Heat transport and magnetic phase transitions of low-dimensional quantum magnets

Xuefeng Sun（孙学峰）

Hefei National Laboratory for Physical Sciences at the Microscale (HFNL),
University of Science and Technology of China (USTC)

中国科学技术大学
University of Science and Technology of China

合肥微尺度物质科学国家实验室(筹)
HEFEI NATIONAL LABORATORY FOR PHYSICAL SCIENCES AT MICROSCALE

Collaborators

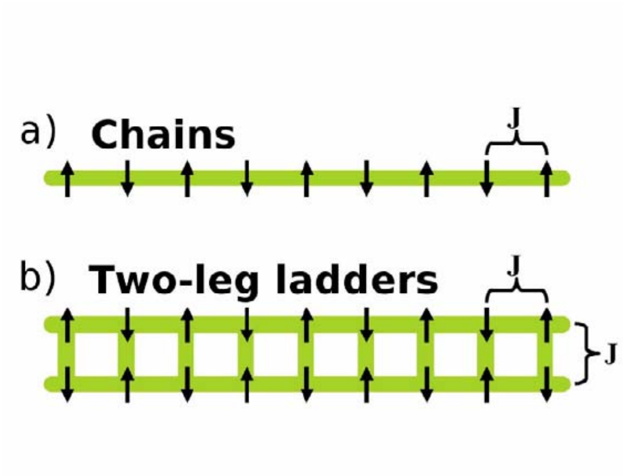
- Z. Y. Zhao（赵志颖）, C. Fan（范诚）, Q. J. Li（李秋菊）, F. B. Zhang（张发宝）, L. M. Chen（陈丽敏）, J. Shi（史俊）， X. Zhao（赵霞）
- H. D. Zhou（周海东）(University of Tennessee)
- Y. Ando, A. A. Taskin (Osaka University)

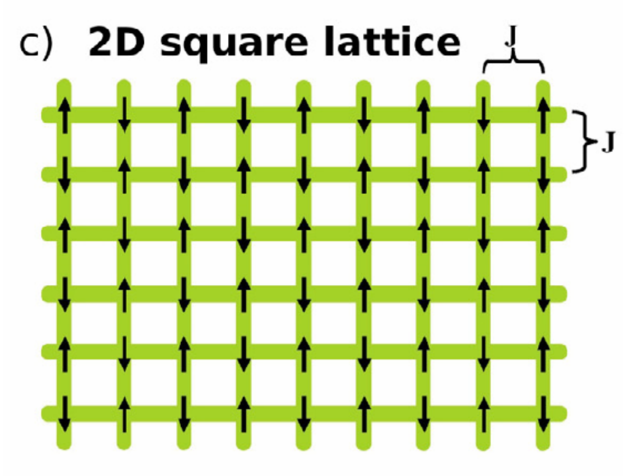
Outlines

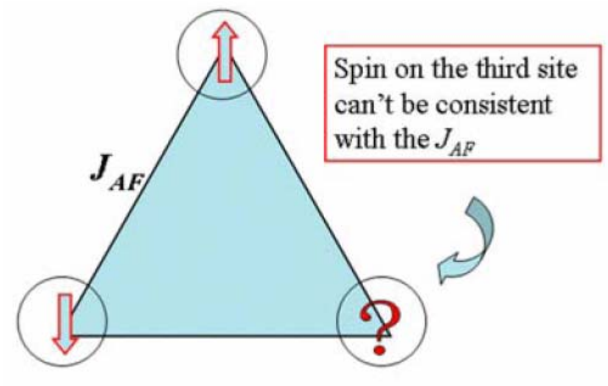
- Research purposes: magnetic heat transport and magnetic transition in quantum magnets
- Advantages of heat transport as a probe
- Complications of heat transport as a probe
- Several results of heat transport study on the low-dimensional magnets

Quantum Magnets

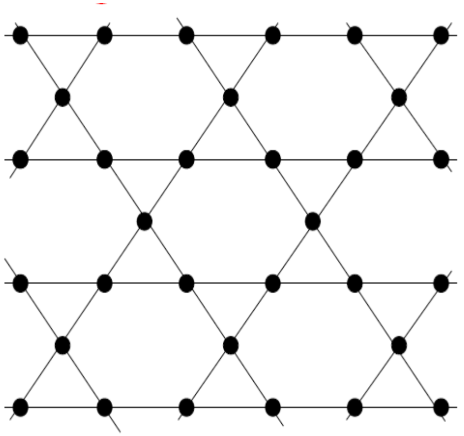
- **Low dimensionality, Small spin number, Spin frustration**







Geometry frustration



Kagome lattice

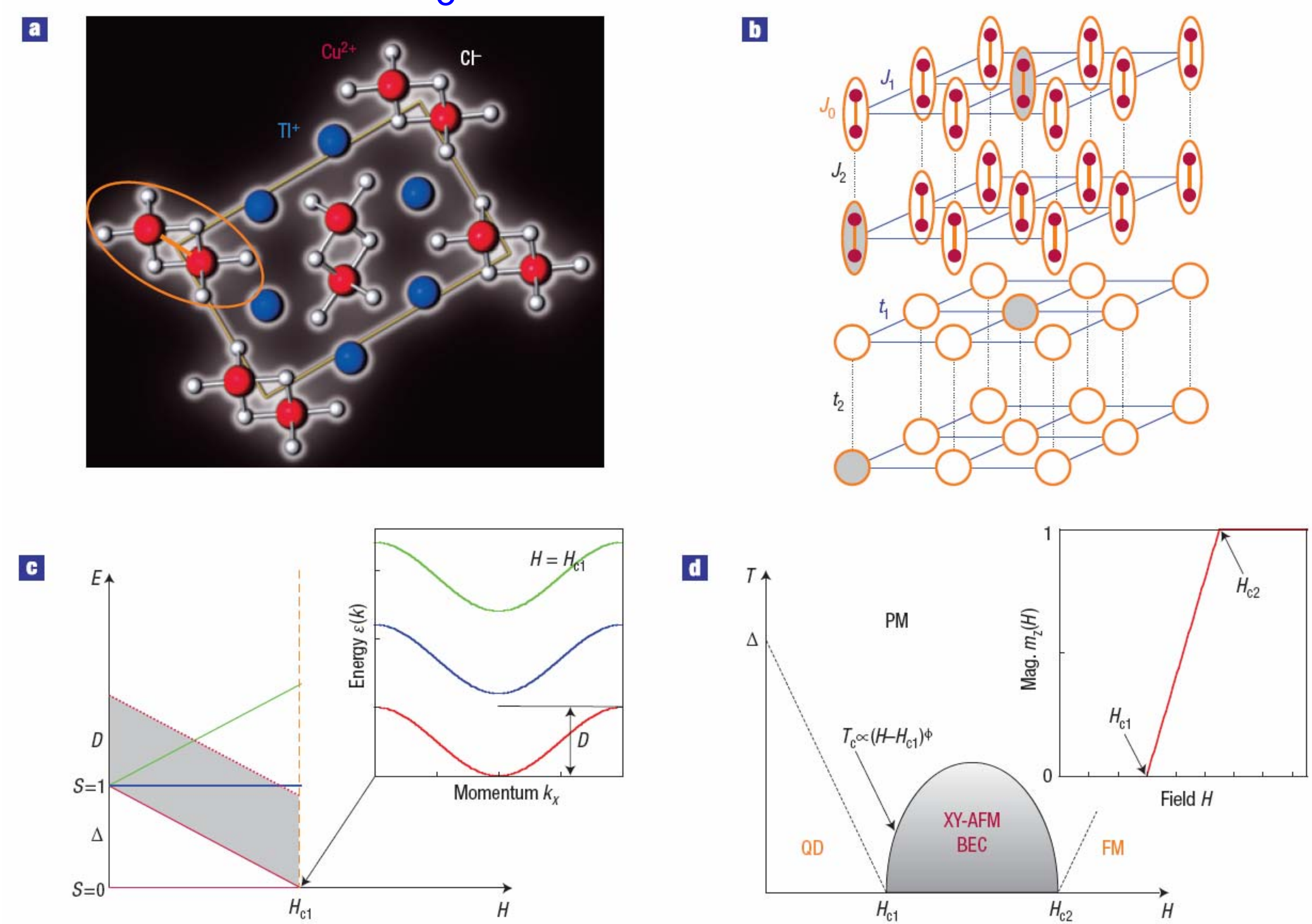
Novel physics in quantum magnets

- **Novel quantum magnetism**
  - Spin liquid, Spin gap, Spin dimer, Haldane gap, ……
  - Quantum phase transitions
- **Peculiar magnetic excitations or quasiparticles**
  - Magnon, Spinon, ……

Field-induced QPTs in spin-gapped materials

3D $TlCuCl_3$

T. Giamarch *et al.*, Nature Phys. **4**, 198 (2008)

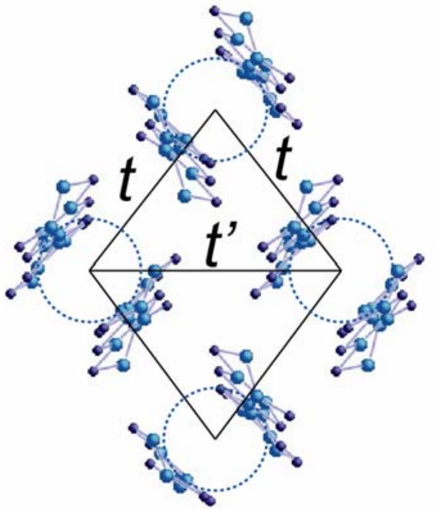


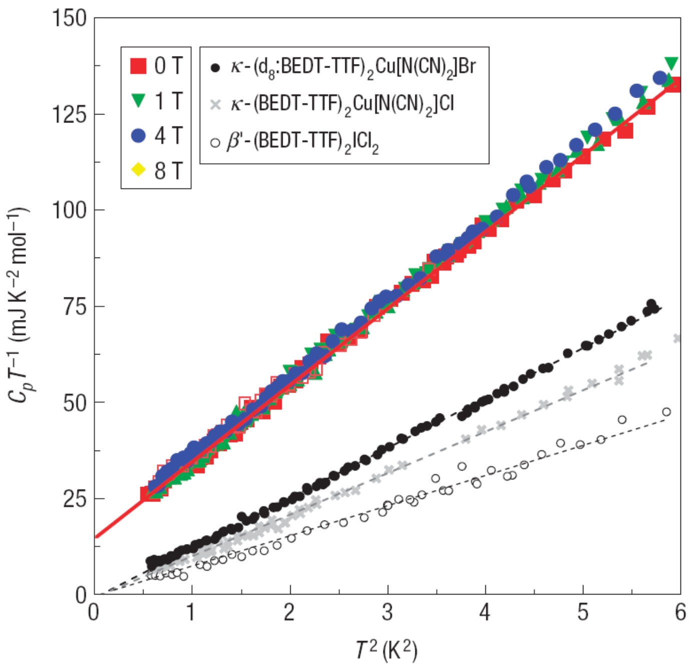
Magnon Bose-Einstein Condensation?

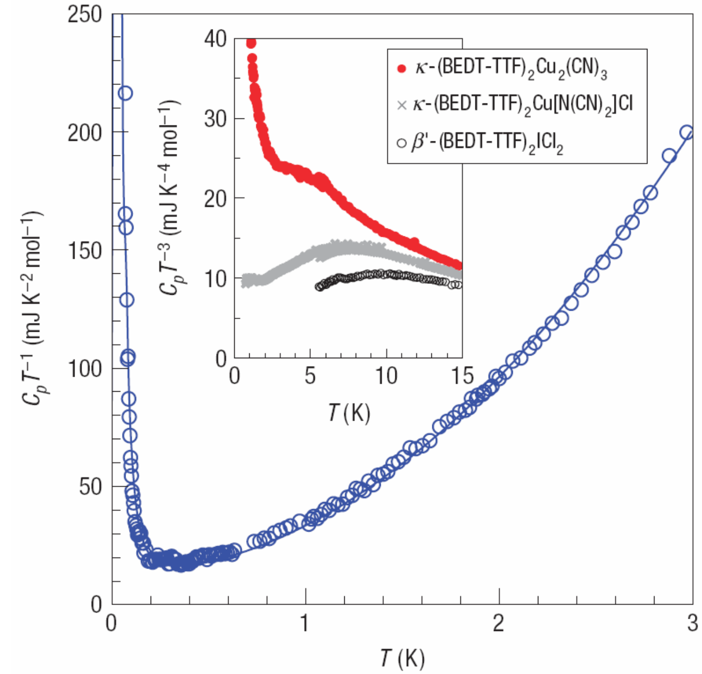
Experimental probe: specific heat

- A direct probe for the nature of ground state and magnetic excitation
- Usually difficult to analyze due to the Schottky term

κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ $\gamma = 15 \,\mathrm{mJ}/\mathrm{K}^2\mathrm{mol}$ Evidence for Gapless spinon?







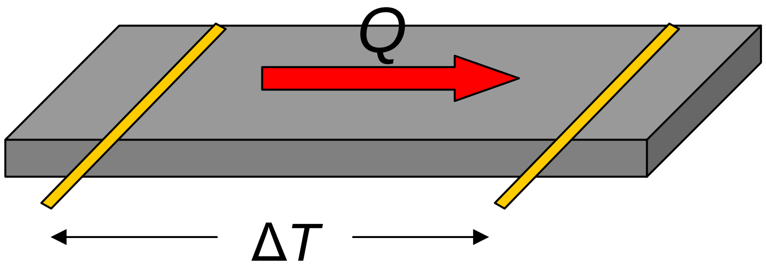
S. Yamashita *et al.*, Nature Phys. **4**, 459 (2008)

Heat transport as a probe

- Thermal conductivity can probe all the extended elementary excitations

$$\kappa = \kappa_{ph} + \kappa_e + \kappa_m$$

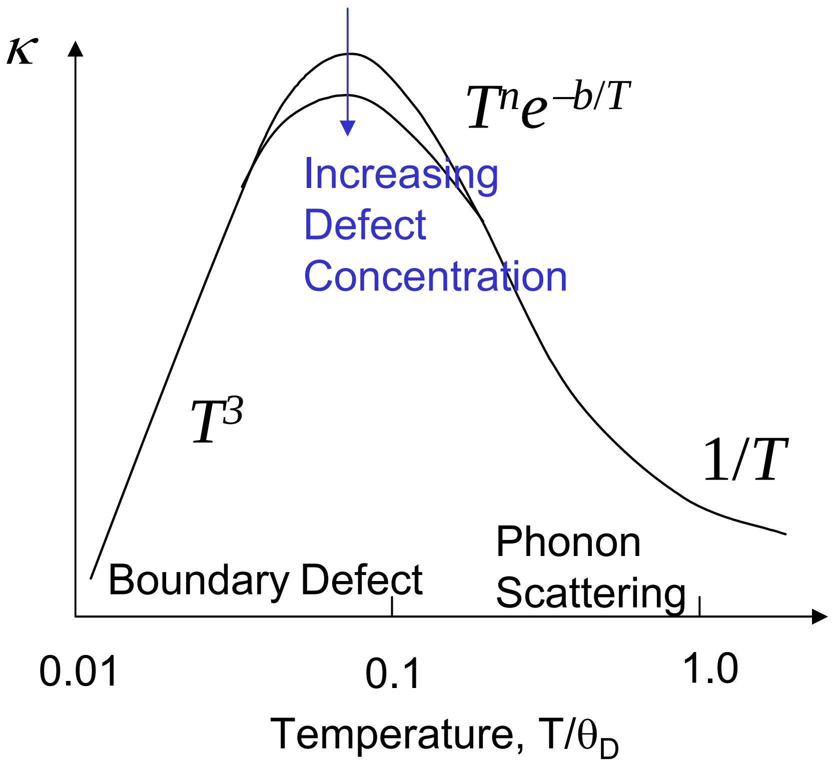
- Insulator: $\kappa_e = 0$

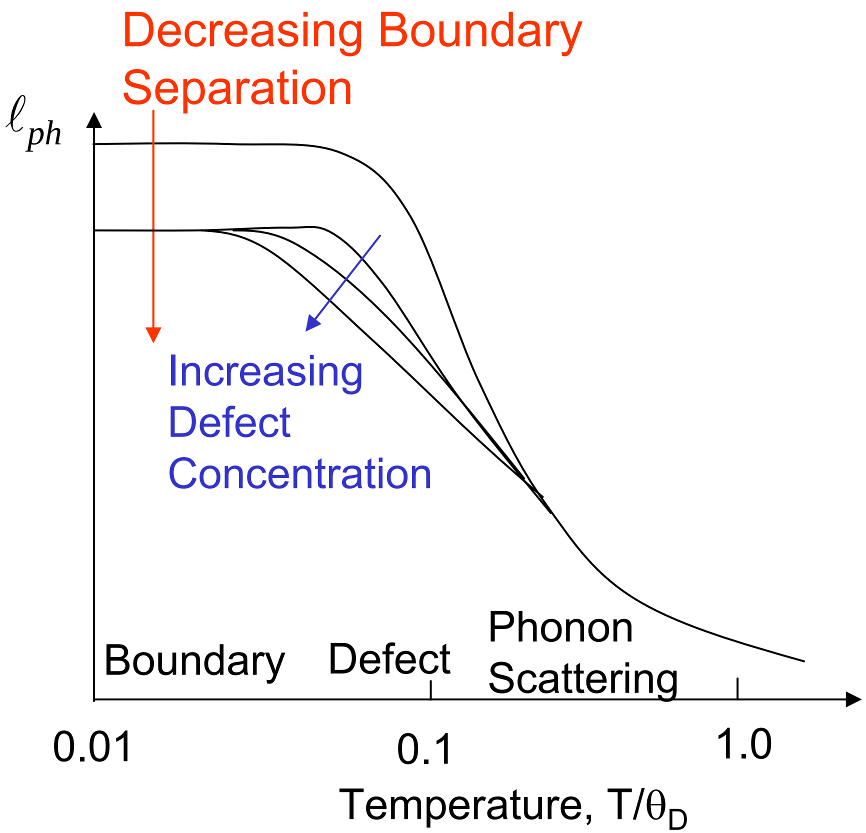


$$\kappa \propto \frac{Q}{\Delta T}$$

Thermal conductivity of insulators: Phonons

$$\kappa = \frac{1}{3} c v l_{ph}$$

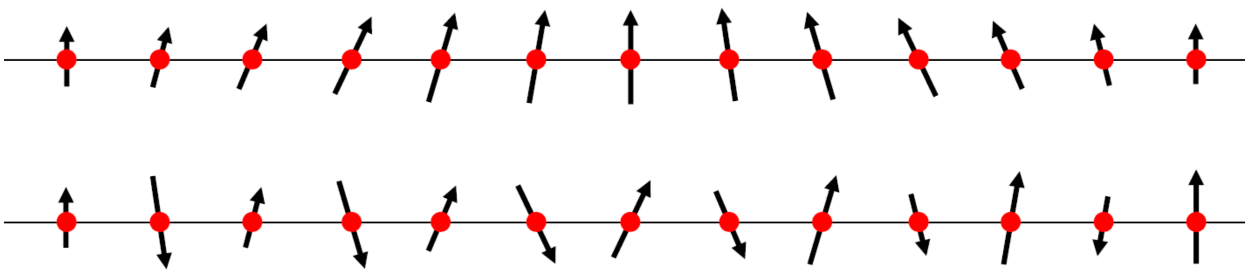




***T^3*: ballistic transport of phonons (boundary scattering limit)**

Magnetic excitations

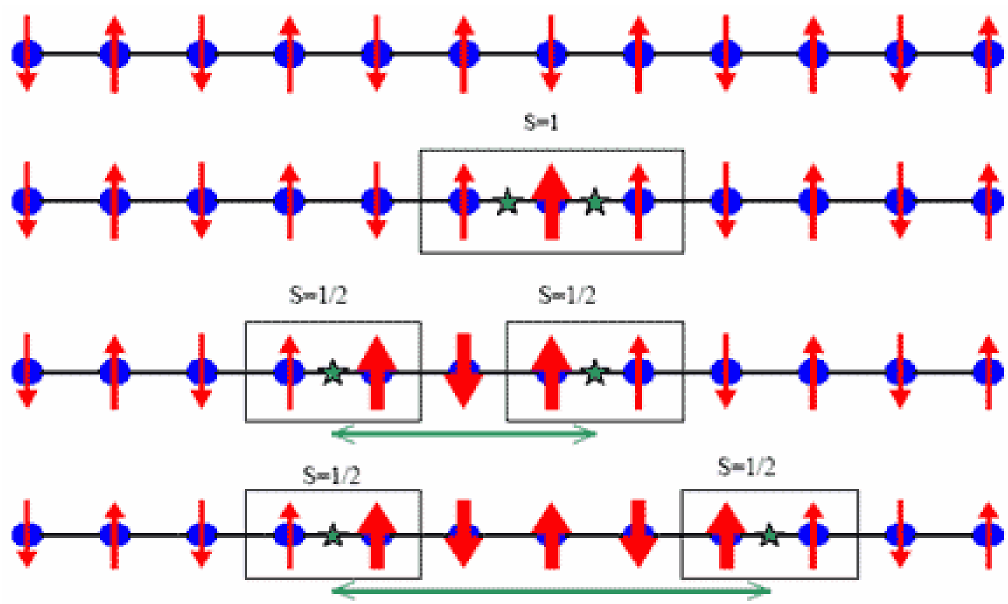
- Magnon：Quantized spin wave in LRO state



$$\varepsilon(k) = 8JS\sin^2(k_x/2)$$

$$\varepsilon(k) = 4JS\left|\sin k_x\right|$$

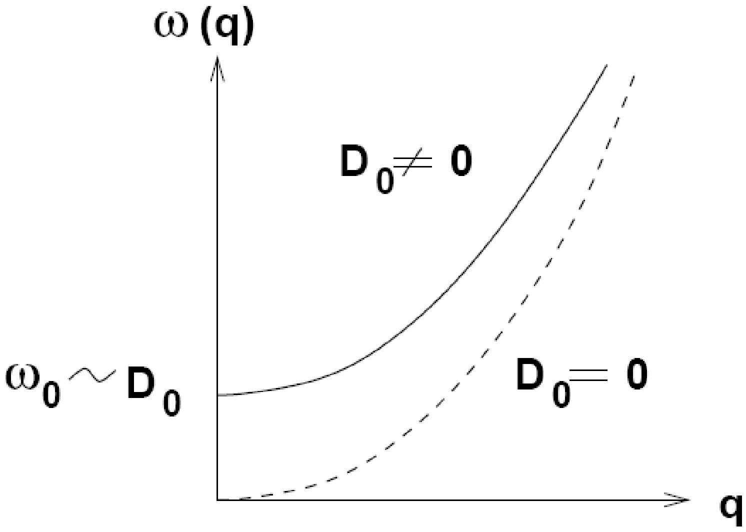
- Spinon：in one-dimensional spin chain



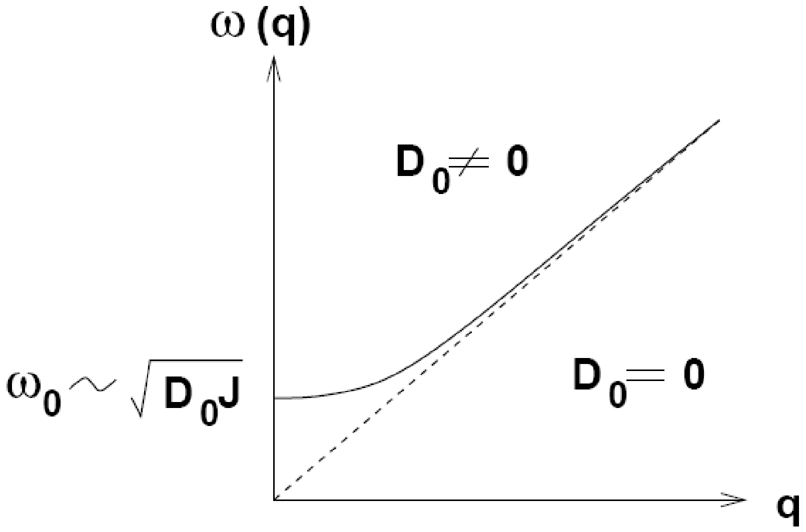
$$\varepsilon(k) = \frac{\pi J}{2}\left|\sin k_x\right|$$

Magnon dispersion and anisotropy

FM



AFM

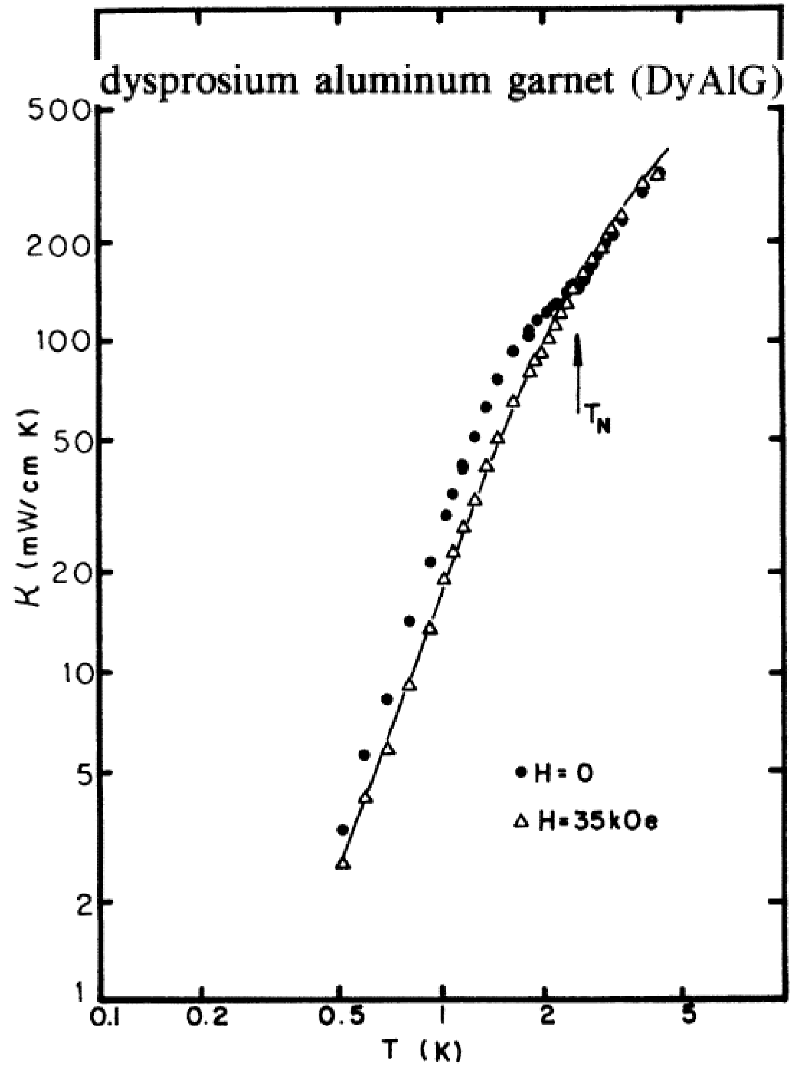


If D_0 = 0, at low temperatures, AFM magnon thermal conductivity

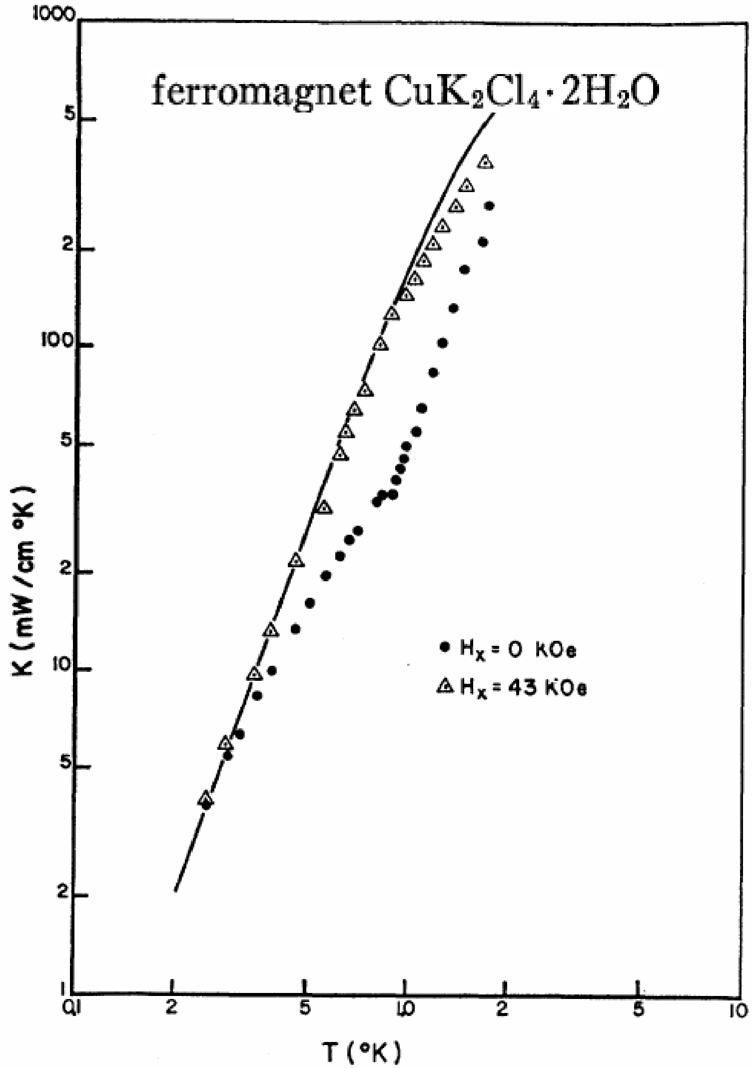
$$\kappa_m = \frac{1}{3} C_m v_m l_m \propto T^3$$

It is a ballistic transport (boundary scattering limit).

Magnons can act as either heat carriers or phonon scatterers

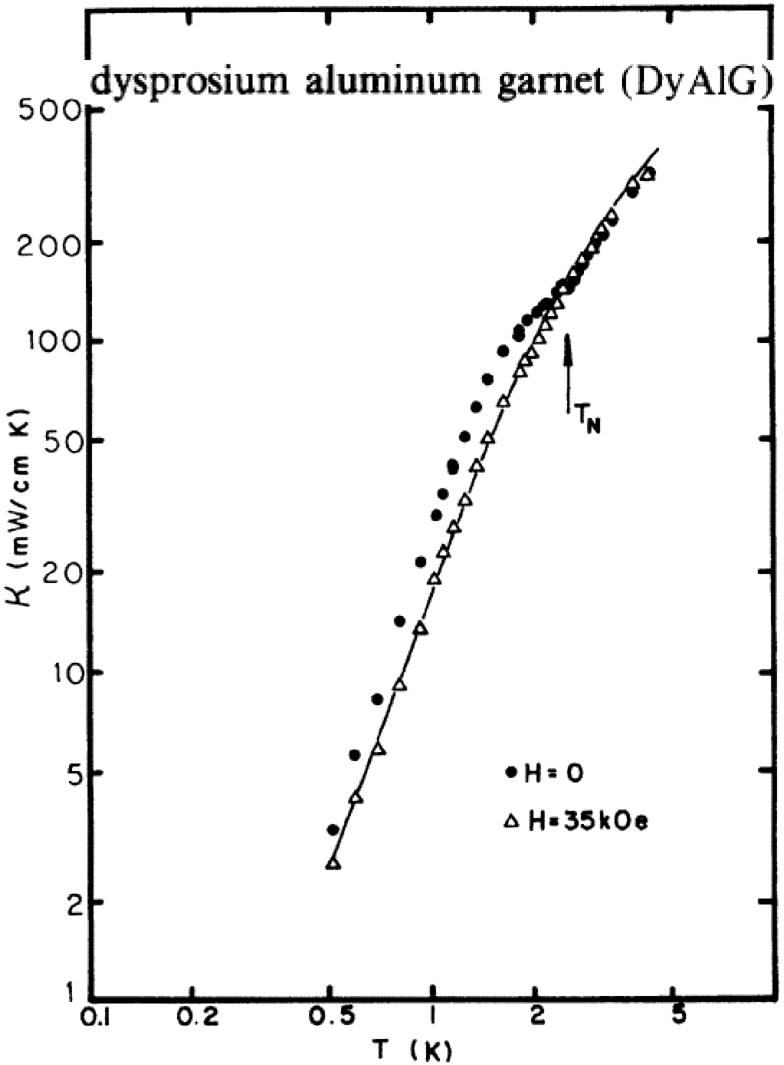


G. S. Dixon *et al.*, PRB **13**, 3121 (1972)

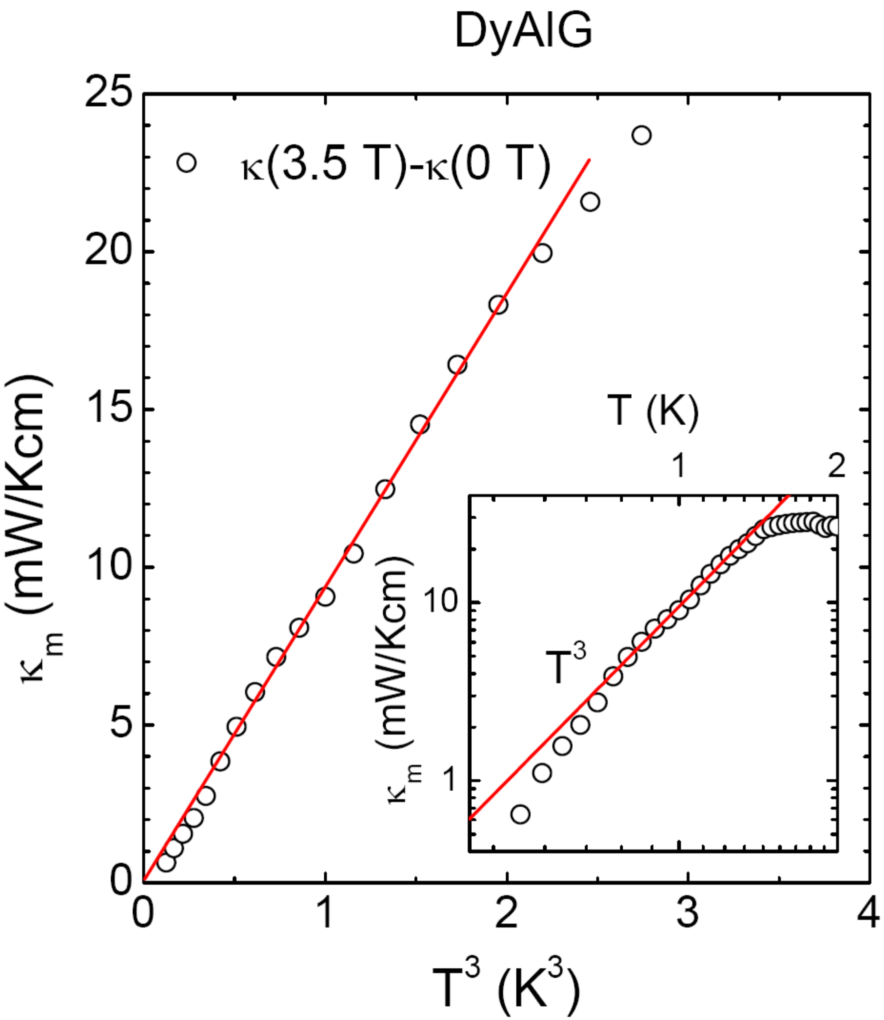


G. S. Dixon *et al.*, PR **185**, 735 (1969)

Magnons can act as either heat carriers or phonon scatterers



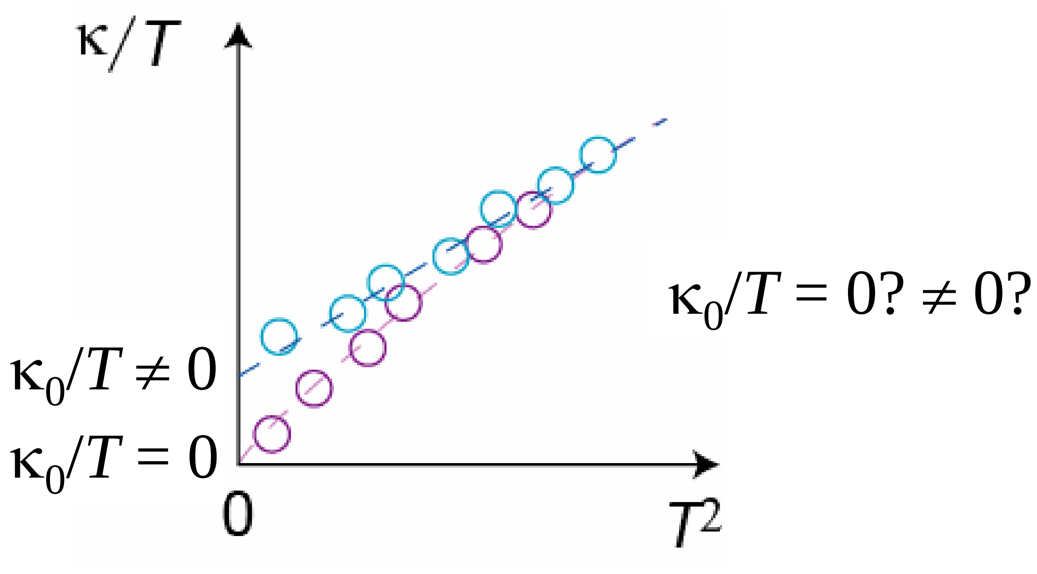
G. S. Dixon *et al.*, PRB **13**, 3121 (1972)



κ_m decrease more quickly than T^3:
anisotropy gap is not zero!

Heat transport can also probe spinons in low-dimensional magnets

- Magnetic excitations can act as heat carriers
- Magnetic thermal conductivity manifests the characteristics of magnetic excitations and is very useful for studying spin liquid and spin gap



Gapped or Gapless spin liquid?
Spinon with a Fermi surface?

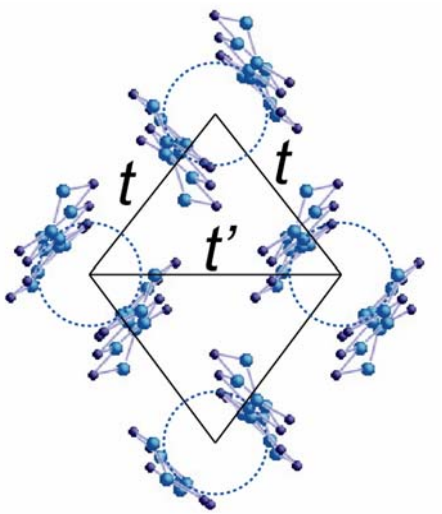
Example: Probe the gap of quantum spin liquid

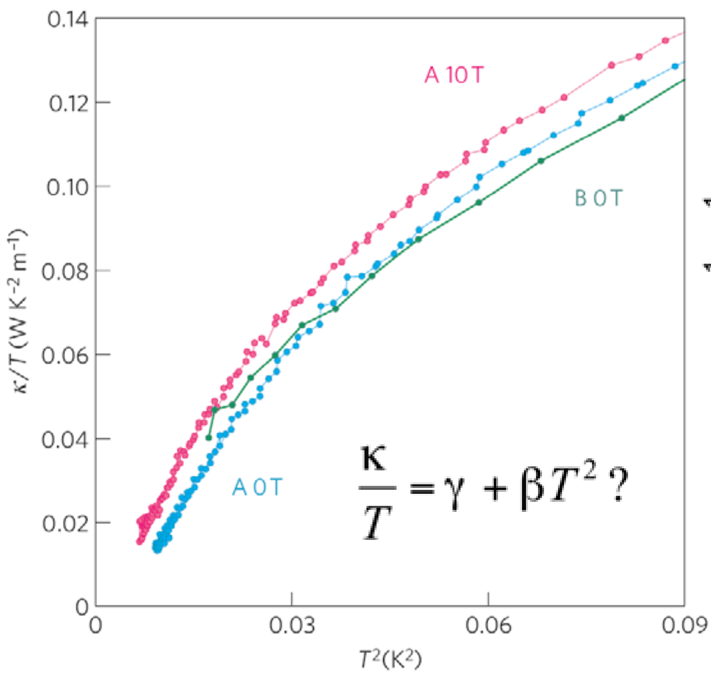
- $\kappa(T)$ at very low temperatures can show the statistical law of magnetic excitations and the information of gap

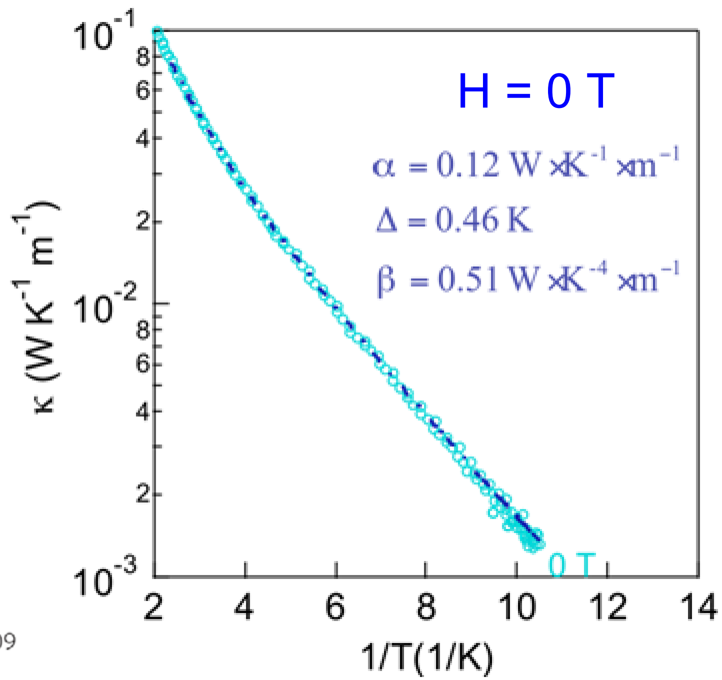
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

$$\gamma = 0$$

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





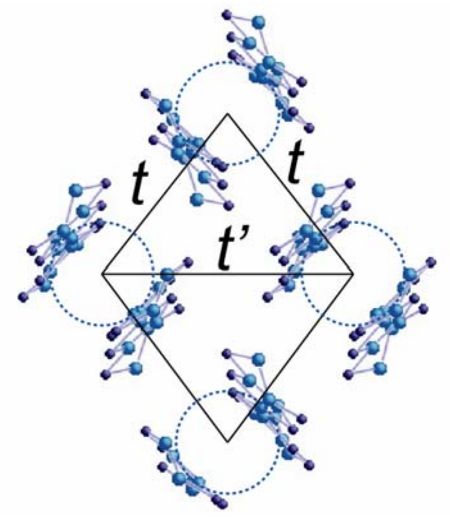


M. Yamashita *et al.*, Nature Phys. **5**, 44 (2009)

Example: Probe the gap of quantum spin liquid

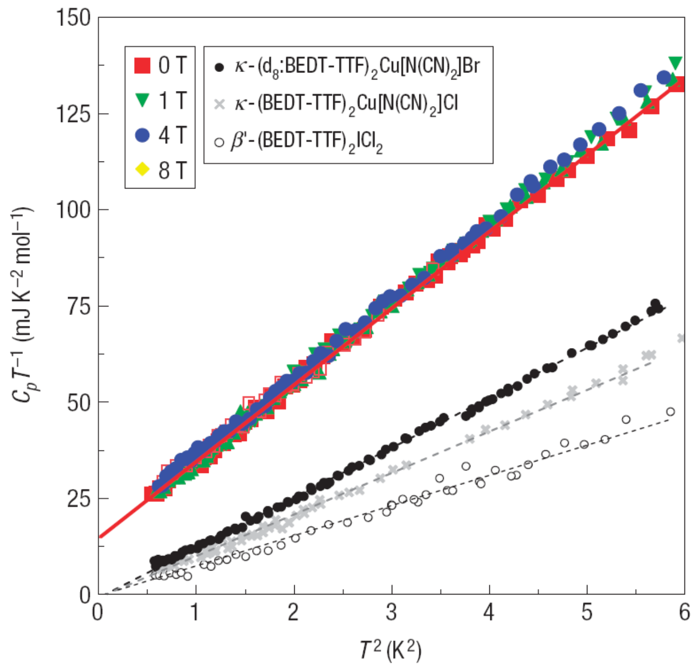
κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$

Inconsistency between specific heat and κ

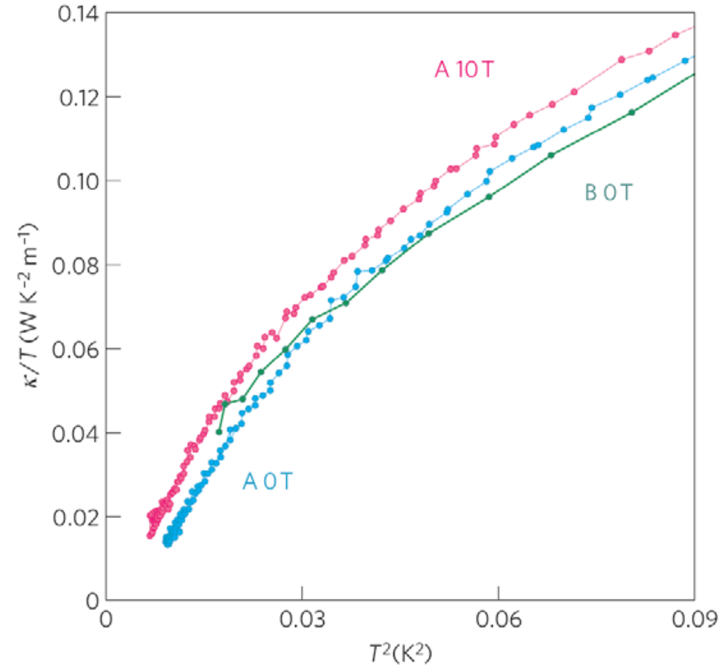


$$\frac{C_p}{T} = \gamma + \beta T^2$$

$\gamma = 15$ mJ/K^2 mol



$\kappa_0 / T = 0$



S. Yamashita *et al.*, Nature Phys. **4**, 459 (2008)

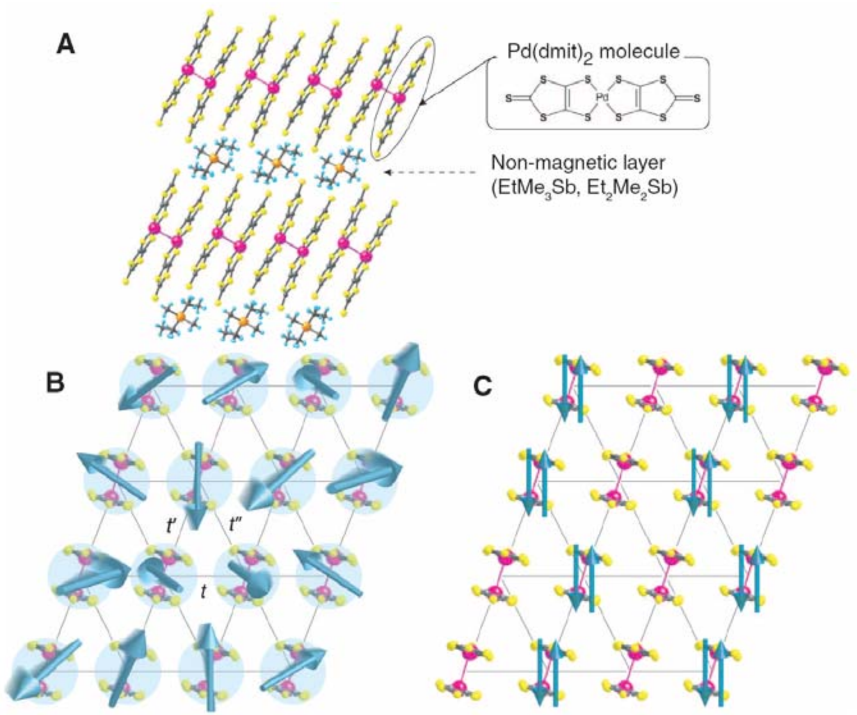
Evidence for Gapless spinon?

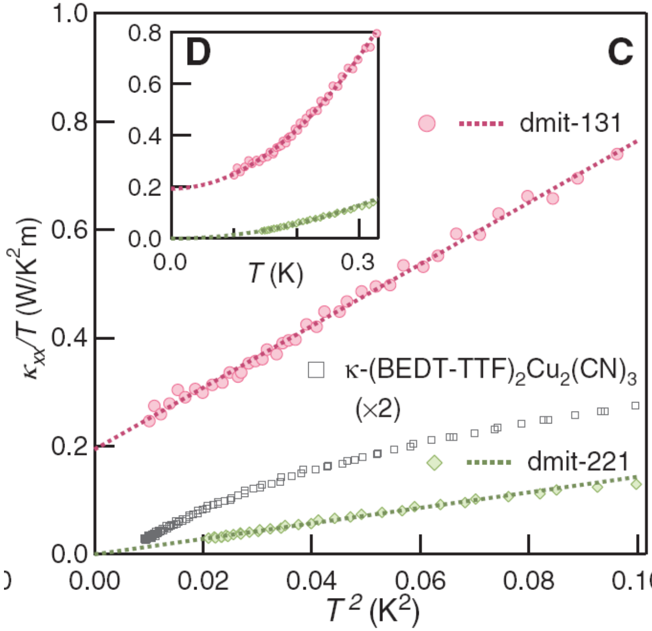
Evidence for a Gapped state?

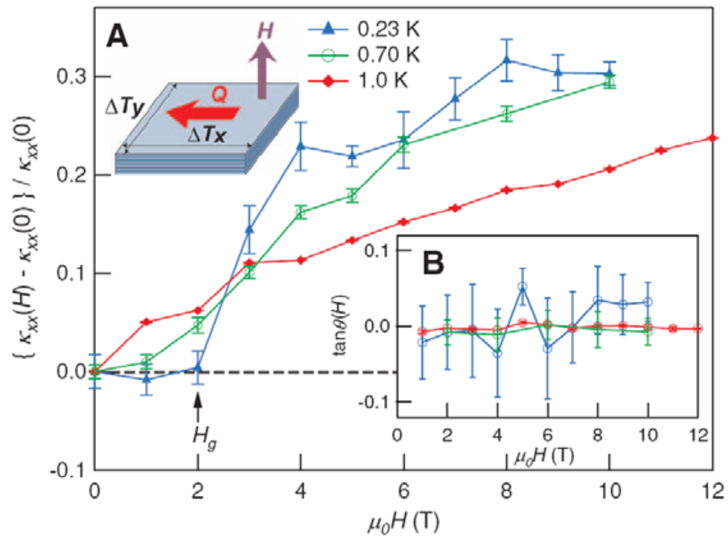
M. Yamashita *et al.*, Nature Phys. **5**, 44 (2009)

Example: Probe the gap of quantum spin liquid

$EtMe_3Sb[Pd(dmit)_2]_2$







$\kappa(T)$: no spin gap（$\gamma \neq 0$）;

$\kappa(H)$: non-zero spin gap

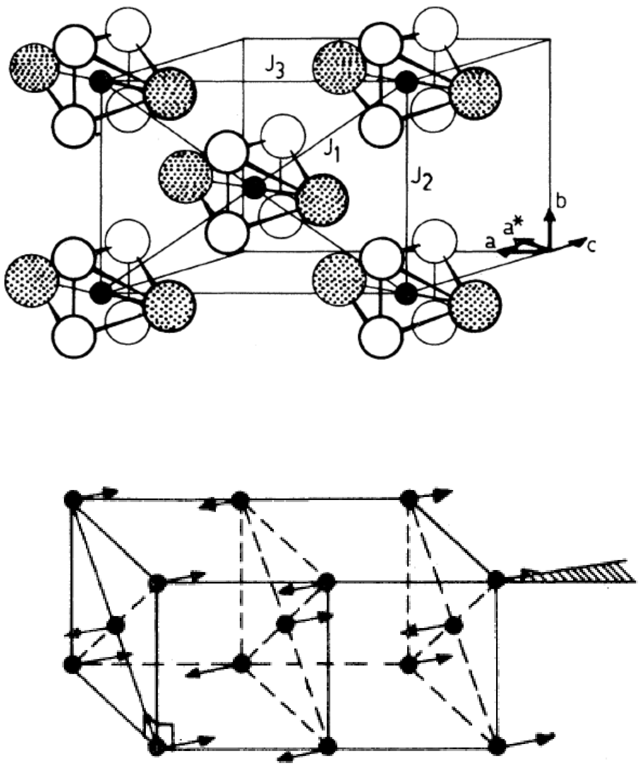
M. Yamashita *et al.*, Science **328**, 1246 (2010)

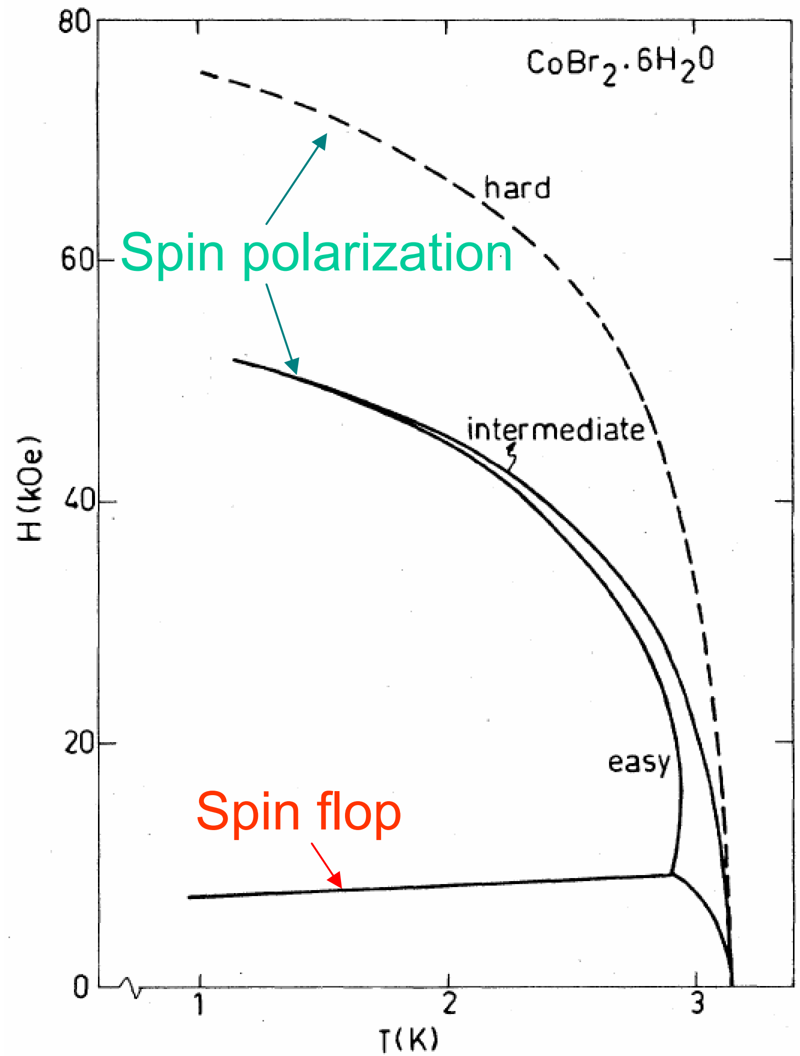
Heat transport is very useful to probe magnetic transitions

- If magnons or spinons can transport, κ_m changes drastically at the magnetic transitions because of the change in magnetic spectrum.
- κ_{ph} also changes at the magnetic transitions, because of the interactions between phonons and magnetic excitations.

Magnetic transitions: An example of 2D XY-AFM

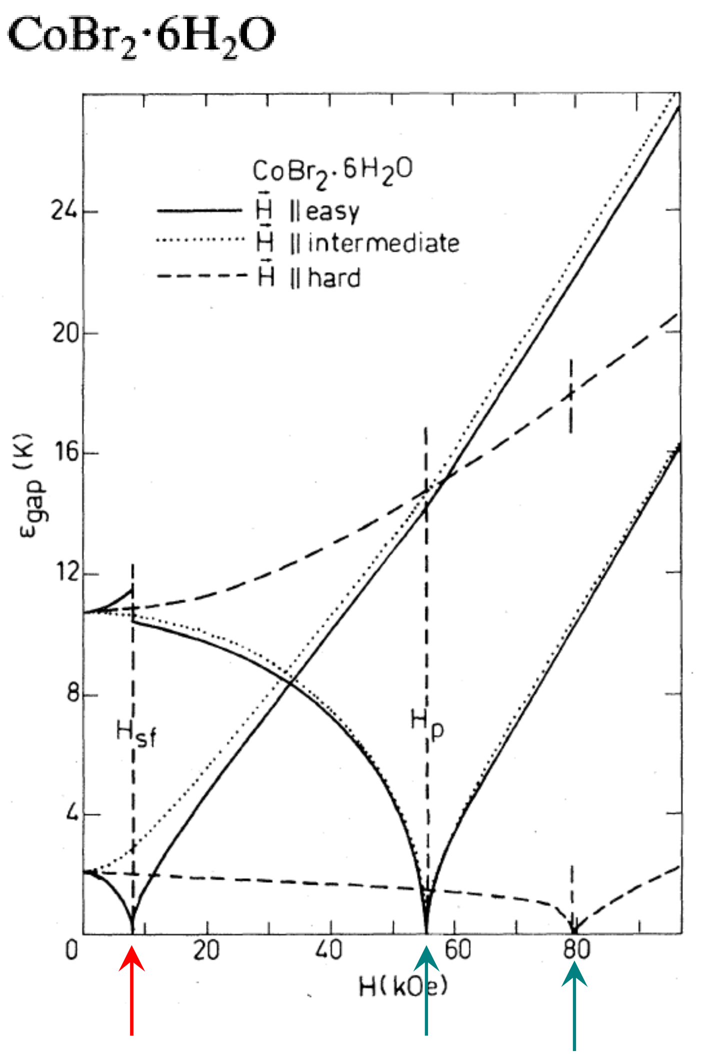
$CoBr_2 \cdot 6H_2O$

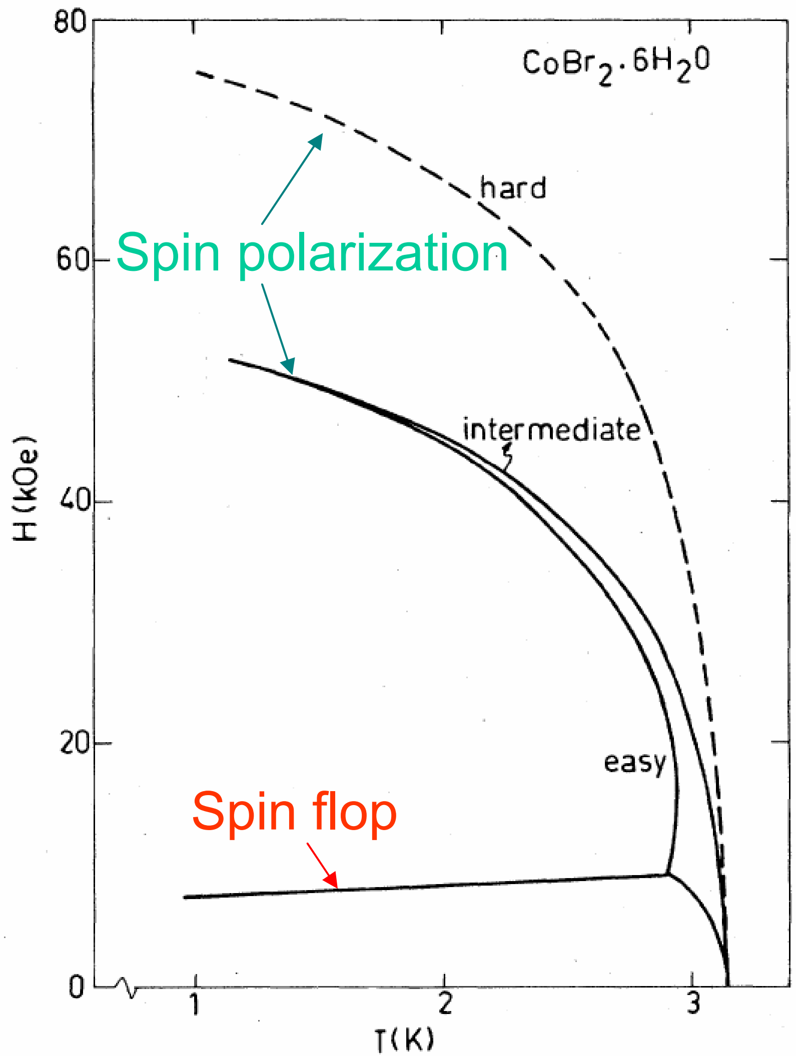




J. A. H. M. Buys and W. J. M. De Jonge,
PRB **25**, 1322 (1982)

Magnetic transitions: An example of 2D XY-AFM





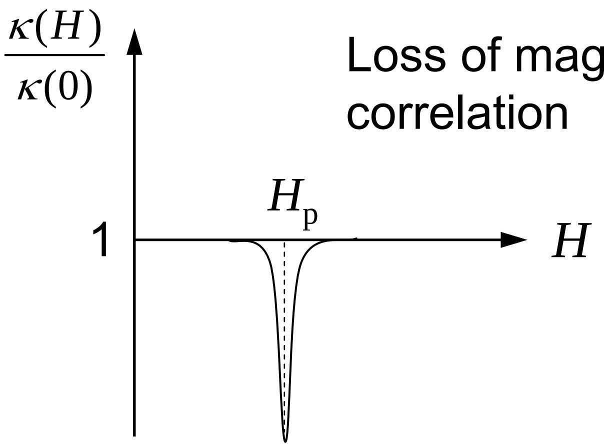
Note that the gaps are reopened after transition.

J. A. H. M. Buys and W. J. M. De Jonge, PRB **25**, 1322 (1982)

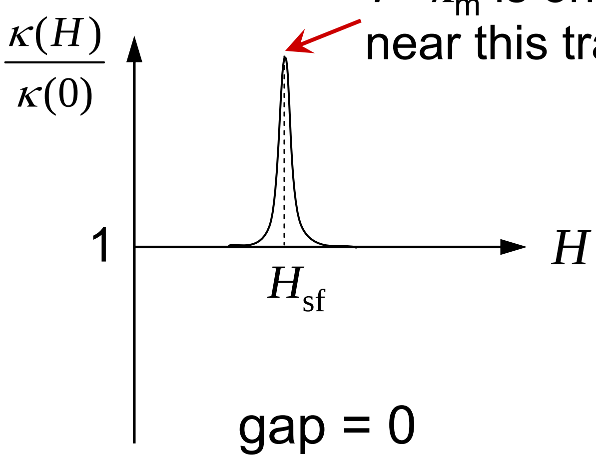
Changes of κ at the magnetic phase transitions

Magnons act as heat carriers

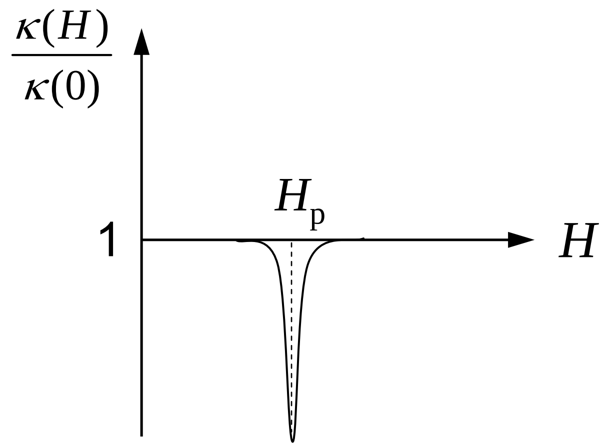
Loss of magnetic correlation



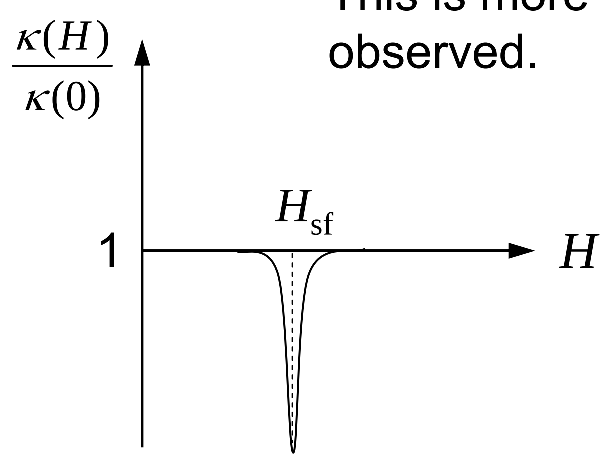
T^3-κ_m is only possible near this transition



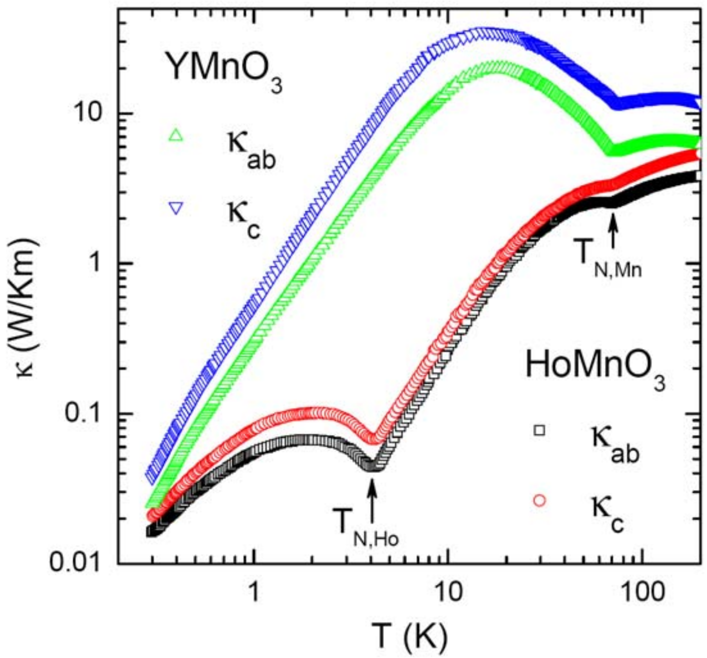
Magnons act as phonon scatterers

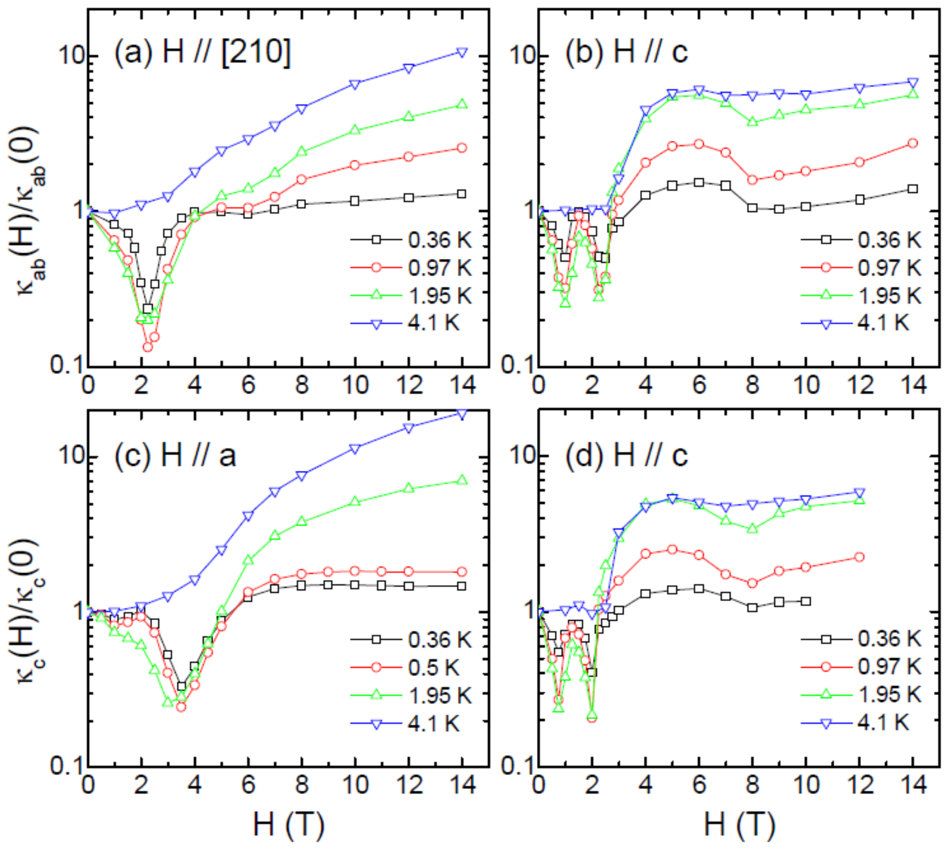


This is more often observed.



Example: Transitions of magnetic structure in $HoMnO_3$

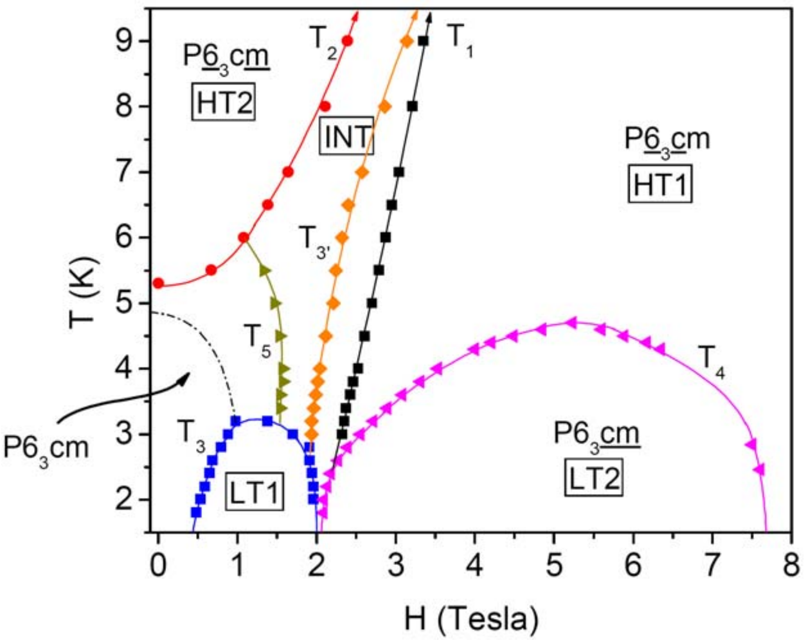




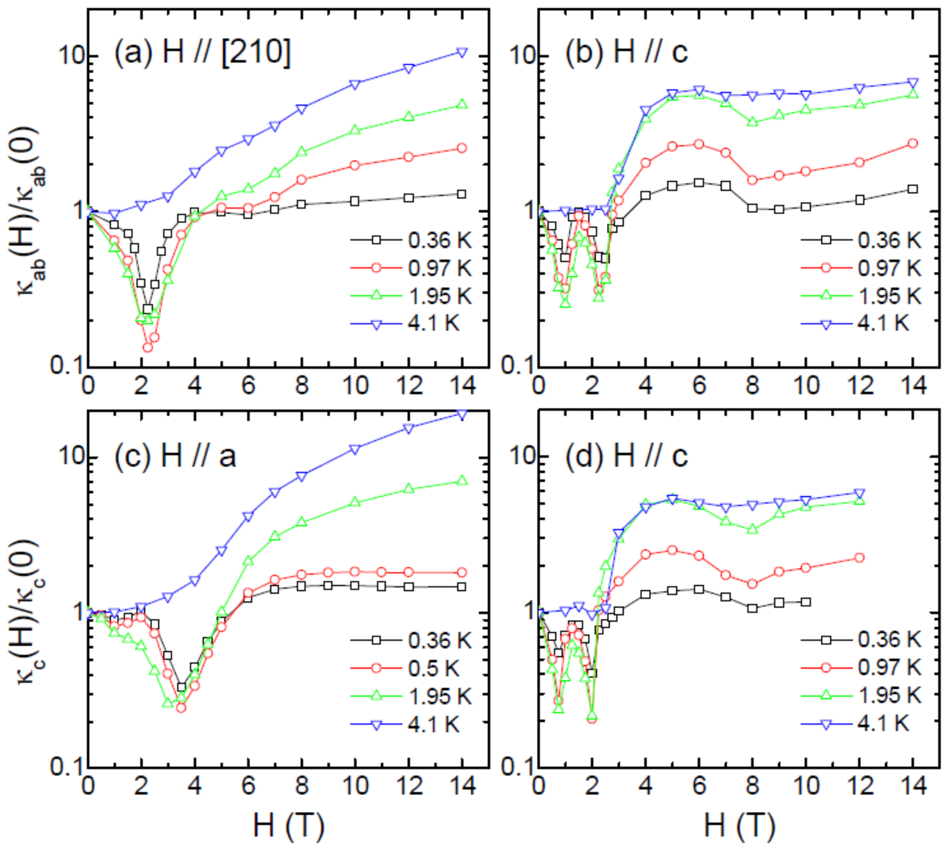
Thermal conductivity can be changed by two orders of magnitude in magnetic fields and the dips of $\kappa(H)$ indicate the spin re-orientations.

X. M. Wang *et al.*, PRB **82**, 094405 (2010)

Example: Transitions of magnetic structure in $HoMnO_3$



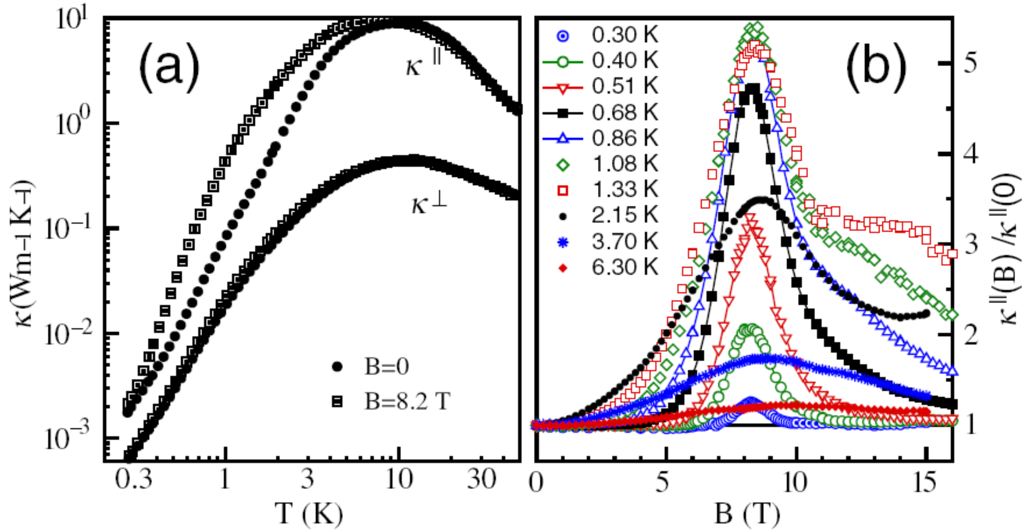
F. Yen *et al*., J. Mater. Res. **22**, 2163 (2007)

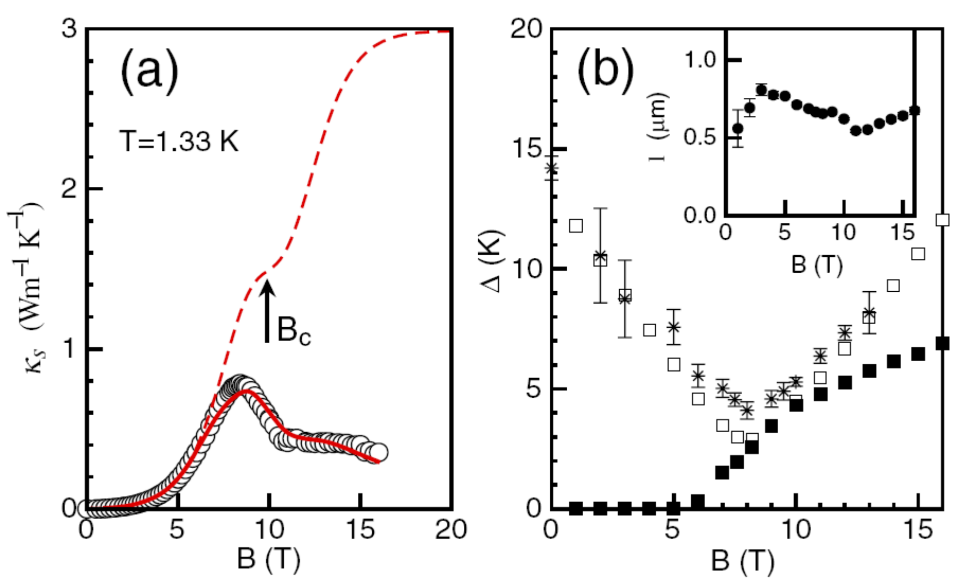


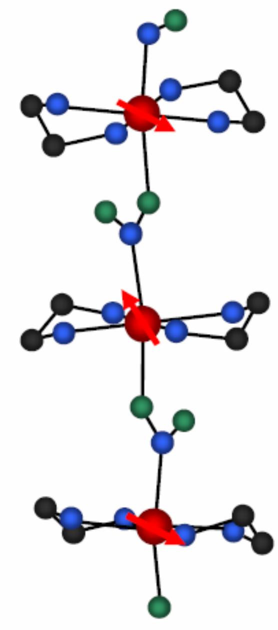
Thermal conductivity can be changed by two orders of magnitude in magnetic fields and the low-field dips of $\kappa(H)$ indicate the spin re-orientations.

X. M. Wang *et al.*, PRB **82**, 094405 (2010)

Example: magnetic transport in $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP)







In zero field, the spin thermal conductivity is negligible at *T* << 12.2K (spin gap).

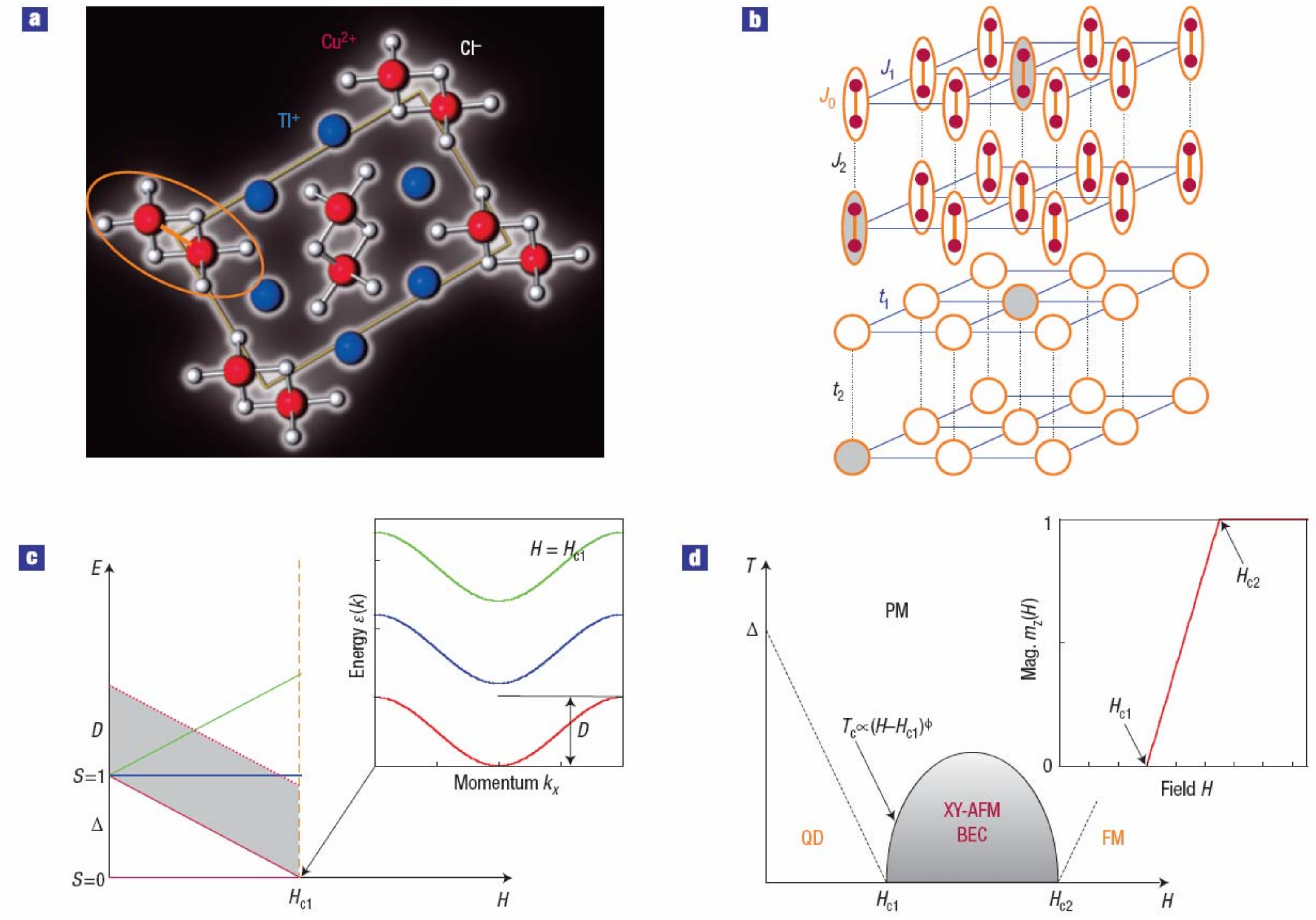
The magnetic thermal conductivity appears in applied magnetic field.

A. V. Sologubenko *et al.*, PRL **100**, 137202 (2008)

Field-induced QPTs in spin-gapped materials

3D $TlCuCl_3$

T. Giamarch *et al.*, Nature Phys. **4**, 198 (2008)

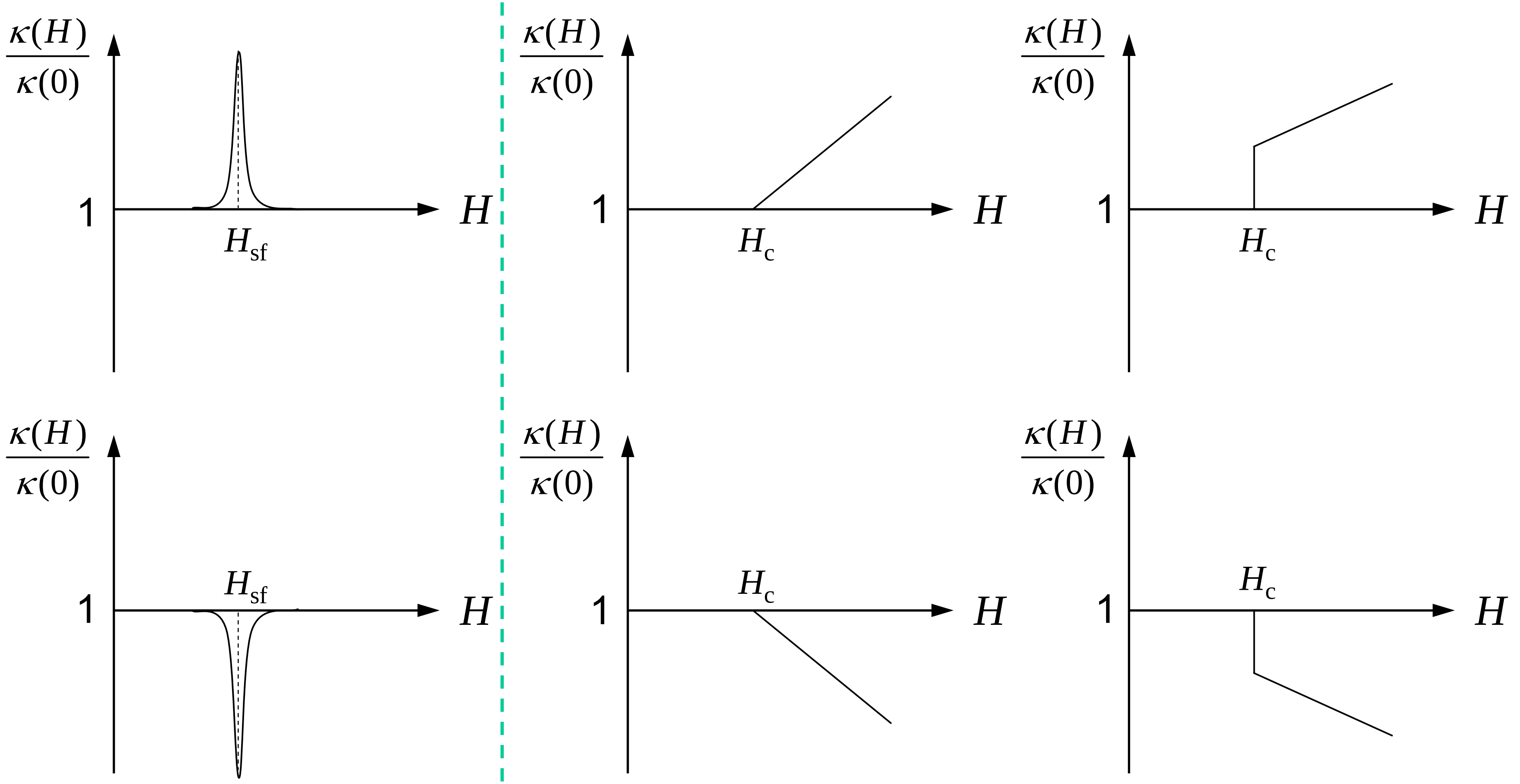


Magnon Bose-Einstein Condensation?

Magnon BEC state is expected to be ungapped

Spin flop

BEC transition



The observation of ballistic magnon transport would be a strong support for BEC.

Heat transport as a probe: shortcomings or complications

- Usually the *T*- or *H*-dependencies of κ is very complicated!

$$\kappa = \kappa_{ph} + \kappa_e + \kappa_m$$

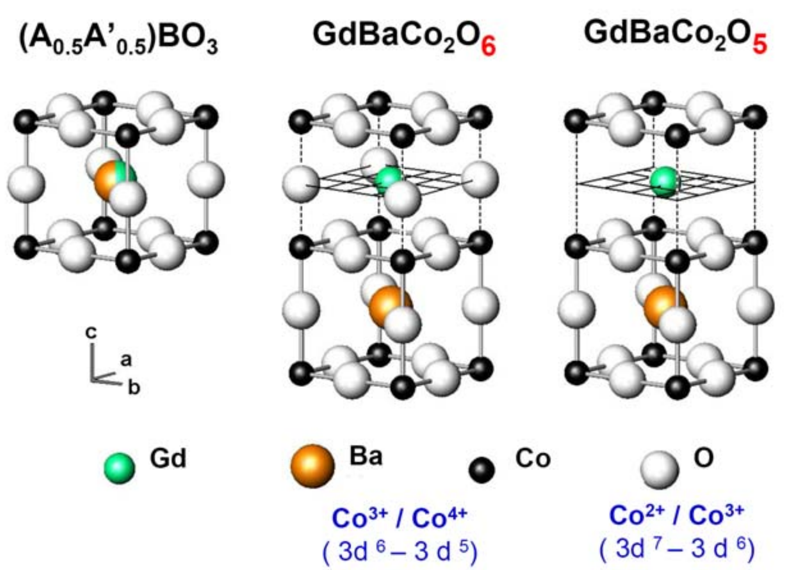
- Insulator: $\kappa_e = 0$
- Phonon conductivity has complicated *T* dependence
- Magnetic thermal conductivity is definitely dependent on field and also has a complicated *T* dependence
- Phonon conductivity can also change with magnetic field because of the interactions between phonons and magnetic excitations

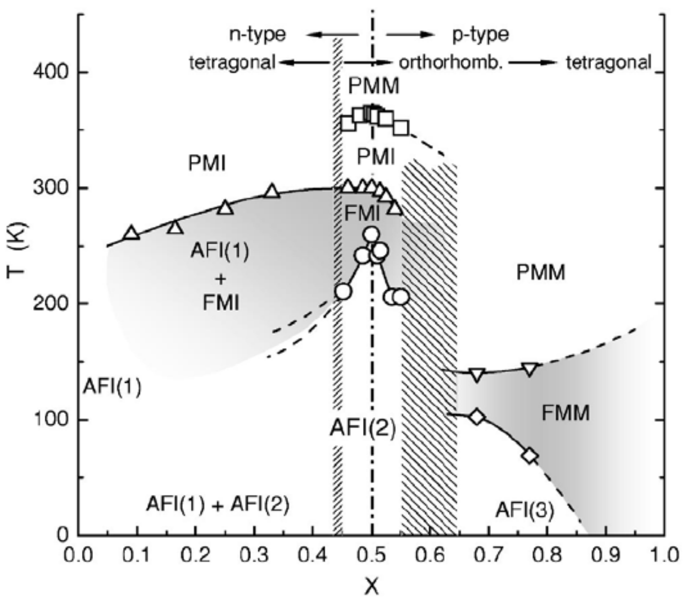
It is in general very difficult to separate κ_{m} and κ_{ph}.

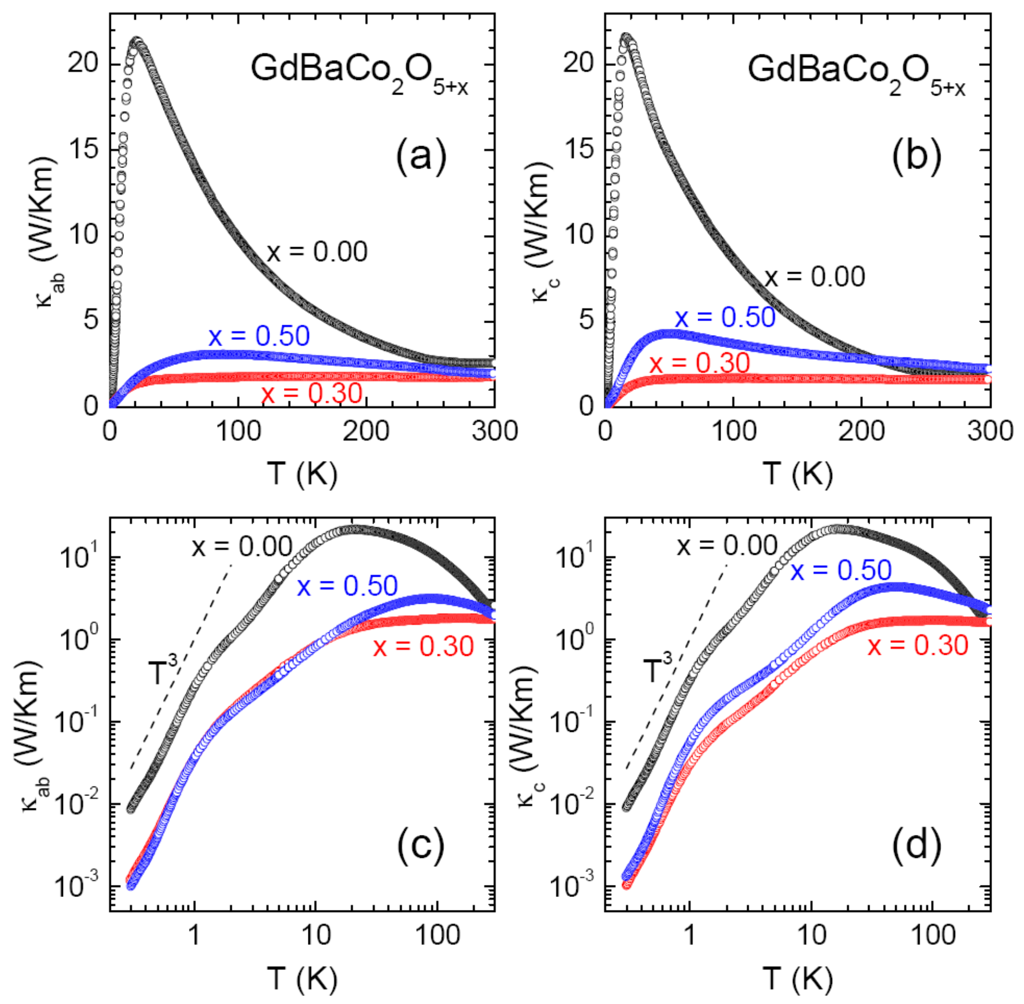
Very low-temperature result is the easiest one to deal with.

Paramagnetic scattering effect on phonons

Large magnetothermal conductivity in $GdBaCo_2O_{5+x}$

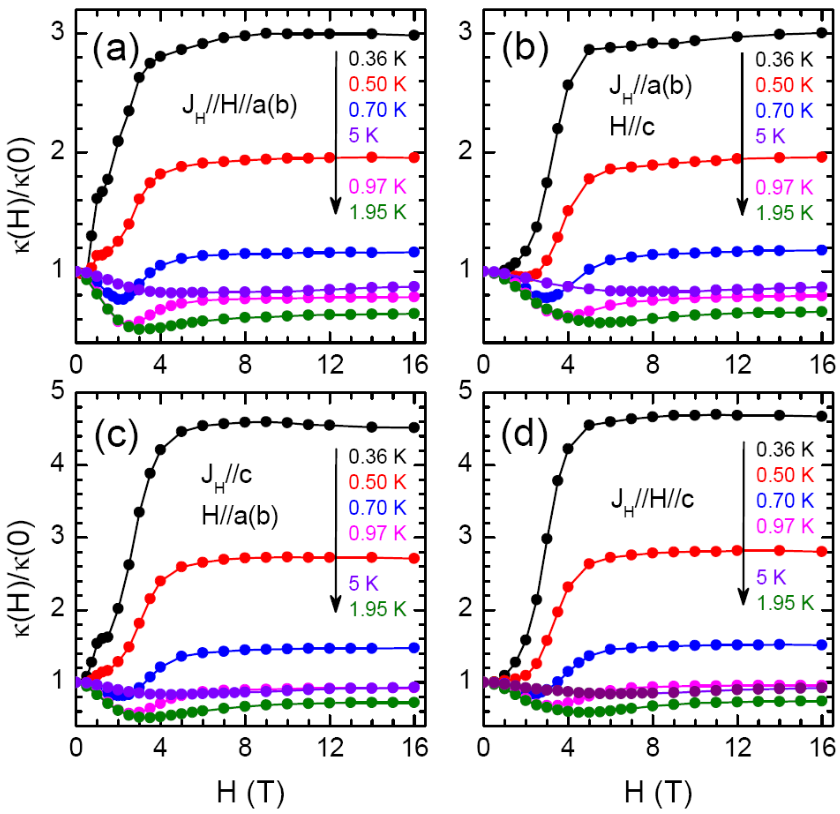


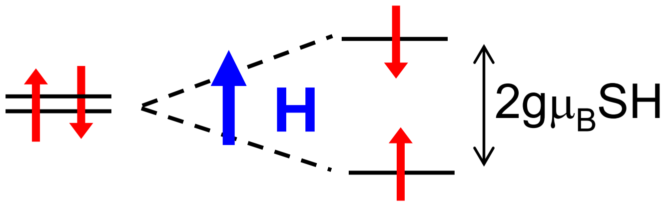


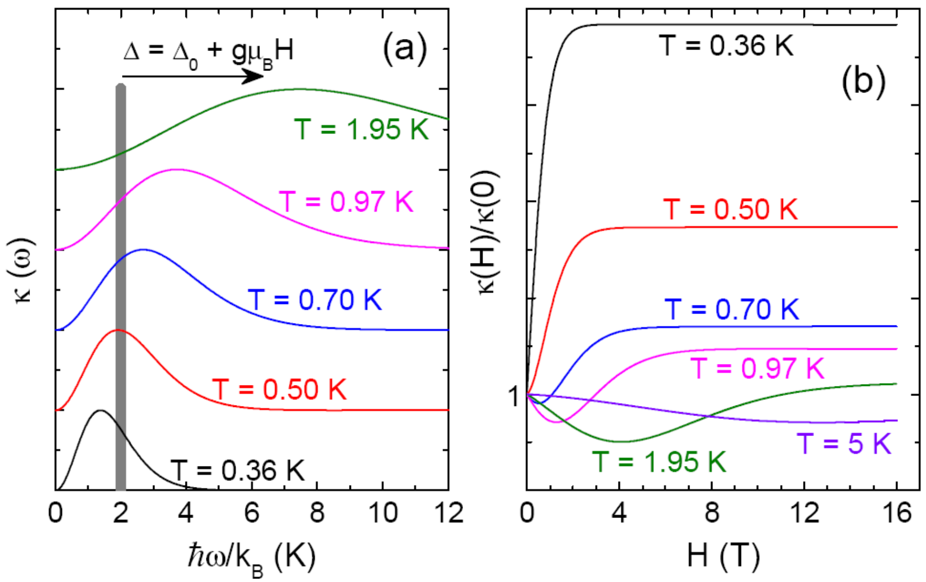


X. F. Sun *et al.*, PRB **77**, 054436 (2008)

Paramagnetic scattering effect







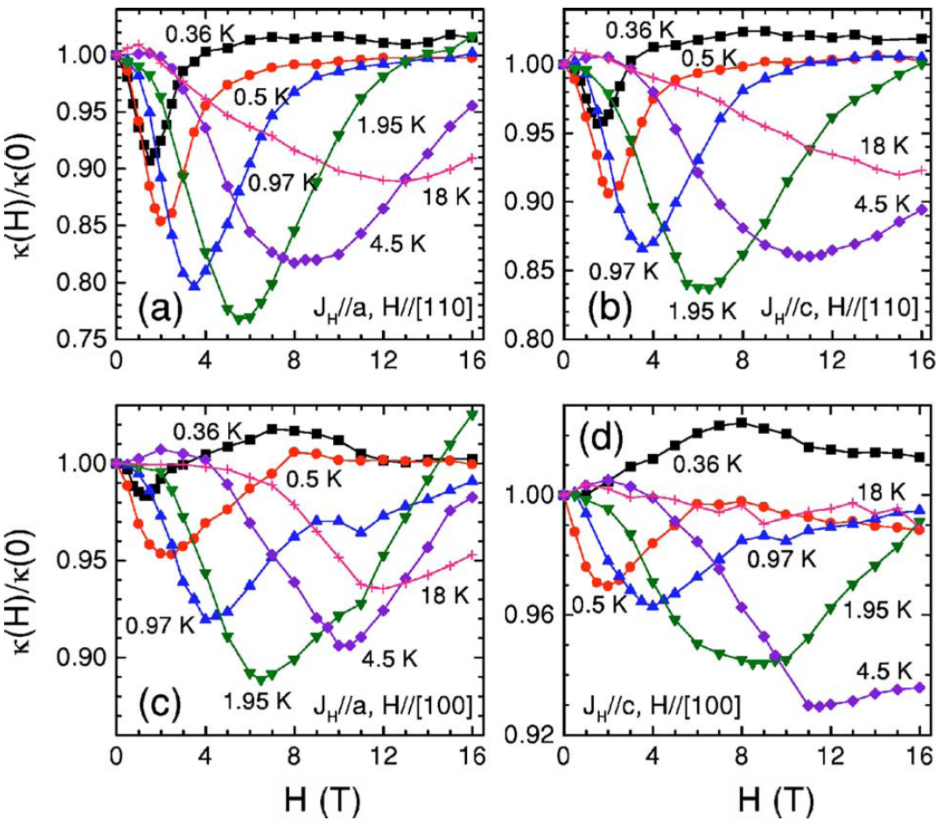
Calculations based on spin-phonon scattering

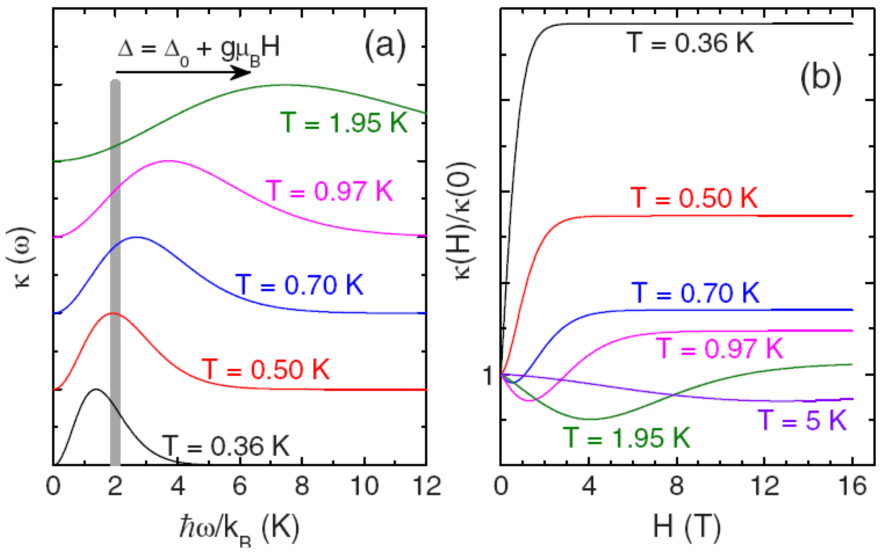
- This indicates strong spin-phonon coupling in transition-metal oxides.

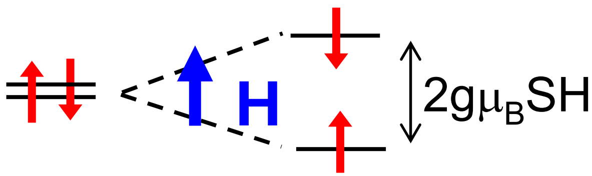
X. F. Sun *et al.*, PRB **77**, 054436 (2008)

Paramagnetic scattering in a parent cuprate

$Pr_{1.3}La_{0.7}CuO_4$







The influence of spin-flop of Cu ions on $\kappa(H)$ is negligible.

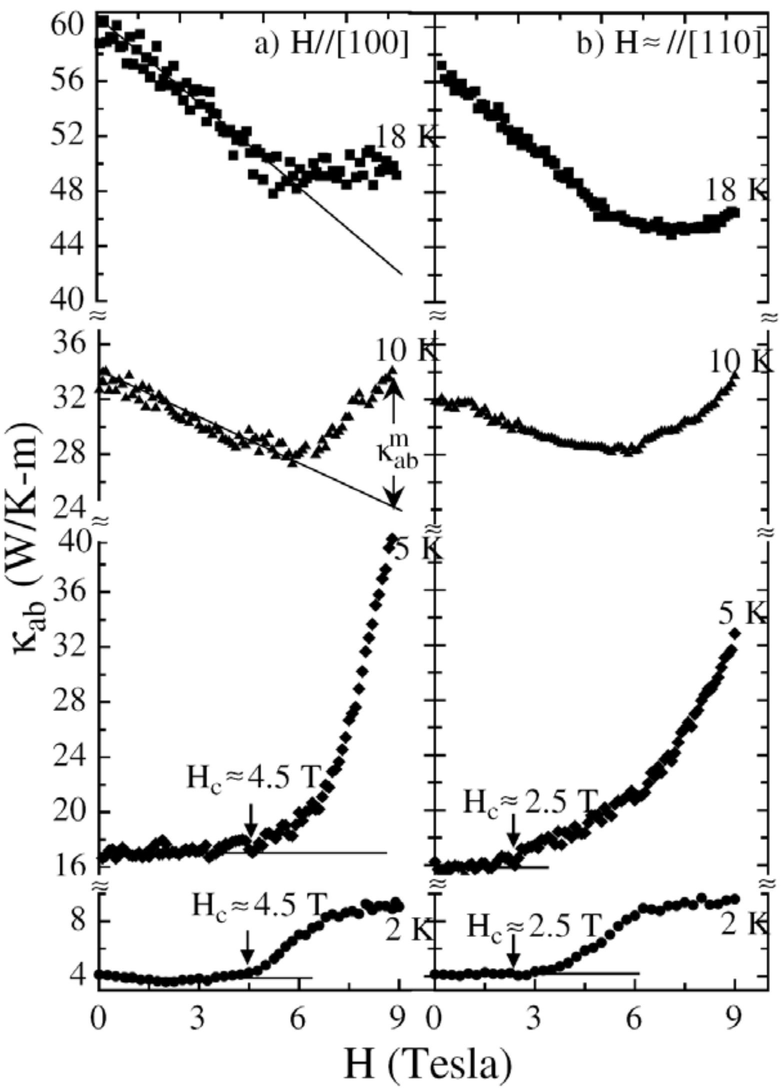
Paramagnetic scattering can induce H-dependence of phonon transport.

X. F. Sun *et al*., PRB **72**, 104501 (2005)

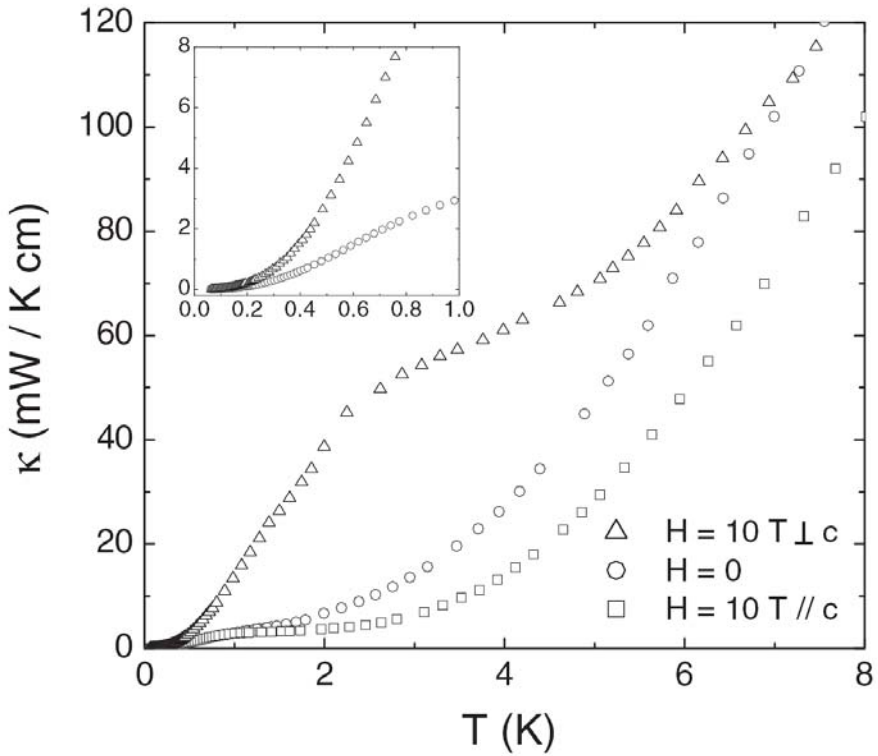
Result I:

Low-*T* heat transport of Nd_2CuO_4

Nd_2CuO_4: strong magnetic field-dependence of κ



Is the high-field increase of κ a contribution of magnon transport?

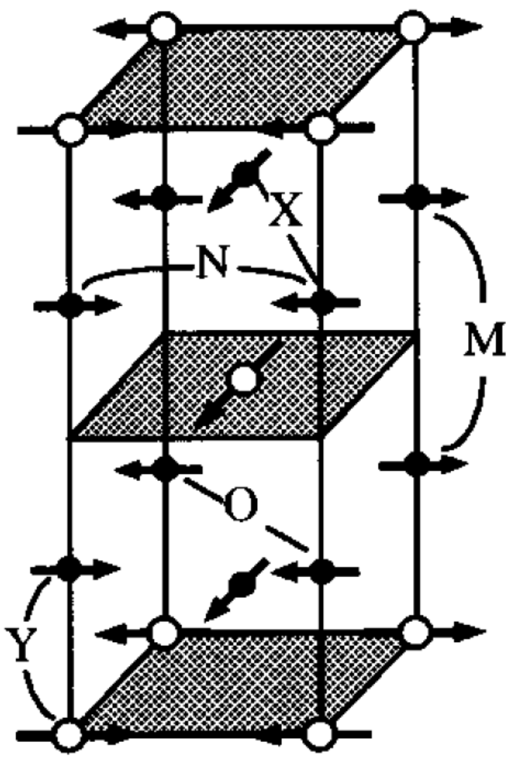


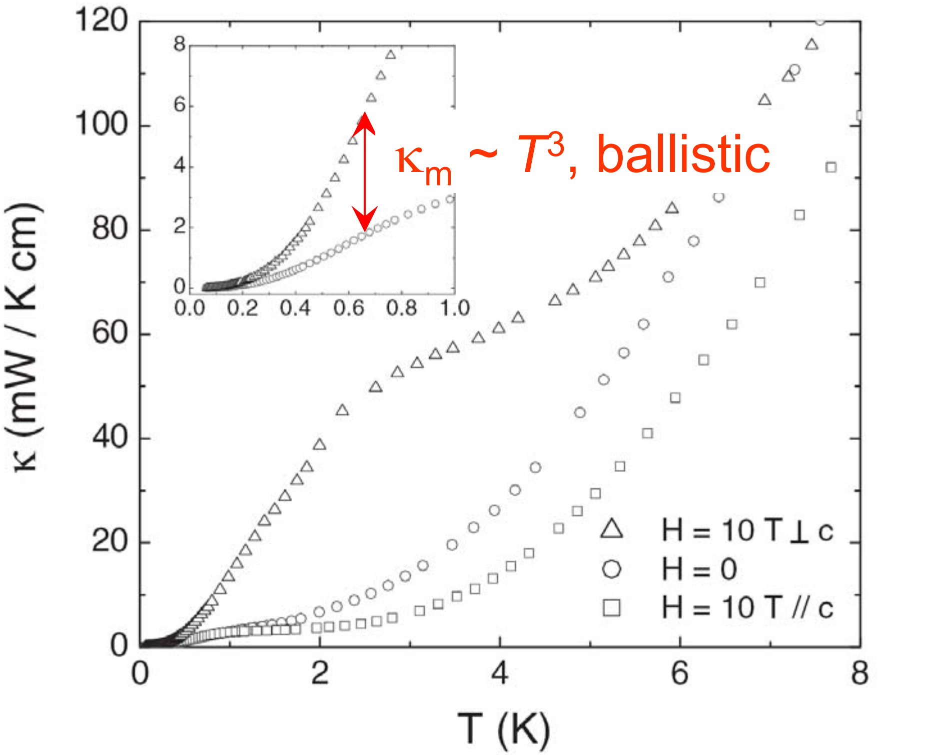
R. Jin *et al.*, PRL **91**, 146601 (2003)

S. Y. Li *et al.*, PRL **95**, 156603 (2005)

Nd magnon heat transport in the Cu spin-flop state?

$T_N^{Cu} = 255\text{K}$ $T_N^{Nd} = 1.5\text{K}$



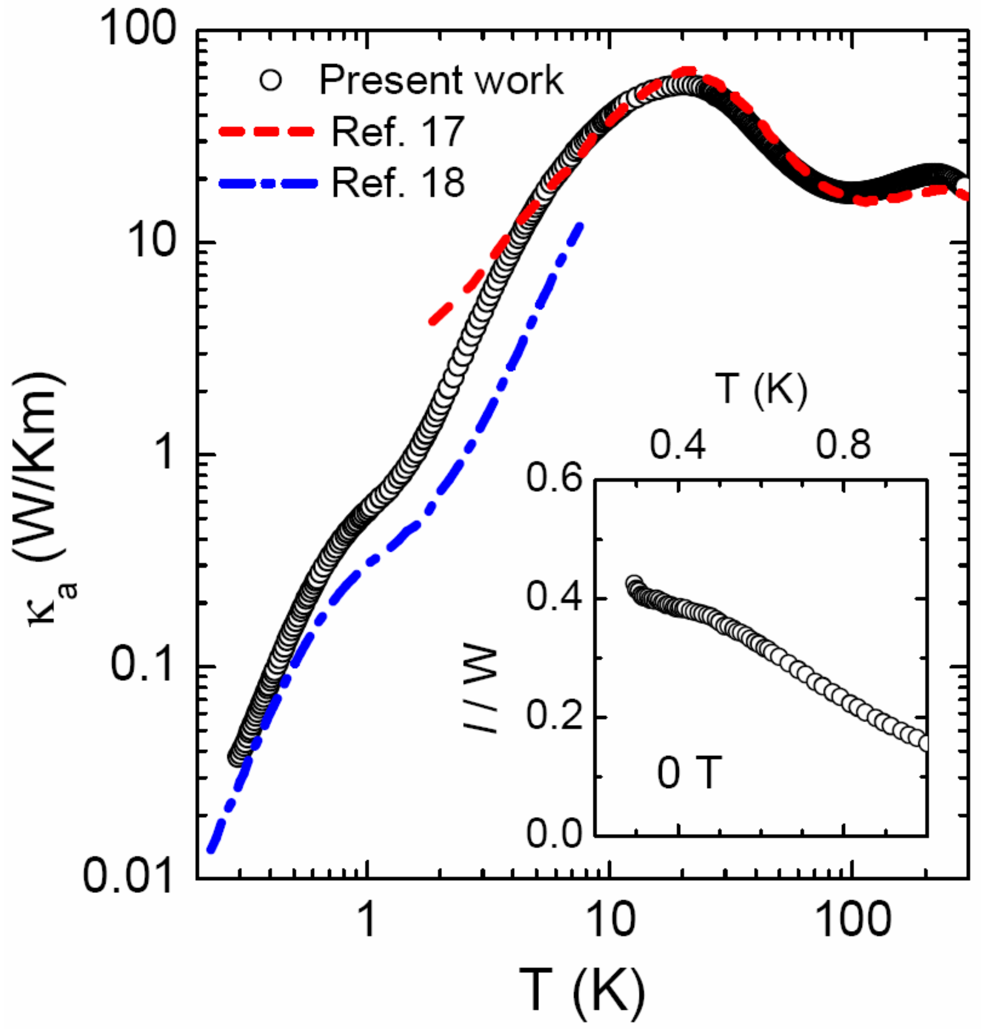


Cu spin flop at ~ 4.5 T for H//[100] and ~ 0.7 T for H//[110]

The explanation is unreasonable since the anisotropy gap is opened at 10 T!

S. Y. Li *et al.*, PRL **95**, 156603 (2005)

Q: Different impacts of the ab-plane and the c-axis fields



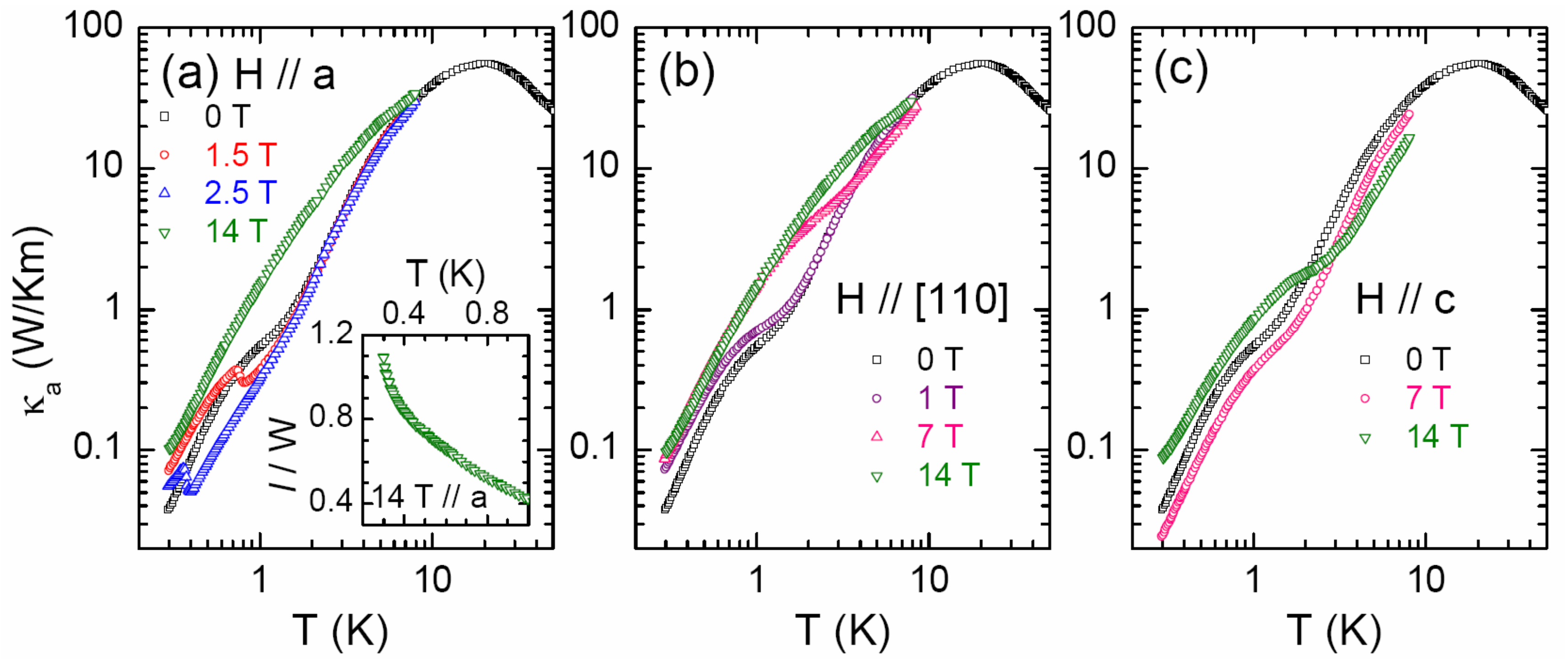
$$\kappa = \frac{1}{3}Cvl$$

$$C = \beta T^3$$

$$W = 2\sqrt{\frac{S}{\pi}}$$

Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

$\kappa(T)$: The direction of field is crucial



The phonon scattering seems to be smeared out in high in-plane field, which means that the magnetic scattering is rather strong in zero field.

Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

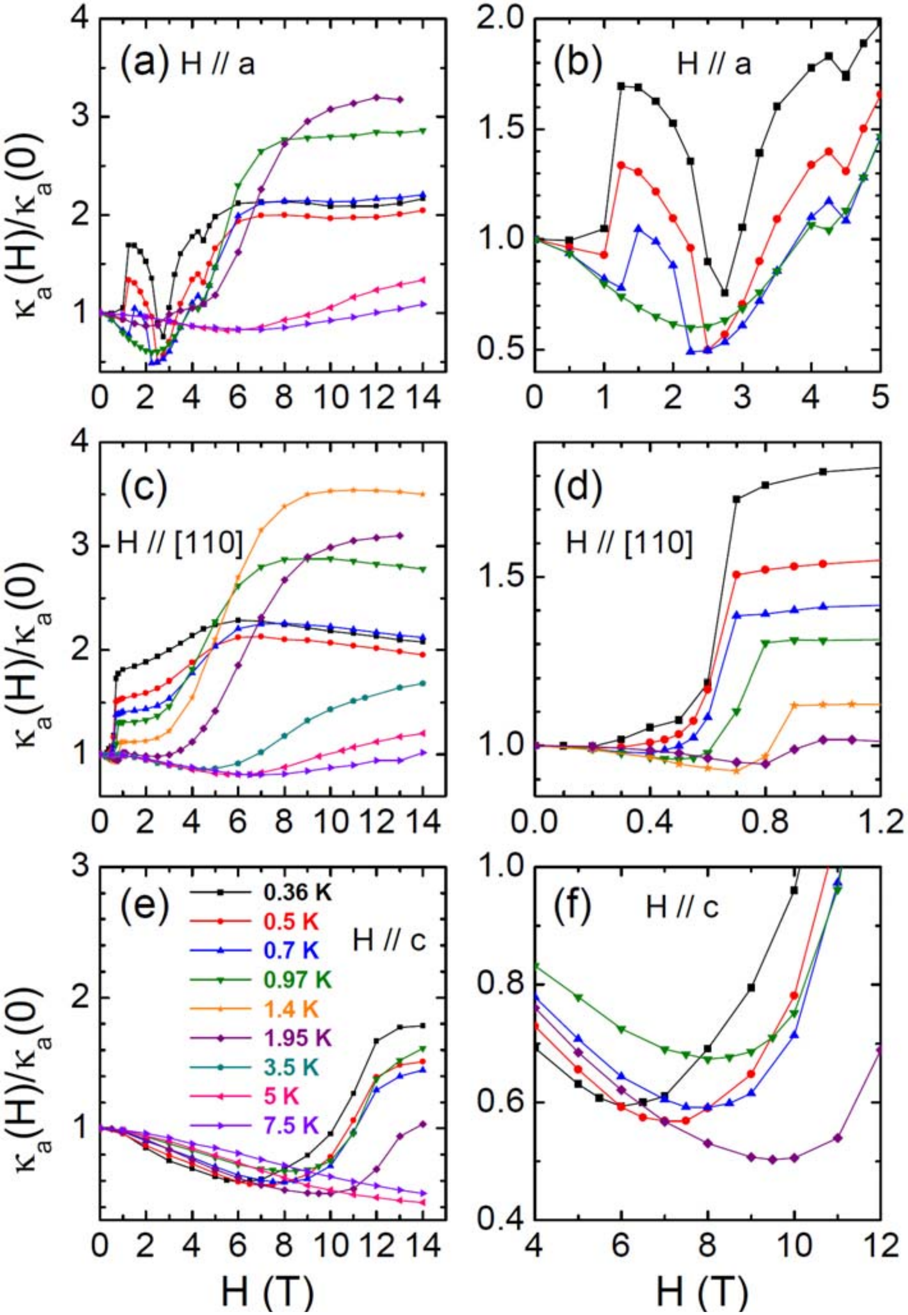
$\kappa(H)$

(i) The c-axis field induces strong changes of thermal conductivity.

Note that the c-axis field does not change the spin structures!

The phonon scattering by paramagnetic moments must be significant!

This effect is usually isotropic.



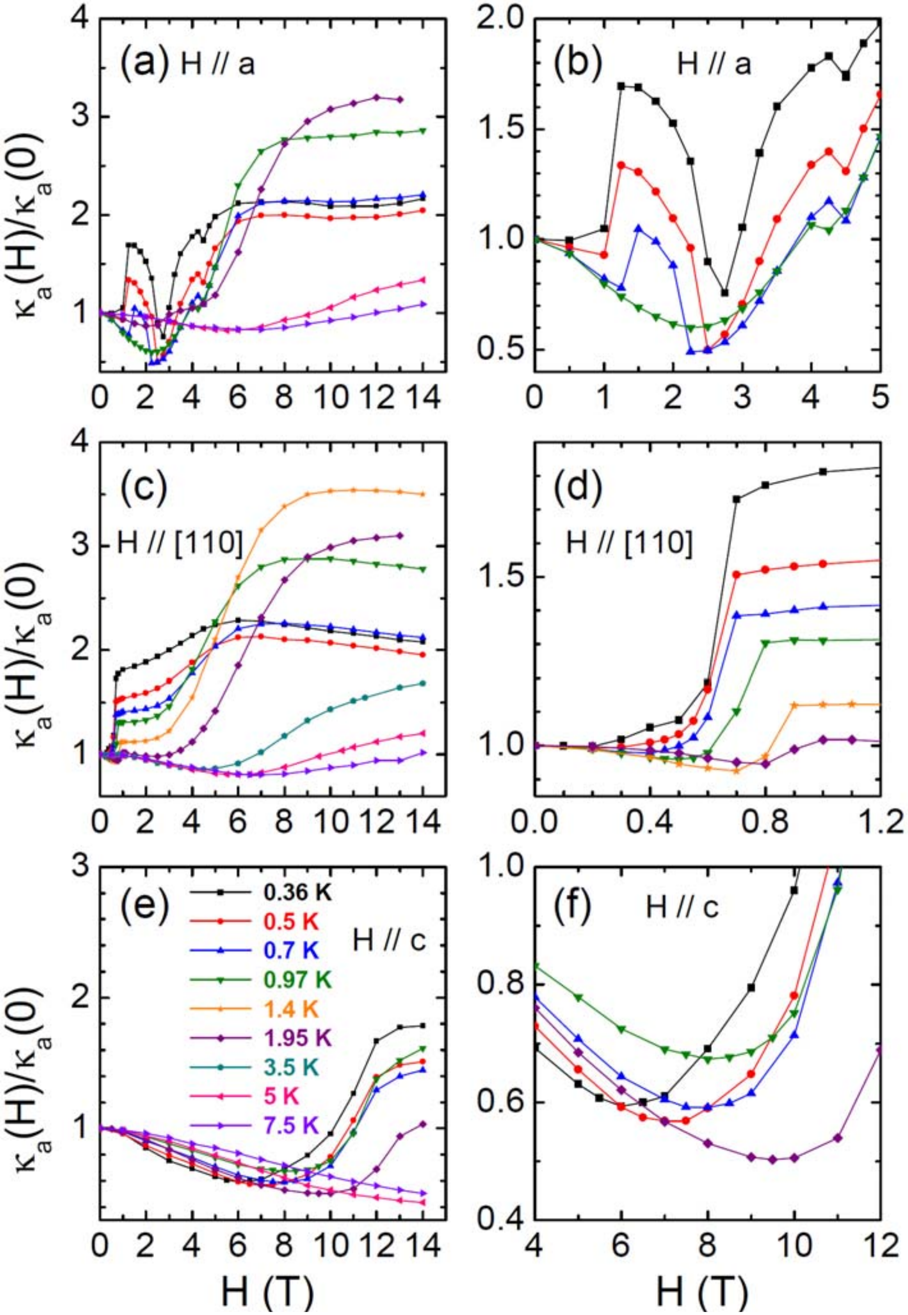
Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

$\kappa(H)$

(ii) The changes of Cu spin structure affect the heat transport very weakly.

Note the 4.5-T spin flop for H//a!

The strong low-field dependence of κ must be related to the changes of Nd spin structure!

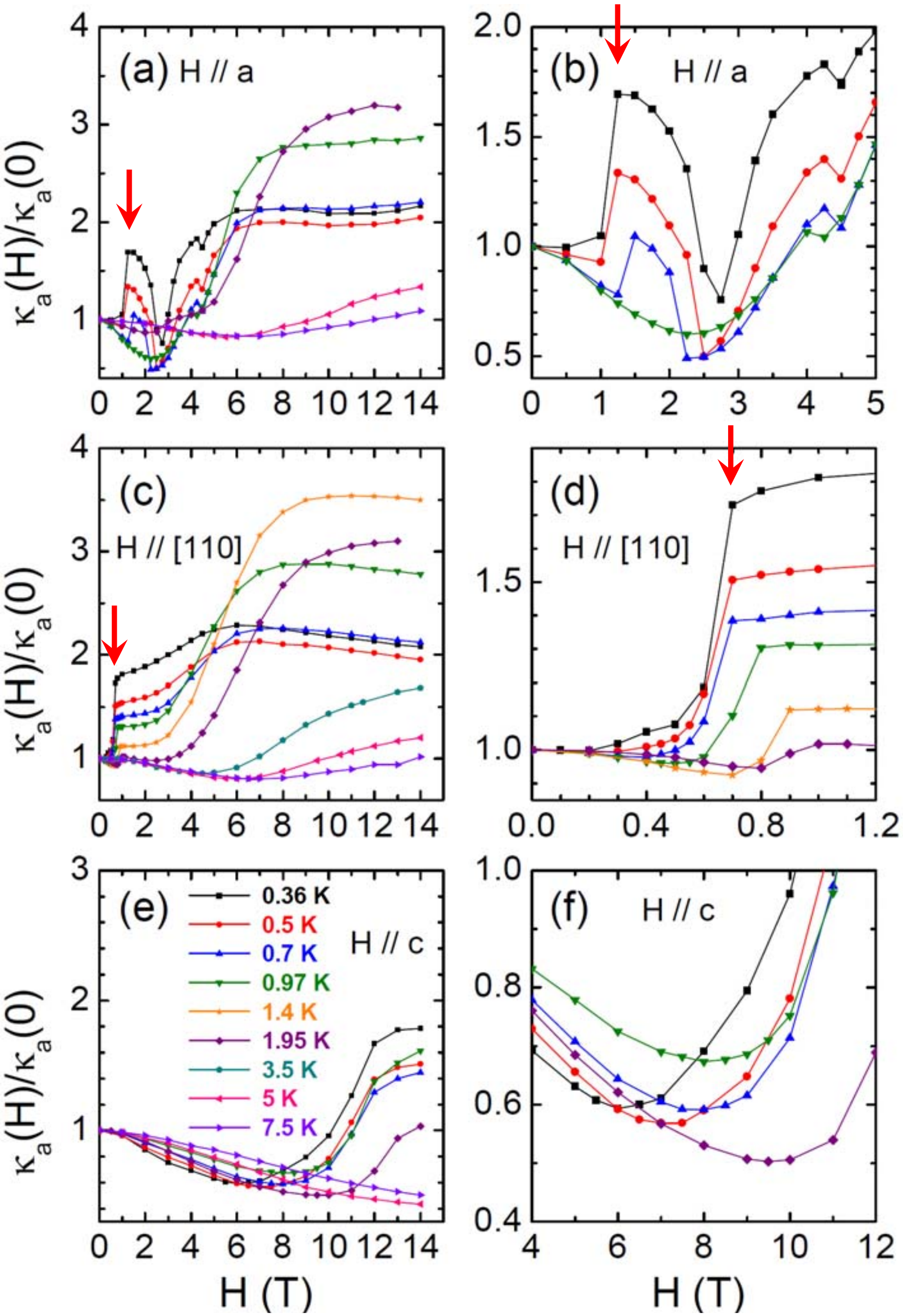


Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

$\kappa(H)$

(iii) Nd magnons can transport heat, but only in low fields.

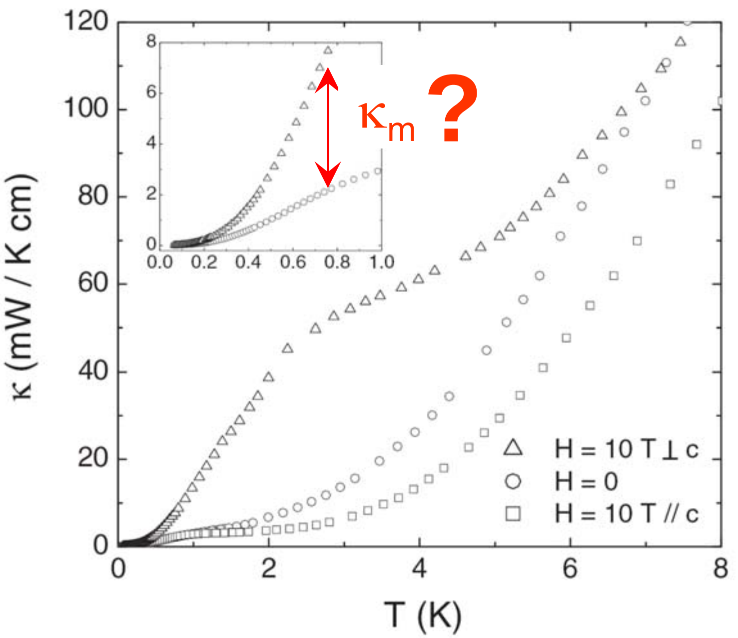
Note the low-field step-like increase of κ!
This is likely due to the spin-flop transition of Nd moments.

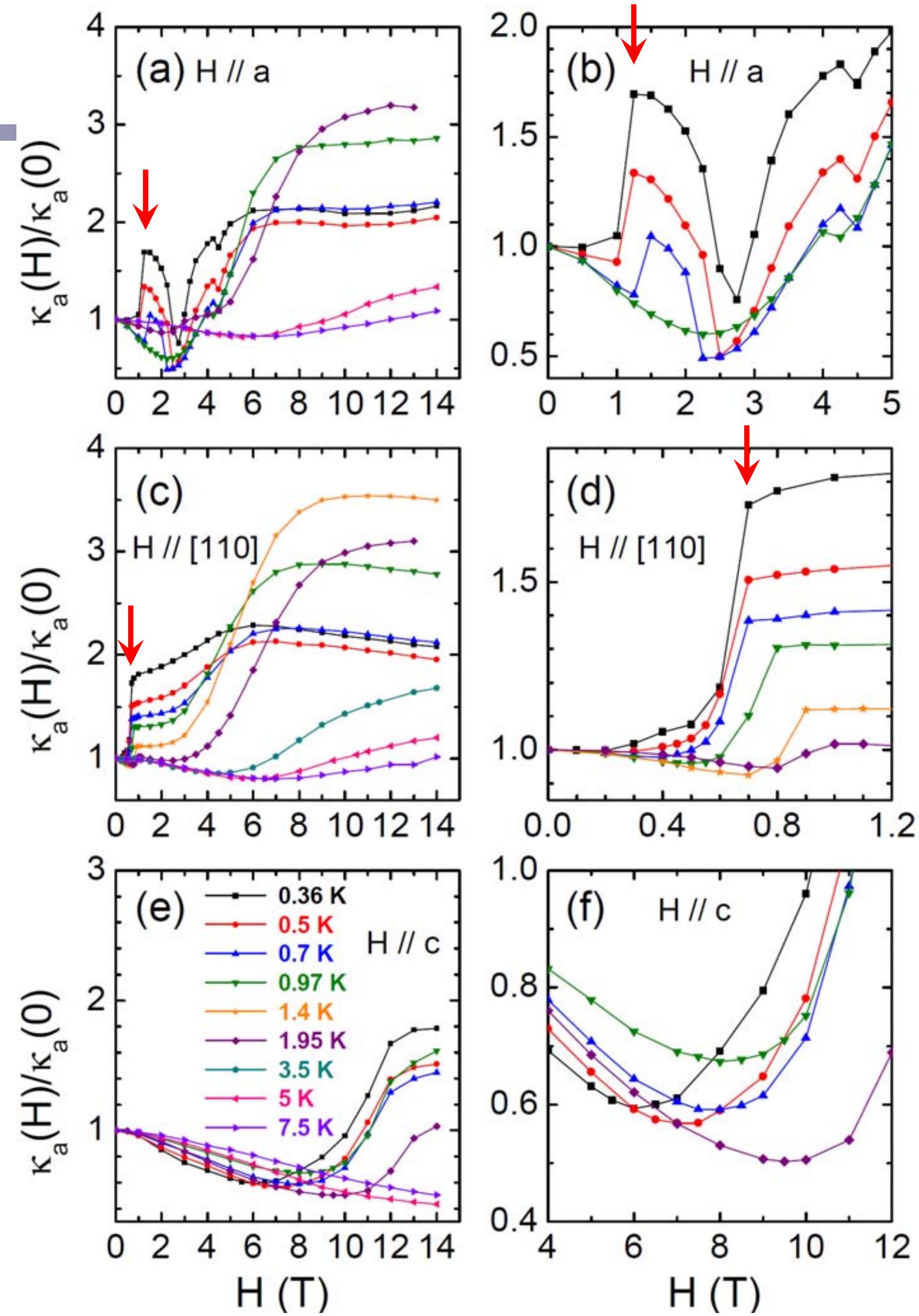


Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

$\kappa(H)$

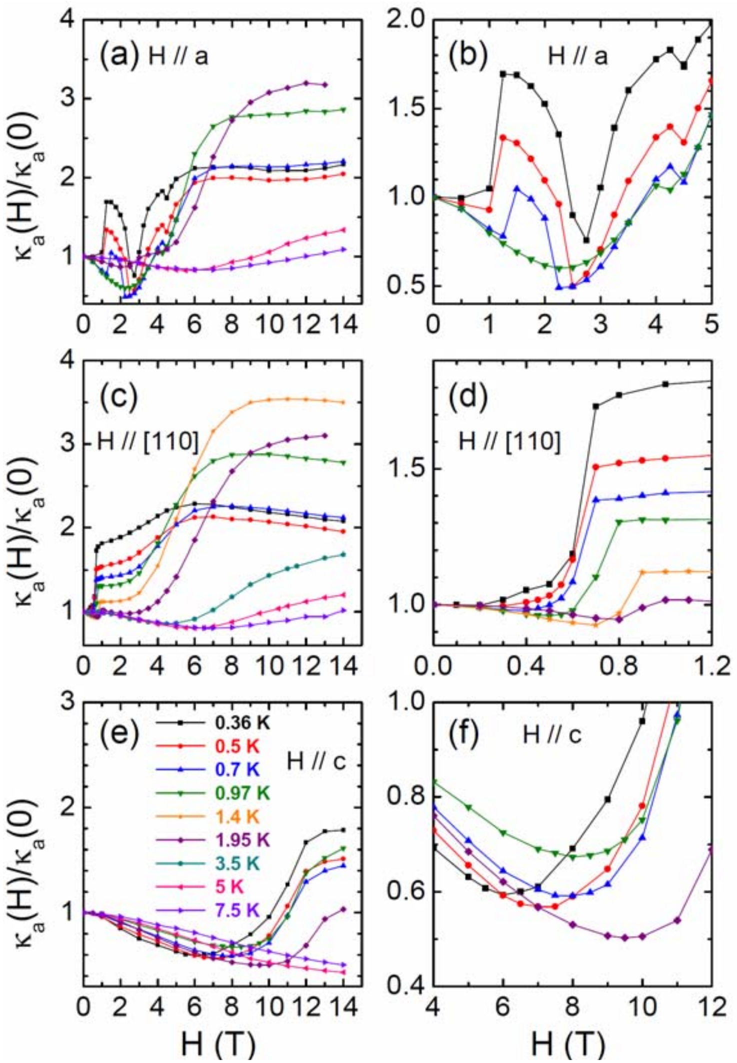
(iv) The conclusion of an earlier work is misleading and was arrived by an accidentally unreasonable choose of magnetic fields.

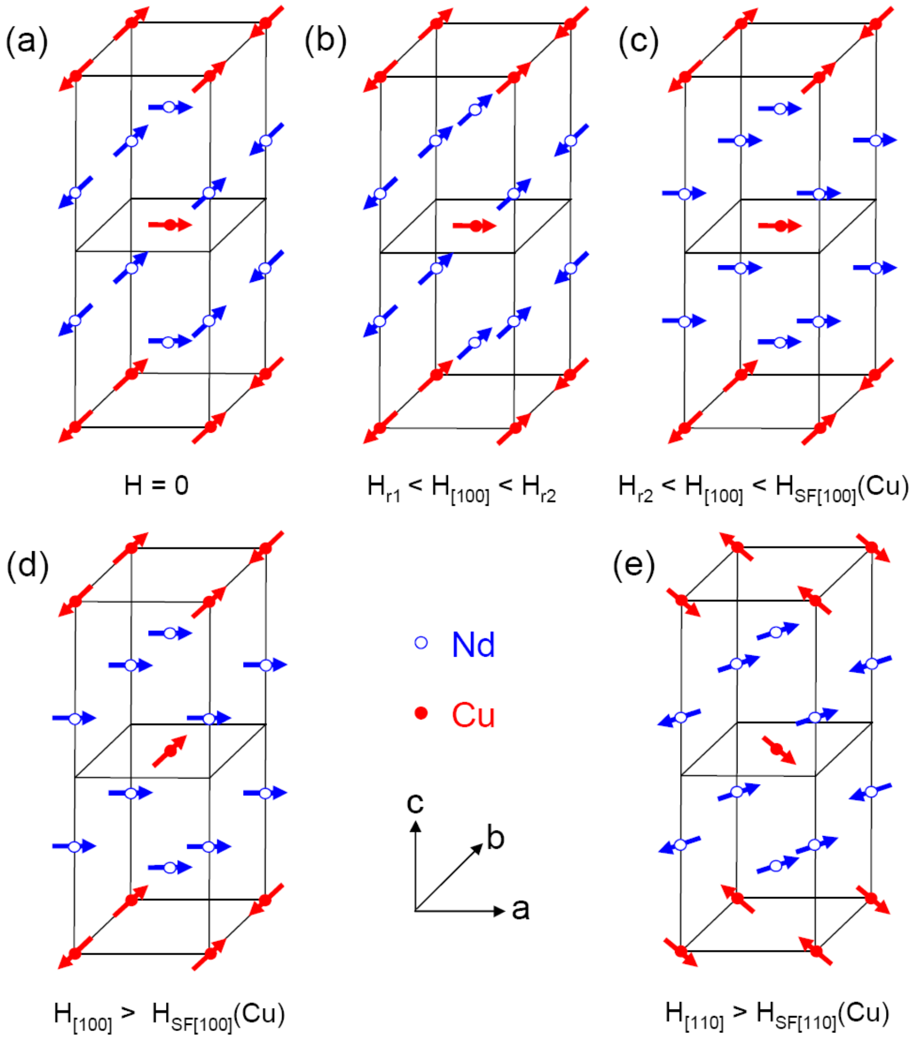




Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

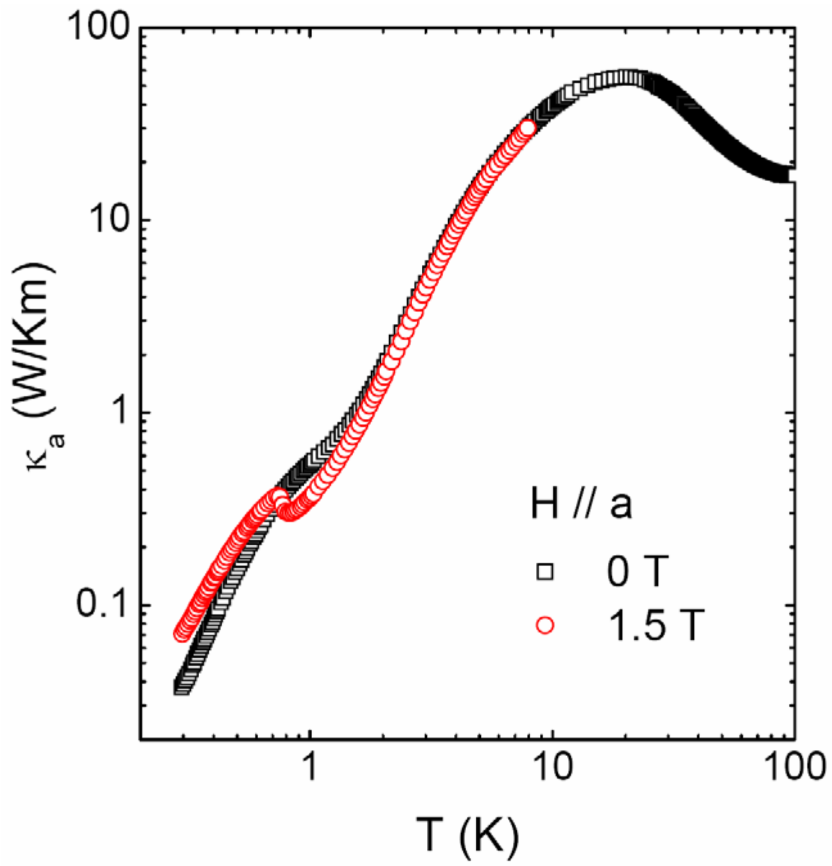
Spin structures and their transitions in magnetic field deduced from the heat transport

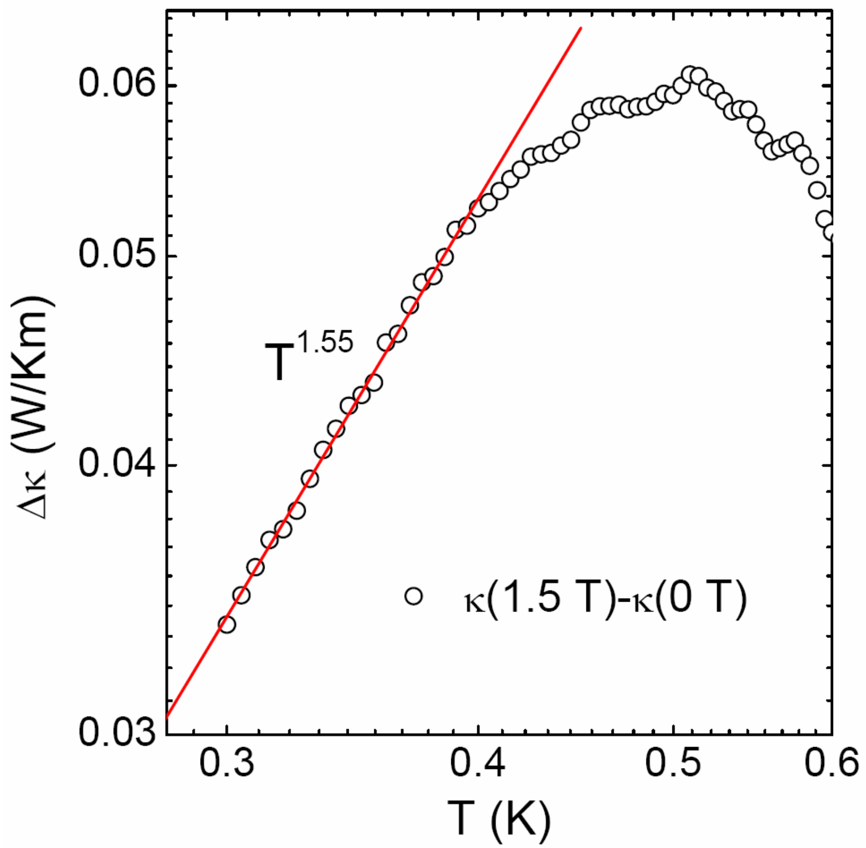




Z. Y. Zhao *et al.*, PRB **83**, 174518 (2011)

Temperature dependence of the magnon conductivity





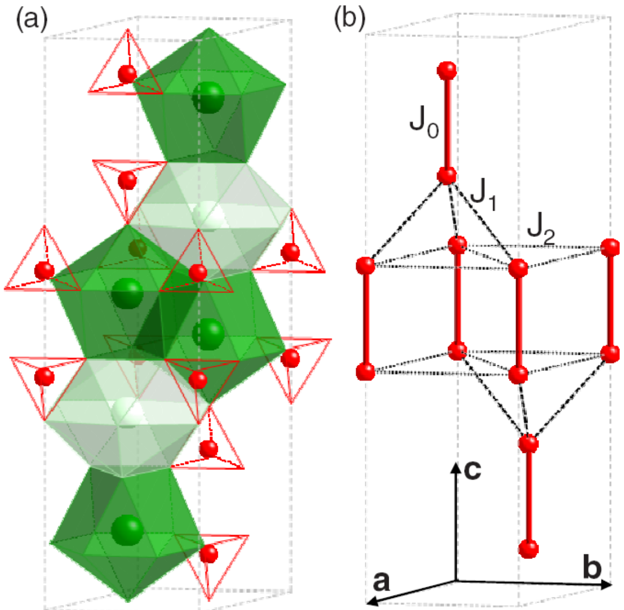
The increase of κ at the Nd^{3+} spin flop does not follow a ballistic behavior: it is mixed with a field-induced change of phonon conductivity.

Summary (I)

- The influence of Cu^{2+} spin flop on thermal conductivity is very weak.
- Nd^{3+} magnons can act as heat carriers in the spin-flop state of Nd^{3+} spins.
- The enhancement of $\kappa(H)$ at high field is mainly due to the weakening of magnetic scattering on phonons, rather than the magnon heat conduction.
- κ_m at the spin-flop field cannot be separated, because of the effect of magnetic scattering.
- There is still no convincing example of the T^3 ballistic magnon transport.

Result II:
Low-*T* heat transport of magnon BEC materials

Example I: $Ba_3Mn_2O_8$

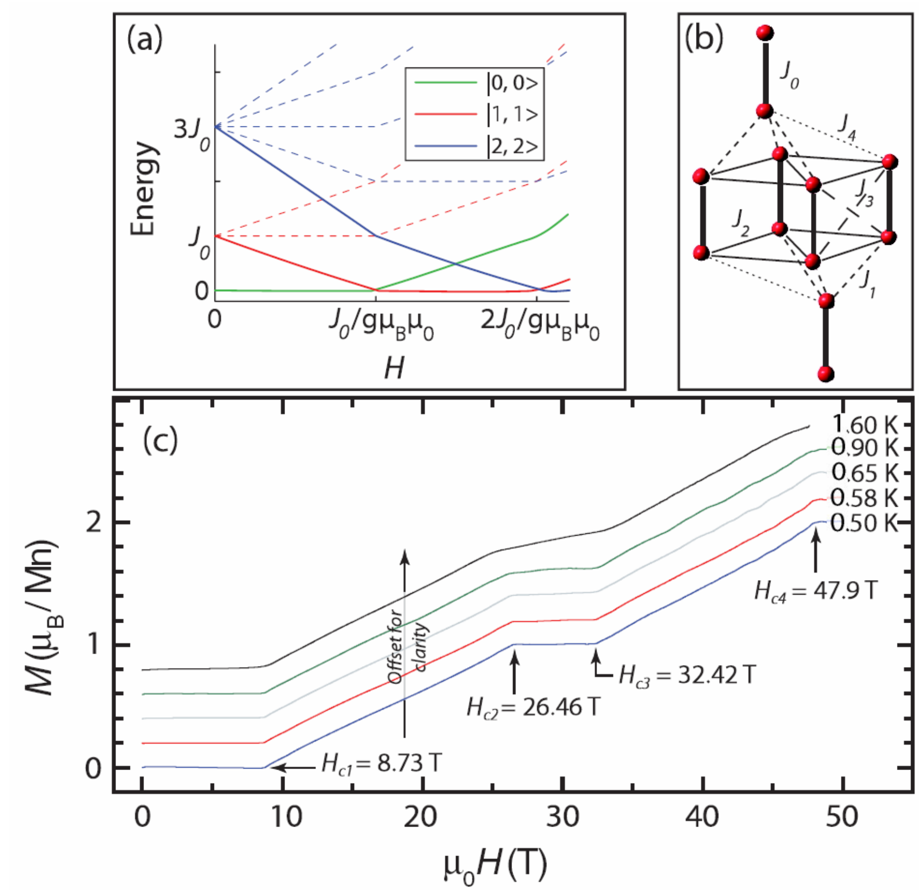


space group: *R3m*

a = 5.711Å, *c* = 21.444Å
Mn^{5+} ion ($3d^2$): *S* =1

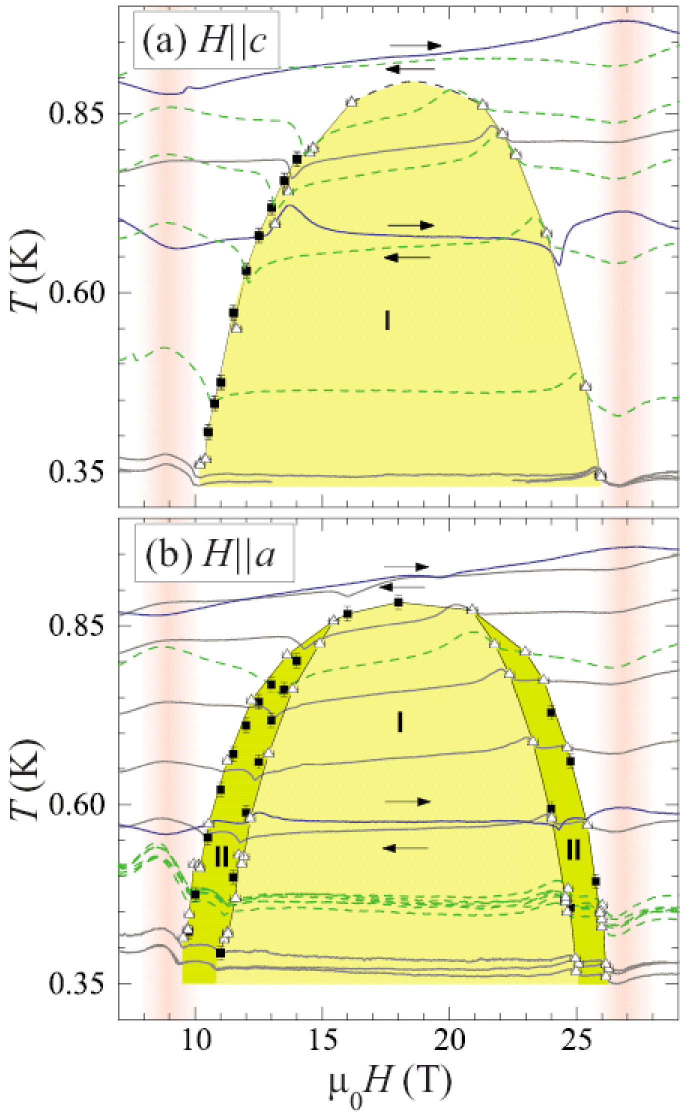
AF dimer exchange J_0 = 1.50~1.65 meV

M. B. Stone *et al.*, PRB **77**, 134406 (2008); E. C. Samulon *et al.*, PRL **103**, 047202 (2009)



Energy spectrum consists of excited triplet and quintuplet states above the singlet ground state.

Phase diagram of $Ba_3Mn_2O_8$

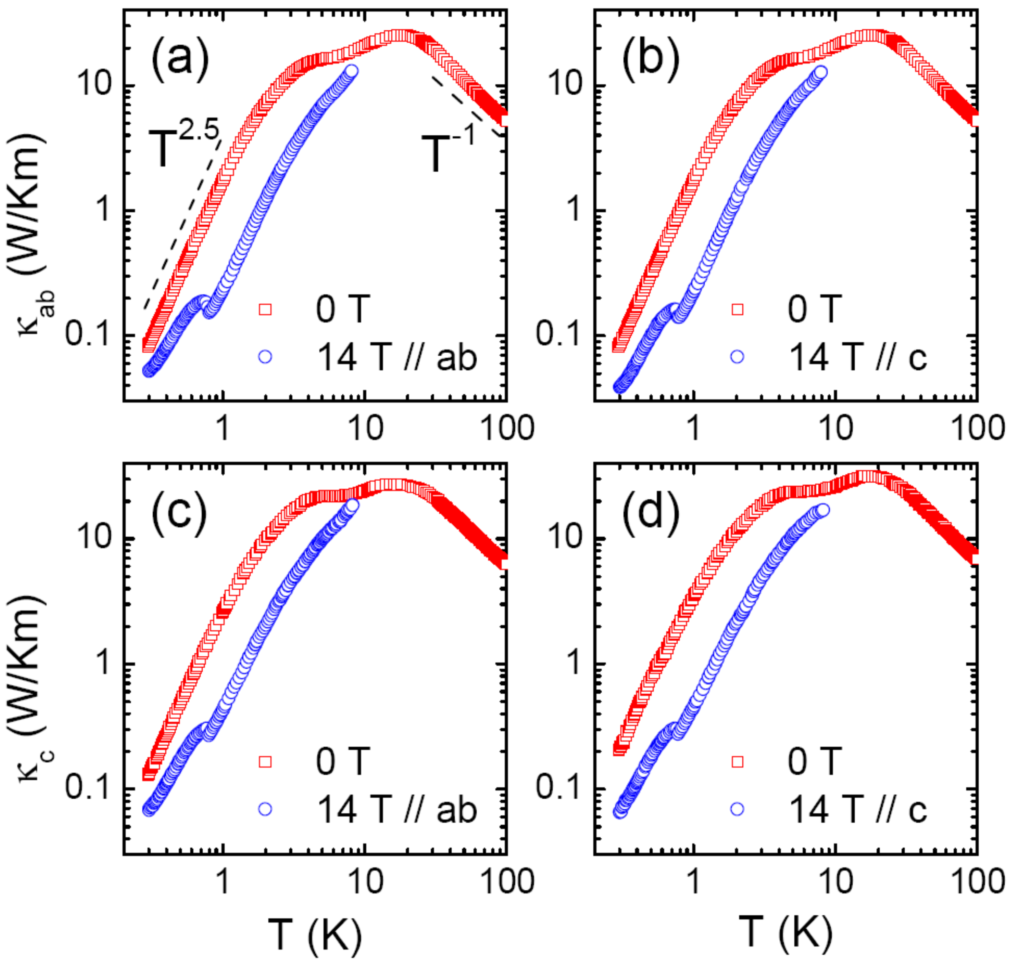


Phase I consists of a spiral structure which is stabilized by the weak interlayer coupling.

Phase II appears to be a fully modulated structure with no moment along the y direction, stabilized by a combination of the single-ion anisotropy and the interlayer coupling.

E. C. Samulon *et al.*, PRL **103**, 047202 (2009); PRB **77**, 214441 (2008)

Heat transport of $Ba_3Mn_2O_8$ single crystals



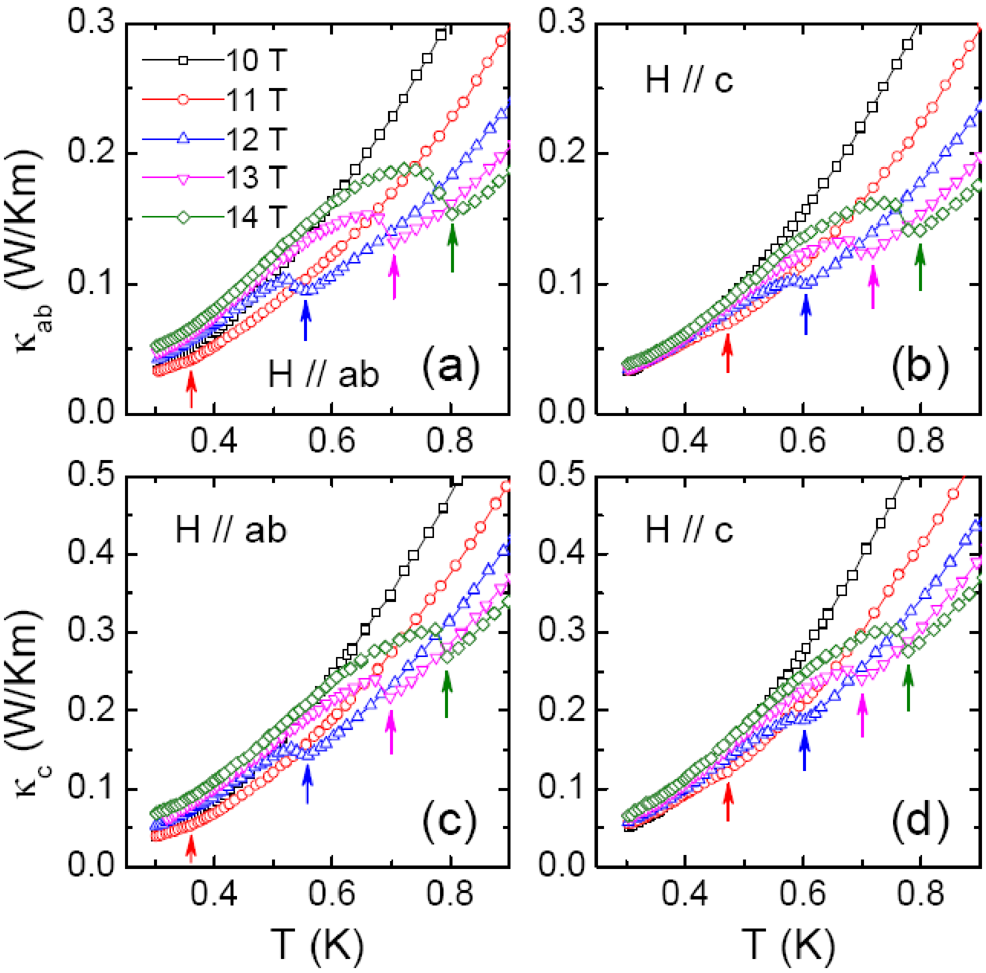
Pure phonon transport at zero field.

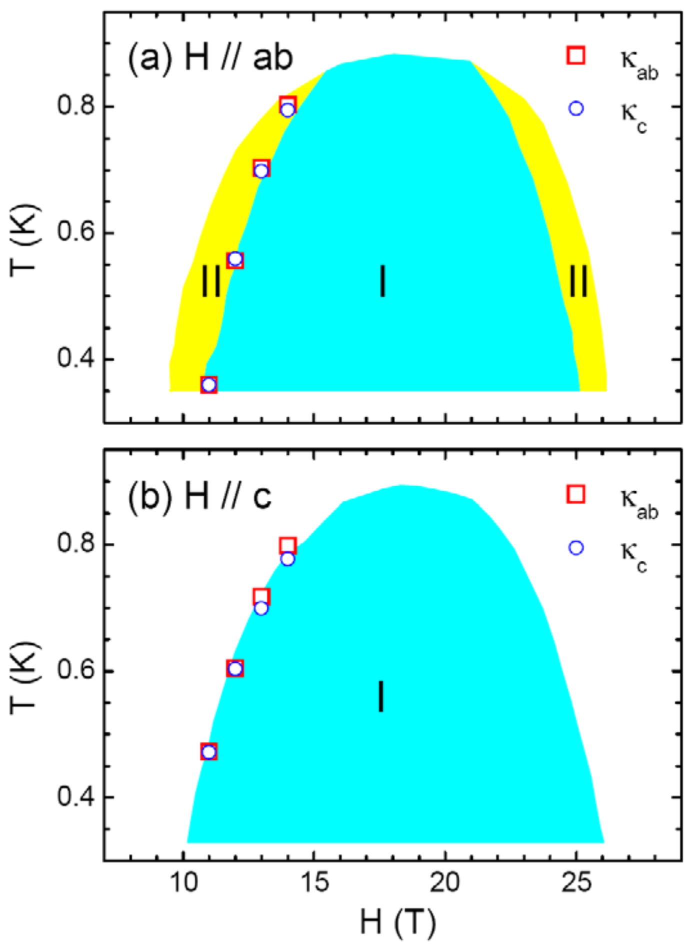
A shoulderlike feature at ~ 6 K: resonant scattering by magnetic excitations.

Low-*T* thermal conductivity is strongly suppressed in high magnetic field: phonon scattering by magnetic excitations.

W. P. Ke *et al.*, PRB **84**, 094440 (2011)

Enhancement of κ below LRO transition

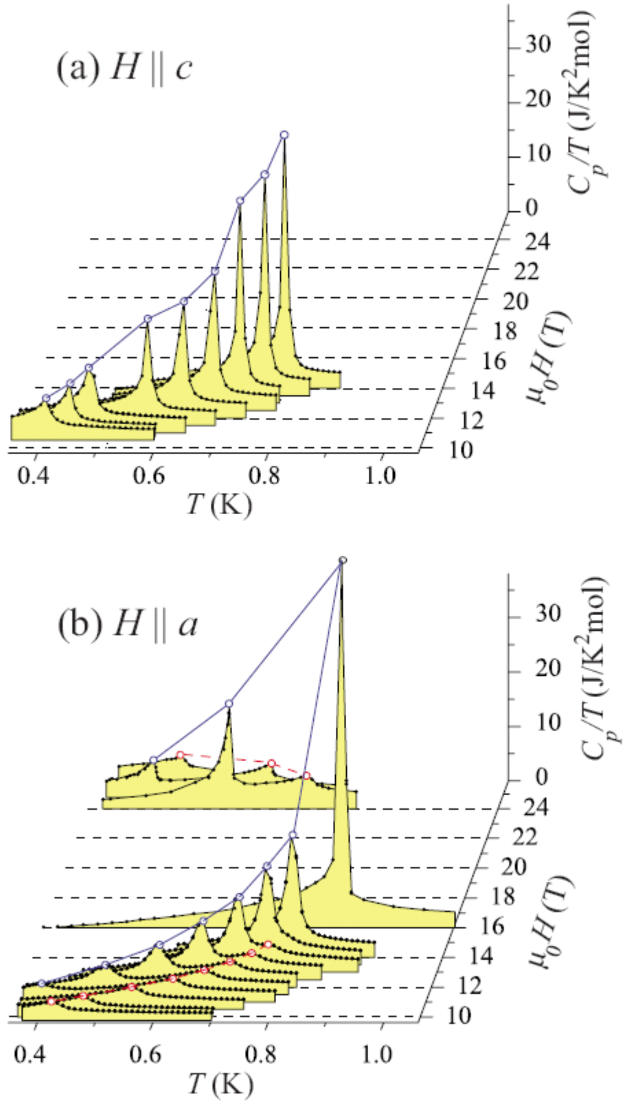


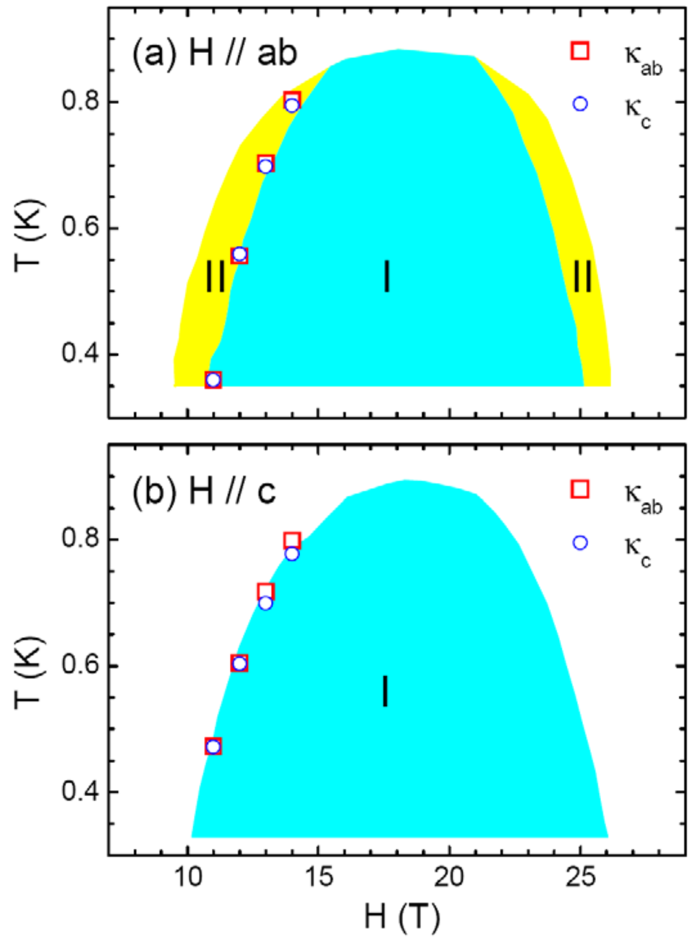


Rather weak enhancement at T_c; κ anomalies appear at the phase boundary of phase I only.

W. P. Ke *et al.*, PRB **84**, 094440 (2011)

Enhancement of κ below LRO transition

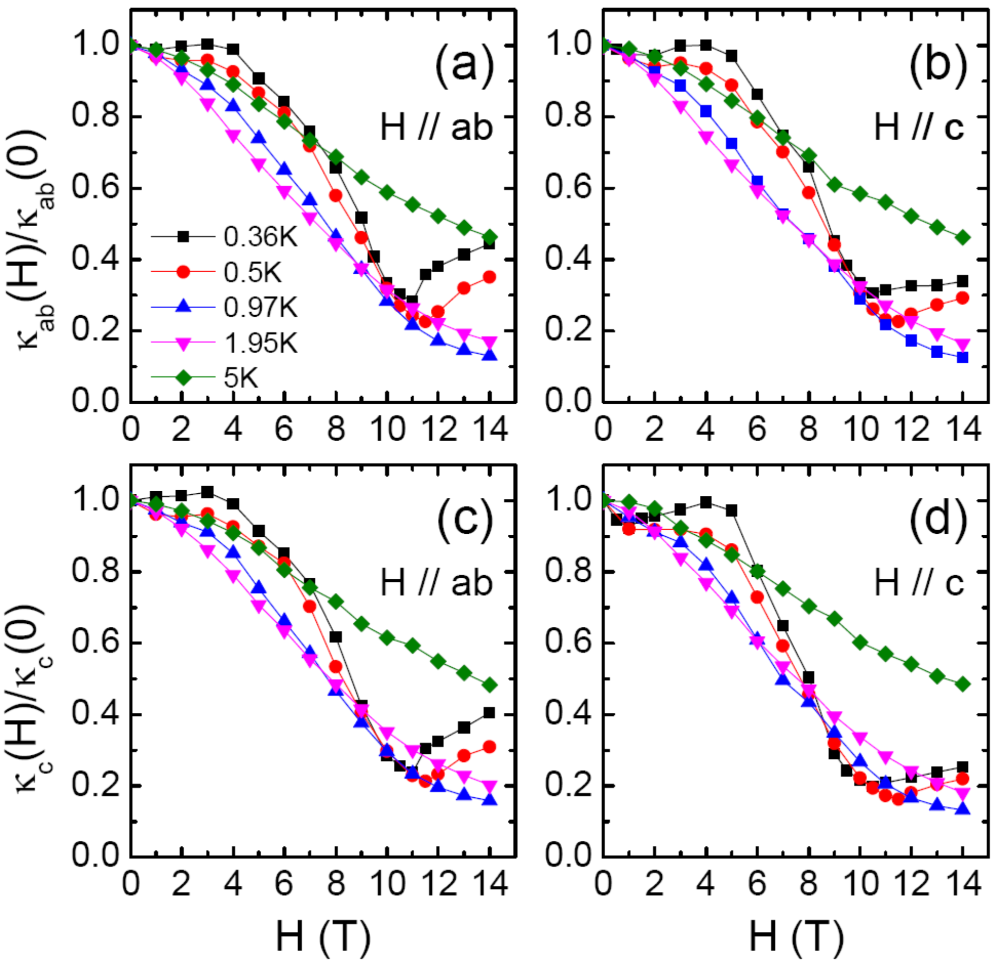




Rather weak enhancement at T_c; κ anomalies appear at the phase boundary of phase I only.

E. C. Samulon *et al.*, PRB **77**, 214441 (2008)

Strong phonon scattering by magnetic excitations



The main role of magnons is strongly scattering phonons.

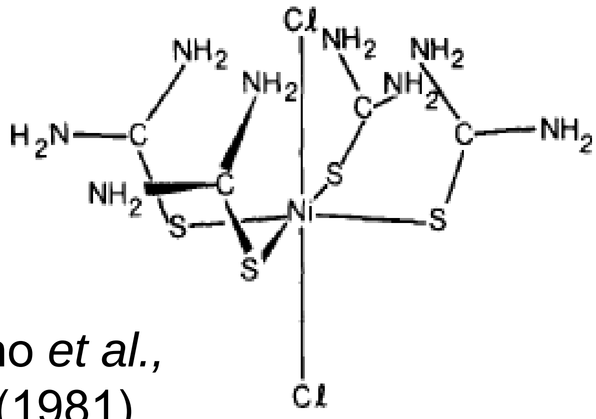
The thermal conductivity in magnon BEC state seems to be smaller than the zero-field phonon conductivity.

The contribution of magnons to heat transport in BEC state cannot be large.

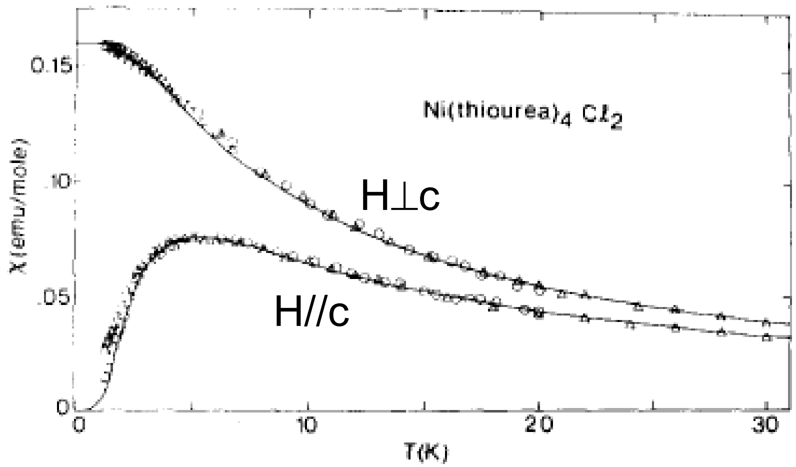
W. P. Ke *et al.*, PRB **84**, 094440 (2011)

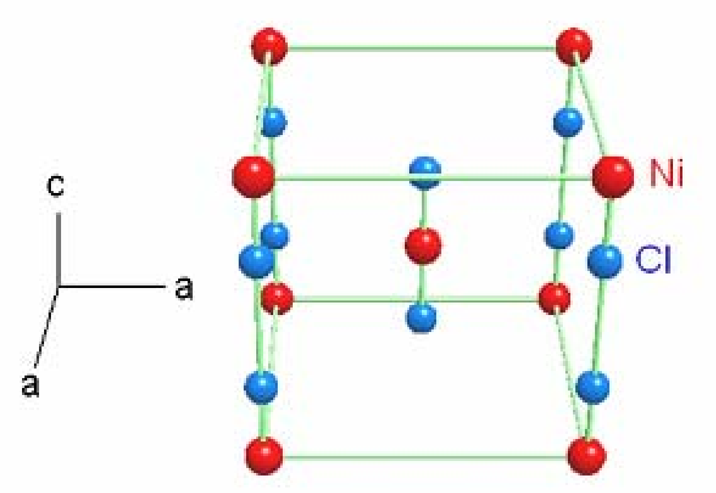
Example II: $NiCl_2$-$4SC(NH_2)_2$ (DTN)

Organic



A. Paduan-Filho *et al.*,
JCP **74**, 4103 (1981)





1D spin chain along the *c* axis

space group: *I*4
tetragonal
$a = b = 9.558$Å, $c = 8.981$Å

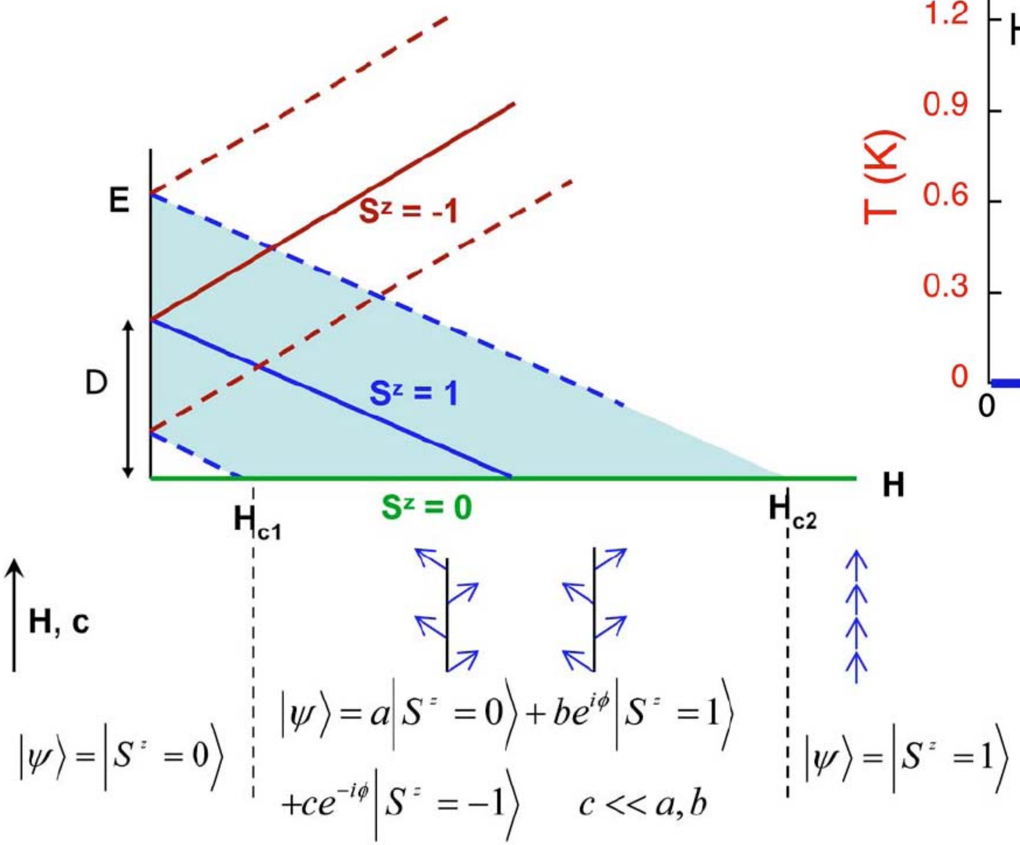
$J_{chain} = 2.2$ K, $J_{plane} = 0.17$ K
$D = 8.9$ K

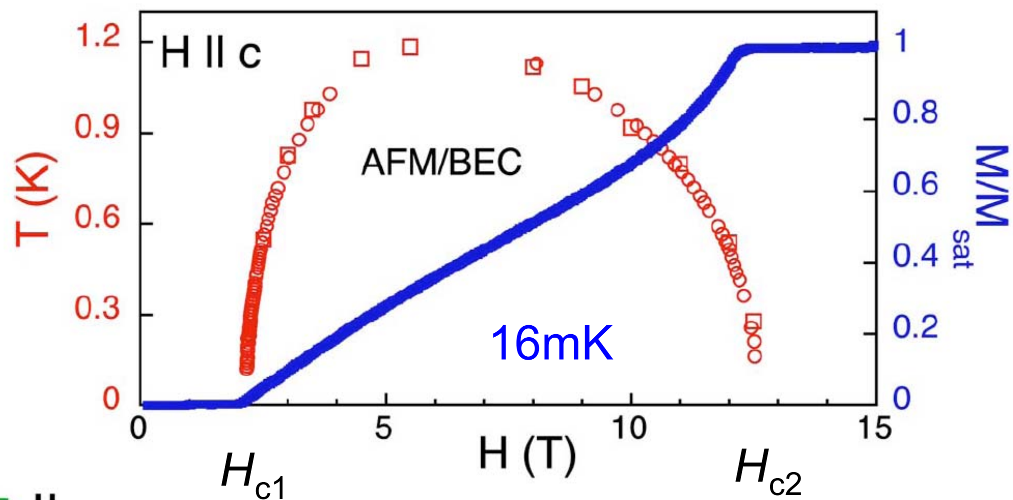
Spin spectrum: anisotropy gap

V. S. Zapf *et al.*, JAP **101**, 09E106 (2007)

Phase diagram

- Field-induced magnetic ordering



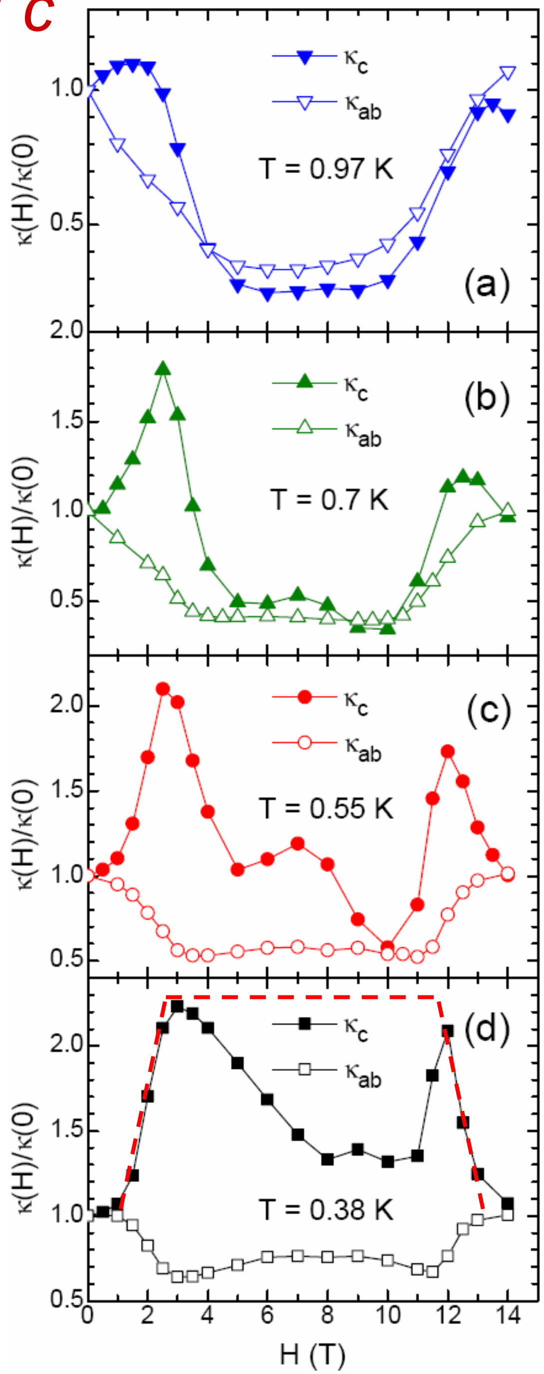


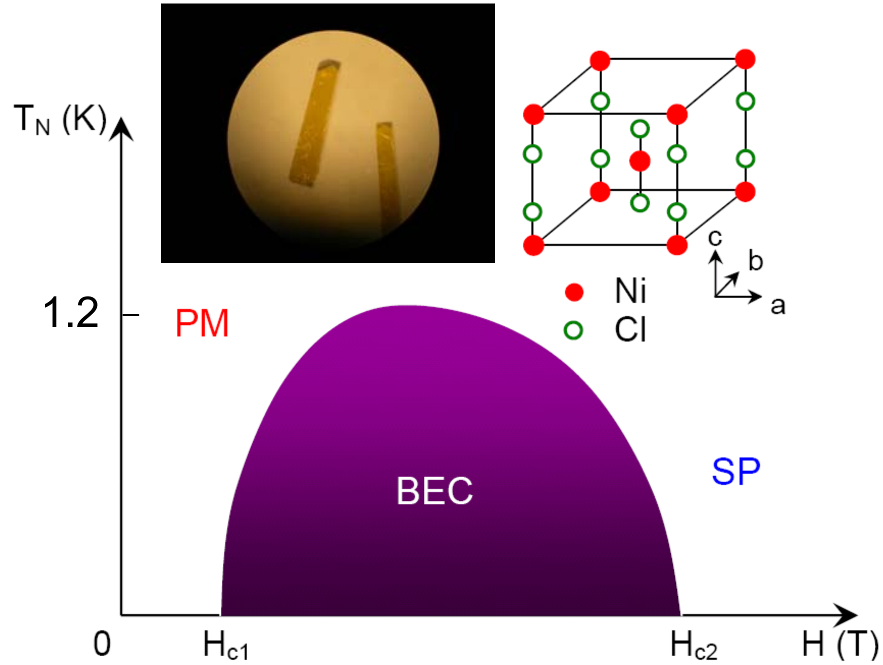
The Ni *S*=1 spin triplet is split by single-ion anisotropy into a $S_z = 0$ ground state and $S_z = \pm 1$ excited states with an energy gap of *D.*

V. S. Zapf *et al.*, JAP **101**, 09E106 (2007)

H // c

Possible large thermal conductivity in magnon BEC state



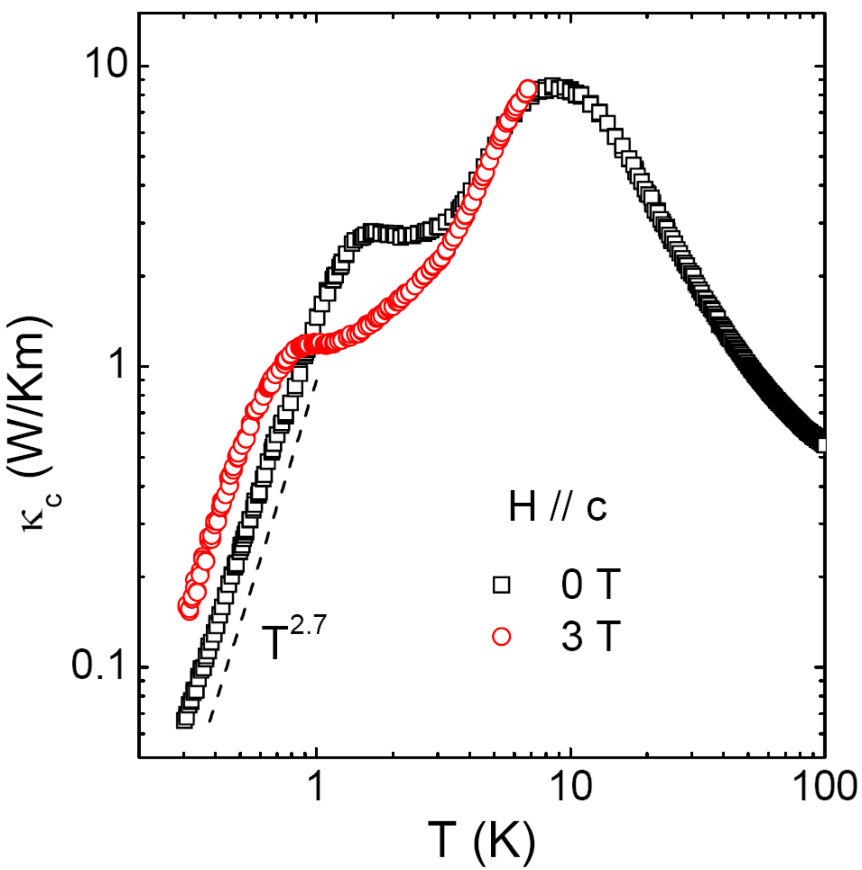


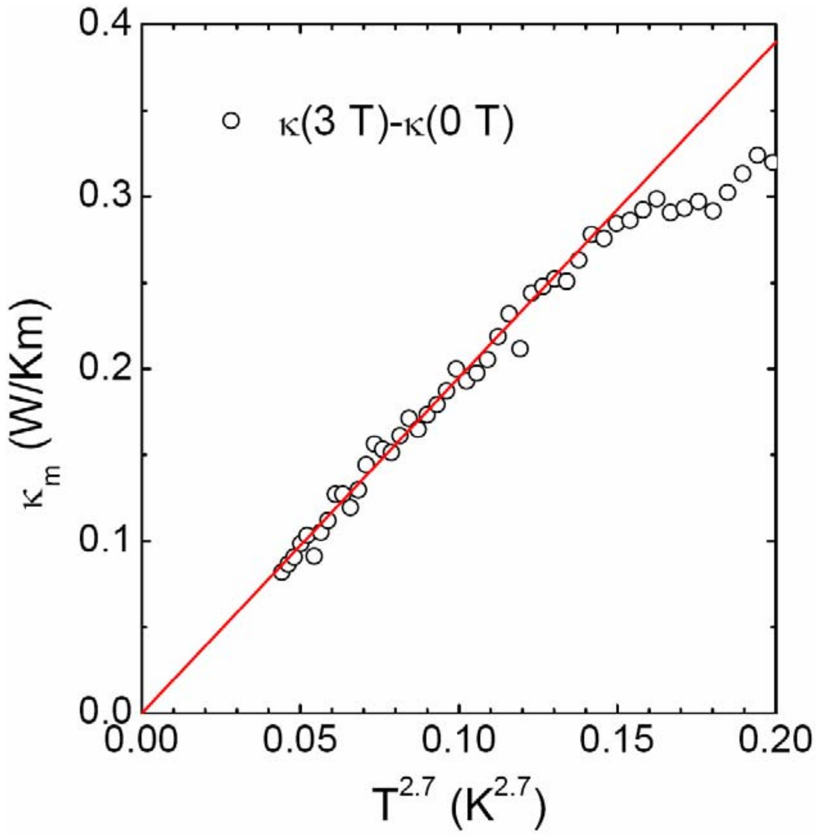
In the *ab* direction, magnons only scatter phonons.

Along the *c* axis, magnons act mainly as phonon scatterers at relatively high temperatures, but change their role to heat carriers upon $T \to 0$.

X. F. Sun *et al.*, PRL **102**, 167202 (2009)
See also: Y. Kohama *et al.*, PRL **106**, 037203 (2011)

Magnon heat transport at the BEC transition





In zero field, the boundary scattering limit is approached at lowest temperature.

At very low temperatures, field-induced increase of κ follows a $T^{2.7}$ dependence.

Magnons are acting as heat carriers in the BEC state.

X. F. Sun *et al.*, unpublished

Summary (II)

- The heat transport behaviors are sensitive to the field-induced QPTs of the spin-gapped materials.
- The field-induced LRO (BEC) state does not necessarily have sizeable magnetic heat conductivity.
- The magnetic excitations can act as either heat carriers or phonon scatterers.
- DTN is a promising material that can show the T^3 ballistic transport of magnons. However, the magnon transport is evidenced only at the critical field.