Title: Experimental Sightings of the Quantum Spin Liquid

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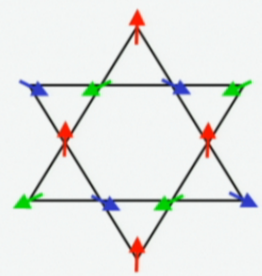
Abstract: New states of matter may be produced if quantum effects and frustration conspire to prevent the ground state from achieving classical order. An example of a new quantum phase is the quantum spin liquid. Such spin liquids cannot be characterized by local order parameters; rather, they are distinctive by their possession of long range quantum entanglement. I will describe recent experimental progress in the quest to study quantum spin liquids in frustrated magnets. The kagome lattice, composed of corner-sharing triangles, is highly frustrated for antiferromagnetic spins. Materials based on the kagome lattice with spin-1/2 are ideal hosts for quantum spin liquid ground states. I will discuss our groupâ€™s work which includes single crystal growth, bulk characterization, and neutron scattering measurements of the S=1/2 kagome lattice material ZnCu3(OH)6Cl2 (also known as herbertsmithite). Our inelastic neutron scattering measurements of the spin correlations in a single crystal sample reveal that the excitations are fractionalized, a hallmark signature of spin liquid physics.

Experimental sightings of the quantum spin liquid

Young Lee (MIT)



4-Corners Southwest Ontario Condensed Matter Symposium
Perimeter Institute, May 2014

Cast and Credits

MIT Physics
Harry Han *
Joel Helton *
Kit Matan
Robin Chisnell
Craig Bonnoit
Dillon Gardner
Andrea Prodi

MIT CMSE
Shaoyan Chu

MIT Chemistry
Dan Nocera
Danna Freedman
Matthew Shores
Emily Nytko
Daniel Grohol

Johns Hopkins / NIST
Tyrel Mcqueen
Collin Broholm
Jose Rodriguez

Oak Ridge
Steve Nagler

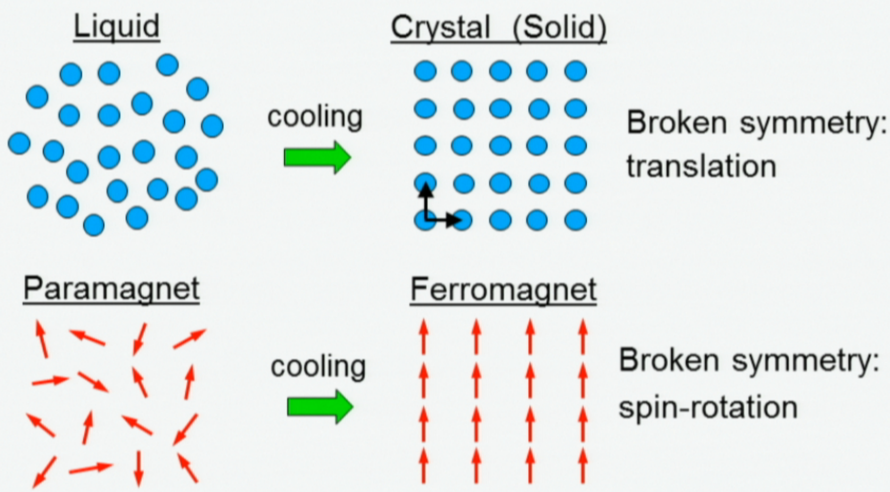
A brief history of magnetic ground states

Ferromagnet

before 4th century BC

Searching for "novel" states in condensed matter

Contrast with conventional ground states:



What if, due to quantum mechanics, the ground state did not break these symmetries?

A simple system of interacting spins:

$$H = J \sum_{\substack{\text{nearest} \\ \text{neighbors}}} \vec{S}_i \cdot \vec{S}_j$$

the "Heisenberg" Hamiltonian

J is the "magnetic exchange" energy:

J < 0 : spins want to point in same direction
- a ferromagnet, like iron

J > 0 : spins want to point in opposite directions
- an antiferromagnet

Competing ideas for the antiferromagnet

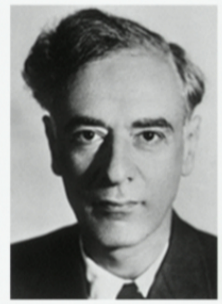
Louis Néel

1970 Nobel Prize for "fundamental work & discoveries concerning antiferromagnetism"

$\downarrow \uparrow \downarrow \uparrow \downarrow \uparrow$

Lev Landau

1962 Nobel Prize for "his pioneering theories of condensed matter"



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

Classical versus Quantum

The fundamental difficulty

$$H = J \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j$$

J < 0 : ferromagnet

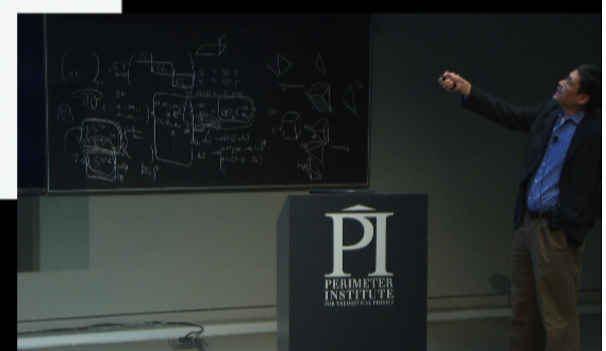
↑ ↑ ↑ ↑ is an eigenstate

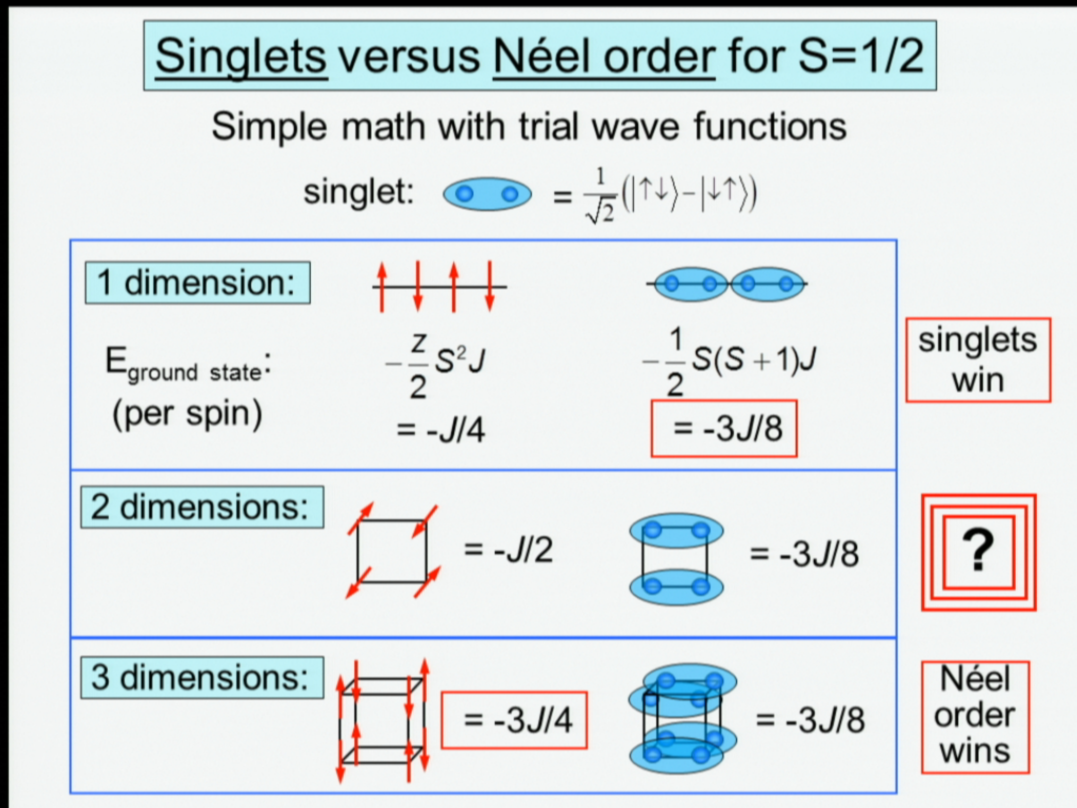
quantum state = classical state

J > 0 : antiferromagnet

↑ ↓ ↑ ↓ is not an eigenstate!

quantum state ≠ classical state




Singlets versus Néel order for S=1/2
Simple math with trial wave functions
singlet: $= \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$
1 dimension:
$E_{ground\ state}$:
(per spin)
$-\frac{z}{2}S^2J$
= -J/4
$-\frac{1}{2}S(S+1)J$
= -3J/8
singlets win
2 dimensions:
= -J/2
= -3J/8
?
3 dimensions:
= -3J/4
= -3J/8
Néel order wins

40 years since Phil Anderson's provocative proposal

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

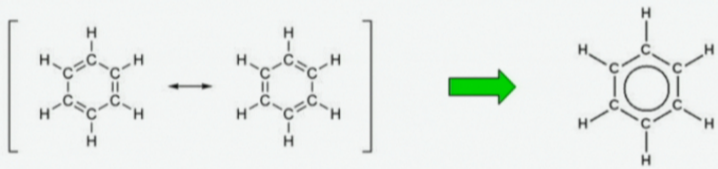
Physics Today 61(4), 8 (2008)

Who or what is RVB?

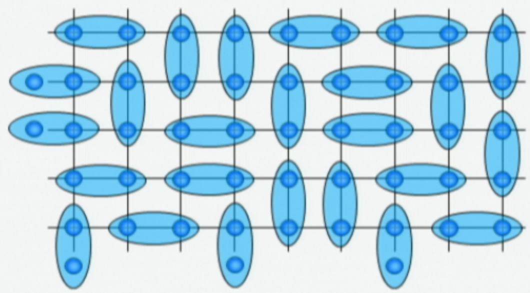
Philip Anderson

RVB = "resonating valence bond" (a quantum spin liquid !)

Analogy to Linus Pauling and benzene:



The quantum spin-liquid:
A new state of matter in two-dimensions



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

Every spin is hidden in a singlet

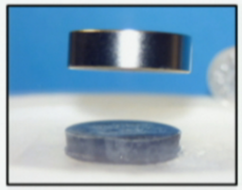
Anderson's RVB state (1973--triangular) (1987--square)

Actual wavefunction is a superposition of many configs.

The ground state does not break conventional symmetries, it is not a crystal (no translational symmetry breaking), and it is not an ordered magnet (no spin-rotation breaking).

What's a quantum spin liquid good for?

- central to theories of high-T_c superconductivity



- state with long-range quantum entanglement
  → proposals for topologically protected qubits

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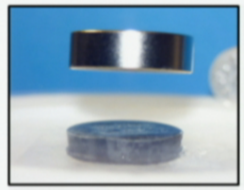
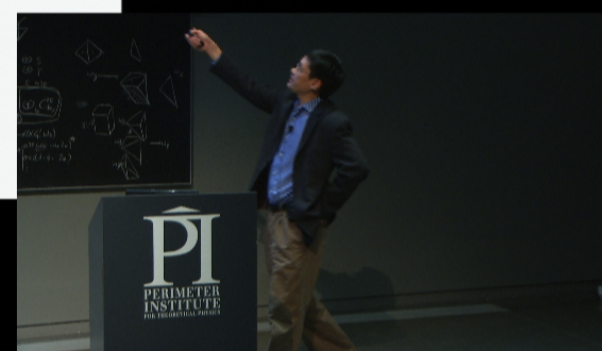
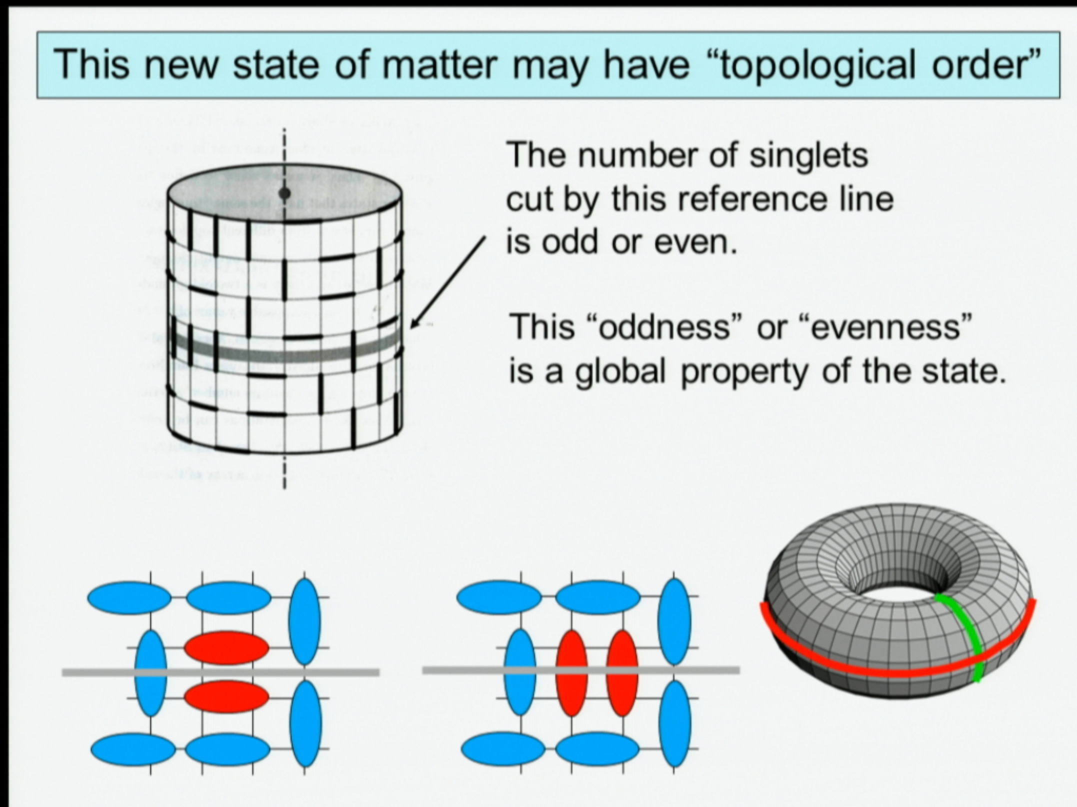


- state with <u>long-range quantum entanglement</u>
  → proposals for topologically protected qubits

- It's fundamental physics
- It's fun
  - strong interplay between theory and experiment




This new state of matter may have “topological order”
The number of singlets
cut by this reference line
is odd or even.
This “oddness” or “evenness”
is a global property of the state.

Big picture: new paradigm for describing quantum matter

Xiao-Gang Wen (Perimeter/MIT)

Long-range entanglement
and topological order
(nice review arXiv:1210.1281)

Examples:
1) Fractional quantum Hall effect
   - experimentally realized
2) Quantum spin liquids
   - experimentally realized (?)

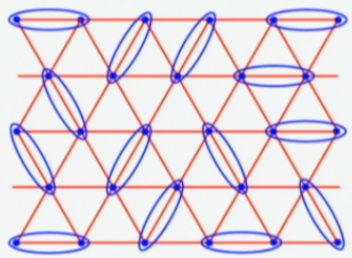
<u>An experimental signature:</u>
Exotic excitations with fractional quantum numbers

Only a handful of quantum spin liquid candidates are known

They all share a common a common feature: **frustration!**

Triangular lattice

Edge sharing



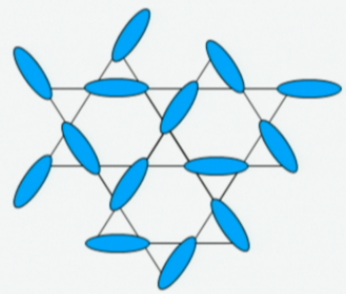
organic compounds

κ-$(ET)_2Cu_2(CN)_3$, $EtMe_3Sb[Pd(dmit)_2]_2$

Kagomé lattice

Corner-sharing

→ more frustrated



$ZnCu_3(OH)_6Cl_2$ (Herbertsmithite)

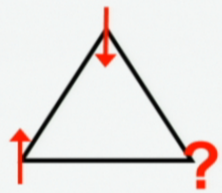
Why deal with frustration?

Problem: Néel order on the square lattice "kills" the spin liquid

Solution: add **frustration**

The main ingredient for geometrical frustration:

Lattices based on triangles



$$H = J \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j$$

$J > 0$ (antiferromagnetic)

MASSACHUSETTS INSTITUTE OF TECHNOLOGY
DEPARTMENT OF PHYSICS

EDUCATION OFFICE
ROOM 4-352

PHONE (617)-253-4842
FAX (617)-258-8319

DOCTORAL GENERAL EXAMINATION PART II (from hell)

Problem 1) Find the ground state of the spin-1/2 Heisenberg model on the kagomé lattice.

$$H = J \sum_{<ij>} \vec{S}_i \cdot \vec{S}_j$$

(with $J > 0$)

(The question is simple to state.)

What's the ground state for the **quantum spin-1/2** kagomé lattice Heisenberg model?

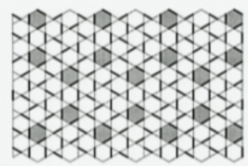
Theoretical consensus: ground-state is not Néel ordered

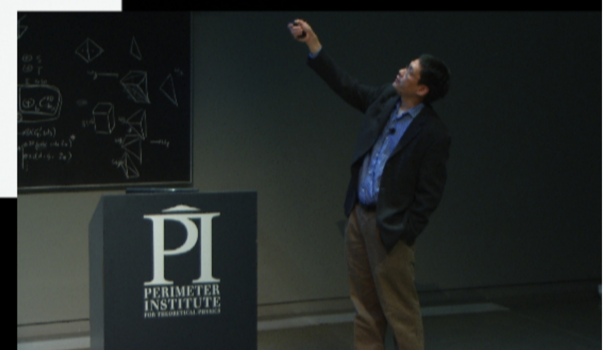
Nature of ground state?

1) Spin liquid (gapped? gapless?)
- Waldtmann *et al.*, Eur. Phys. J. B 2, 501 (1998)
  → spin-gap ≤ J/20
- Ran, Hermele, Lee, Wen, *et al.*, PRL 98, 117205 (2007)
  → gapless Dirac fermions
- Yan, Huse, White, Science 332, 1173 (2011)
  → spin gap, Z_2 topological order

2) Valence bond crystal

Nikolic & Senthil (2003)
Singh & Huse (2008)





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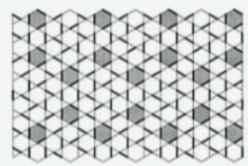


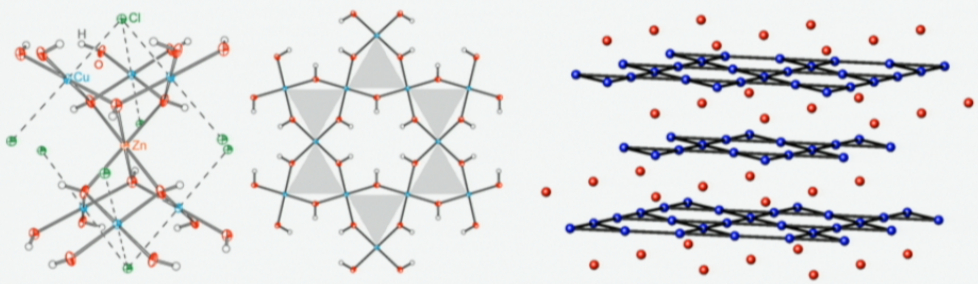
Experiments to the rescue: ideal S=1/2 kagomé lattice material <u>Herbertsmithite</u> ($ZnCu_3(OH)_6Cl_2$)

S=1/2 Cu^{2+} kagomé layers separated by non-magnetic Zn^{2+} layers

Collaboration with Dan Nocera (MIT, Chemistry)

Shores *et al*., J. Am. Chem. Soc. **127**, 13 462 (2005)

Helton *et al*, Phys. Rev. Lett., **98**, 107204 (2007)



This has the ideal kagomé structure

From neutron, NMR, μSR, and thermodynamic measurements on powders, we know that herbertsmithite ***is not*** :

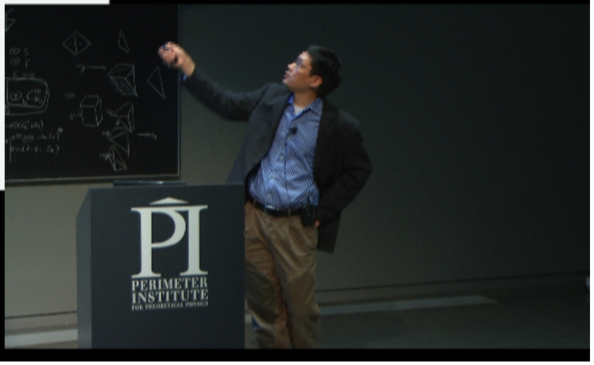
Helton *et al*, PRL (2007), Mendels *et al*, PRL (2007)

1) It is not Néel ordered or frozen (down to T=J/3000)
2) It is not gapped (at least down to Δ=J/170)

If the above two statements can be made without qualifiers, then the state ***is*** a quantum spin-liquid.

Sherlock Holmes to Watson:
"When you have eliminated the impossible, whatever remains, however improbable, must be the truth"

Deducing a spin liquid by ruling out other possibilities
- not completely satisfying



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What is a positive signature for a quantum spin liquid?

→ The excitations are "**exotic**"

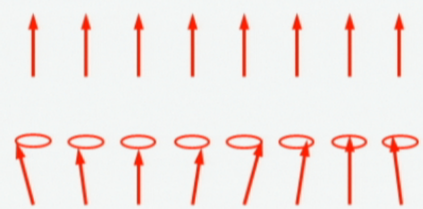
What's a typical, "**non-exotic**" excitation?

→ a spin-wave

Classical picture:

Ground state (total spin NS)

Lowest excited state (NS-1)



The $\Delta S=1$ is shared among all spins.

The energy $\hbar\omega$ of the wave depends on the wavelength

What is a positive signature for a quantum spin liquid?

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Classical picture:

Ground state (total spin NS)

Lowest excited state (NS-1)

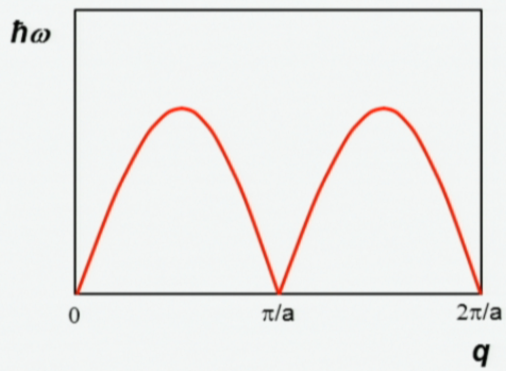
The $\Delta S=1$ is shared among all spins.

The energy $\hbar\omega$ of the wave depends on the wavelength $\boldsymbol{q}$.

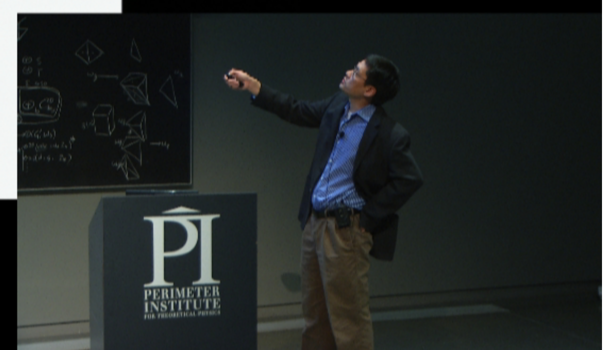
"Non-exotic" spin-wave excitations (cont'd)

The S=1 quantum of excitation is called a "magnon."
- this emergent quasiparticle is a boson

Magnon dispersion relation in an antiferromagnet



Spontaneously broken continuous symmetry
$\rightarrow$ massless Goldstone boson



What is a positive signature for a quantum spin liquid?

$\rightarrow$ Fractionalized excitations -- ***Spinons***

Well understood in one-dim S=1/2 antiferromagnetic Heisenberg chain

Ground state: (H. Bethe, 1931)

superposition of singlets, short-range AF order

What is a positive signature for a quantum spin liquid?

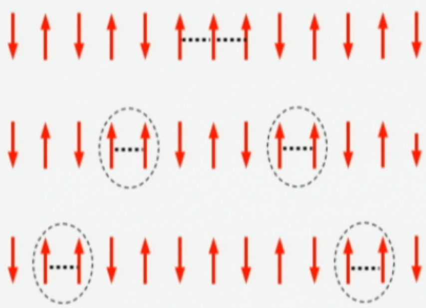
→ Fractionalized excitations -- ***Spinons***

Well understood in one-dim S=1/2 antiferromagnetic Heisenberg chain

Ground state: (H. Bethe, 1931)

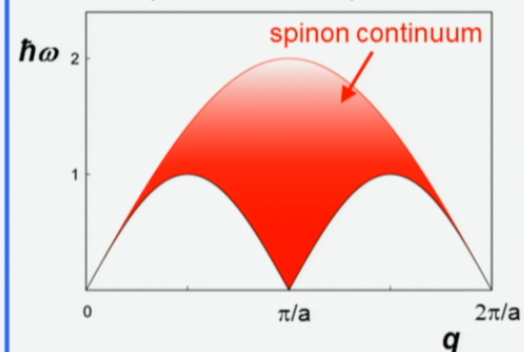
superposition of singlets, short-range AF order

An S=1 excitation fractionalizes into two S=1/2 *spinons*

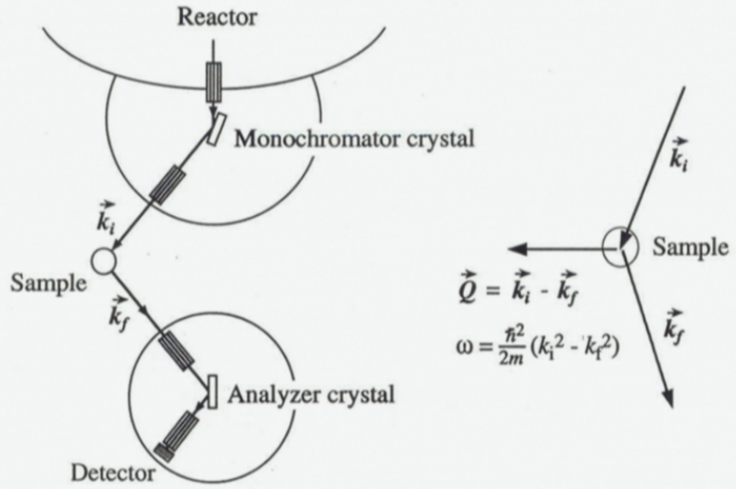


the spinons are *deconfined*

Spin excitation spectrum

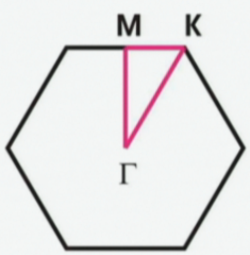


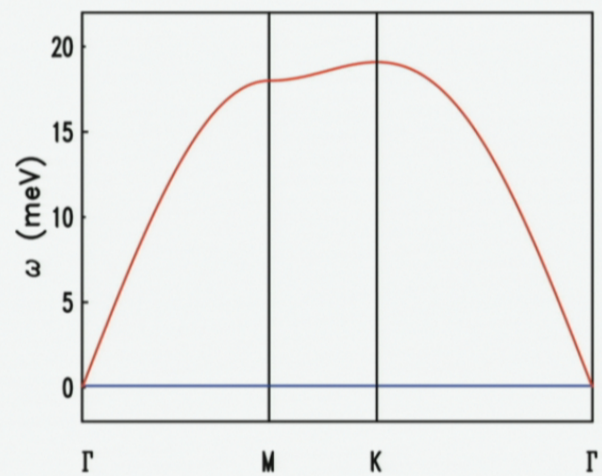
Triple-axis neutron scattering technique



$$\textbf{Intensity} \propto \mathbf{S}^{\alpha\beta}(\vec{Q},\omega) = \frac{1}{2\pi\hbar}\int dt\, e^{-i\omega t}\,\frac{1}{N}\sum_{\vec{R}\vec{R}'} e^{i\vec{Q}\cdot(\vec{R}-\vec{R}')} < S^{\alpha}_{\vec{R}}(t)\,S^{\beta}_{\vec{R}'}(0) >$$

Textbook example: a classical spin wave dispersion





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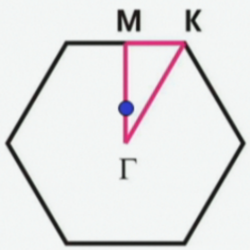

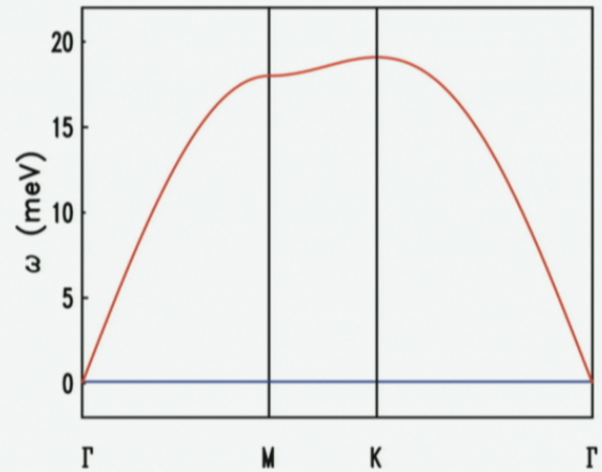

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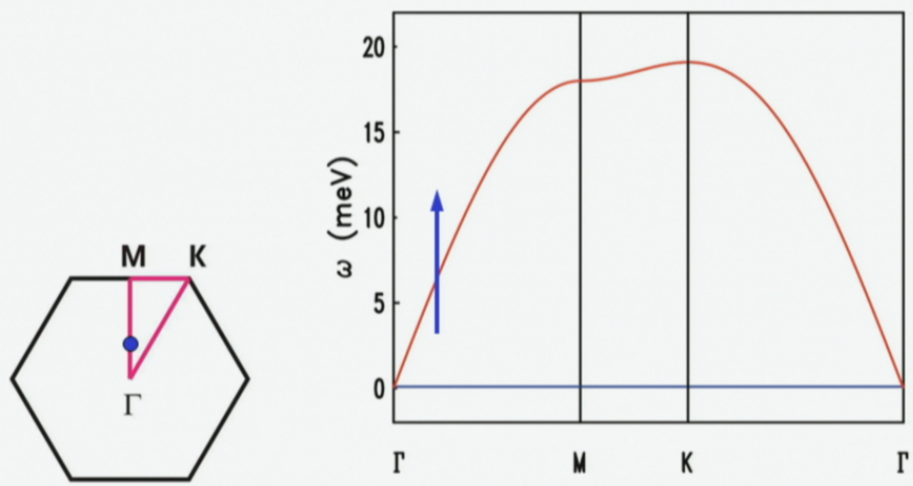

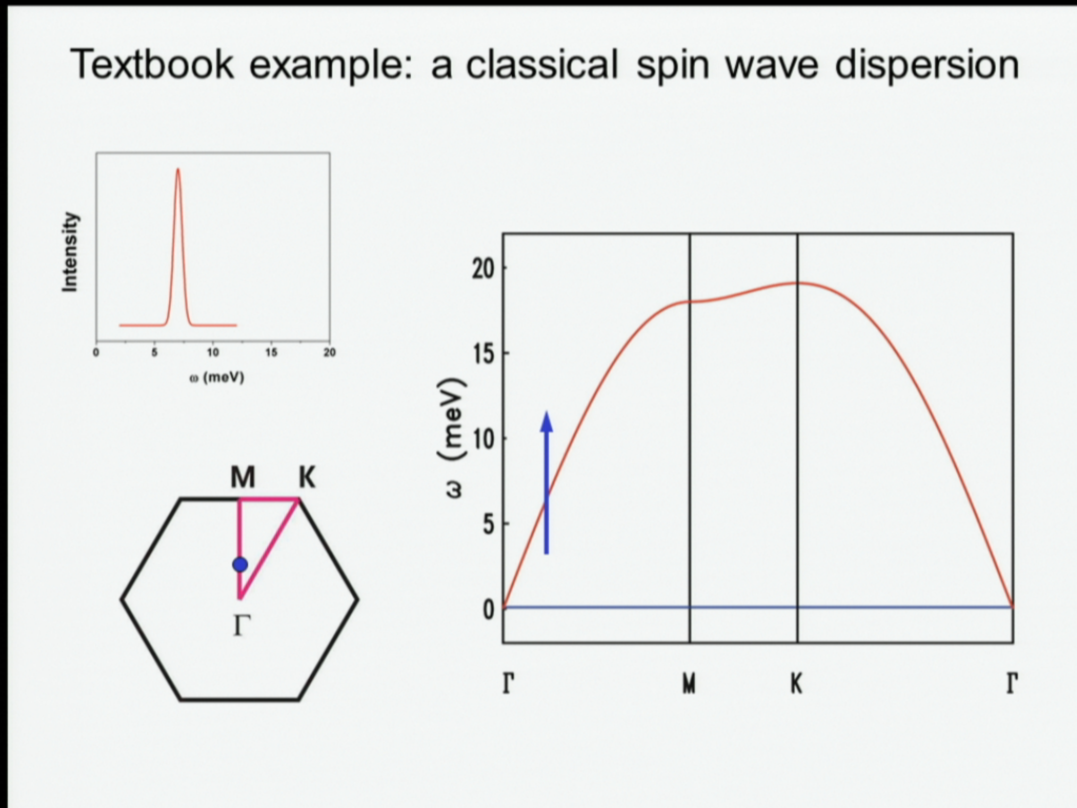
Textbook example: a classical spin wave dispersion
Intensity
ω (meV)
0
5
10
15
20
M
K
Γ
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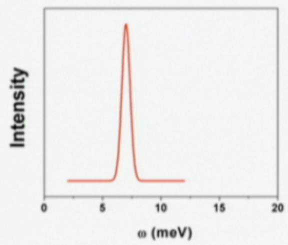
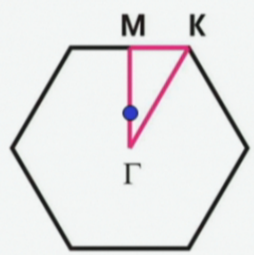
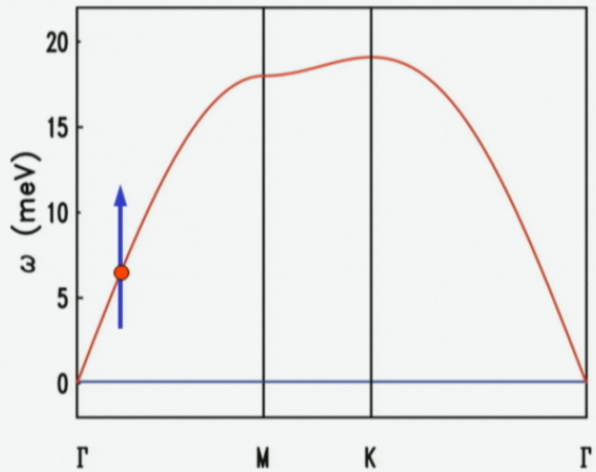
Textbook example: a classical spin wave dispersion







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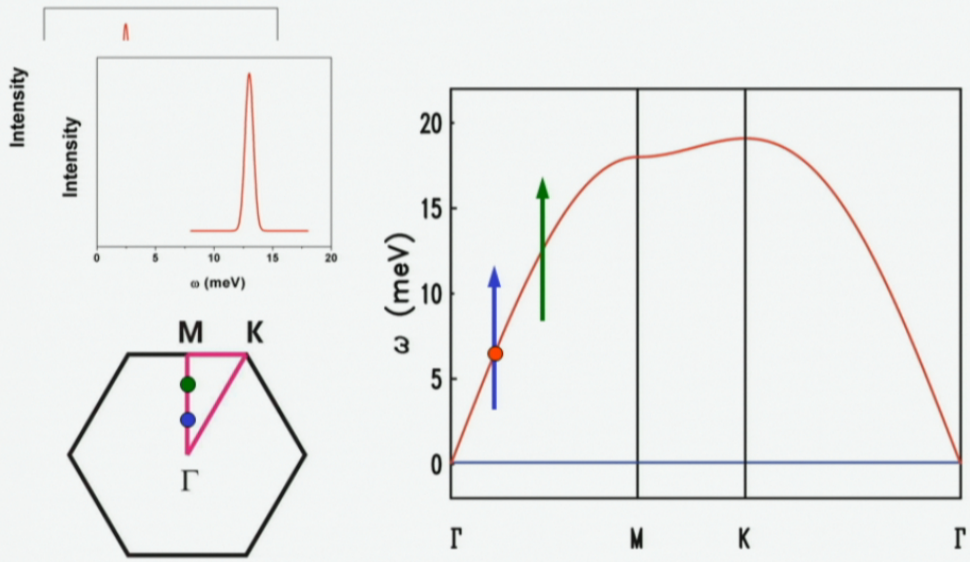

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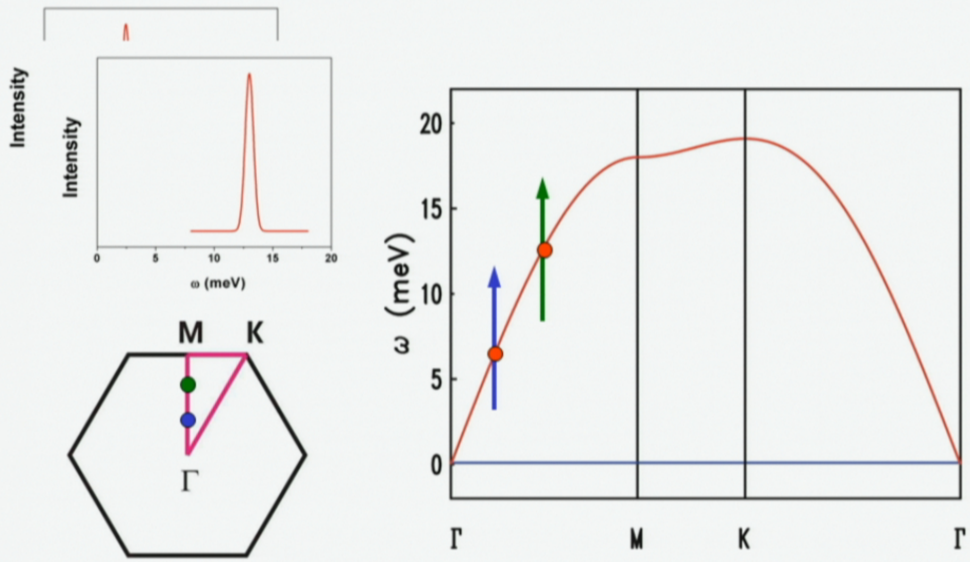


Does the S=1/2 kagome lattice have
a quantum spin liquid ground state?

The lynchpin experiment:

Inelastic neutron scattering on **single crystals**

(find deconfined spinons in a 2D magnet !)

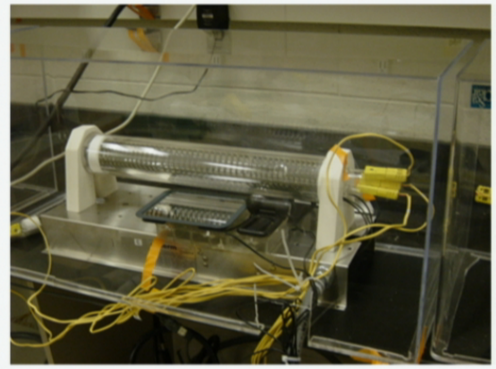
Obstacles for this experiment are daunting:

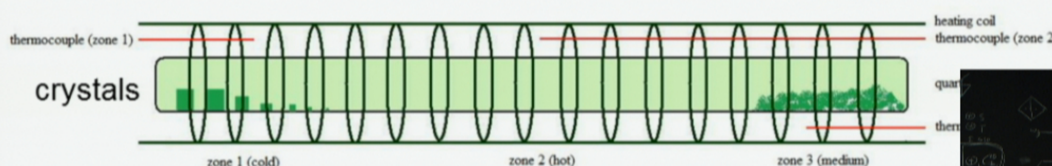
1) Small spin value S=1/2
2) No sharp peaks expected in ω or $\boldsymbol{Q}$
3) No recipe for crystals

The urgent need for single crystals

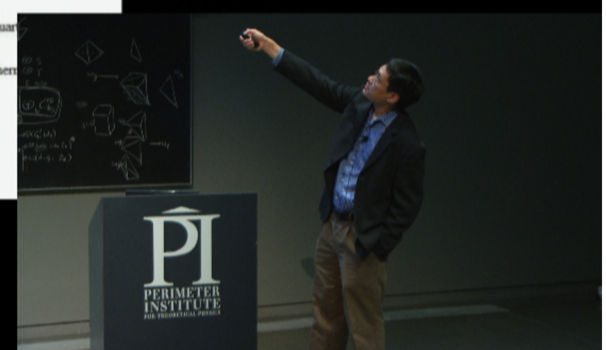
Harry's heroic efforts

Inventing and perfecting a new crystal growth method --the "hydrothermal zone"





Typical temp: 175 C 185 C 180 C



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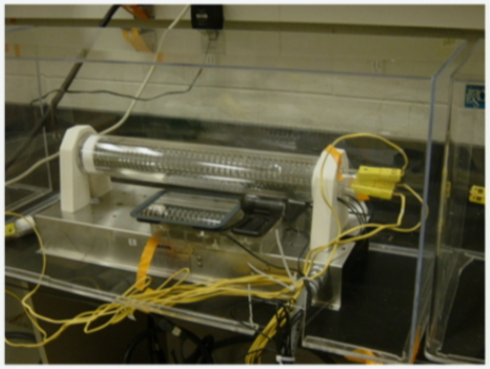

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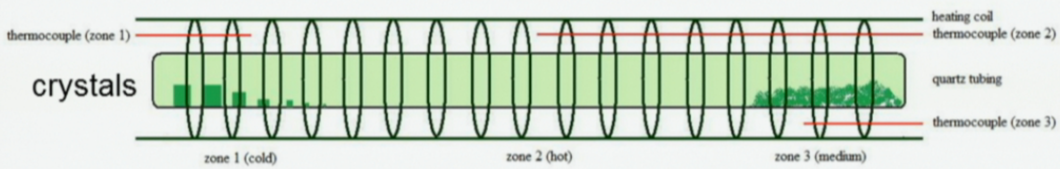
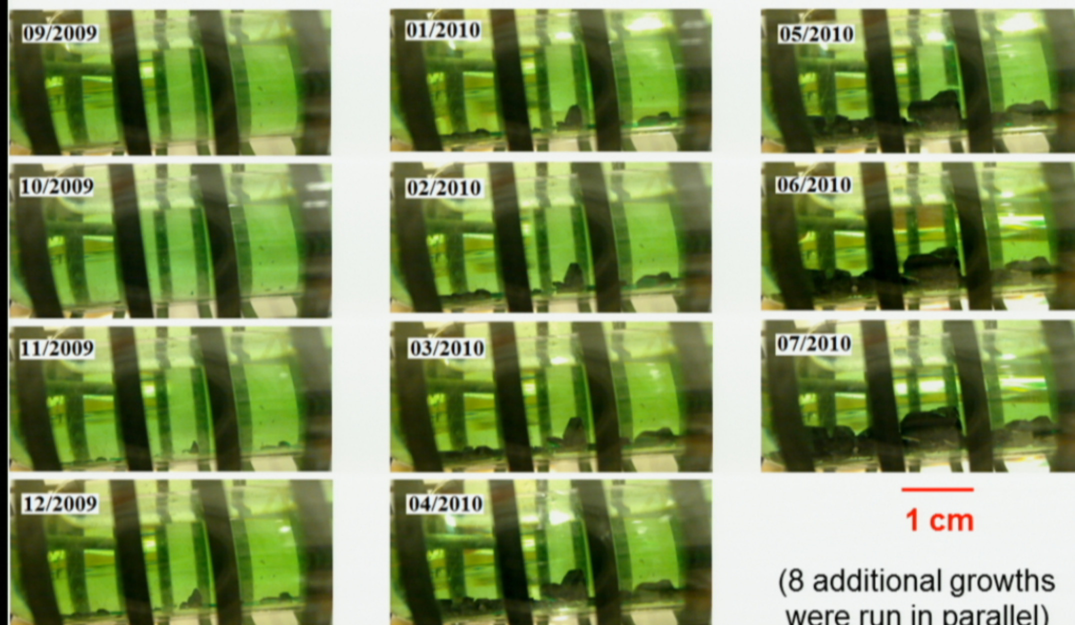


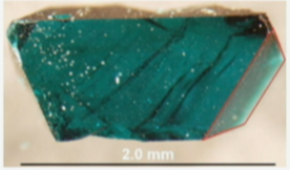
Typical temp: 175 C 185 C 180 C

A crazy thing to do... unless it works!



(8 additional growths were run in parallel)

Crystals now available to explore the physics



$ZnCu_3(OH)_6Cl_2$
herbertsmithite single crystals

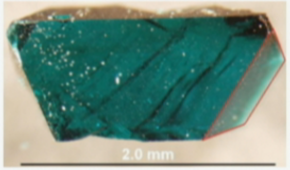
Crystals used in collaborations:

1) Raman scattering – D. Wulferding, et al, PRB 82, 144412 (2010)
2) NMR – T. Imai, et al, PRB 84, 020411 (2011), Fu – next talk
3) μSR – O. Ofer, et al, J. Phys. Cond. Mat. 23, 164207 (2011)
4) Optical conductivity – N. Gedik, et al, PRL (2013)

This talk:

1) Improved characterization of $ZnCu_3(OH)_6Cl_2$
2) The spin excitations using neutrons
3) High field thermodynamics

Crystals now available to explore the physics



$ZnCu_3(OH)_6Cl_2$
herbertsmithite single crystals

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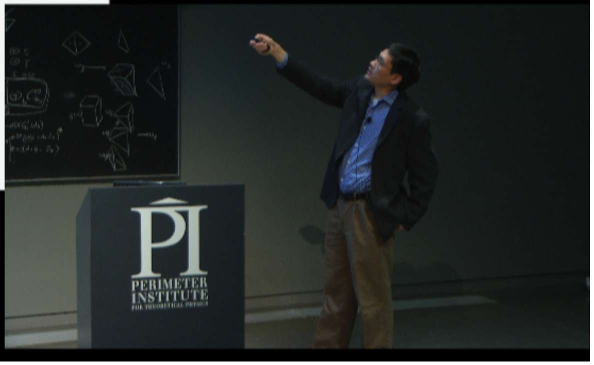
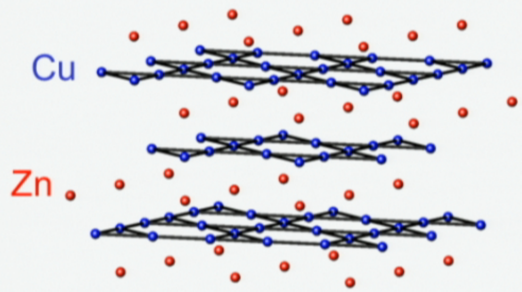
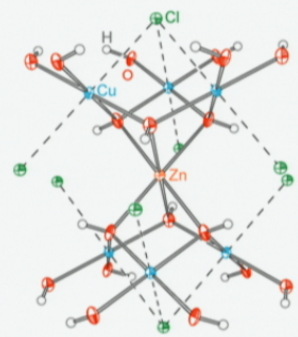


The question of impurities: Does "anti-site" disorder exist in herbertsmithite?

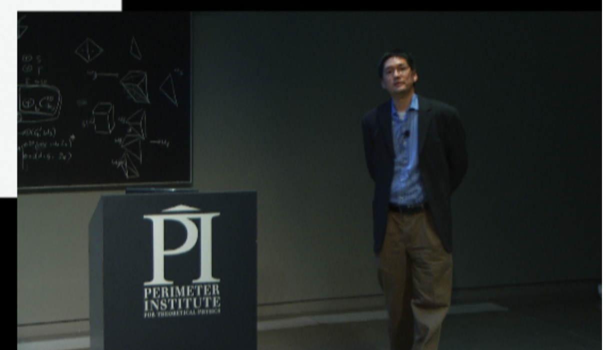
Perhaps Cu and Zn ions swap locations (at the 5% level) ?

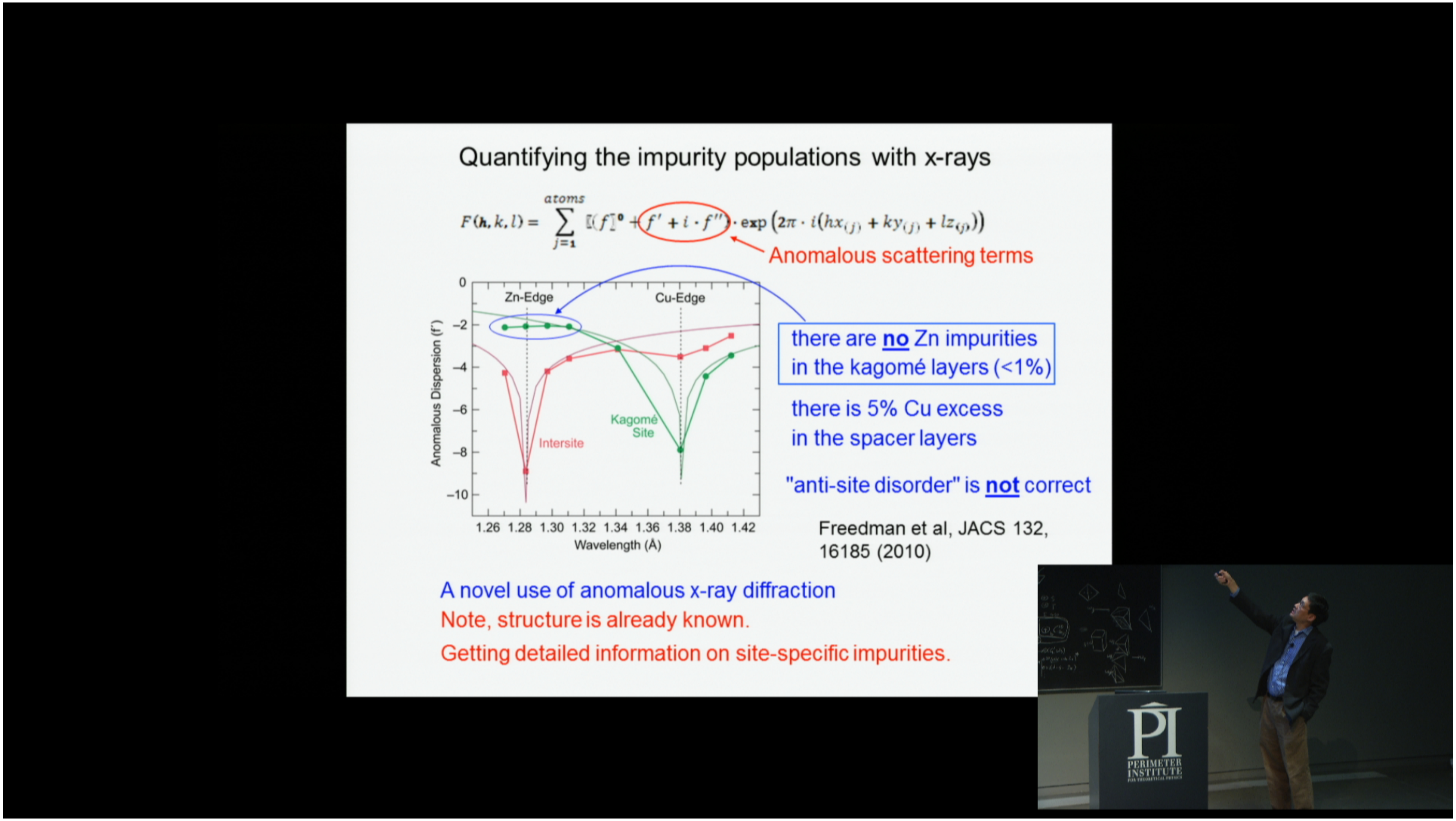
Zn in the kagomé layer would be a strong form of disorder
Cu in the spacer layer would be less so.





A difficult question in materials science





Quantifying the impurity populations with x-rays
$F(h,k,l) = \sum_{j=1}^{atoms} [(f)^0 + (f' + i \cdot f'') \cdot \exp(2\pi \cdot i(hx_{(j)} + ky_{(j)} + lz_{(j)}))$
Anomalous scattering terms
Zn-Edge
Cu-Edge
Anomalous Dispersion (f')
Intersite
Kagomé Site
Wavelength (Å)
there are no Zn impurities in the kagomé layers (<1%)
there is 5% Cu excess in the spacer layers
"anti-site disorder" is not correct
Freedman et al, JACS 132, 16185 (2010)
A novel use of anomalous x-ray diffraction
Note, structure is already known.
Getting detailed information on site-specific impurities.
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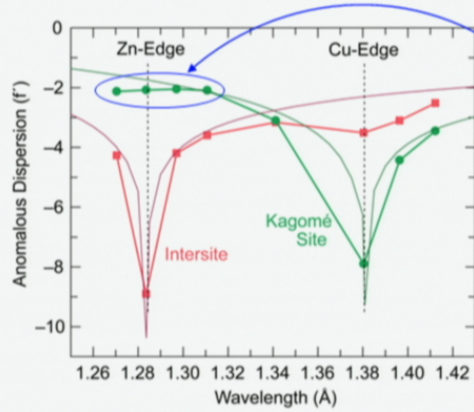
Anomalous scattering terms



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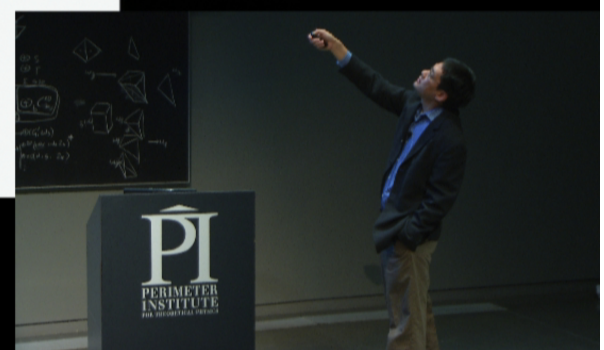

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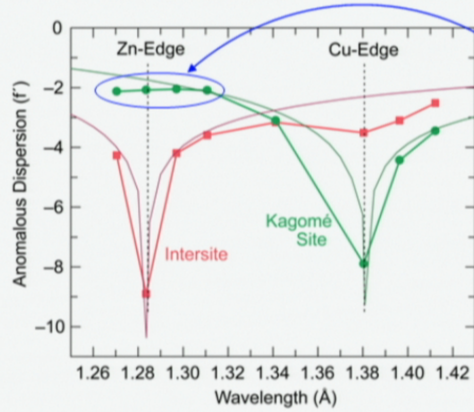

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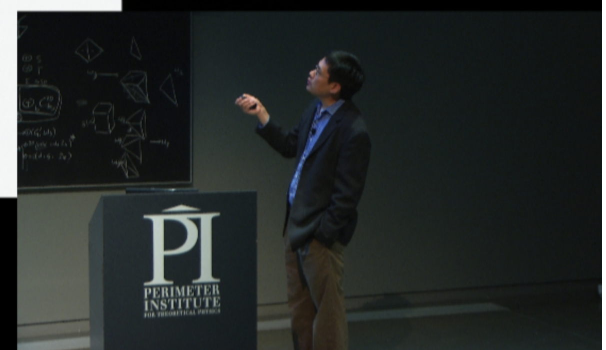


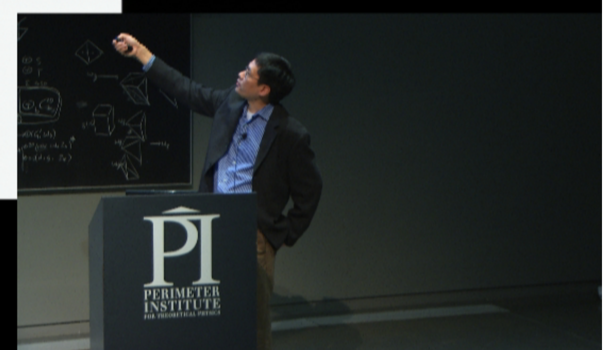
Herbertsmithite is now exceedingly well characterized:

1) The kagome geometry of Cu ions is ideal (all bonds equivalent)

2) No observed magnetic ordering, spin freezing, or spin-gap down to low temperatures and energies

3) No Zn impurities in the Cu kagome layers
   - ~5% excess Cu S=1/2 impurities in-between layers

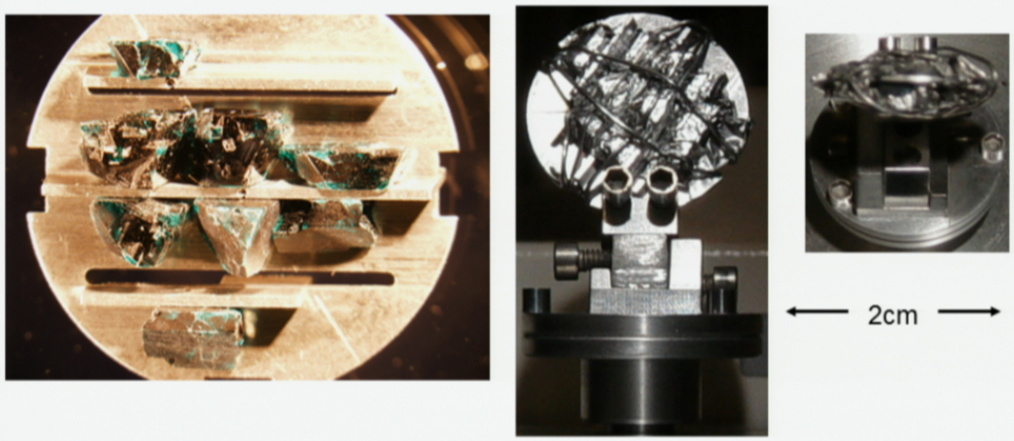
4) The Heisenberg model is a good approximation
   - easy-axis exchange at the ~10% level
   - DM interaction at the ~10% level



Towards a neutron sample (Harry's heroic efforts, part II)

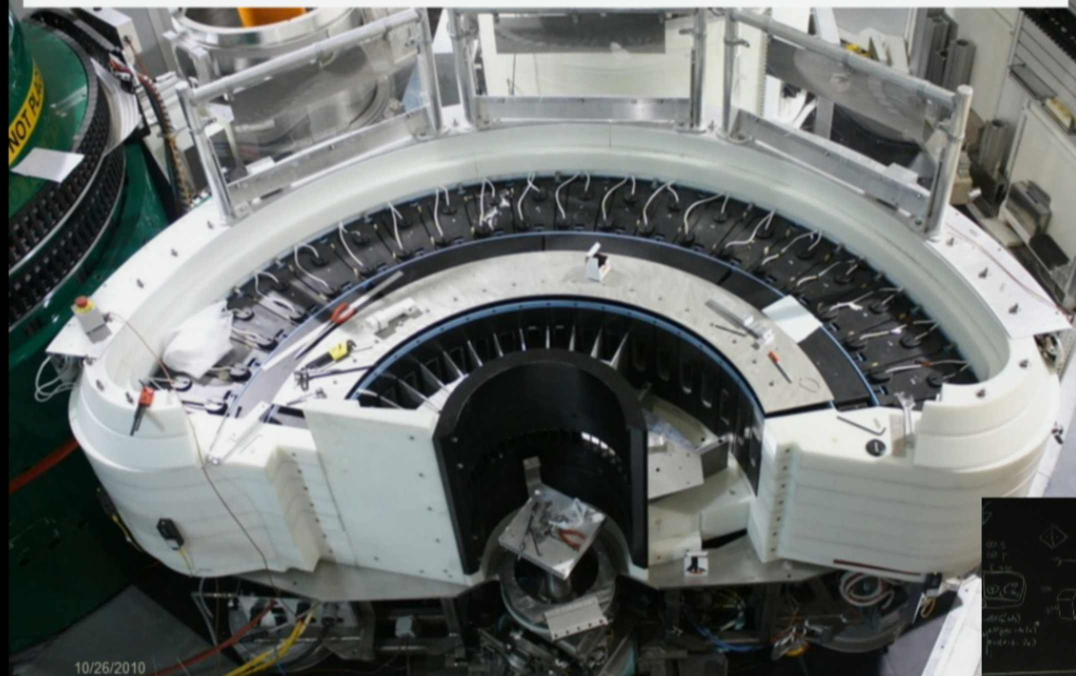
Comounting 15 deuterated herbertsmithite crystals with total mass **1.25g**

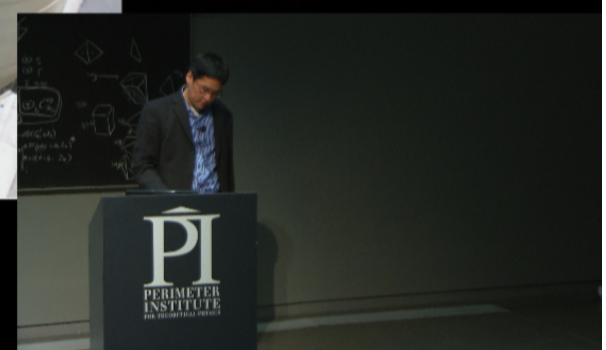
The "single" crystal for neutron scattering



All crystals coaligned within 2 degrees
(the divergence of the neutron beam is ~1-2 degrees)

New high-throughput cold triple-axis instrument **MACS** at NIST Center for Neutron Research



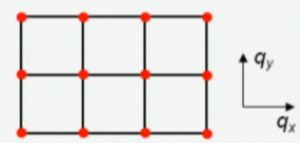


Expectations for the magnetic excitation spectrum

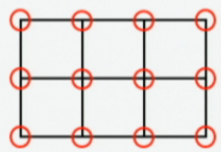
S=1/2 square lattice

ordinary spin-waves

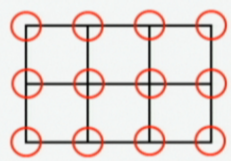
ω=0 slice



ω=J/5



ω=J/2

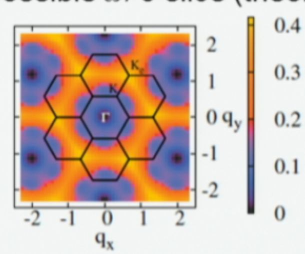


S=1/2 kagomé lattice

???

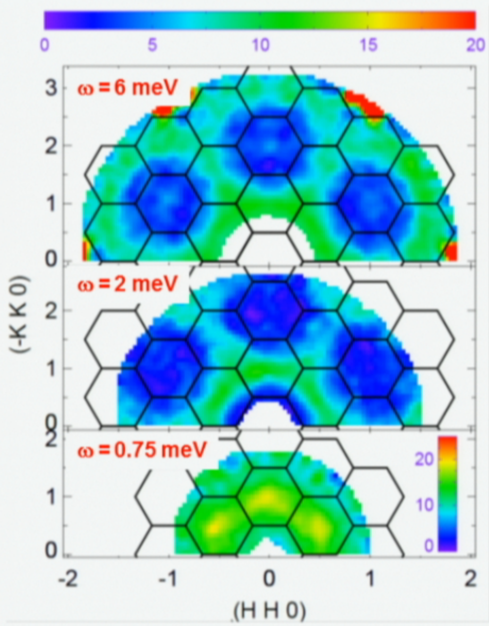
ω=0
no Bragg peaks

A possible ω≠0 slice (theory)



Tchernyshyov et al (2010)

Spin correlations of the S=1/2 kagomé antiferromagnet



T Han, YL, et al,
Nature 492, 406 (2012)

T = 1.6 K

magnetic coupling J~17 meV

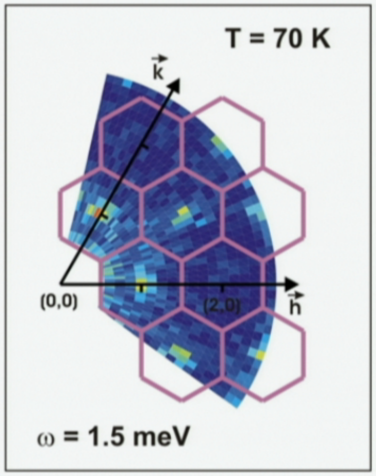
The observed scattering is:

- intrinsic to the kagomé spins
  (not just due to impurities)
- diffuse
  → no sharp dispersion surfaces !

Plots of $S(\vec{Q},\omega)$ (background measured with empty sample holder and subtracted)

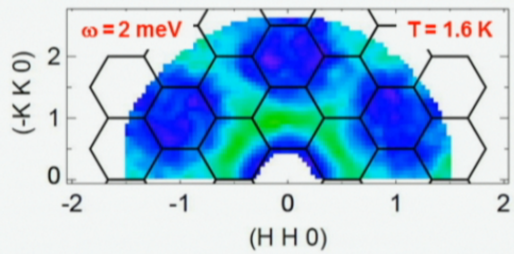
The S=1/2 data are completely novel

S=5/2 $KFe_3(OH)_6(SO_4)_2$



Even above T_N, the spins show the tendency for order

S=1/2 $ZnCu_3(OD)_6Cl_2$

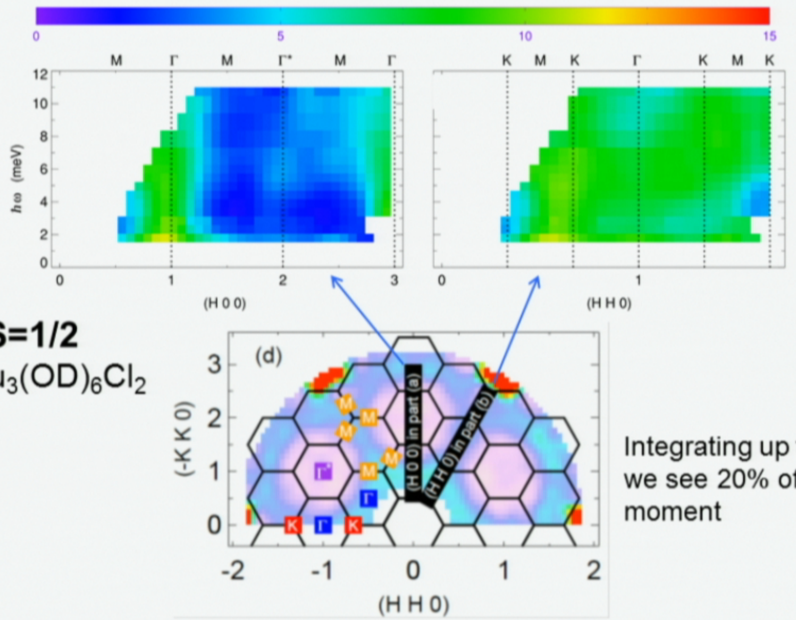


Here,
- there is no T_N
- no tendency for order (even at T << J)

In fact, the scattering looks like that expected for deconfined **<u>spinon</u>** excitations!

A continuum of spin excitations in a two-dim magnet

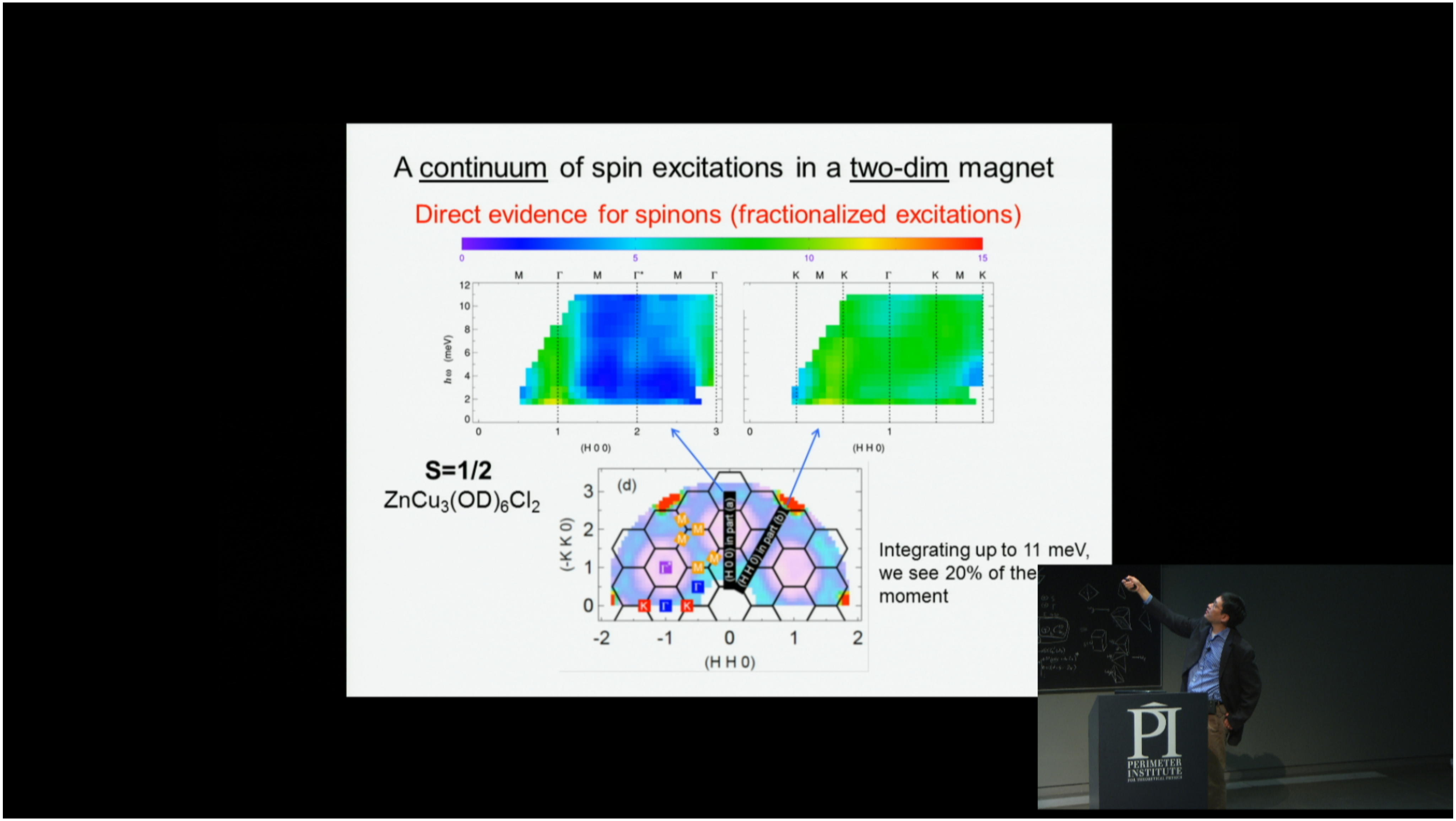
Direct evidence for spinons (fractionalized excitations)



S=1/2

$ZnCu_3(OD)_6Cl_2$

Integrating up to 11 meV, we see 20% of the total moment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S=1/2
ZnCu$_3$(OD)$_6$Cl$_2$
(H 0 0)
(H H 0)
(-K K 0)
(d)
Integrating up to 11 meV,
we see 20% of the
moment
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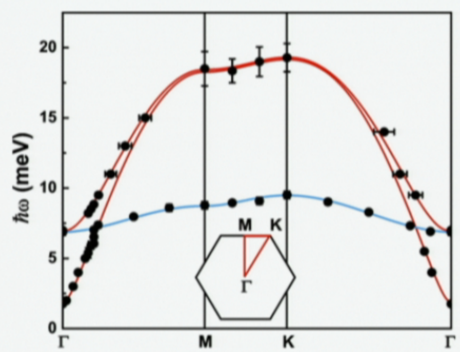
Contrast: a tale of two kagomé

S=5/2 $KFe_3(OH)_6(SO_4)_2$

ordered ground state

S=1 magnons

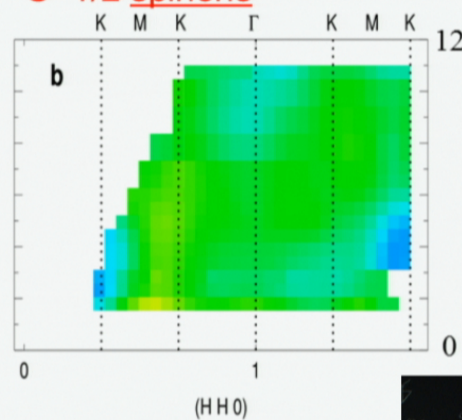


K Matan, YL, *et al.*,
PRL 96, 247201 (2006)

S=1/2 $ZnCu_3(OH)_6Cl_2$

spin liquid ground state

S=1/2 spinons



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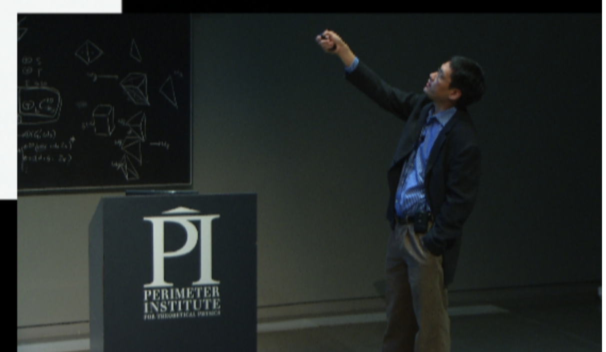
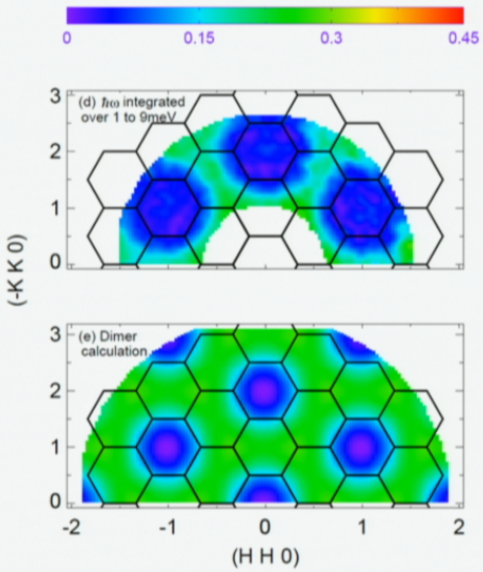
Nature 492, 406 (2012)



Making sense of the pattern in ***Q***-space

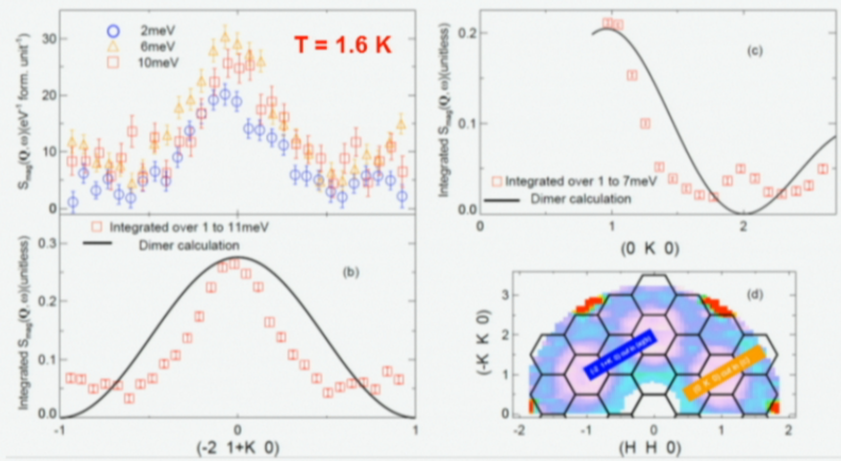


Energy integrated scattering
(if integrated over all energies, this would be the equal-time S(***Q***) → a snapshot of the correlations)

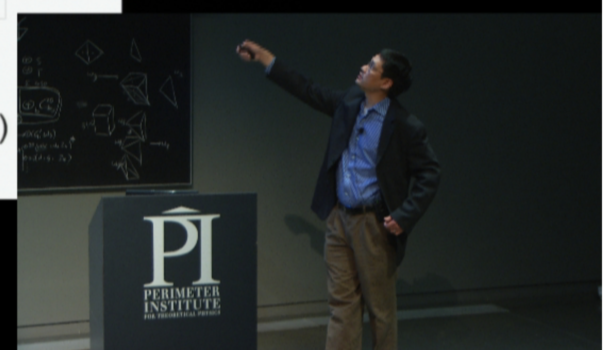
Calculation for uncorrelated nearest neighbor singlets

Spin correlations in herbertsmithite:
similar to short-range, resonating valence bond (RVB) state

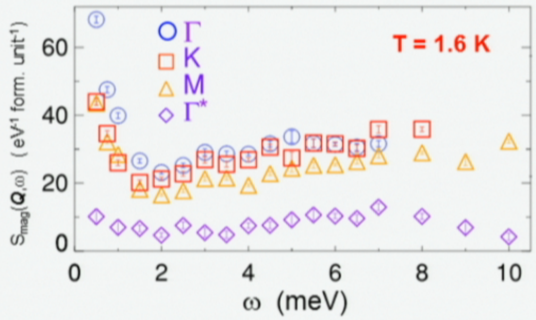
The spin correlations are short range,
though longer than just nearest neighbors

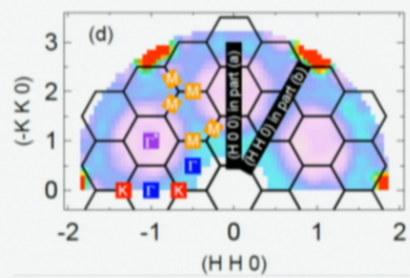


Spin correlation length ~ 5 Å
(various line-scans in ***Q*** fit to Lorentzian lineshapes)



Energy dependence at high symmetry positions





Above 2 meV: rather flat spectrum

Below 2 meV: increase in scattering at select ***Q***'s

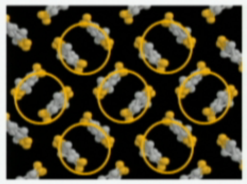
(re: scaling seen with powder)

No spin gap observed (at any ***Q*** position)
down to 0.25 meV (~J/70)

→ a gapless spin liquid, with short-range spin correlations (?)

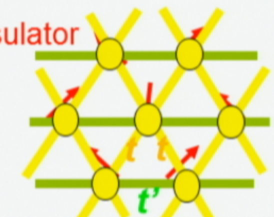
Intriguing point: the spin liquid materials observed so far appear to be gapless

κ-type organic salts



dimer model

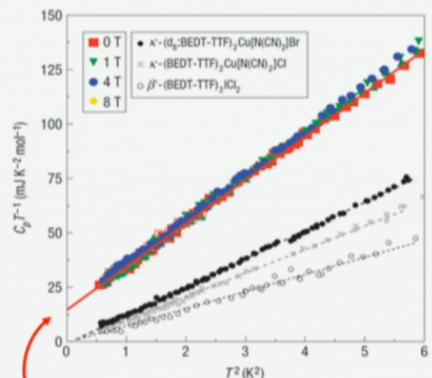
Mott insulator



anisotropic triangular lattice

$t' / t = 0.5 \sim 1.1$

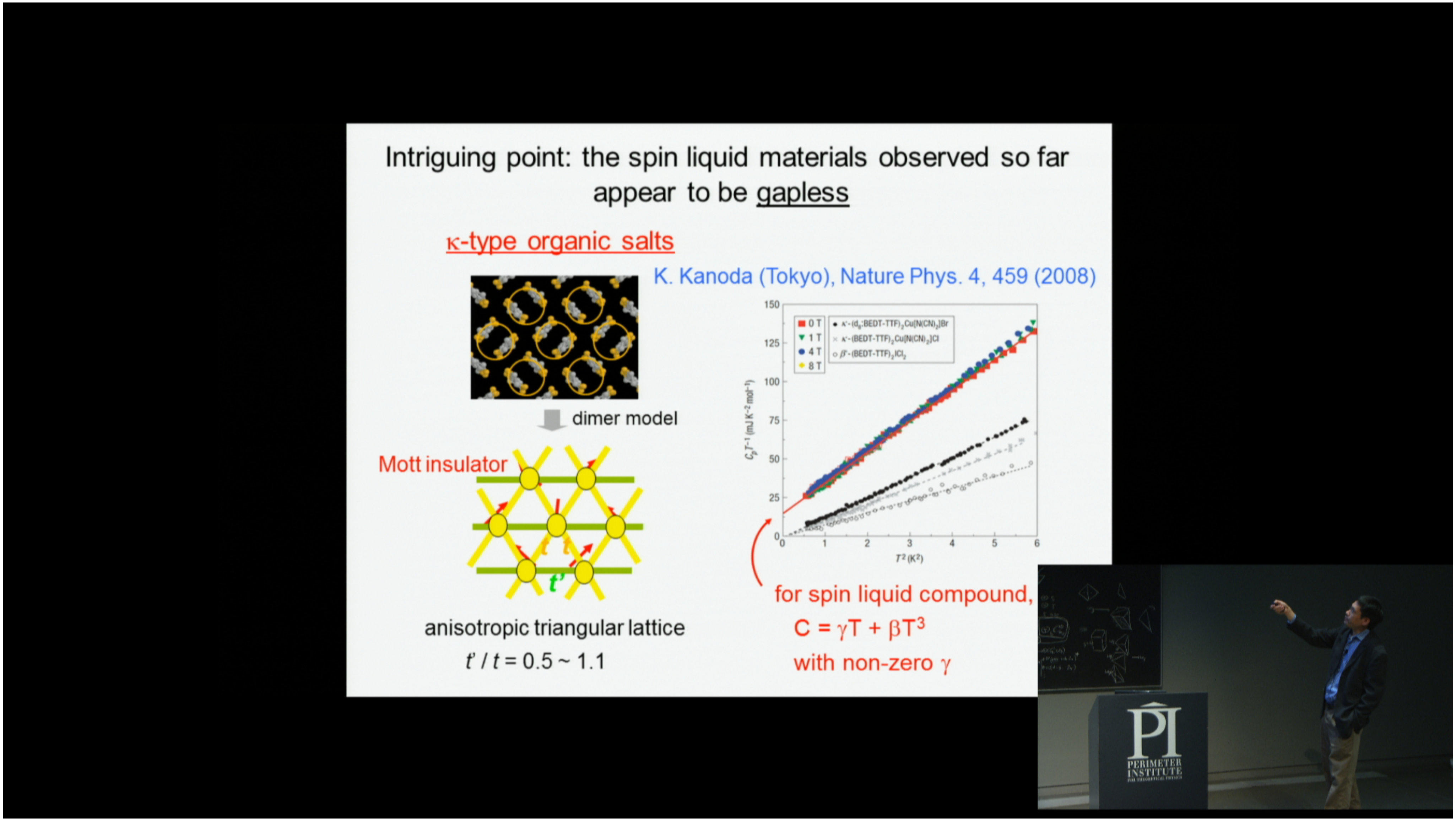
K. Kanoda (Tokyo), Nature Phys. 4, 459 (2008)



for spin liquid compound,

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

with non-zero γ



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K. Kanoda (Tokyo), Nature Phys. 4, 459 (2008)
dimer model
Mott insulator
t
t
t'
anisotropic triangular lattice
t' / t = 0.5 ~ 1.1
0 T
1 T
4 T
8 T
κ'-(d8:BEDT-TTF)2Cu[N(CN)2]Br
κ'-(BEDT-TTF)2Cu[N(CN)2]Cl
β'-(BEDT-TTF)2ICl2
CpT−1 (mJ K−2 mol−1)
T2 (K2)
0
25
50
75
100
125
150
0
1
2
3
4
5
6
for spin liquid compound,
C = γT + βT3
with non-zero γ
PI
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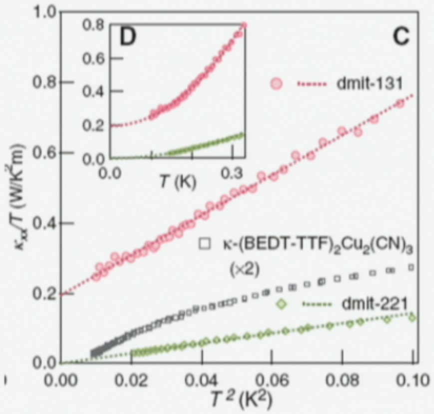
More gapless-ness: thermal conductivity of dmit organic salts

M. Yamashita et al, Science 328, 1246 (2010)



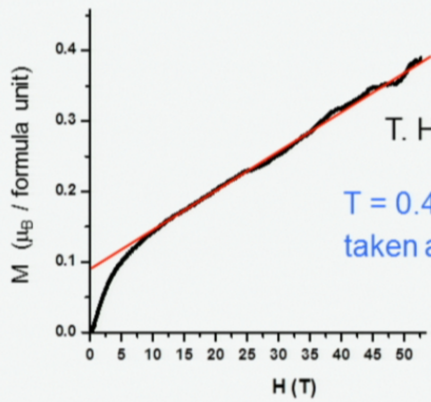
$$\kappa_{xx}^{\mathrm{spin}} = C_s v_s \ell_s / 3$$

mean free path reaches 500 inter-spin spacings

→ consistent with a spin liquid with a spinon Fermi surface

High field thermodynamics

How to disentangle the effects due to impurities?



T. Han, et al, arXiv:1402.2693

T = 0.4 K
taken at NHMFL, LANL

Can “saturate” excess Cu impurities in fields > 10 Tesla
- weakly interacting with each other ($\theta_{CW} \approx -1.5$ K)
(but, cannot ignore coupling (~J/10) to intrinsic Cu moments)

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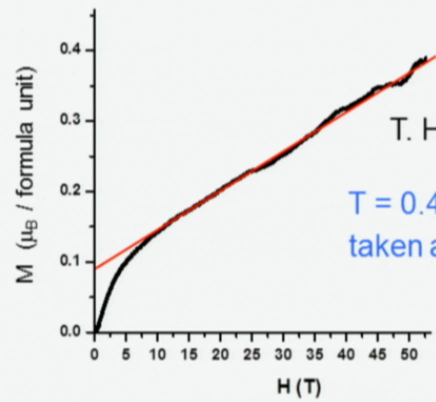

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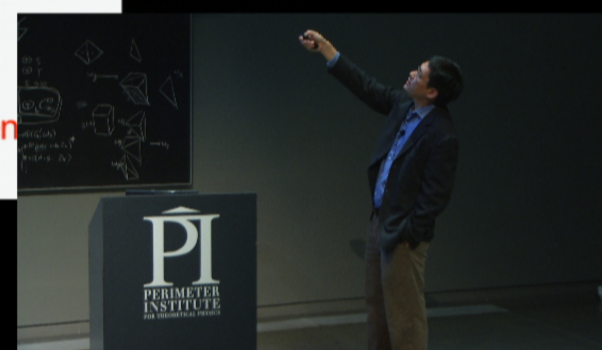
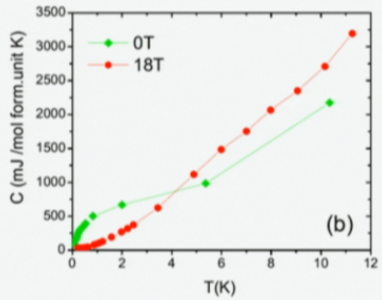
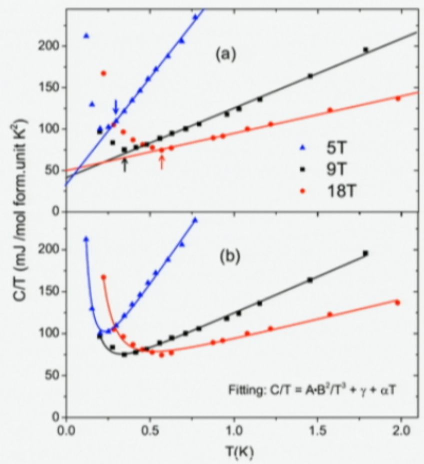


High field specific heat reveals substantial T-linear term





H = 9 T	H = 18 T
γ = 37	47 mJ/mol K²
α = 87	48 mJ/mol K³

→ comparable to γ seen in organic QSL's
yields Wilson ratio of ~6

- No evidence for gap at low energies (for H > 10 T)
- Can high field stabilize a spinon Fermi surface?

Exotic phase: the quantum spin liquid

Exotic-ness, part I: fractionalized excitations ✓

Exotic-ness, part II: the emergent gauge field

PRL **99**, 156402 (2007) PHYSICAL REVIEW LETTERS week ending 12 OCTOBER 2007

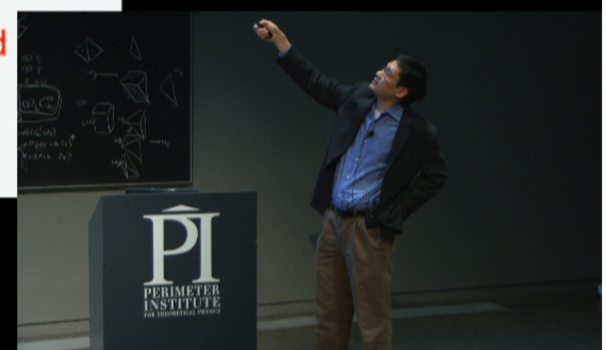
Power-Law Conductivity inside the Mott Gap: Application to κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

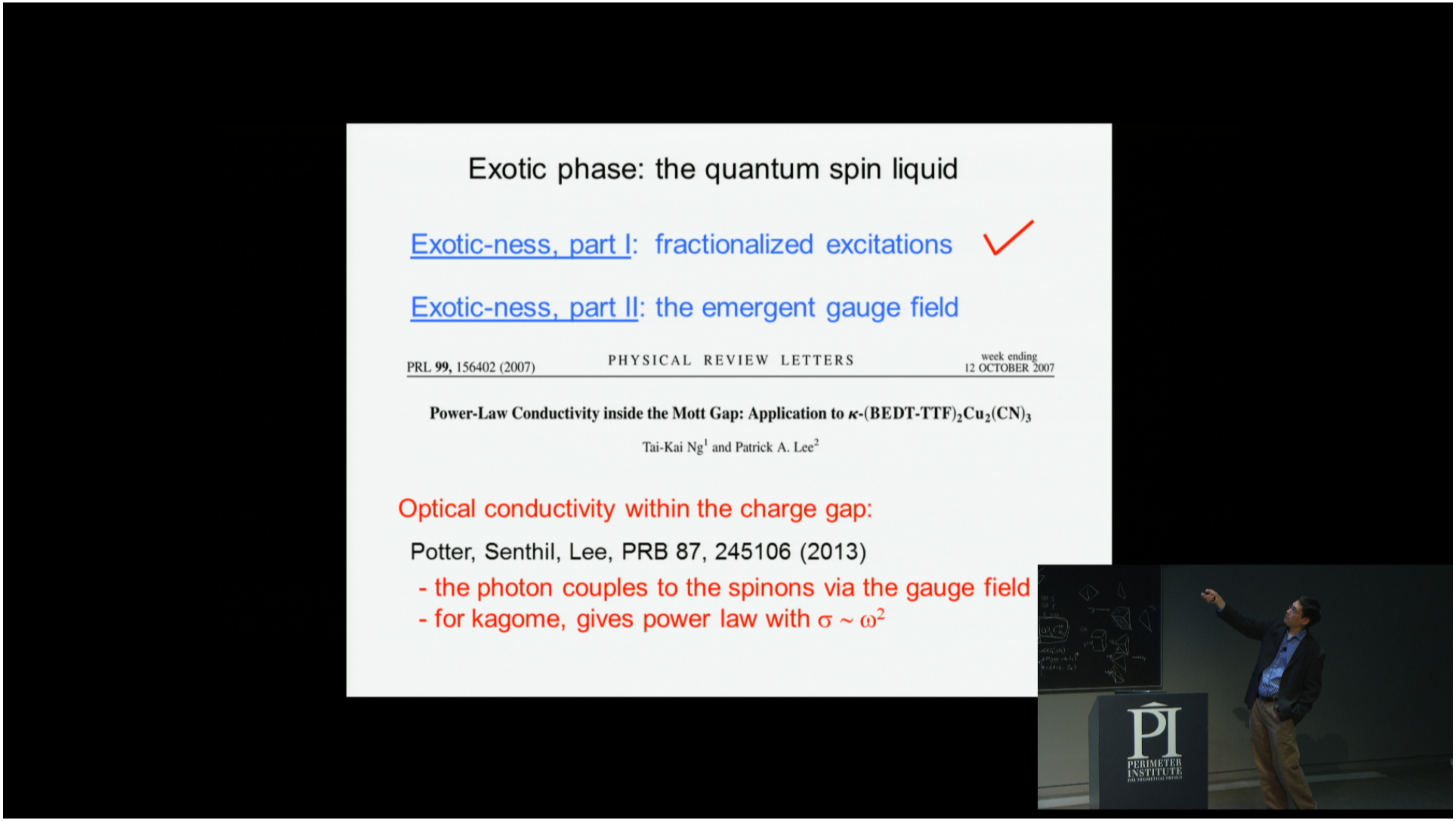
Tai-Kai Ng[1] and Patrick A. Lee[2]

Optical conductivity within the charge gap:

Potter, Senthil, Lee, PRB 87, 245106 (2013)

- the photon couples to the spinons via the gauge field
- for kagome, gives power law with $\sigma \sim \omega^2$

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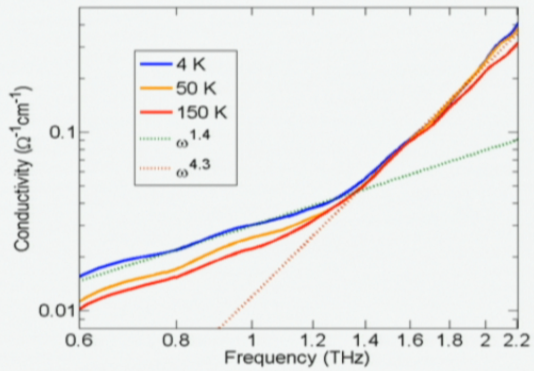
Optical conductivity measurements on our crystals

Pilon, Gedik, et al, PRL 111, 127401 (2013)



In-plane conductivity at low temperature:

low frequency response has power law with $\omega^{1.4}$

- consistent with a gapless spin liquid phase

The million dollar question:

Can we claim "discovery" of a quantum spin liquid in Herbertsmithite?

Among all of the "spin liquid" compounds, Herbertsmithite is special:

1) Kagome is theoretically well motivated
2) Hamiltonian (and impurities) are well characterized
3) Exotic spinon continuum observed (neutron $S(\boldsymbol{Q},\omega)$)
4) Need more theories that closely match the material
5) New probes to detect topological degeneracy and long-range entanglement

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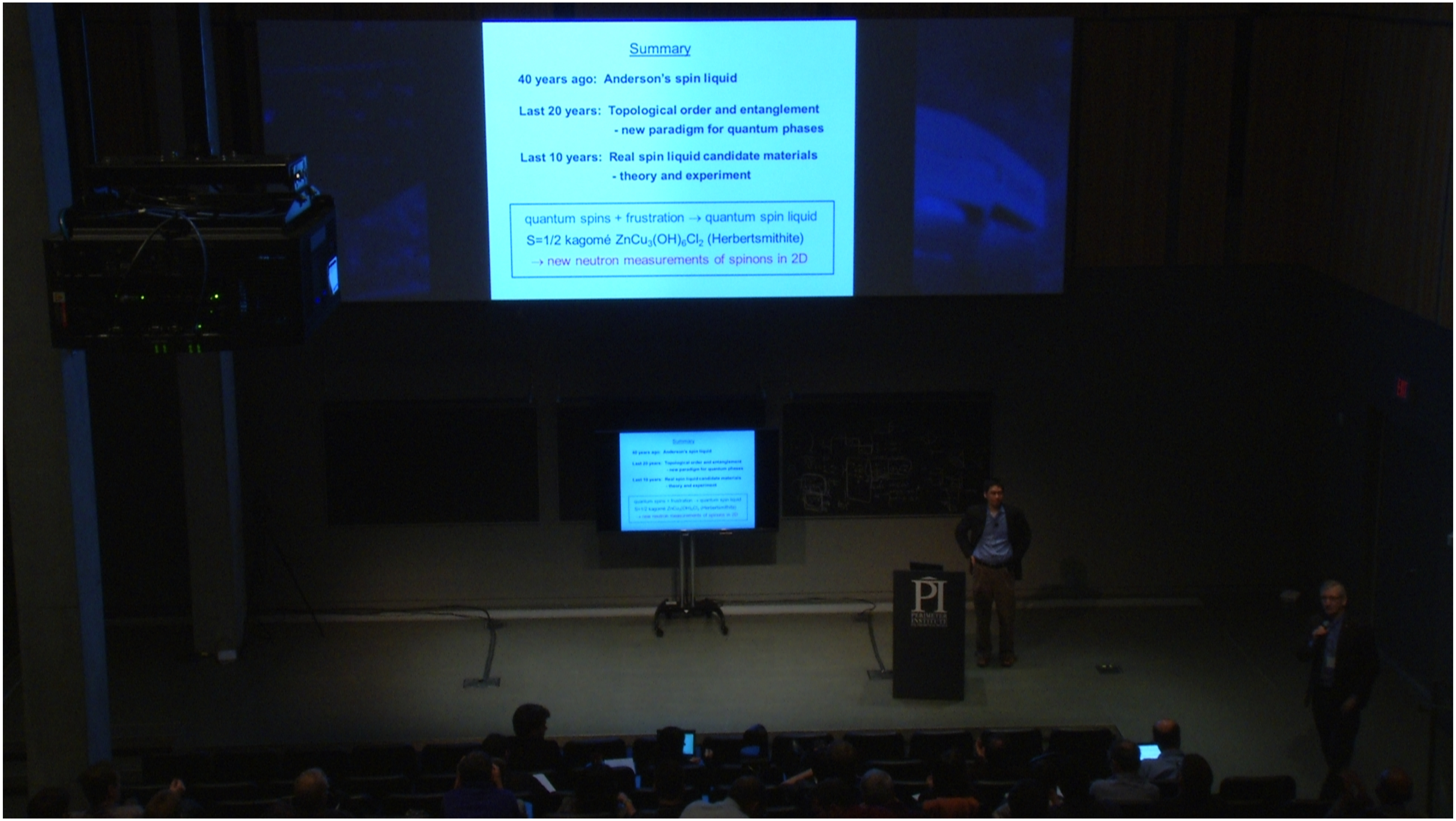
Summary

40 years ago: Anderson's spin liquid

Last 20 years: Topological order and entanglement
- new paradigm for quantum phases

Last 10 years: Real spin liquid candidate materials
- theory and experiment

quantum spins + frustration $\rightarrow$ quantum spin liquid
S=1/2 kagomé $ZnCu_3(OH)_6Cl_2$ (Herbertsmithite)
$\rightarrow$ new neutron measurements of spinons in 2D


Summary
40 years ago: Anderson's spin liquid
Last 20 years: Topological order and entanglement
- new paradigm for quantum phases
Last 10 years: Real spin liquid candidate materials
- theory and experiment
quantum spins + frustration → quantum spin liquid
S=1/2 kagomé ZnCu3(OH)6Cl2 (Herbertsmithite)
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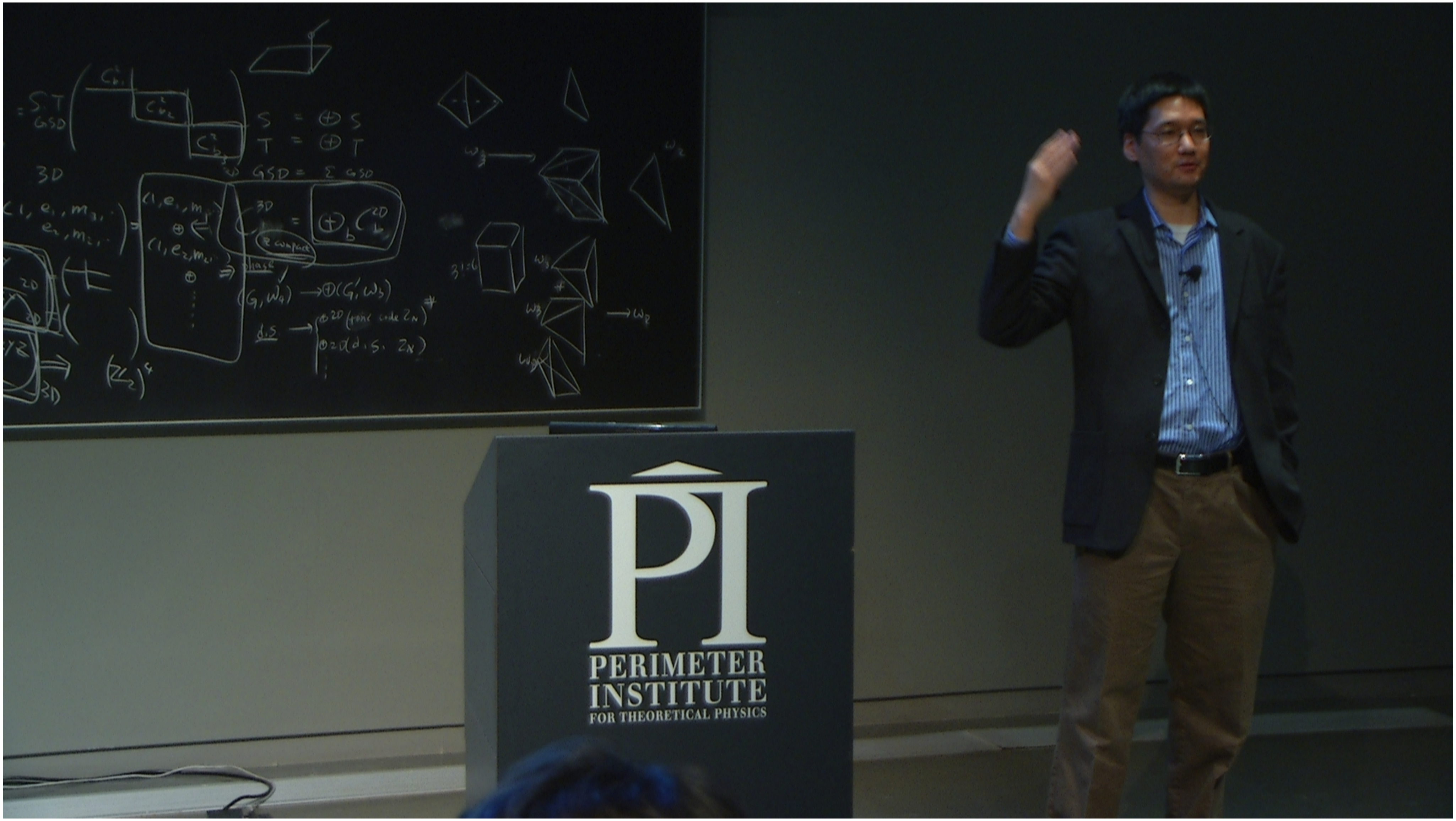
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