Title: Possible observation of photons and monopoles in the frustrated pyrochlore Yb2Ti2O7 and Pr2Zr2O7

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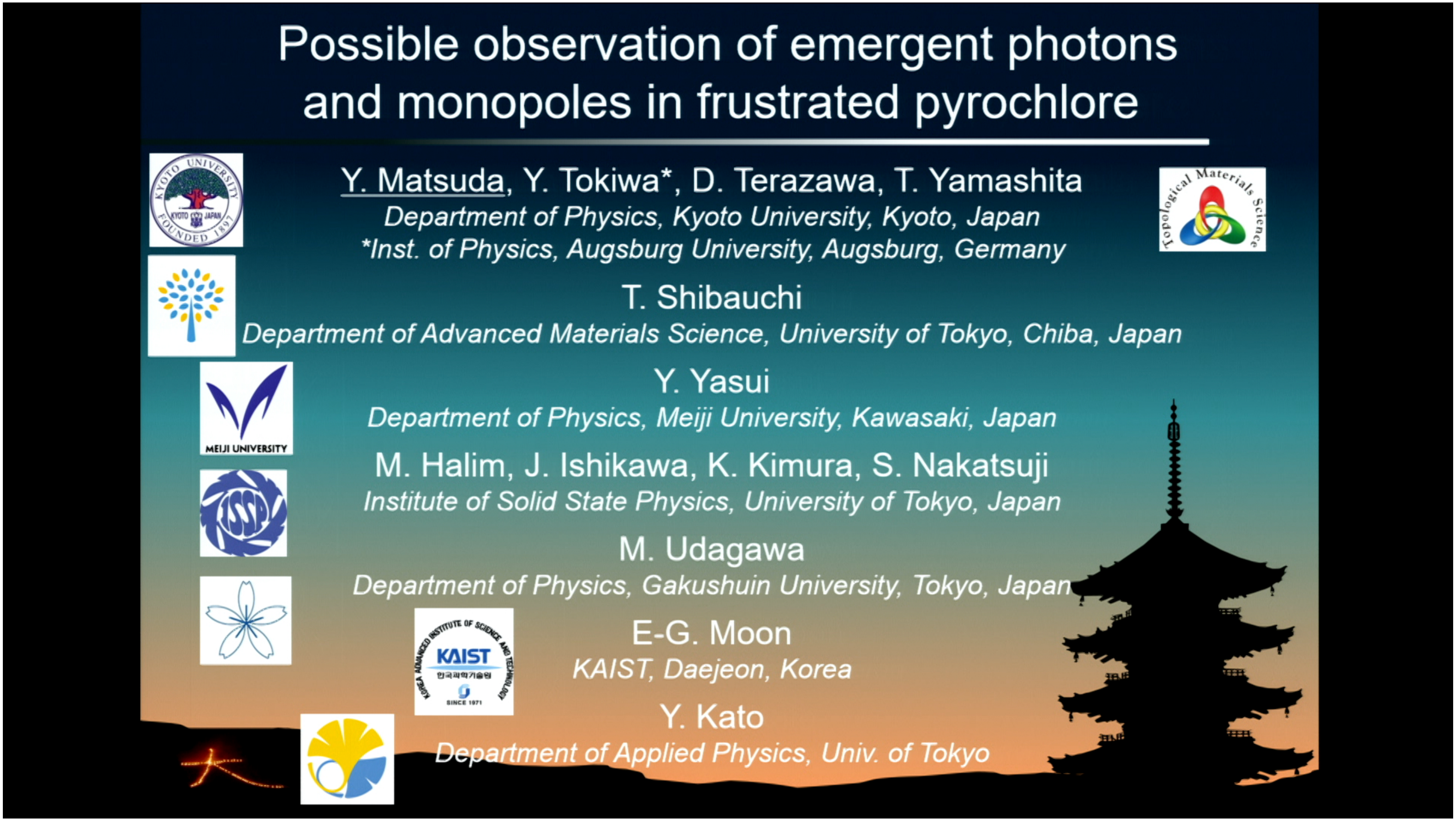
Abstract: We report highly unusual heat conduction generated by the spin degrees of freedom in spin liquid states of the pyrochlore magnets Yb2Ti2O7 and Pr2Zr2O7. In Yb2Ti2O7, the excitations propagate a long distance without being scattered, in contrast to the diffusive nature of classical monopoles. In Pr2Zr2O7, the thermal conductivity unexpectedly shows a dramatic enhancement at very low temperature. The low-lying excitations are discussed in terms of a possible emergent photons, coherent gapless spin excitations in a spin-ice manifold.



Possible observation of emergent photons and monopoles in frustrated pyrochlore

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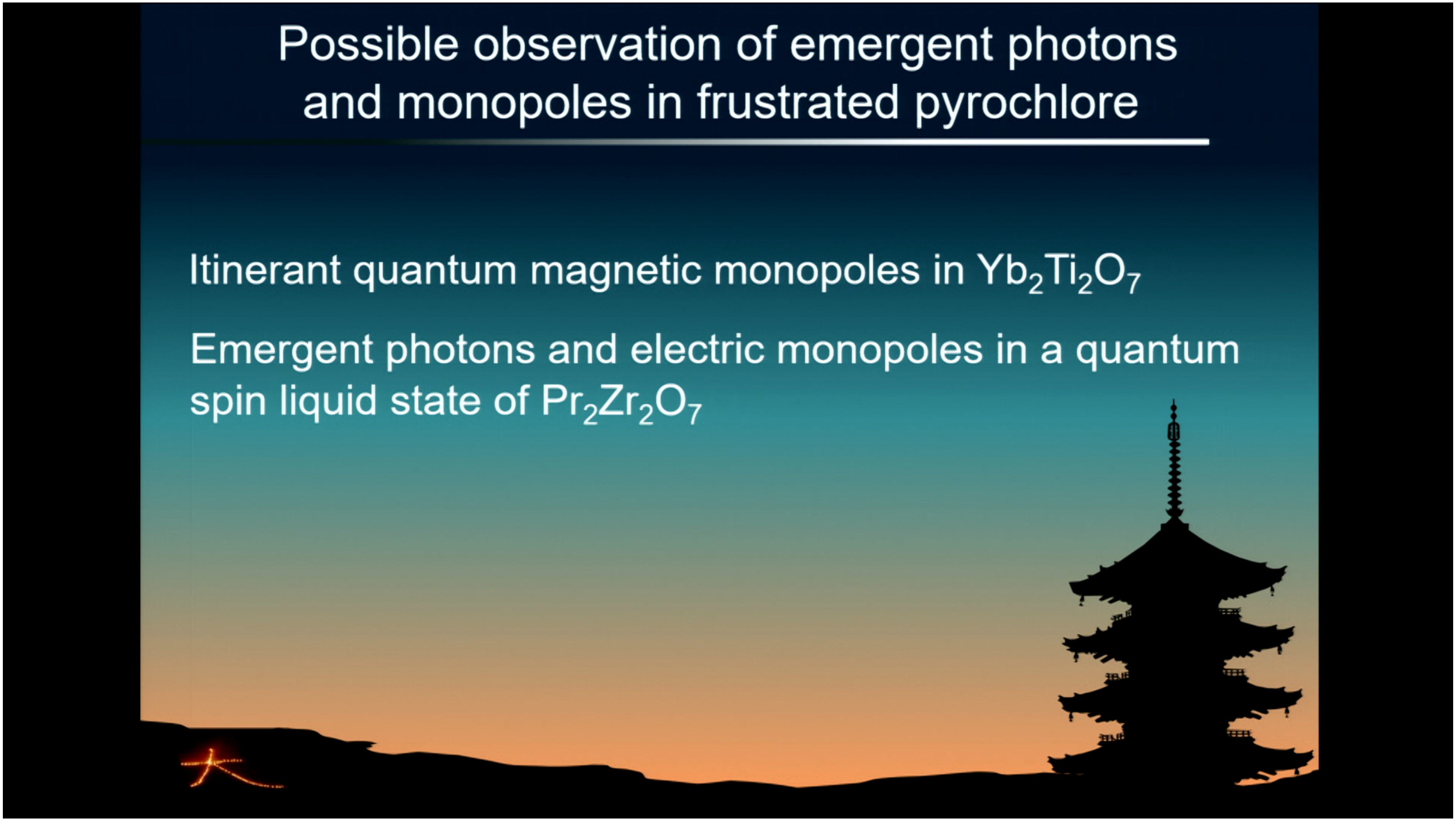


Possible observation of emergent photons and monopoles in frustrated pyrochlore

Itinerant quantum magnetic monopoles in $Yb_2Ti_2O_7$

Emergent photons and electric monopoles in a quantum spin liquid state of $Pr_2Zr_2O_7$

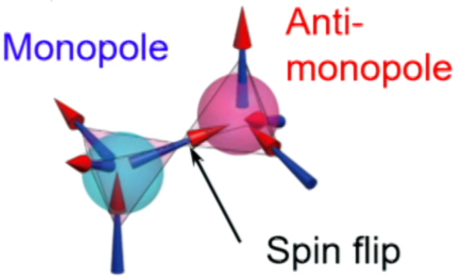
Elementary excitations from classical spin-ice

Classical spin ice   Ising term only

$Ho_2Ti_2O_7$, $Dy_2Ti_2O_7$

$$H = J_{zz} \sum_{<i,j>} S_i^z S_j^z \qquad J_{zz} > 0$$

Excitation: spin flip along the local easy axes



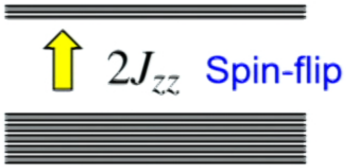
3-in 1-out
1-in 3-out
M-monopole pair

Magnetic monopoles

$$\nabla \cdot \boldsymbol{B} = \rho_m$$

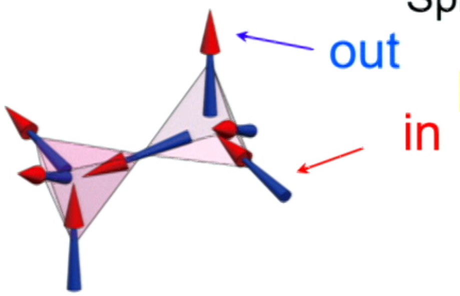
M-monopole pairs generate emergent ***B***-field

M-monopoles propagate diffusively

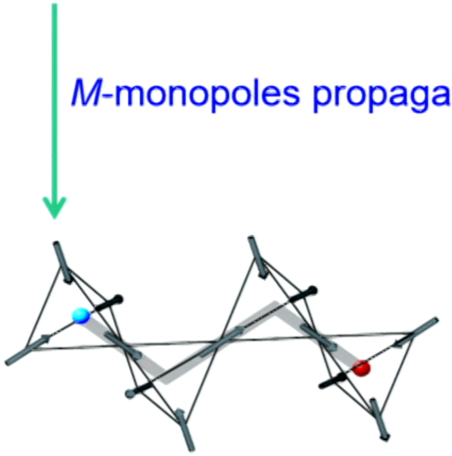


Ground state
2-in 2-out

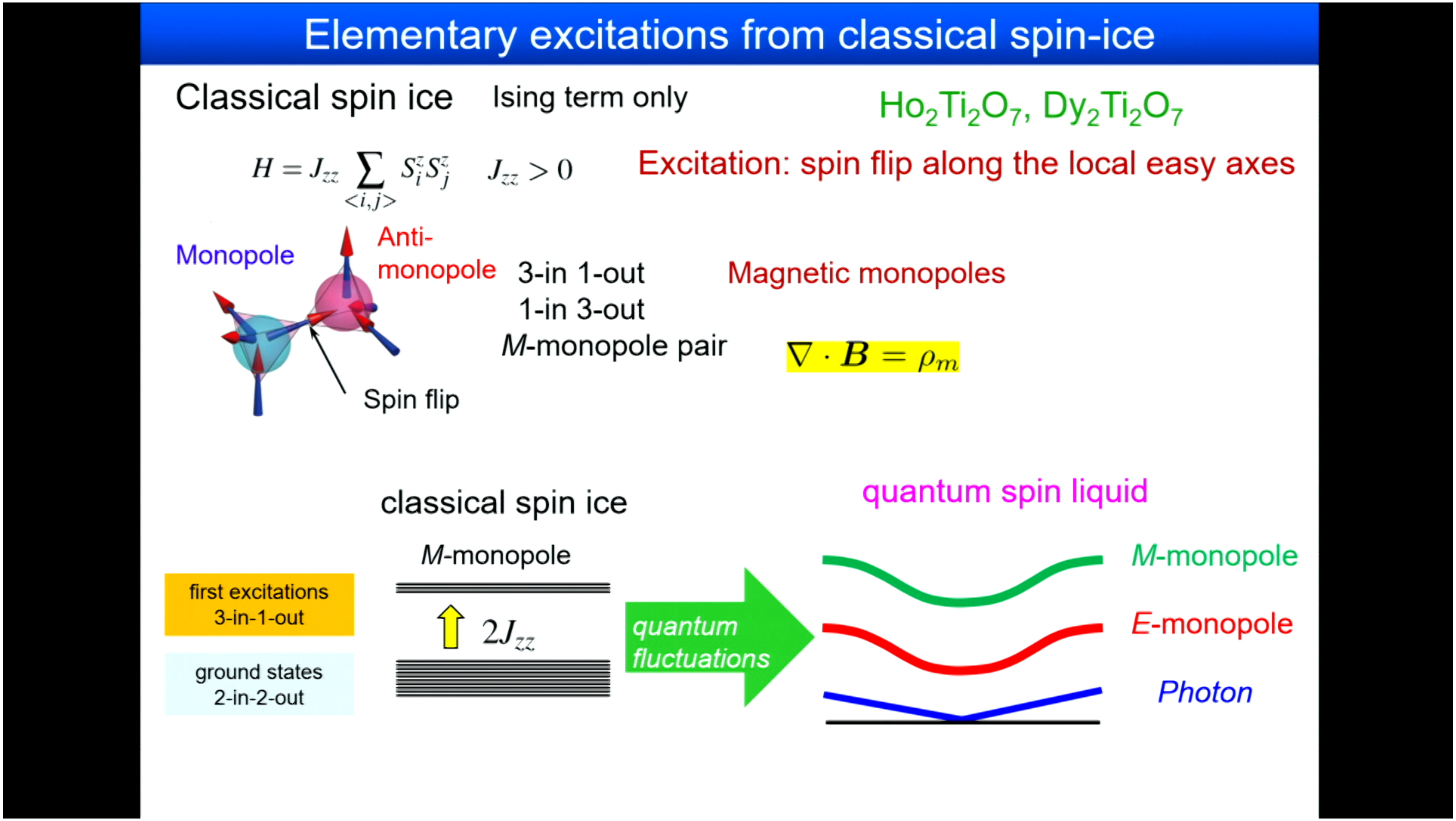
Spin-ice manifold



$$\nabla \cdot \boldsymbol{B} = 0$$



C. Castelnovo, R. Moessner and S. L. Sondhi, Nature **451**, 42-45 (2008).



Elementary excitations from classical spin-ice

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3-in 1-out
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M-monopole pair

Magnetic monopoles

$$\nabla \cdot \boldsymbol{B} = \rho_m$$

classical spin ice

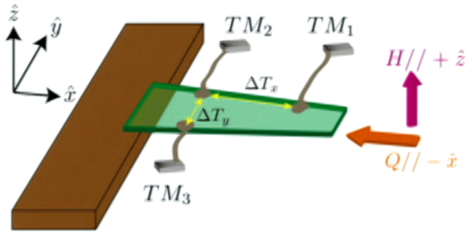
quantum spin liquid

first excitations
3-in-1-out

ground states
2-in-2-out

Thermal conductivity in a QSL state

Thermal conductivity



$$\mathbf{j}_{\mathrm{T}} = \kappa(-\nabla T)$$

$$\kappa = \begin{pmatrix} \kappa_{xx} & \kappa_{xy} \\ -\kappa_{xy} & \kappa_{xx} \end{pmatrix}$$

Thermal conductivity κ_{xx}

$$\kappa_{xx} = C v_s \ell$$

Thermal conductivity can sensitively probe low energy "*itinerant*" excitations at low temperature.

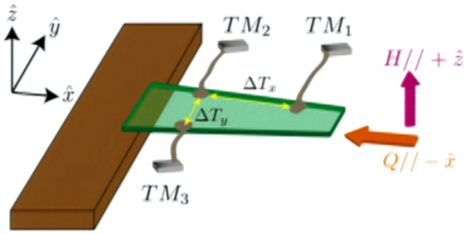
Not affected by localized impurities
Not contaminated by Schottky contribution

Quantum spin liquids often transport heat well.

Thermal conductivity in a QSL state

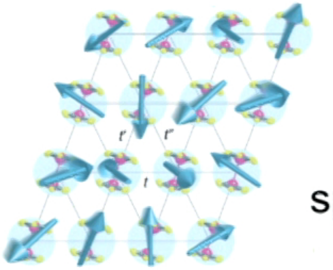
Thermal conductivity

$\mathbf{j}_{\mathrm{T}} = \kappa(-\nabla T)$



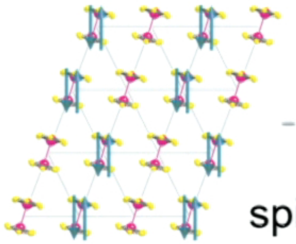
$\mathbf{j}_{\mathrm{T}} = \kappa(-\nabla T)$

$\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$ Spin liquid

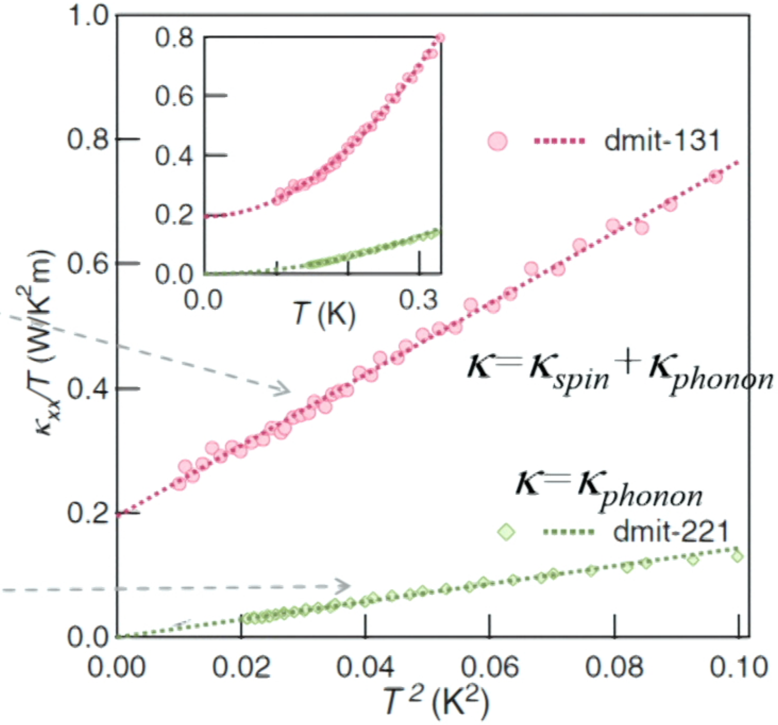


spin 1/2

$\mathrm{Et_2Me_2Sb[Pd(dmit)_2]_2}$ Charge order



spin 0

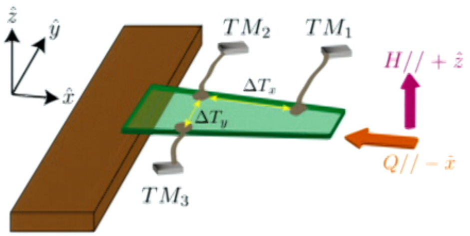


M. Yamashita *et al*. Science **328**, 1246 (2010)

Thermal conductivity in a QSL state

Thermal conductivity

$\mathbf{j}_{\mathrm{T}} = \kappa(-\nabla T)$



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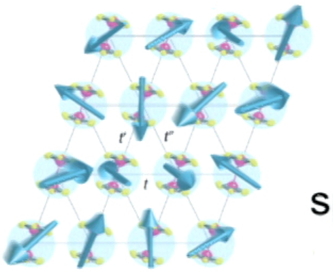
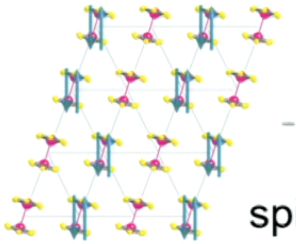
$\mathrm{EtMe_3Sb[Pd(dmit)_2]_2}$ Spin liquid

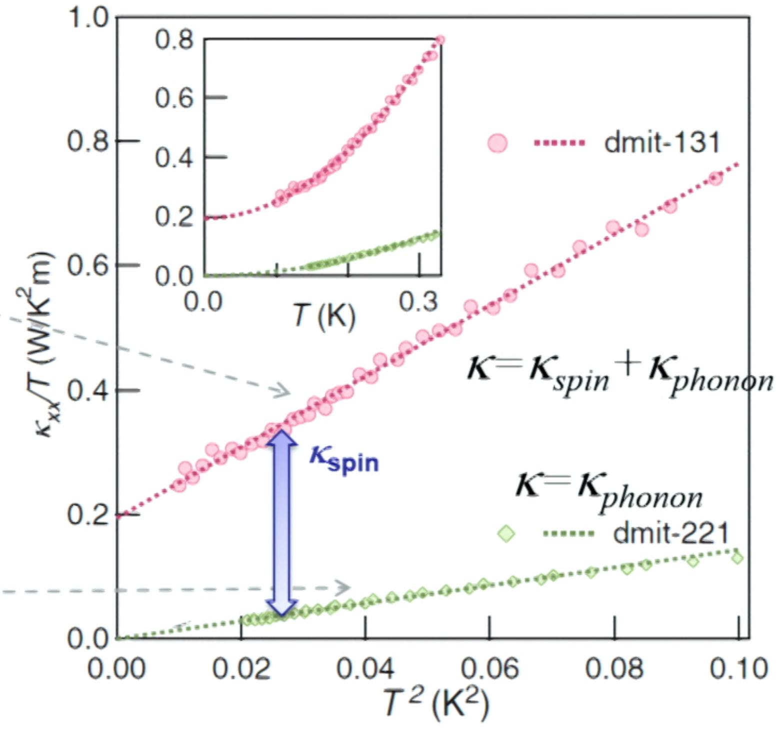


spin 1/2

$\mathrm{Et_2Me_2Sb[Pd(dmit)_2]_2}$ Charge order



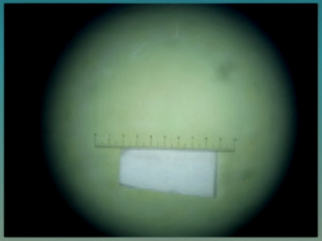
spin 0



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

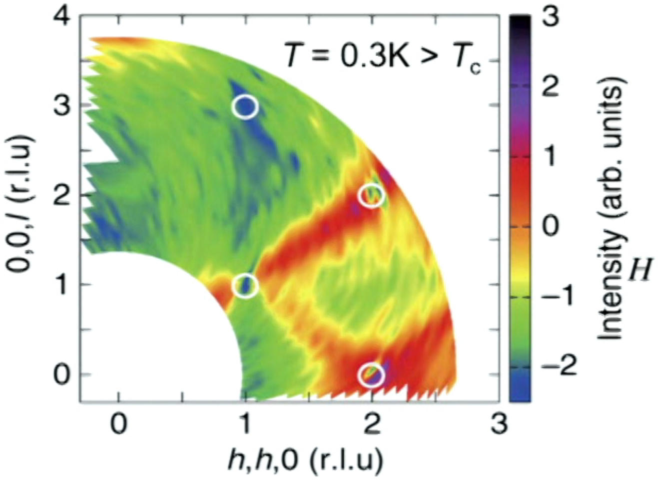
Possible observation of emergent photons and monopoles in frustrated pyrochlore

Itinerant magnetic monopoles in $Yb_2Ti_2O_7$



Emergent photons and monopoles in a quantum spin liquid state of $Pr_2Zr_2O_7$

Quantum spin ice state in $Yb_2Ti_2O_7$



pseudospin-1/2 of Yb ions

pinch points

➡ Spin ice (2-in-2-out) correlations

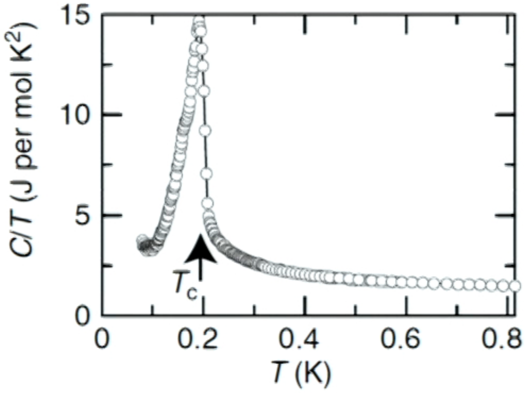
✓ disappear below T_c and above 4 K

L. –J. Chang *et al.* , Nature Commun. 3, 992 (12).

$$H = \sum_{<i,j>} \{J_{zz} S_i^z S_j^z - J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+) + J_{z\pm}[(S_i^z S_j^+ + S_i^z S_j^-) + (i \leftrightarrow j)]\}$$

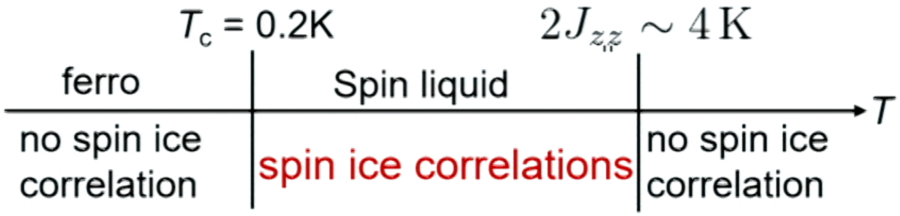
$J_{zz} \sim 2\,\mathrm{K}$	Ising	
$J_{\pm} \sim 0.58\,\mathrm{K}$	xy	Quantum fluctuations
$J_{z\pm} \sim 1.7\,\mathrm{K}$	off-diagonal	

K. Ross *et al.* Phy. Rev. X 1, 021002 (11)
J. Hodges *et al.* J. Phys Cond. Matter 13, 9301 (01)



Y. Tokiwa, Nature Commun (16)

- FM ordering below 0.2K (neutron)
- Magnetization along [100] with 1.1μ_B/Yb

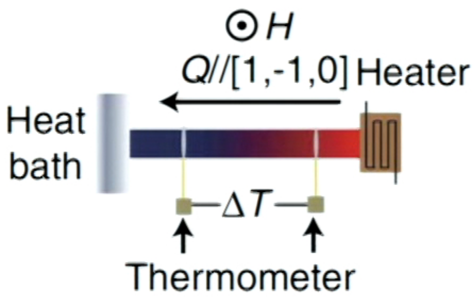


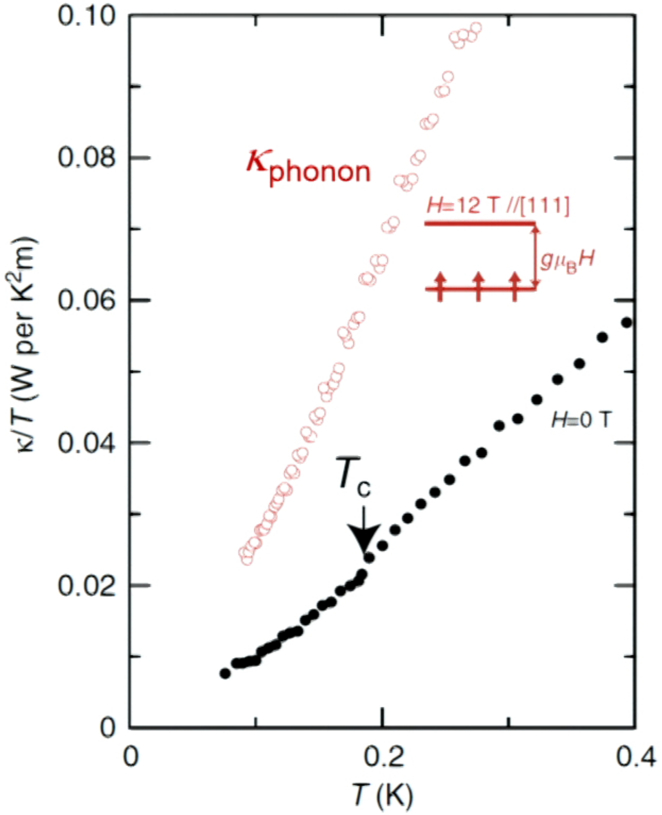
T_c = 0.2K

$2J_{zz} \sim 4\,\mathrm{K}$

ferro	Spin liquid	
no spin ice correlation	spin ice correlations	no spin ice correlation

Thermal conductivity in $Yb_2Ti_2O_7$



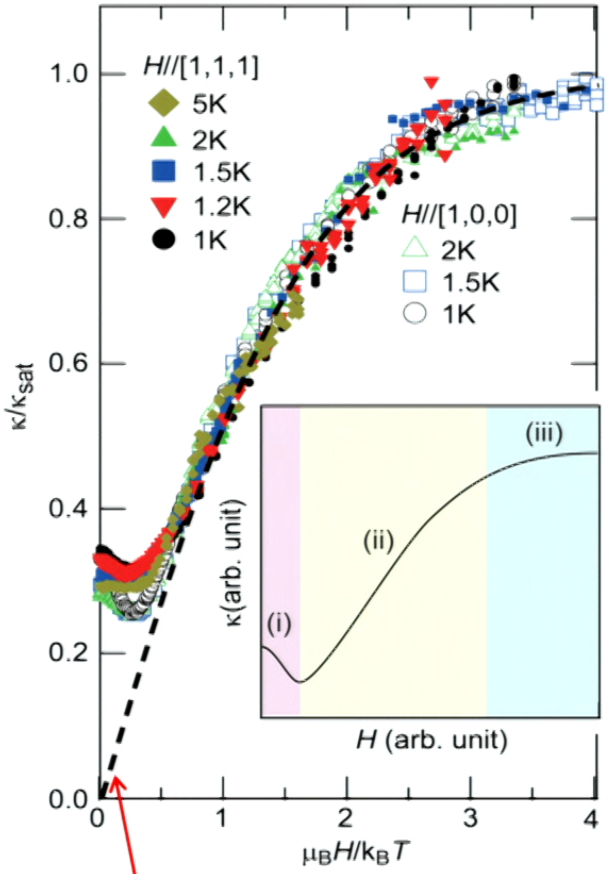


Y. Tokiwa, Nature Commun. (2016)

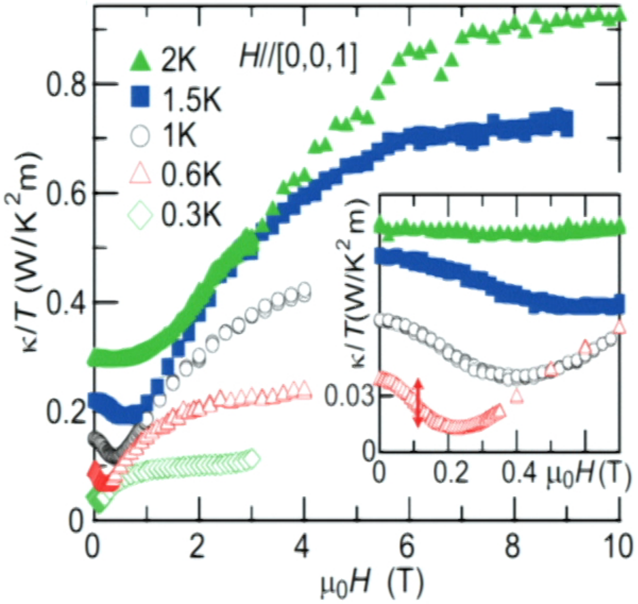
Enhancement of the thermal conductivity by magnetic field

Suppression of spin-phonon scattering by spin polarization

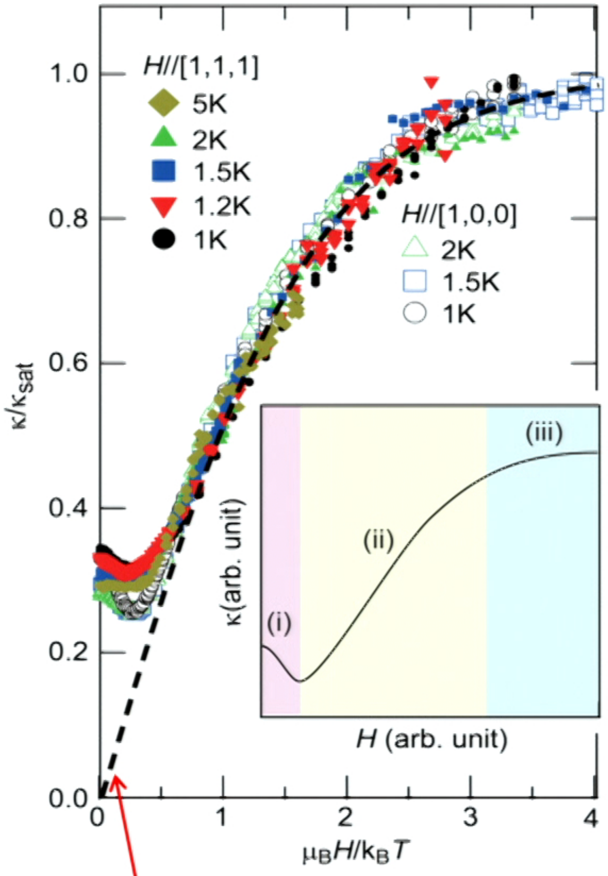
H-dependence of thermal conductivity in $Yb_2Ti_2O_7$



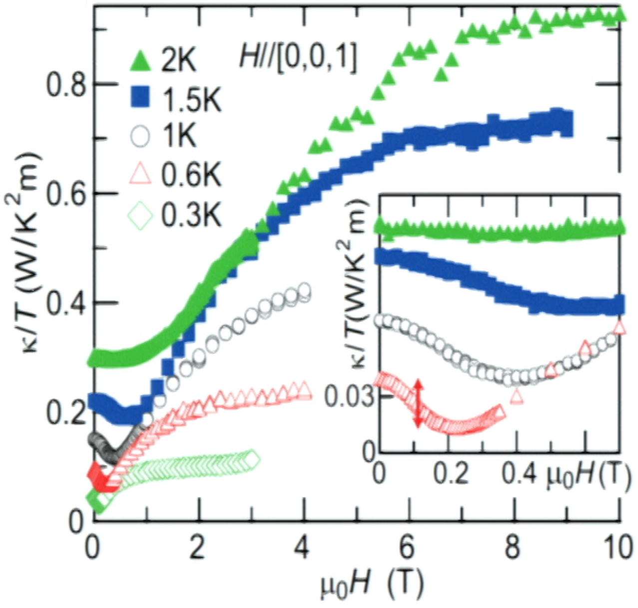
Brillouin function



H-dependence of thermal conductivity in $Yb_2Ti_2O_7$

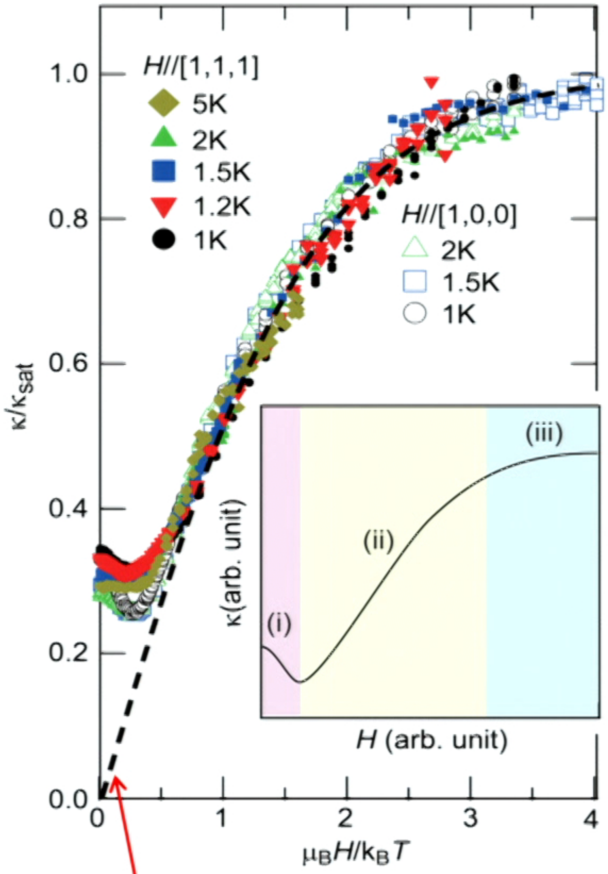


Brillouin function



H-dependence of thermal conductivity in $Yb_2Ti_2O_7$

(ii) and (iii) regimes



Brillouin function

Thermal conductivity is well scaled by *H/T*

→ • Thermally excited paramagnetic spins are important

Regime (iii) $g\mu_B H \gg J_{zz}, k_B T$

Perfect alignment of spins

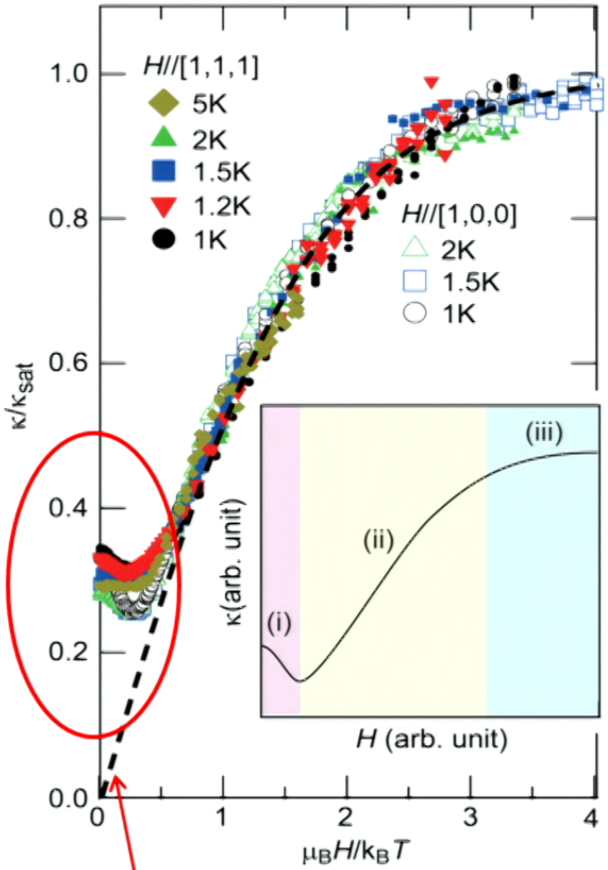
→ purely *phononic*, *H*-independent

Regime (ii) $g\mu_B H \sim k_B T$

Phonons are scattered by *thermally excited spins*

Regime (ii) and (iii) explained by phonon thermal conductivity

H-dependence of thermal conductivity in $Yb_2Ti_2O_7$



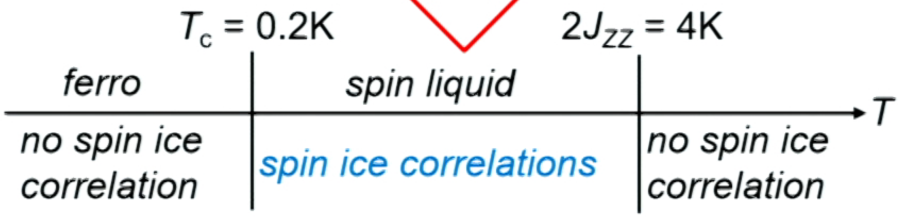
Brillouin function

Regime (i)

Spin-phonon scattering suppressed with H

→Initial reduction cannot be explained by phonon contribution

The initial reduction appears only in the regime of spin ice correlations



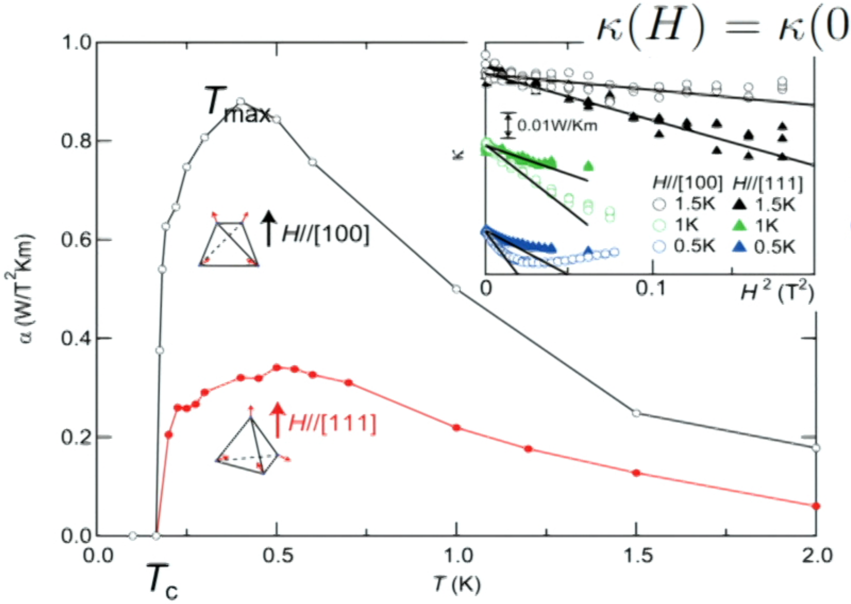
Reduction of κ at low field

most likely magnetic monopole excitations

Suppression of monopole thermal conductivity by H in $Yb_2Ti_2O_7$

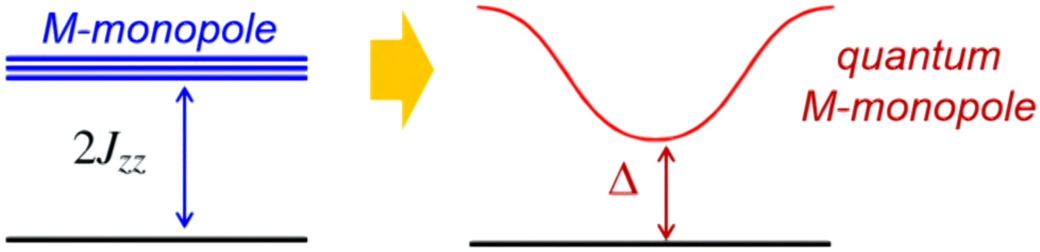
$$\kappa(H) = \kappa(0) - \alpha H^2 \ (\alpha > 0)$$

measure of the suppression of *M*-monopoles



Y. Tokiwa, Nature Commun. (2016)

Maximum at T_{max}~0.5K→ T_{max}~*M*-particle excitation energy $\Delta \ (< 2J_{zz} = 4\,\mathrm{K})$



Classical

M-monopole

$2J_{zz}$

quantum

M-monopole

Δ

$\sim 2J_{z\pm}$

Mean free path of *M*-monopole

~200 times distance of neighboring tetrahedra

M-monopole excitations disappear at T_c

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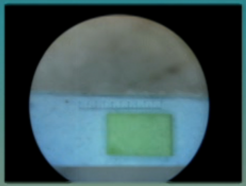


Y. Tokiwa *et al*. a preprint.

Emergent photon

M. Hermele, M.P.A. Fisher and L. Balents, Phys. Rev. B **69**, 064404 (04).
O. Benton, O. Sikora and N. Shannon, Phys Rev. B 86, 075154 (12).
M. J. P. Gingras and P.A. McCarty, Rep. Prog. Phys. 77, 056501 (14).

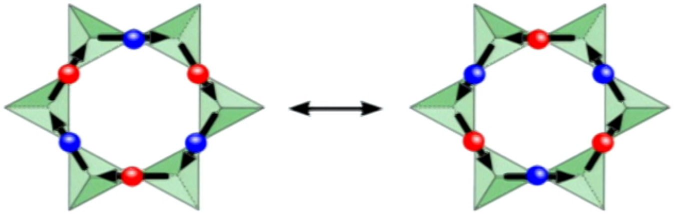
Quantum fluctuations

$$H = \sum_{<i,j>} \{ \underline{J_{zz} S_i^z S_j^z} + \underline{J_\pm (S_i^+ S_j^- + S_i^- S_j^+)} \}$$

Ising

xy Quantum fluctuations

$Yb_2Ti_2O_7$
$Pr_2Zr_2O_7$
$Tb_2Ti_2O_7$



$$H_p = -g \sum_{\hexagon} (S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- + h.c.) \quad g = \frac{12 J_\pm^3}{J_{zz}^2},$$

Quantum fluctuations lift the degeneracy of classical spin ice

⇨ **Quantum spin liquid**

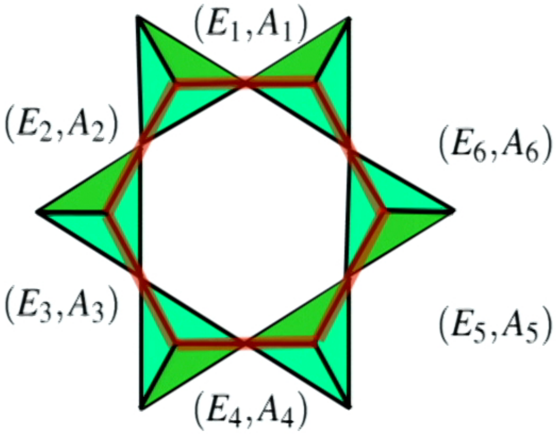
Electromagnetism spin

$E \longleftrightarrow S^z$ Ising

$A \longleftrightarrow S^\pm$ XY

$[E, A] = i\hbar \qquad S^\pm \propto e^{\pm iA} \longrightarrow [S^z, S^\pm] = \pm S^\pm$

$$(\nabla_{\hexagon} \times A)_{\mathbf{rr'}} = \sum_{i=1}^{6} A_i = B$$

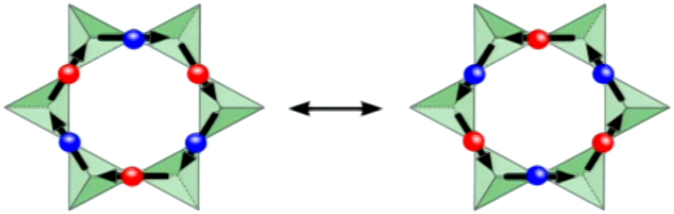


Quantum fluctuations

$$H = \sum_{<i,j>} \{ \underbrace{J_{zz} S_i^z S_j^z}_{\text{Ising}} + J_{\pm} \underbrace{(S_i^+ S_j^- + S_i^- S_j^+)}_{\text{xy}} \}$$

Quantum fluctuations

$Yb_2Ti_2O_7$
$Pr_2Zr_2O_7$
$Tb_2Ti_2O_7$



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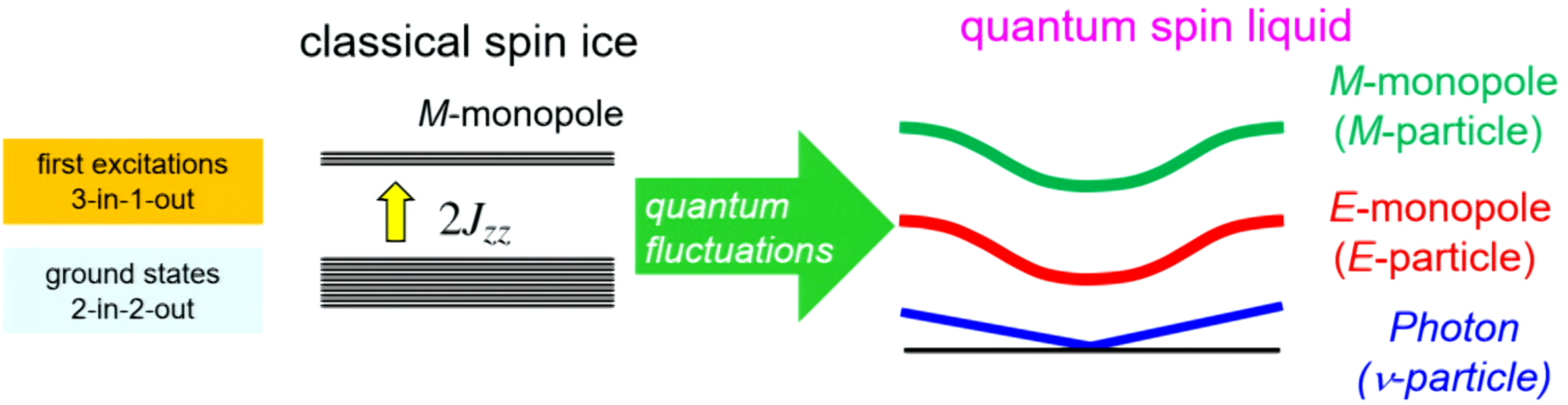
Quantum fluctuations lift the degeneracy of classical spin ice ⇨ **Quantum spin liquid**



classical spin ice

quantum spin liquid

first excitations
3-in-1-out

ground states
2-in-2-out

M-monopole

$2J_{zz}$

quantum fluctuations

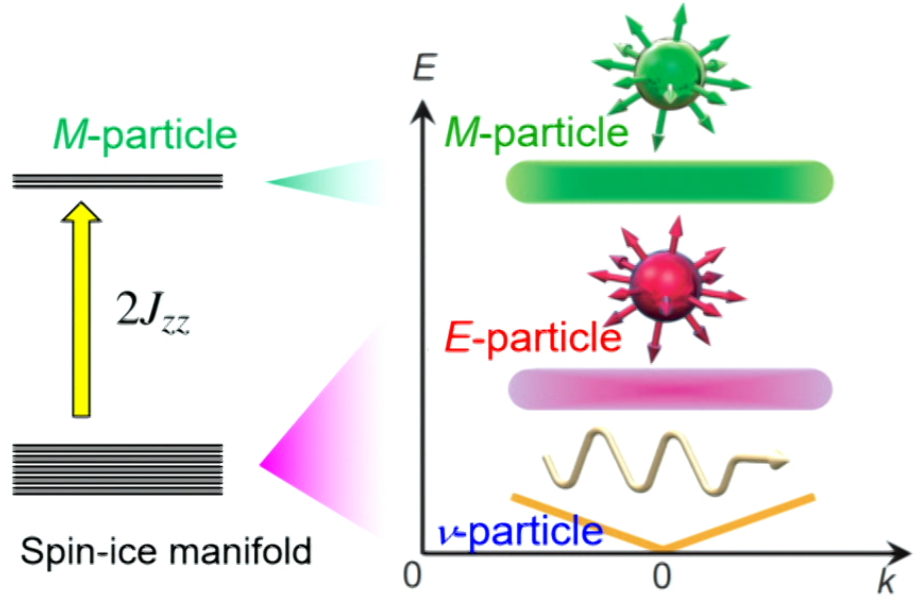
M-monopole
(*M*-particle)

E-monopole
(*E*-particle)

Photon
(ν-particle)

Elementary excitations in a quantum spin liquid

$$H = \sum_{<i,j>} \{J_{zz} S_i^z S_j^z + J_{\pm}(S_i^+ S_j^- + S_i^- S_j^+)\}$$



classical spin ice

Quantum spin liquid

M-pairs (creation of ***B***-field)

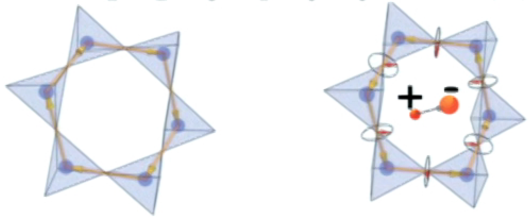
Spin flip along the local easy axes

E-pairs (creation of ***E***-field)

Spin excitations perp. to local easy axes

Topological defect

$$S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- \propto \cos(\mathcal{E})$$

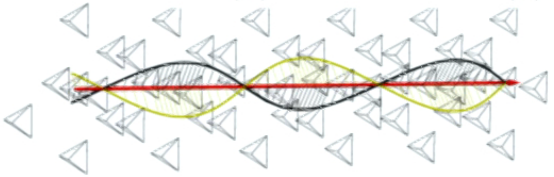


$\nu-$ particle (gapless linear excitation)

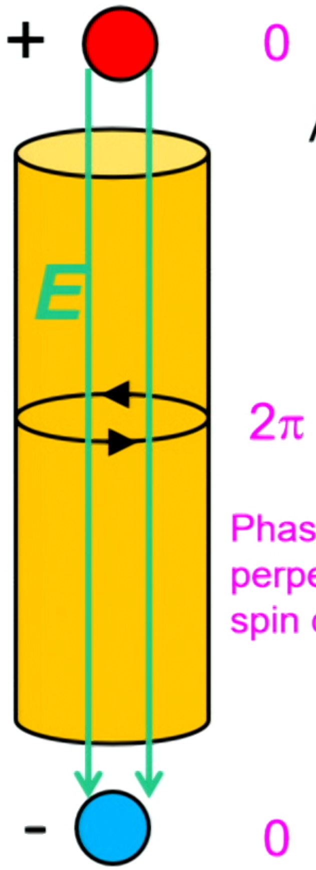
mediates dynamical interaction

between *E*- and *M*- particles

$$\mathcal{H}_{\mathrm{U(1)}} = \frac{\mathcal{U}}{2} \sum_{\langle \mathbf{rr}' \rangle} [(\nabla_{\bigcirc} \times \mathcal{A})_{\mathbf{rr}'}]^2 + \frac{\mathcal{K}}{2} \sum_{\langle \mathbf{ss}' \rangle} \mathcal{E}_{\mathbf{ss}'}^2$$

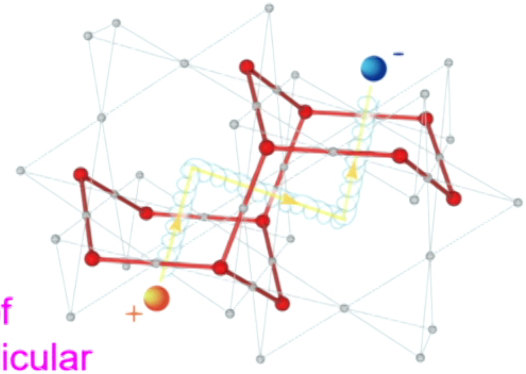


Elementary excitations in a quantum spin liquid



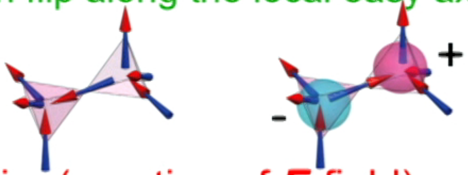
Phase of perpendicular spin component

A pair of *topological defects*,
E-particles



M-pairs (creation of ***B***-field)

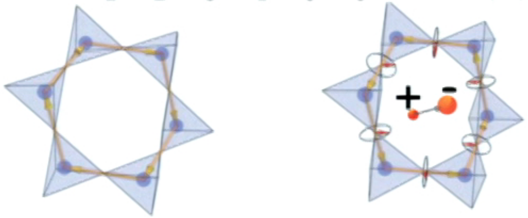
Spin flip along the local easy axes



E-pairs (creation of ***E***-field)

Spin excitations perp. to local easy axes
Topological defect

$$S_1^+ S_2^- S_3^+ S_4^- S_5^+ S_6^- \propto \cos(\mathcal{E})$$



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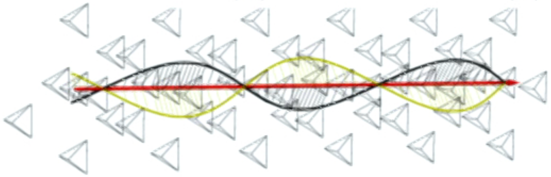
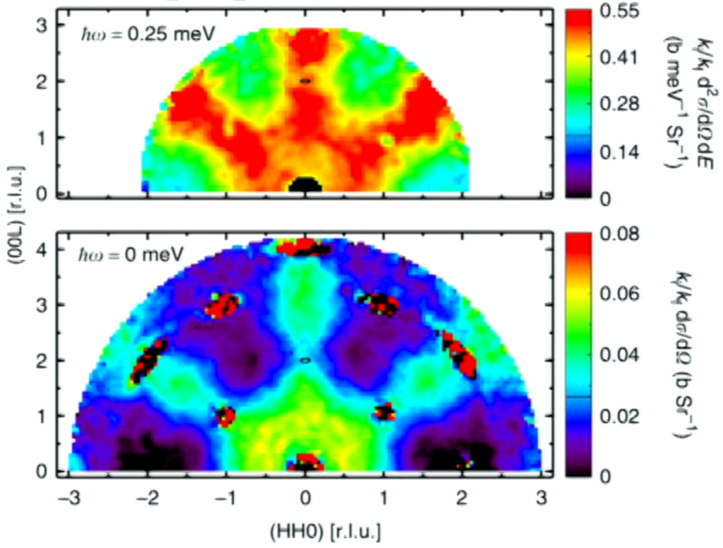
between *E*- and *M*- particles

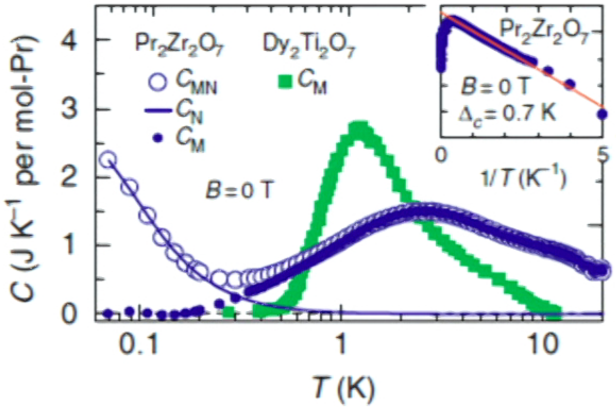
$$\mathcal{H}_{\mathsf{U(1)}} = \frac{\mathcal{U}}{2}\sum_{\langle \mathbf{rr'}\rangle}[(\nabla_{\circ} \times \mathcal{A})_{\mathbf{rr'}}]^2 + \frac{\mathcal{K}}{2}\sum_{\langle \mathbf{ss'}\rangle}\mathcal{E}_{\mathbf{ss'}}^2$$



$Pr_2Zr_2O_7$

$Pr_2Zr_2O_7$ No magnetic order down to 50 mK (NMR)





K. Kimura *et al.*, Nature Commun. **4**, 1934 (2013).

Neutron scattering

Pinch points below 1.6 K

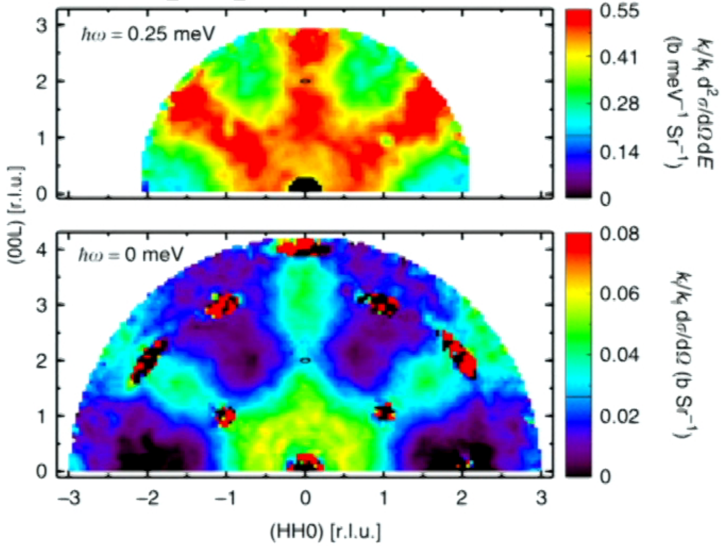
⇨ Spin ice (2-in-2-out) correlations

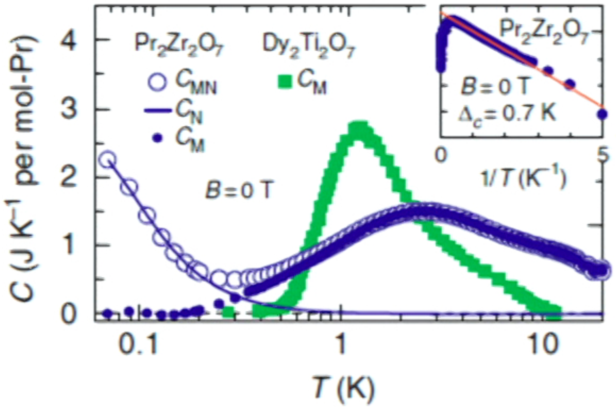
Inelastic neutron: over 90% weight

⇨ Strong quantum fluctuations

$Pr_2Zr_2O_7$

$Pr_2Zr_2O_7$ No magnetic order down to 50 mK (NMR)





K. Kimura *et al.*, Nature Commun. **4**, 1934 (2013).

Pr^{3+} non-Kramers doublet

$$\mathcal{H} = \sum_{\langle i,j \rangle} \left[J_{zz} S_i^z S_j^z + J_\perp (S_i^x S_j^x + S_i^y S_j^y) \right]$$

Specific heat

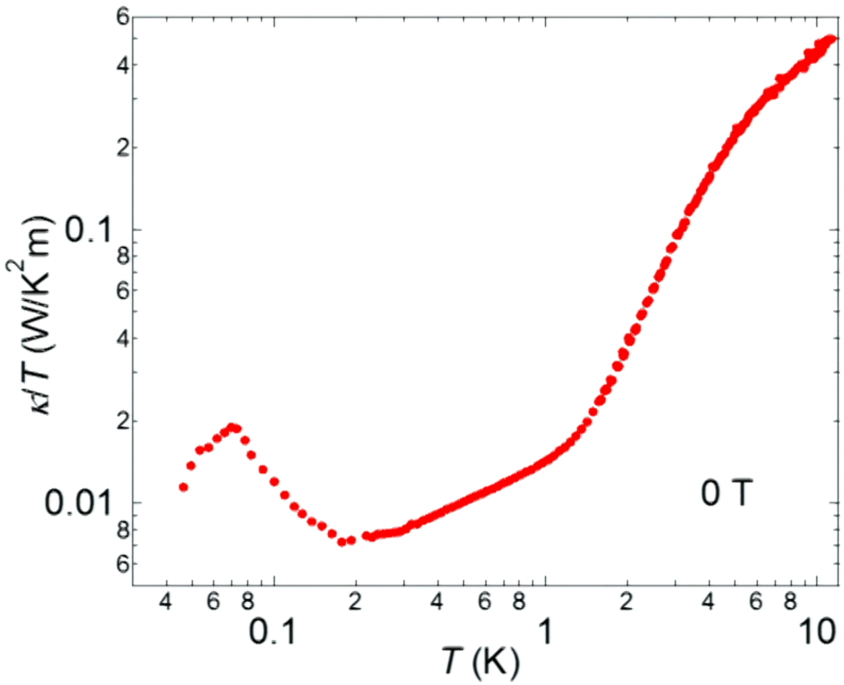
$J_{zz} \sim 7\,\mathrm{K}$ and $J_\perp \sim 0.4 J_{zz}$

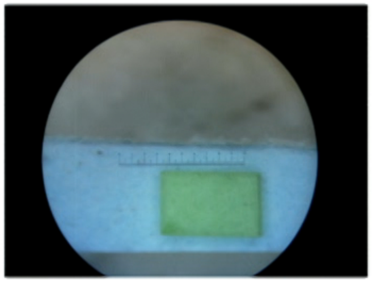
⇨ *M*-particles Y. Kato and S. Onoda PRL **115**, 077202 (15)

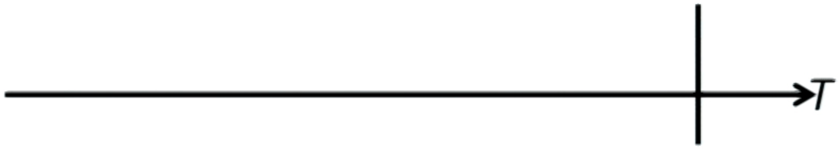
Large nuclear contribution at low T

difficulty in detecting *E*- and ν- particles

Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$







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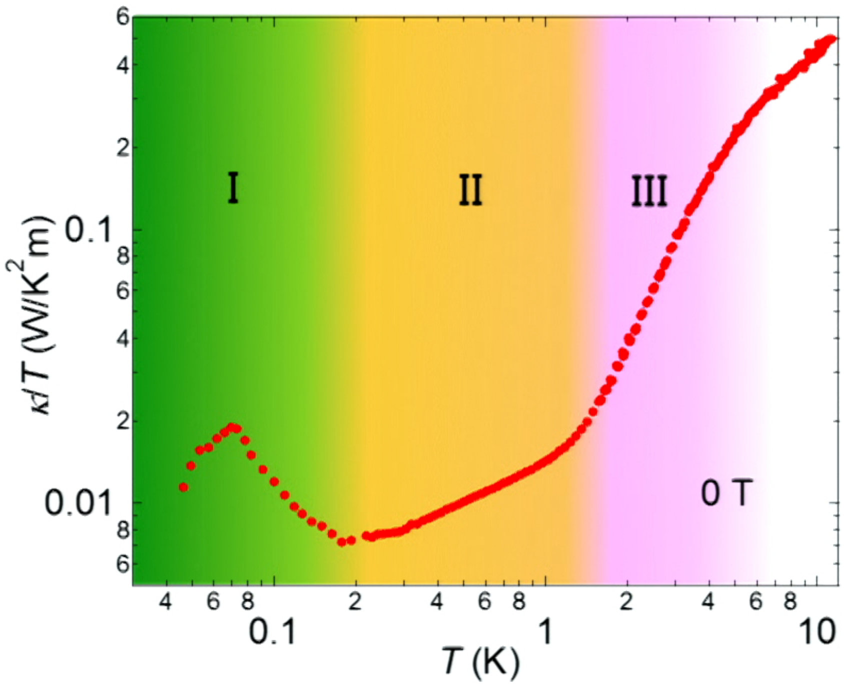

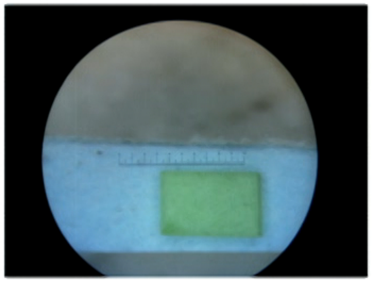

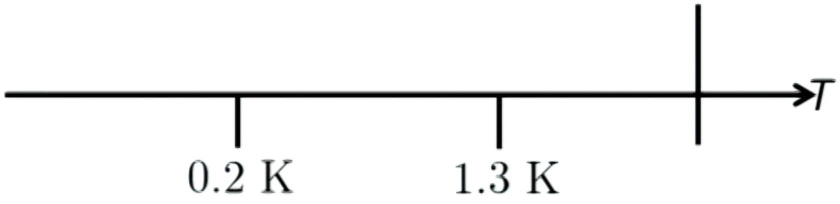

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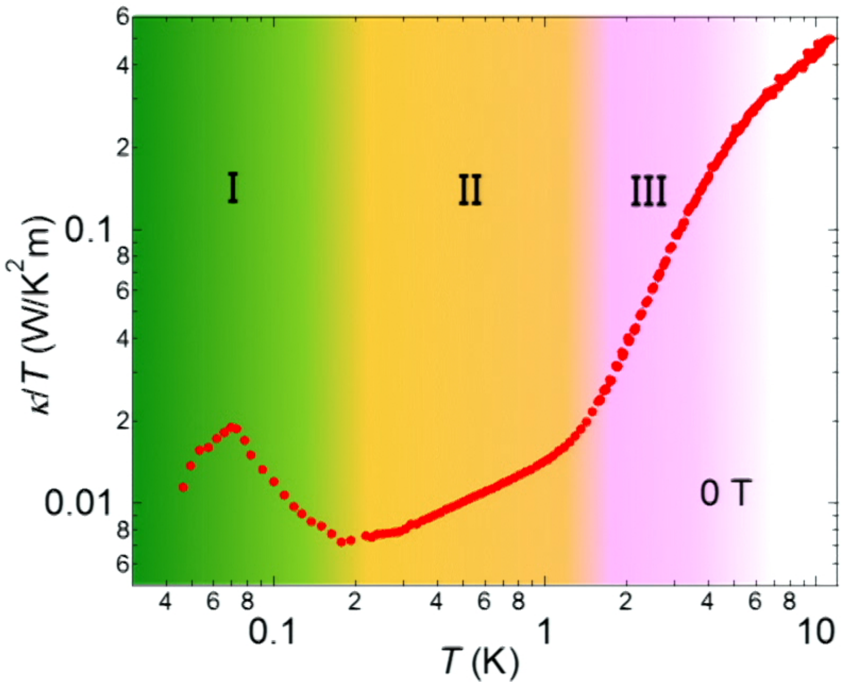

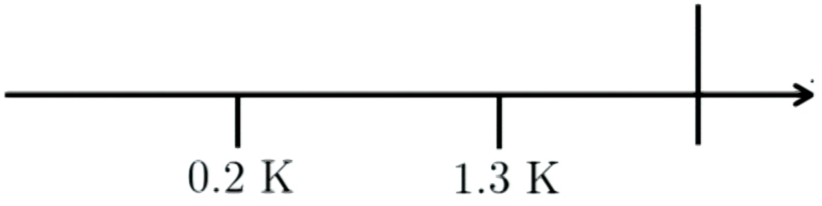

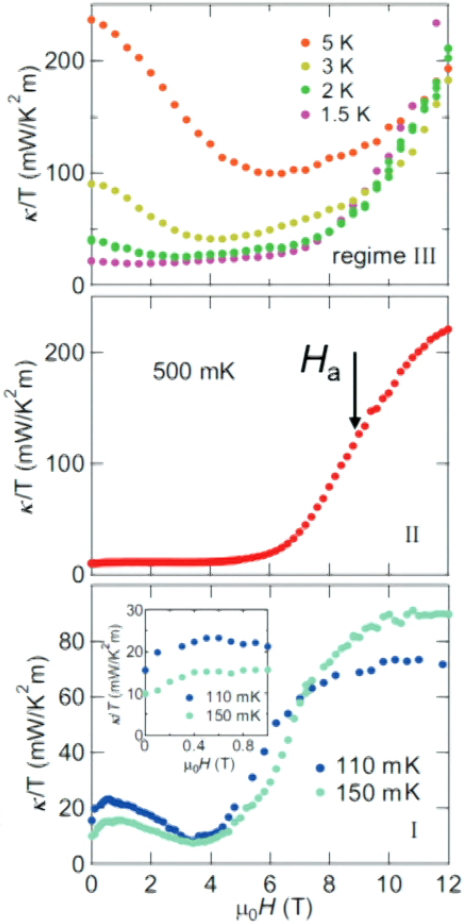

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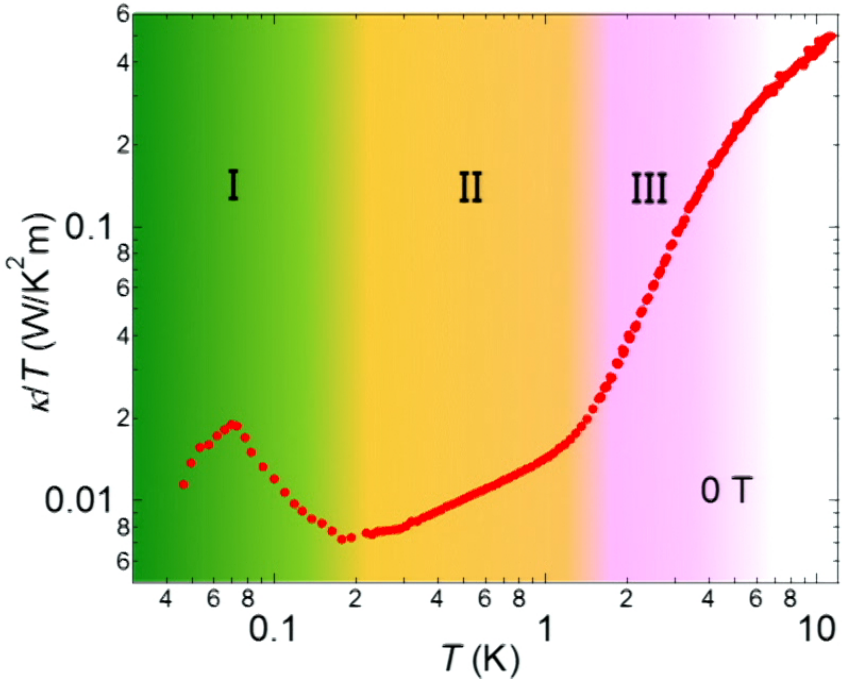
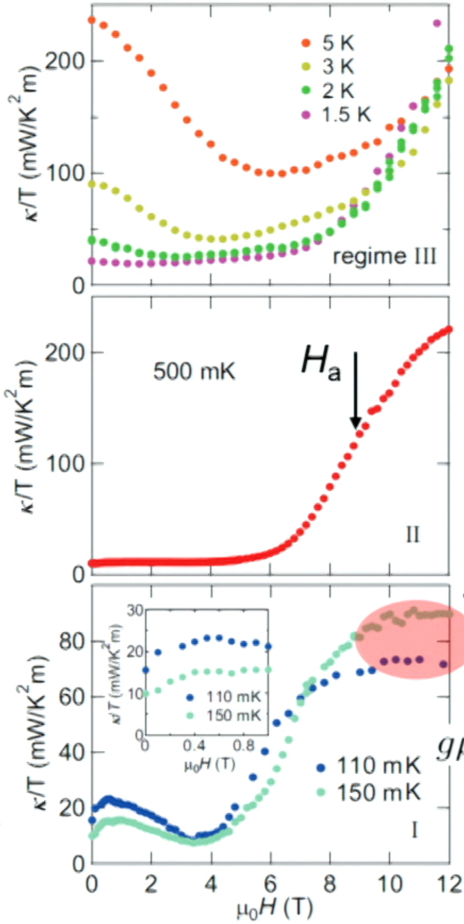
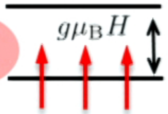


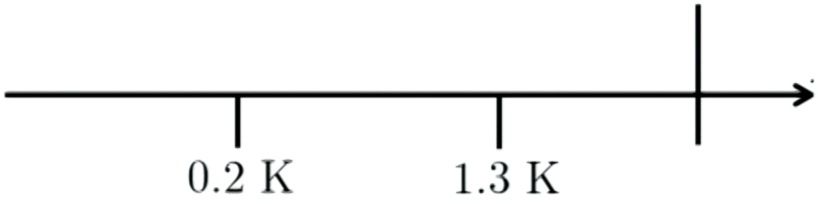




$g\mu_B H \gg J_{\parallel},$

$k_B T$

purely *phononic*



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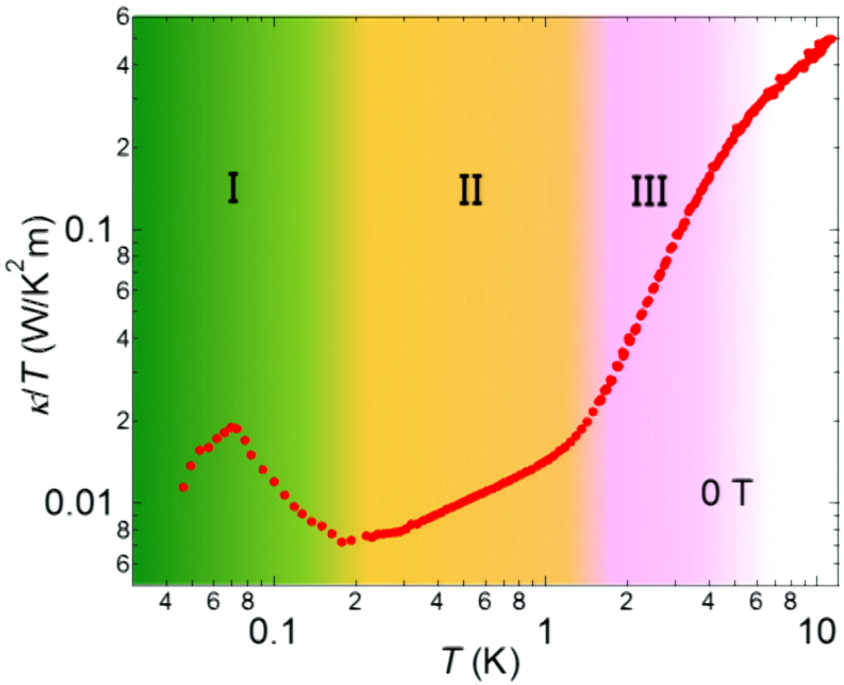
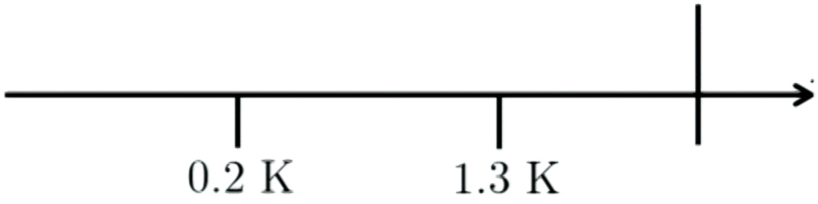
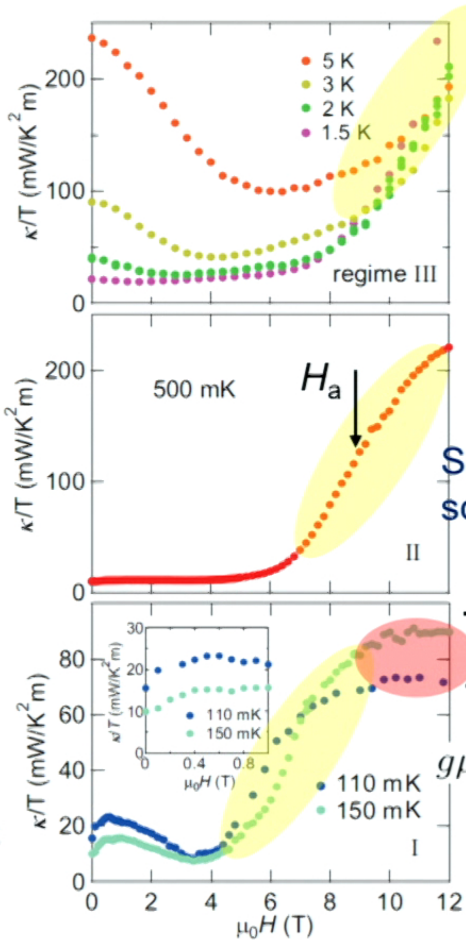
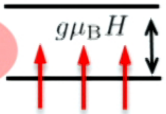






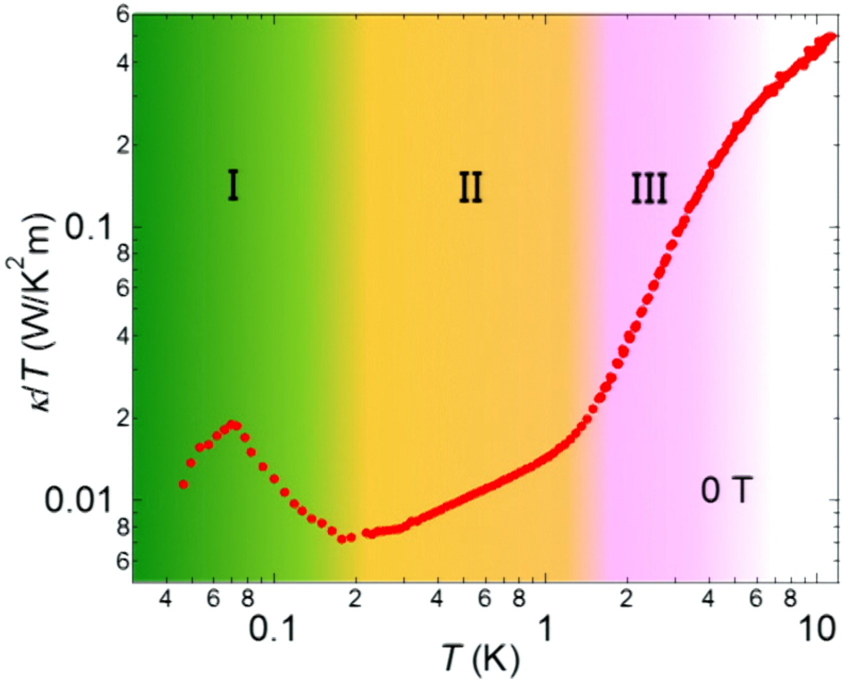
Spin-phonon scattering

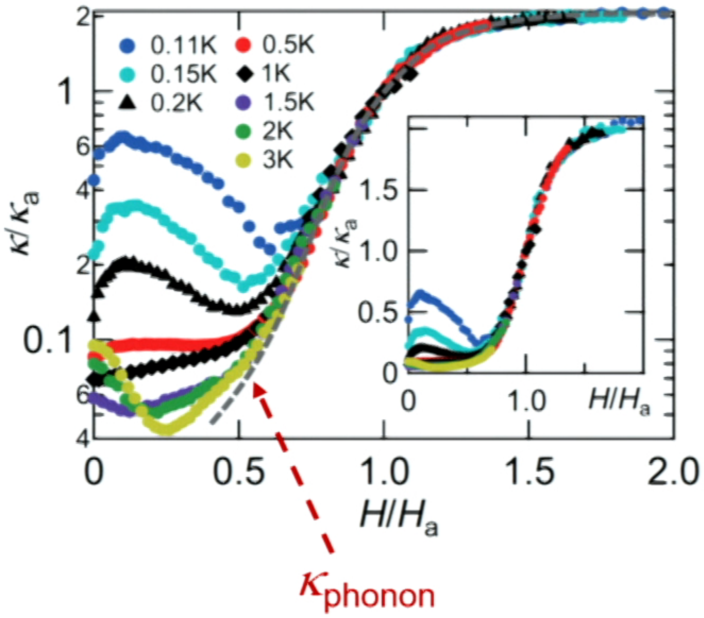


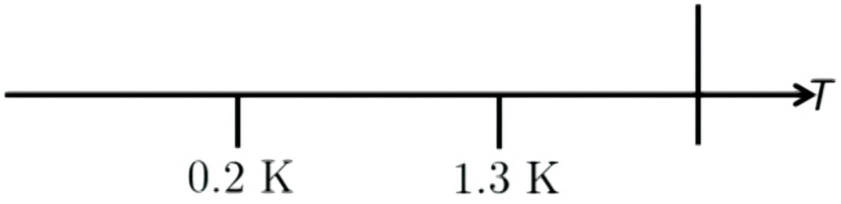
$g\mu_B H \gg J_{\parallel}, k_B T$

purely *phononic*

Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$







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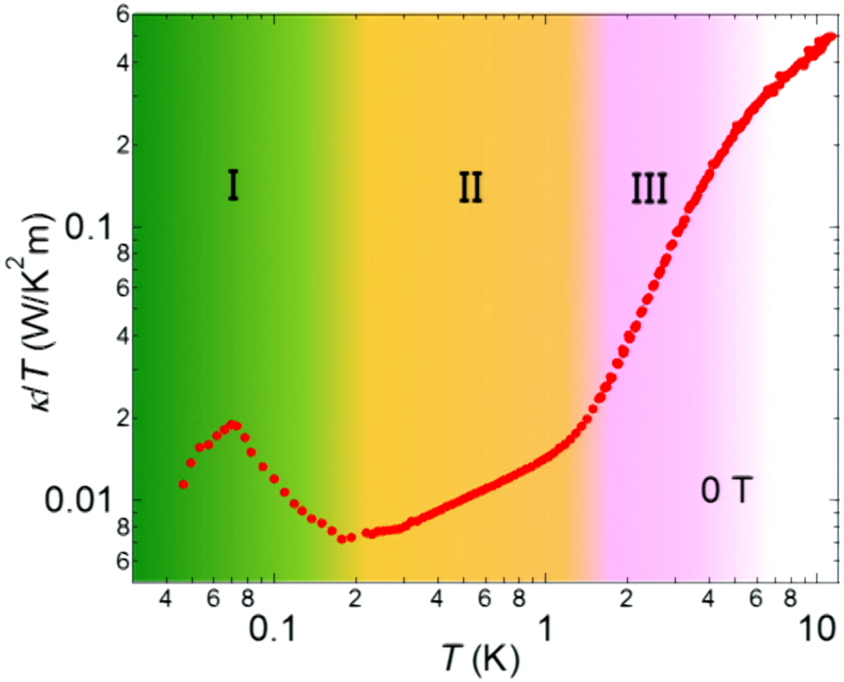
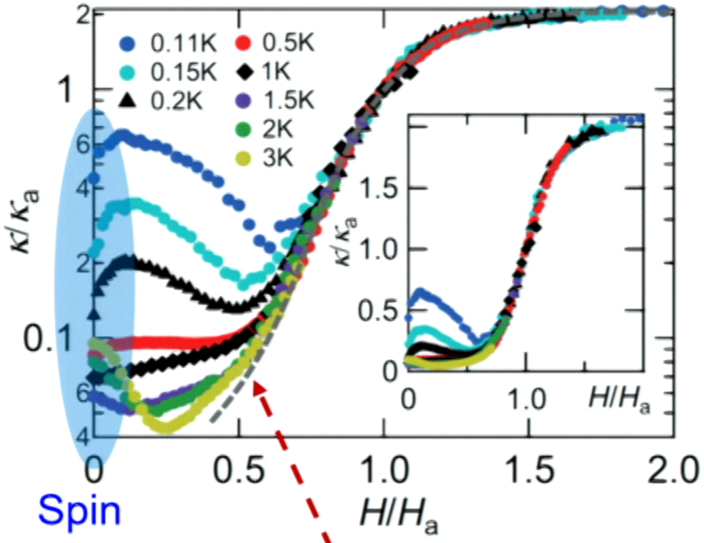
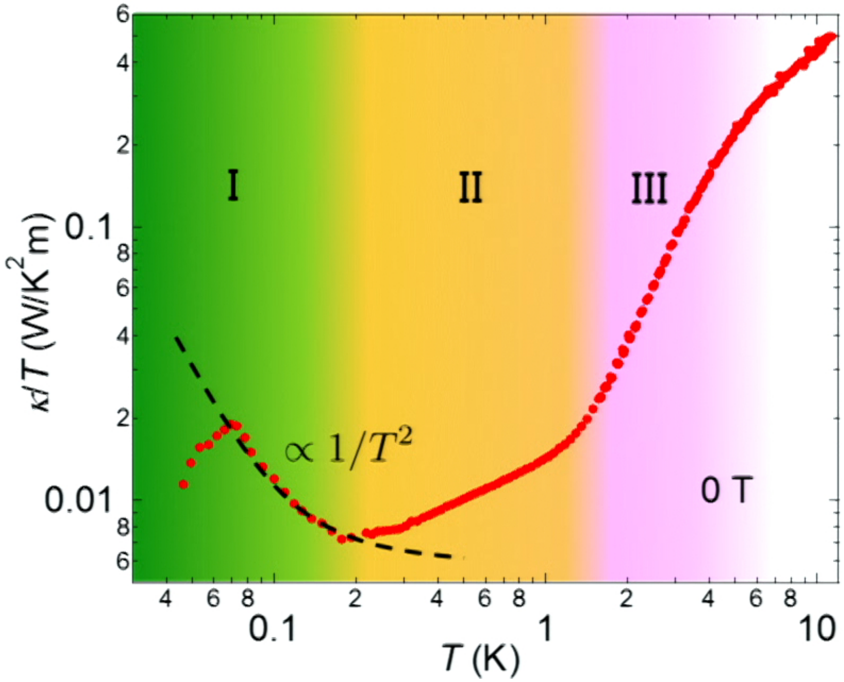


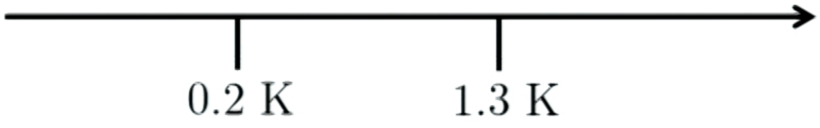


Spin contribution dominates at *H*=0

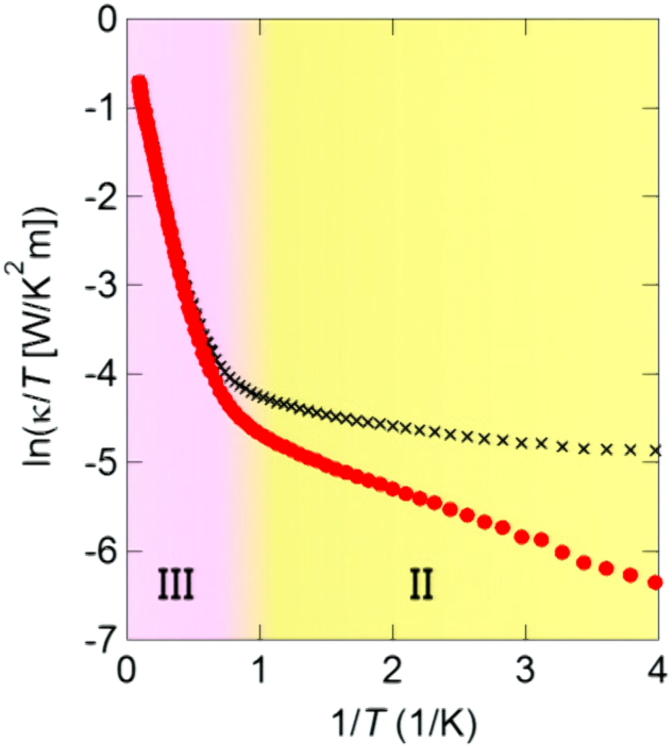
κ_{phonon}

Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$



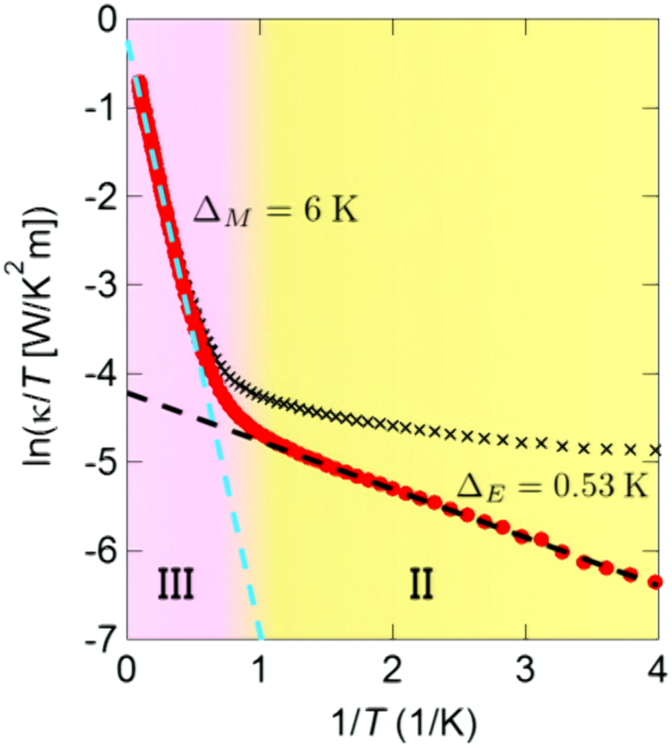


Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$



- Arrhenius plot $\kappa/T \propto \exp(-\Delta/k_\mathrm{B}T)$
  - ✓ In regime III
  - ✓ In regime II

Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$



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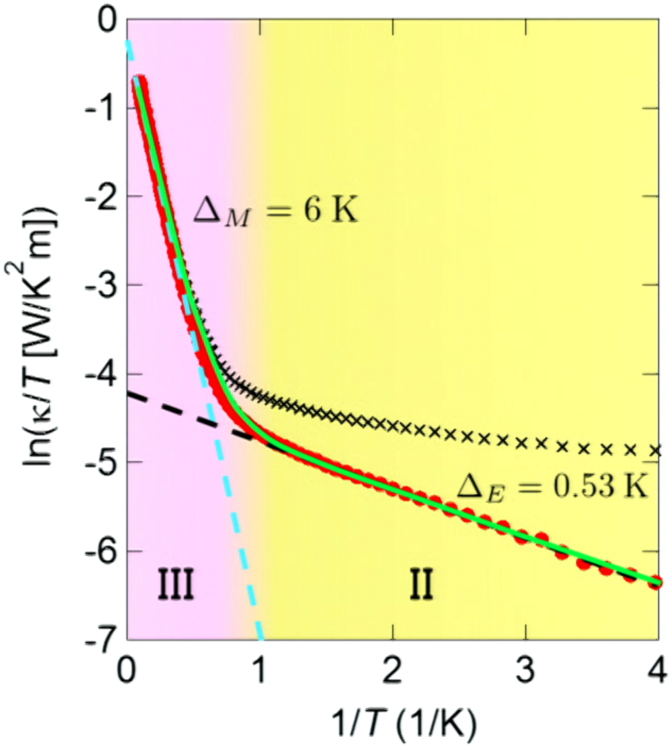


- Arrhenius plot $\kappa/T \propto \exp(-\Delta/k_\mathrm{B}T)$

  ✓ In regime III

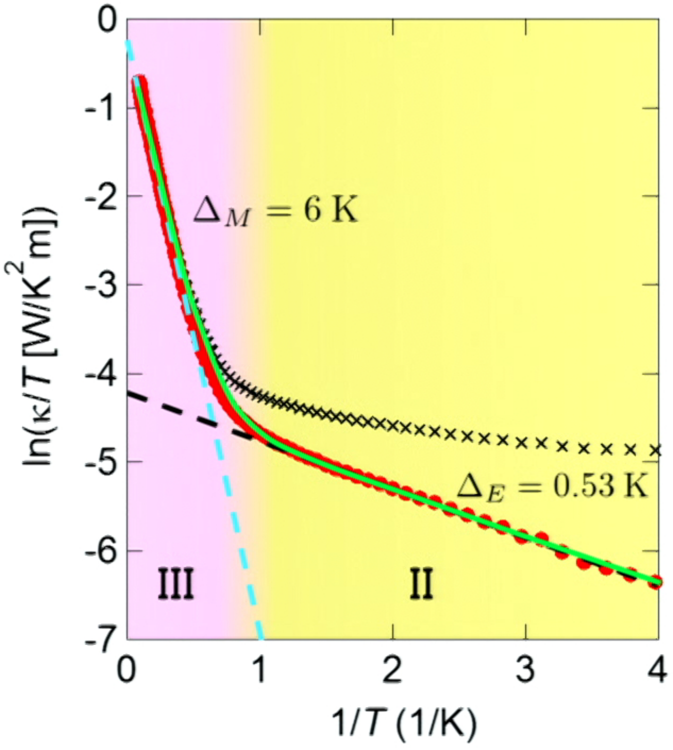
  $\longrightarrow \quad \Delta_M = 6\,\mathrm{K}$

  ✓ In regime II

  $\longrightarrow \quad \Delta_E = 0.53\,\mathrm{K}$

→ Two different types of *gapped* excitations

Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$



- Arrhenius plot $\kappa/T \propto \exp(-\Delta/k_{\mathrm{B}}T)$

✓ In regime III **M-particle**

$\longrightarrow$ $\Delta_M = 6\,\mathrm{K}$

Excitation energy of *M*-particle

$\sim J_{zz} \sim 7\,\mathrm{K}$

✓ In regime II **E-particle**

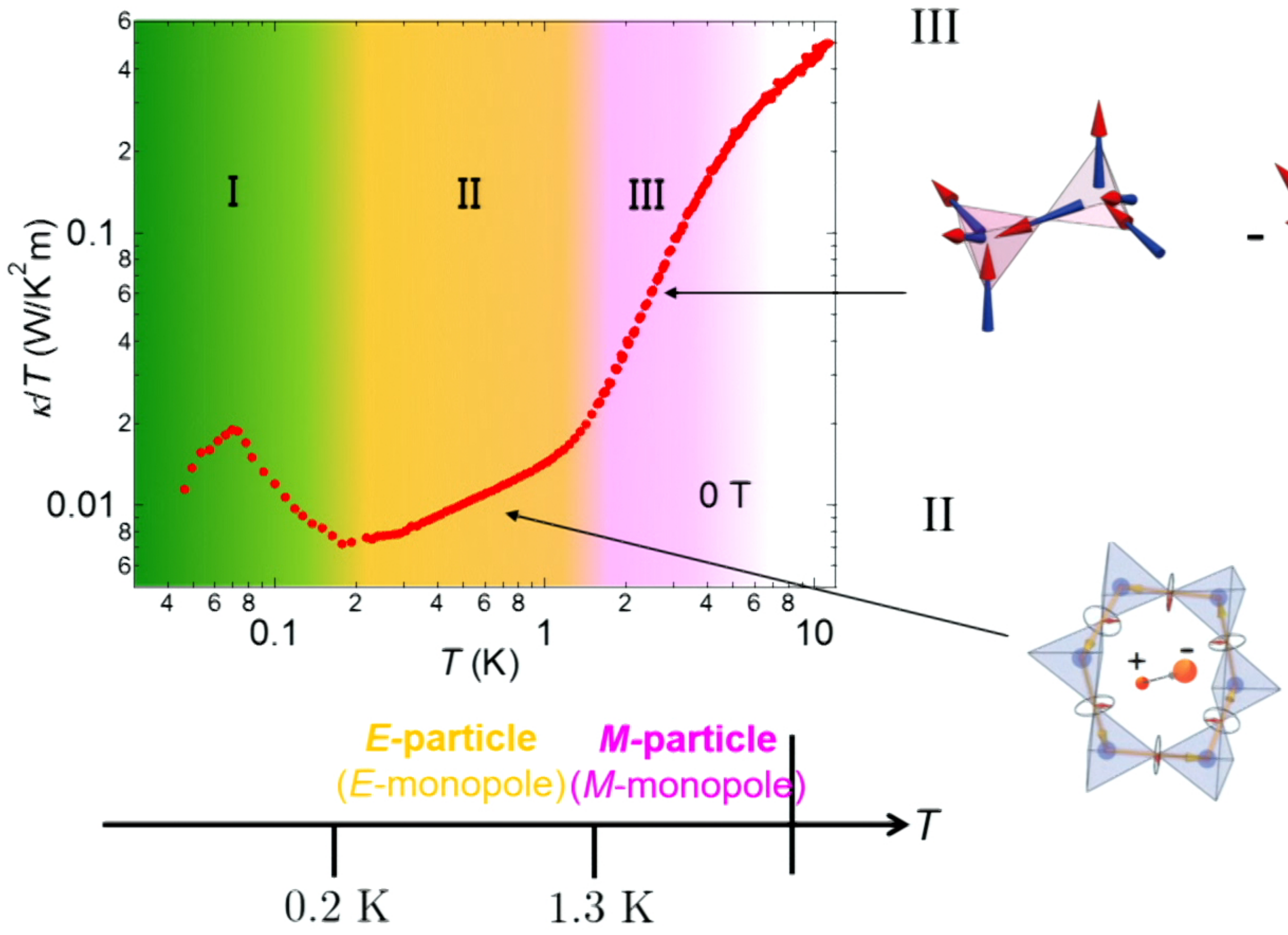
$\longrightarrow$ $\Delta_E = 0.53\,\mathrm{K}$

Excitation energy of *E*-particle

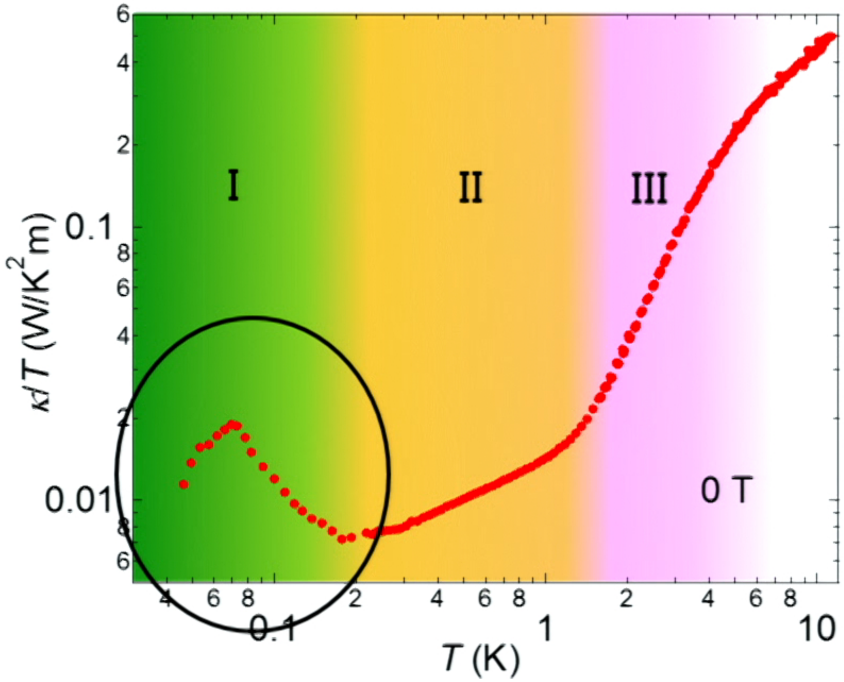
$\sim 3\dfrac{J_\perp^3}{J_{zz}^2} \sim 0.6\,\mathrm{K}$

➡ Two different types of *gapped* excitations

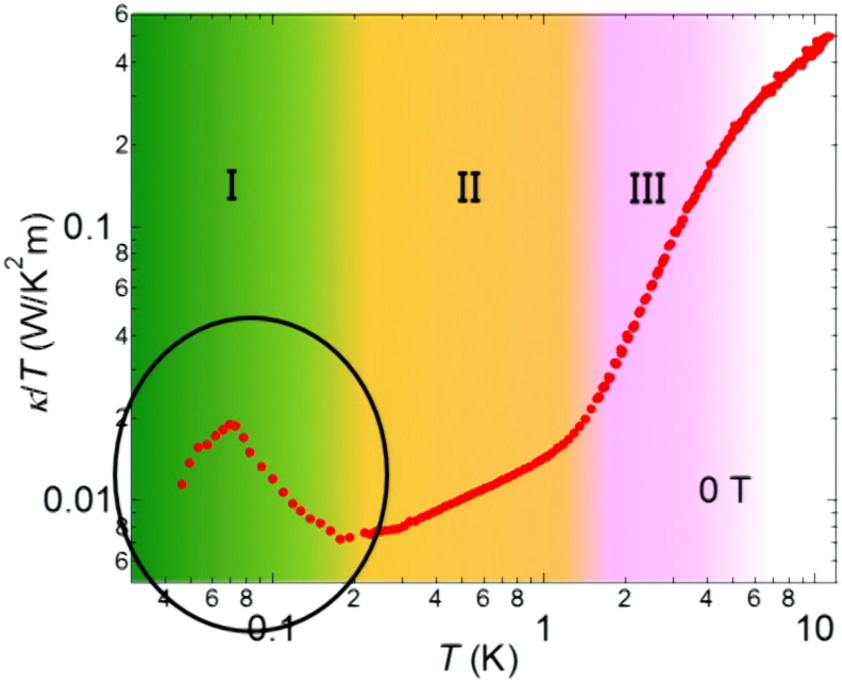
Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$

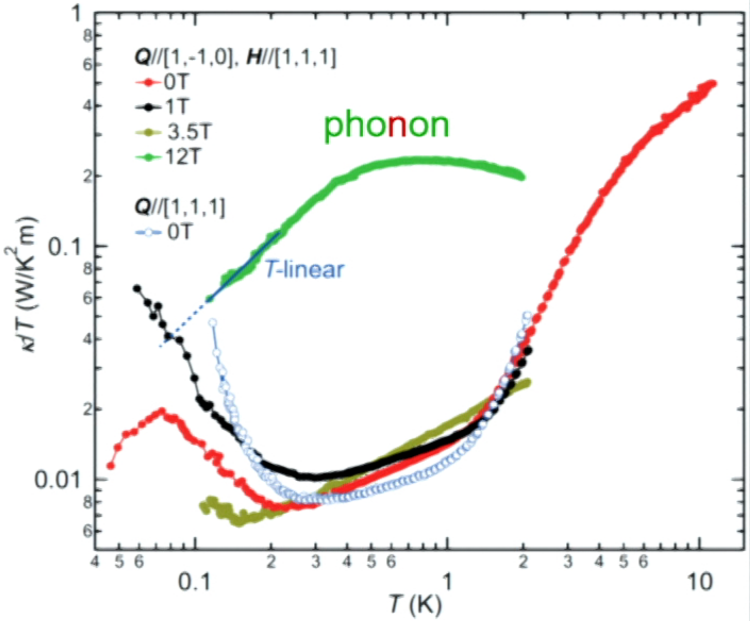


Low temperature enhancement of κ/T in regime I



Low temperature enhancement of κ/T in regime I





Enhancement cannot be attributed to phonon!

1. κ/T exceeds κ_{phon}/T
2. $\kappa_{\text{phon}}/T = (1/3)(C_{\text{phon}}/T)v_{\text{phon}}\ell_{\text{phon}}$ cannot show double peak structure.
3. κ/T of different crystal with a similar size is very different.
   (ℓ_{phon} is limited by sample size).

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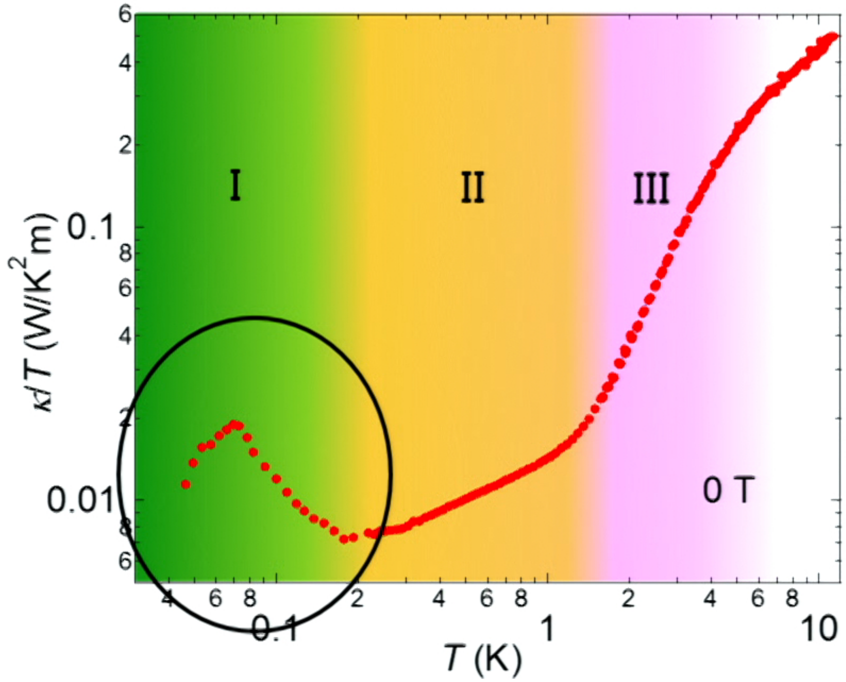

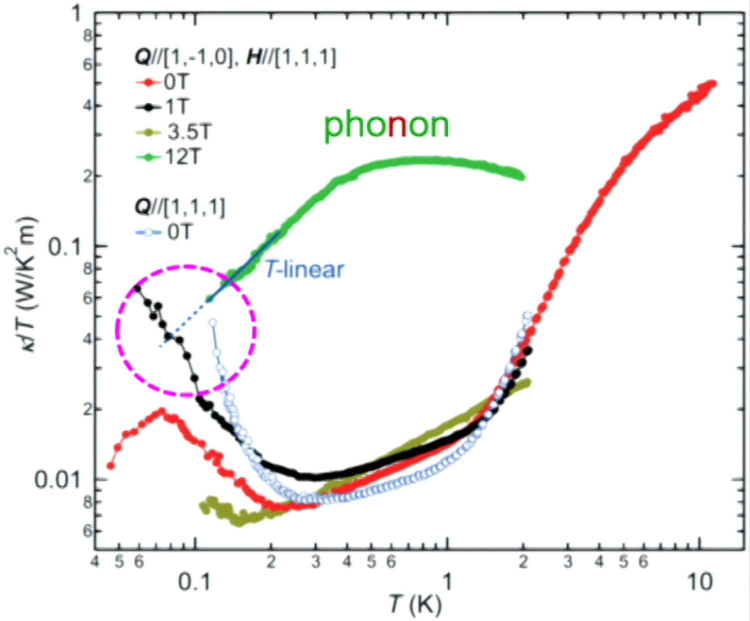

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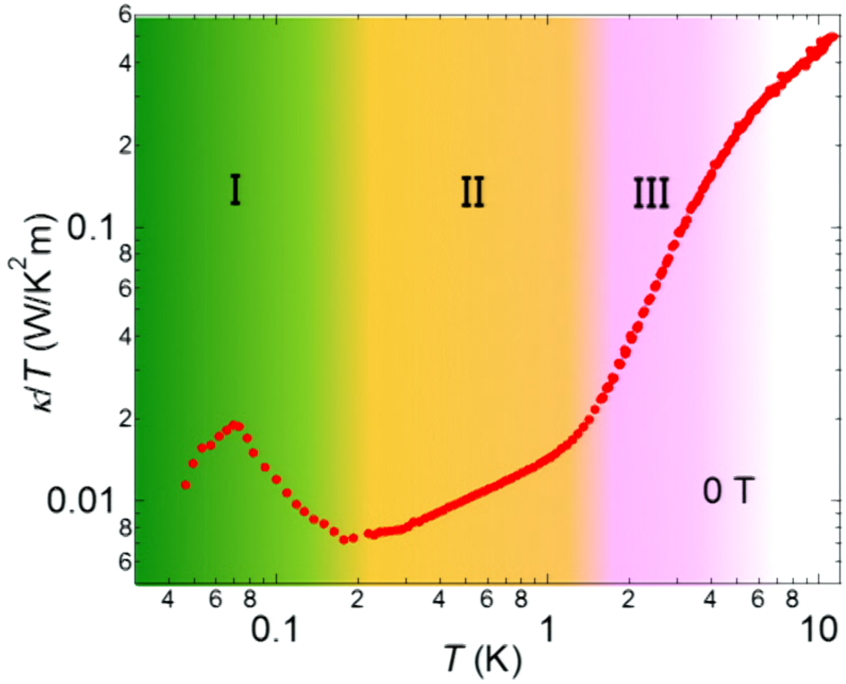
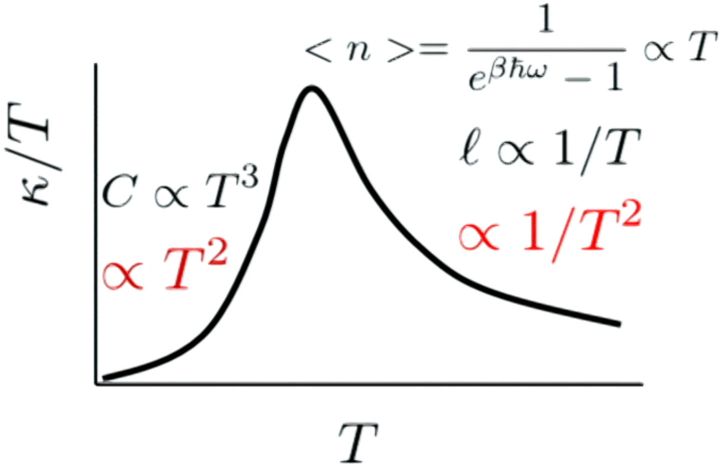
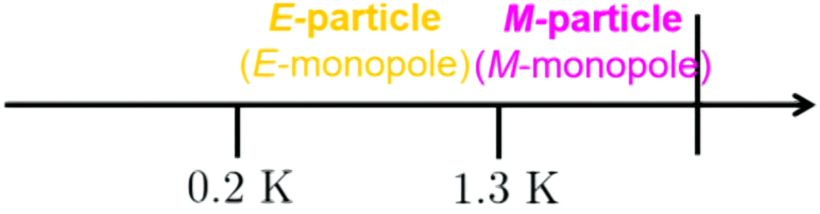
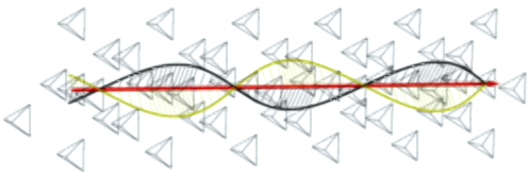
Temperature dependence of thermal conductivity of Pr$_2$Zr$_2$O$_7$



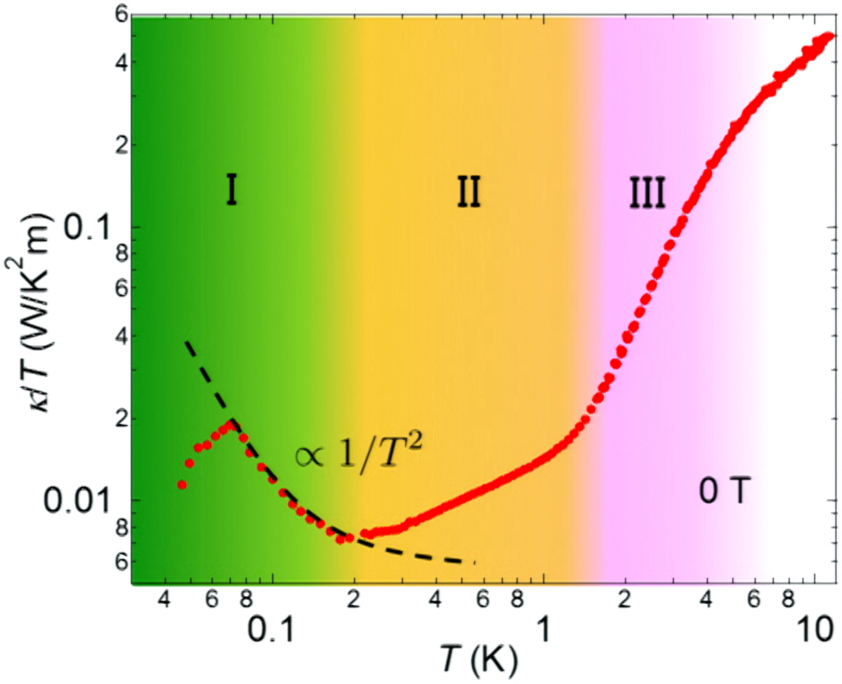
Regime-I







Temperature dependence of thermal conductivity of $Pr_2Zr_2O_7$



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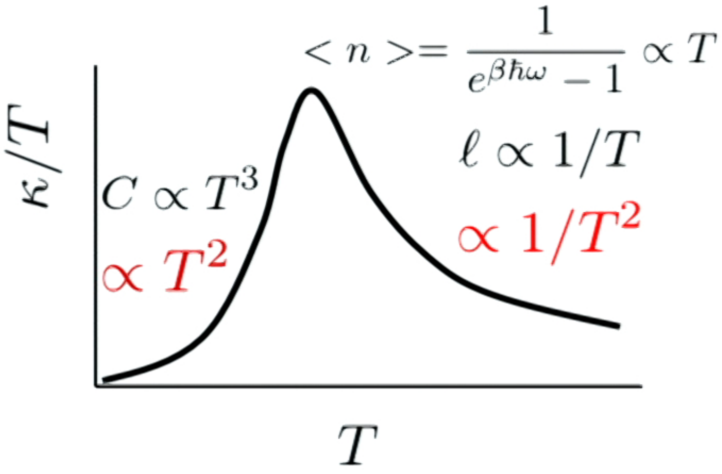
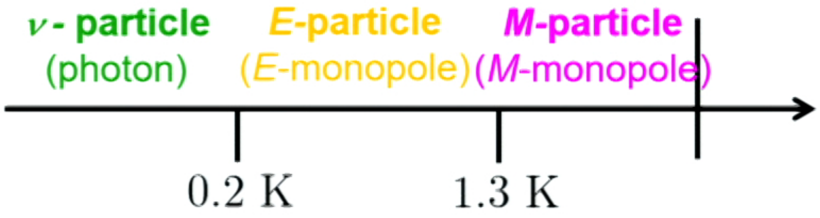
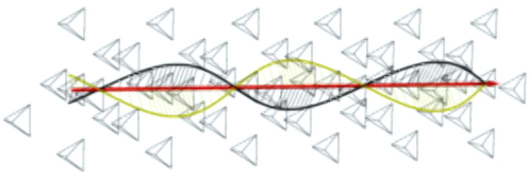



ν - particle (photon)

***E*-particle** (*E*-monopole)

***M*-particle** (*M*-monopole)



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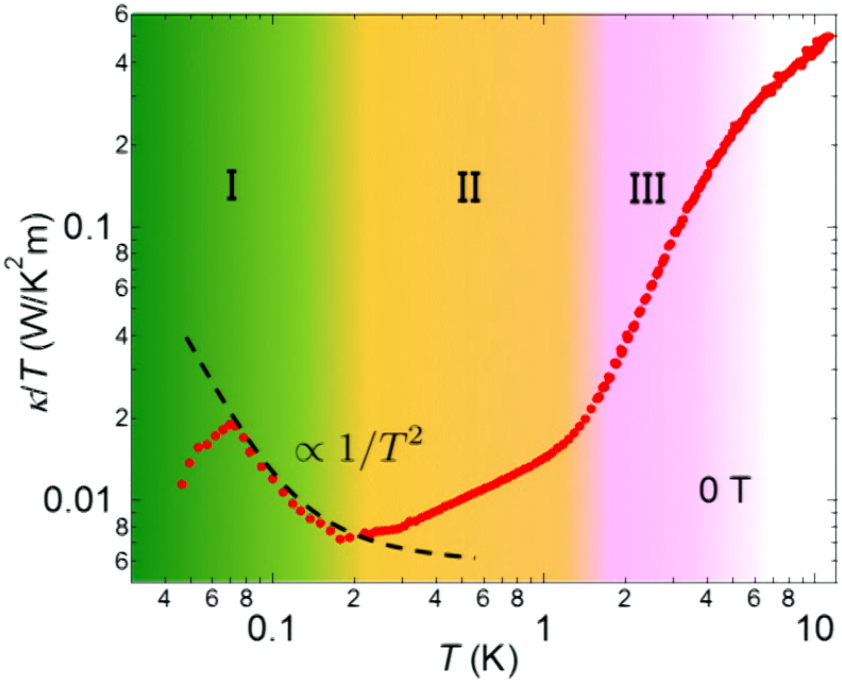
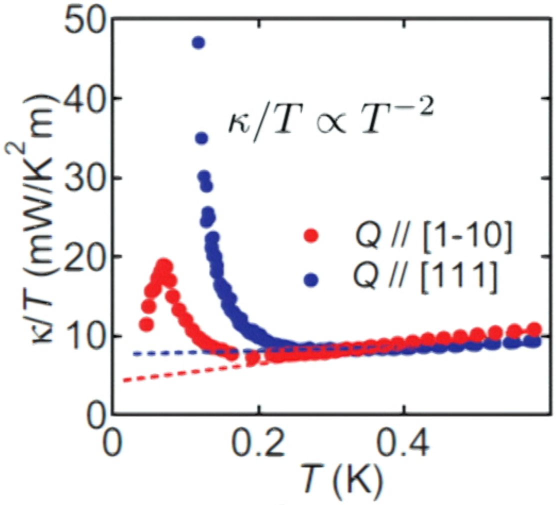
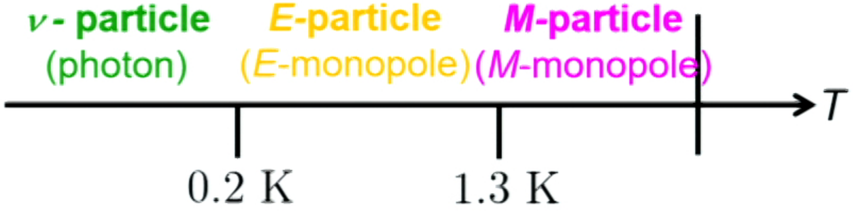


Regime-I



$\kappa/T \propto T^{-2}$

Extremely sensitive to sample quality



ν - particle (photon)

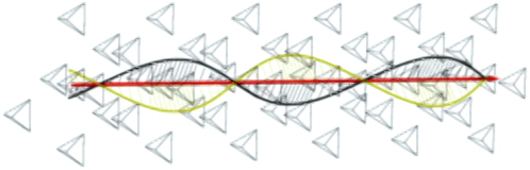
E-particle (E-monopole)

M-particle (M-monopole)

T

0.2 K

1.3 K



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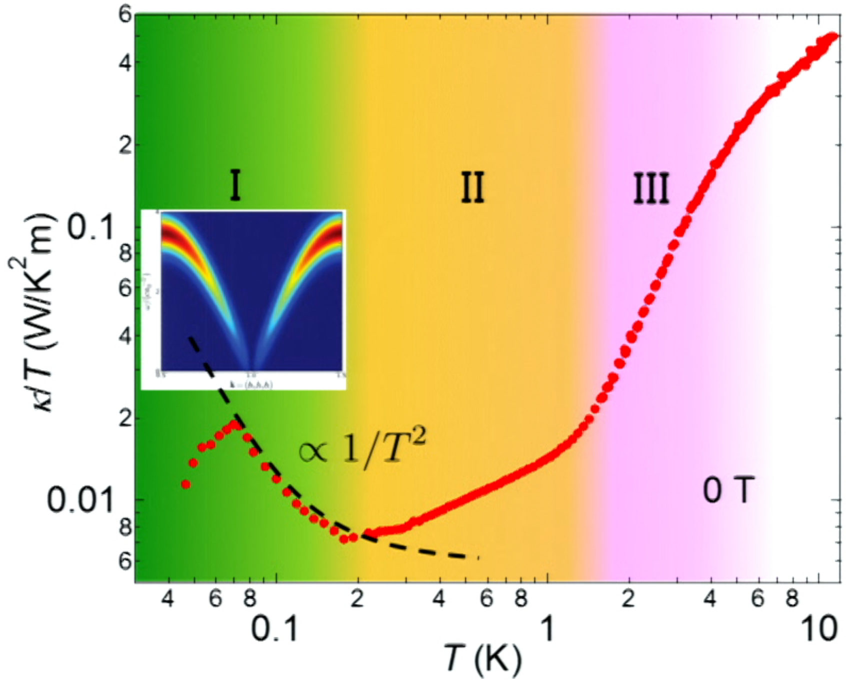
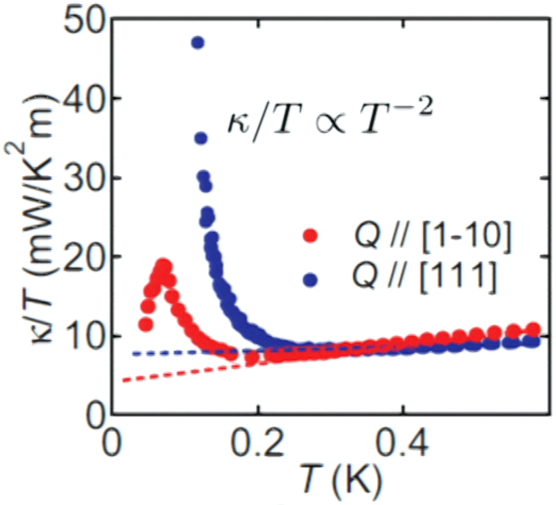


Regime-I



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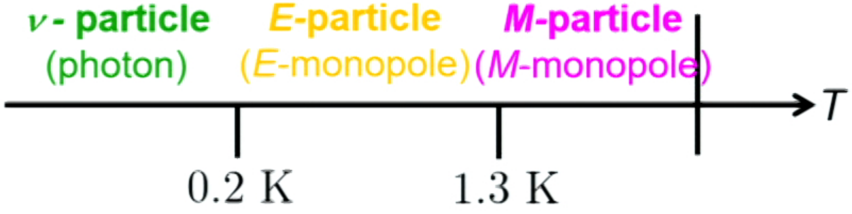

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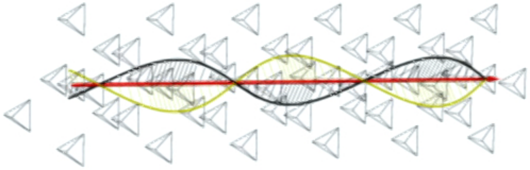
T

0.2 K

1.3 K



Summary

Detecting emergent photon and monopoles in a quantum spin liquid with spin-ice correlations by thermal conductivity

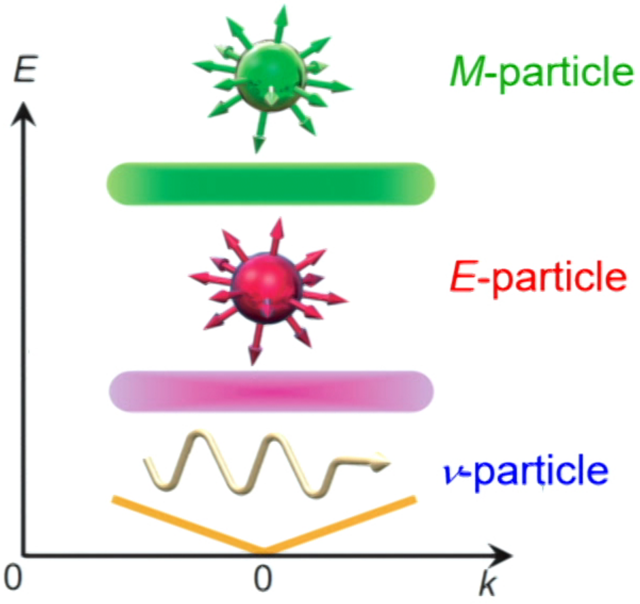
$Yb_2Ti_2O_7$

Itinerant magnetic monopoles (*M*-particle)

$Pr_2Zr_2O_7$

In addition to
magnetic monopoles (*M*-paricles)

Emergent photon (ν-particle) and electric monopoles (*E*-particle)



M- and *E*-particles ⇨ Quasiparticle fractionalization

Emergent ν- and *E*-particles ⇨ Gauge field fluctuations