Driven complex matter

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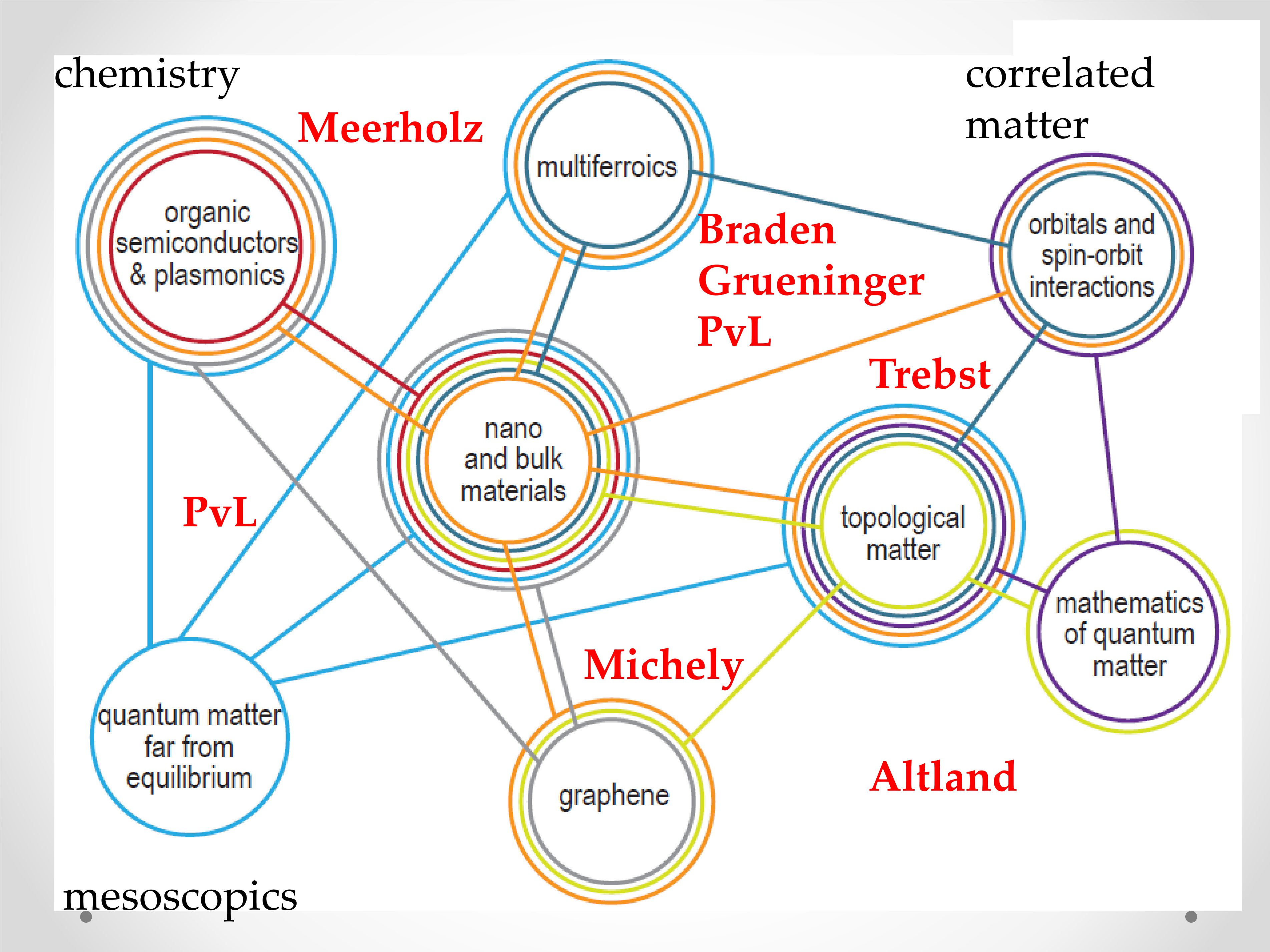
UoC: Excellence University

The University of Cologne's **institutional strategy "Meeting the Challenge of Change and Complexity"** aims at strengthening and further developing the university's research profile, at establishing funding programmes for cutting edge research and at integrating new career-promoting structures and support measures. The strategy also includes the further development of regional and international research networks and exchange programs, the promotion of Gender Equality and a set of measures to promote research-based teaching.

QM2 Quantum matter and materials

"Quantum Matter and Materials" (QM2) is a fascinating field of research driven both by the intellectual challenge and the promise for applications. Within QM2, researchers of mathematics, experimental and theoretical physics, anorganic and physical chemistry and crystallography collaborate to unravel the properties of quantum matter. The mathematical structures underlying topological matter, the prospect to functionalize "Dirac matter" like graphene, new states of matter arising from spin-orbit interactions, quantum matter far from thermal equilibrium, the use of nanostructured materials for wide ranges of applications, and the development of organic electronics, are some of the research topics important for QM2.

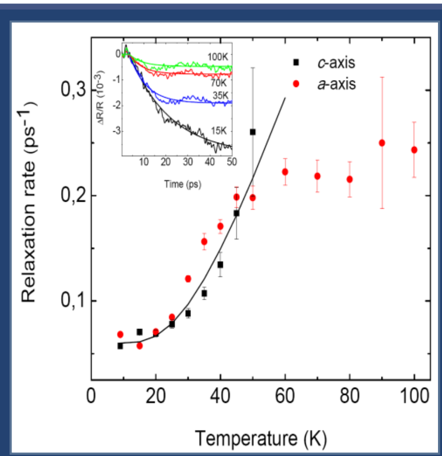


chemistry
Meerholz
multiferroics
correlated
matter
organic
semiconductors
& plasmonics
Braden
Grueninger
PvL
orbitals and
spin-orbit
interactions
Trebst
nano
and bulk
materials
PvL
topological
matter
mathematics
of quantum
matter
Michely
quantum matter
far from
equilibrium
graphene
Altland
mesoscopics

Dynamical properties of matter

Mapping interactions & processes on the time domain

- Electron-electron
- Electron-phonon
- Electron-magnon
- Phonon-magnon
- Energy, charge, and spin transfer
- Spin, phonon, orbital coherence
- Pair breaking
- Charge separation
- …



Magnon assisted hopping in $TbMnO_3$

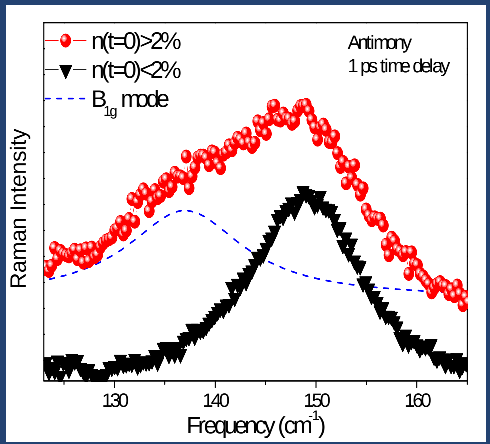
→ *Charge – Magnon interaction*
→ *k=π Electromagnon energy*

I.P. Handayani et al., JPCM 2013

Dynamical properties of matter

Controlling states of matter

- Novel states of matter
- Ultrafast melting of lattice, spin, charge ordering
- Switching magnetism
- Switching ferroelectricity
- Stripes & induced superconductivity
- Metastable states
- …



Induced phase transition in A3 elemental metals

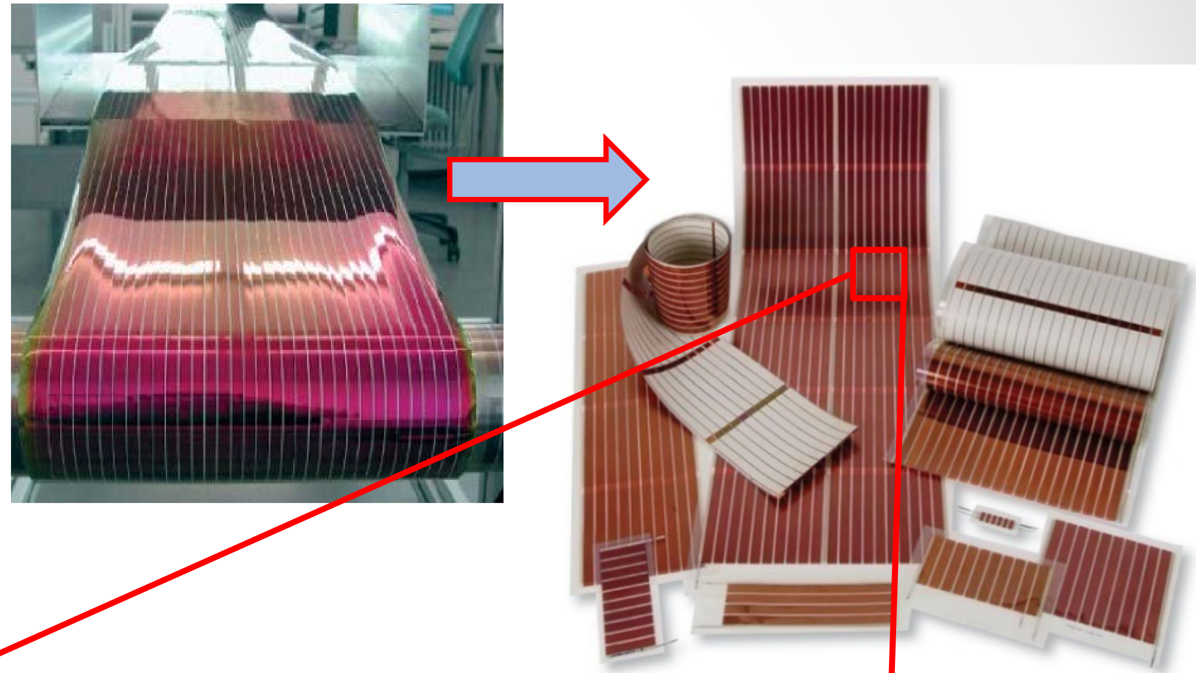
→ *Non-thermal state*
→ *Destabilization Jones-Peierls state*
→ *New Jones-Peierls distortion*

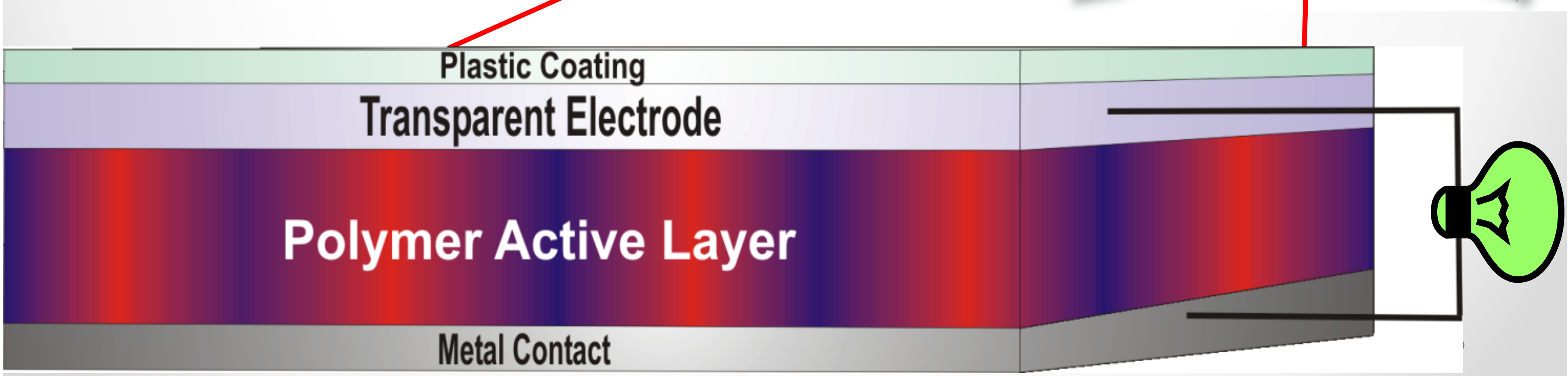
D. Fausti et al., PRB-RC 2009

Charge separation in OPV

Advantages:

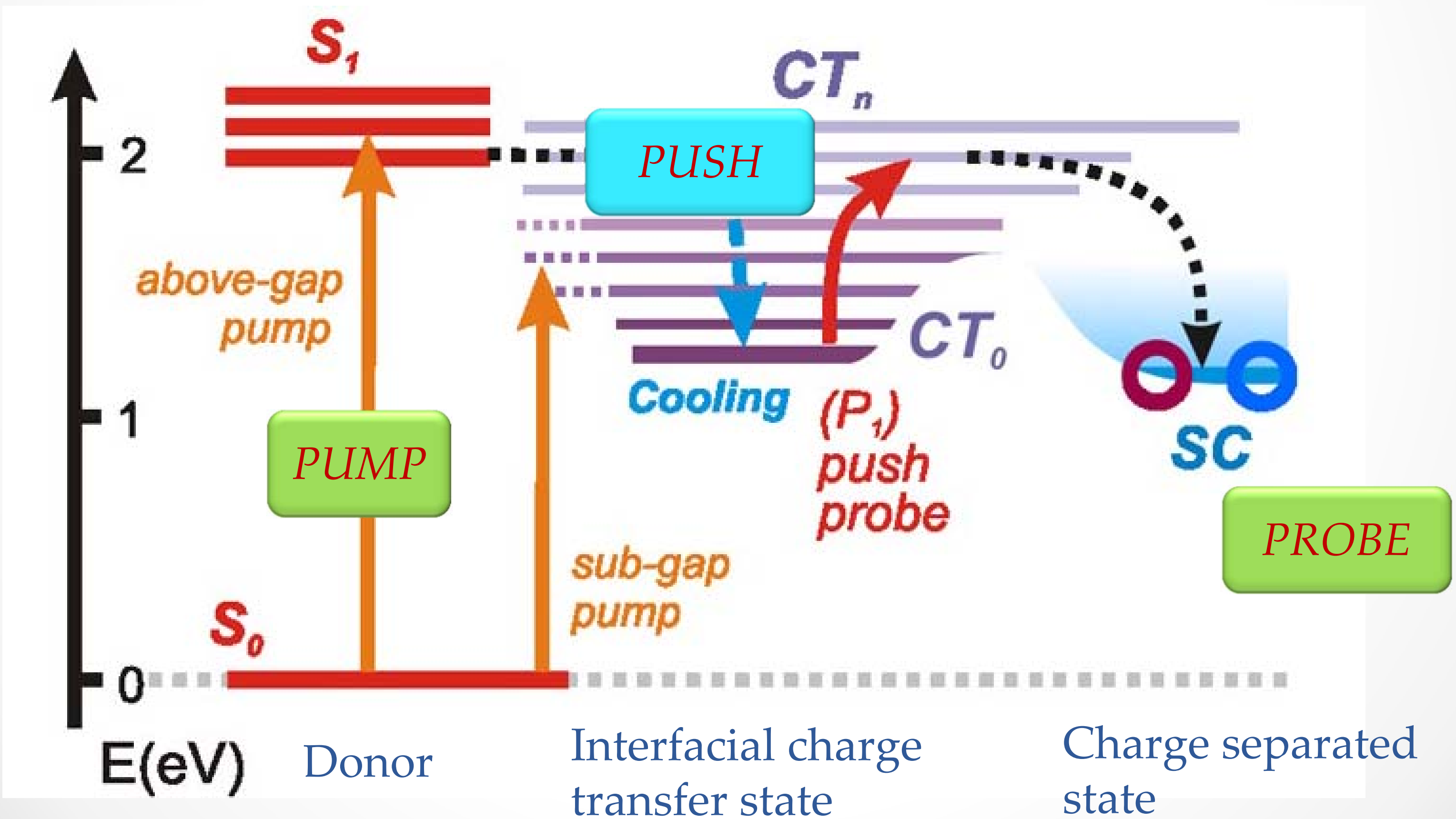
- ✓ Inexpensive
- ✓ Lightweight
- ✓ Flexible
- ✓ Tunable properties
- ✓ Printing-processable





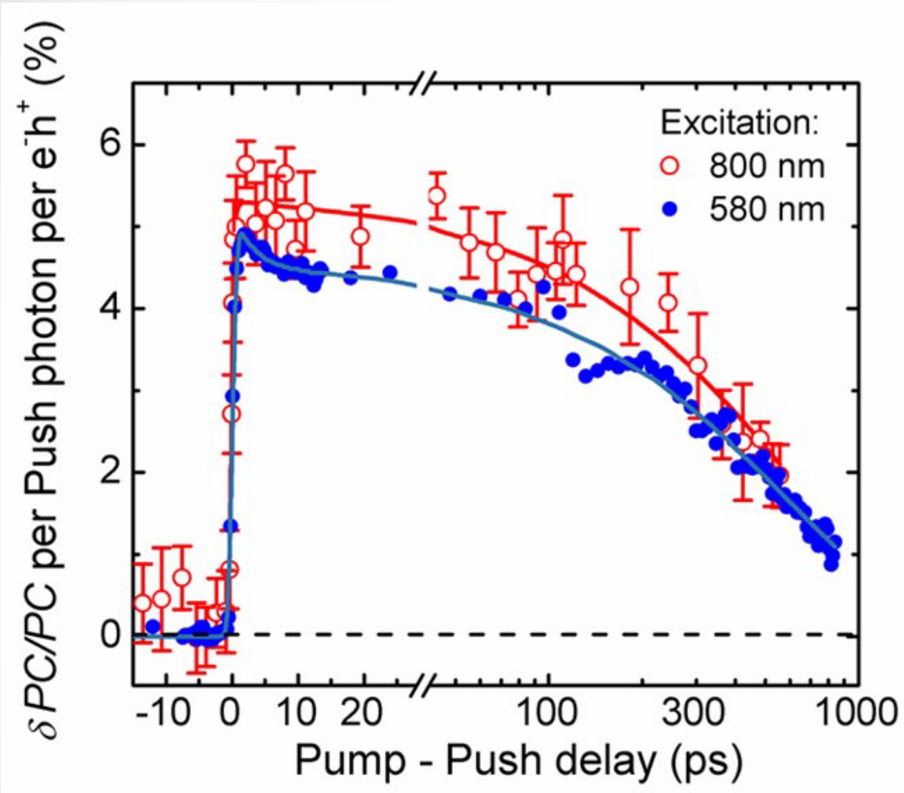
Charge separation in OPV

Action occurs at the donor-acceptor interface.
What determines the efficiency ?

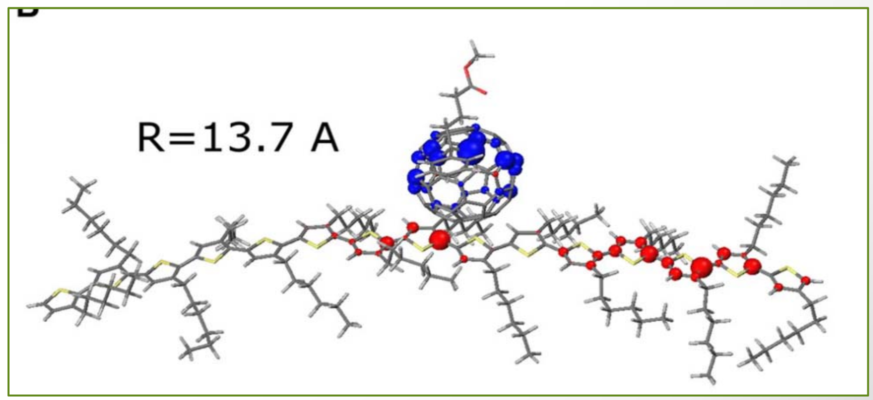


Similar for acceptor: Bakulin et al., Funct. Adv. Mat. 2010

Charge separation in OPV



Delocalization drives charge generation efficiency



Bakulin et al. Science **335,** 1340 (2012).

Ultrafast magnetism in EuO

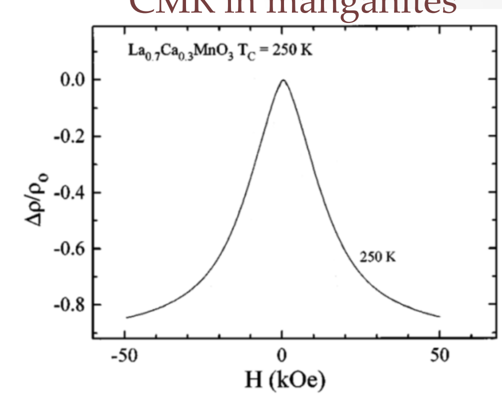
Interplay between magnetism and conductivity

RKKY interaction
- → GMR read heads
- → Kondo physics

Double exchange
- → CMR in manganites
- → Heavy fermion systems

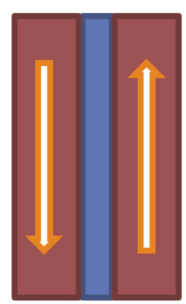
Spintronics
- → Doped semiconductors
- → EuO

CMR in manganites



M.F. Hundley *et al.* APL **67**, 860 (1995)

spin valve

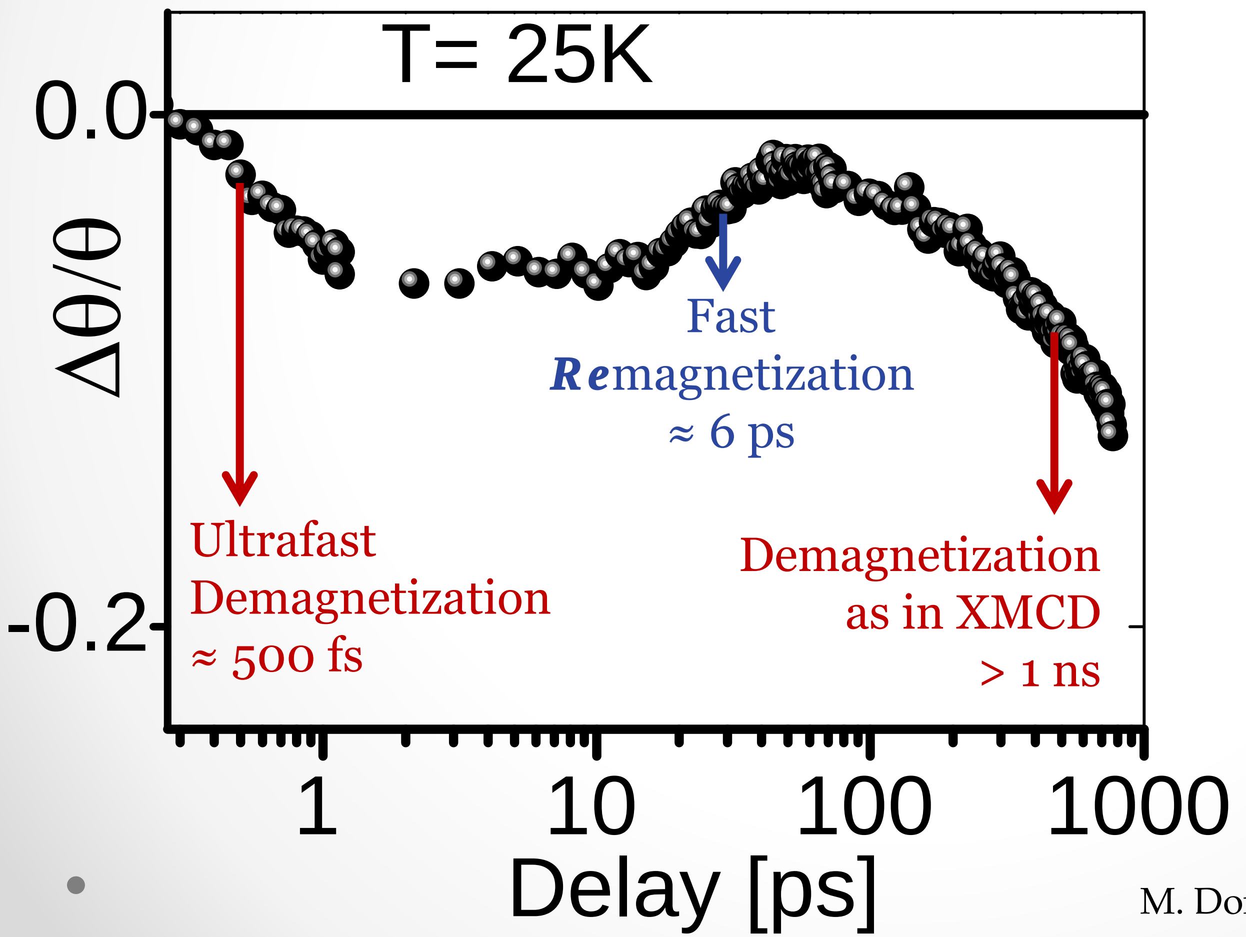


GMR

hard drive

Ultrafast magnetism in EuO

Ultrafast switching of magnetism in a magnetic semiconductor:
Interplay between $5d^1$ electrons, $4f^6$ holes and $4f^7$ moments



First picosecond
- hot $5d^1$ electrons
- Elliot-Yafet scattering

10 ps timescale
- $4f^6$-$5d^1$
- magnetic polaron formation

Nanosecond timescale
- $4f^6$,$4f^7$
- spin-lattice relaxation
- anisotropic CF fluctuations

M. Donker et al., unpubl. 2013

Dynamic heat transport in low dimensional quantum magnets

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QM²

Universität zu Köln

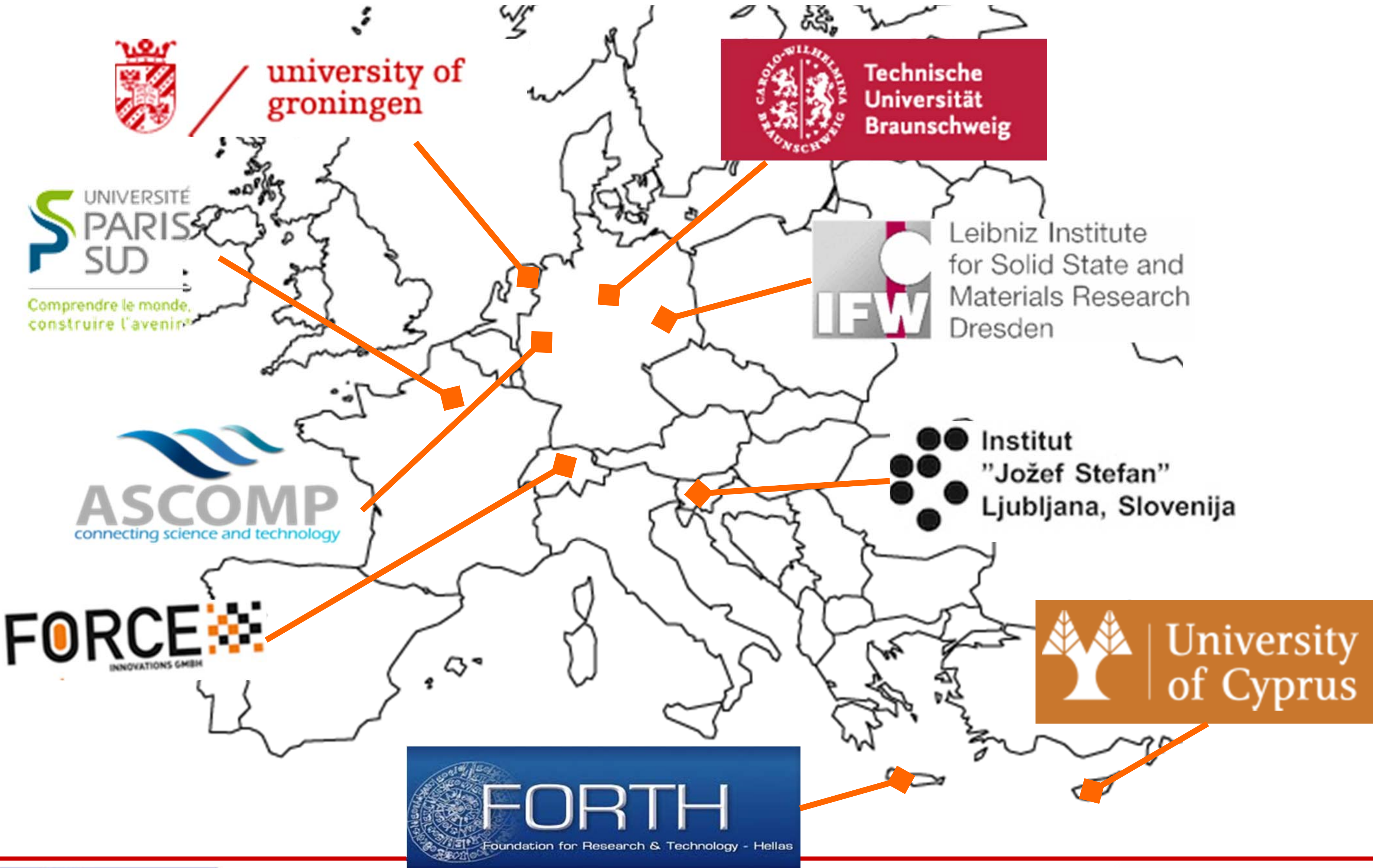
Dynamic heat transport in low dimensional quantum magnets.

Paul H.M. van Loosdrecht

Optical Condensed Matter Physics

Zernike Institute for Advanced Materials

The LOTHERM collaboration



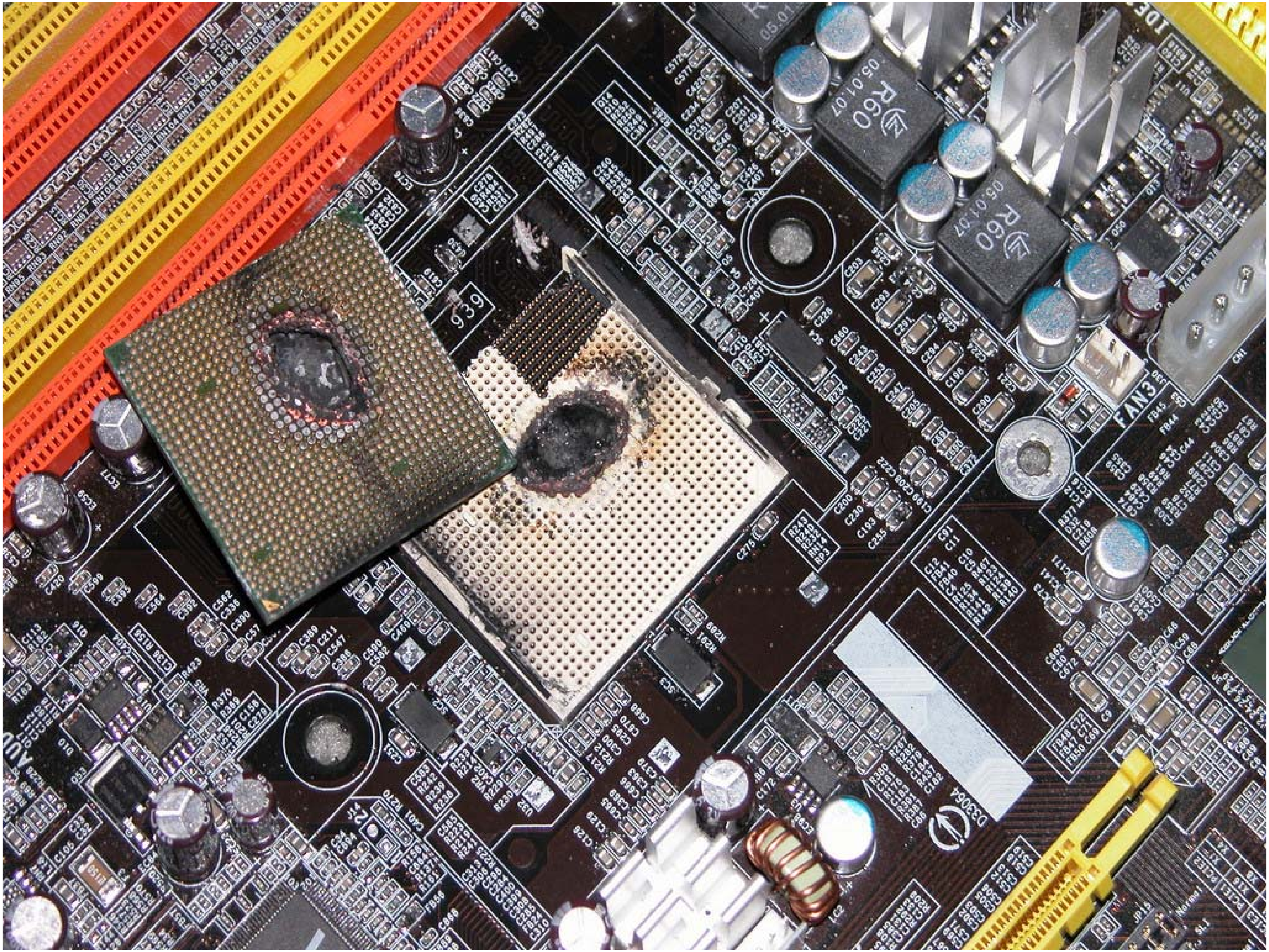
LOTHERM
Magnetic-mode heat transport

university of groningen / zernike institute for advanced materials

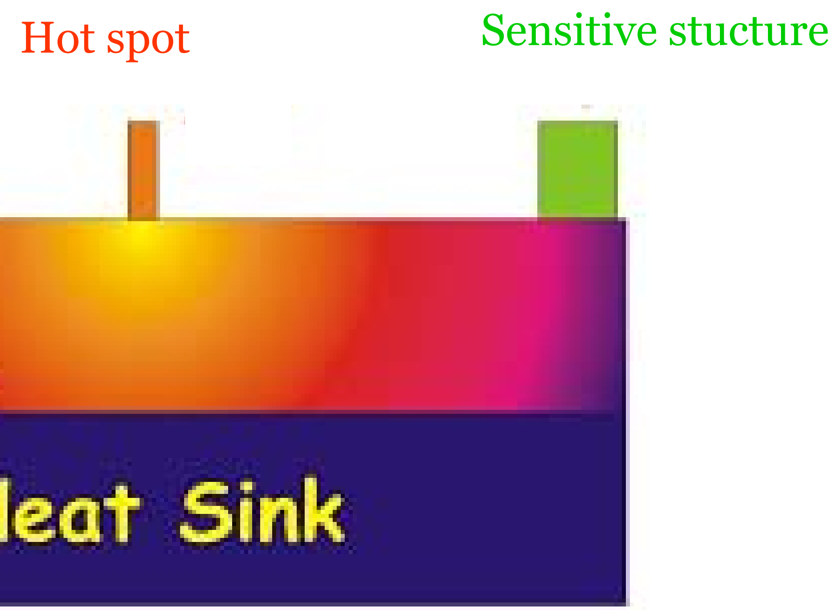
Heat management

"Managing heat generation within IC's will be a crucial issue in developing the next generation of electronics."

International Technology Roadmap for Semiconductors, ITRS-2009

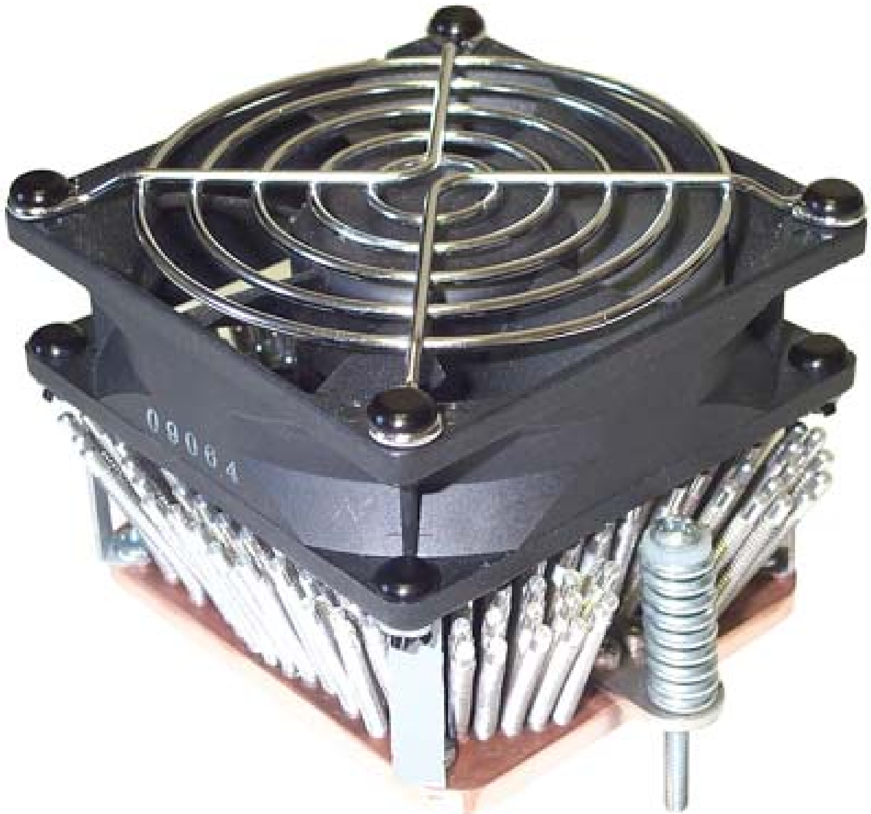

R60
05.01.07
R60
05.01.07
939
FAN3

Heat management



Solutions ?

Buy a big fan

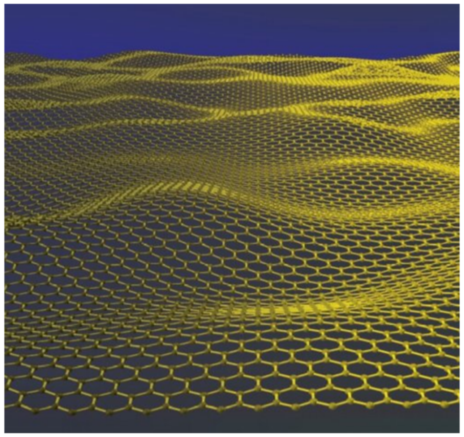


Use a champion heat conductor

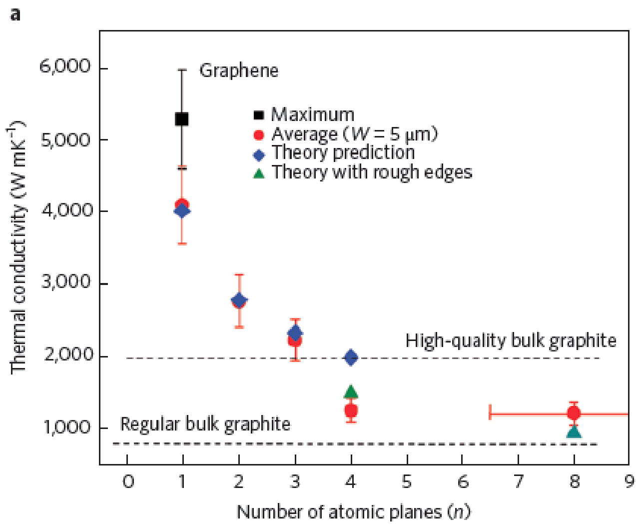
Diamond
2000 W/mK

Graphene 5000 W/mK

Hope



Even more Hope

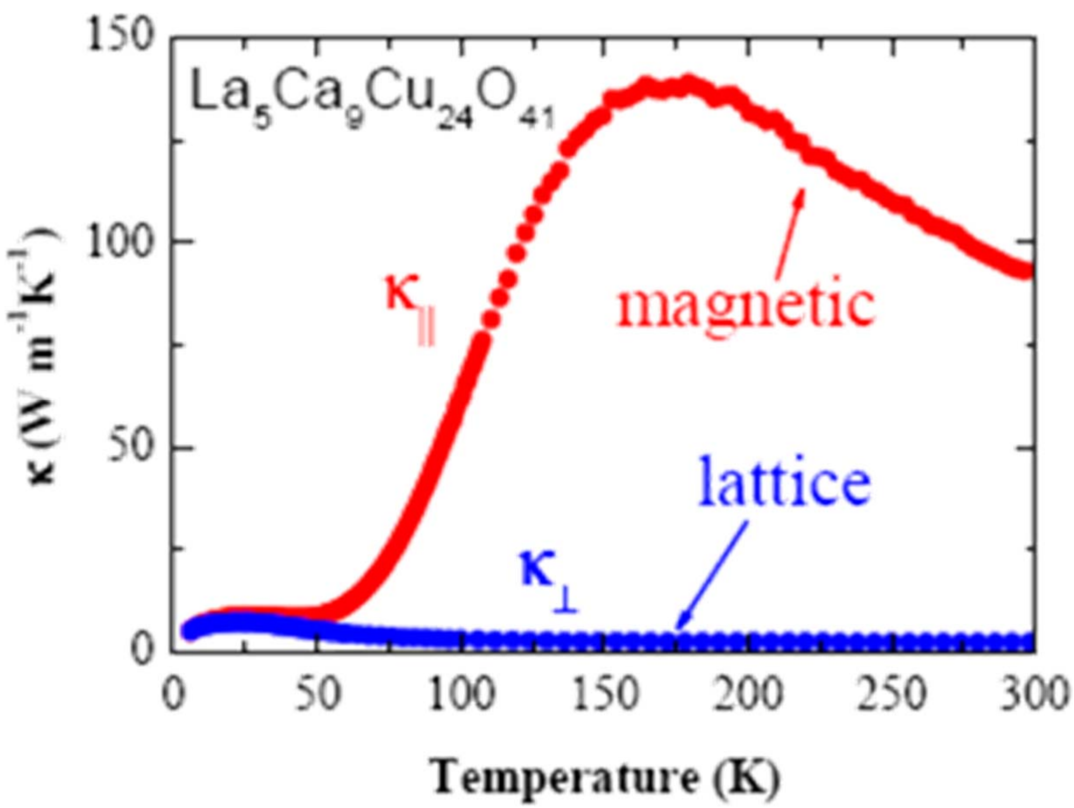


Balandin, Nature Materials 2011

Expensive

Also good charge conductor

Quantum magnets ?



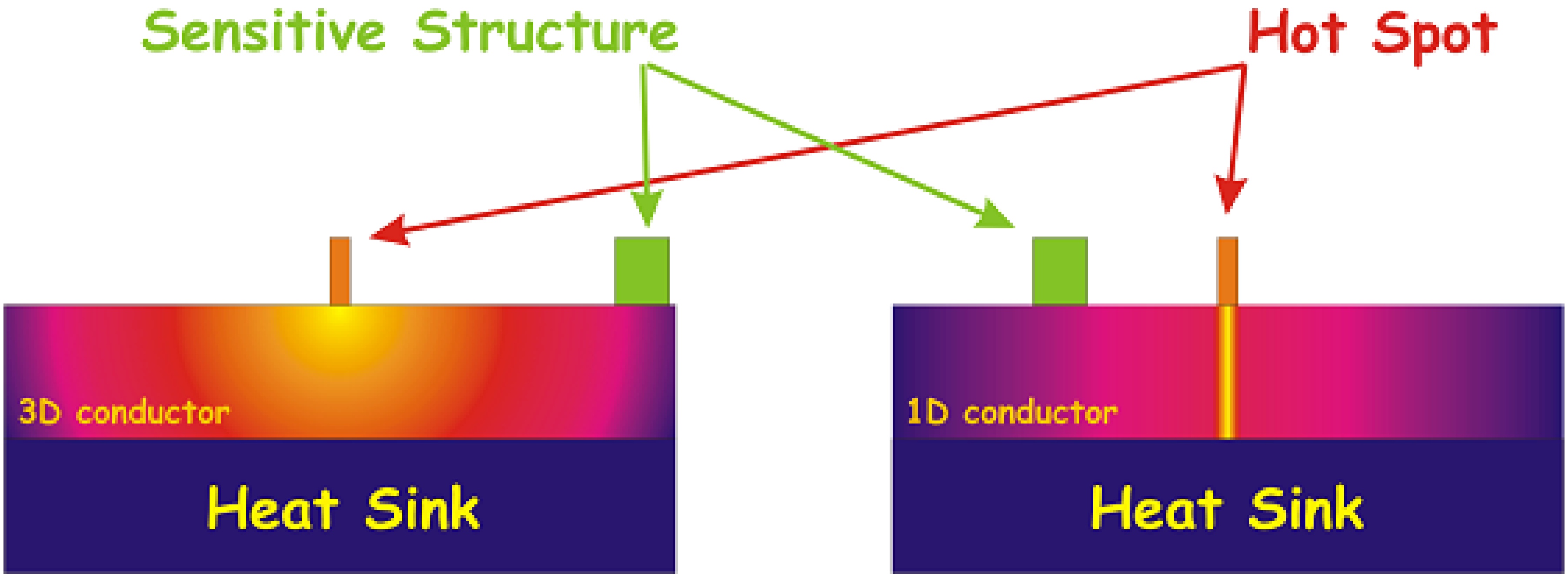
LCCO:

- *140 W/mK*
- *Good charge insulator*
- *1 dimensional*

Steel	50 W/mK
Copper	350 W/mK
Silicon	100 W/mK
SiO_2	1 W/mK
SiC	400 W/mK
Diamond	1000 W/mK

C. Hess et al., Phys. Rev. **B 64**, 184305 (2001).

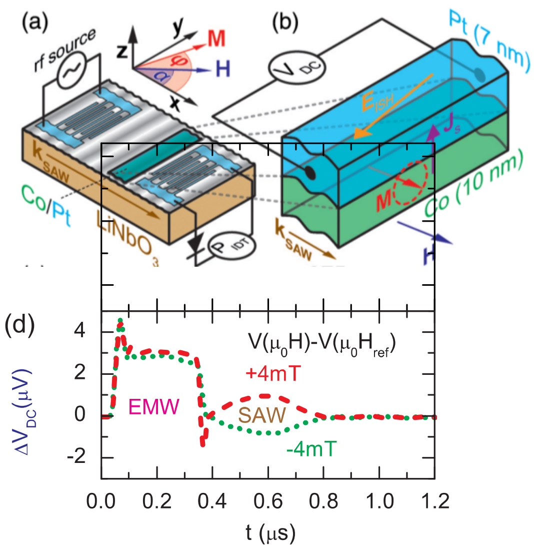
Better cooling → Higher densities



Low dimensional quantum magnets

- Good heat conductivity (> 100 $Wm^{-1}K^{-1}$)
- Unidirectional
- Electrically insulating

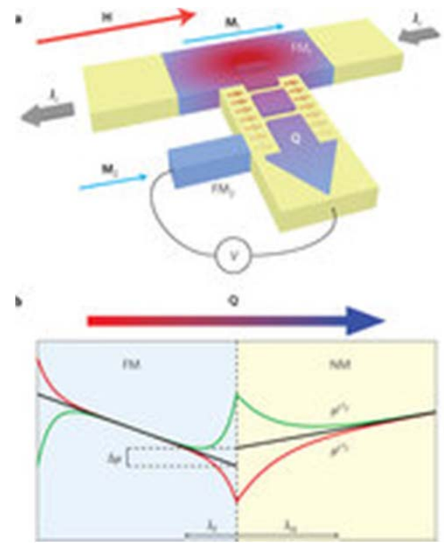
Spin pumping with coherent elastic waves



Weiler, PRL 2012

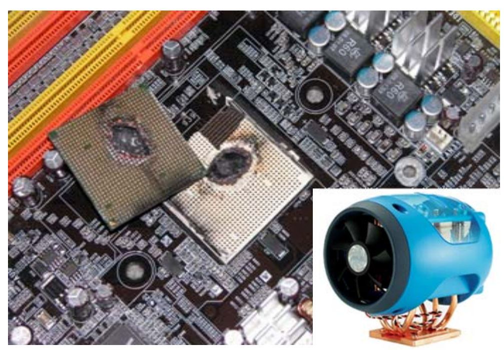
PHONON-MAGNON COUPLING

Spin Seebeck Effect



Bauer, Nat. Mat. 2012

Heat management

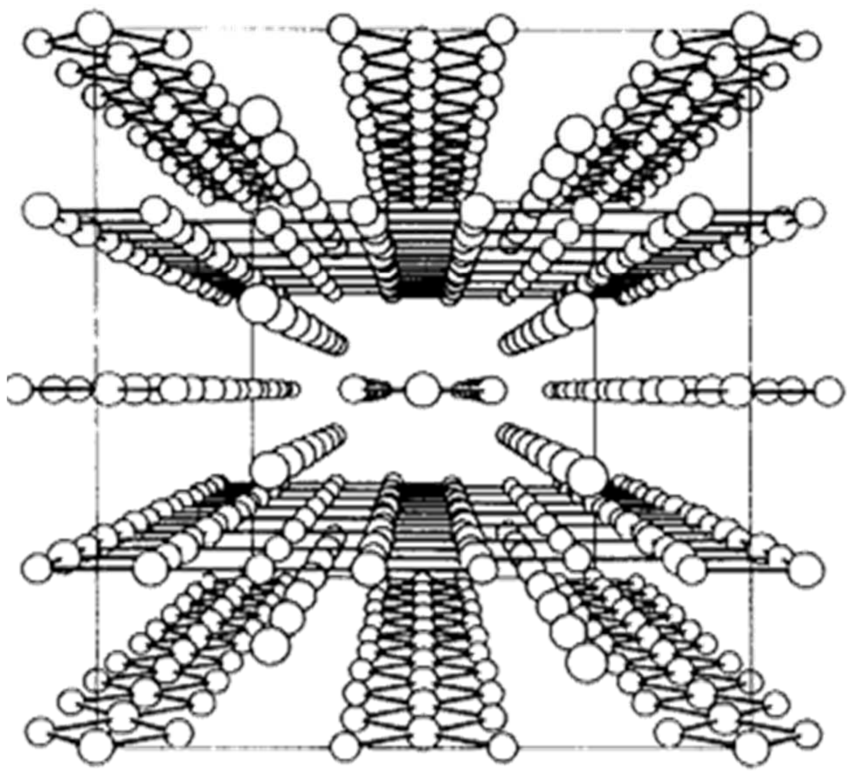


$(Sr,La,Ca)_{14}Cu_{24}O_{41}$

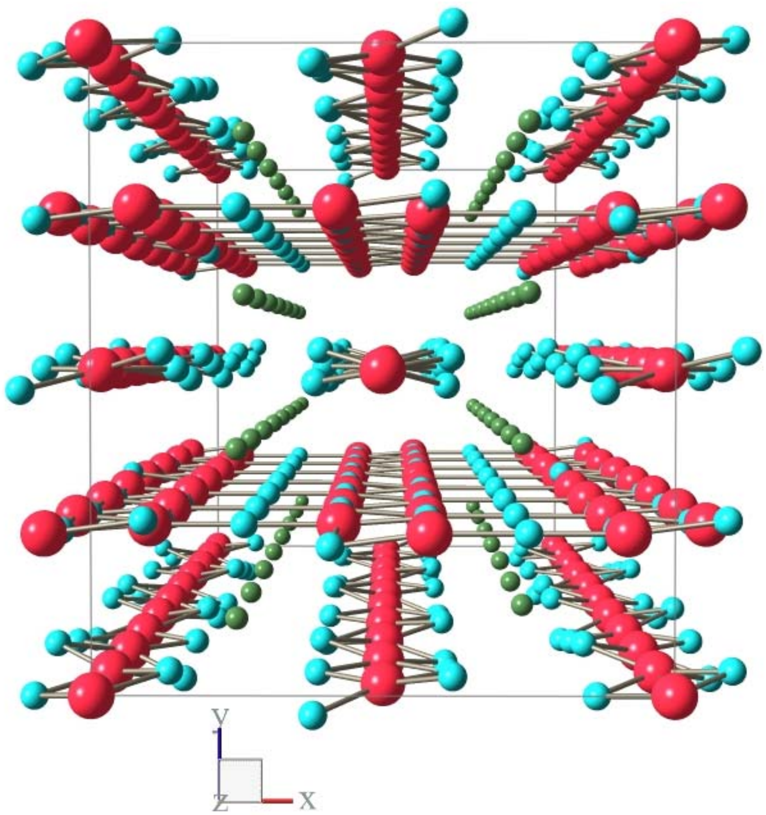
Mat. Res. Bull., Vol. 23, pp. 1355-1365, 1988. Printed in the USA.
0025-5408/88 $3.00 + .00

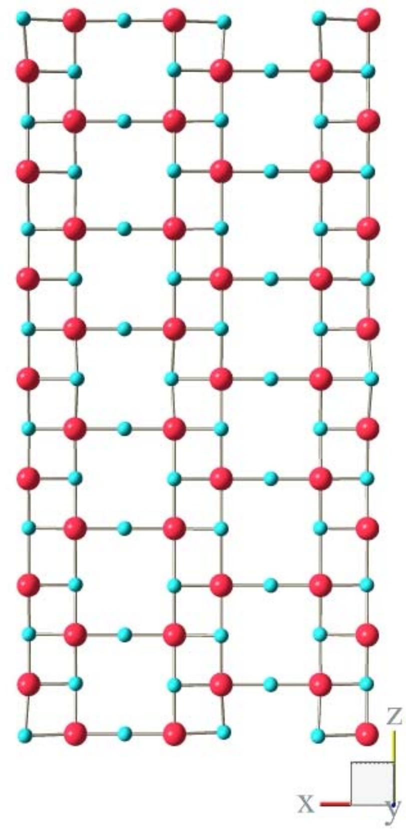
THE INCOMMENSURATE STRUCTURE OF $(Sr_{14-x}Ca_x)Cu_{24}O_{41}$ $(0 < x \sim 8)$
A SUPERCONDUCTOR BYPRODUCT

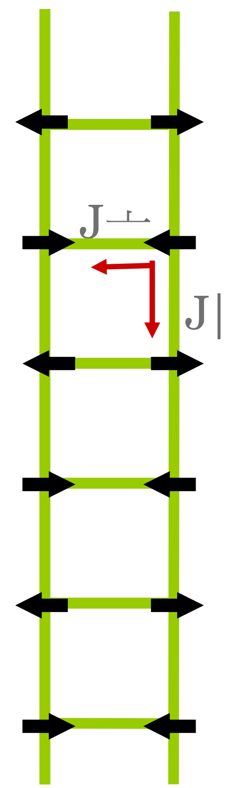
E. M. McCarron, III*, M. A. Subramanian, J. C. Calabrese
and R. L. Harlow*



$(Sr,La,Ca)_{14}Cu_{24}O_{41}$

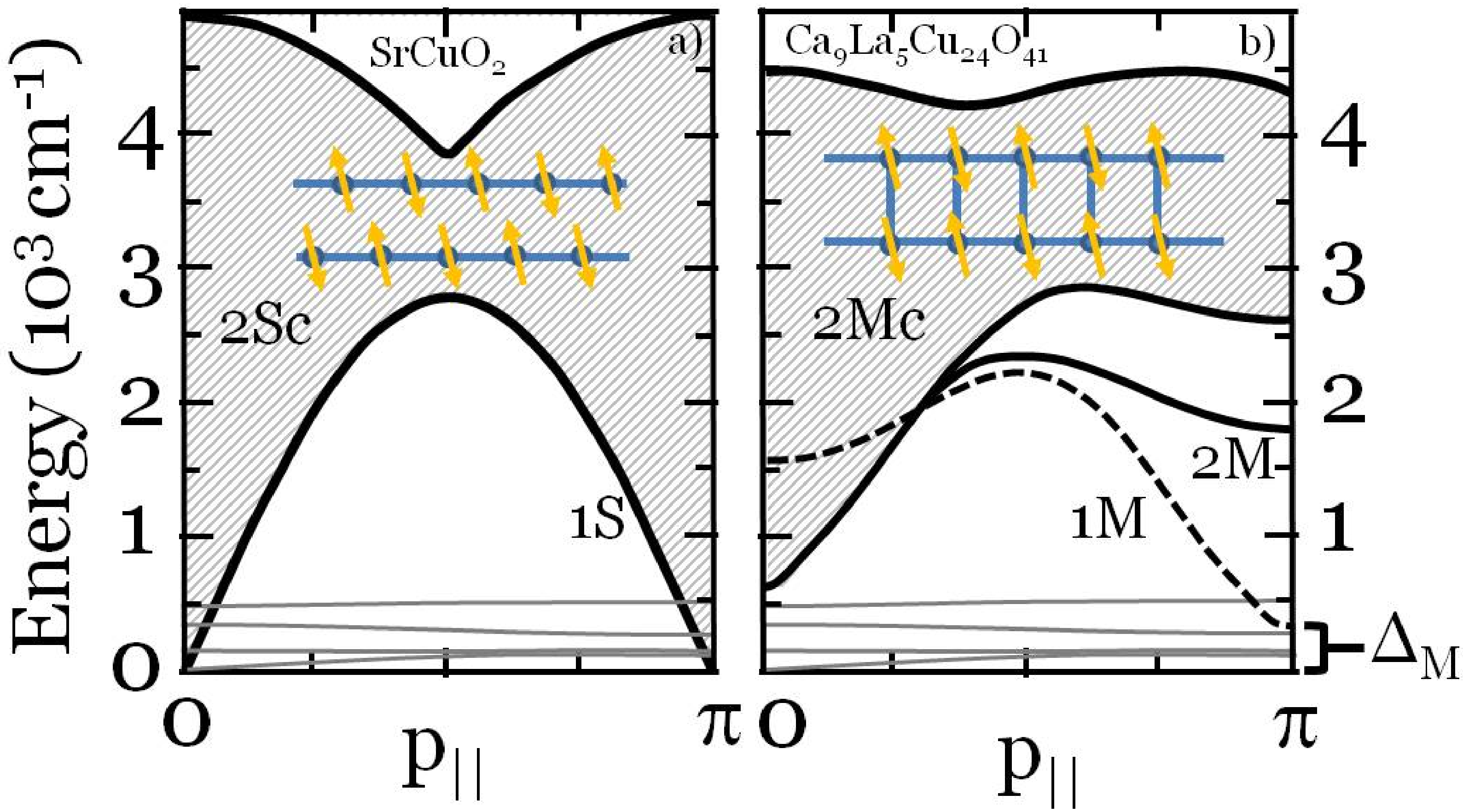






$J_{//}$=130 meV
$J_{\perp}$= 70 meV
Δ=32 meV

Magnetic excitations in chains and ladders



Chain:
2-Spinon excitations

Ladder:
2-Magnon excitations

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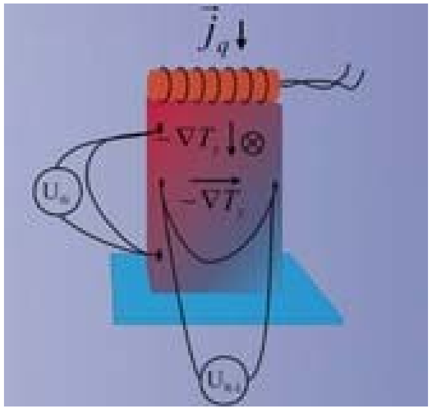
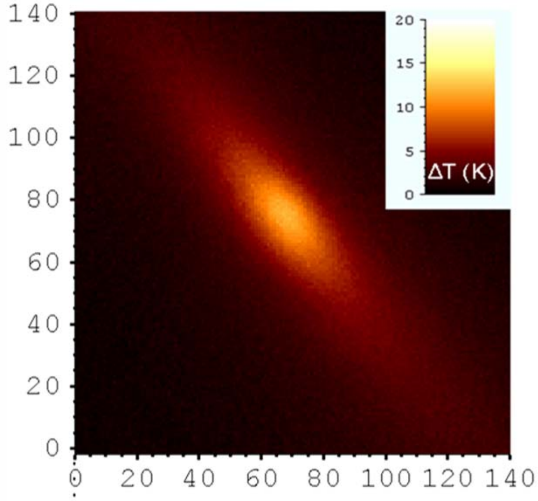
Measuring heat transport

- Traditional approach
  - Slow
  - Not suitable for films
- 3ω approach
  - Not fast enough
  - Elaborate
- Optical approach
  - Fast
  - Also on thin films
  - Easy





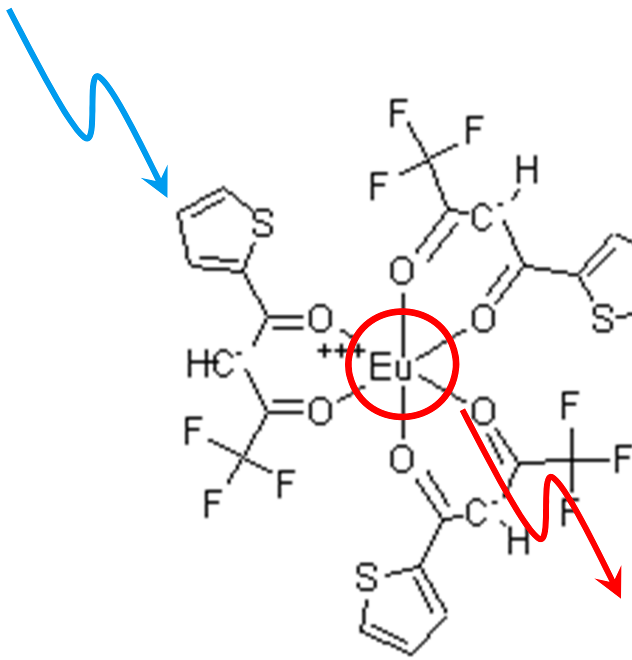
Otter et al., JMMM 321, 796 (2009)

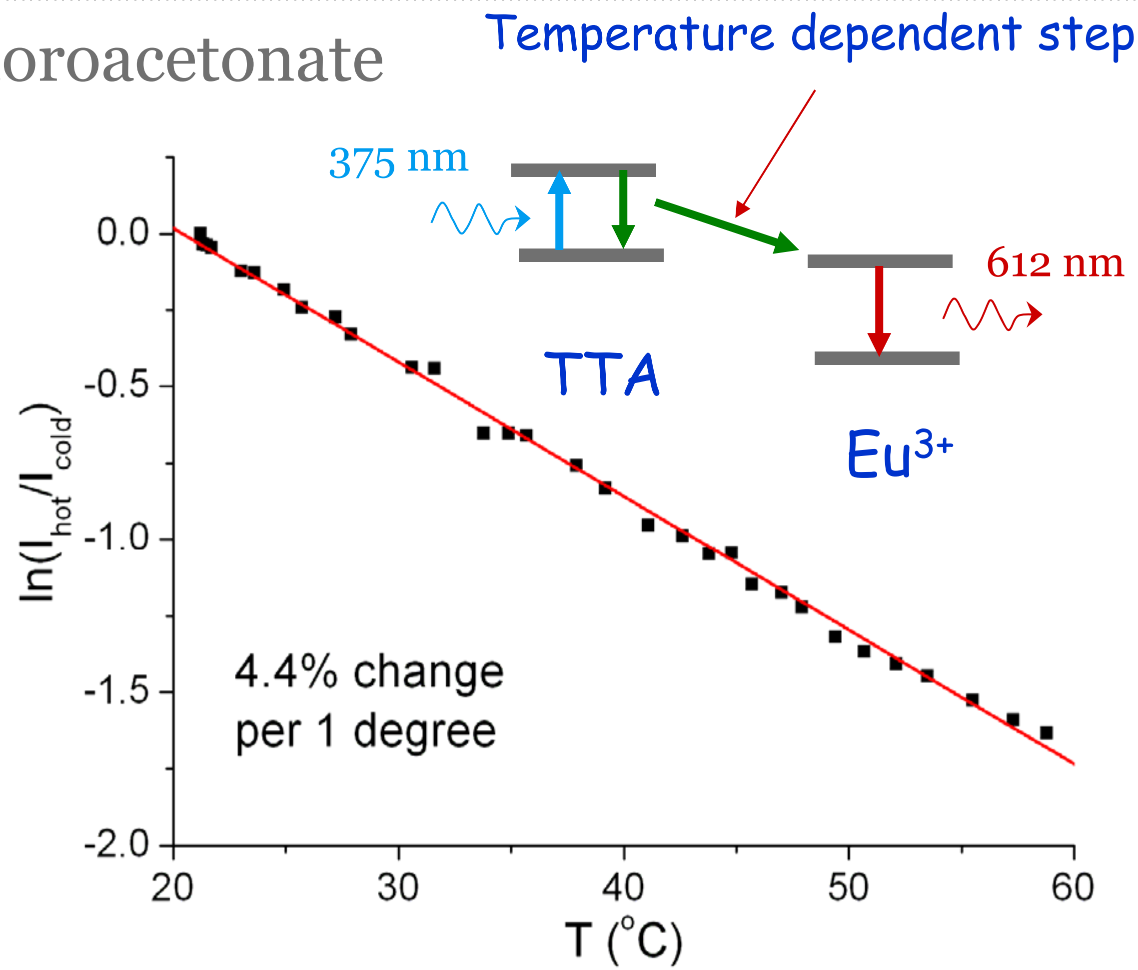
Measuring heat conductivity requires a heat source and temperature measurement

- Heat source: Laser
- Temperature detector: T-dependent luminescence

Optical temperature detection

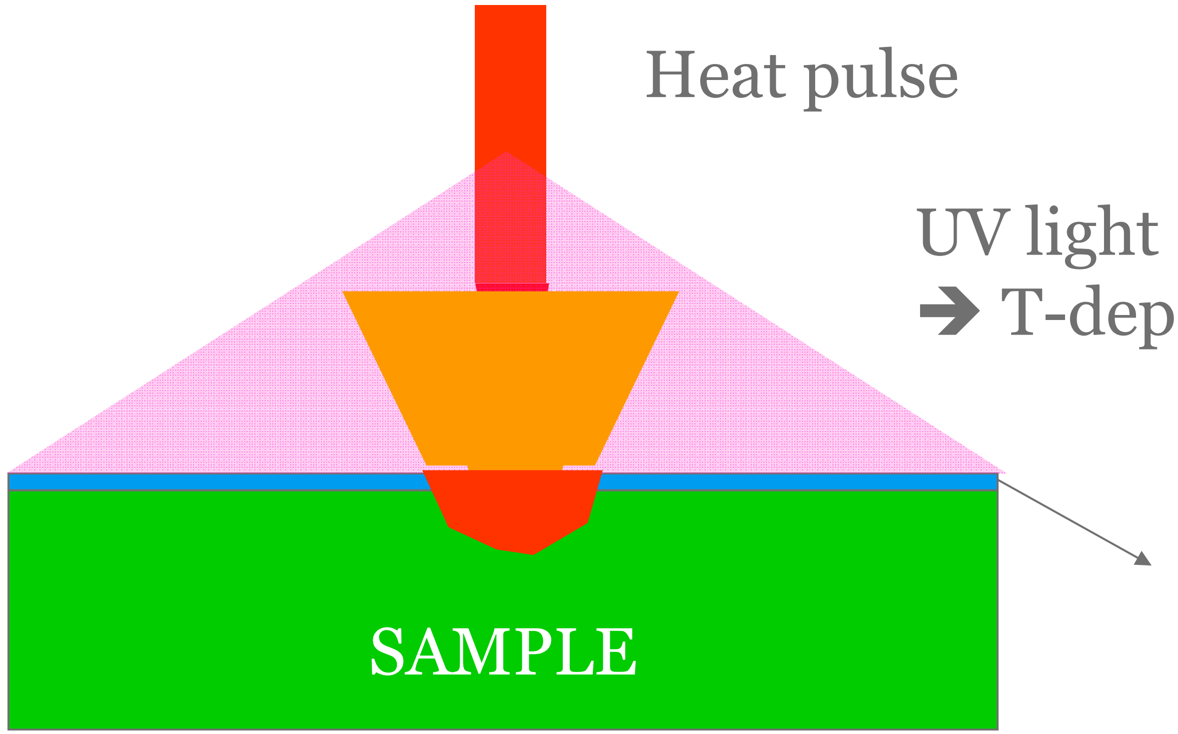
EuTTA: Eu-thenoyltrifluoroacetonate





→ Spin coat EuTTA + dPMMA layer on crystals

Imaging heat transport



Otter, PvL, et al. JMMM **321**, 796 (2009); Patent EP11163209 (T-Storm);

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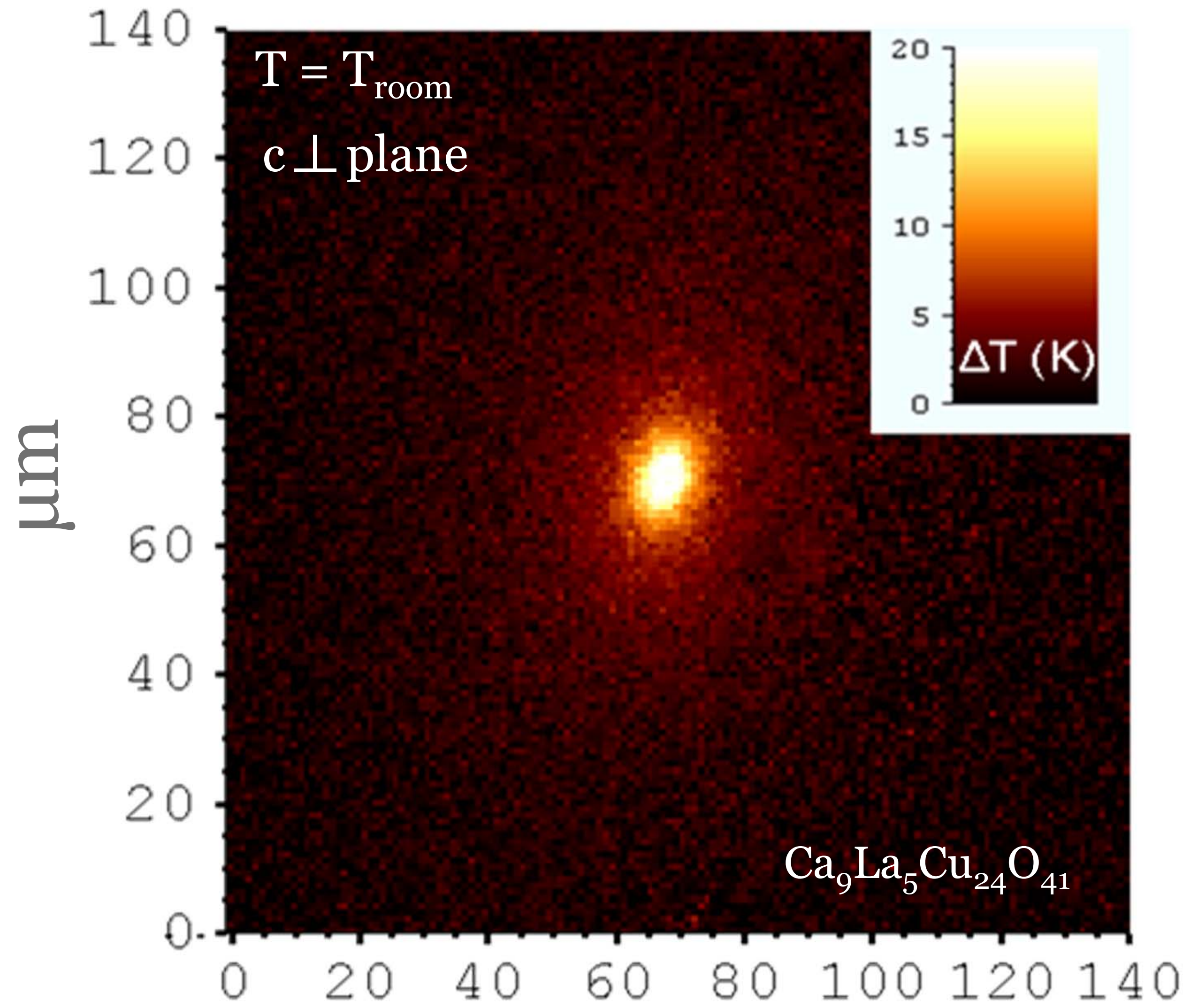
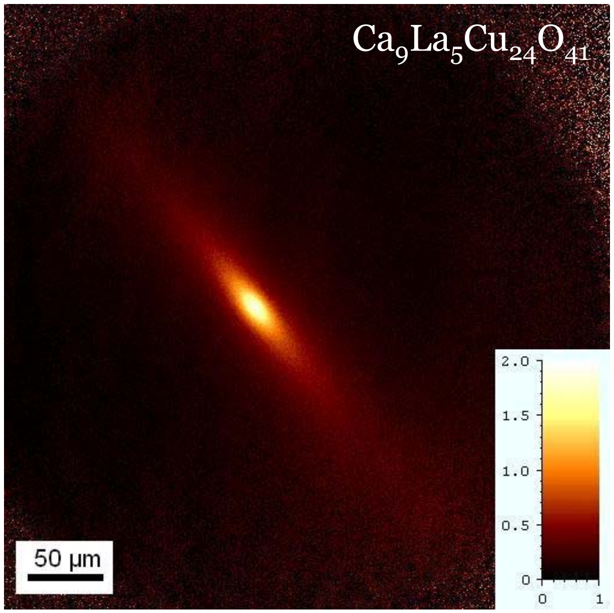
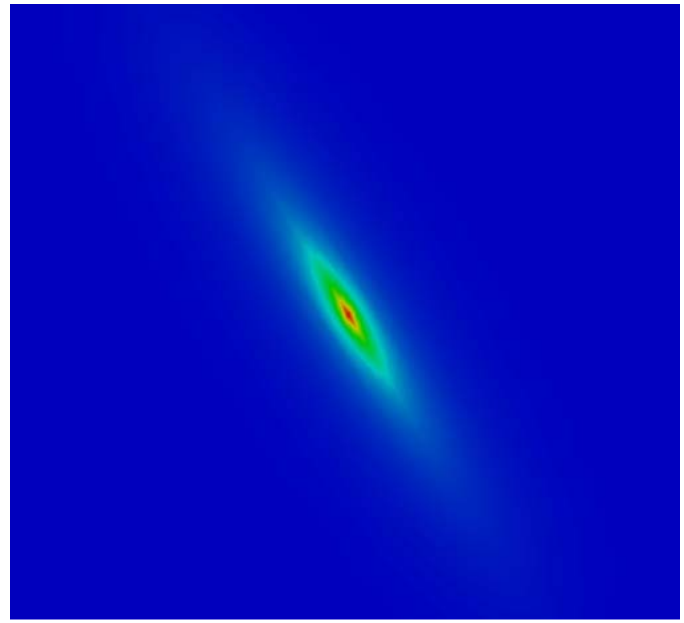
Static thermal imaging: ab-plane



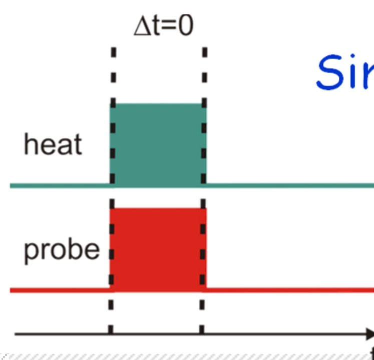
Static thermal imaging: ac-plane



Experiment (RUG)

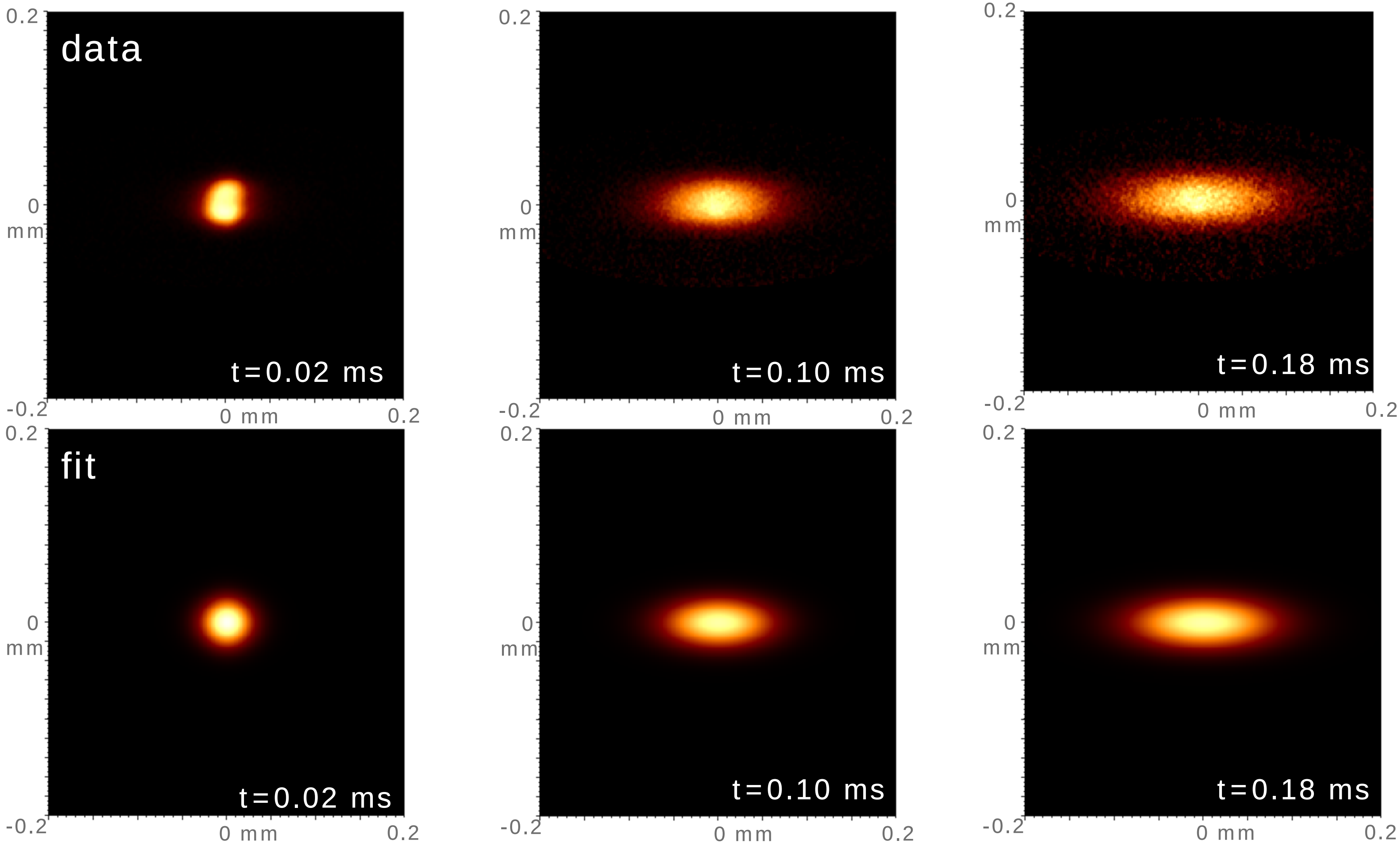


Simulation (ASCOMP)



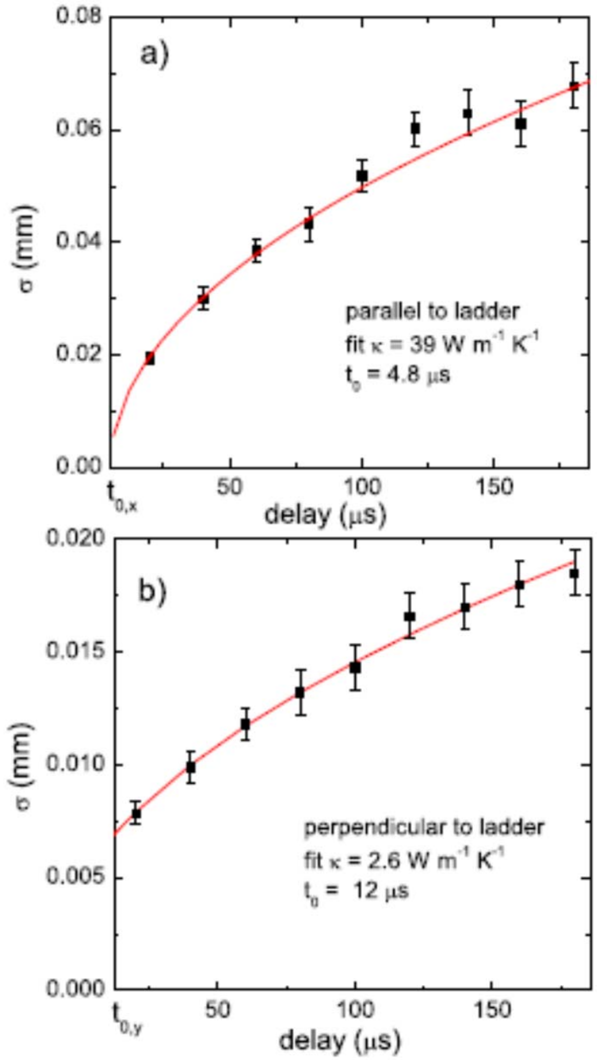
$$\frac{\partial T}{\partial t} = \nabla\left(\overleftrightarrow{D}\nabla \cdot T\right)$$

Heat transport in $La_5Ca_9Cu_{24}O_{41}$



Otter, PvL, et al. IJHMT **55**, 2531 (2012).

Extraction of heat transport coefficients



Analysis using diffusion model

Width: $\sigma_\nu = \sqrt{2D_\nu \cdot (t - t_{0,\nu})}$

$\kappa = \rho \cdot C_V \cdot D$

Perpendicular to ladder

κ = 2.6 W/m·K

Good agreement with DC

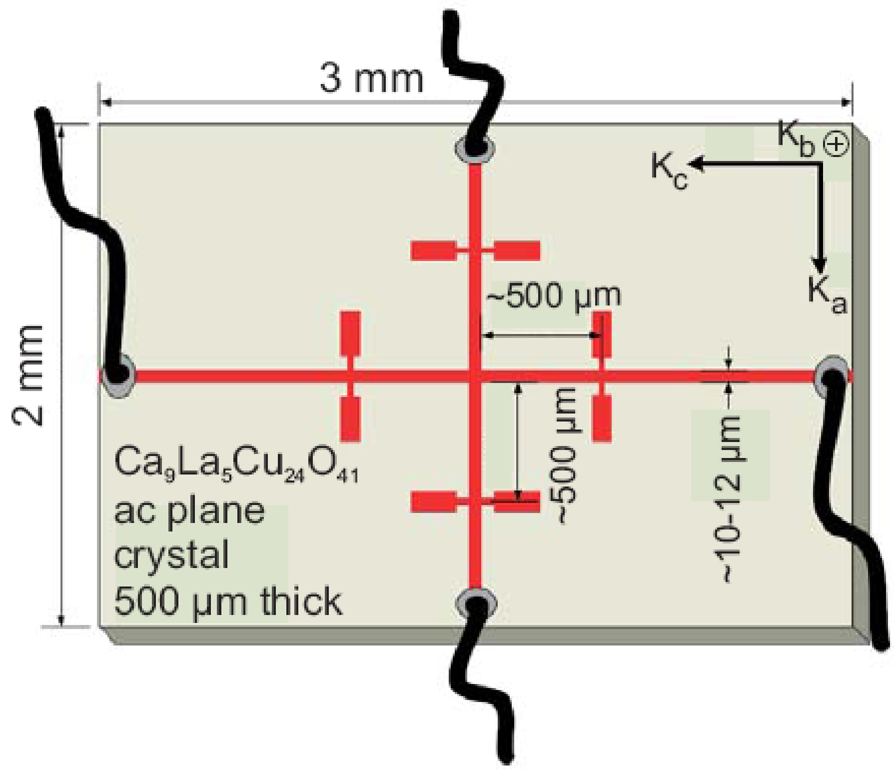
Parallel to ladder

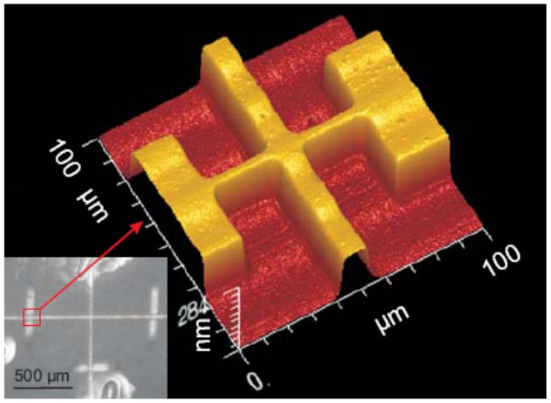
κ = 39 W/m·K

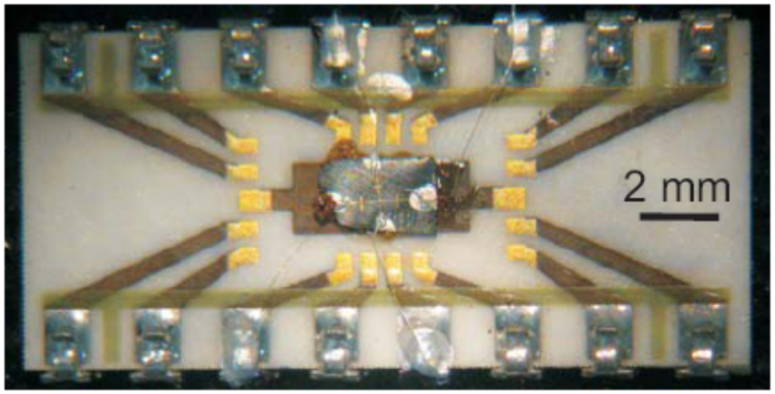
Bad agreement with DC

DC: κ = 80 W/m·K

Application in a device geometry





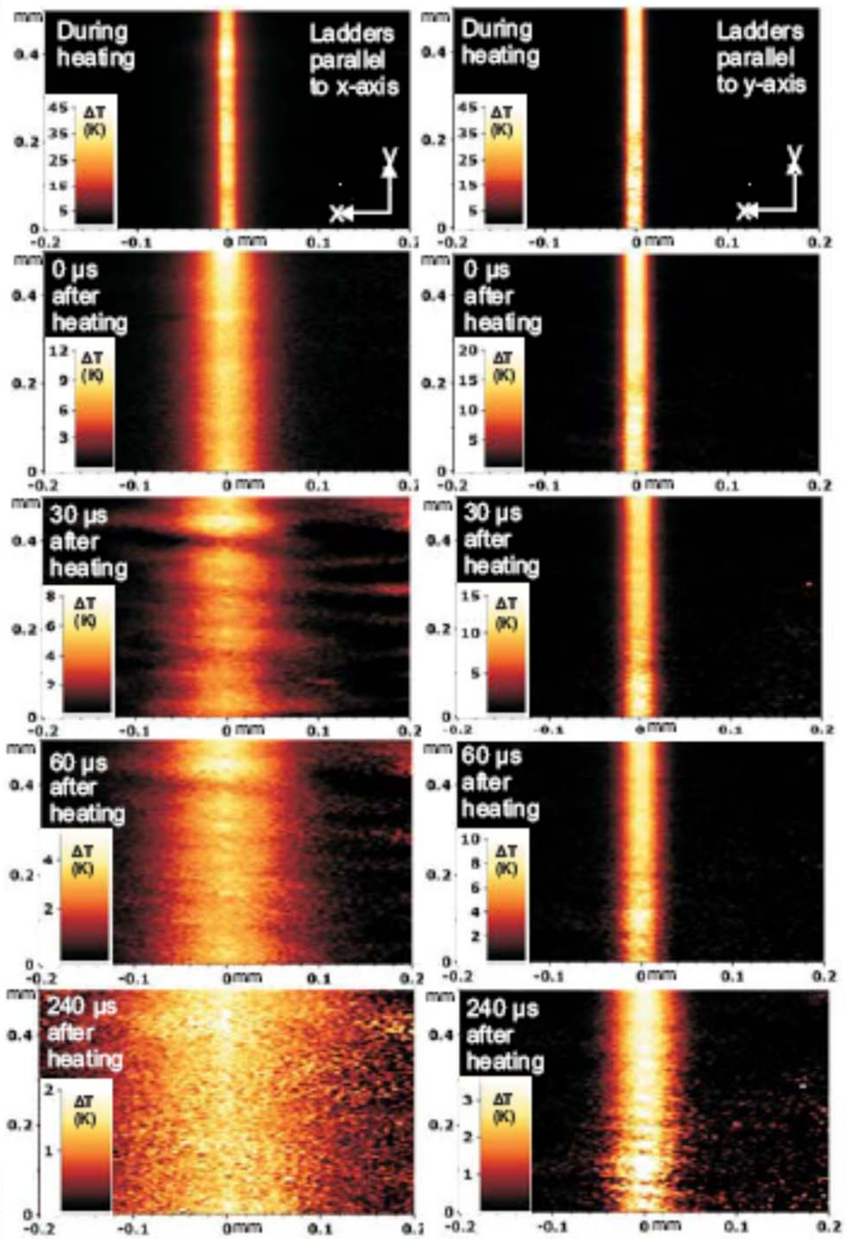


Hot strips along
a and c direction

Application in a device geometry

Strip perp. to ladder

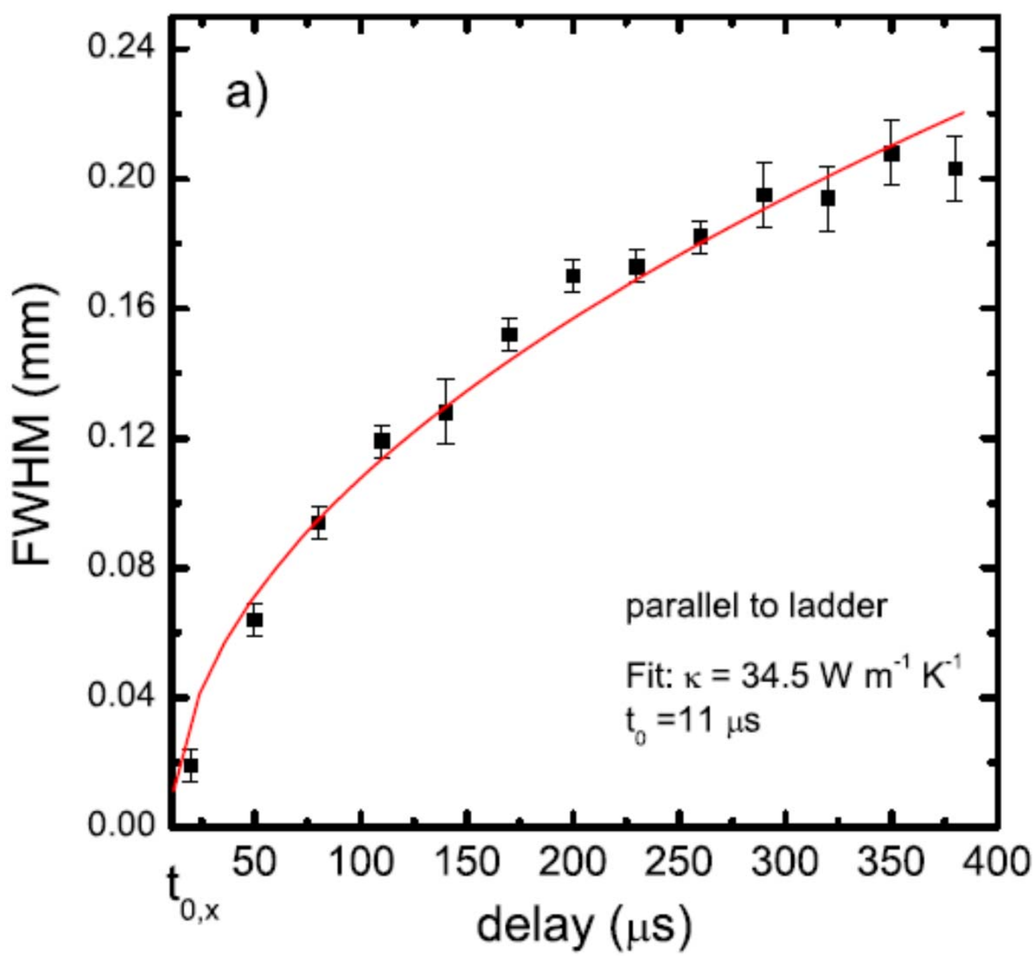
Good
(magnon) conduction
along ladder



Strip along ladder

Bad
(phonon) conduction
perp to ladder

Application in a device geometry



Heat conductivity

- Along a: 2.8 W/mK

- Along ladder (c-dir)

34.5 W/mK

Again: Discrepancy with DC !!!

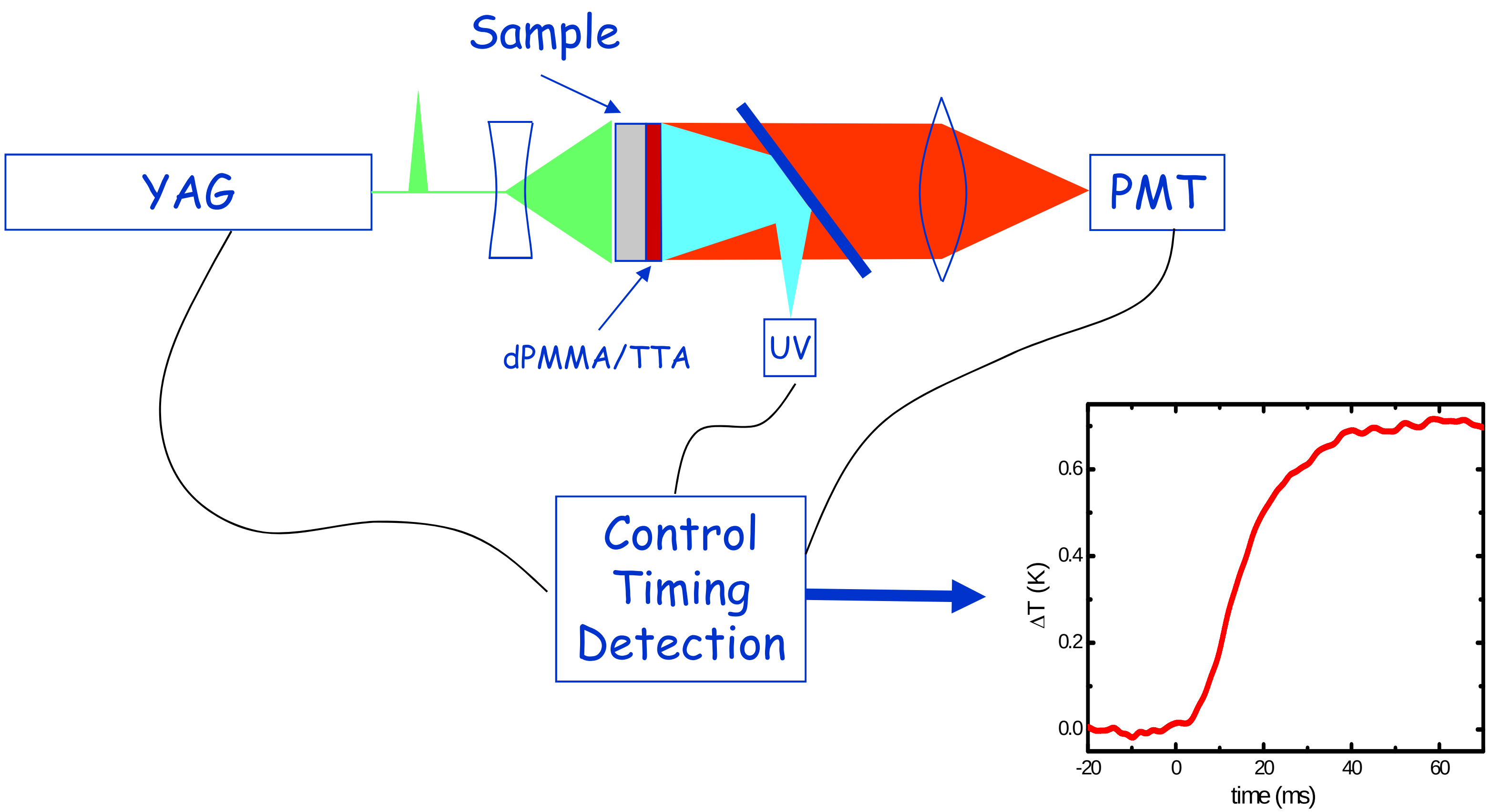
Surface vs. Bulk

Discrepancy maybe due to surface effects?

Let us look at the bulk transport, again using optics

Detection of bulk heat diffusion

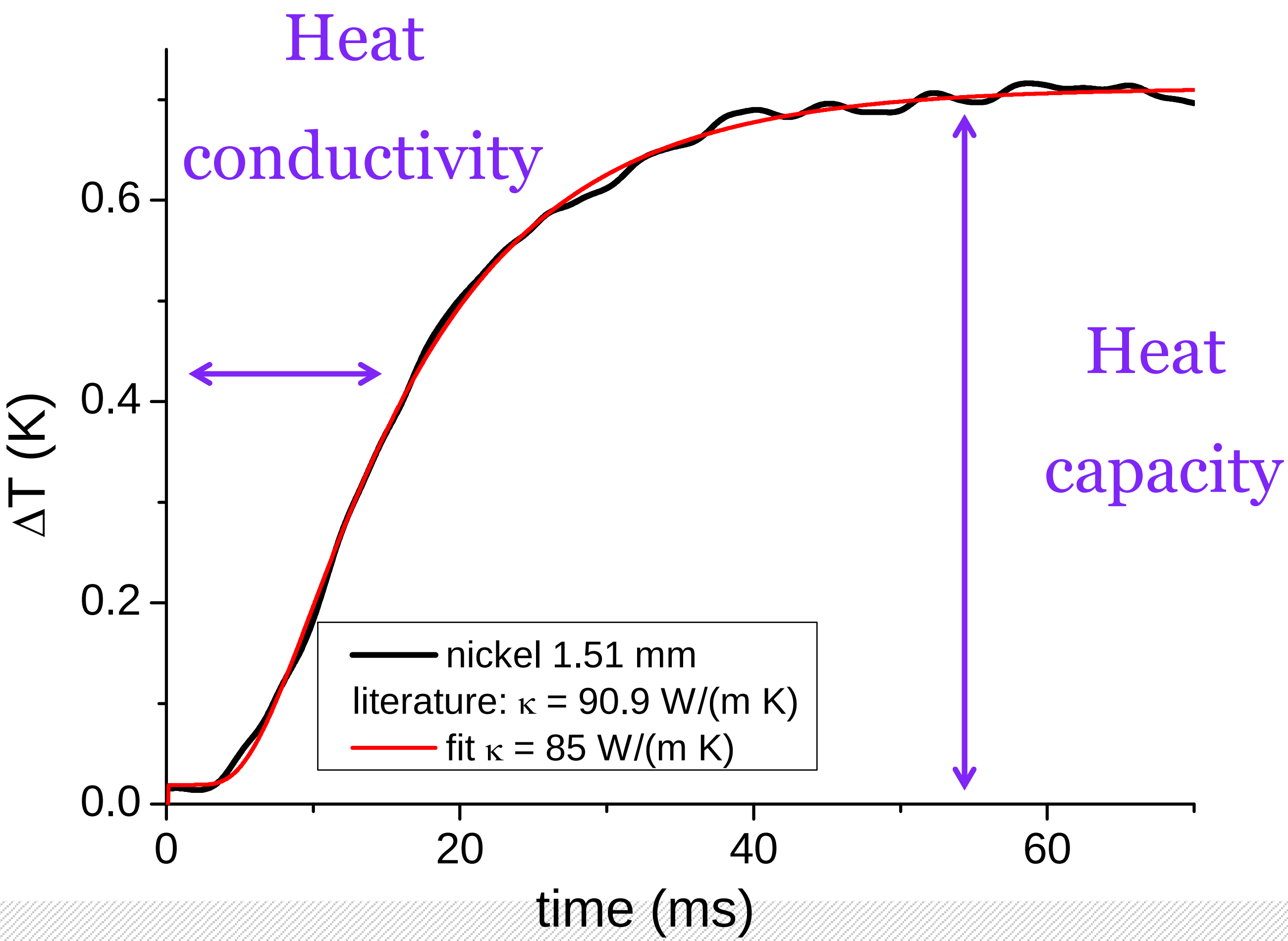
Heat diffusion set-up



Test experiment on Ni single Xtal

W.J. Parker et al, J. Appl. Phys. **32**, 1679 (1961).

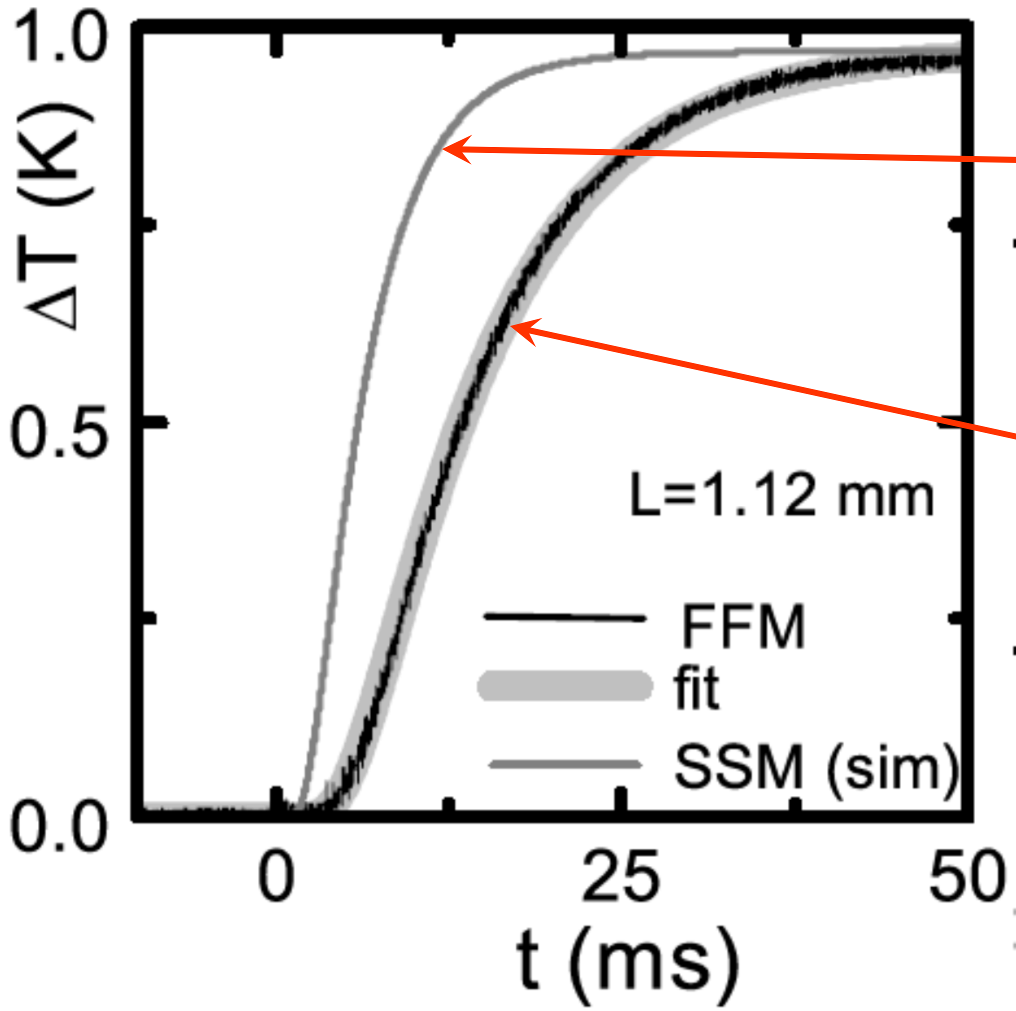
$$\Delta T(t) = 1 + 2\sum_{m=1}^{\infty}(-1)^m \exp(-m^2\pi^2 Dt/L^2)$$



$$\frac{\partial T}{\partial t} = D\frac{\partial^2 T}{\partial z^2}$$

$$D = \frac{\kappa}{\rho \cdot C_p}$$

Bulk diffusion $Ca_9La_5Cu_{24}O_{41}$



Expectation using DC value

Measured curve

➔ *Again discrepancy: Not a surface effect!*

Let's think again

We would like to probe magnon transport

One needs magnons for this

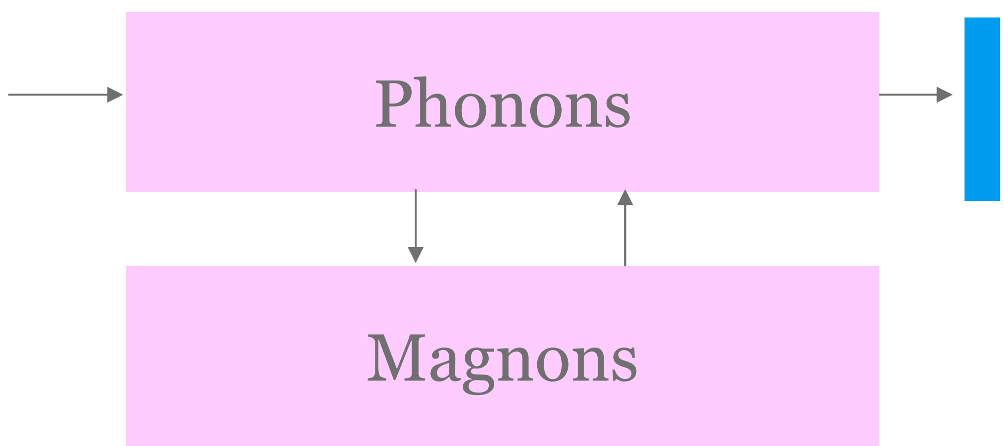
The experiments inject and detects phonons, not magnons !!!

In DC experiments, lots of time for equilibration

Optical experiment = Dynamic experiment !!!

Two particle diffusion

Heat in
(optical, Joule heating)



EuTTA
Detection

How fast is the conversion into magnons ???

Two particle diffusion model

Coupled diffusion equations

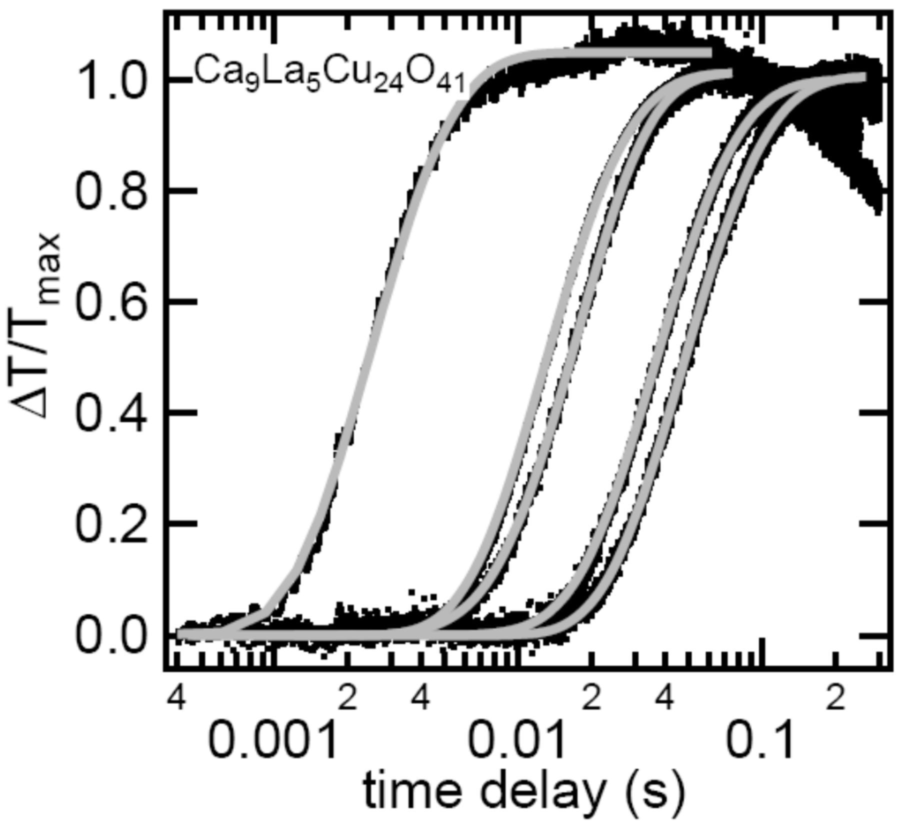
$$C_L\,\partial_t T_L = \kappa_L\,\partial_x^2 T_L - g\left(T_L - T_M\right)$$

$$C_M\,\partial_t T_M = \kappa_M\,\partial_x^2 T_M + g\left(T_L - T_M\right)$$

$$g = \frac{C_L C_M}{C_L + C_M}\cdot \tau_{LM}$$

Solution T(x=L,t) depends on thickness sample

Thickness dependence



Room T

Thickness l.t.r.

0.4 mm

1.1

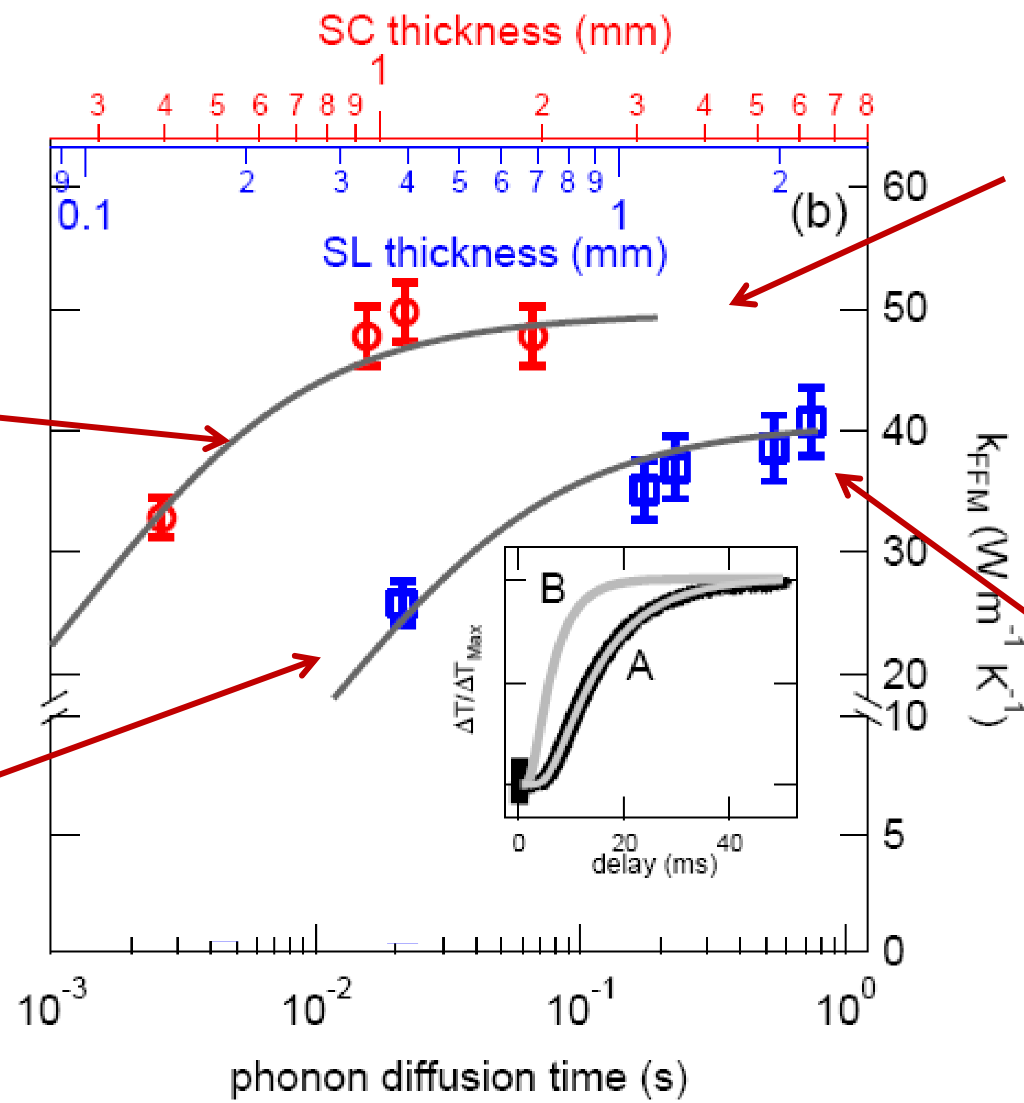
1.25

1.9

2.3

Fits to a 3D two particle heat transport model

Comparison to chain material



Chain (SrCuO$_2$)

= DC value

Chain

< 1 ns

Ladder

< DC value

Ladder

400 µs

$$\tau_d = 0.48 \frac{L^2 C_p}{\pi^2 k_b}.$$

Dynamic heat conductivity

› Chain material: No large differences with DC xpt

› Ladder material
  - Equilibration time: 300 μs
  - ‘saturation’ value ~ half the DC value

The long time is not necessarily a surprise

PHYSICAL REVIEW B VOLUME 15, NUMBER 3 1 FEBRUARY 1977

Effect of magnon-phonon thermal relaxation on heat transport by magnons*

D. J. Sanders† and D. Walton
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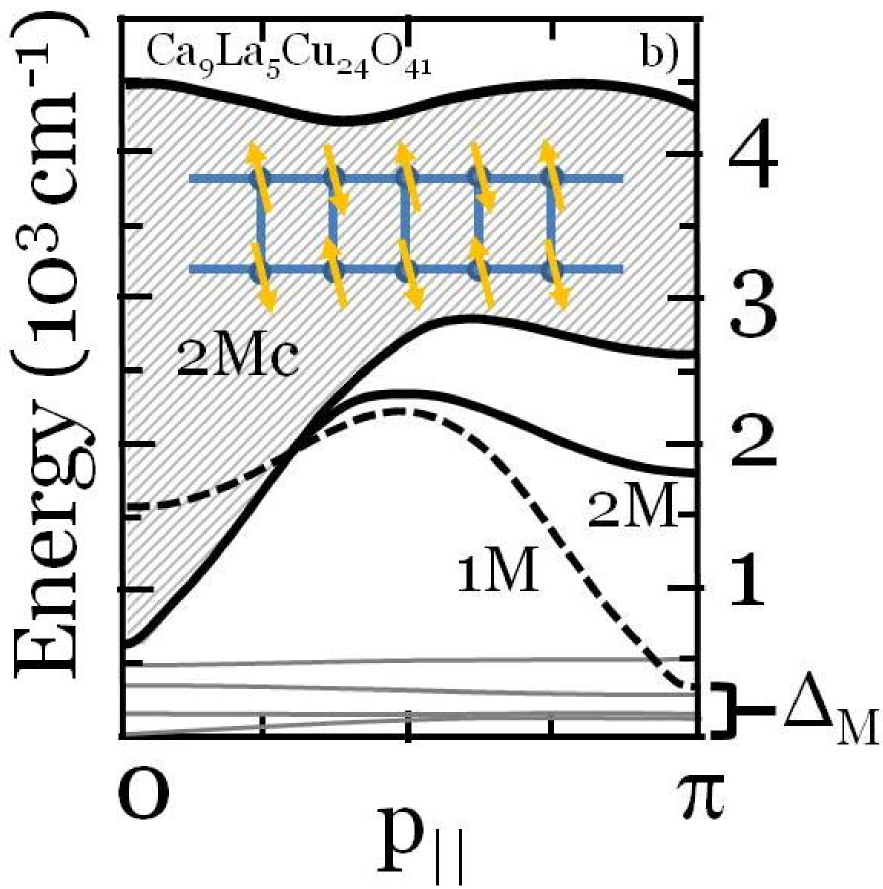
(Received 24 June 1976)

Alternatively, it should be noted[34] that τ_{mp} varies as B_2^2 so that τ_{mp} for MnF_2 should be roughly 100 times longer than that for YIG or ~10^{-4} sec.

For MnF_2 phonon-magnon scattering time is estimated to be about 100 μs

Why slow



Heat mostly injected as acoustic phonons

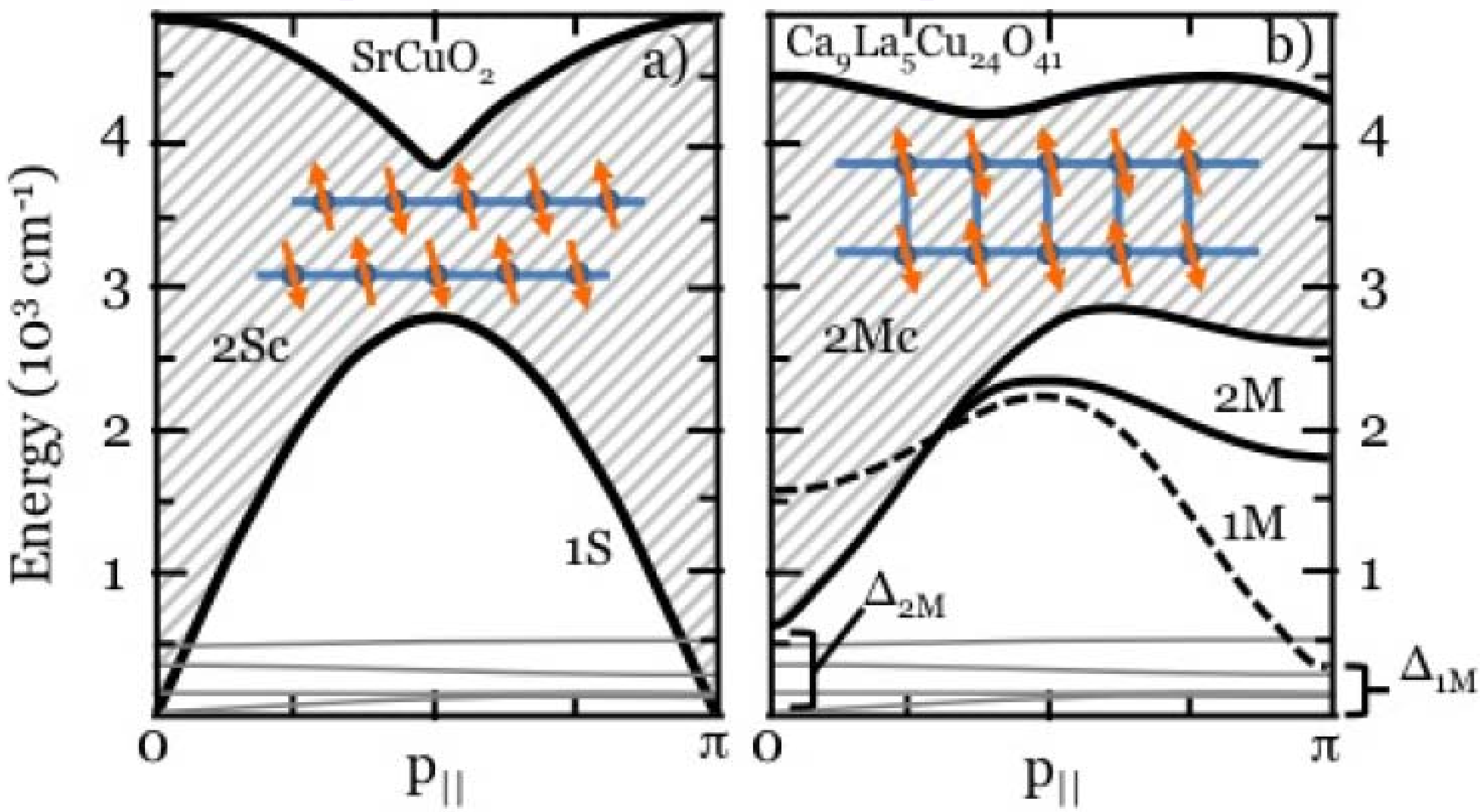
Magnon gap $\Delta_M \sim 450$ K

Phonon-magnon conversion violates

- Energy conservation
- Spin conservation
- Momentum conservation

Why not in the spin chain material ?

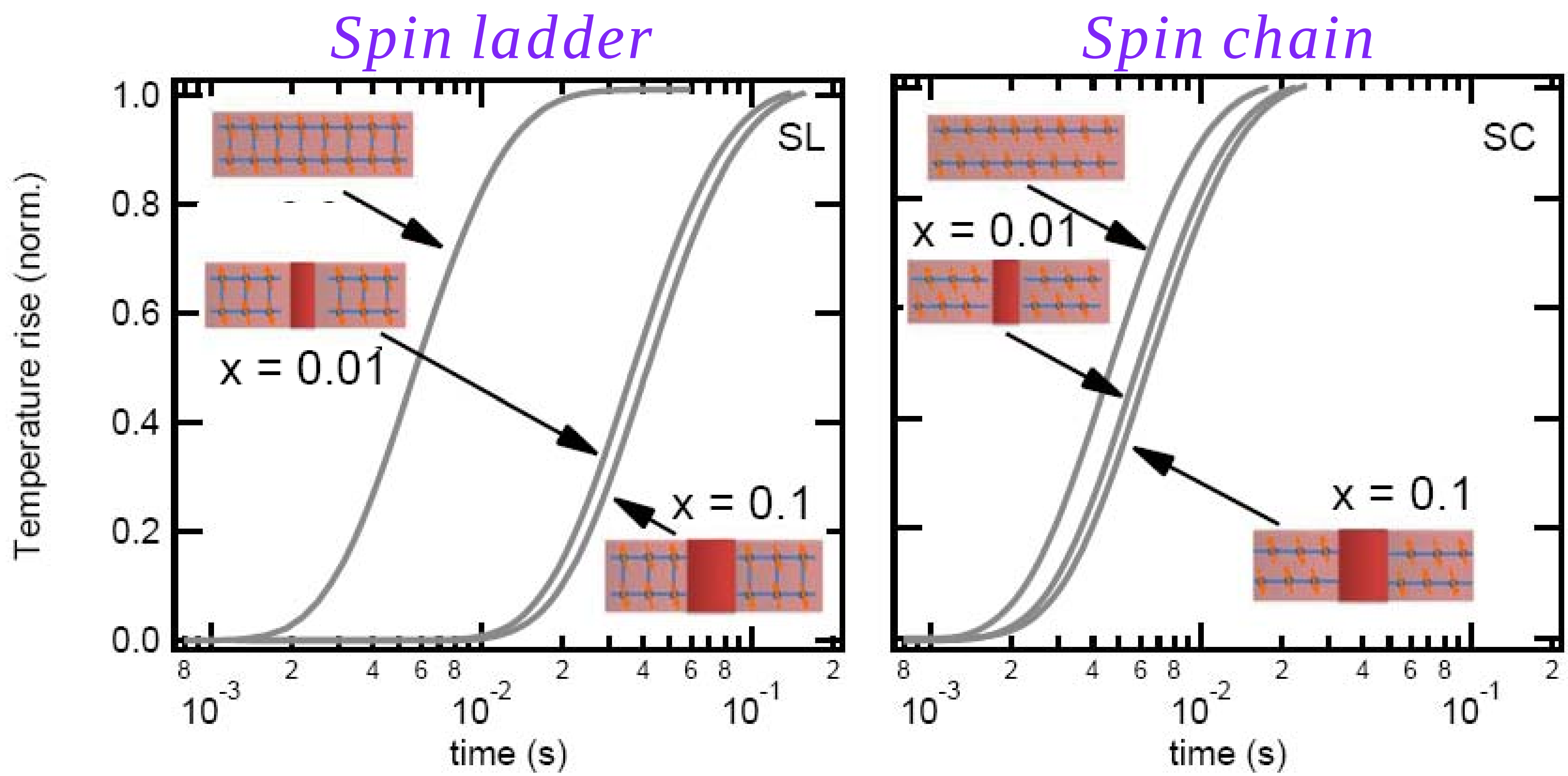
Spin chain *Spin ladder*



Final value discrepancy

Defect (finite spin chains/ladders) limited

Dynamic conductivity



Conclusions

Quantum magnets for heat transport

› High thermal conductivity
› uni-directional
› electrically insulating

Diffusion and thermal imaging provide all-optical heat transport detection methods

› straightforward with a fast response time
› bonus: heat capacity
› applicable to bulk and thin films and easy to integrate in pump-probe methods

Dynamic heat transport

› Allows for a direct measurement of phonon-spin interaction
› Coupling in ladder material very weak due to general conservation laws (E,k,S)

RUG

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Thank you for your attention