



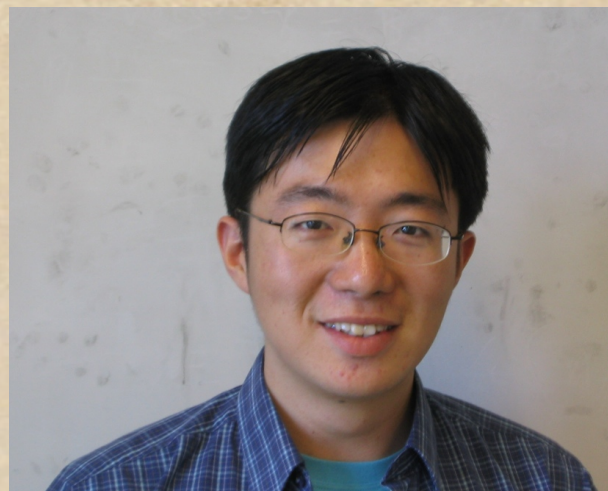
Detecting collective excitations of quantum spin liquids

Talk online: sachdev.physics.harvard.edu





Yang Qi
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arXiv:0809.0694



Ribhu Kaul
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Roger Melko
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Max Metlitski
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arXiv:0808.0495

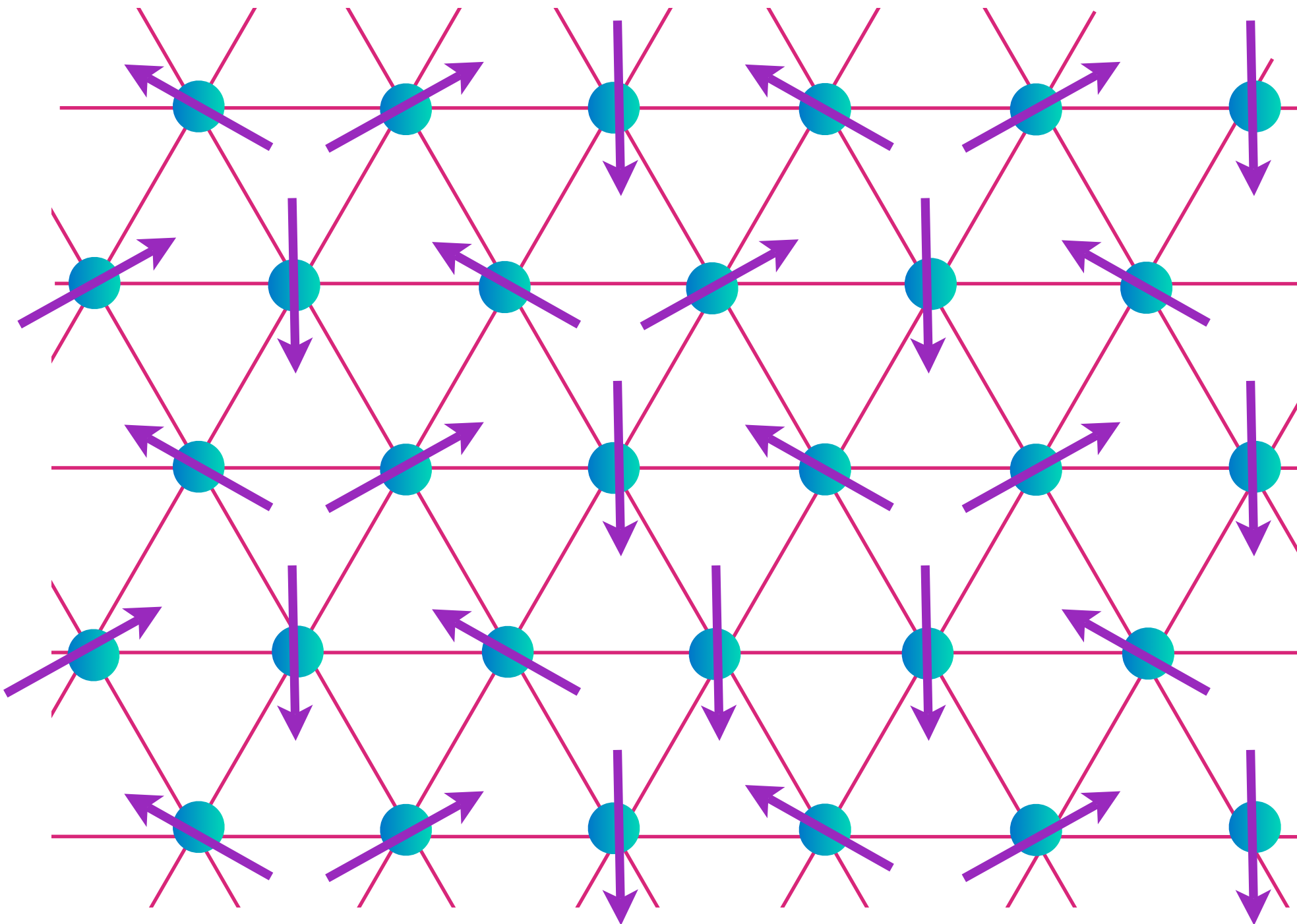
Collective excitations of quantum matter

-  Fermi liquid - *zero sound and paramagnons*
-  Superfluid - *phonons and vortices*
-  Quantum hall liquids - *magnetoplasmons*
-  Antiferromagnets - *spin waves*

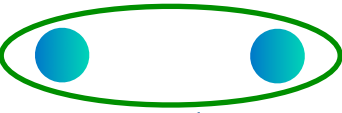
Collective excitations of quantum matter

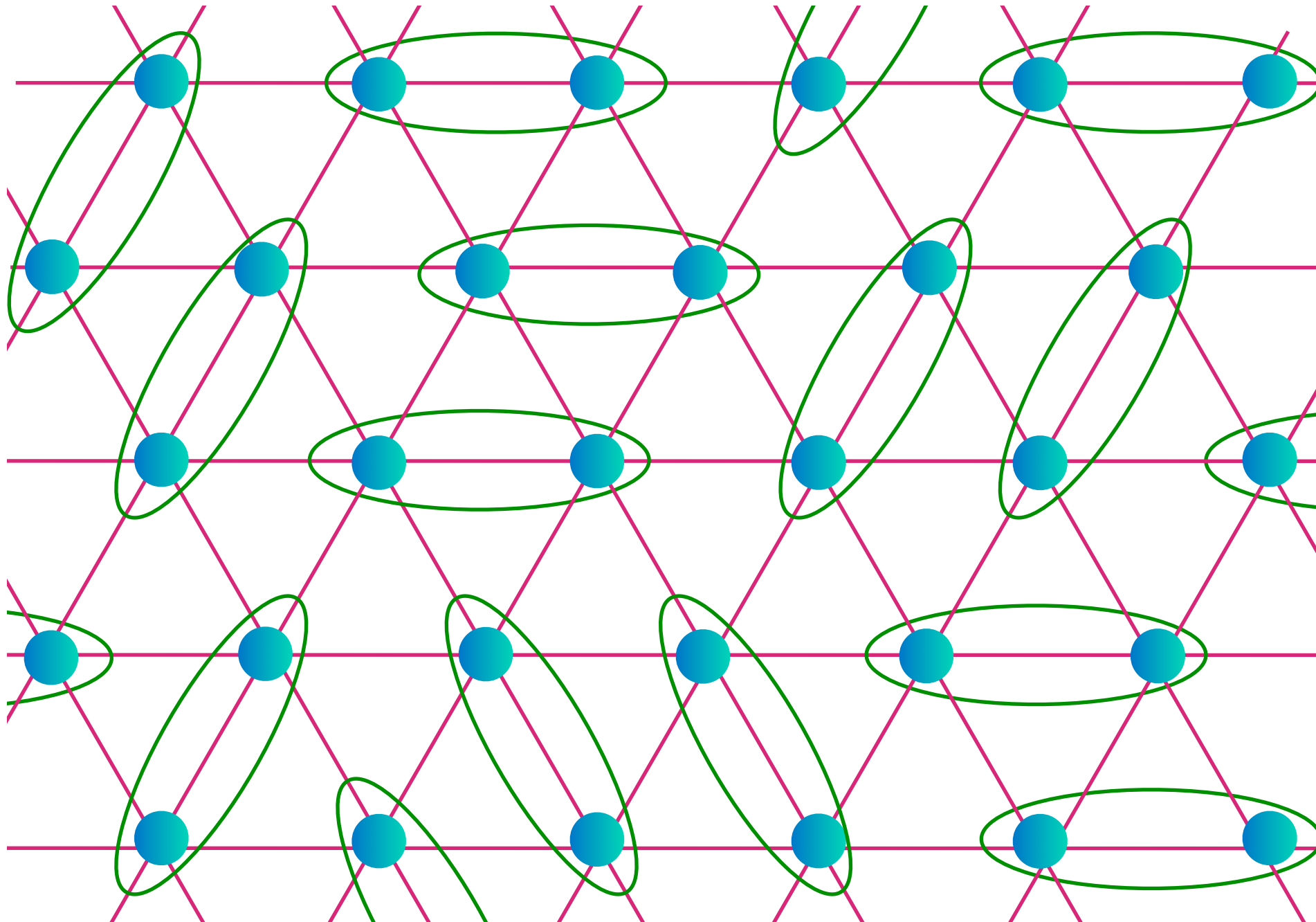
-  **Fermi liquid** - *zero sound and paramagnons*
-  **Superfluid** - *phonons and vortices*
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-  **Antiferromagnets** - *spin waves*
-  **Spin liquids** - *visons and “photons”*

Antiferromagnet

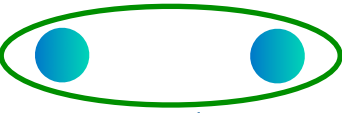


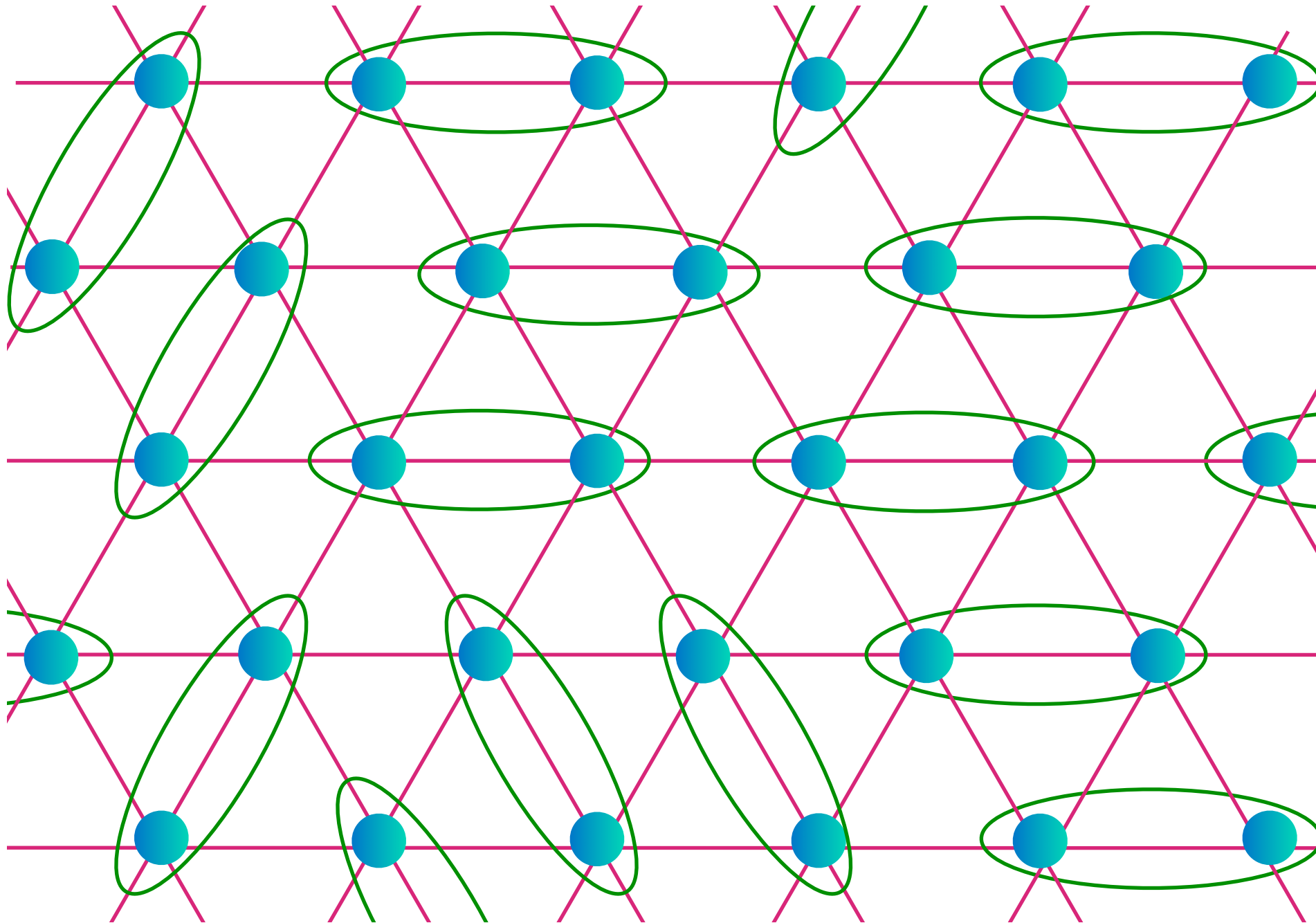
Spin liquid


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



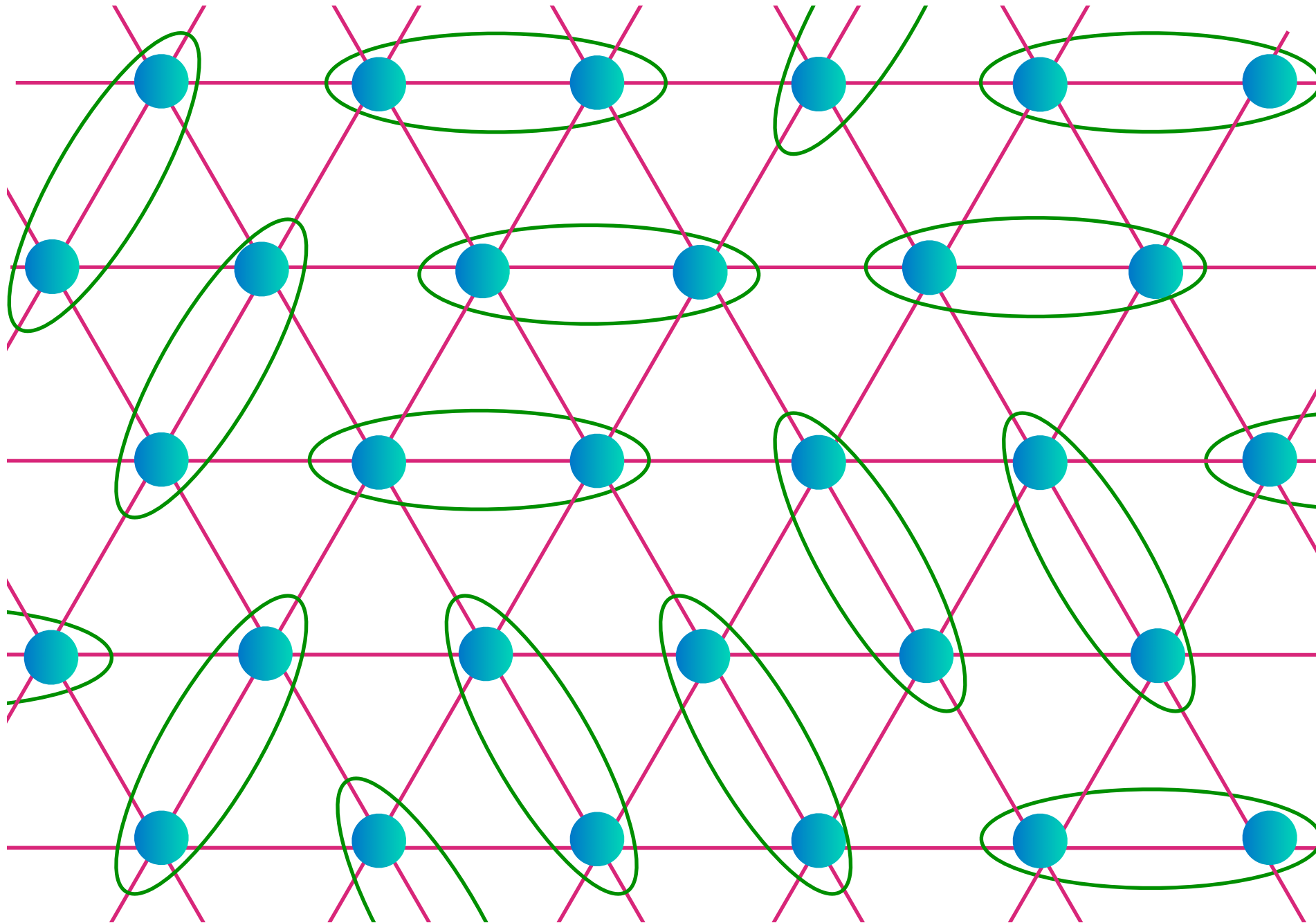
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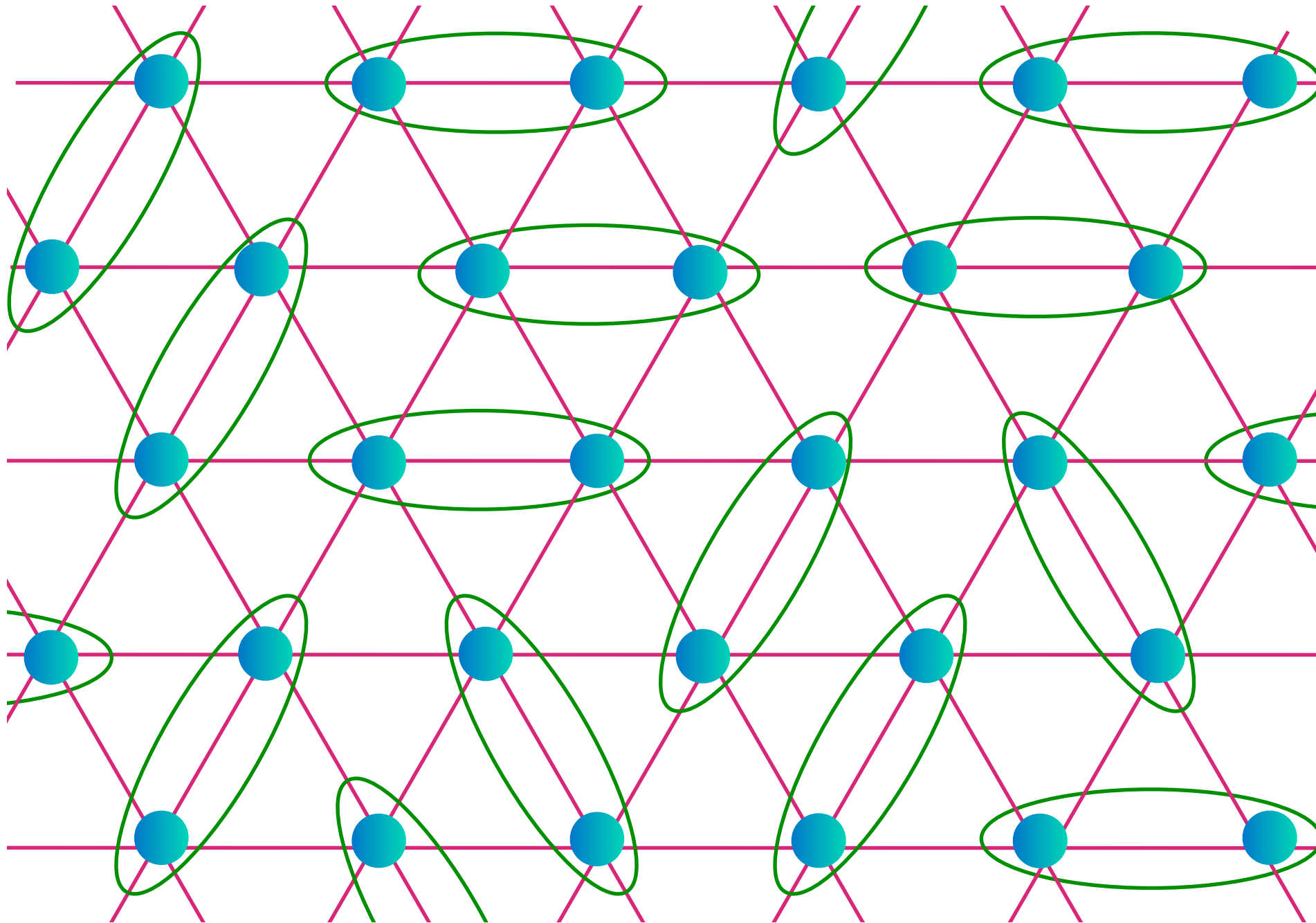
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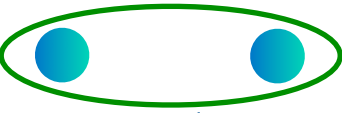


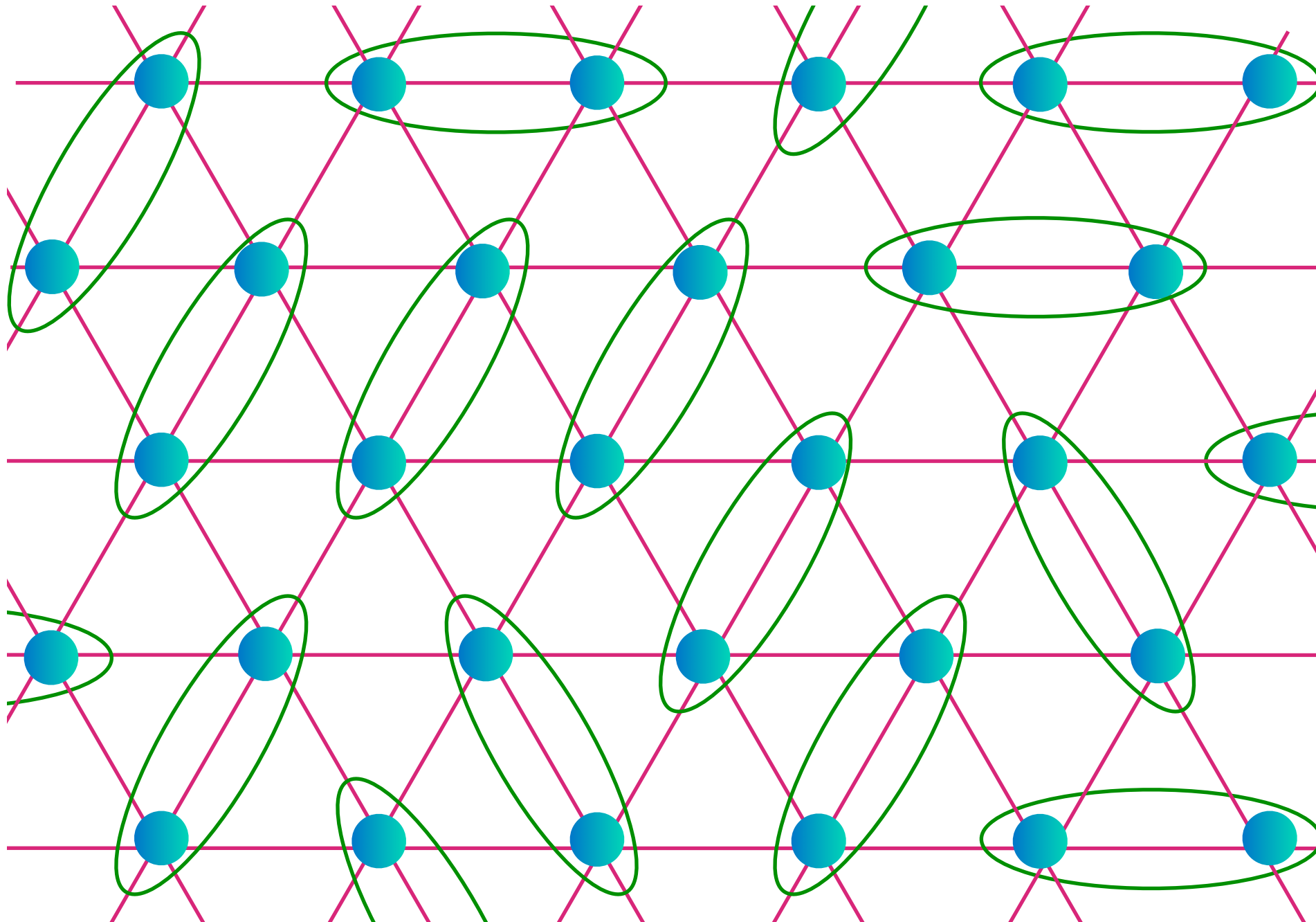
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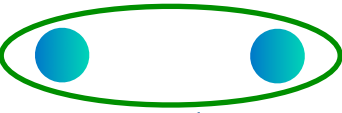


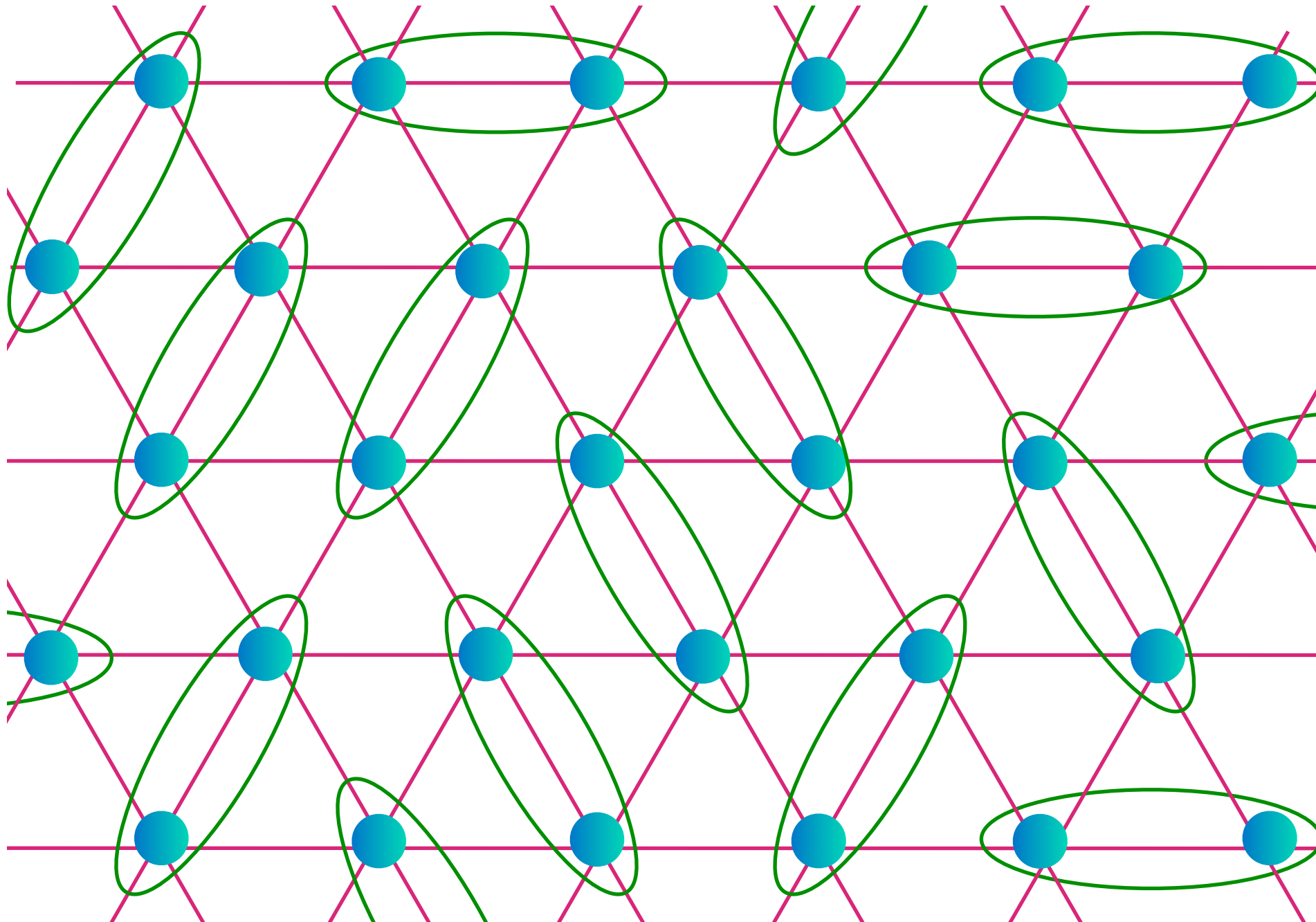
Spin liquid


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



Spin liquid


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General approach

Look for spin liquids across
continuous (or weakly first-order)
quantum transitions from
antiferromagnetically ordered states

Outline

1. Collective excitations of spin liquids in two dimensions

Photons and visons

2. Detecting the vison

Thermal conductivity of $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

3. Detecting the photon

Valence bond solid order around Zn impurities

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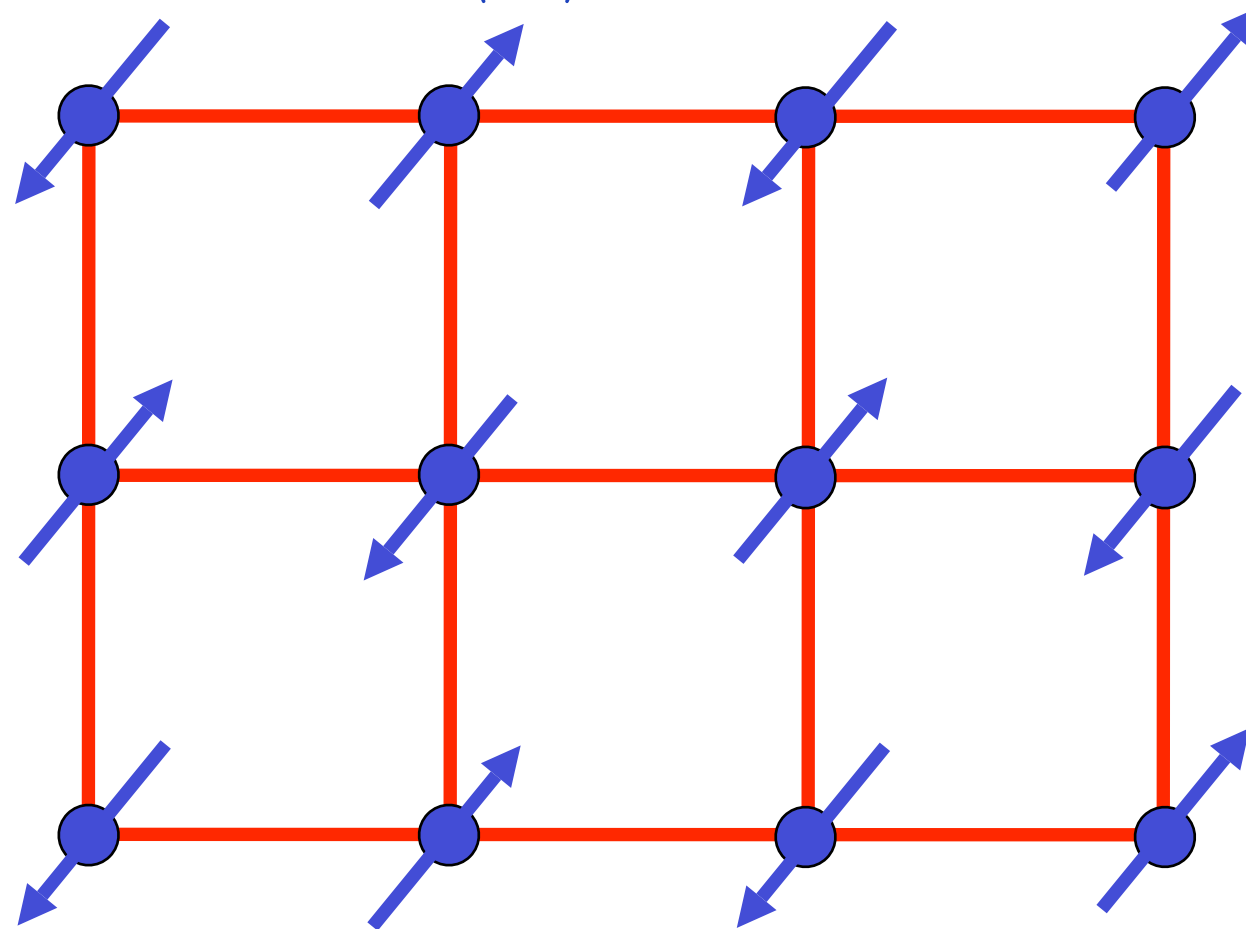
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Valence bond solid order around Zn impurities

Square lattice antiferromagnet

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



Ground state has long-range Néel order

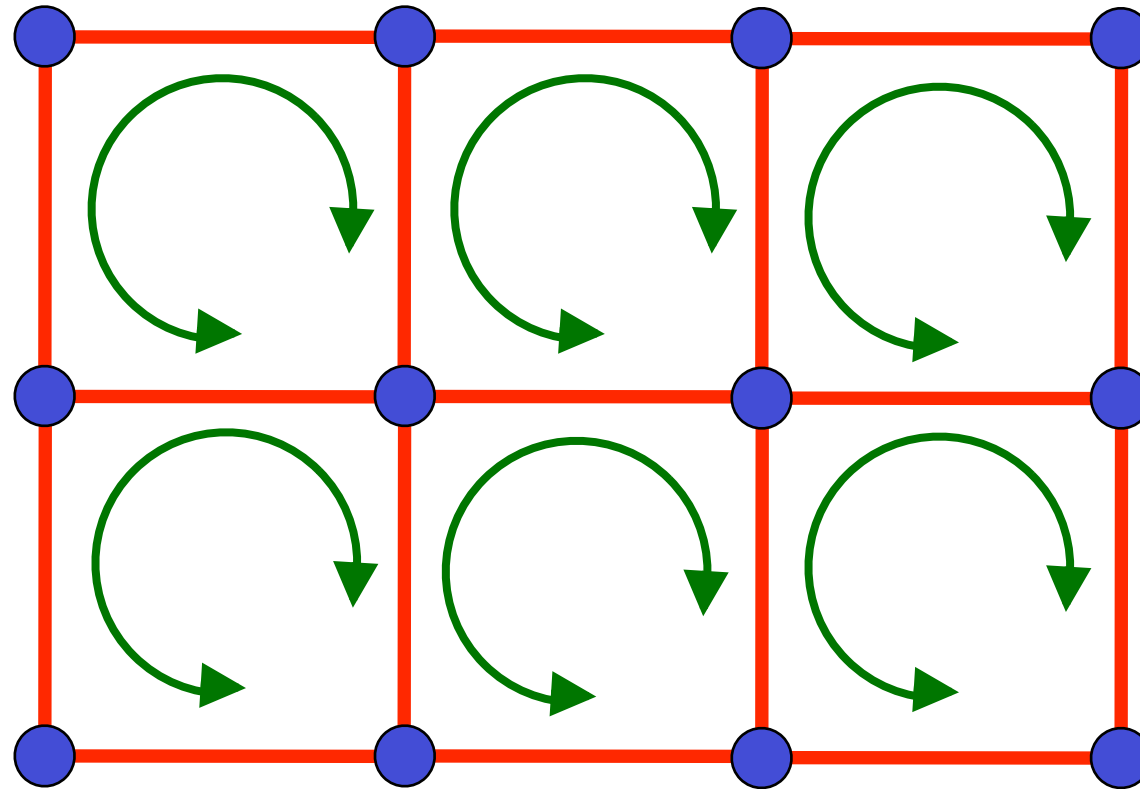
Order parameter is a single vector field $\vec{\varphi} = \eta_i \vec{S}_i$

$\eta_i = \pm 1$ on two sublattices

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

Square lattice antiferromagnet

$$H = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - Q \sum_{\langle ijkl \rangle} \left(\vec{S}_i \cdot \vec{S}_j - \frac{1}{4} \right) \left(\vec{S}_k \cdot \vec{S}_l - \frac{1}{4} \right)$$



Destroy Neel order by perturbations which
preserve full square lattice symmetry

A.W. Sandvik, *Phys. Rev. Lett.* **98**, 2272020 (2007).

R.G. Melko and R.K. Kaul, *Phys. Rev. Lett.* **100**, 017203 (2008).

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

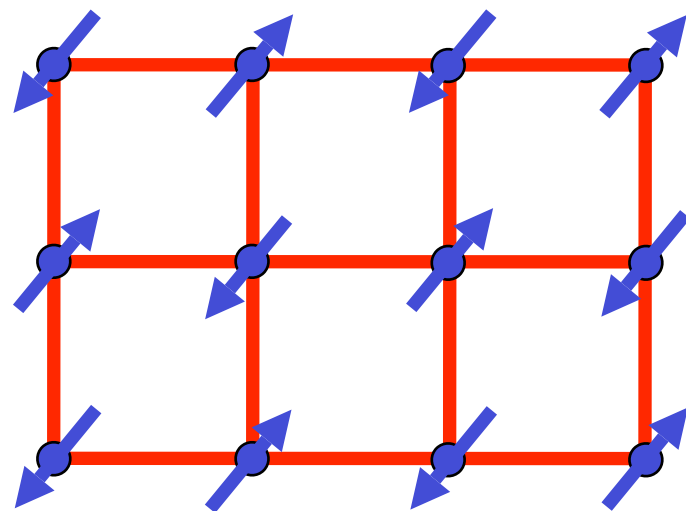
Low energy spinon theory for “quantum disordering” the Néel state is the CP^1 model

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

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



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

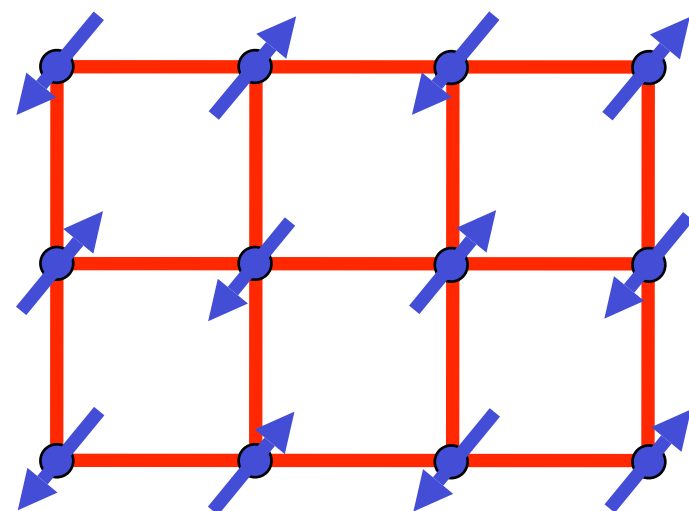
Néel state

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

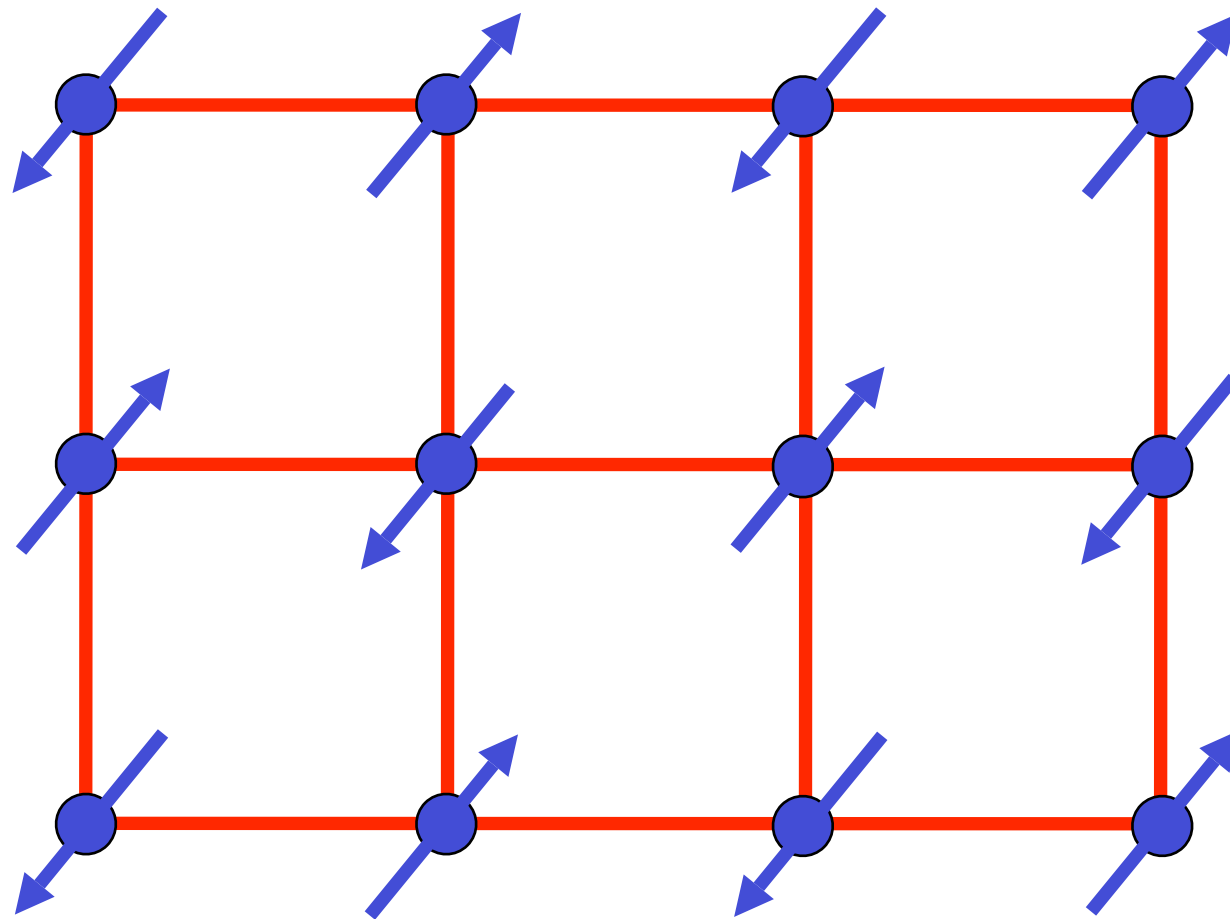
[Unstable to valence bond solid (VBS) order]

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

s_c

s

From the square to the triangular lattice

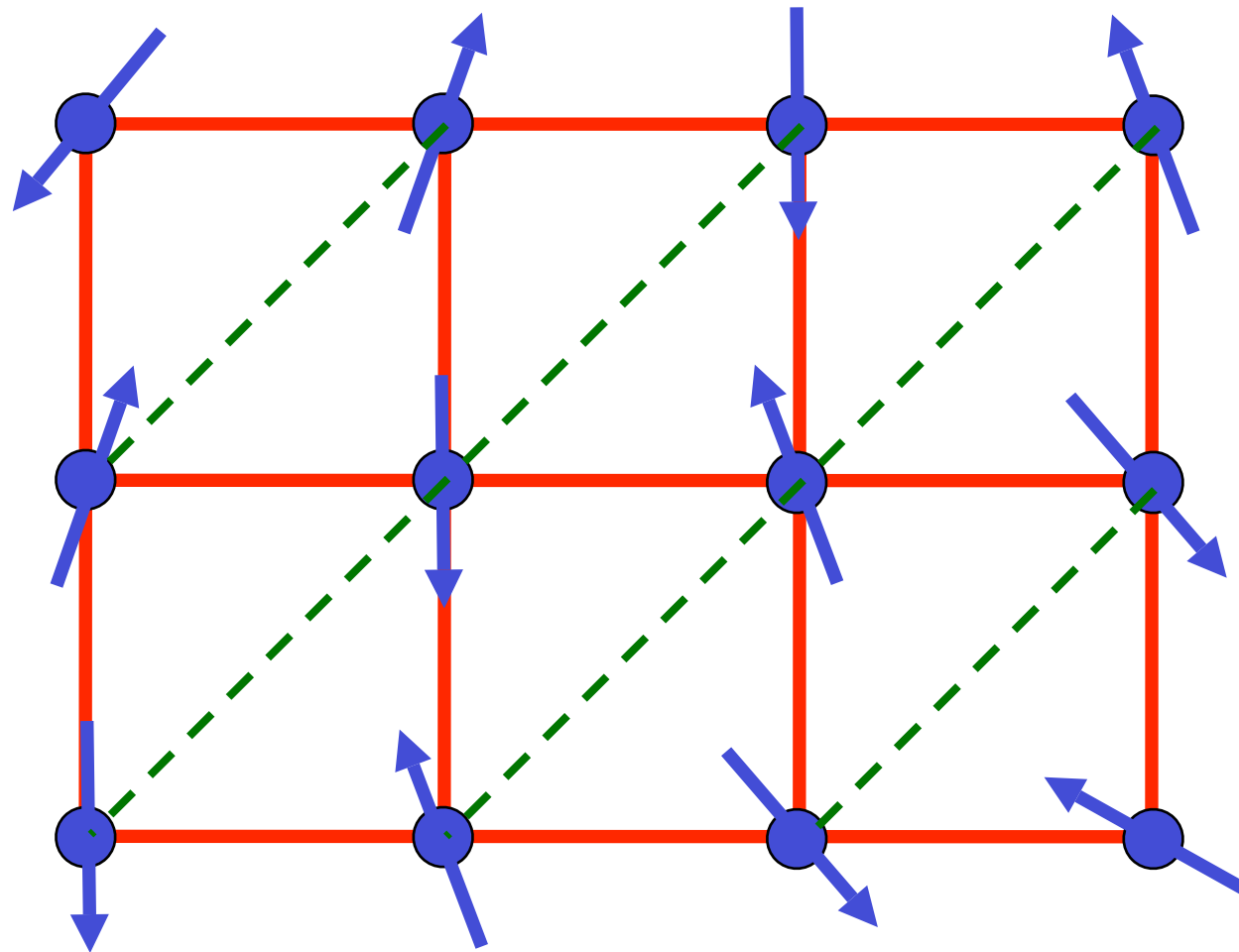


A spin density wave with

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

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

From the square to the triangular lattice



A spin density wave with

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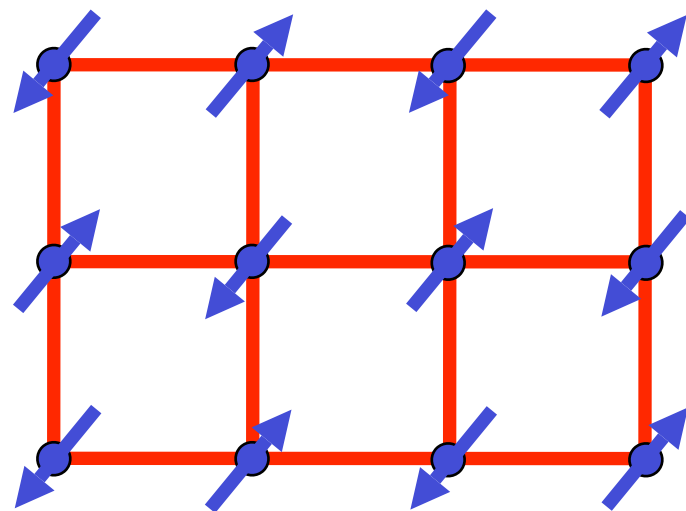
and $\mathbf{K} = (\pi + \Phi, \pi + \Phi)$.

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

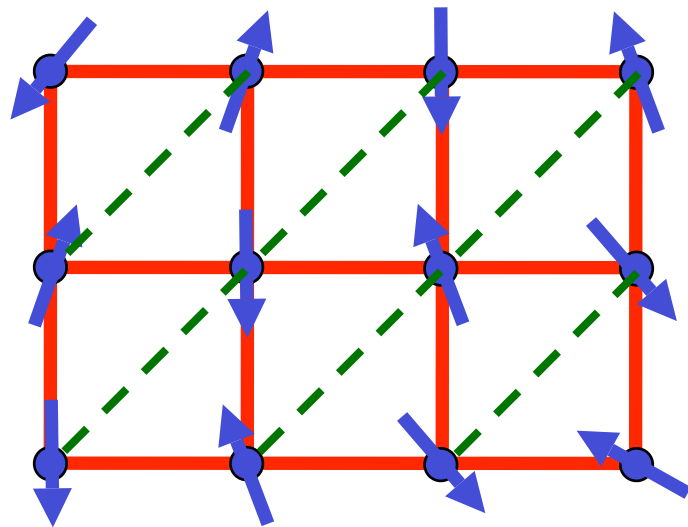
Spin liquid with a
“**photon**” collective mode

[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 ?

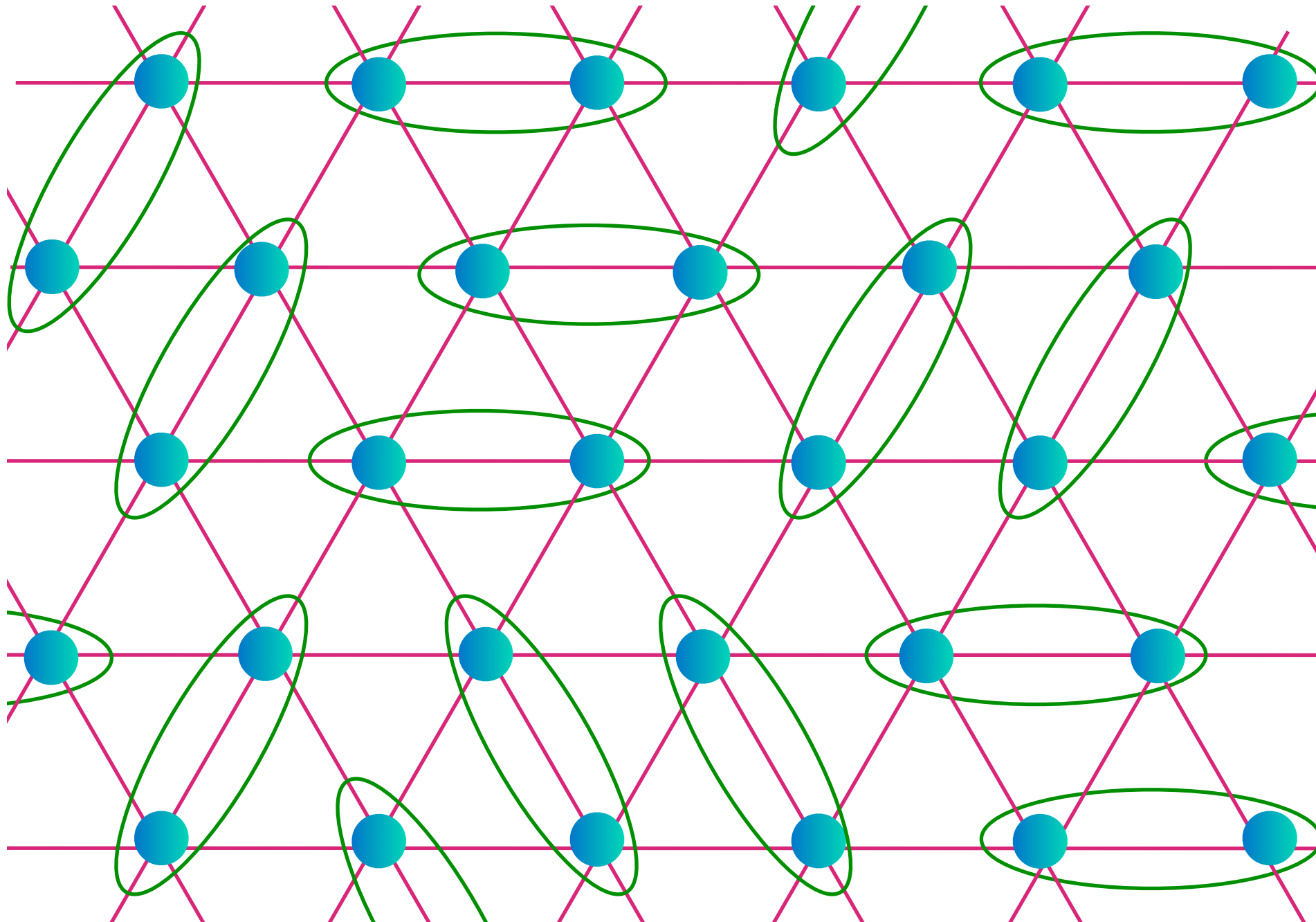
A vison is an Abrikosov vortex in the spinon pair field Φ .

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

What is a vison ?

Z_2 Spin liquid

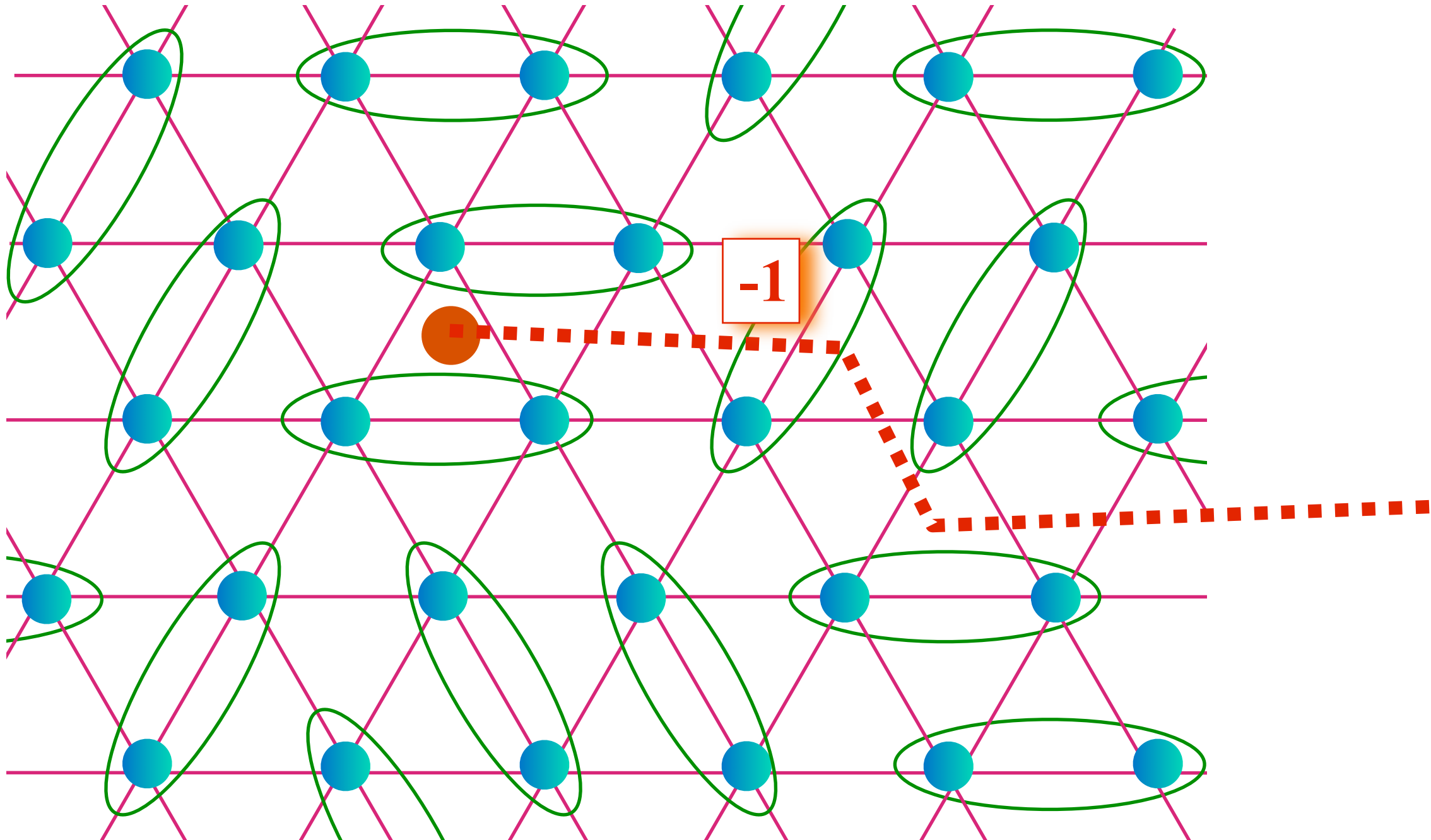
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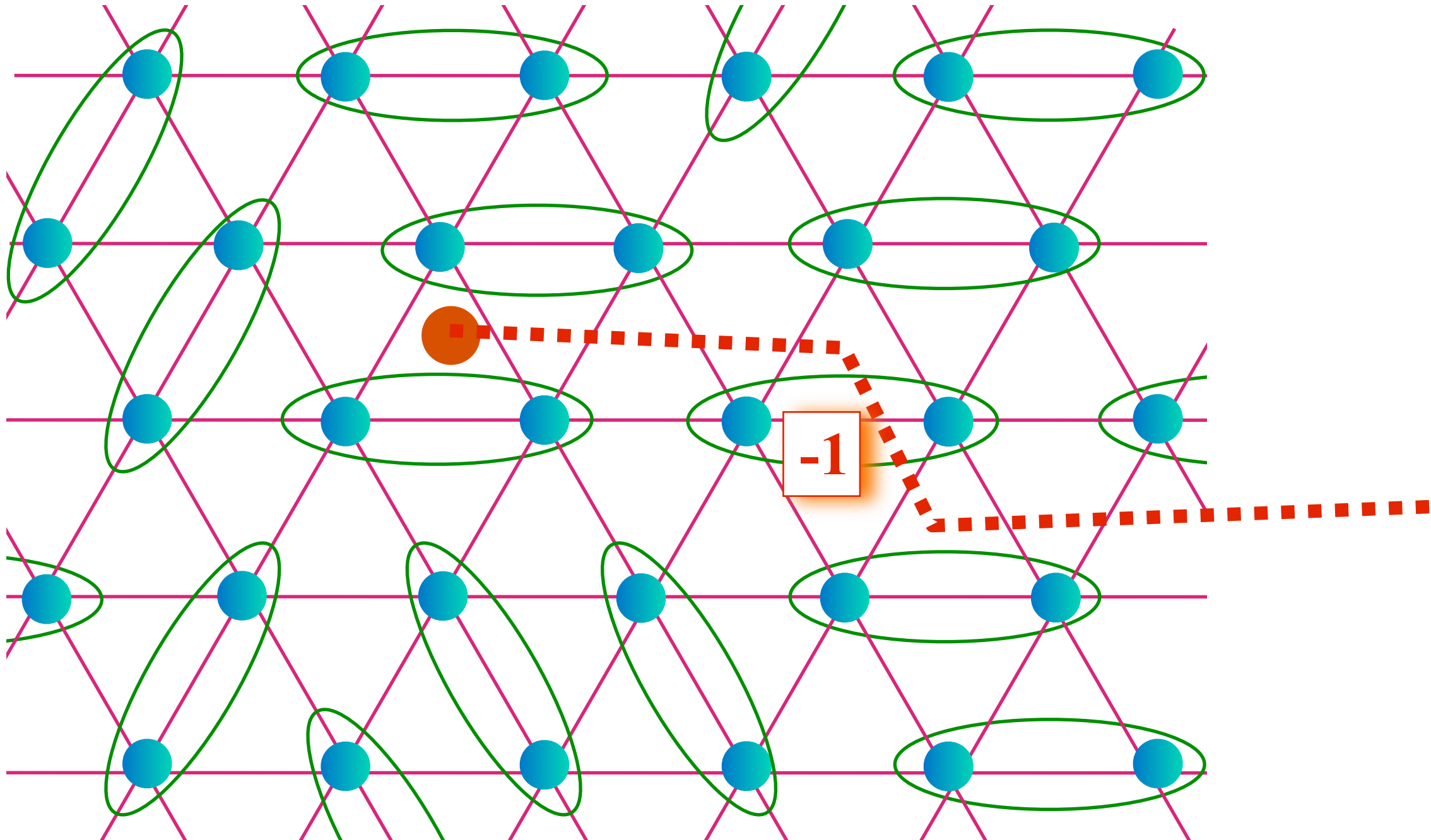


N. Read and B. Chakraborty, *Phys. Rev. B* **40**, 7133 (1989)
N. Read and S. Sachdev, *Phys. Rev. Lett.* **66**, 1773 (1991)

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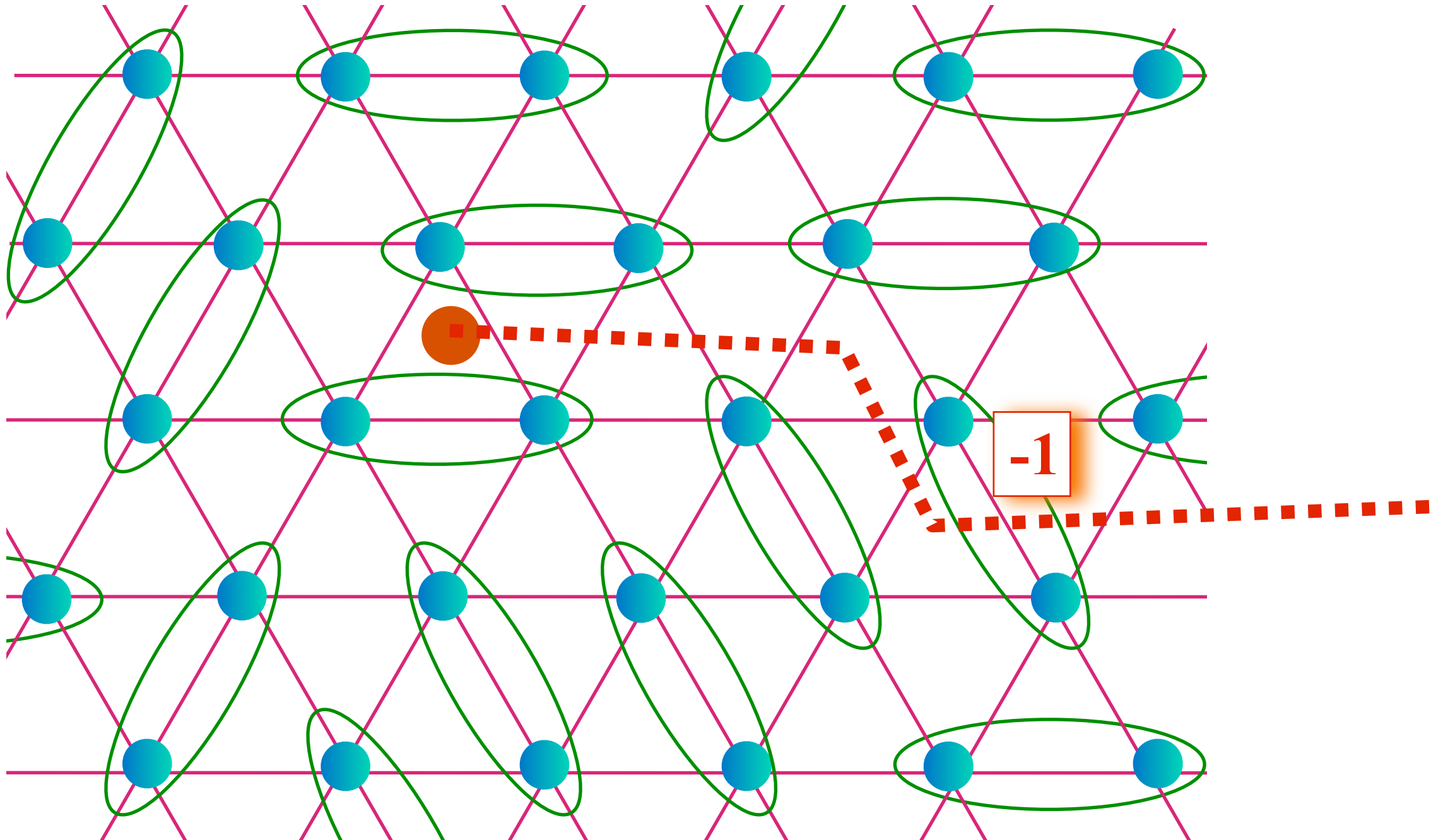
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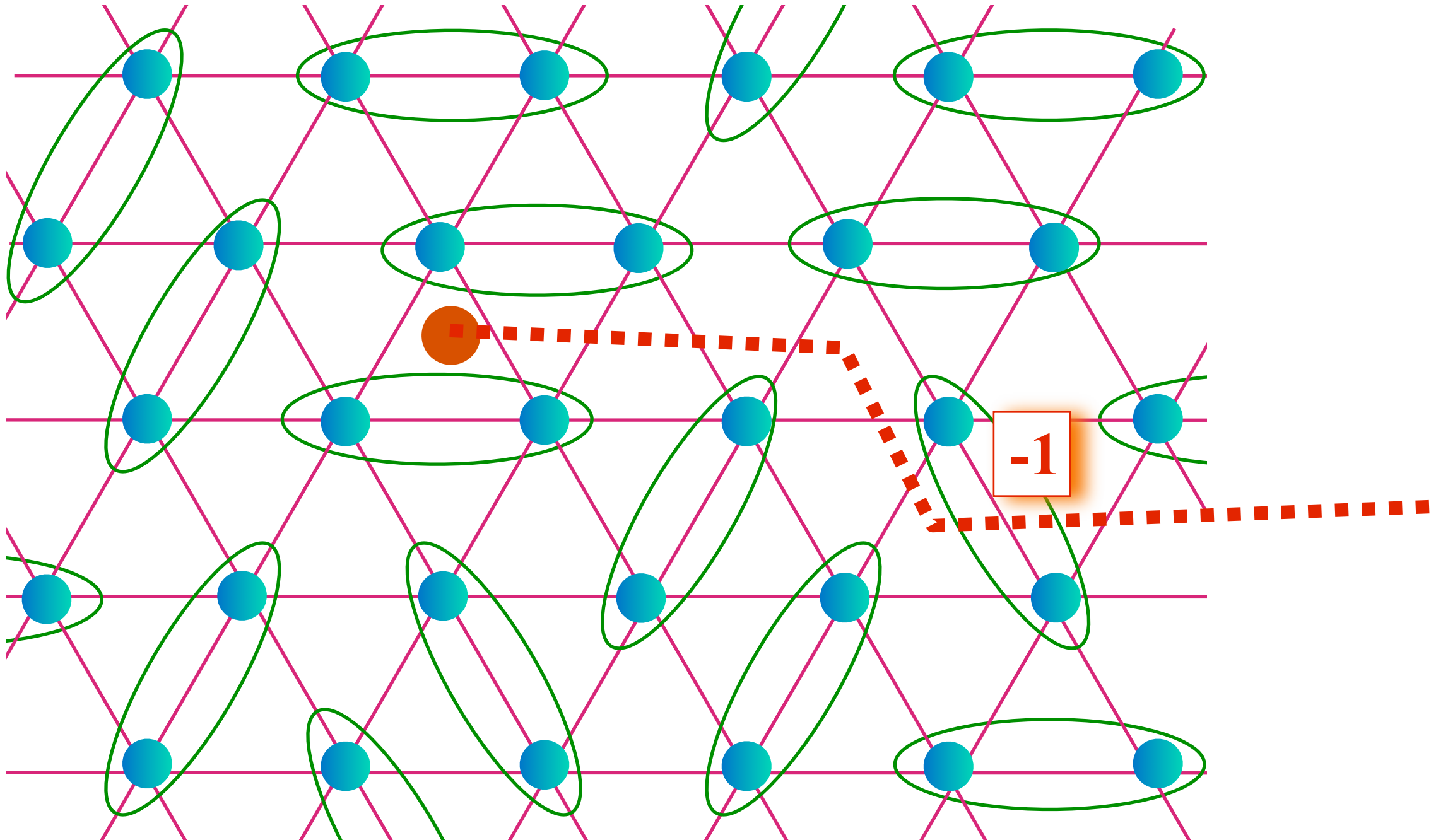


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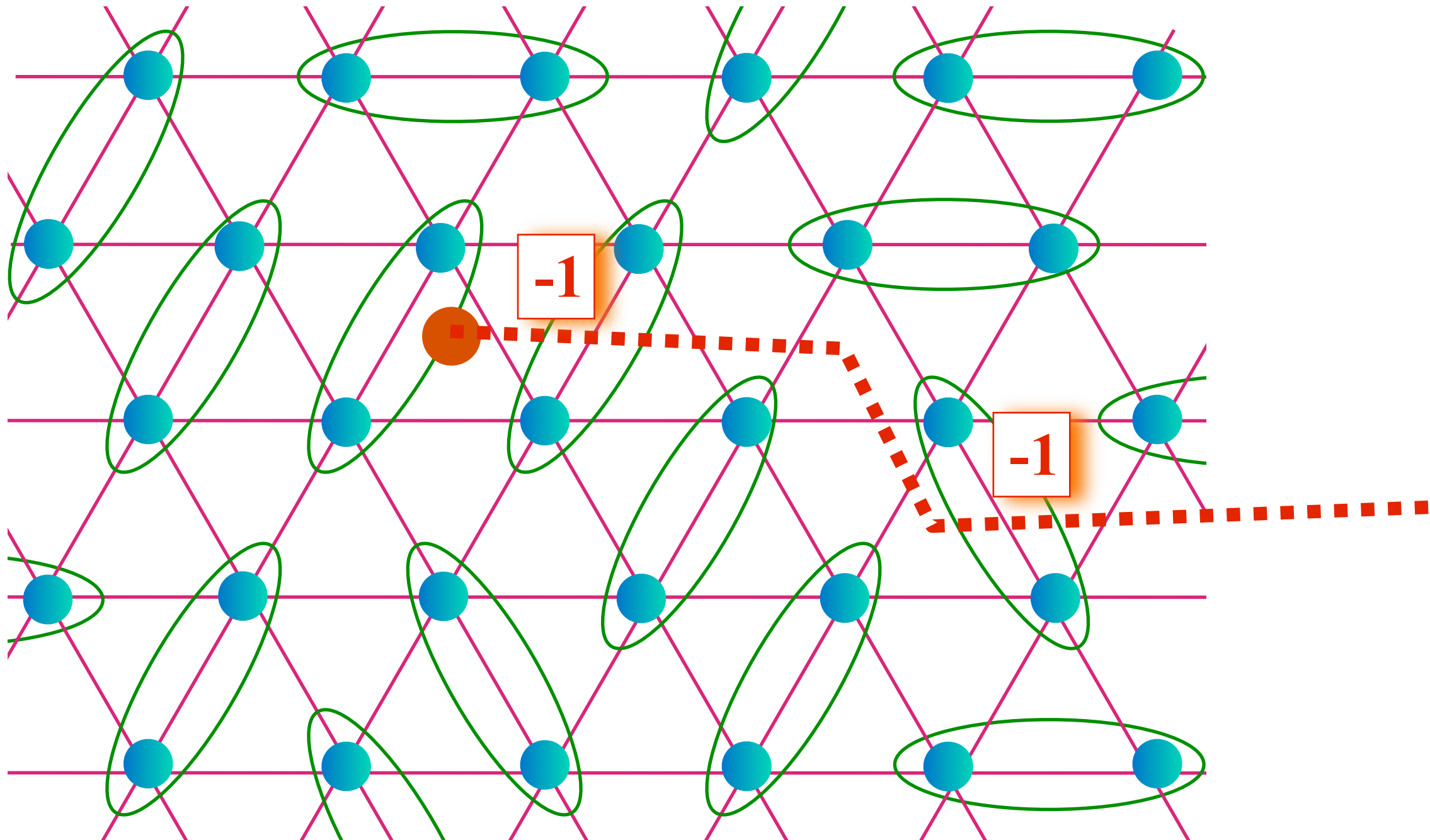


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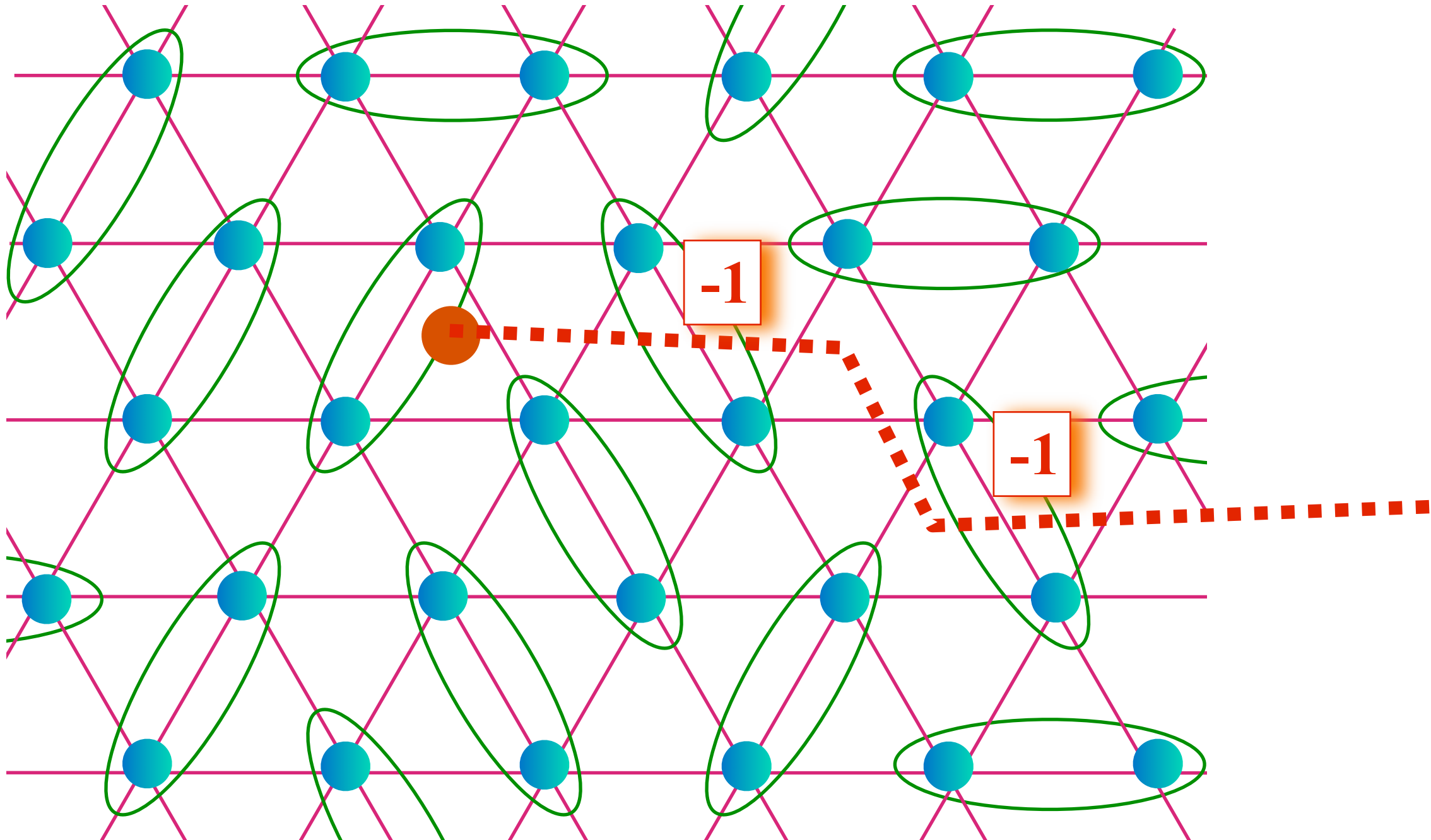


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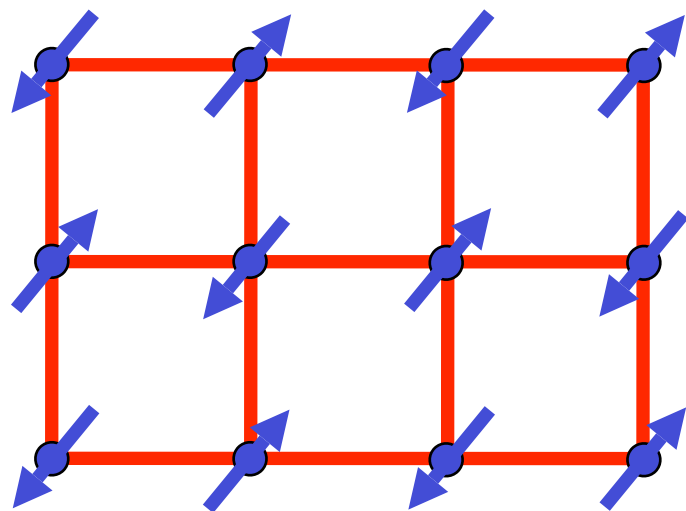
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N. Read and B. Chakraborty, *Phys. Rev. B* **40**, 7133 (1989)
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Global phase diagram



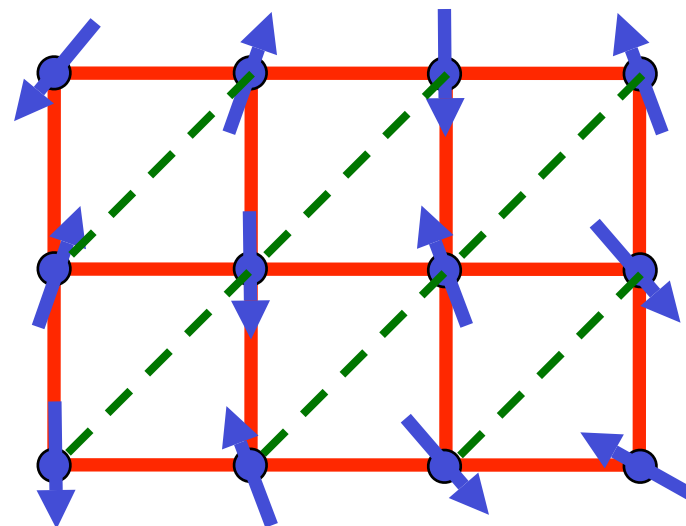
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Néel state

Spin liquid with a
“photon” collective mode

[Unstable to valence bond solid (VBS) order]

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



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

non-collinear Néel state

Z_2 spin liquid with a
vison excitation

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

$\tilde{s}$

s

Mutual Chern-Simons Theory

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

The spinons and visons have mutual semionic statistics, and this leads to the continuum theory:

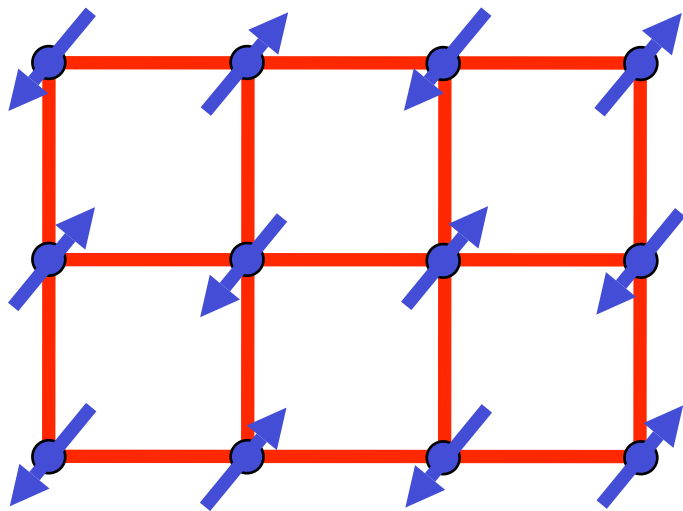
$$\begin{aligned} \mathcal{S} = \int d^2x d\tau & \left[c^2 |(\nabla_x - iA_x)z_\alpha|^2 + |(\partial_\tau - iA_\tau)z_\alpha|^2 + s |z_\alpha|^2 + \dots \right. \\ & + \tilde{c}^2 |(\nabla_x - iB_x)v|^2 + |(\partial_\tau - iB_\tau)v|^2 + \tilde{s} |v|^2 + \dots \\ & \left. + \frac{i}{\pi} \epsilon_{\mu\nu\lambda} B_\mu \partial_\nu A_\lambda \right] \end{aligned}$$

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This theory fully accounts for all the phases, including their global topological properties and their broken symmetries. It also completely describe the quantum phase transitions between them.

Global phase diagram



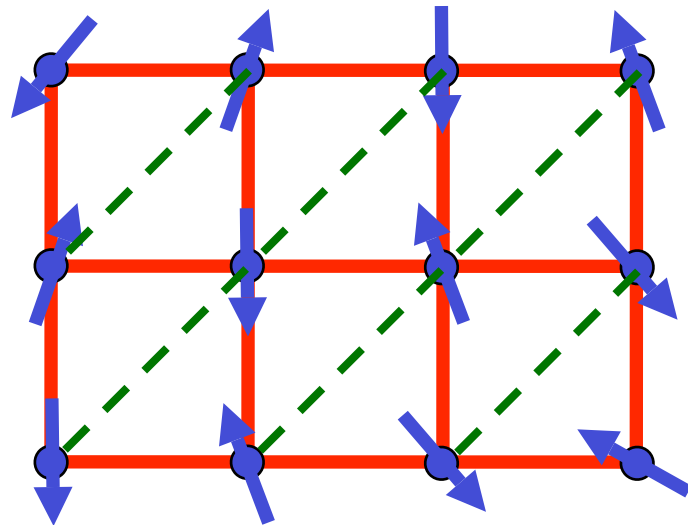
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Low energy states on a torus:

- Z_2 spin liquid has a 4-fold degeneracy.
- Non-collinear Néel state has low-lying tower of states described by a broken symmetry with order parameter S_3/Z_2 .
- “Photon” spin liquid has a low-lying tower of states described by a broken symmetry with order parameter S_1/Z_2 . This is the VBS order $\sim v^2$.
- Néel state has a low-lying tower of states described by a broken symmetry with order parameter $S_3 \times S_1 / (U(1) \times U(1)) \equiv S_2$. This is the usual vector Néel order.

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1. Collective excitations of spin liquids in two dimensions

Photons and visons

2. Detecting the vison

Thermal conductivity of $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

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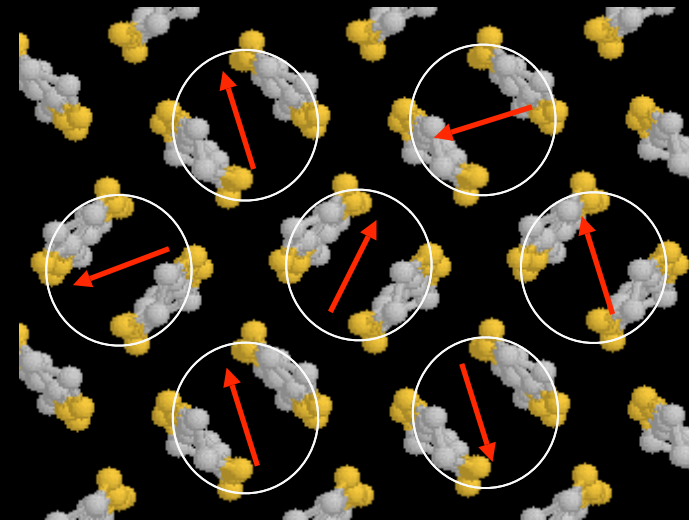
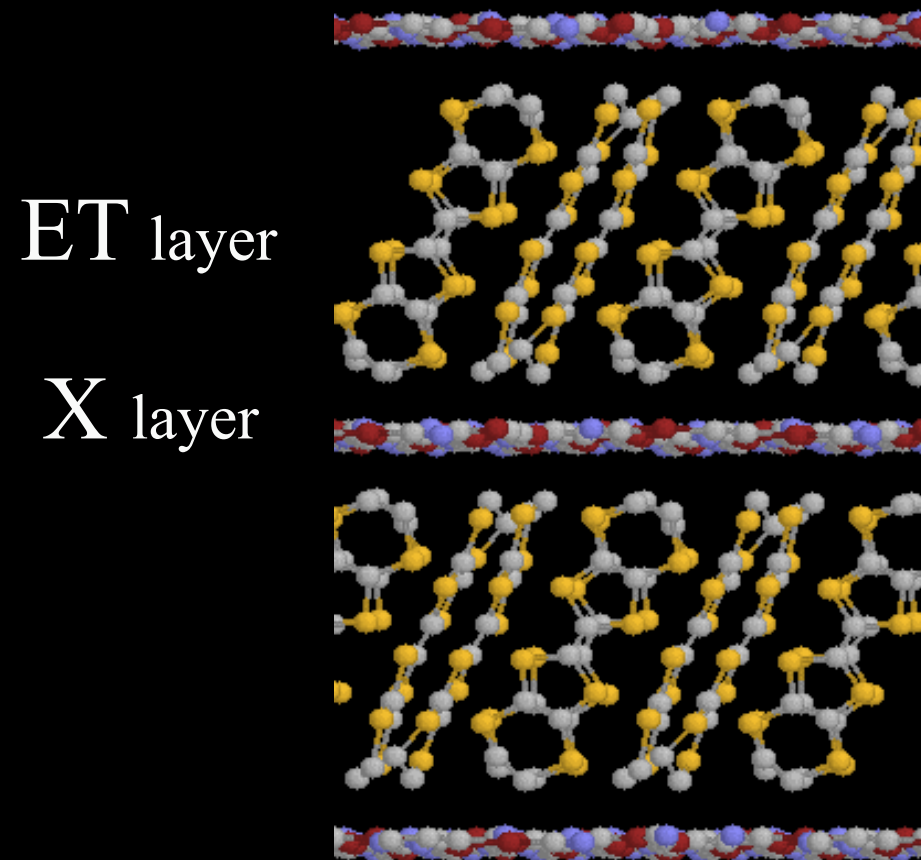
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The following slides and
thermal conductivity data
are from:

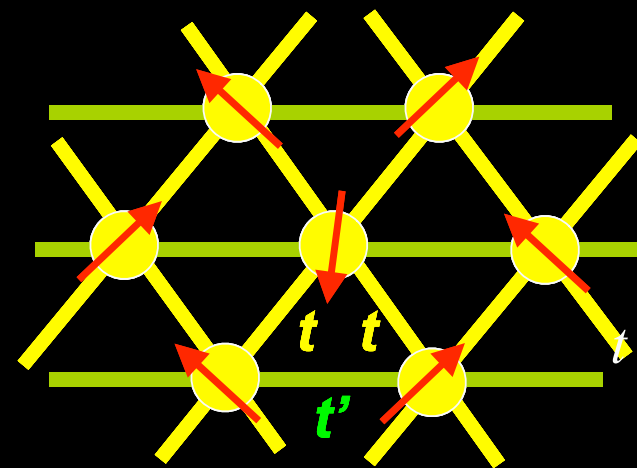
M. Yamashita, H. Nakata, Y. Kasahara, S. Fujimoto, T. Shibauchi, Y. Matsuda, T. Sasaki, N. Yoneyama, and N. Kobayashi, preprint and 25th International Conference on Low Temperature Physics, SaM3-2, Amsterdam, August 9, 2008.

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



Kino & Fukuyama

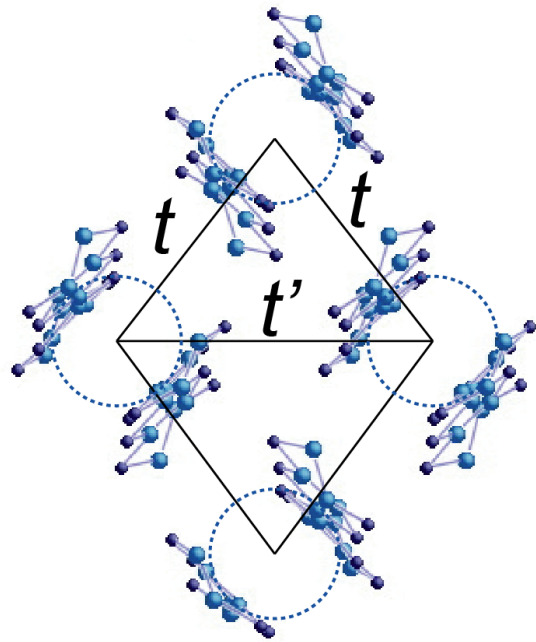
dimer model



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

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

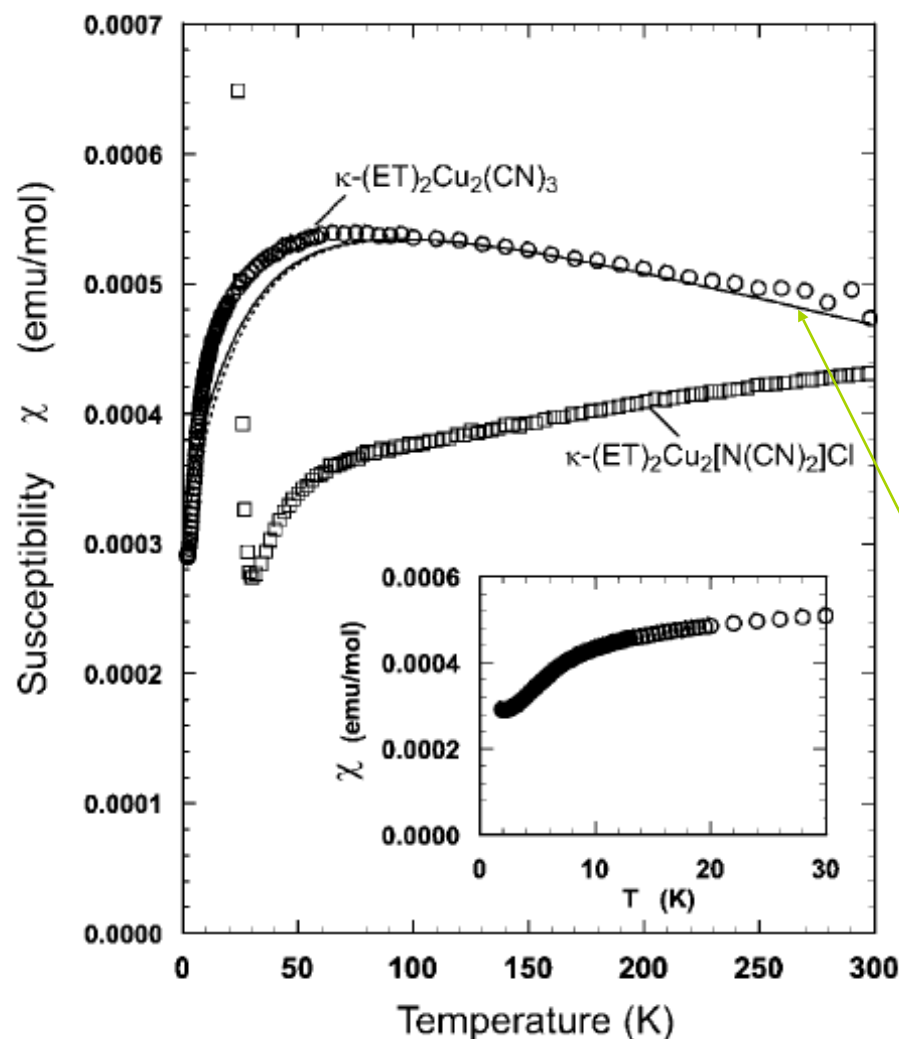


- face-to-face pairs of BEDT-TTF molecules form dimers by strong coupling.

- 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.

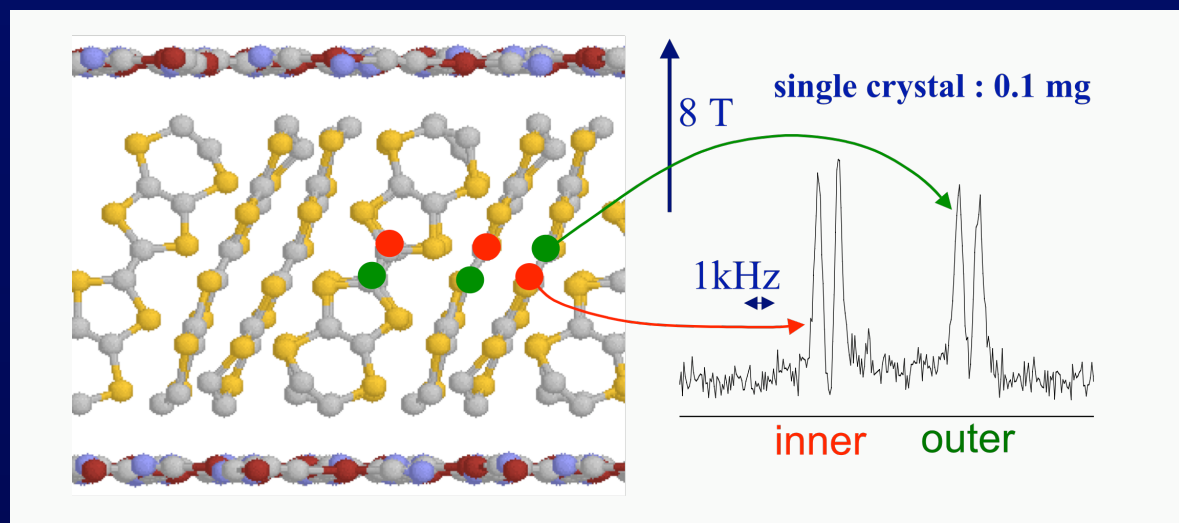
Heisenberg High-T Expansion
(PRL, 71 1629 (1993))

$J \sim 250$ K

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

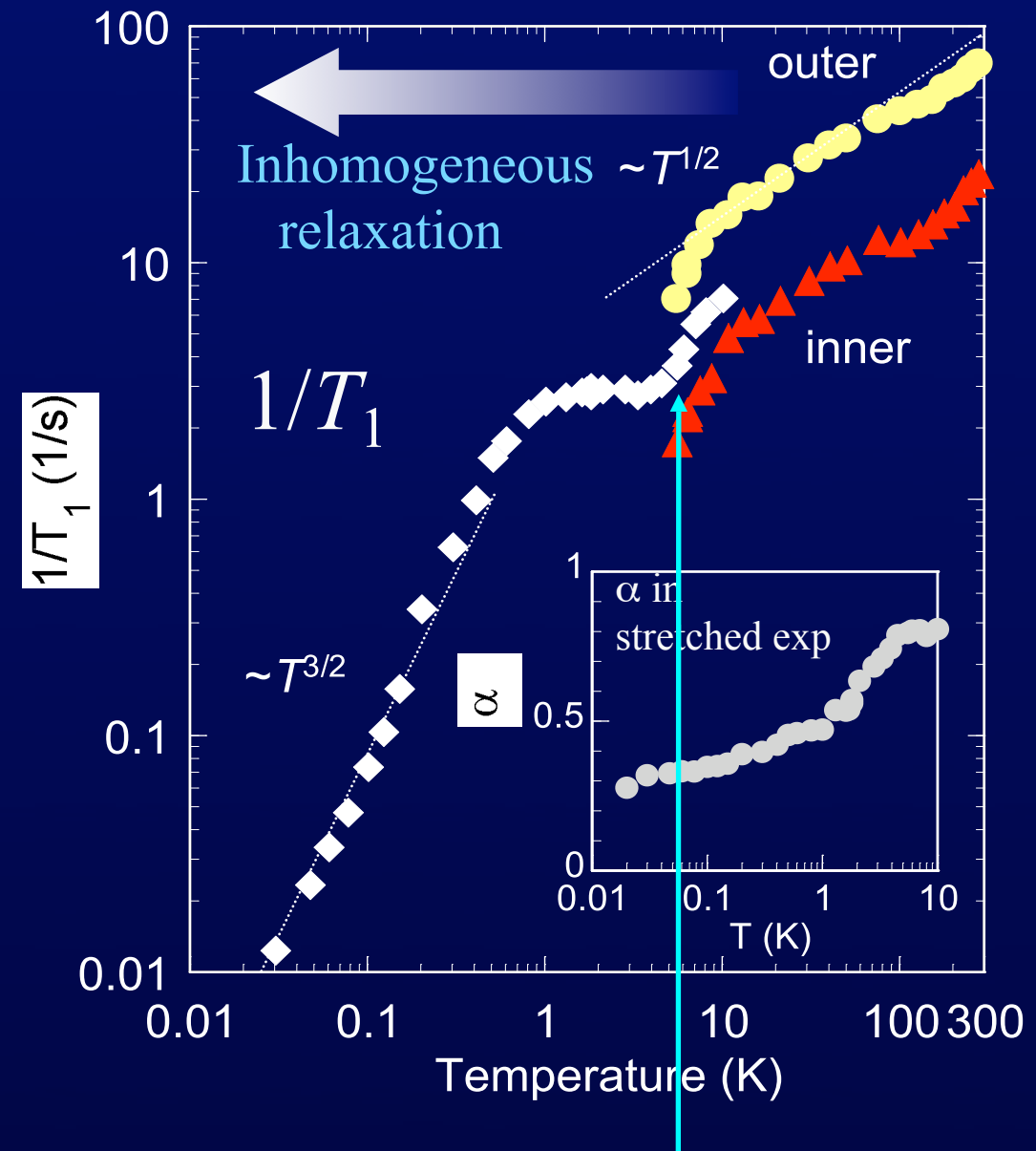
^{13}C NMR relaxation rate

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

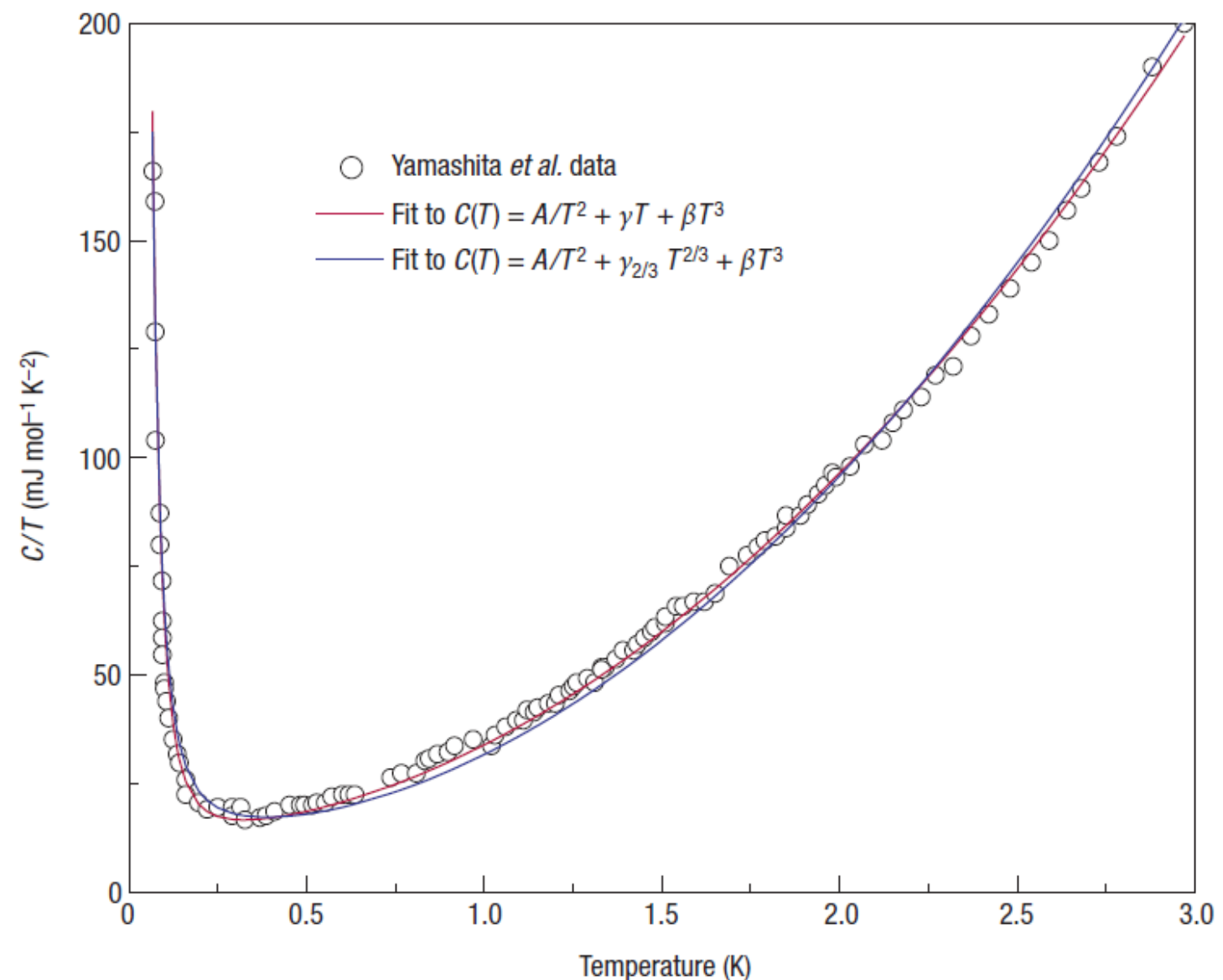
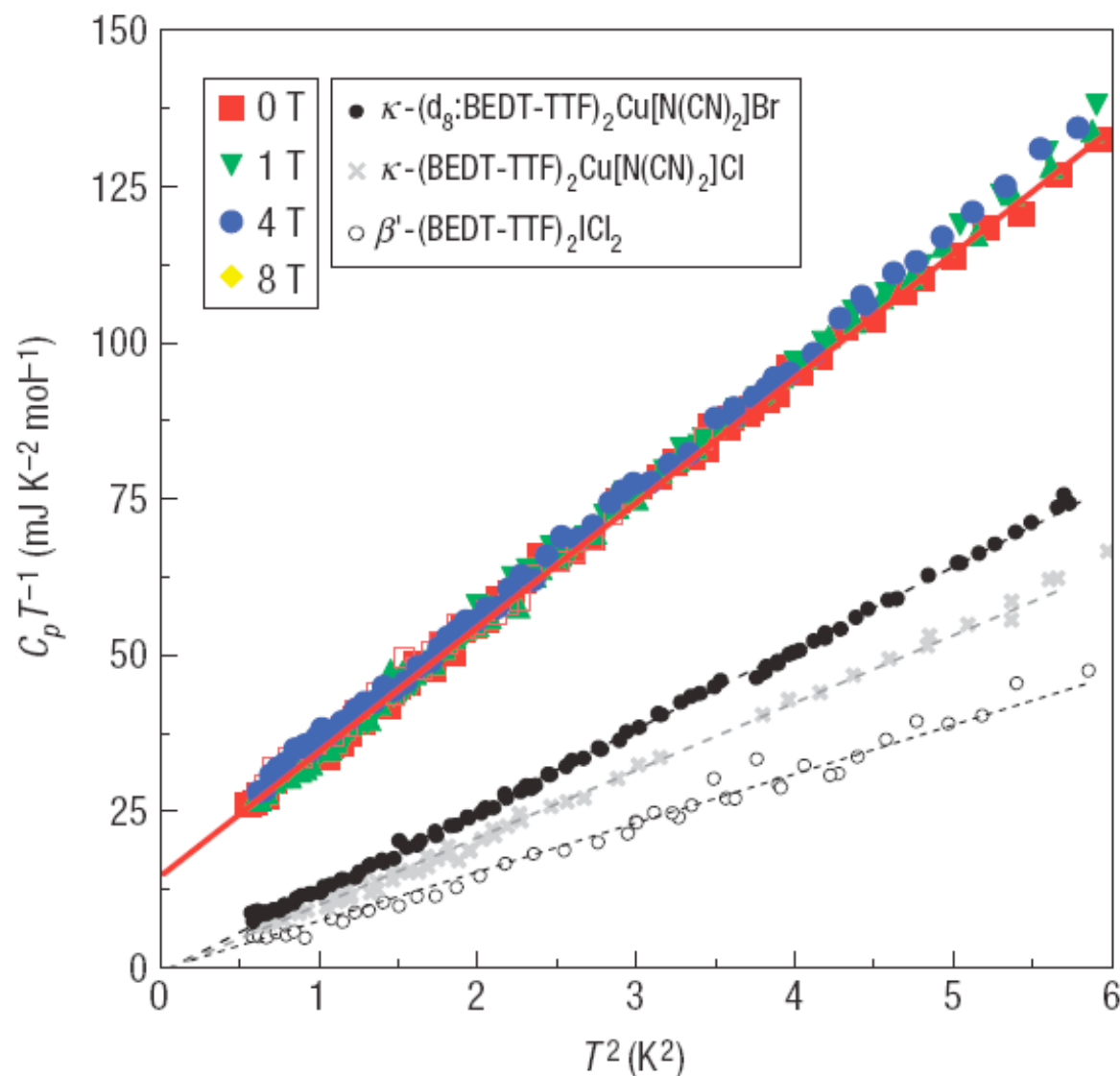
Heat capacity measurements

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

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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?

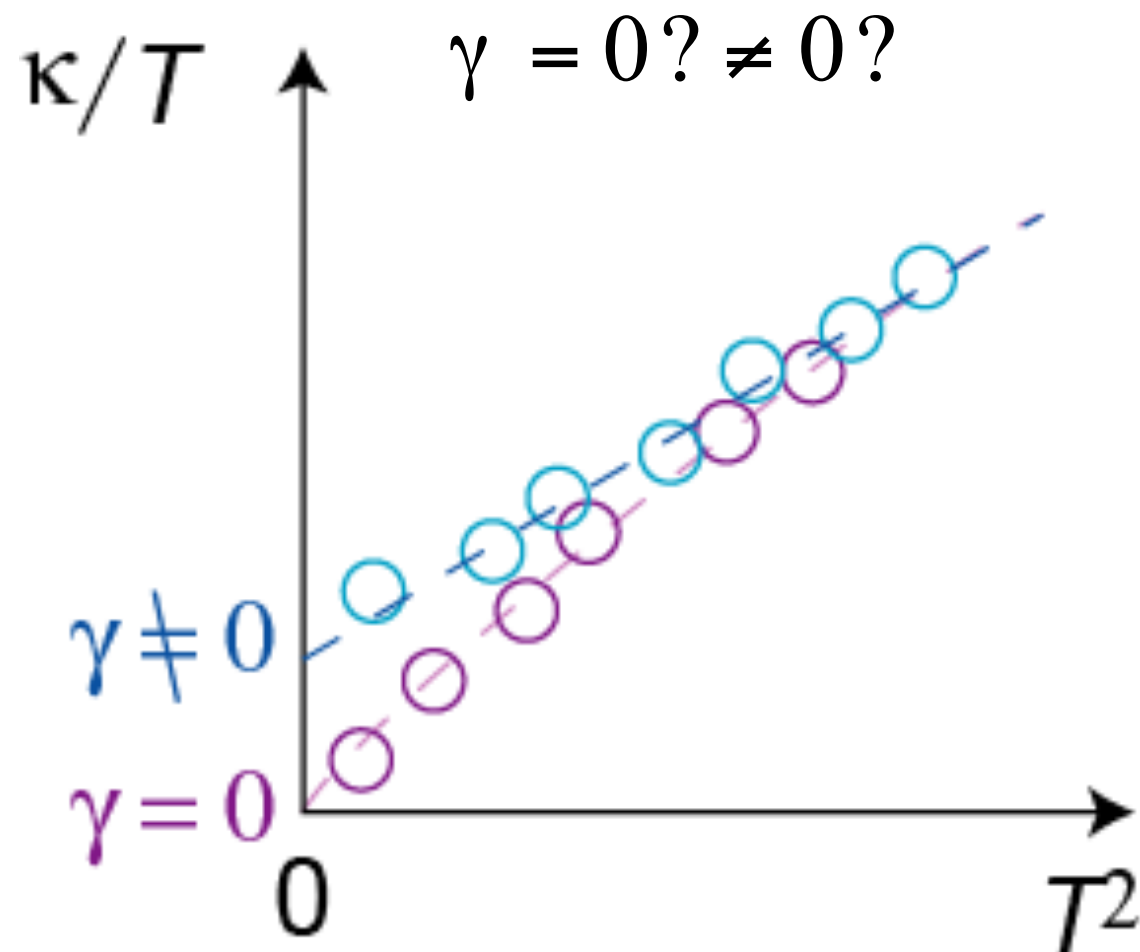


Thermal-Transport Measurements

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.

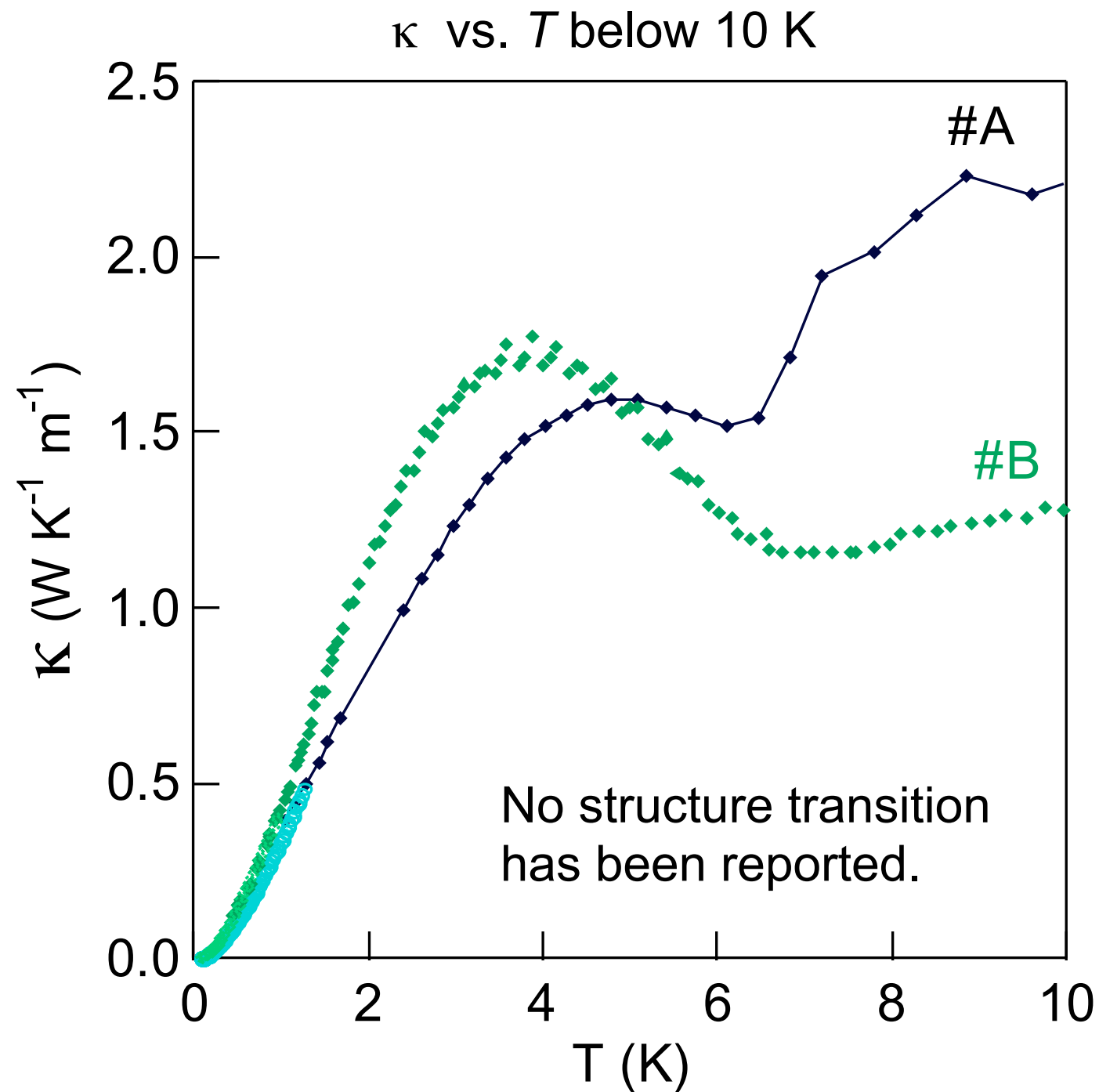


$$\frac{\kappa}{T} \approx \frac{1}{T} (C \times v) \propto \gamma + \beta T^2$$

$C \propto \gamma T + \beta T^3$
 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

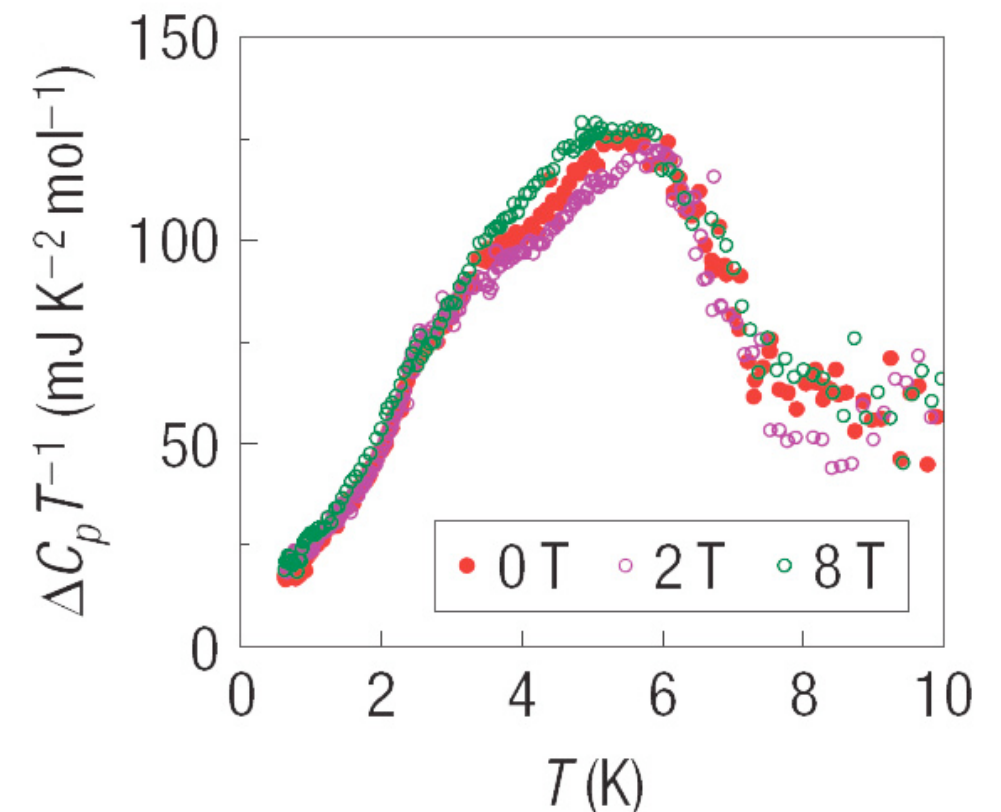


Similar to

- $1/T_1$ 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?

Thermal Conductivity below 300 mK

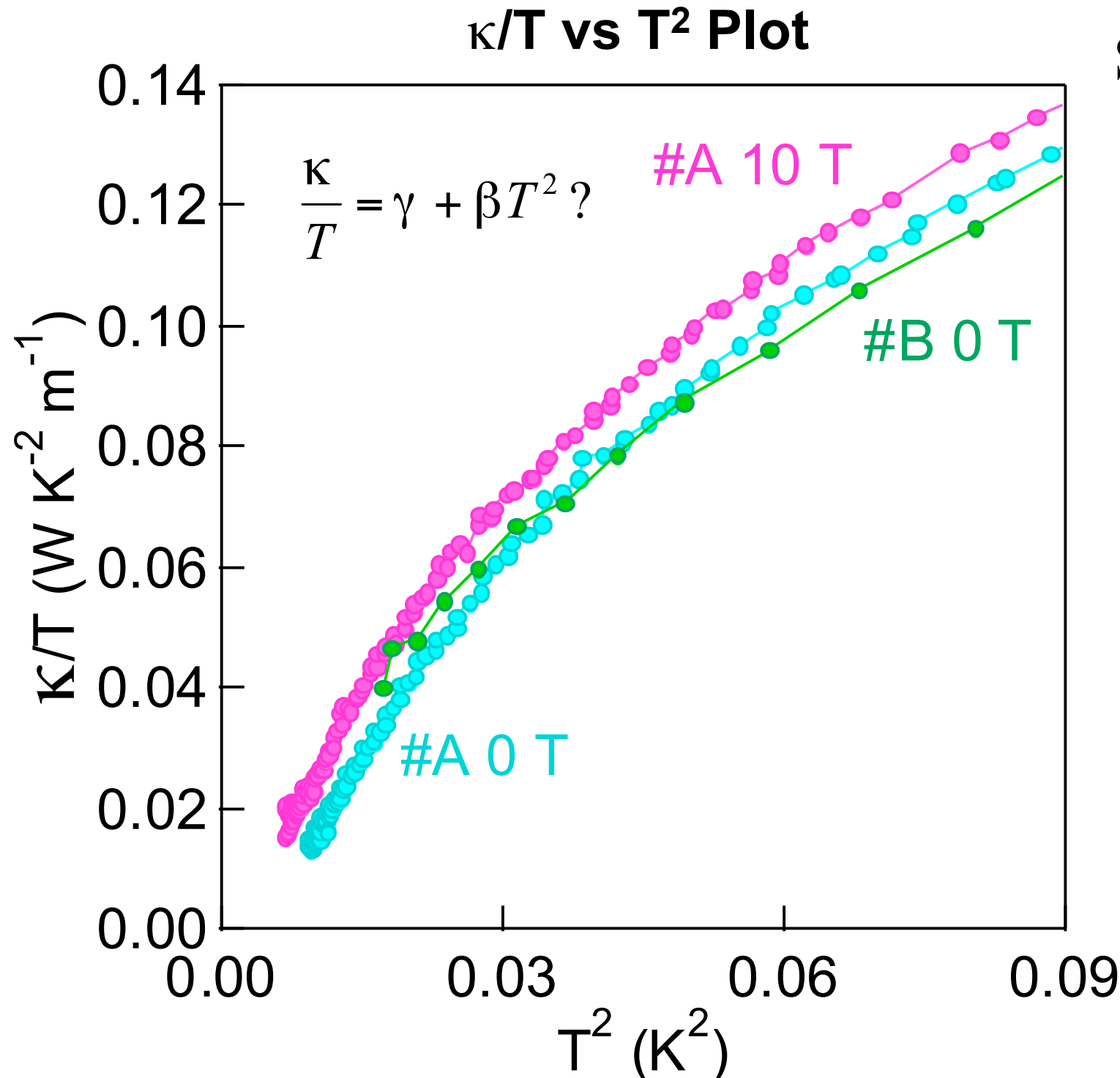
- Convex, non- T^3 dependence in κ
- Magnetic fields enhance κ



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

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

Substantial portion of κ_{mag} in κ

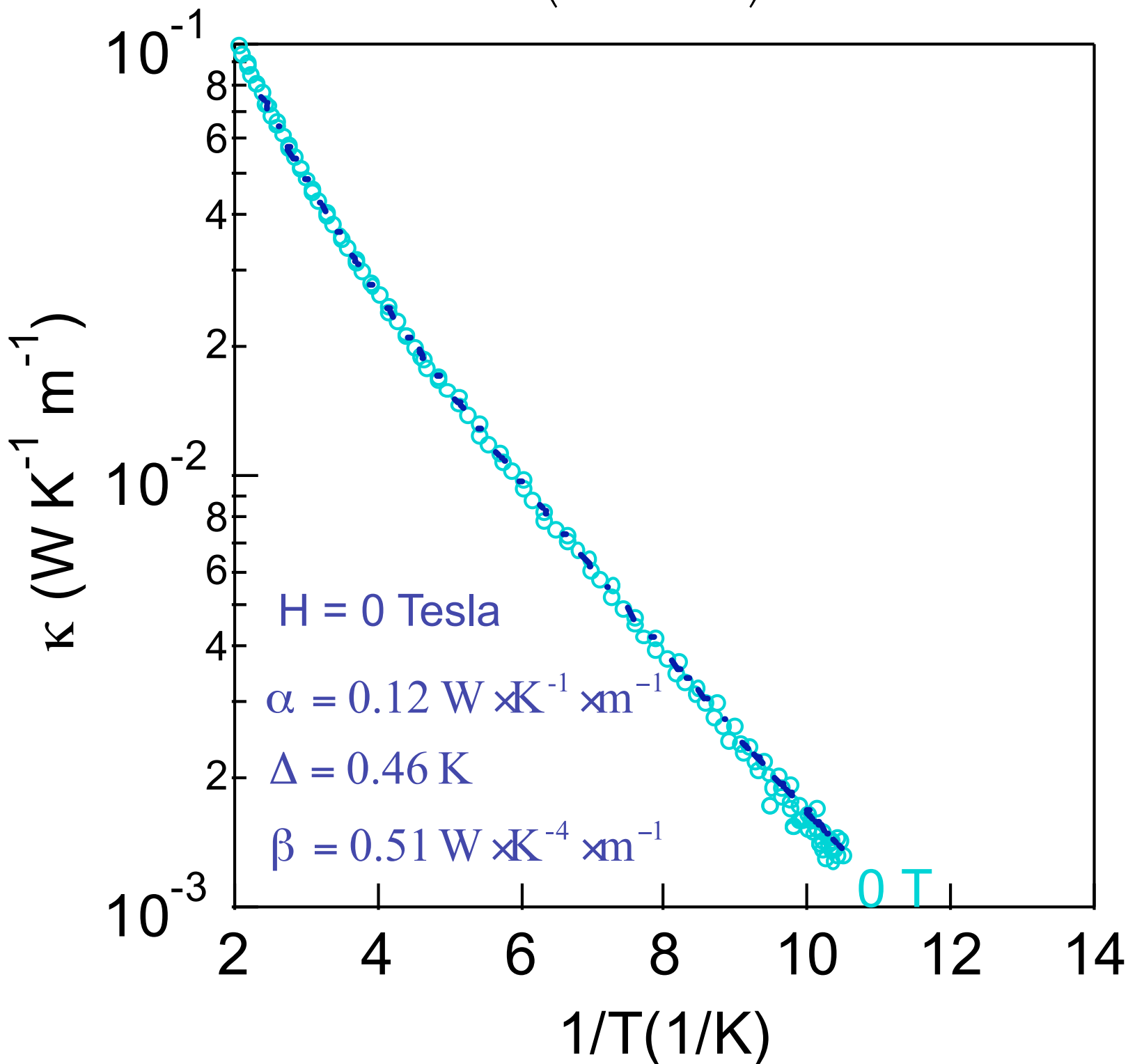


$$\underline{\gamma = 0}$$

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

➤ $\Delta = 0.46$ K $\sim J/500$

How do we reconcile power-law T dependence
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I. Thermal conductivity is dominated by vison transport

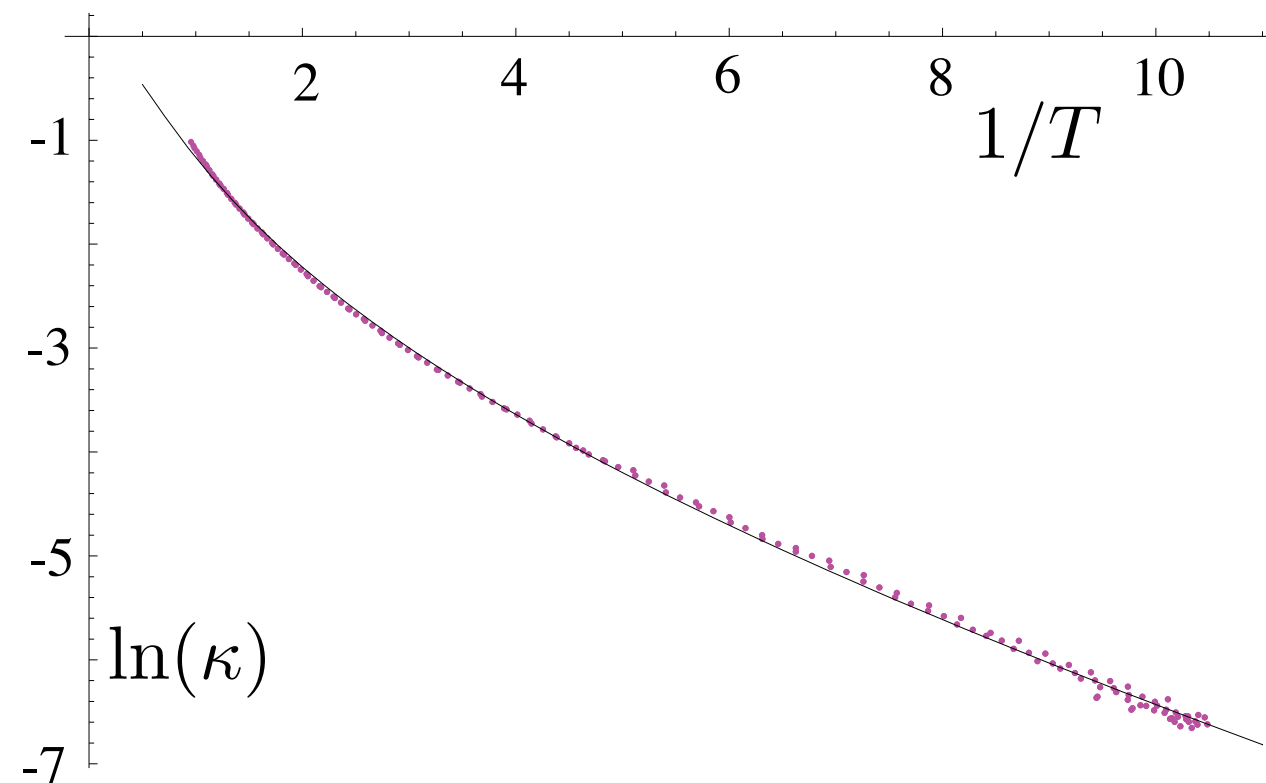
The thermal conductivity (per layer) of N_v species of slowly moving visons of mass m_v , above an energy gap Δ_v , scattering off impurities of density n_{imp} is

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

where T_v is of order the vison bandwidth.

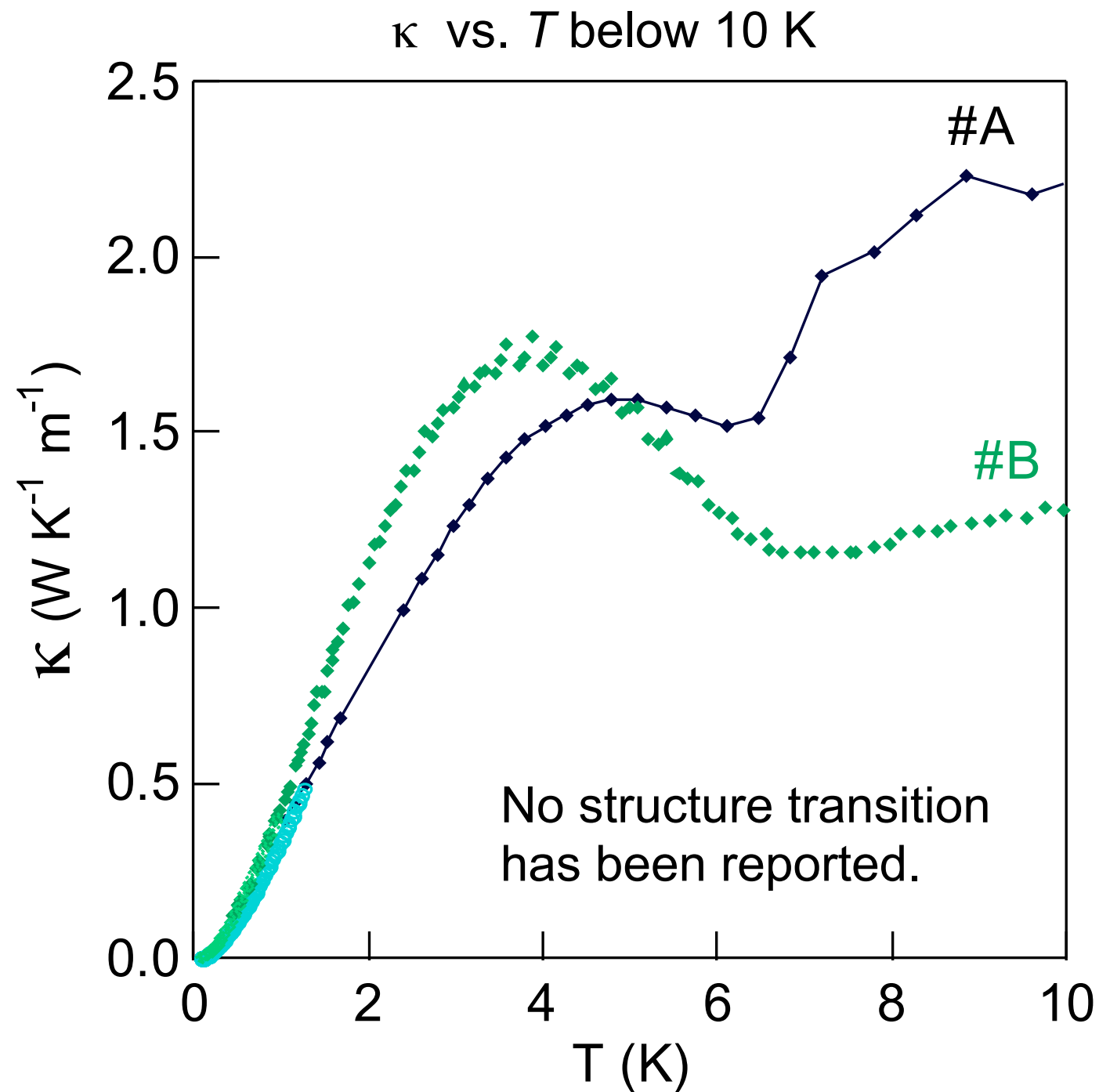
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Best fit to data yields, $\Delta_v \approx 0.24$ K and $T_v \approx 8$ K.

Thermal Conductivity below 10K

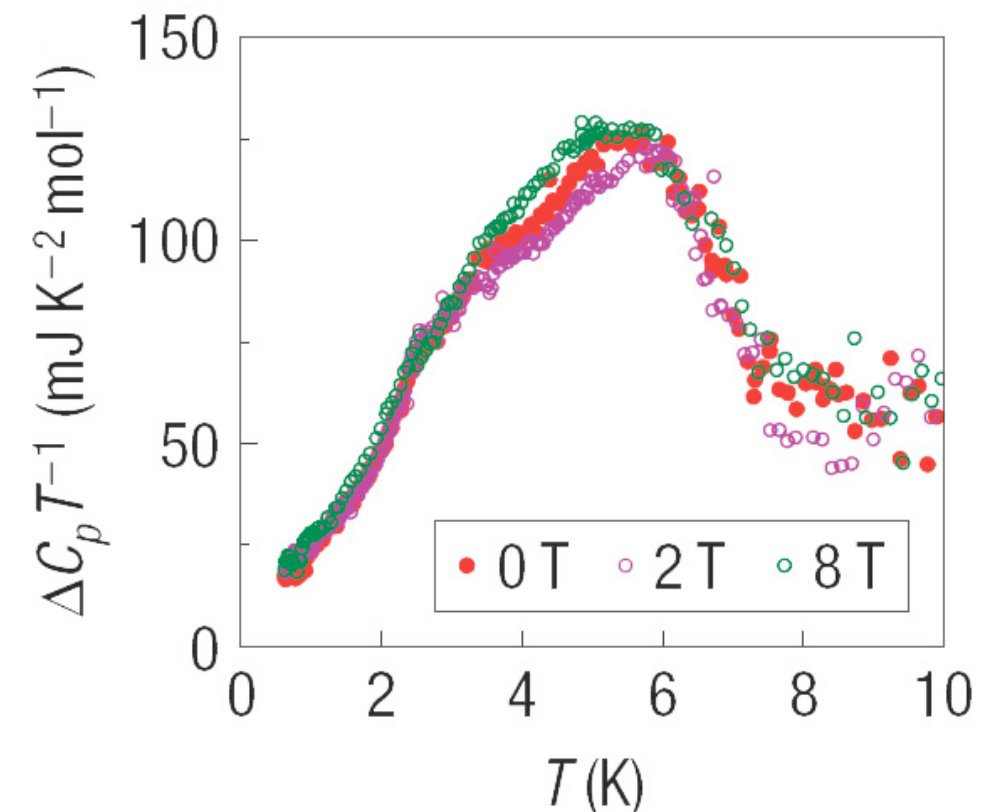


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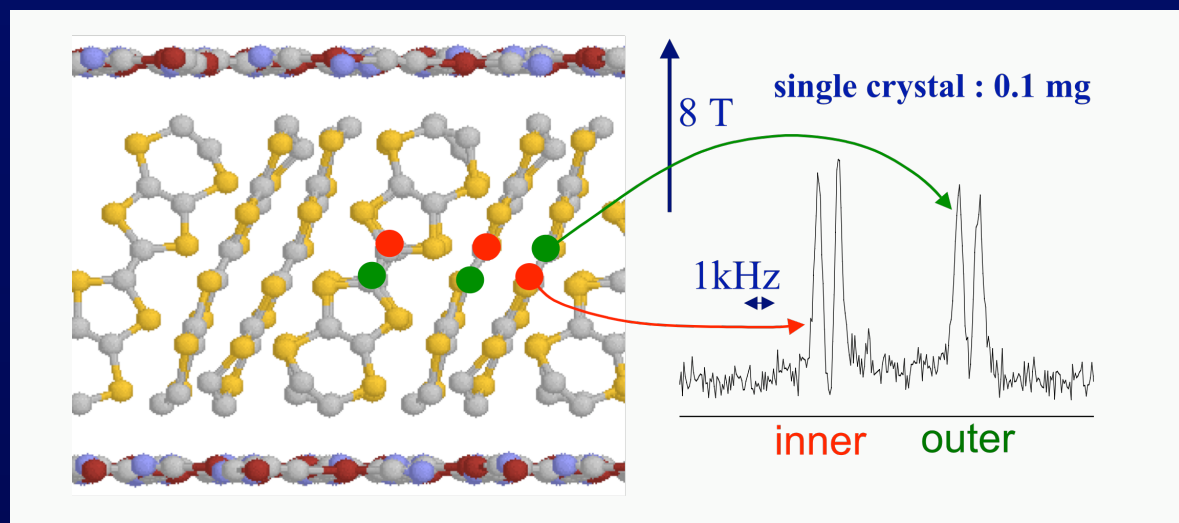
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Spin excitation in $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

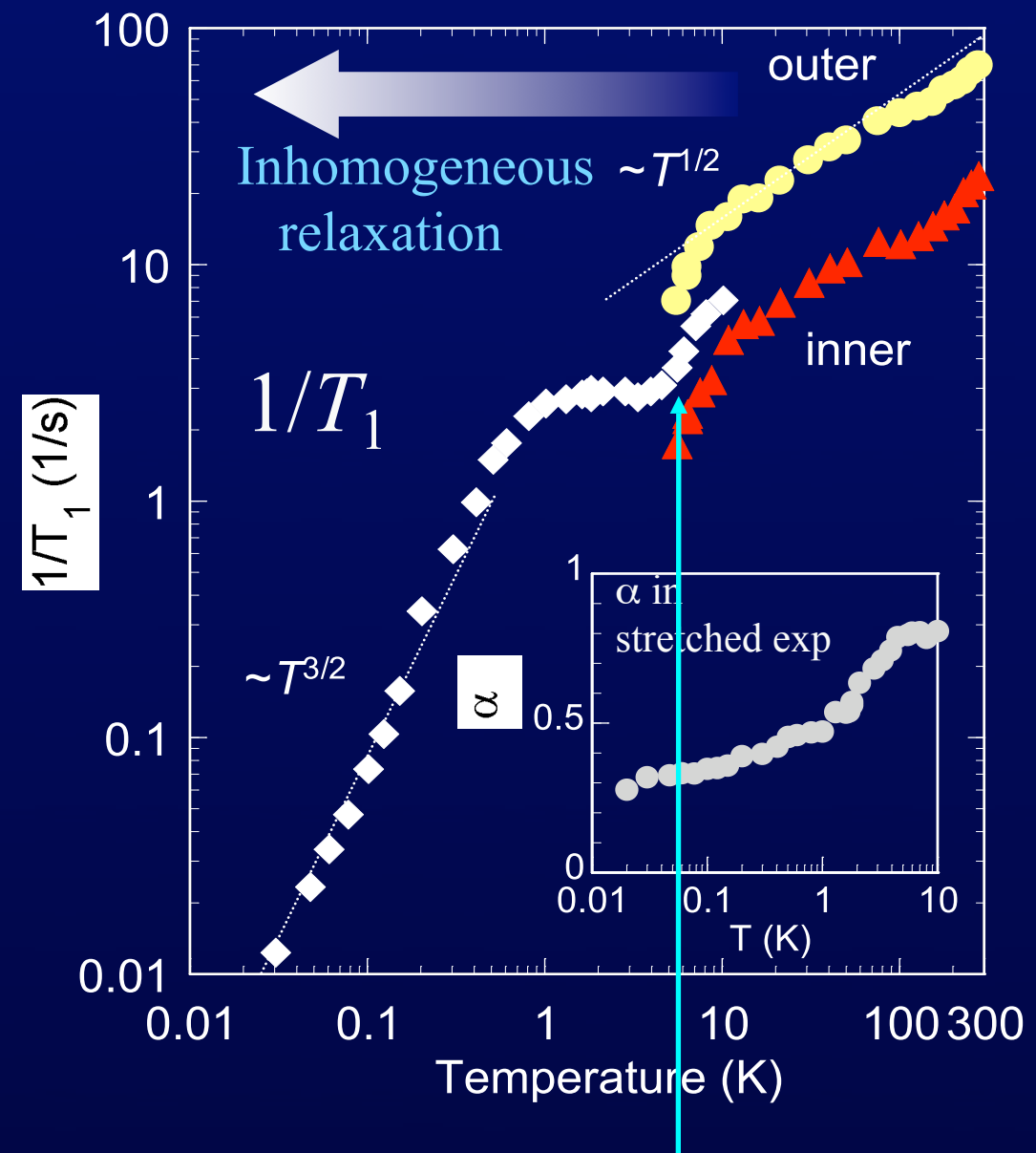
^{13}C NMR relaxation rate

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

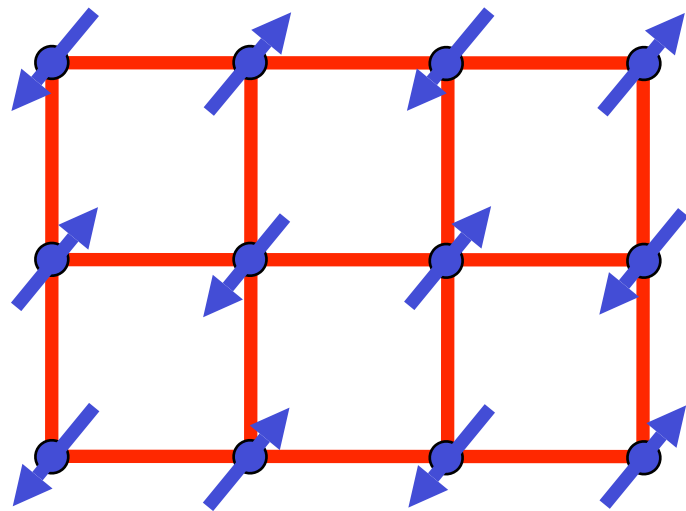
The quantum-critical region of magnetic ordering on the triangular lattice is described by the $O(4)$ model and has

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

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 .)

Global phase diagram



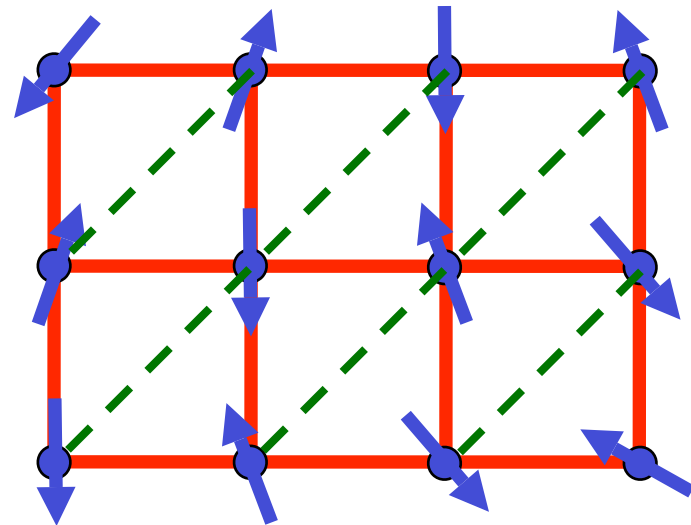
$$\langle z_\alpha \rangle \neq 0, \langle v \rangle \neq 0$$

Néel state

Spin liquid with a
“photon” collective mode

[Unstable to valence bond solid (VBS) order]

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

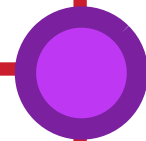


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non-collinear Néel state

Z_2 spin liquid with a
vison excitation

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



s

$\tilde{s}$

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

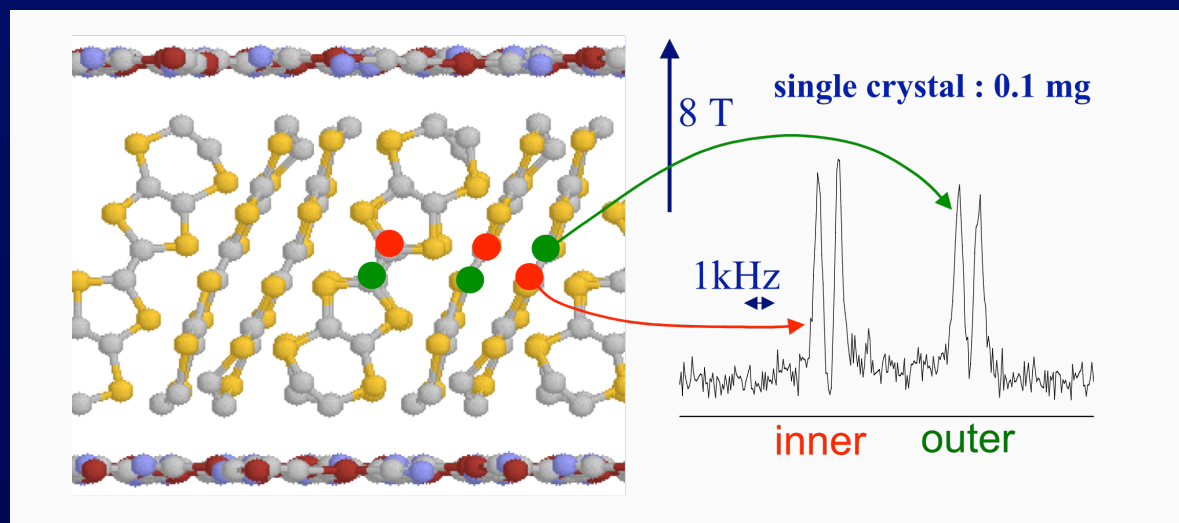
$$\frac{1}{T_1} \sim T^{\eta_{\text{cs}}}$$

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

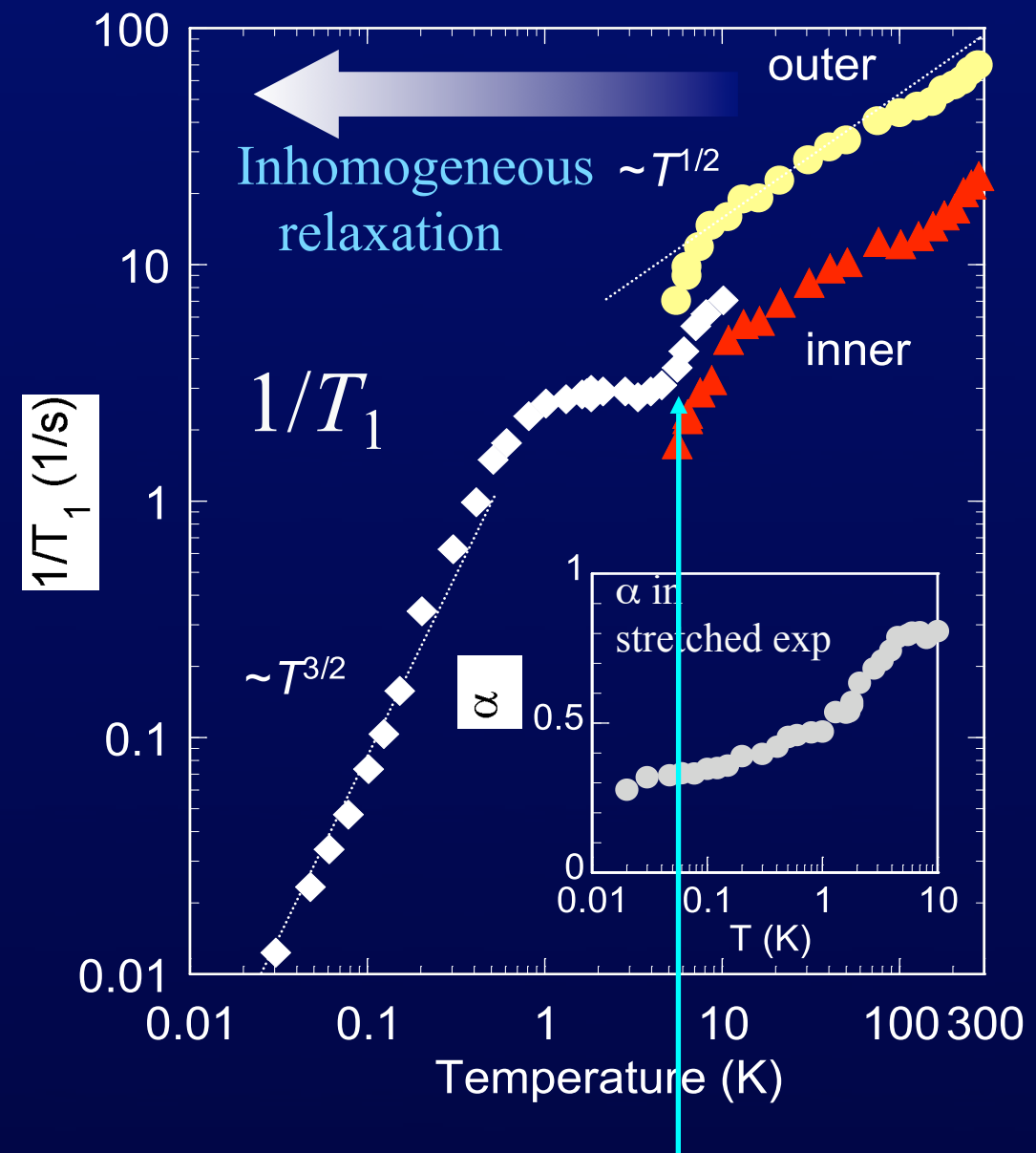
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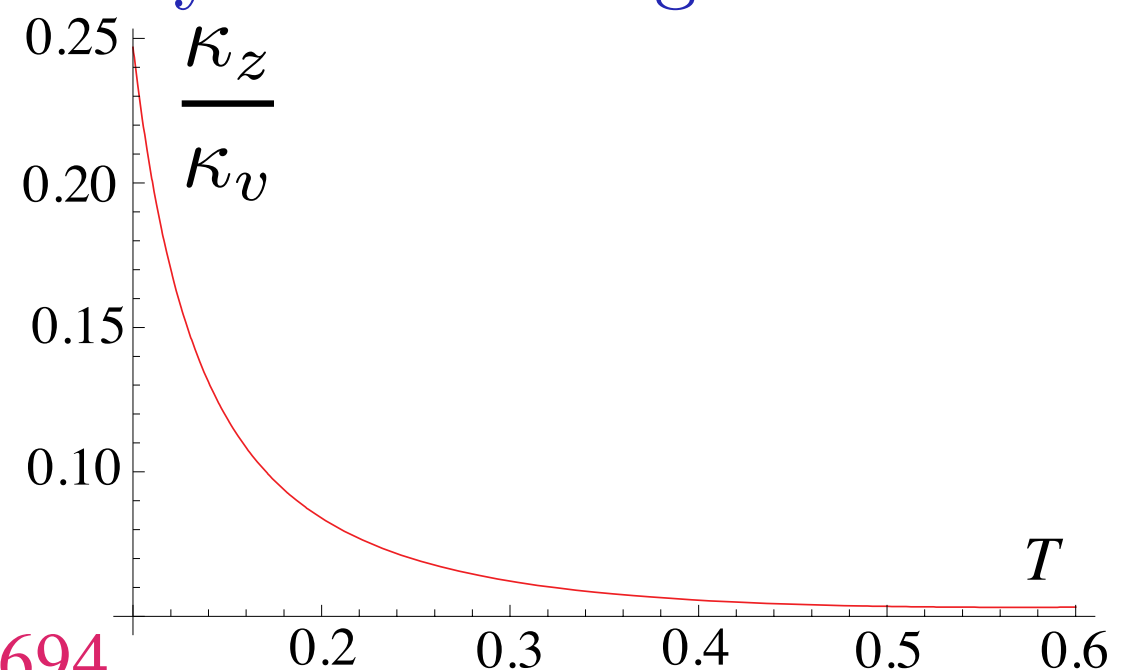


Anomaly at 5-6 K

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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.



Outline

1. Collective excitations of spin liquids in two dimensions

Photons and visons

2. Detecting the vison

Thermal conductivity of $\kappa\text{-(ET)}_2\text{Cu}_2(\text{CN})_3$

3. Detecting the photon

Valence bond solid order around Zn impurities

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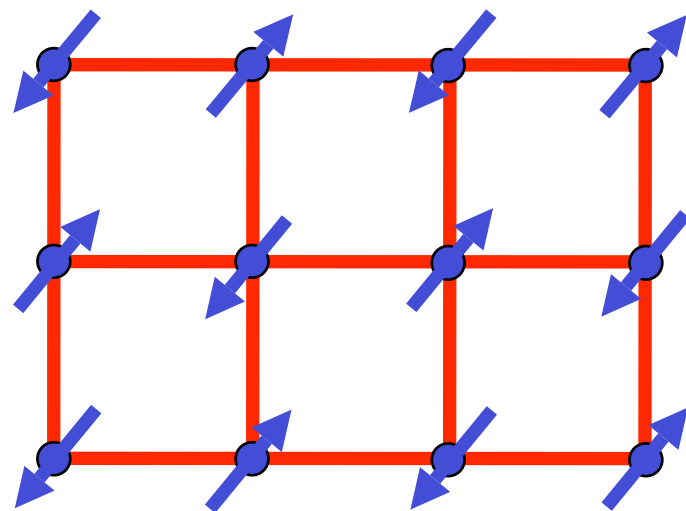
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s_c

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s

Non-perturbative effects in U(1) spin liquid

N. Read and S. Sachdev, *Phys. Rev. Lett.* **62**, 1694 (1990)

T. Senthil, A. Vishwanath, L. Balents, S. Sachdev and M.P.A. Fisher, *Science* **303**, 1490 (2004).

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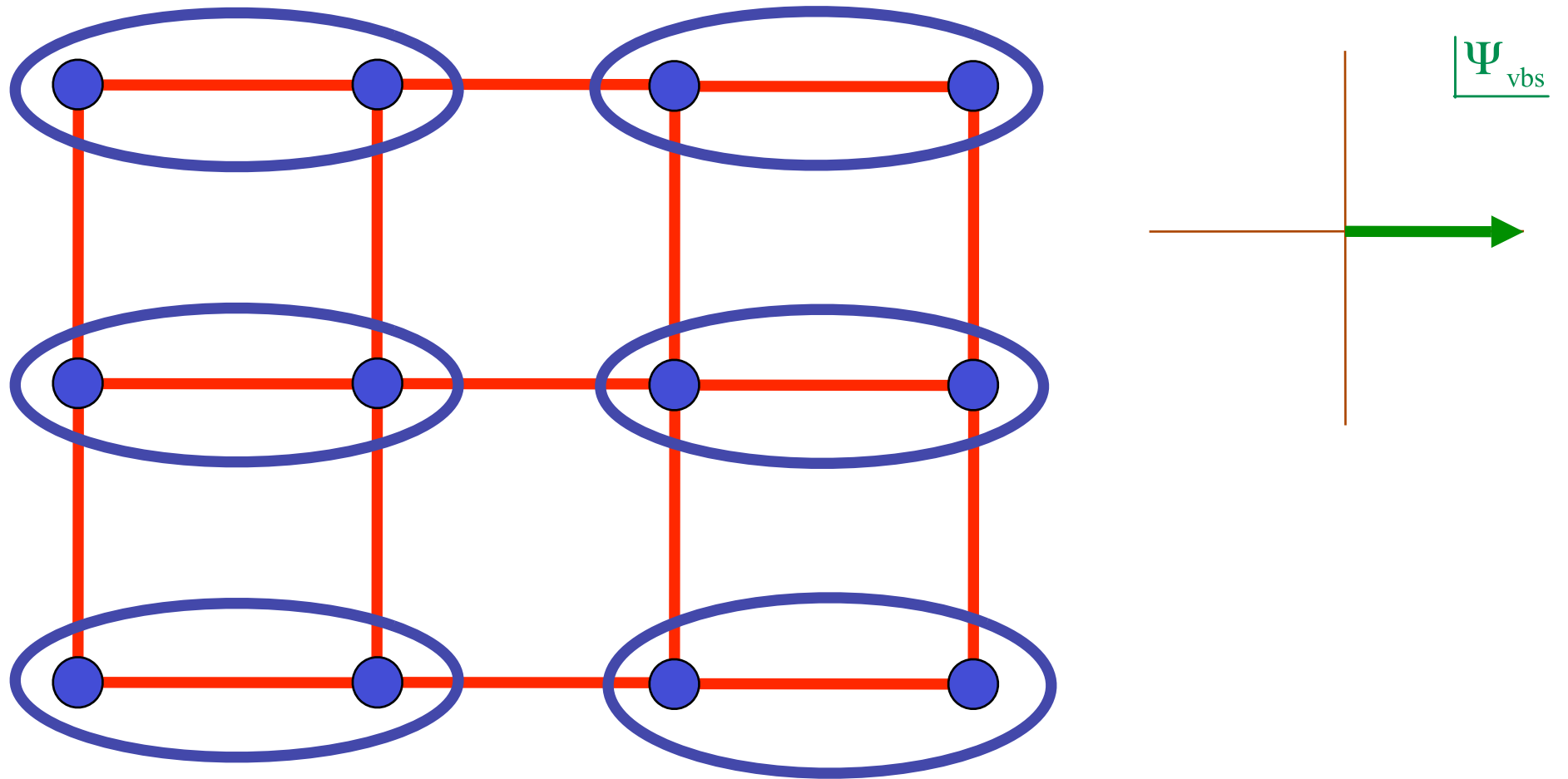
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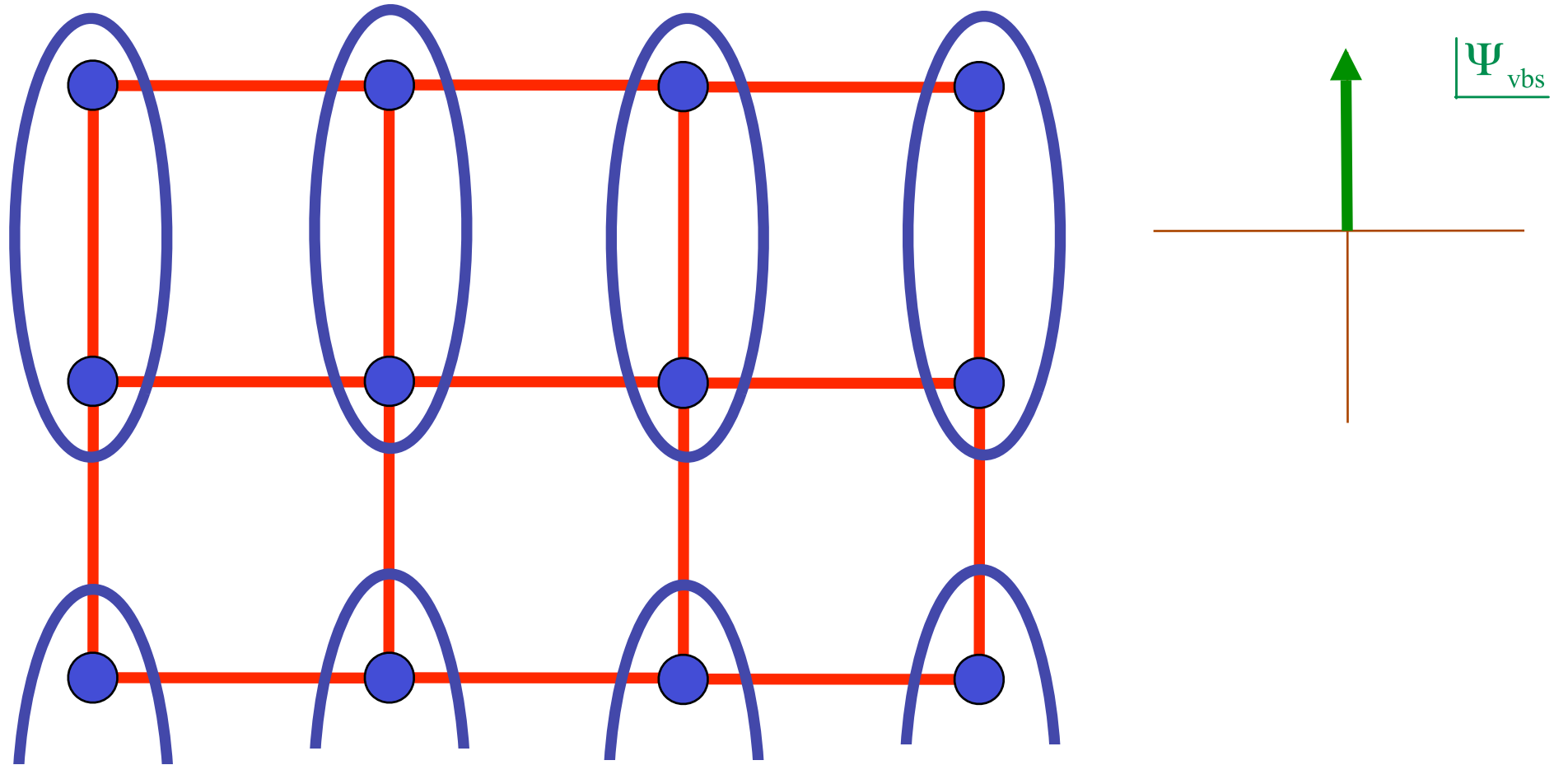
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Order parameter of VBS state



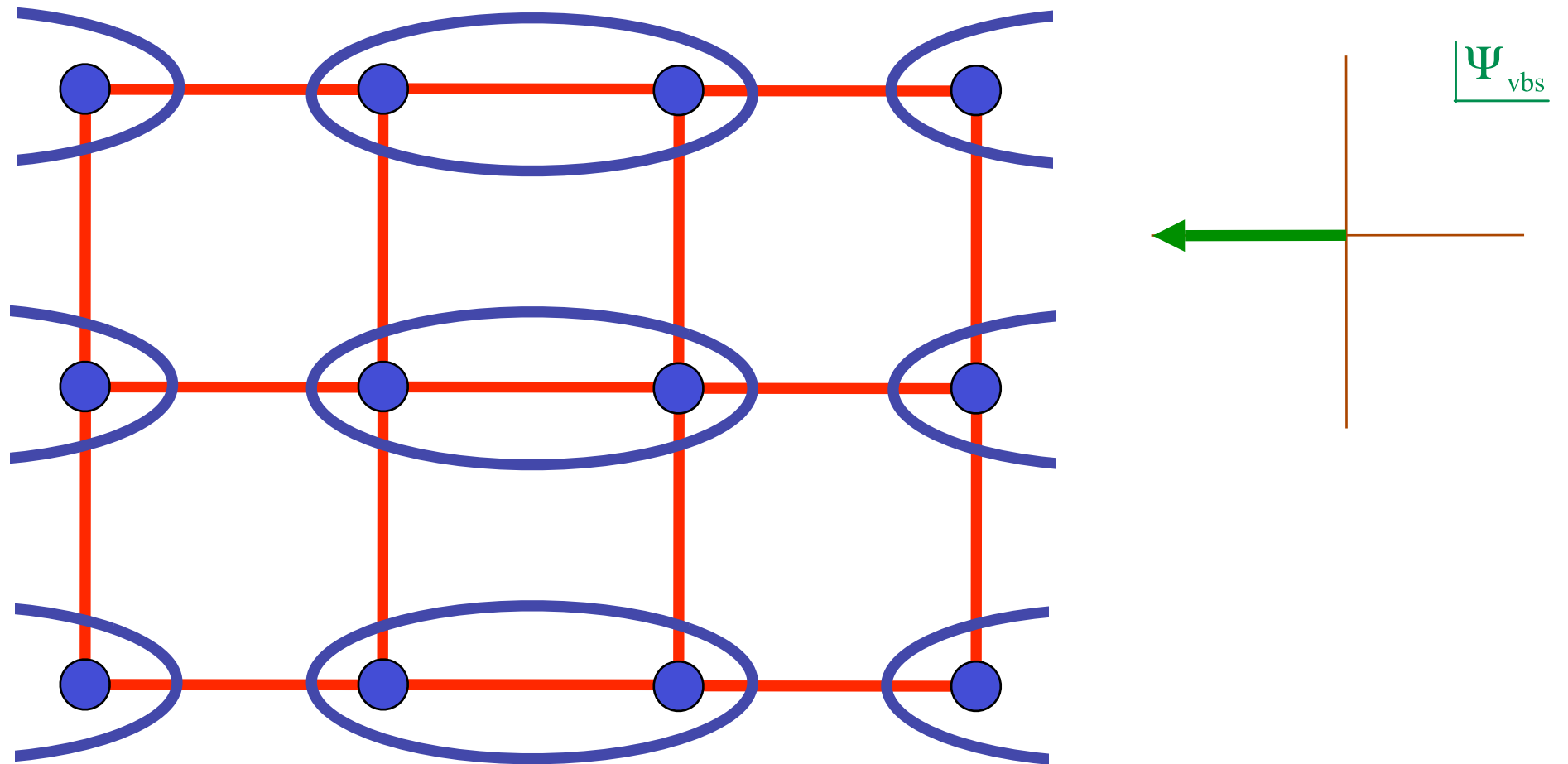
$$\Psi_{\text{vbs}}(i) = \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j e^{i \arctan(\mathbf{r}_j - \mathbf{r}_i)}$$

Order parameter of VBS state



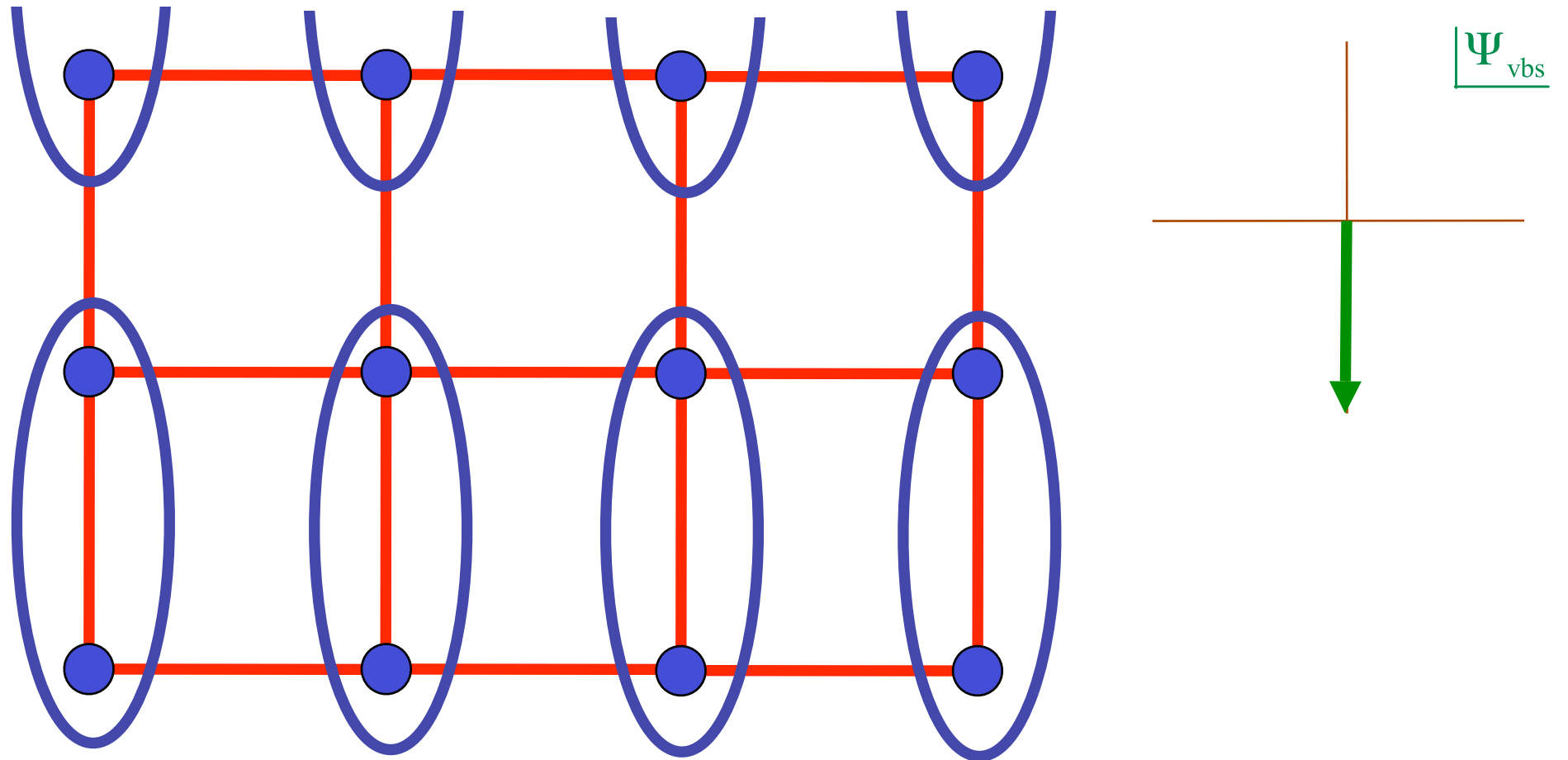
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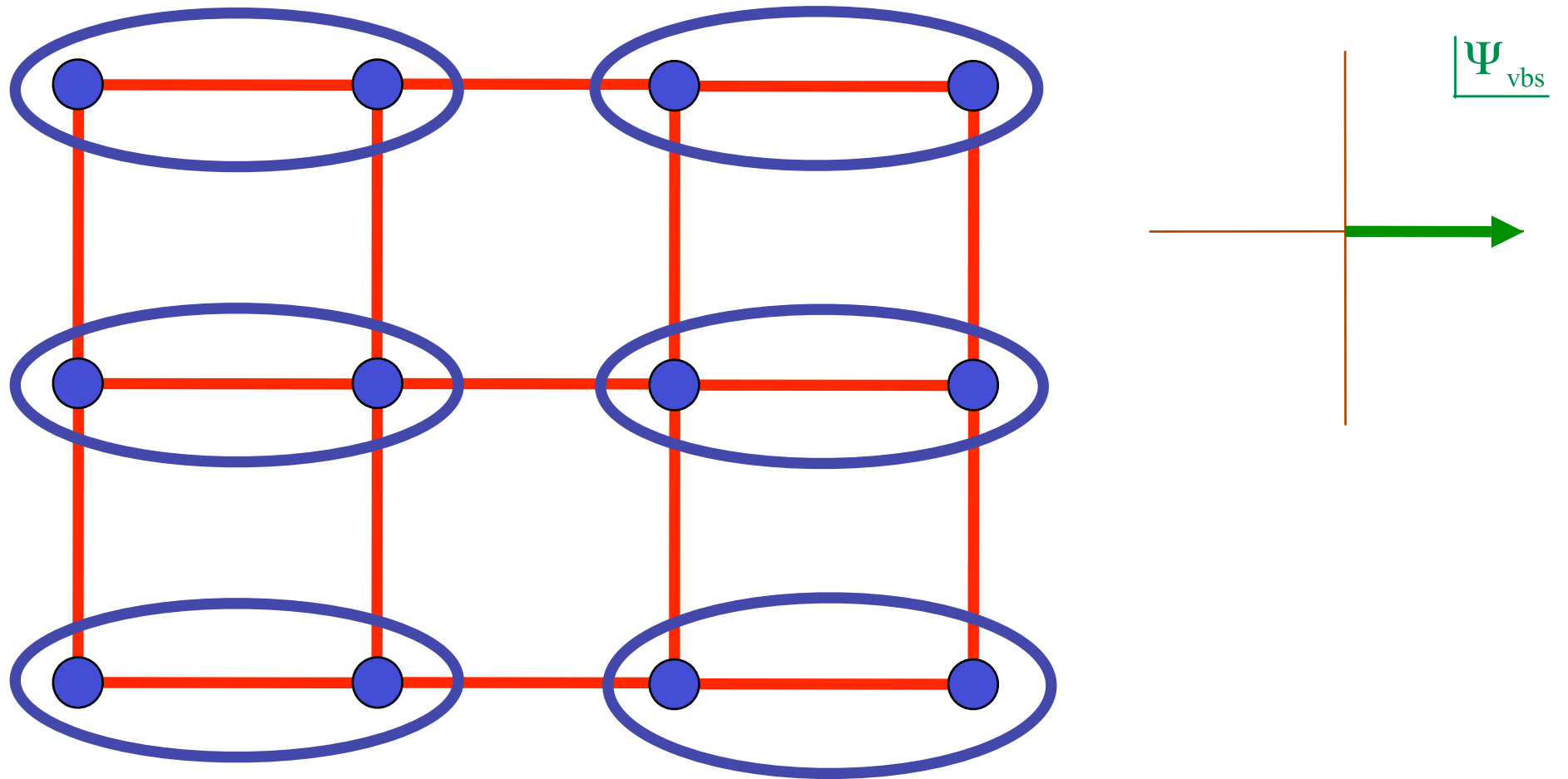
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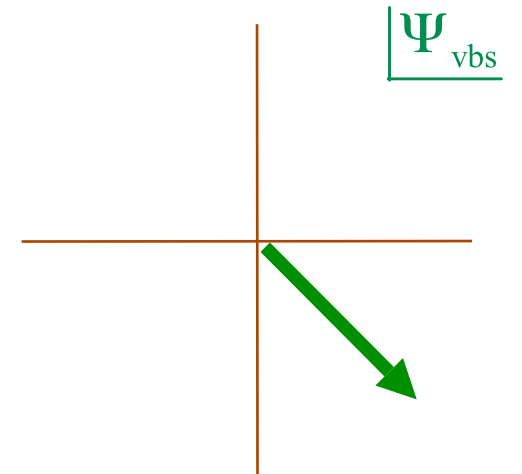
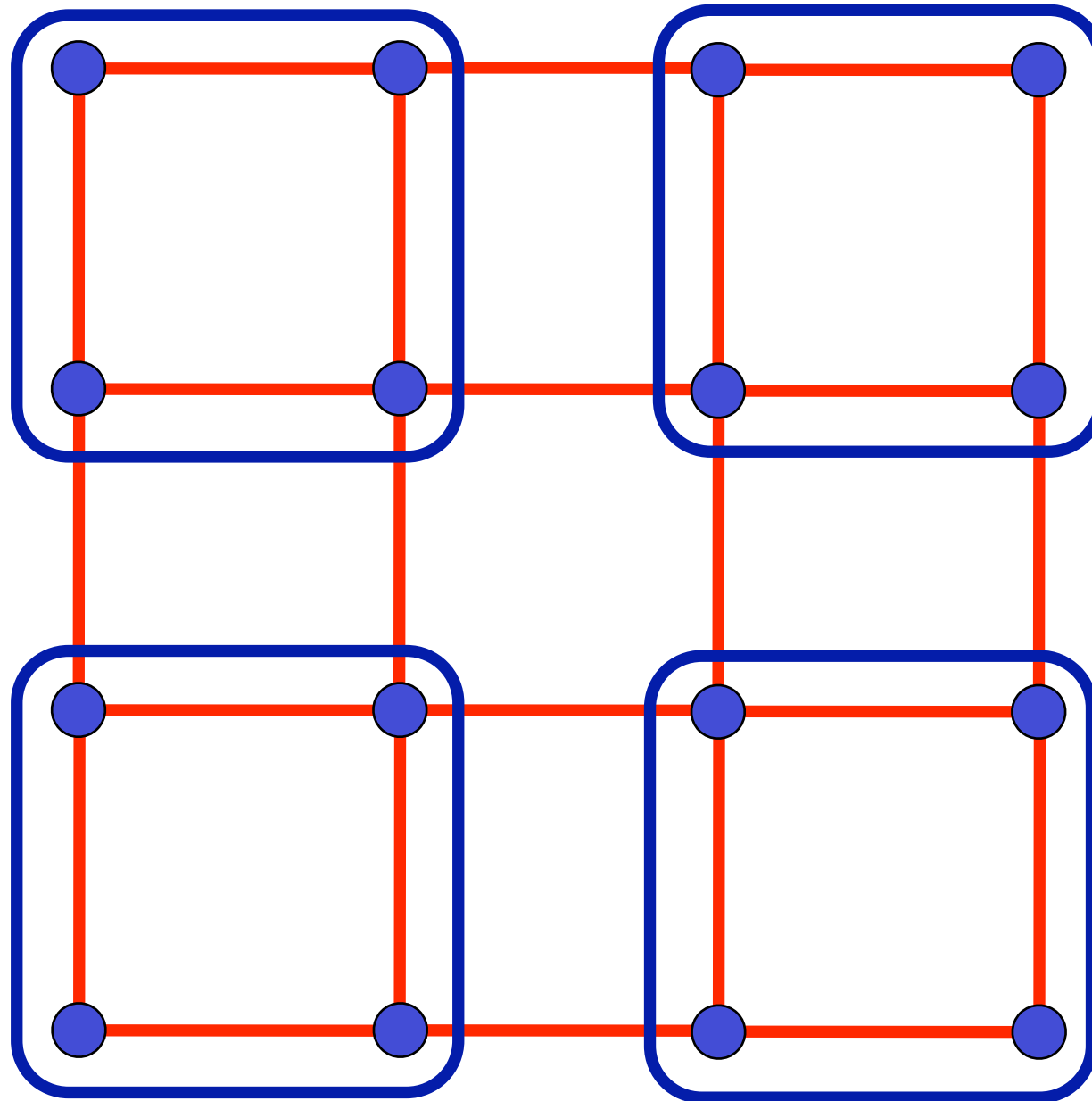
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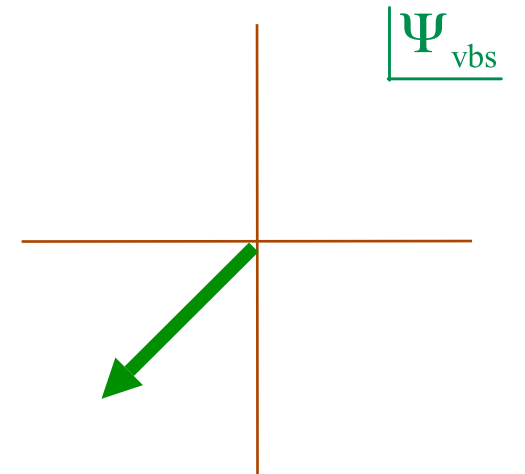
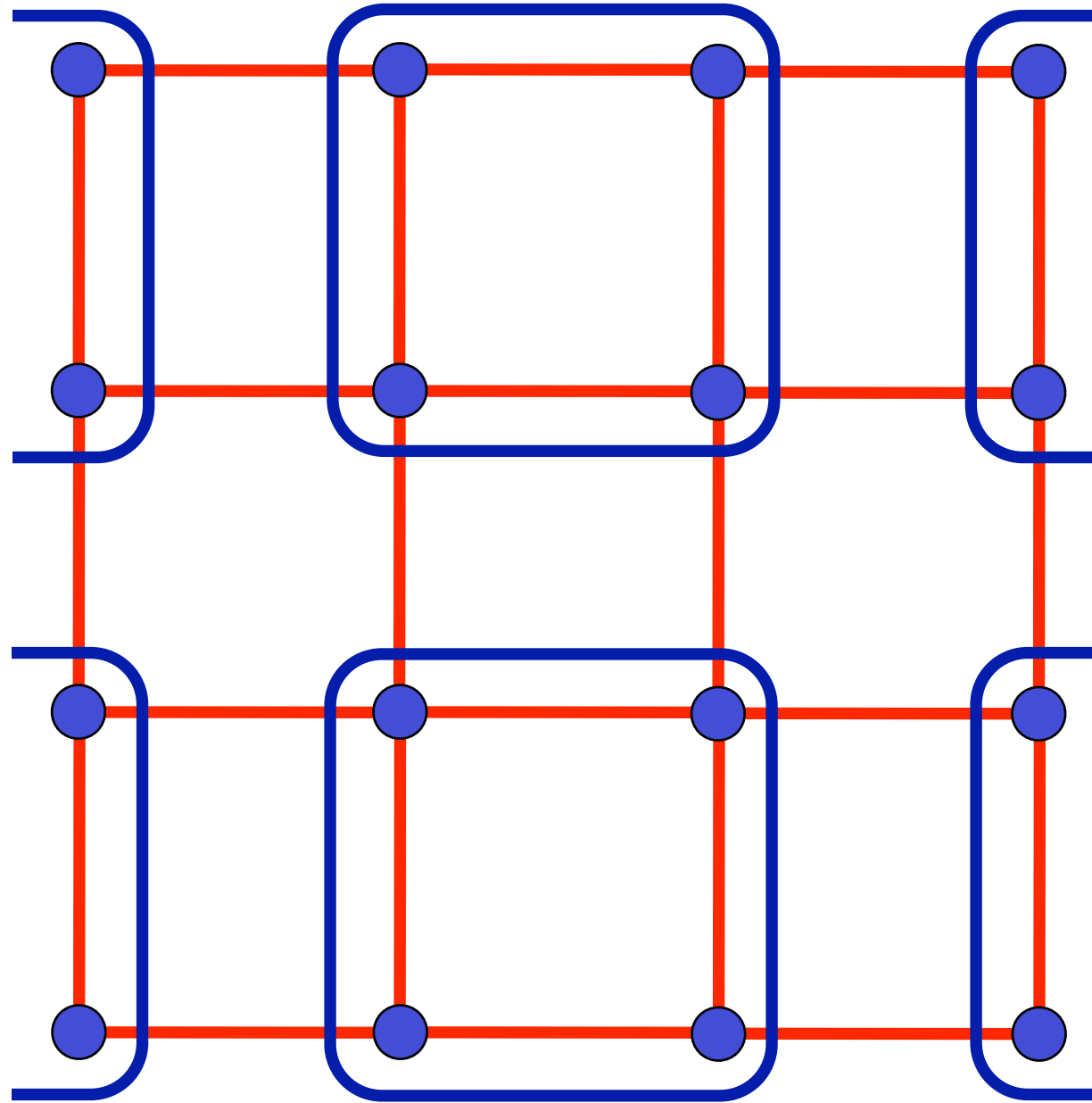
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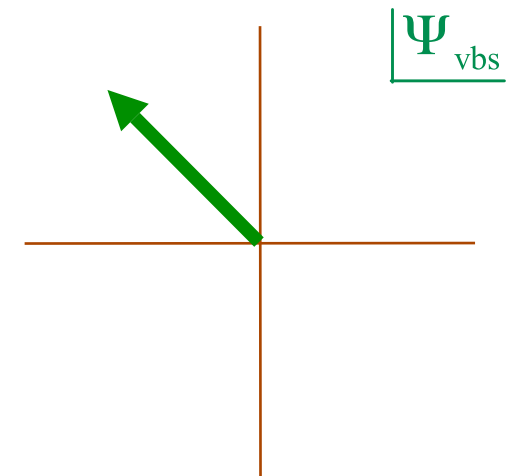
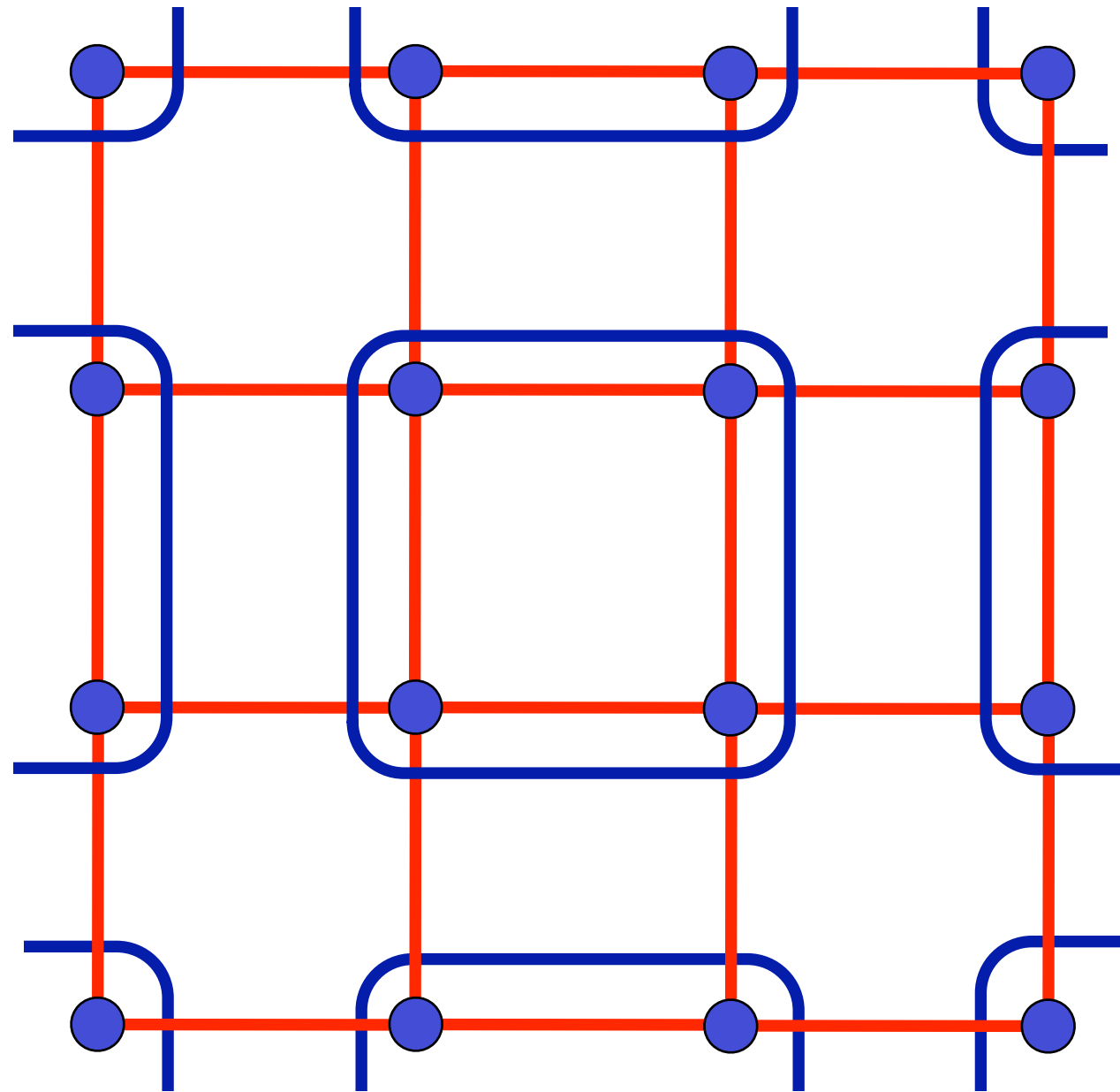
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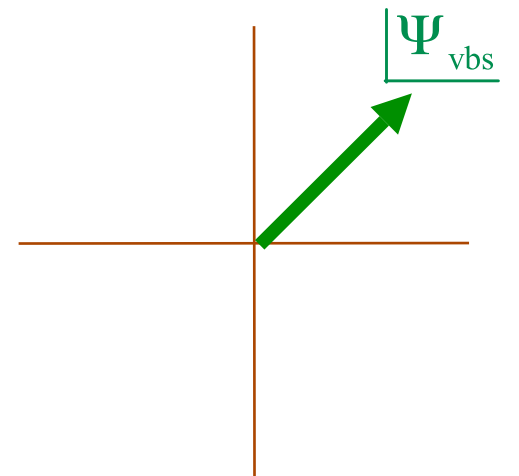
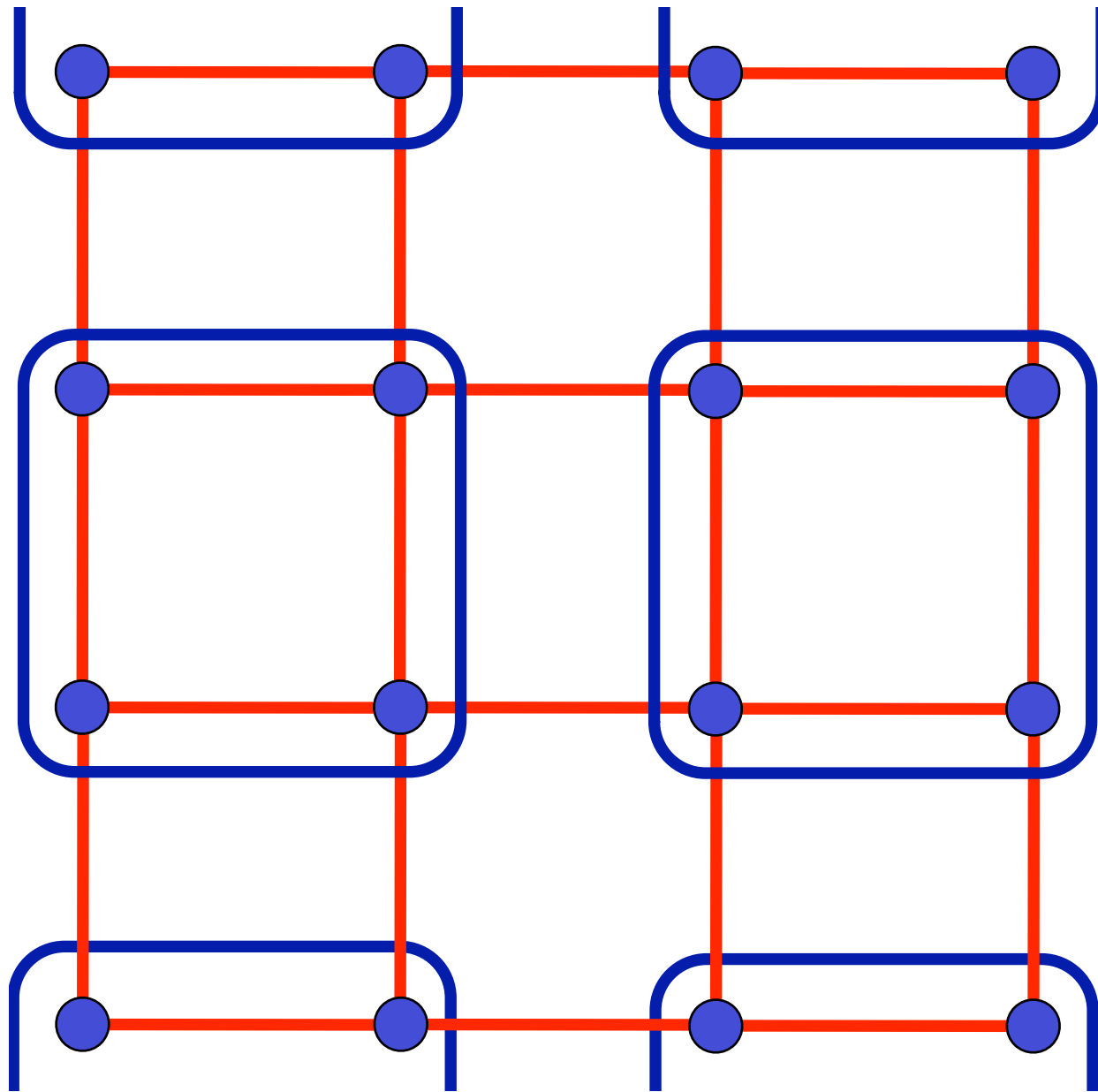
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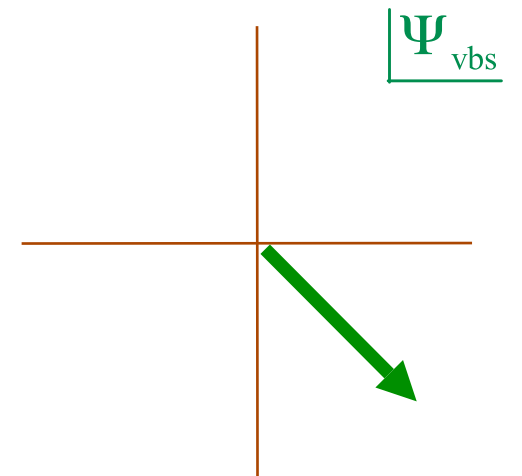
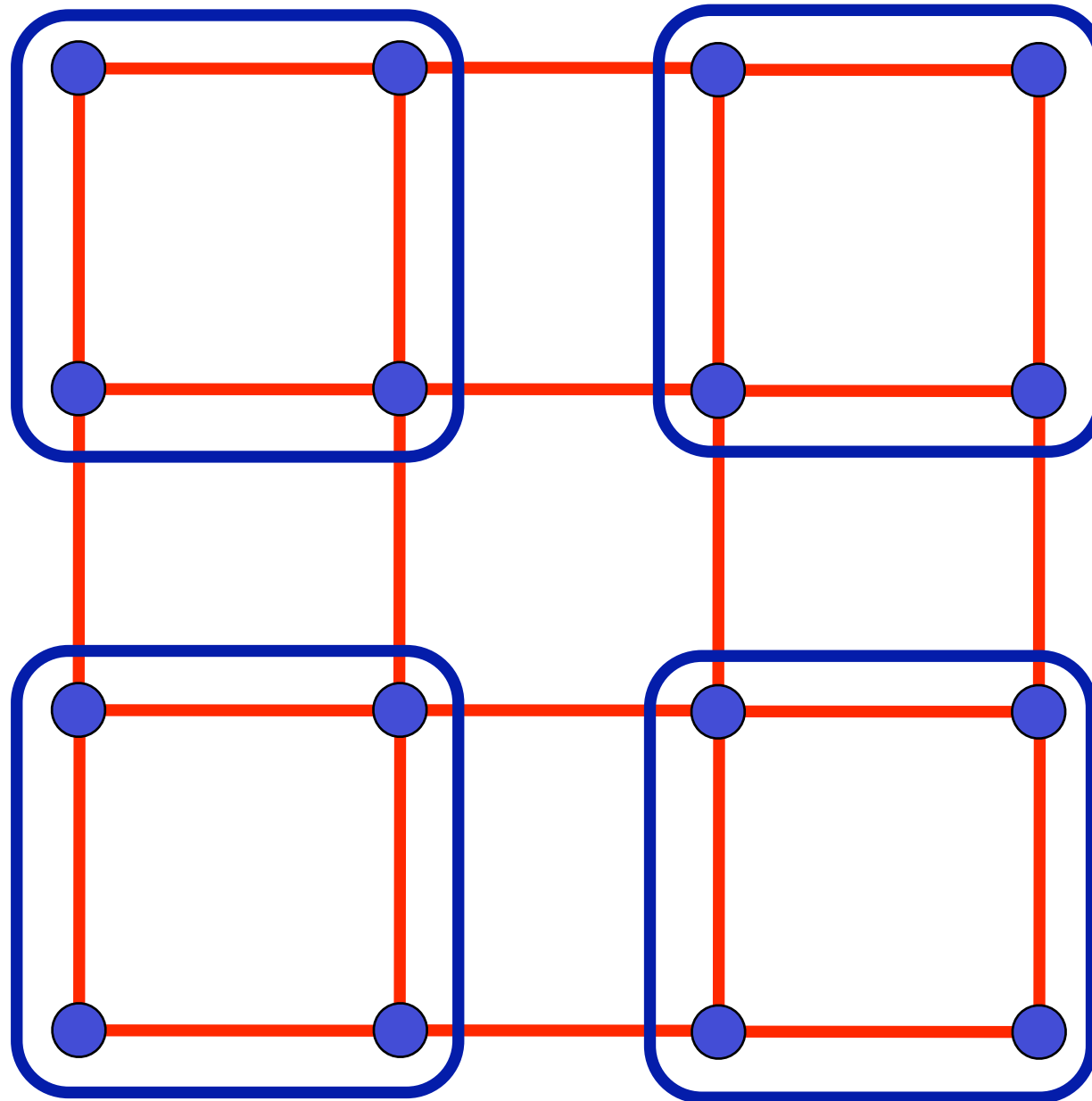
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Non-perturbative effects in U(1) spin liquid

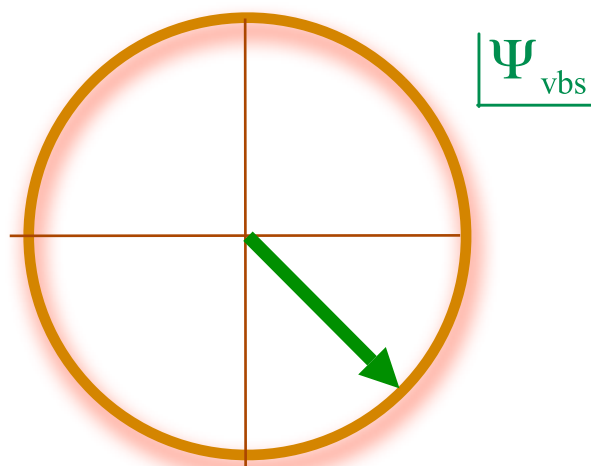
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- The (nearly) gapless photon is the Goldstone mode associated with the emergent circular symmetry



$$\Psi_{\text{vbs}} \rightarrow \Psi_{\text{vbs}} e^{i\theta}.$$

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$$\mathcal{H}_{\text{SU}(2)} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - Q \sum_{\langle ijkl \rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} \right) \left(\mathbf{S}_k \cdot \mathbf{S}_l - \frac{1}{4} \right)$$

Quantum Monte Carlo simulations display convincing evidence for a transition from a

Neel state at small Q
to a
VBS state at large Q

A.W. Sandvik, *Phys. Rev. Lett.* **98**, 2272020 (2007).

R.G. Melko and R.K. Kaul, *Phys. Rev. Lett.* **100**, 017203 (2008).

F.-J. Jiang, M. Nyfeler, S. Chandrasekharan, and U.-J. Wiese, arXiv:0710.3926

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$|\text{Im}[\Psi_{\text{vbs}}]|$

Distribution of VBS
order Ψ_{vbs} at large Q

$\text{Re}[\Psi_{\text{vbs}}]$

*Emergent circular
symmetry is
evidence for $U(1)$
photon and
topological order*

Non-magnetic (Zn) impurity in the U(1) spin liquid

- The dominant perturbation of the impurity is a “Wilson line” in the time direction

$$\mathcal{S}_{\text{imp}} = i \int d\tau A_\tau(\mathbf{r} = 0, \tau)$$

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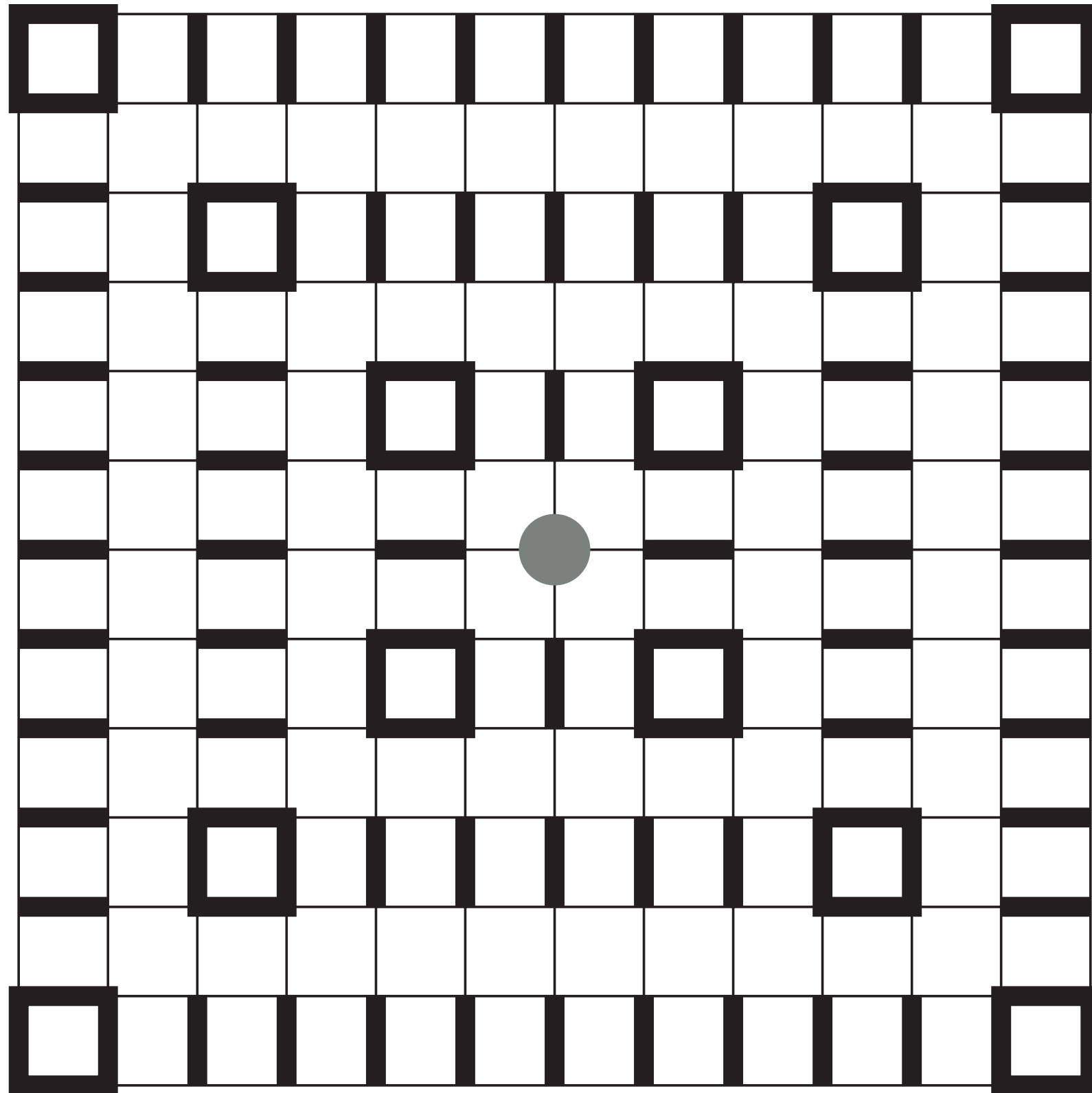
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- Upon approaching the impurity the VBS (monopole) operator has a vortex-like winding in its OPE:

$$\lim_{\mathbf{r} \rightarrow 0} \Psi_{\text{vbs}}(\mathbf{r}, \tau) \sim |\mathbf{r}|^\alpha e^{i\theta}$$

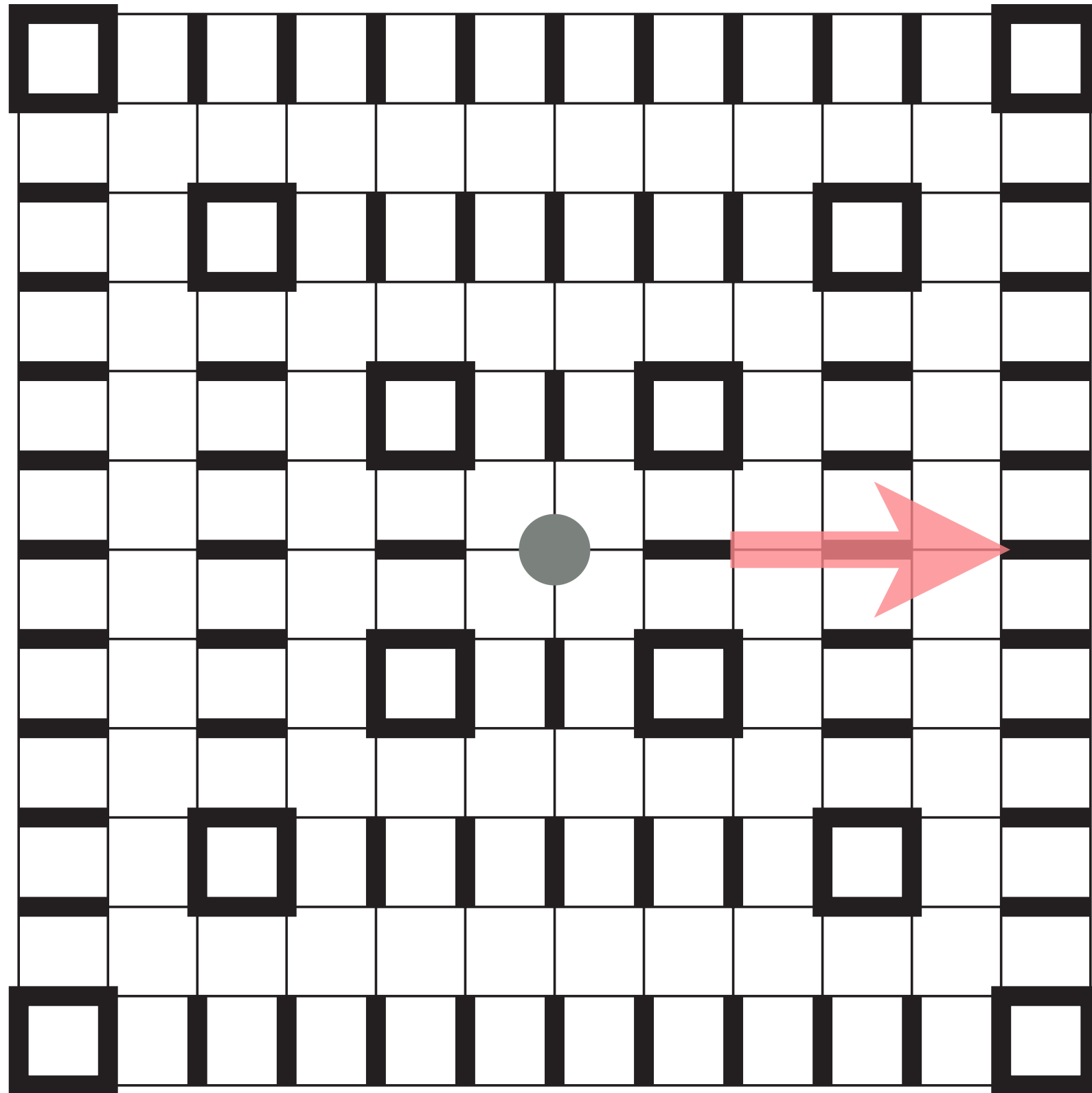
where $\theta = \arctan(y/x)$.

Schematic of VBS order around impurity



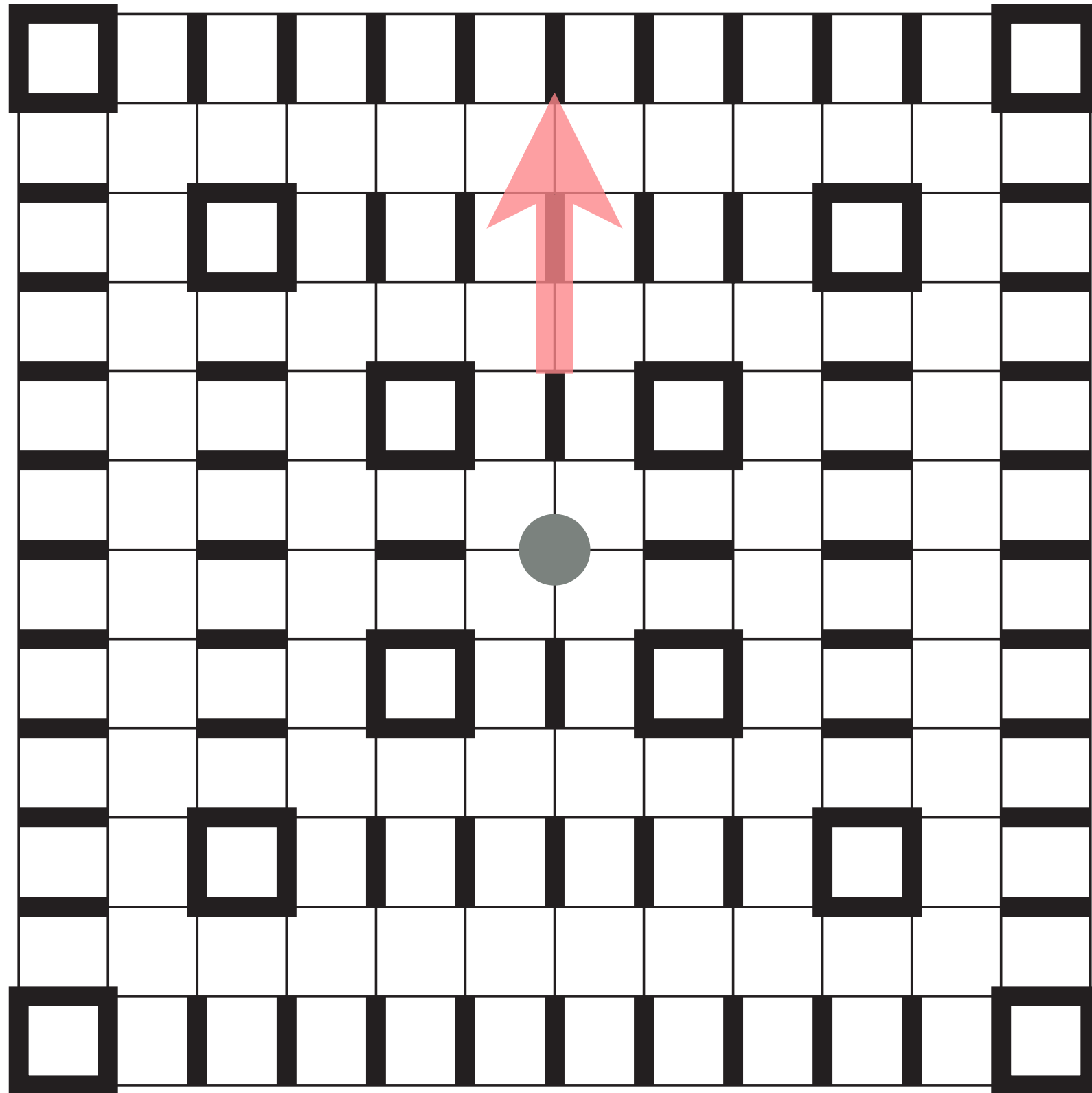
Bulk VBS order is columnar

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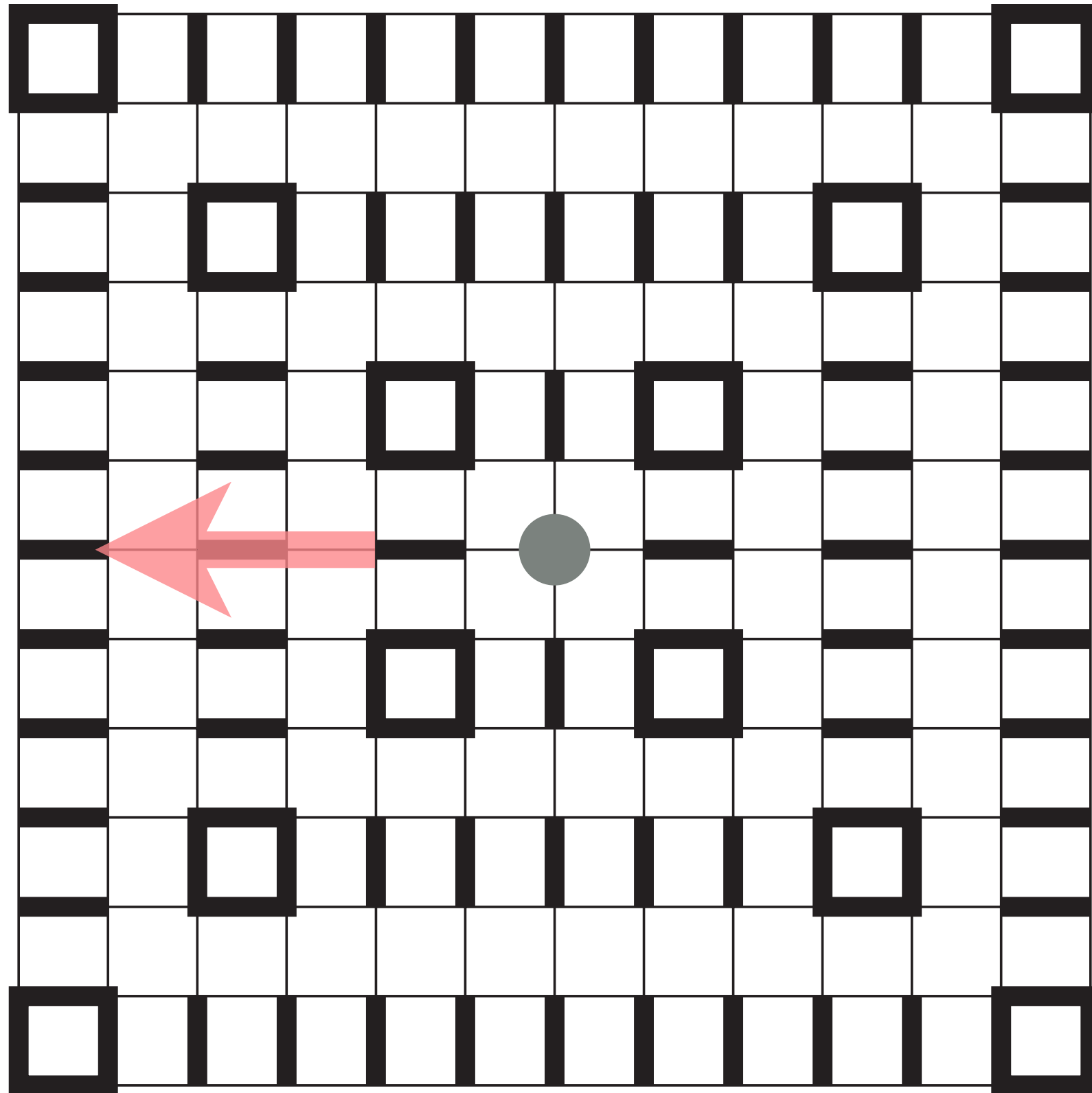
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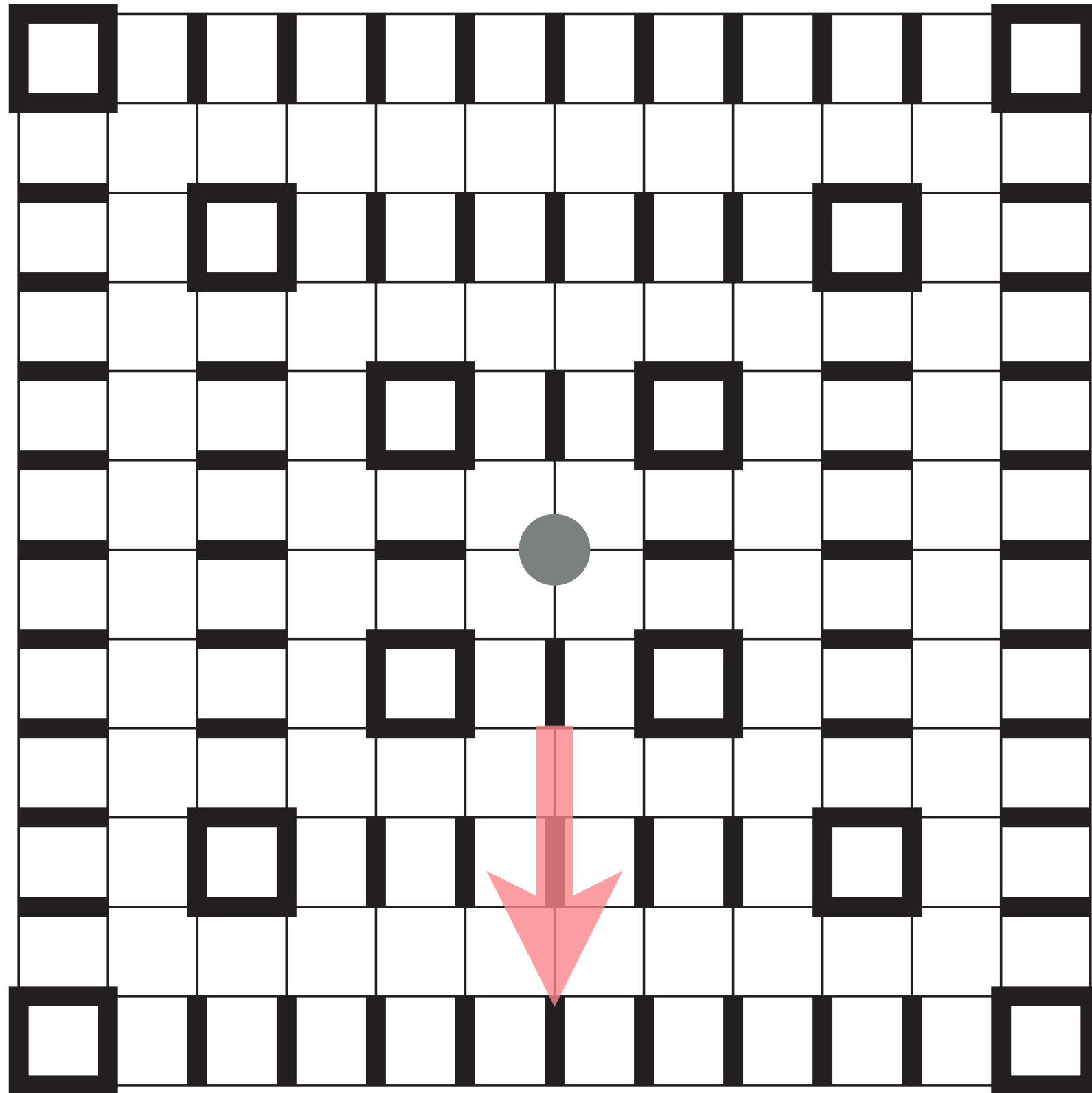
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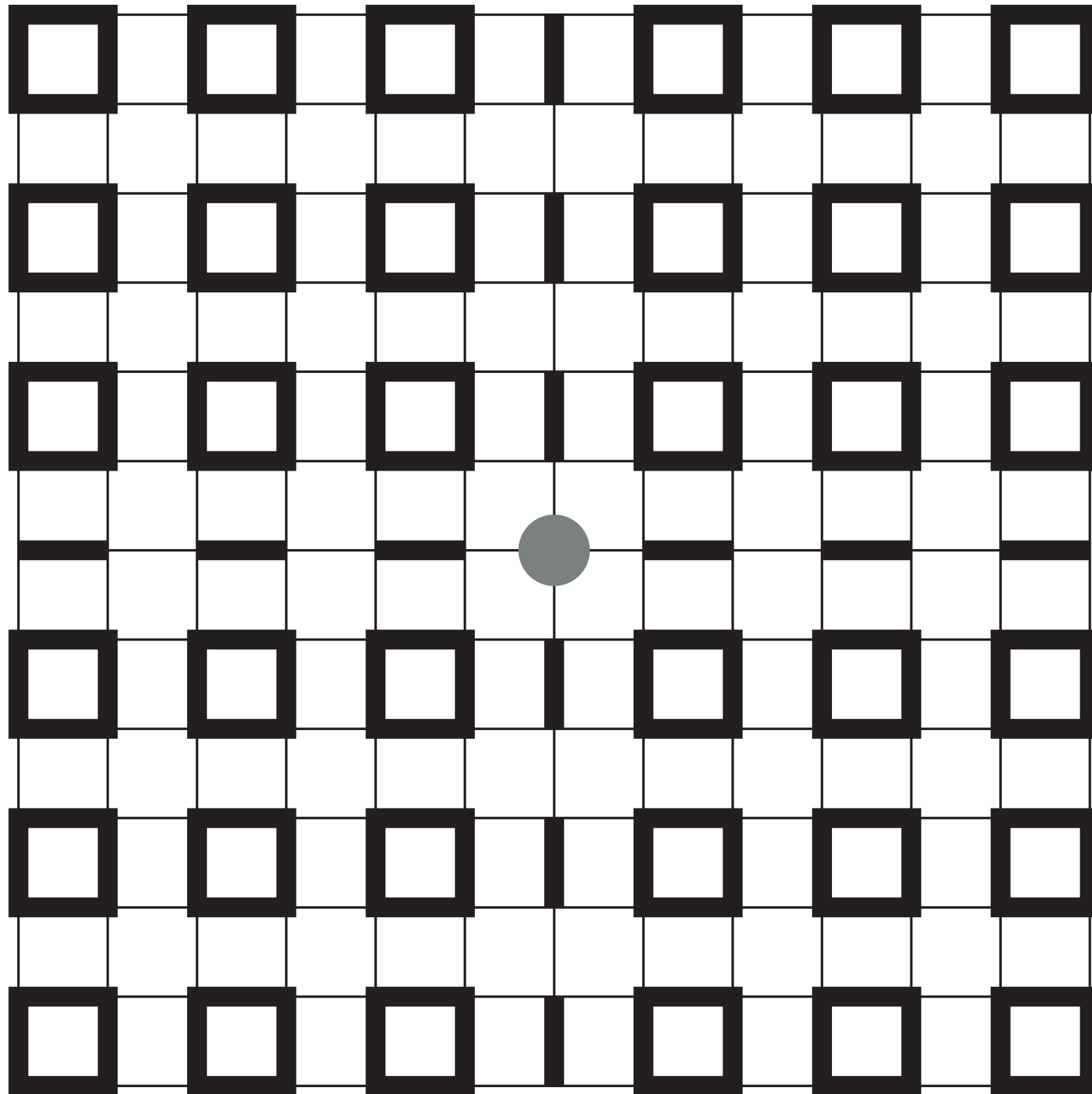
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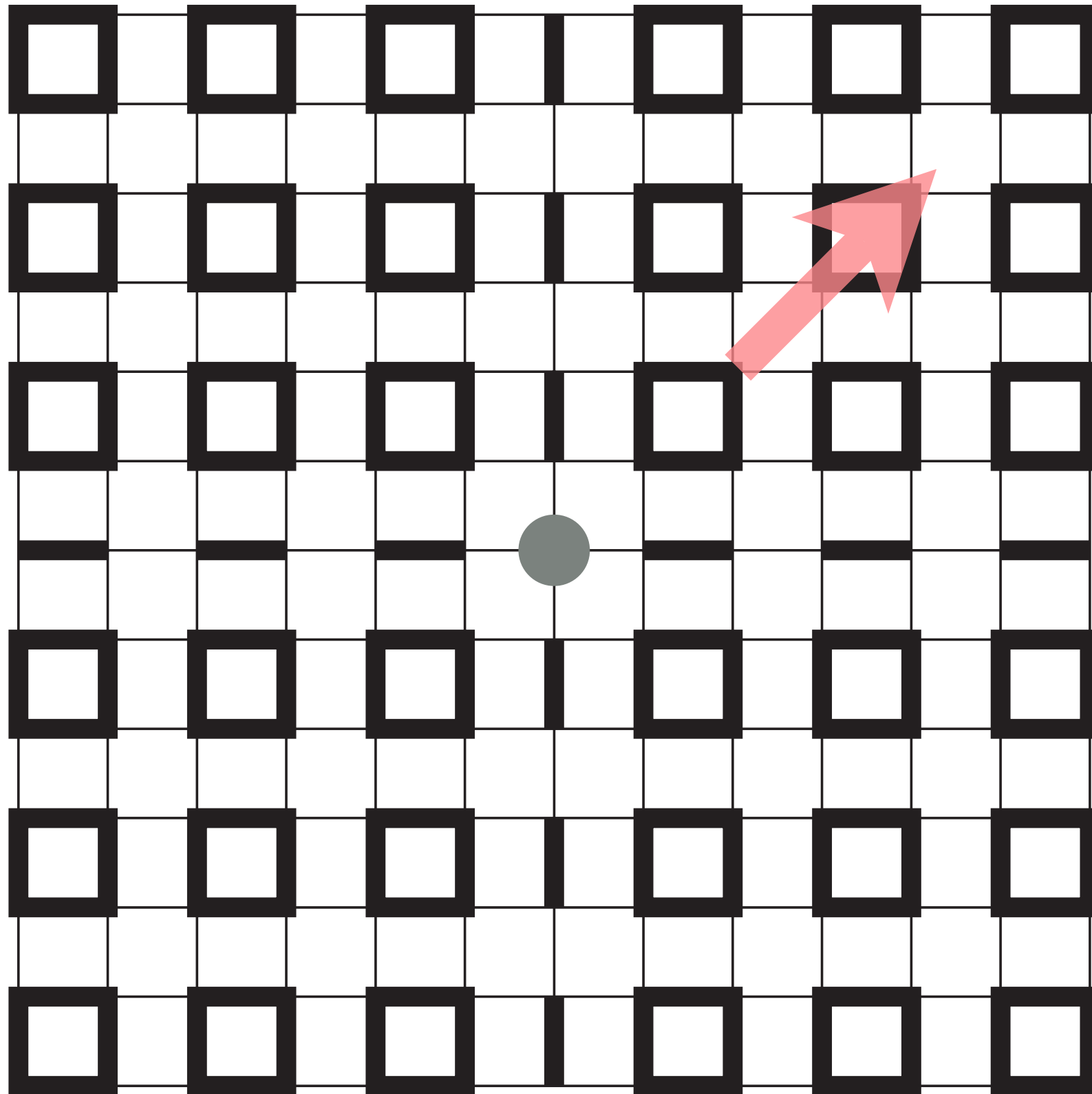
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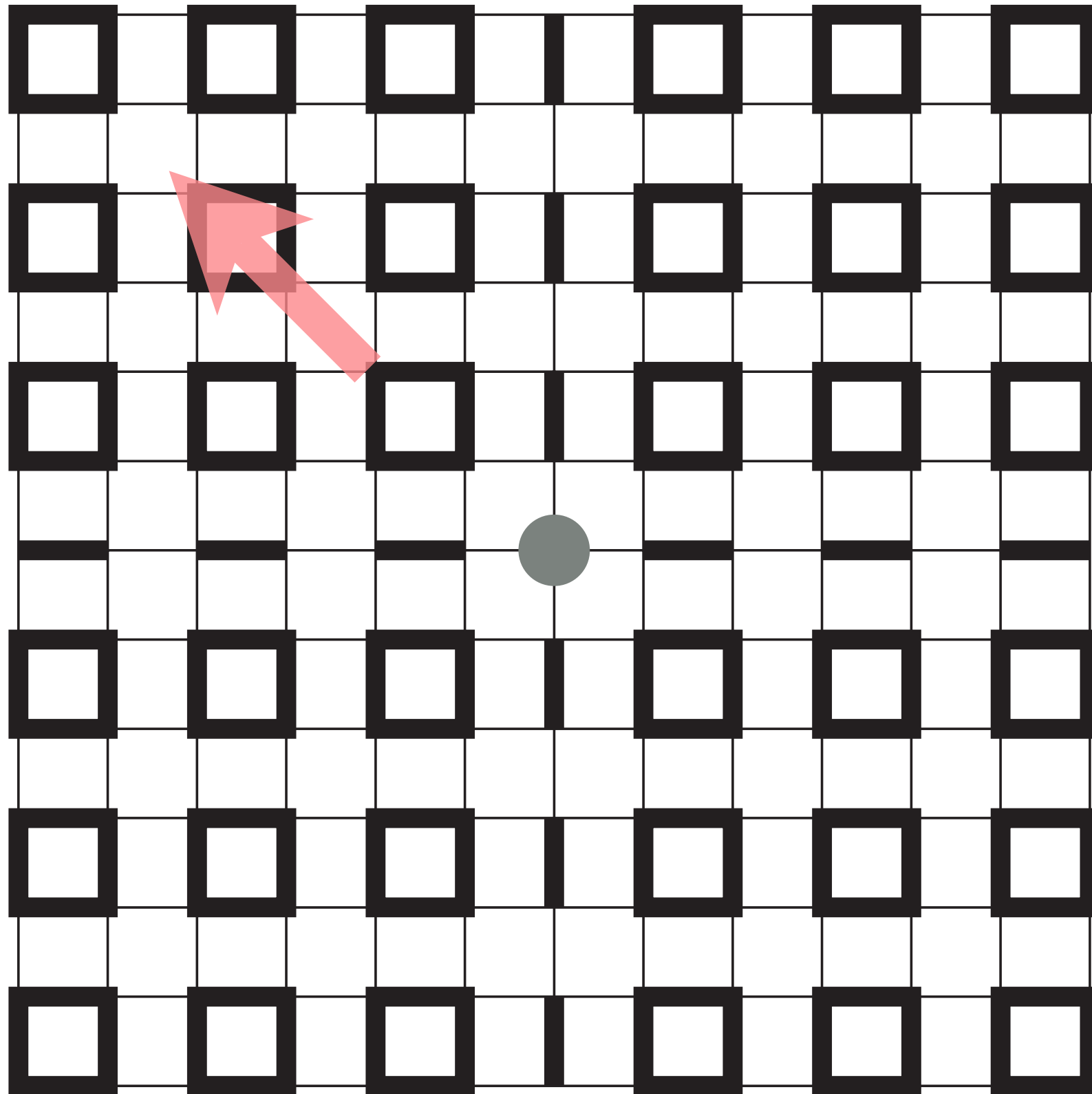
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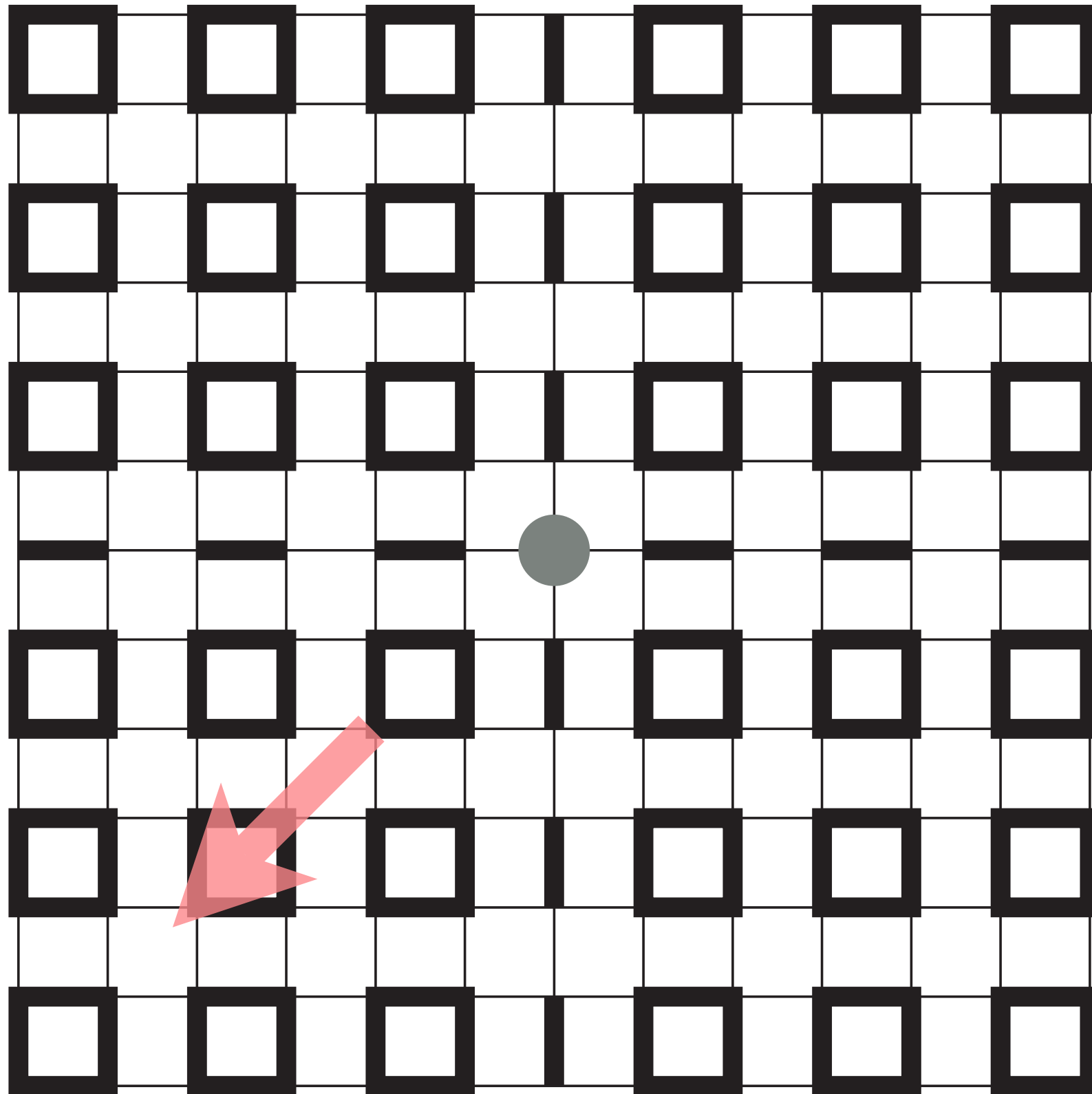
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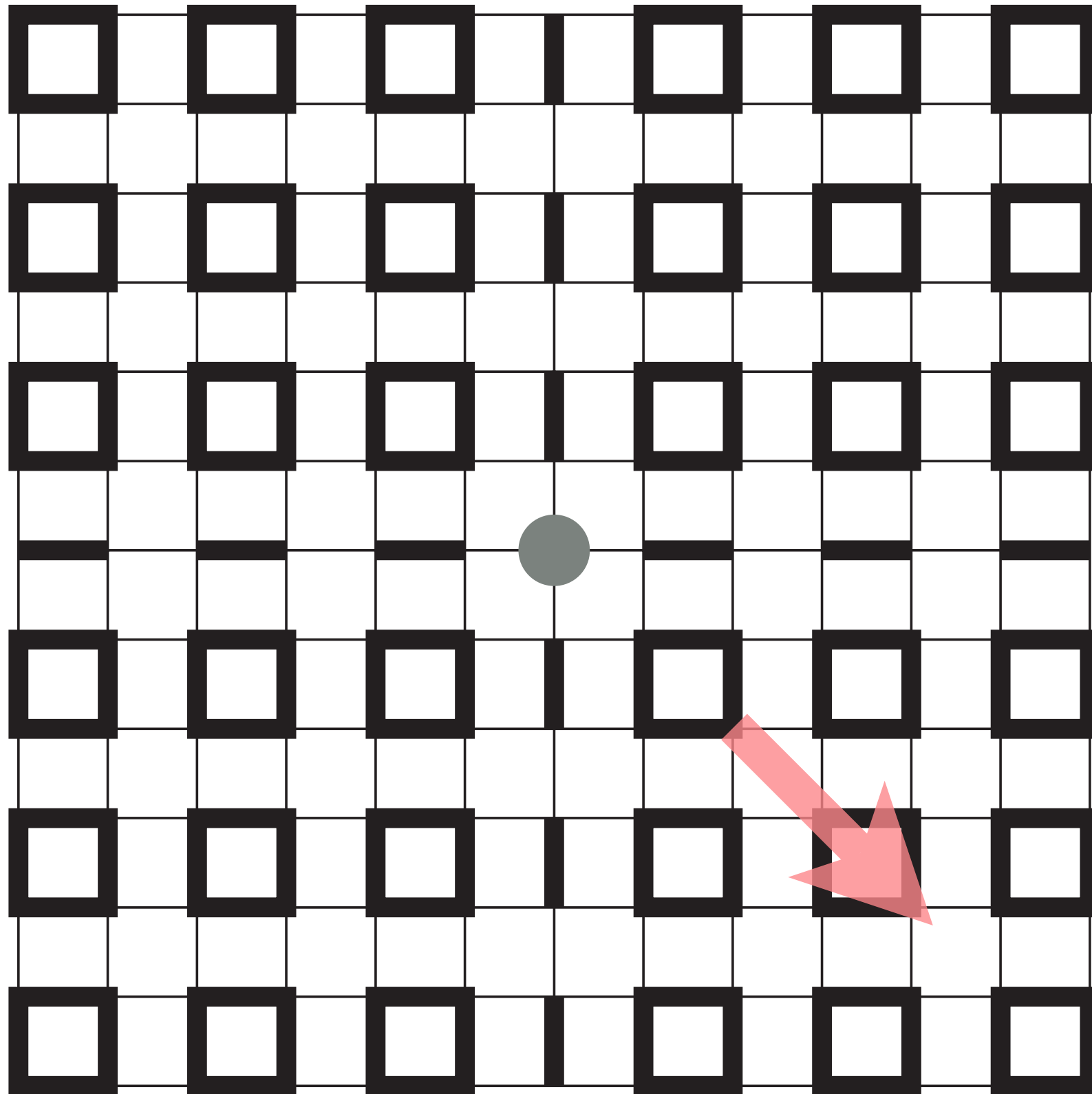
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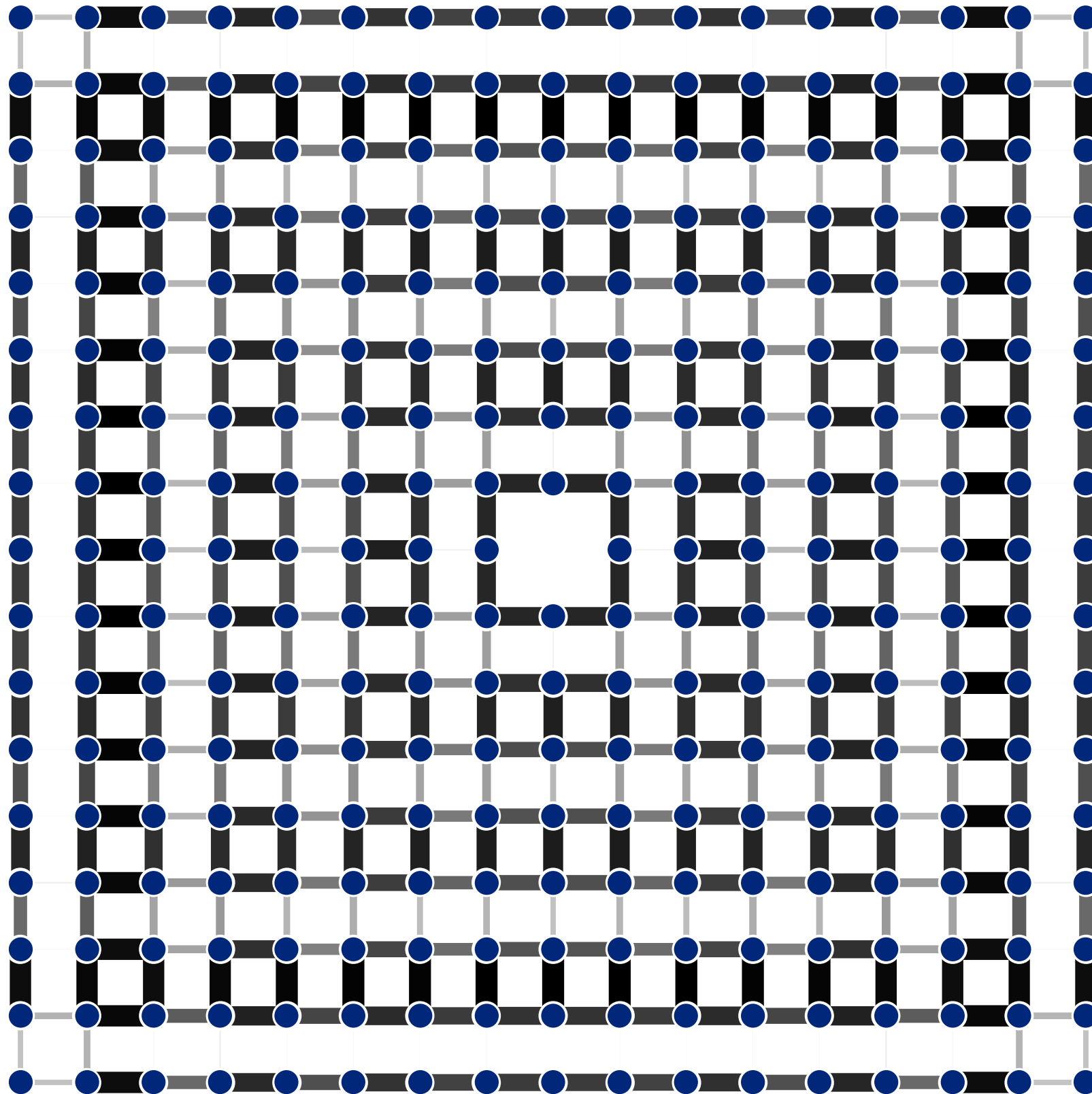
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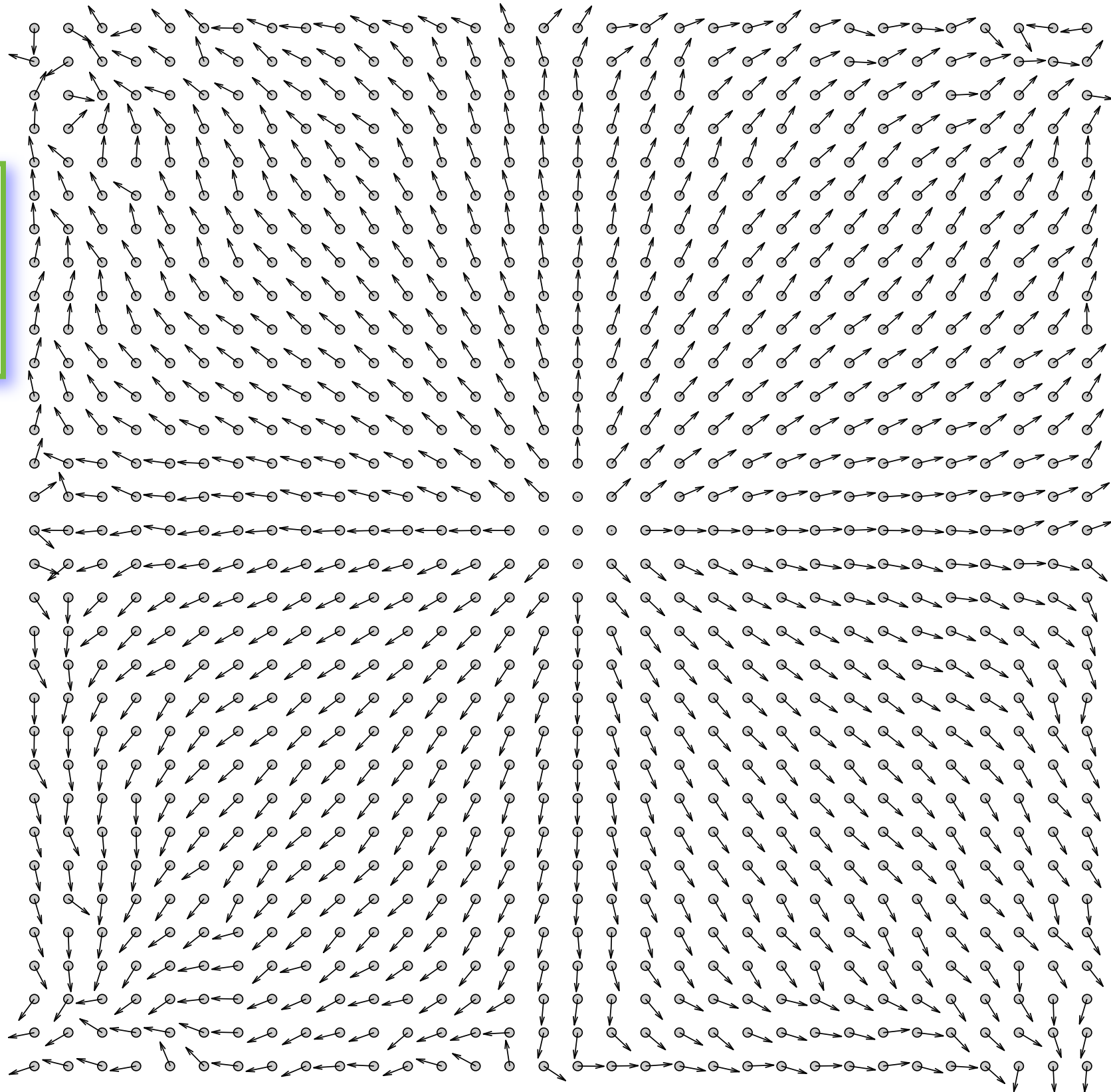
Bulk VBS order is plaquette

Bond order from QMC of J-Q model



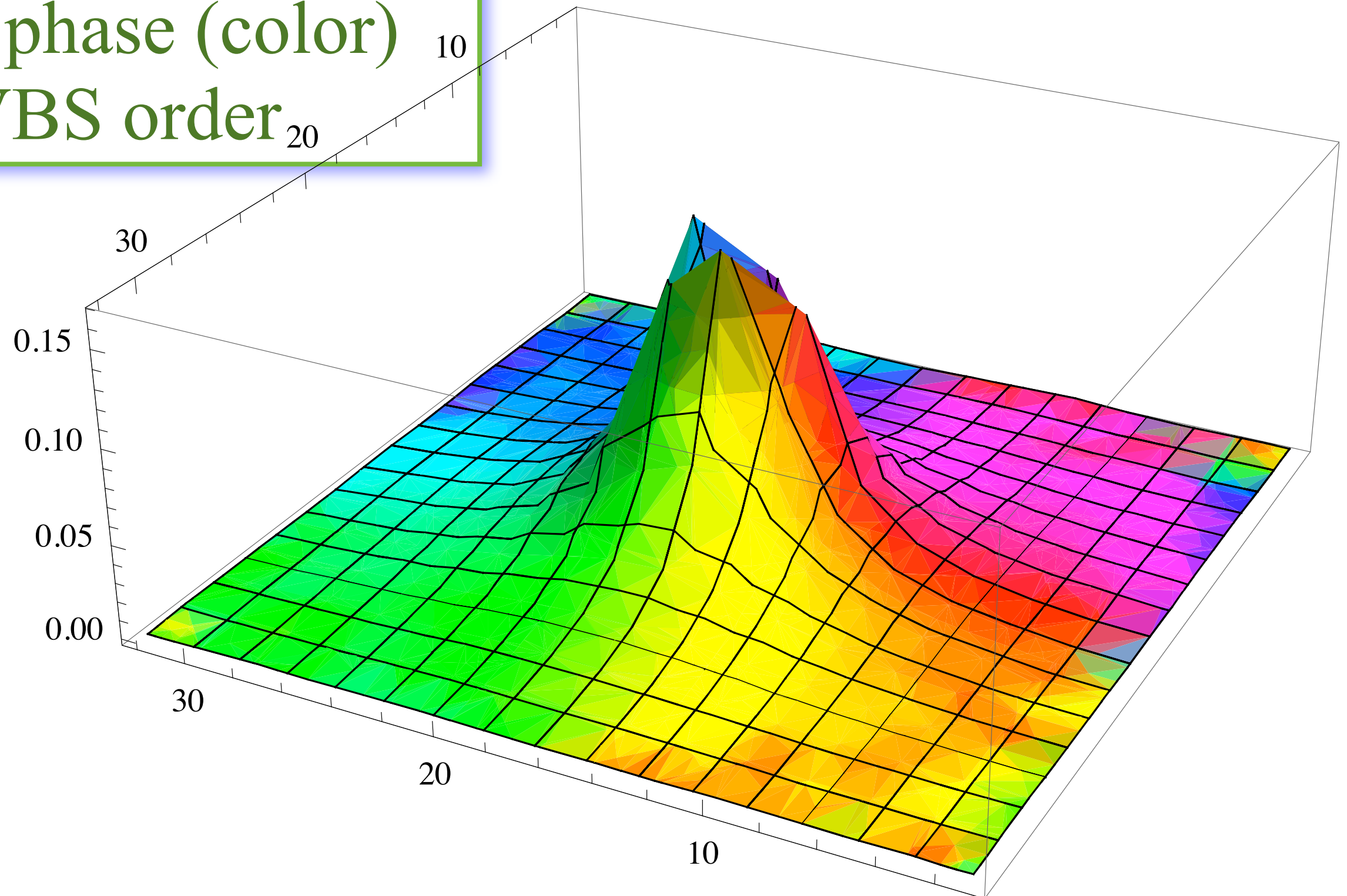
Bond order from QMC of J-Q model

Phase of
VBS order



Bond order from QMC of J-Q model

Amplitude (height)
and phase (color)
of VBS order



Conclusions

- Vison and spinon excitations of a Z_2 spin liquid: possible explanation for NMR and thermal conductivity measurements on κ -(ET)₂Cu₂(CN)₃
- Signature of photon in circular distribution of VBS order around Zn impurity: possibly relevant for STM studies of bond order in underdoped cuprates.

