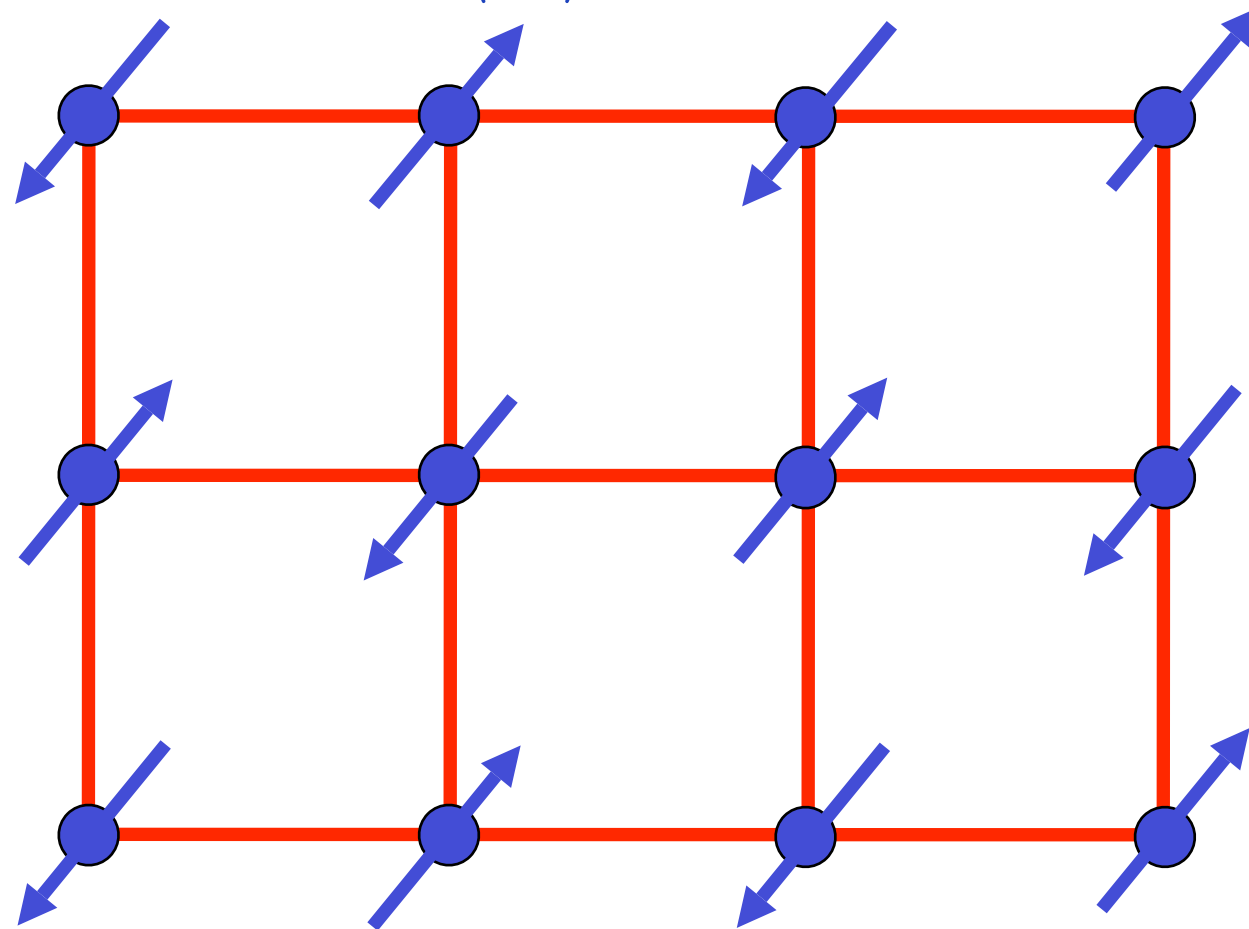
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Thermal conductivity of κ -(ET) $_2$ Cu $_2$ (CN) $_3$

Square lattice antiferromagnet

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



Ground state has long-range Néel order

Order parameter is a single vector field $\vec{\varphi} = \eta_i \vec{S}_i$

$\eta_i = \pm 1$ on two sublattices

$\langle \vec{\varphi} \rangle \neq 0$ in Néel state.

Theory for loss of Neel order

Write the spin operator in terms of
Schwinger bosons (spinons) $z_{i\alpha}$, $\alpha = \uparrow, \downarrow$:

$$\vec{S}_i = z_{i\alpha}^\dagger \vec{\sigma}_{\alpha\beta} z_{i\beta}$$

where $\vec{\sigma}$ are Pauli matrices, and the bosons obey the local constraint

$$\sum_{\alpha} z_{i\alpha}^\dagger z_{i\alpha} = 2S$$

Effective theory for spinons must be invariant under the U(1) gauge transformation

$$z_{i\alpha} \rightarrow e^{i\theta} z_{i\alpha}$$

Perturbation theory

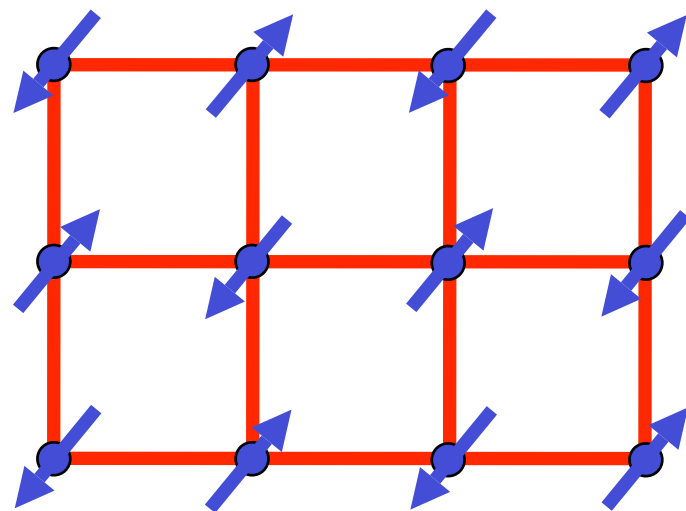
Low energy spinon theory for “quantum disordering” the Néel state is the CP^1 model

$$\mathcal{S}_z = \int d^2x d\tau \left[c^2 |(\nabla_x - iA_x)z_\alpha|^2 + |(\partial_\tau - iA_\tau)z_\alpha|^2 + s |z_\alpha|^2 + u (|z_\alpha|^2)^2 + \frac{1}{4e^2} (\epsilon_{\mu\nu\lambda} \partial_\nu A_\lambda)^2 \right]$$

where A_μ is an emergent U(1) gauge field (the “**photon**”) which describes low-lying spin-singlet excitations.

Phases:

$\langle z_\alpha \rangle \neq 0$	$\Rightarrow$	Néel (Higgs) state
$\langle z_\alpha \rangle = 0$	$\Rightarrow$	Spin liquid (Coulomb) state



$$\langle z_\alpha \rangle \neq 0$$

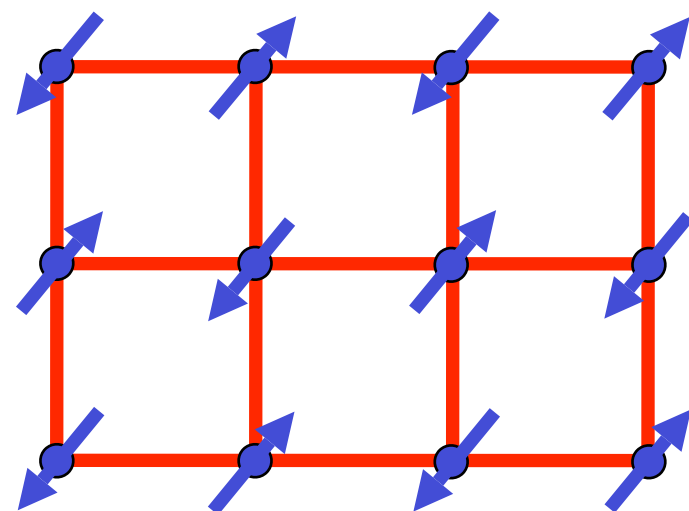
Néel state

s_c

Spin liquid with a
“**photon**” collective mode

$$\langle z_\alpha \rangle = 0$$

s



$$\langle z_\alpha \rangle \neq 0$$

Néel state

Spin liquid with a
“**photon**” collective mode

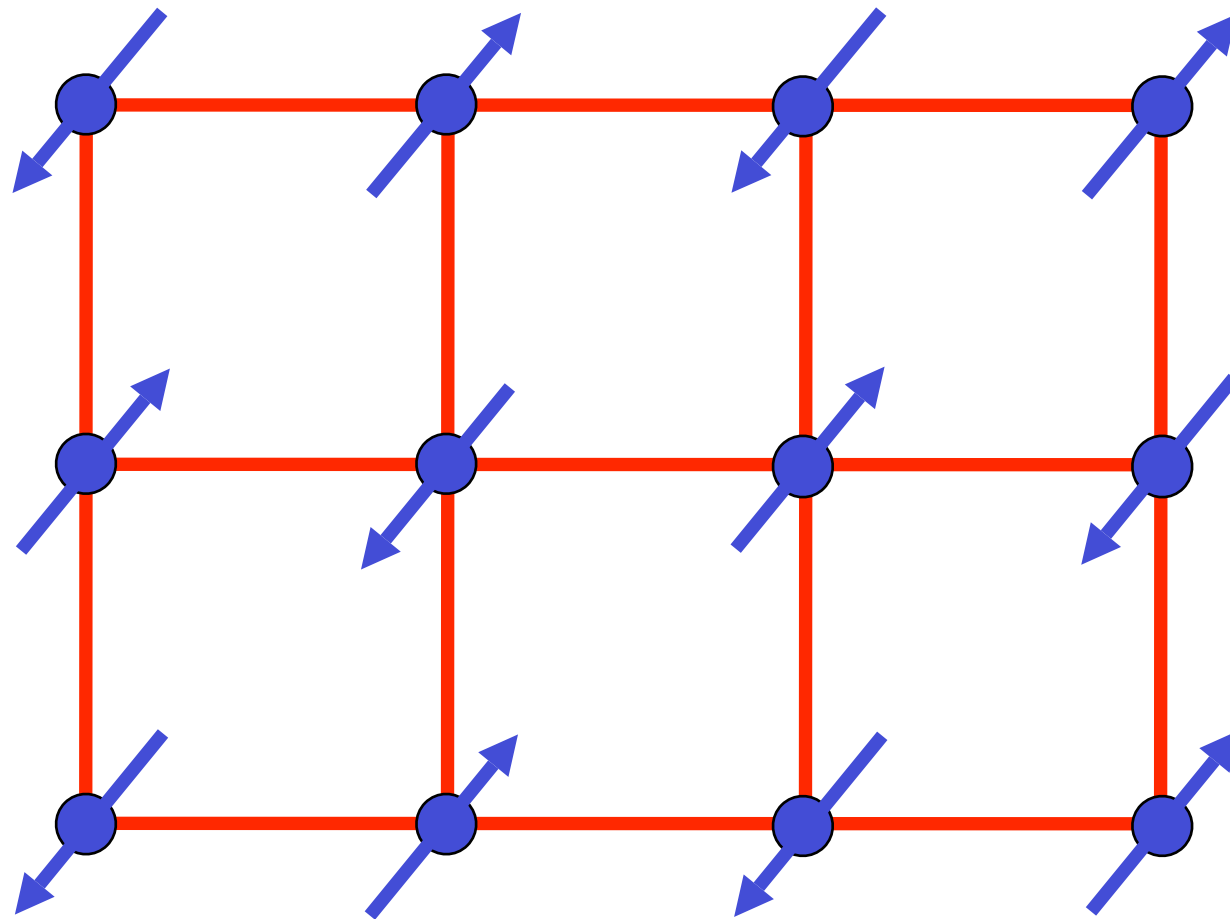
[Unstable to valence bond solid (VBS) order]

$$\langle z_\alpha \rangle = 0$$

s_c

s

From the square to the triangular lattice

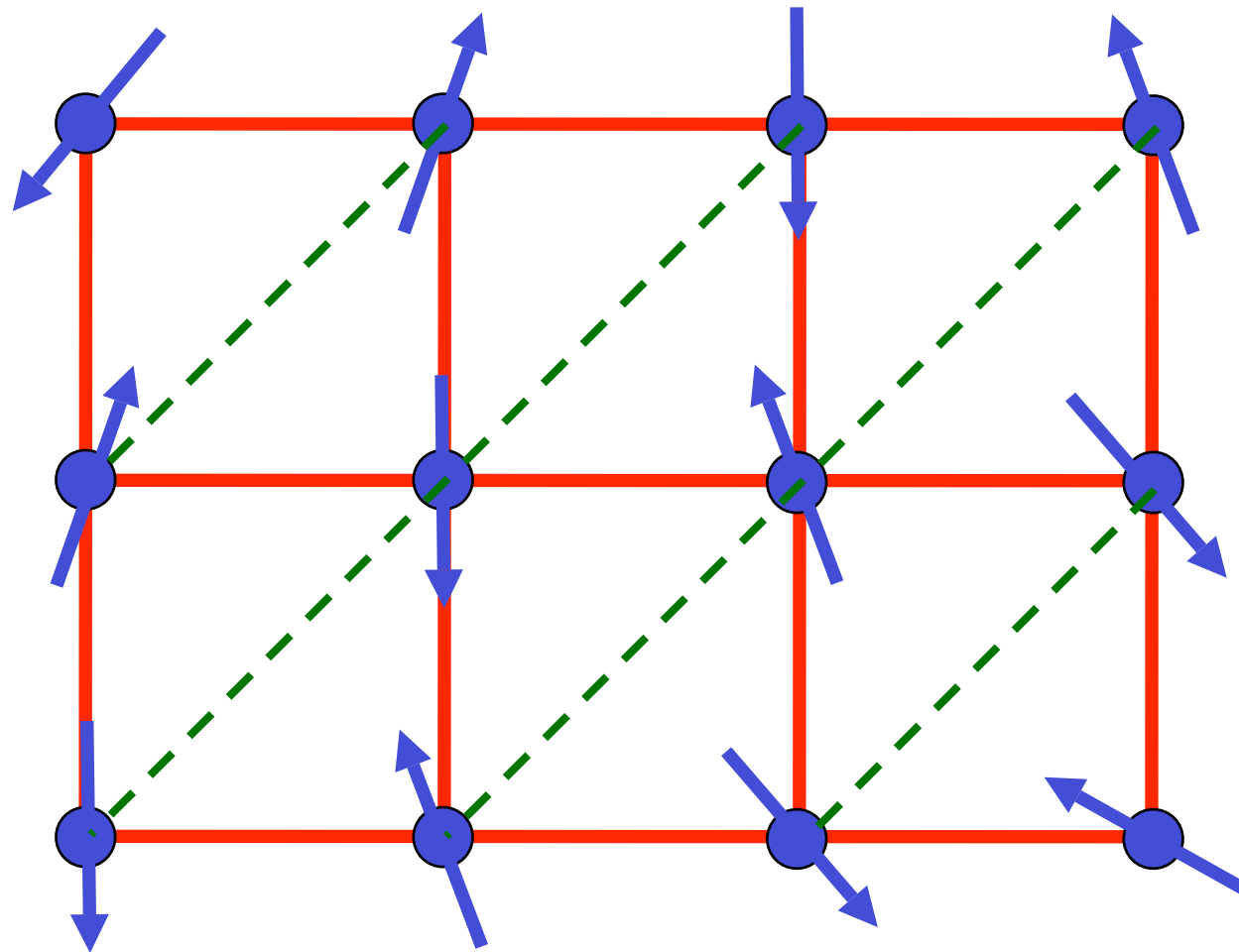


A spin density wave with

$$\langle \vec{S}_i \rangle \propto (\cos(\mathbf{K} \cdot \mathbf{r}_i), \sin(\mathbf{K} \cdot \mathbf{r}_i))$$

and $\mathbf{K} = (\pi, \pi)$.

From the square to the triangular lattice



A spin density wave with

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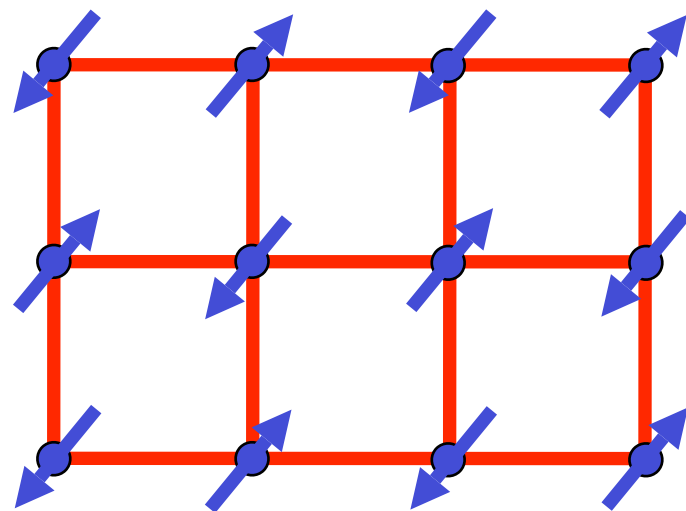
and $\mathbf{K} = (\pi + \Phi, \pi + \Phi)$.

Interpretation of non-collinearity Φ

Its physical interpretation becomes clear from the allowed coupling to the spinons:

$$\mathcal{S}_{z,\Phi} = \int d^2r d\tau [\lambda \Phi^* \epsilon_{\alpha\beta} z_\alpha \partial_x z_\beta + \text{c.c.}]$$

Φ is a spinon pair field



$$\langle z_\alpha \rangle \neq 0$$

Néel state

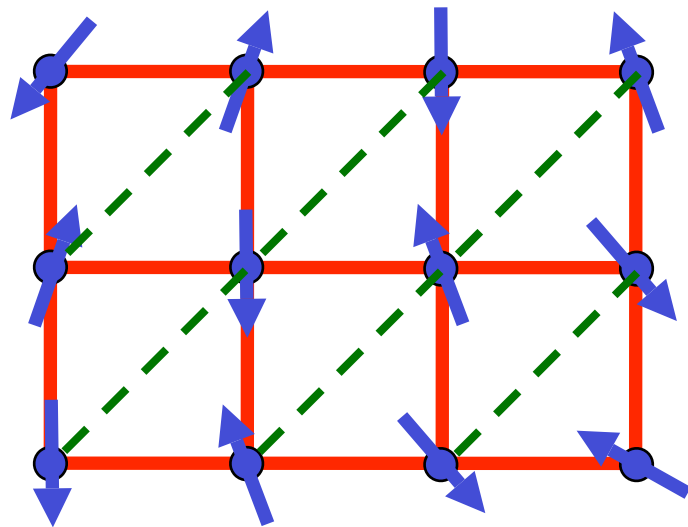
Spin liquid with a
“**photon**” collective mode

[Unstable to valence bond solid (VBS) order]

$$\langle z_\alpha \rangle = 0$$

s_c

s



$\langle z_\alpha \rangle \neq 0$, $\langle \Phi \rangle \neq 0$
 non-collinear Néel state

Z_2 spin liquid with a
vison excitation

$\langle z_\alpha \rangle = 0$, $\langle \Phi \rangle \neq 0$

s_c

s

What is a vison ?

A vison is an Abrikosov vortex in the spinon pair field Φ .

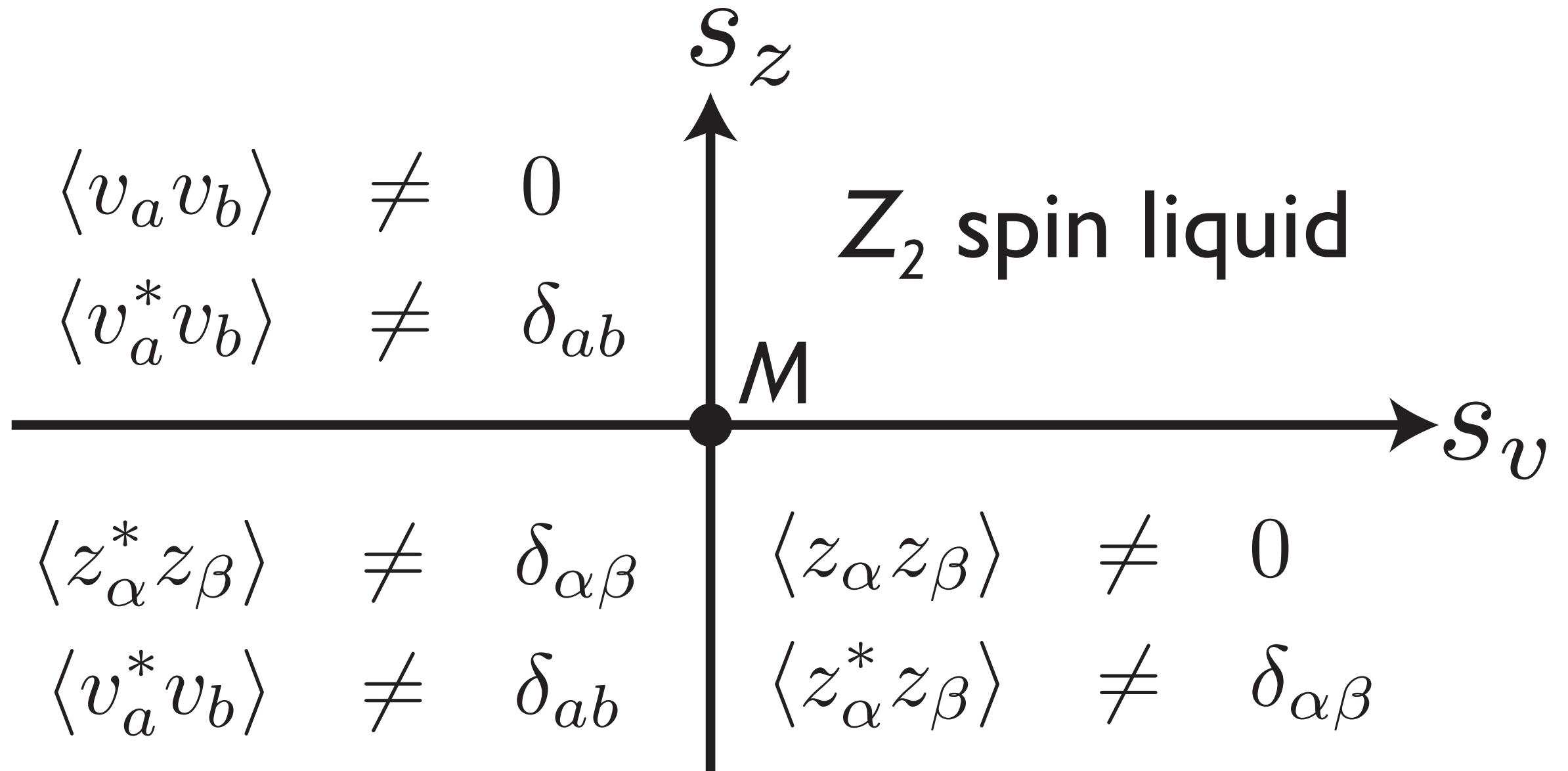
In the Z_2 spin liquid, the vison is $S = 0$ quasiparticle with a finite energy gap

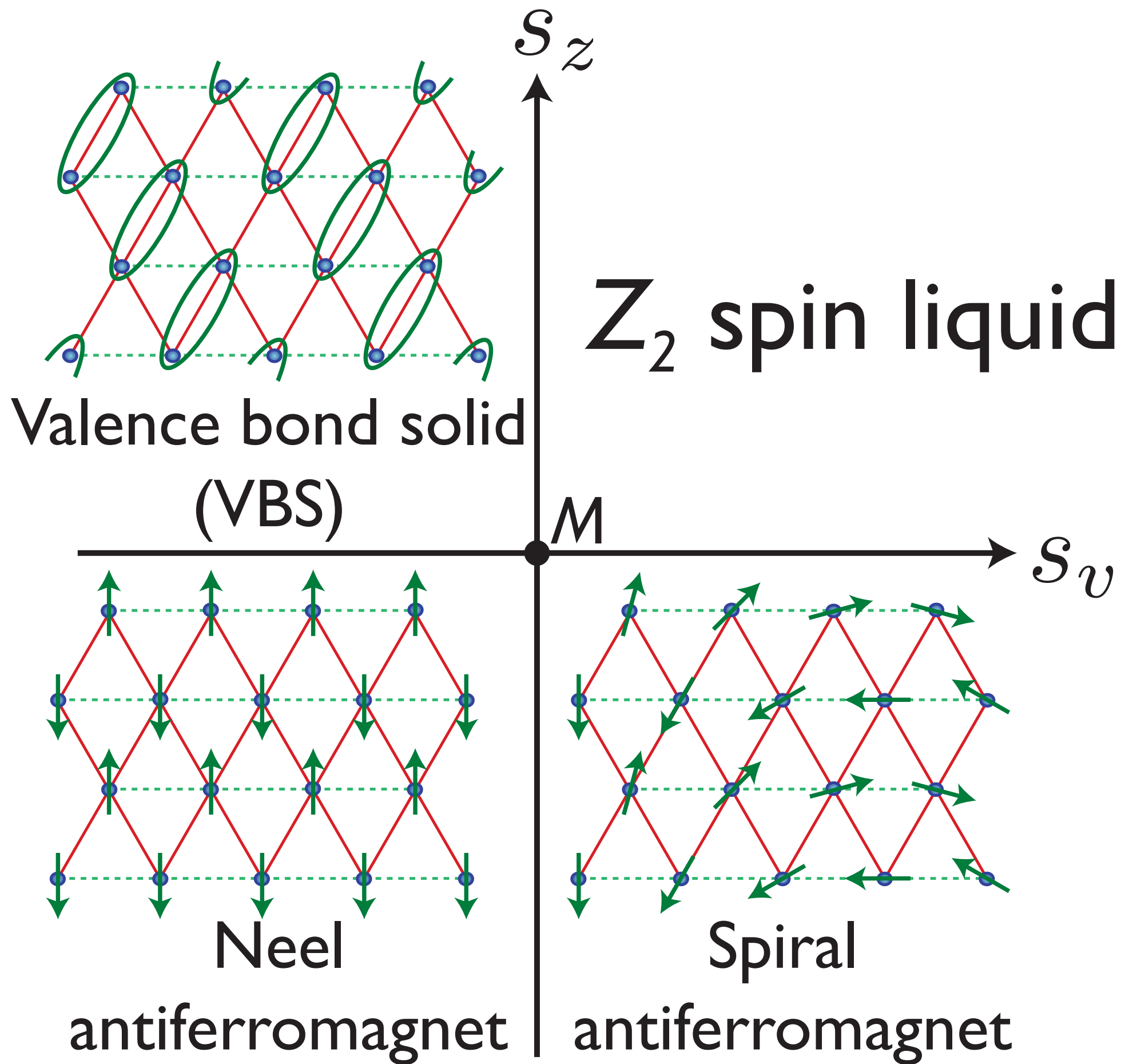
Mutual Chern-Simons Theory

Express theory in terms of the physical excitations of the Z_2 spin liquid: the spinons, z_α , and the visons. After accounting for Berry phase effects, the visons can be described by complex fields v_a , which transforms non-trivially under the square lattice space group operations.

The spinons and visons have mutual semionic statistics, and this leads to the mutual CS theory at $k = 2$:

$$\begin{aligned}\mathcal{L} &= \sum_{\alpha=1}^2 \left\{ |(\partial_\mu - ia_\mu)z_\alpha|^2 + s_z |z_\alpha|^2 \right\} \\ &+ \sum_{a=1}^{N_v} \left\{ |(\partial_\mu - ib_\mu)v_a|^2 + s_v |v_a|^2 \right\} \\ &+ \frac{ik}{2\pi} \epsilon_{\mu\nu\lambda} a_\mu \partial_\nu b_\lambda + \dots\end{aligned}$$





Outline

1. Review of experiments

Phases of the $S=1/2$ antiferromagnet on the anisotropic triangular lattice

2. The Z_2 spin liquid and its excitations

Spinons and visons

3. Field theory for spinons and visons

Berry phases and the doubled Chern-Simons theory

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How do we reconcile power-law T dependence
in $1/T_1$ with activated thermal conductivity ?

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I. Thermal conductivity is dominated by vison transport

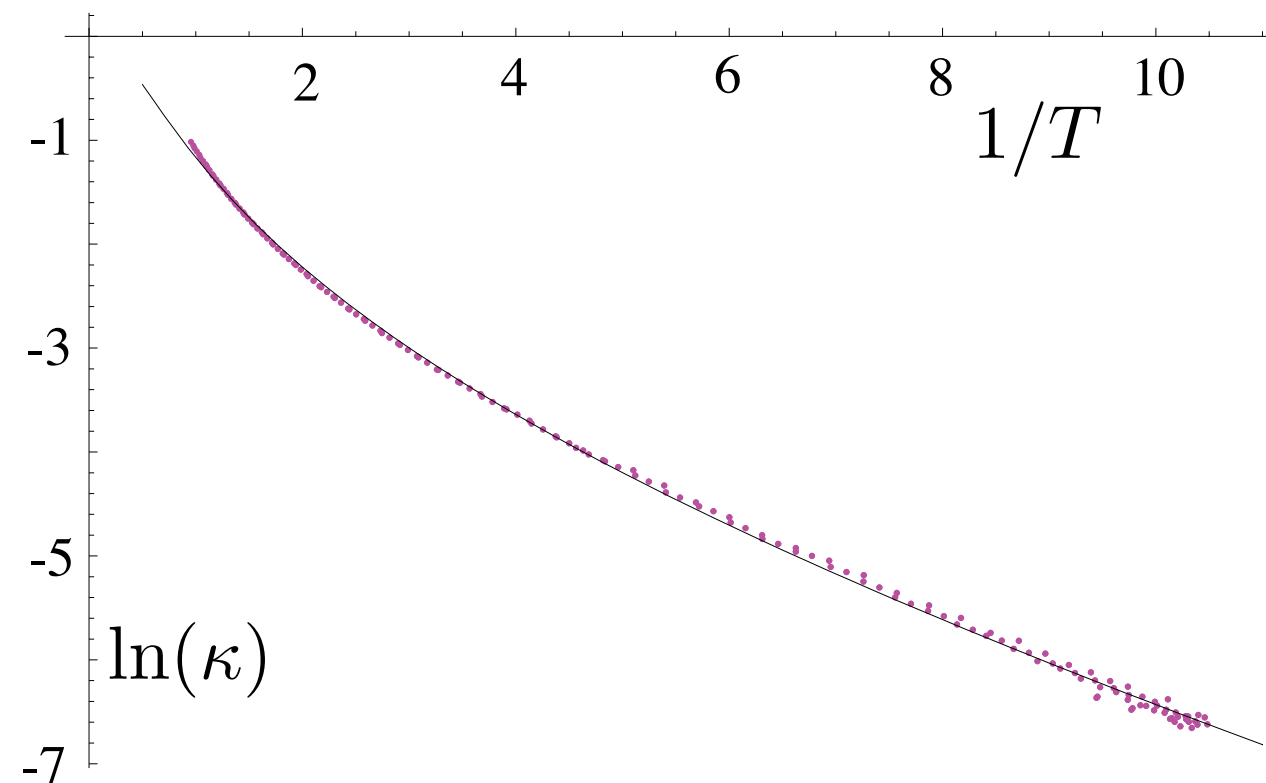
The thermal conductivity (per layer) of N_v species of slowly moving visons of mass m_v , above an energy gap Δ_v , scattering off impurities of density n_{imp} is

$$\kappa_v = \frac{N_v m_v k_B^3 T^2 \ln^2(T_v/T) e^{-\Delta_v/(k_B T)}}{4\pi \hbar^3 n_{\text{imp}}}.$$

where T_v is of order the vison bandwidth.

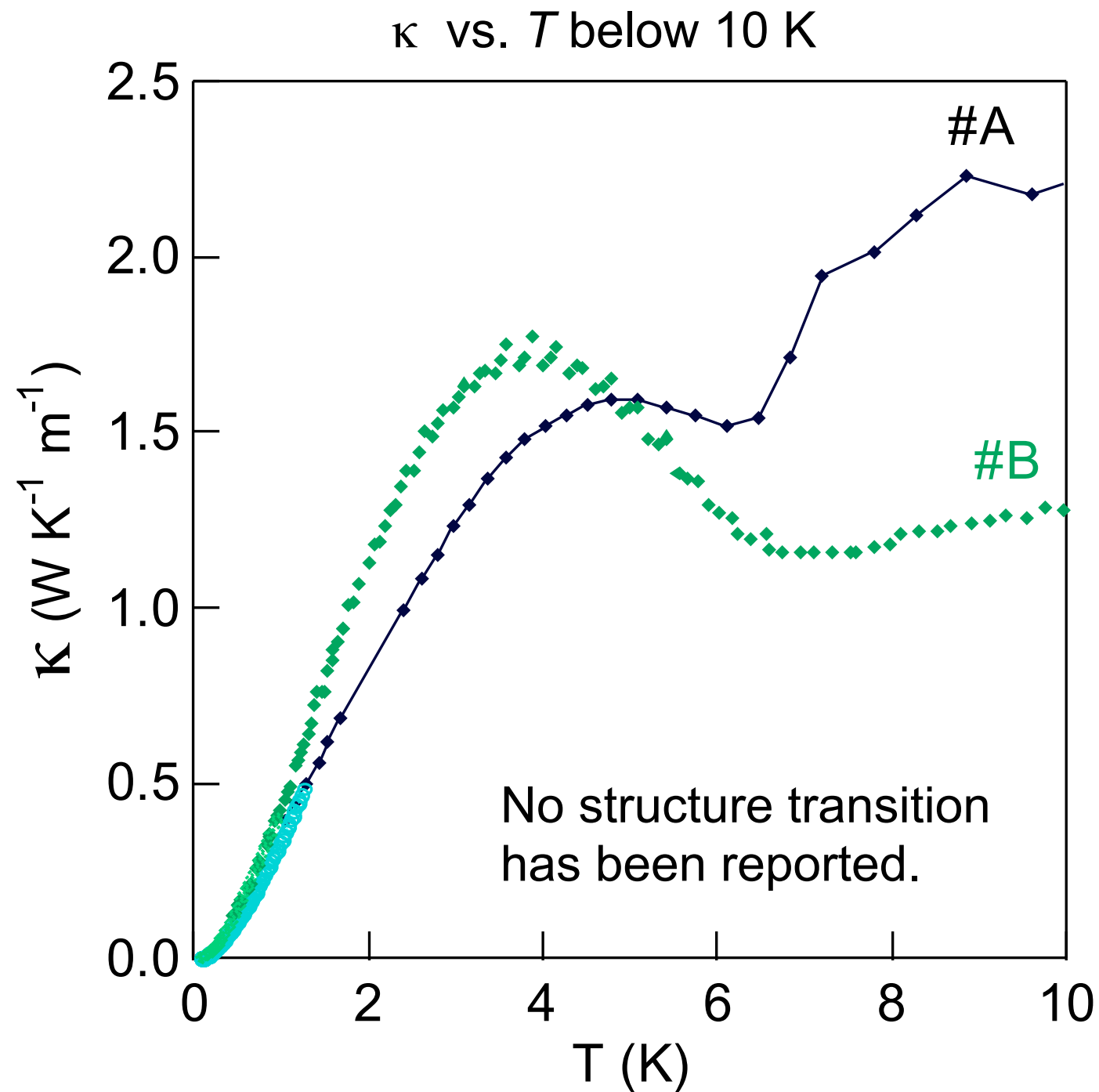
How do we reconcile power-law T dependence in $1/T_1$ with activated thermal conductivity ?

I. Thermal conductivity is dominated by vison transport



Best fit to data yields, $\Delta_v \approx 0.24$ K and $T_v \approx 8$ K.

Thermal Conductivity below 10K

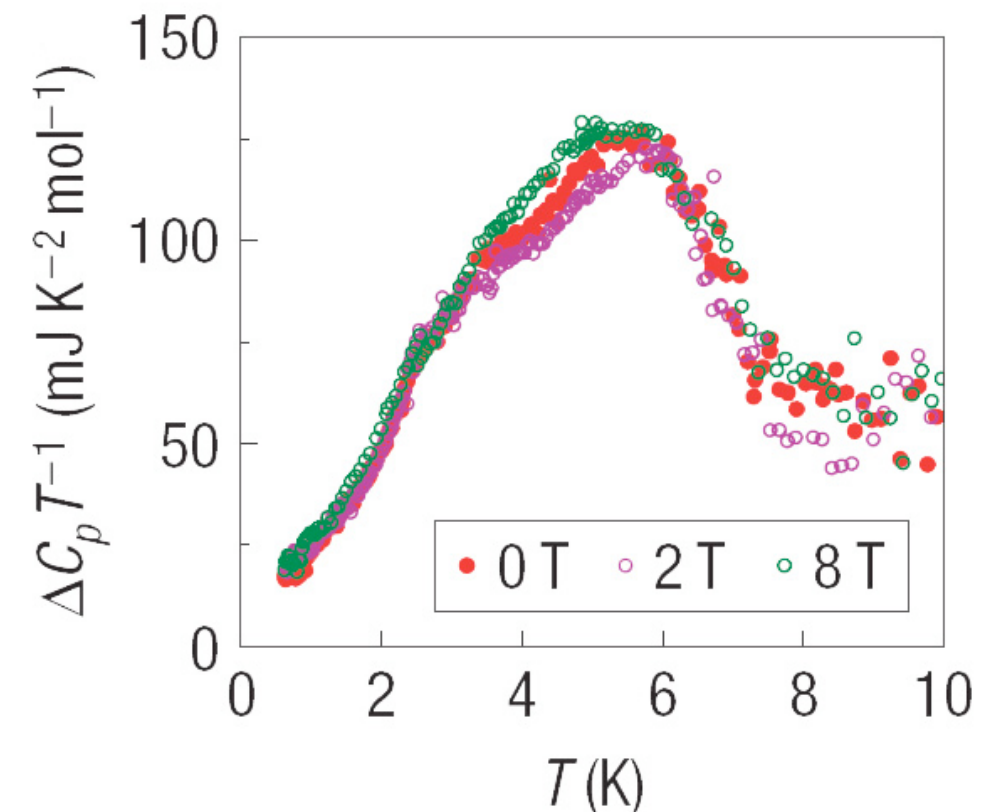


Similar to

- $1/T_1$ by ^1H NMR
- Heat capacity



Difference of heat capacity between
 $X = \text{Cu}_2(\text{CN})_3$ and $\text{Cu}(\text{NCS})_2$
(superconductor).



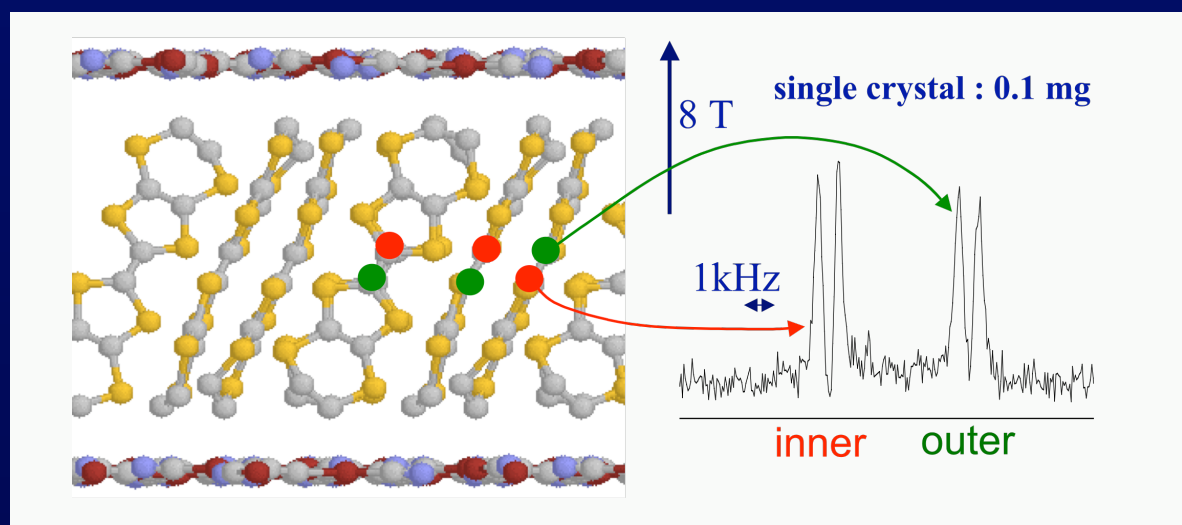
S. Yamashita, et al., Nature Physics **4**, 459 - 462 (2008)

- Magnetic contribution to κ
- Phase transition or crossover?
 - Chiral order transition?
 - Instability of spinon Fermi surface?

Spin excitation in $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

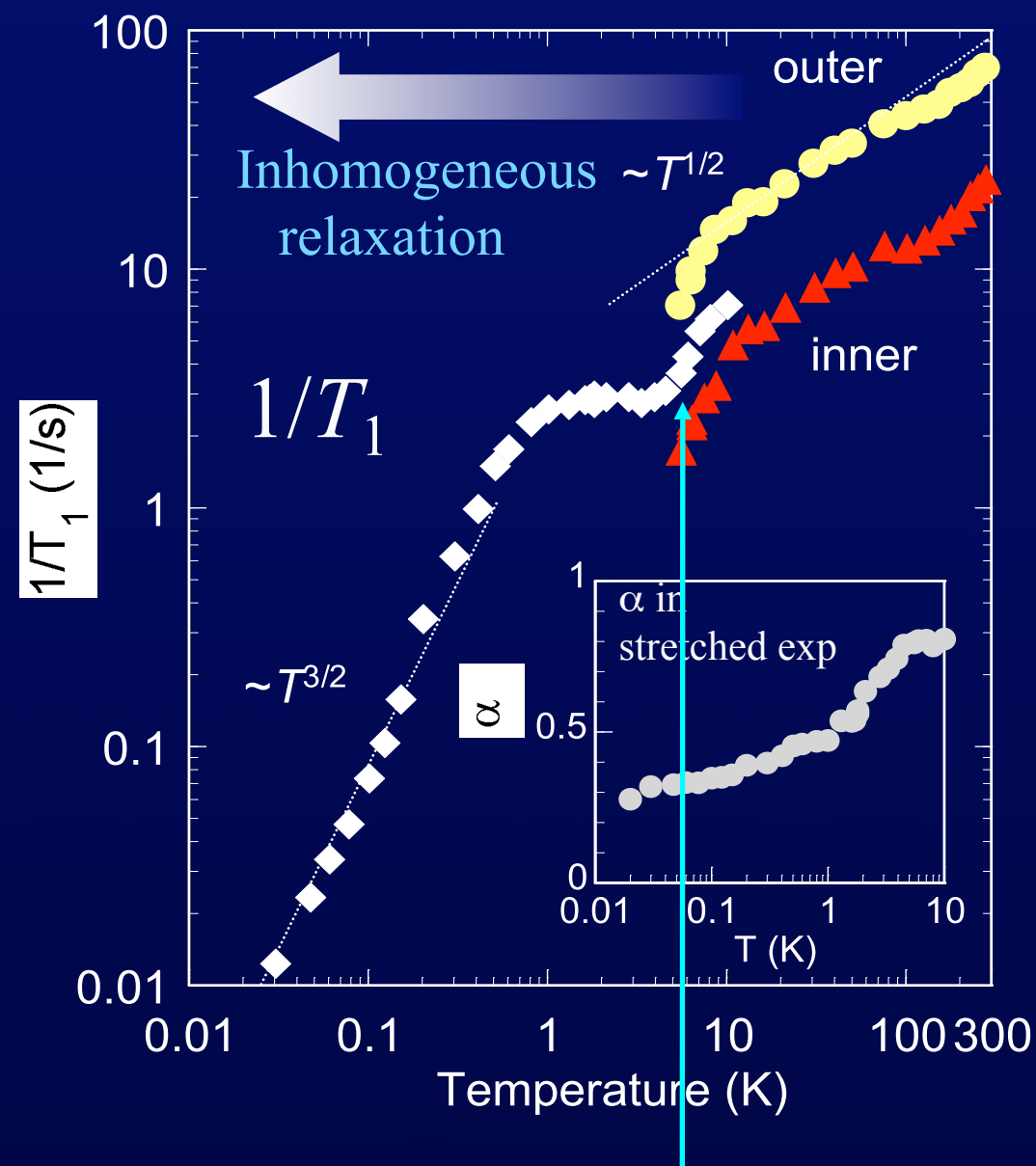
^{13}C NMR relaxation rate

Shimizu *et al.*, PRB 70 (2006) 060510



$1/T_1 \sim$ power law of T

Low-lying spin excitation at low-T



Anomaly at 5-6 K

How do we reconcile power-law T dependence in $1/T_1$ with activated thermal conductivity ?

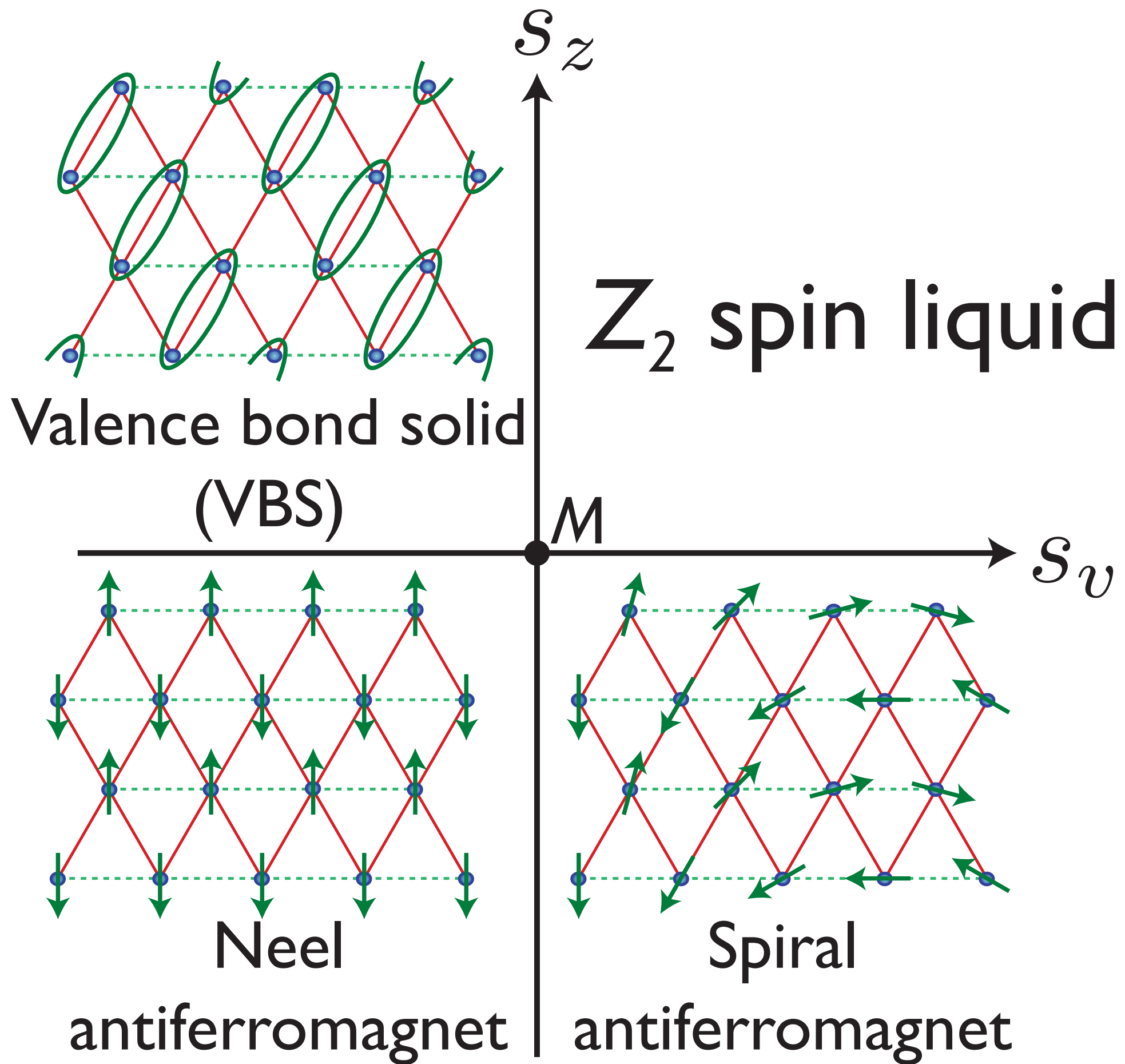
II. NMR relaxation is caused by spinons close to the quantum critical point between the non-collinear Néel state and the Z_2 spin liquid

The quantum-critical region of magnetic ordering on the triangular lattice is described by the $O(4)$ model and has

$$\frac{1}{T_1} \sim T^{\bar{\eta}}$$

where $\bar{\eta} = 1.374(12)$. This compares well with the observed $1/T_1 \approx T^{3/2}$ behavior.

(Note that the singlet gap is associated with a spinon-pair excitations, which is distinct from the vison gap, Δ_v .)



How do we reconcile power-law T dependence in $1/T_1$ with activated thermal conductivity ?

II. NMR relaxation is caused by spinons close to the quantum critical point between the non-collinear Néel state and the Z_2 spin liquid

At higher $T > \Delta_v$, the NMR will be controlled by the spinon-vision multicritical point, described by the mutual Chern-Simons theory. This multicritical point has

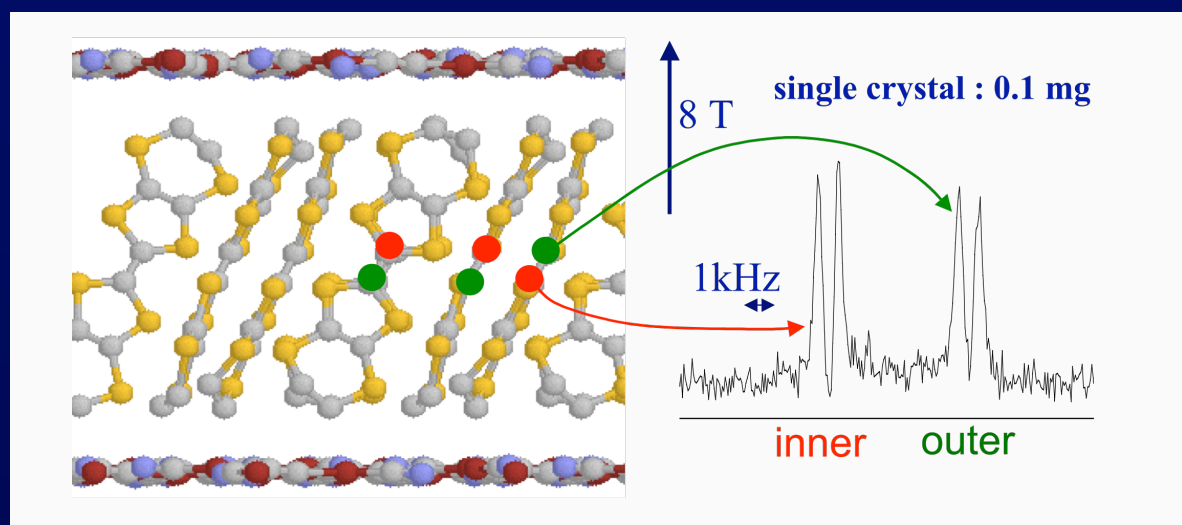
$$\frac{1}{T_1} \sim T^{\eta_{\text{cs}}}$$

We do not know the value η_{cs} accurately, but the $1/N$ expansion (and physical arguments) show that $\eta_{\text{cs}} < \bar{\eta}$.

Spin excitation in $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

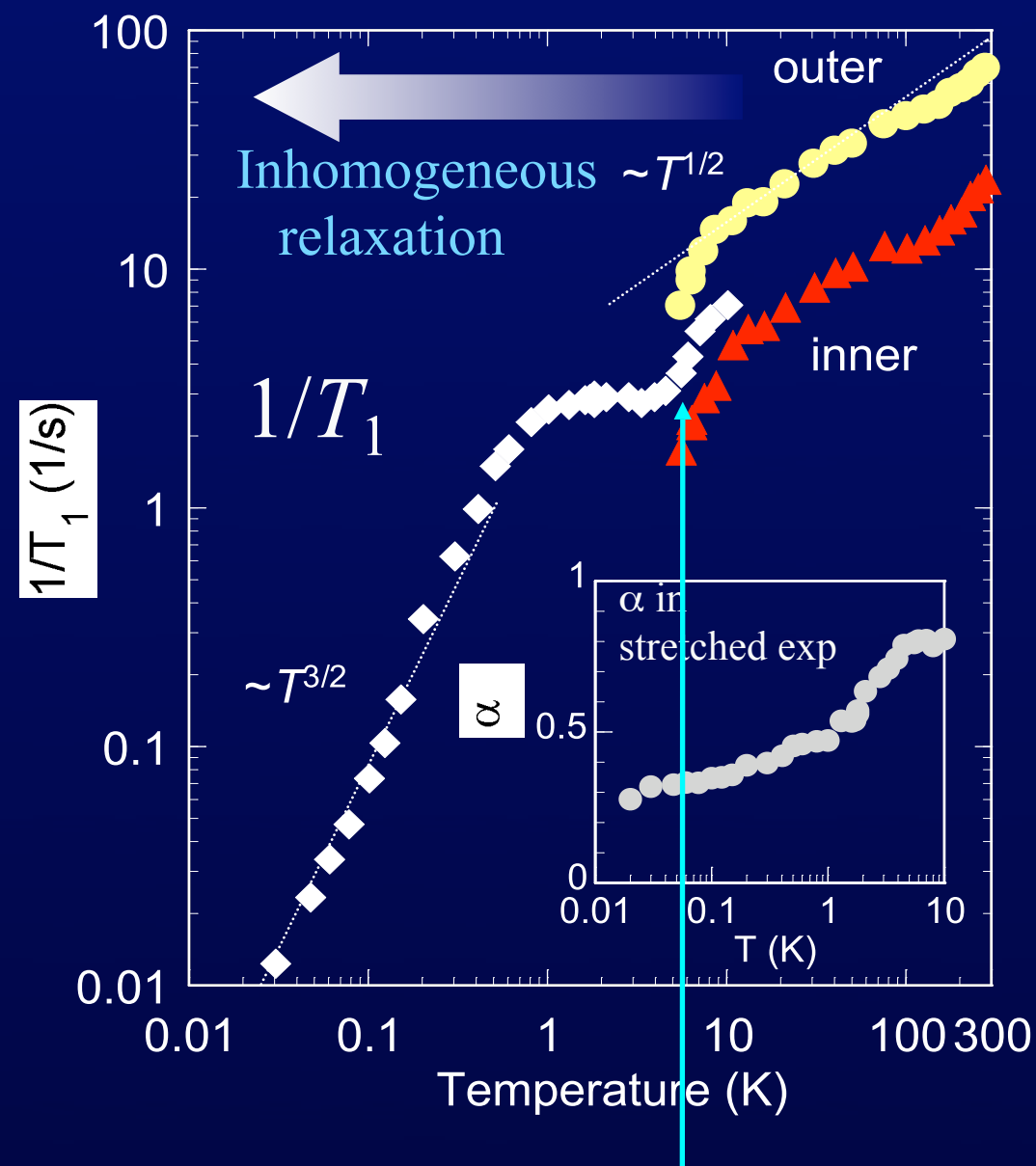
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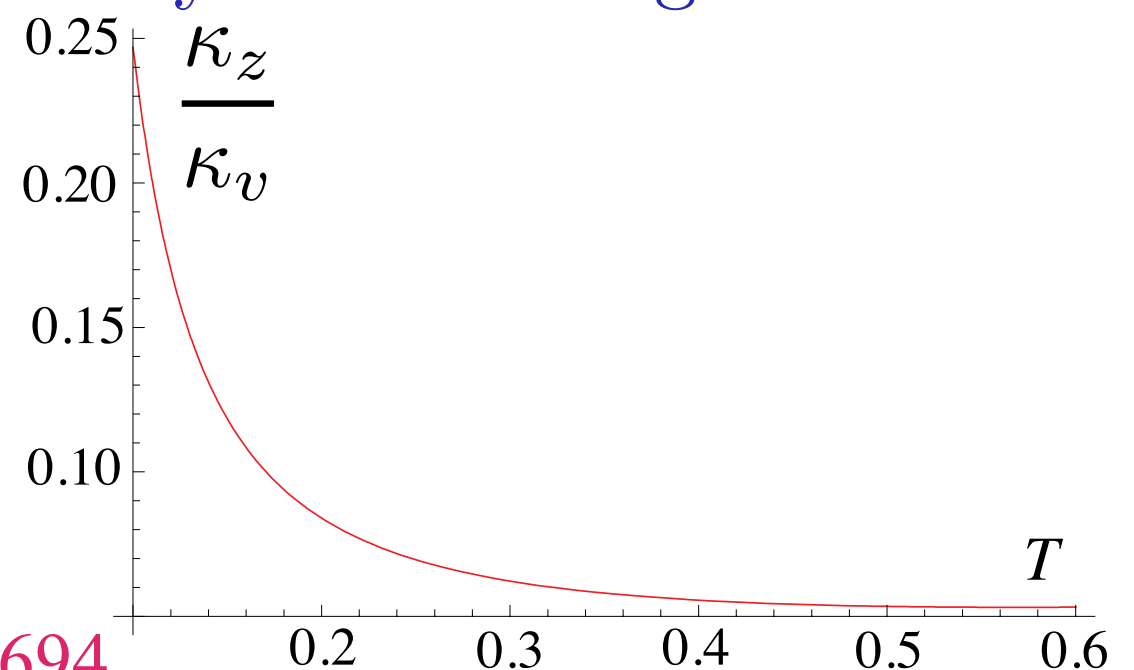


Anomaly at 5-6 K

How do we reconcile power-law T dependence in $1/T_1$ with activated thermal conductivity ?

III. The thermal conductivity of the visons is larger than the thermal conductivity of spinons

We compute the spinon thermal conductivity (κ_z) by the methods of quantum-critical hydrodynamics (developed recently using the AdS/CFT correspondence). Because the spinon bandwidth ($T_z \sim J \sim 250$ K) is much larger than the vison mass/bandwidth ($T_v \sim 8$ K), the vison thermal conductivity is much larger over the T range of the experiments.



Conclusions

- Global phase diagram obtained by condensing vision and spin excitations of the simplest Z_2 spin liquid
- Possible explanation for NMR and thermal conductivity measurements on κ -(ET)₂Cu₂(CN)₃