Experimental study of spinon-phonon coupling in spin chain cuprates

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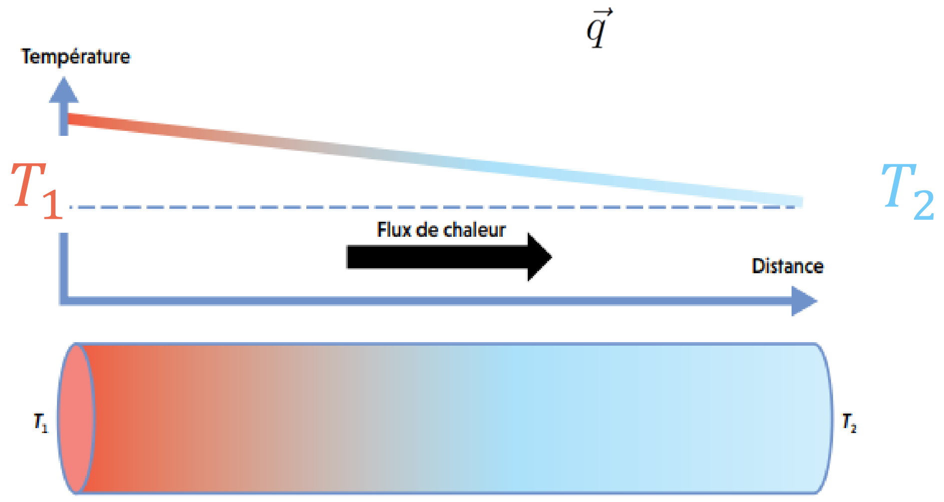
*Submitted to PRB, Dec 2022

Transport coefficients From macroscopic…

Perturbation	Response	Coefficient (tensor)
Magnetic field	Magnetization	Magnetic susceptibility μ
Electric potential	Electric current	Electric conductivity σ
Temperature gradient	Heat flow	Heat conductivity κ

$$\vec{q} = -\kappa \vec{\nabla} T$$

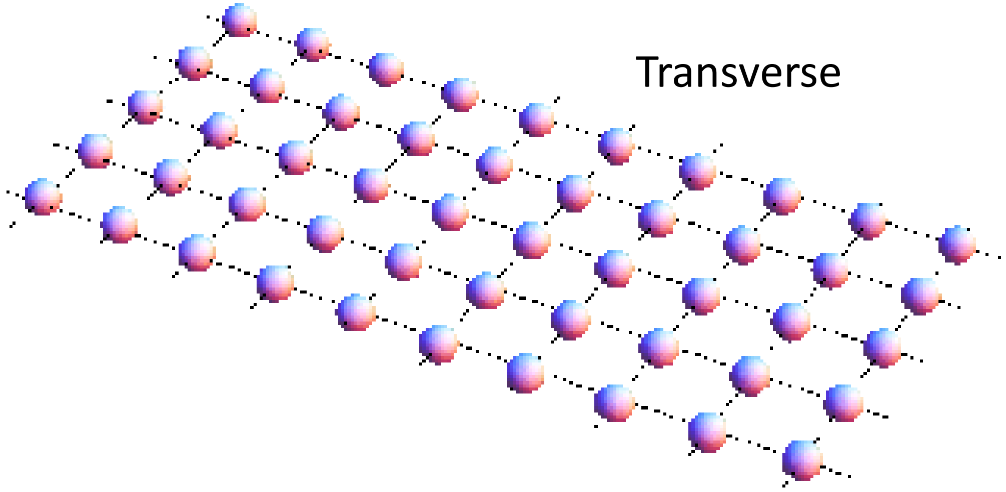


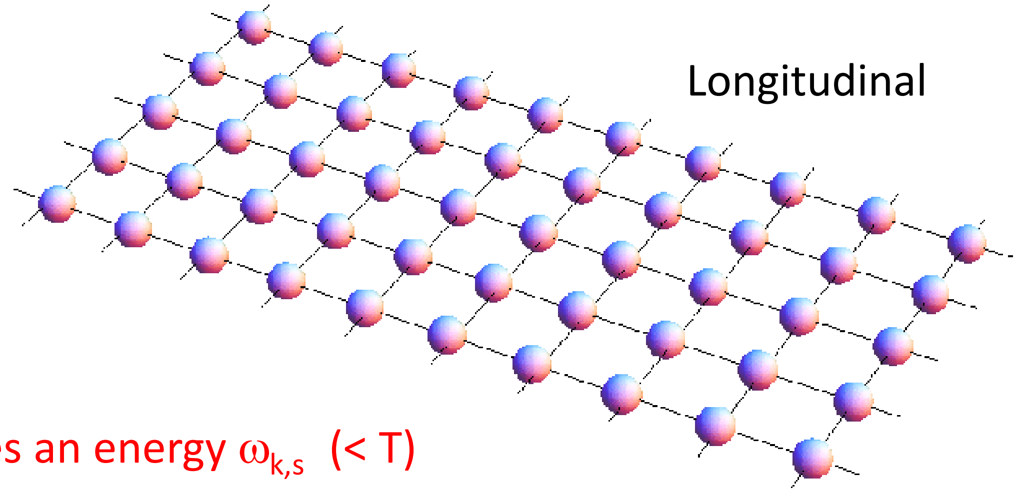
Transport coefficients … to microscopics

Insulators : no contribution from electrons

$$\kappa \approx \frac{1}{\Omega} \sum_{k,s} C_{k,s}\; v_{k,s}\; \ell_{k,s}$$

mean free path





$$v\;\tau = \ell$$

Relaxation time

$$v_{k,s} = \frac{\partial \omega_{k,s}}{\partial \vec{k}}$$

Velocity of the particles

$$C_{k,s} = k_{\mathrm{B}}\; \frac{x^2\; e^x}{(e^x - 1)^2}, \quad x = \omega_{k,s}/T$$

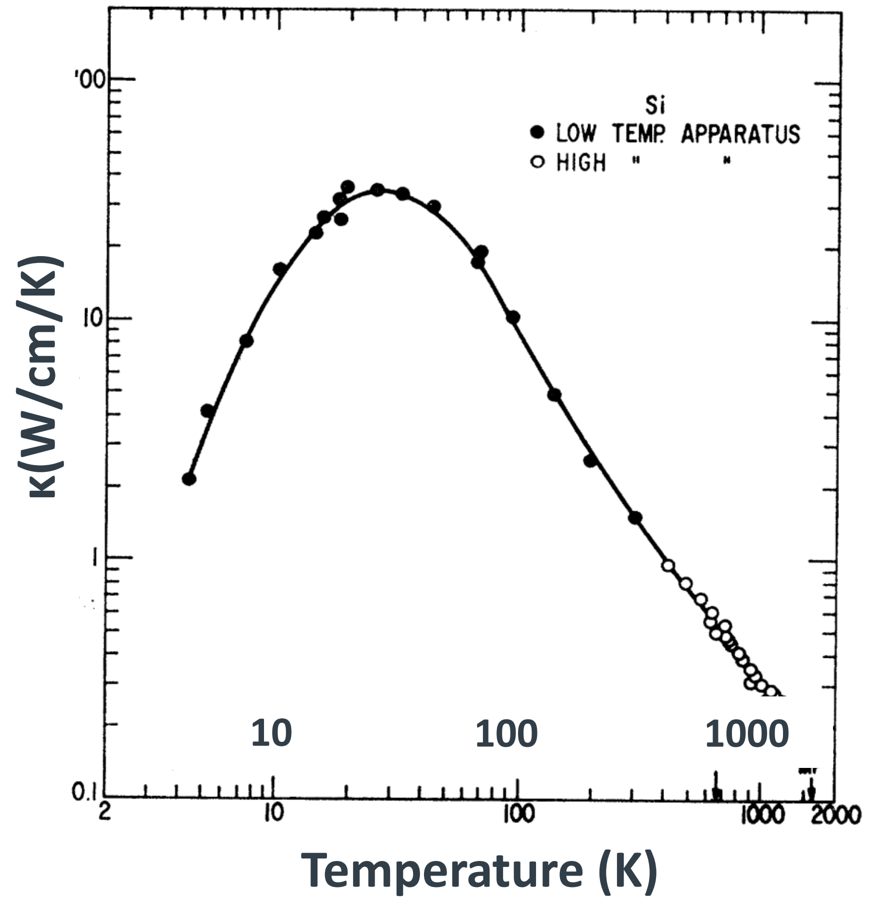
Each populated mode contributes an energy $\omega_{k,s}$ (< T)

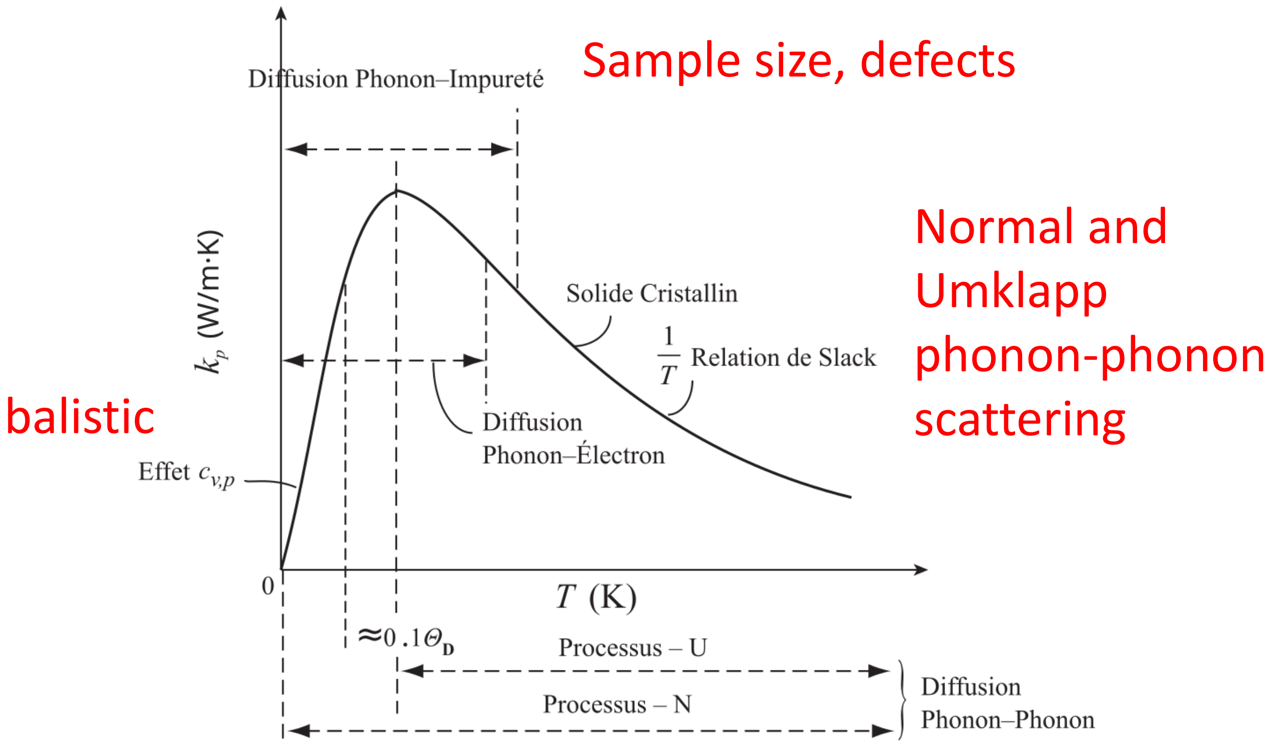
Acoustic modes only !!

Introduction

Ex : heat conductivity in Silicon

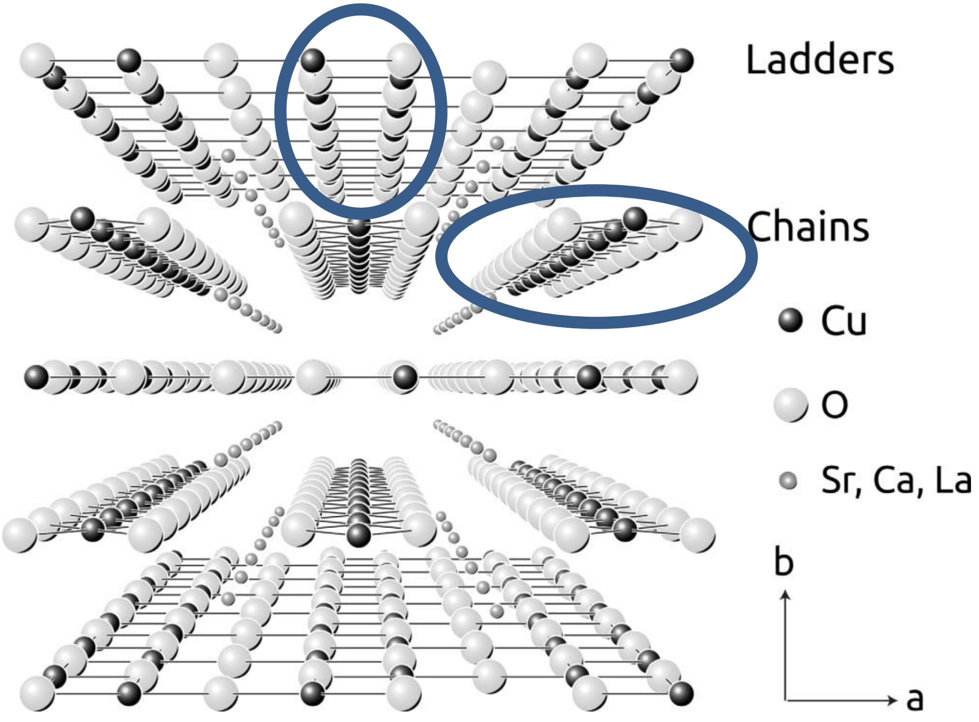
$$\tau_{tot}^{-1} = \tau_S^{-1} + \tau_D^{-1} + \tau_N^{-1} + \tau_U^{-1}$$

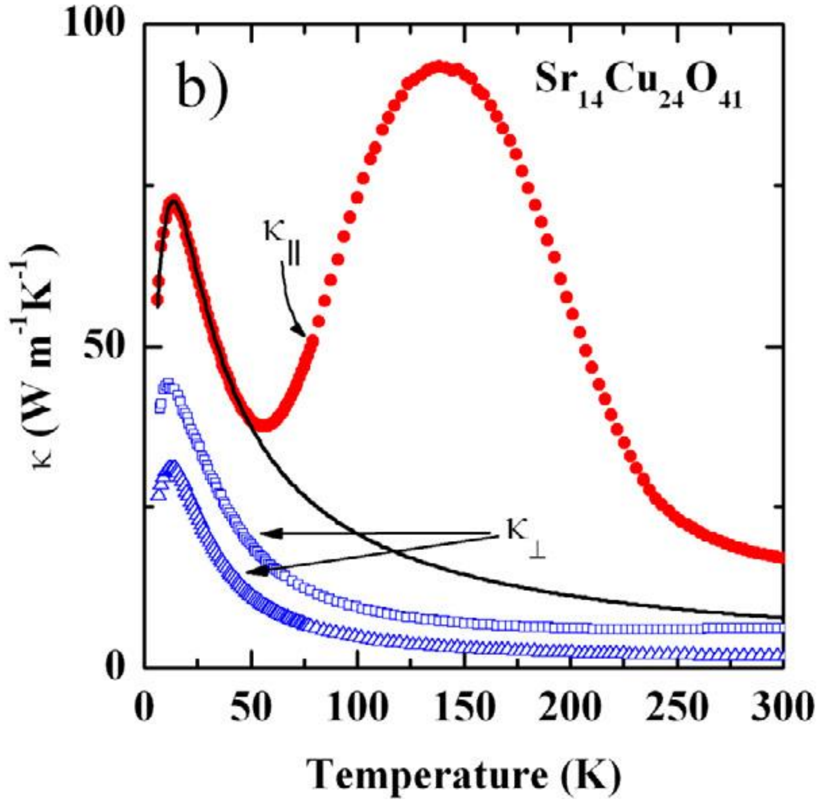




Glassbrenner & Slack, Phys. Rev. 134, A1058 (1964)
Kaviani, Heat transfert Physics (2012)

In addition to phonons, emergent particles propagate along chains and ladders (spin ½ Cu^{2+}) and participate to heat transport. κ is strongly anisotropic.





Hess et al., Physics Reports 811 (2019)

New materials : $Sr_{14}Cu_{24}O_{41}$

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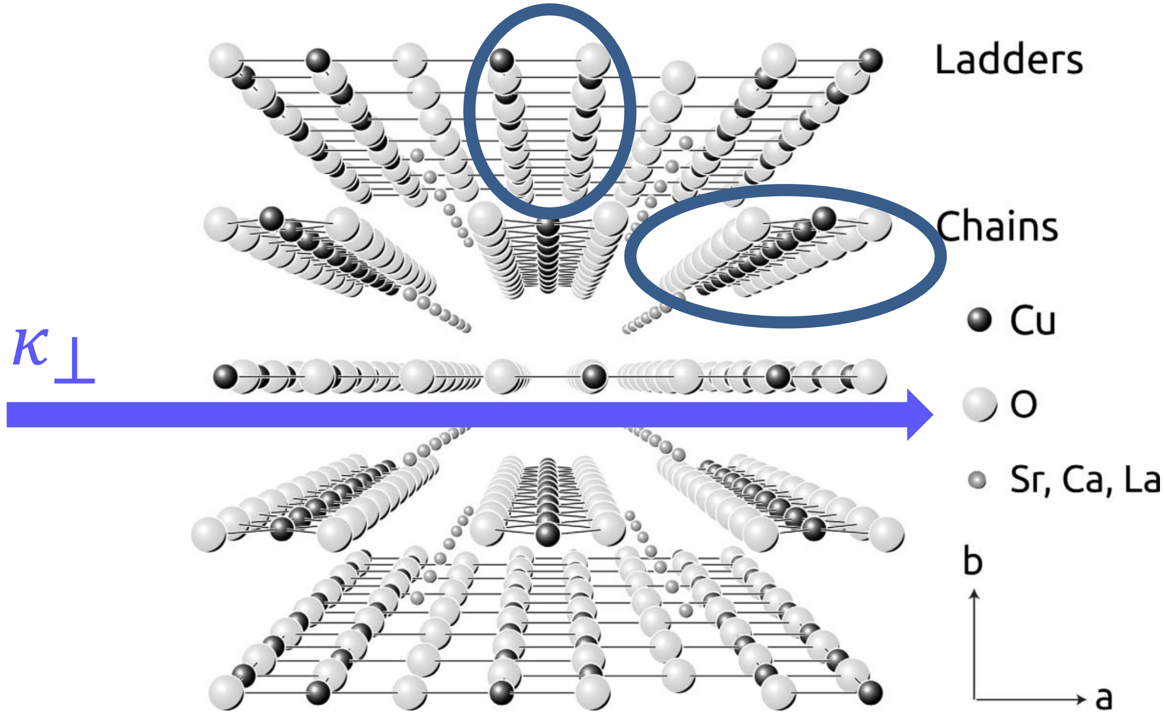

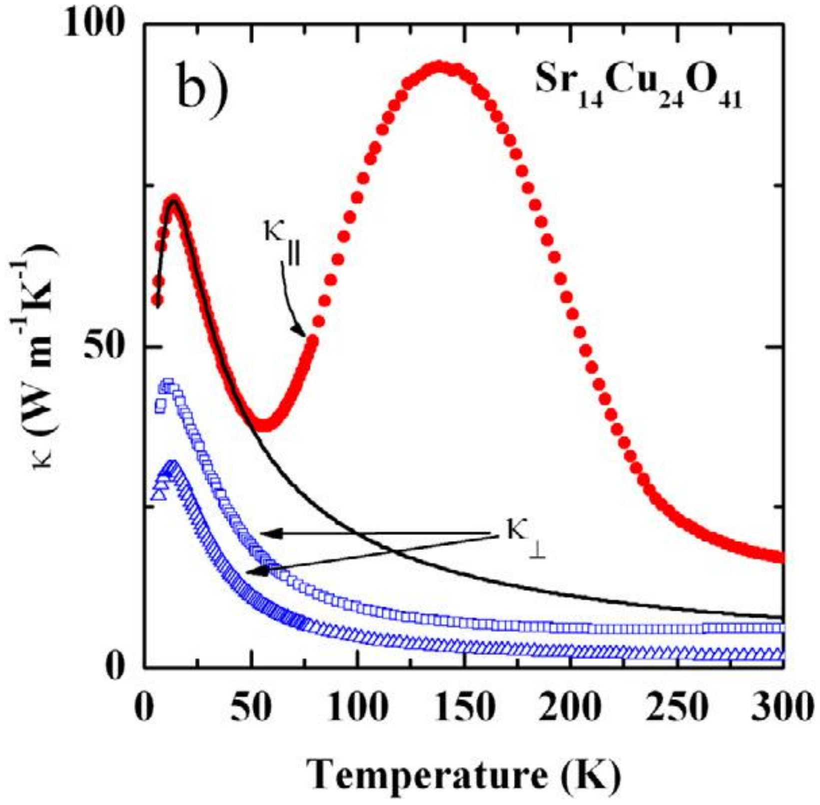

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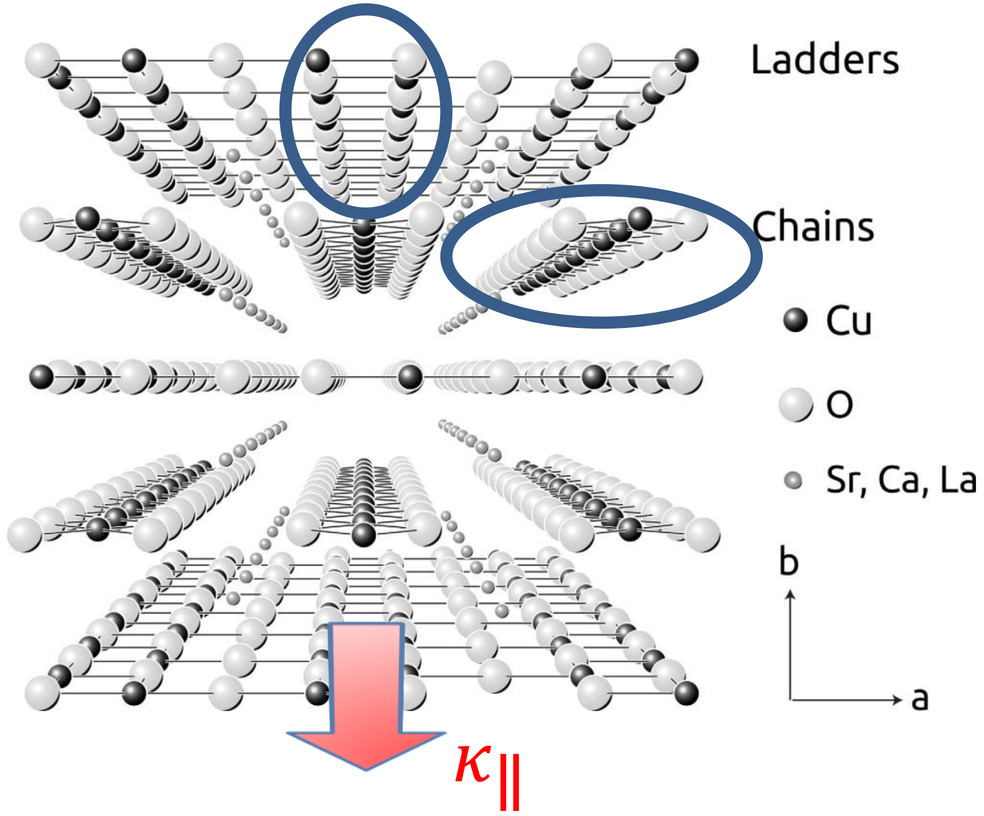

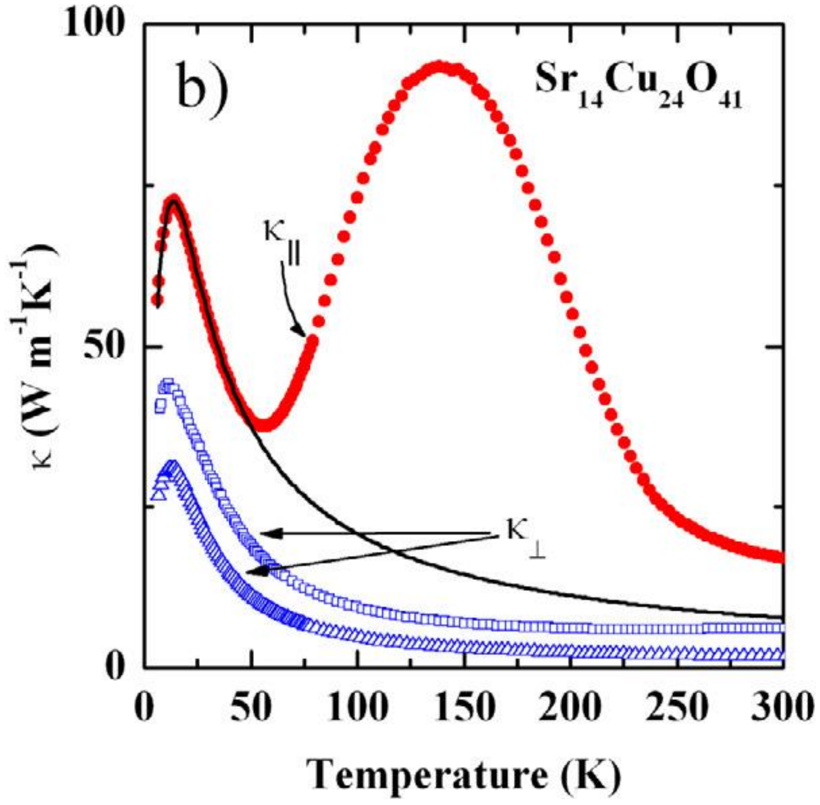

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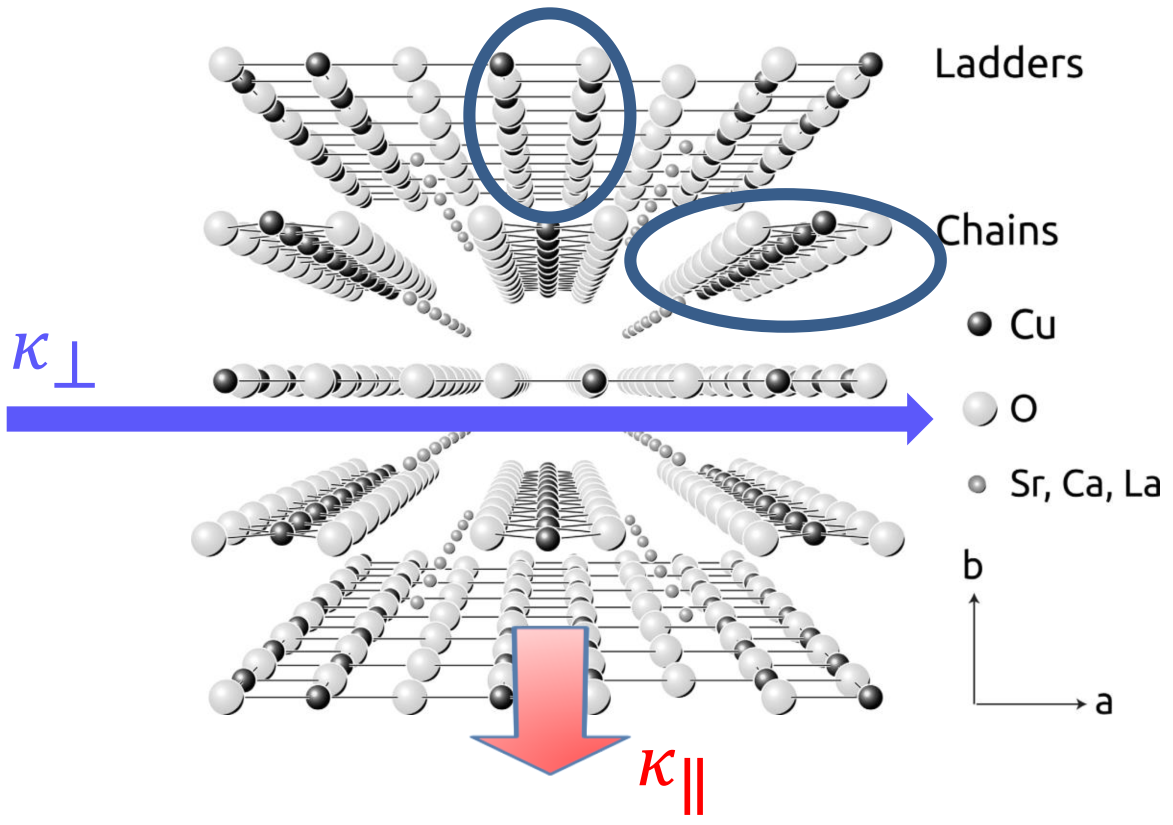
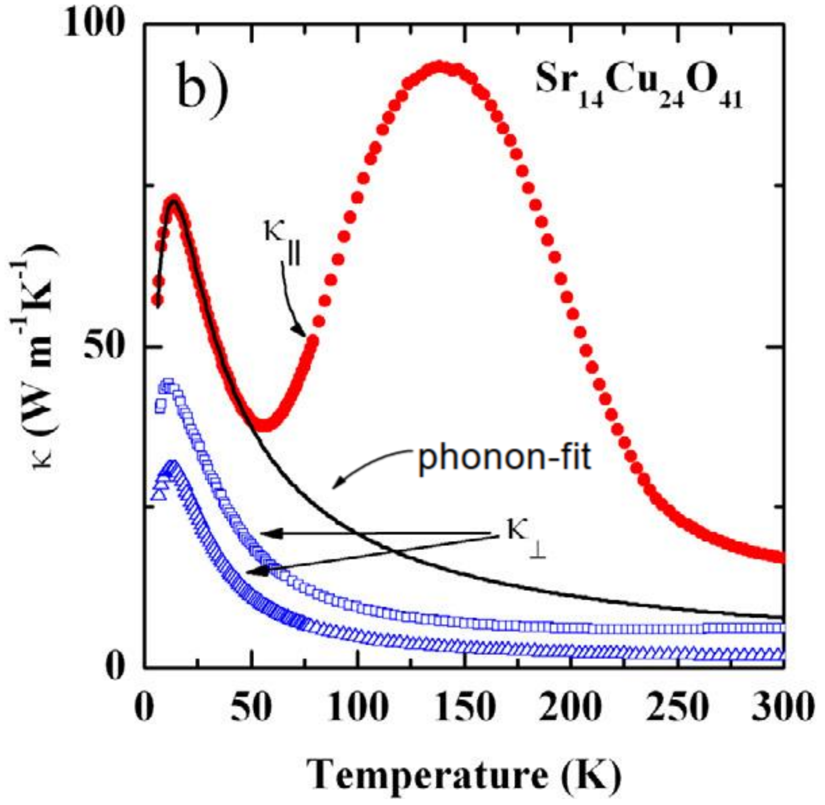
$$\kappa_c = \kappa_{ph} + \kappa_{mag} \longrightarrow \kappa_{mag} = \kappa_{\parallel} - \kappa_{\perp}$$

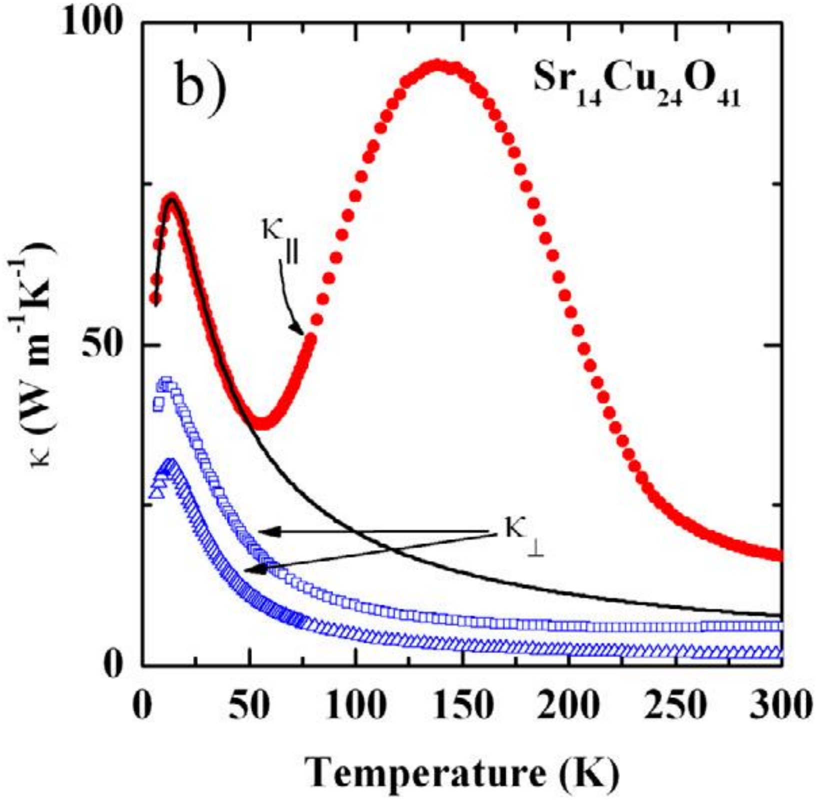


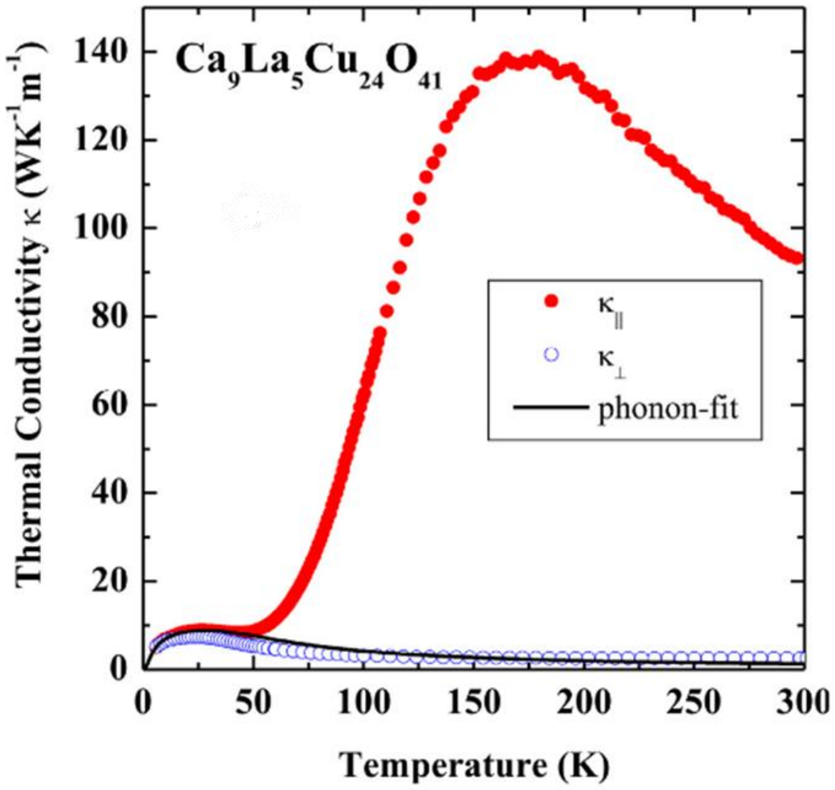


Hess et al., Physics Reports 811 (2019)

but also $Ca_9La_5Cu_{24}O_{41}$

$$\kappa_c = \kappa_{ph} + \kappa_{mag} \longrightarrow \kappa_{mag} = \kappa_{\parallel} - \kappa_{\perp}$$





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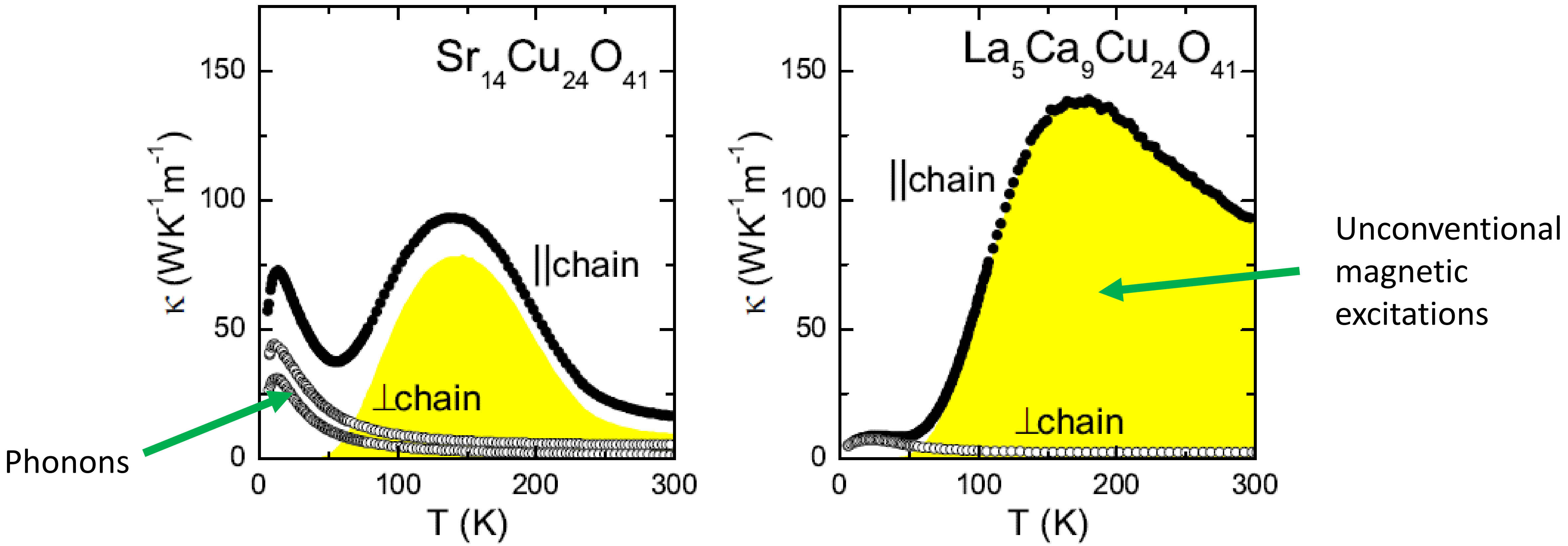
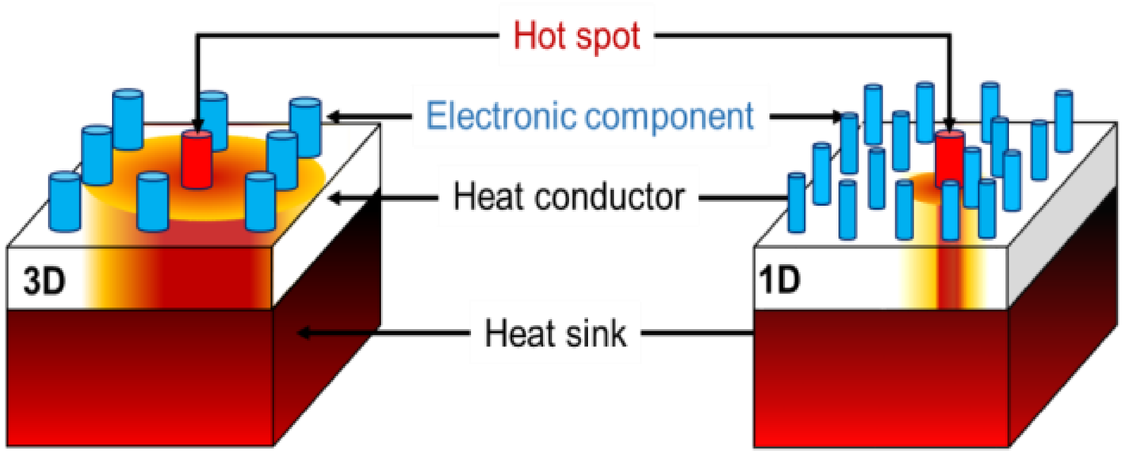
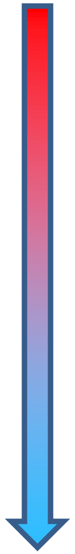


Hlubeck, PhD thesis

Applications ?



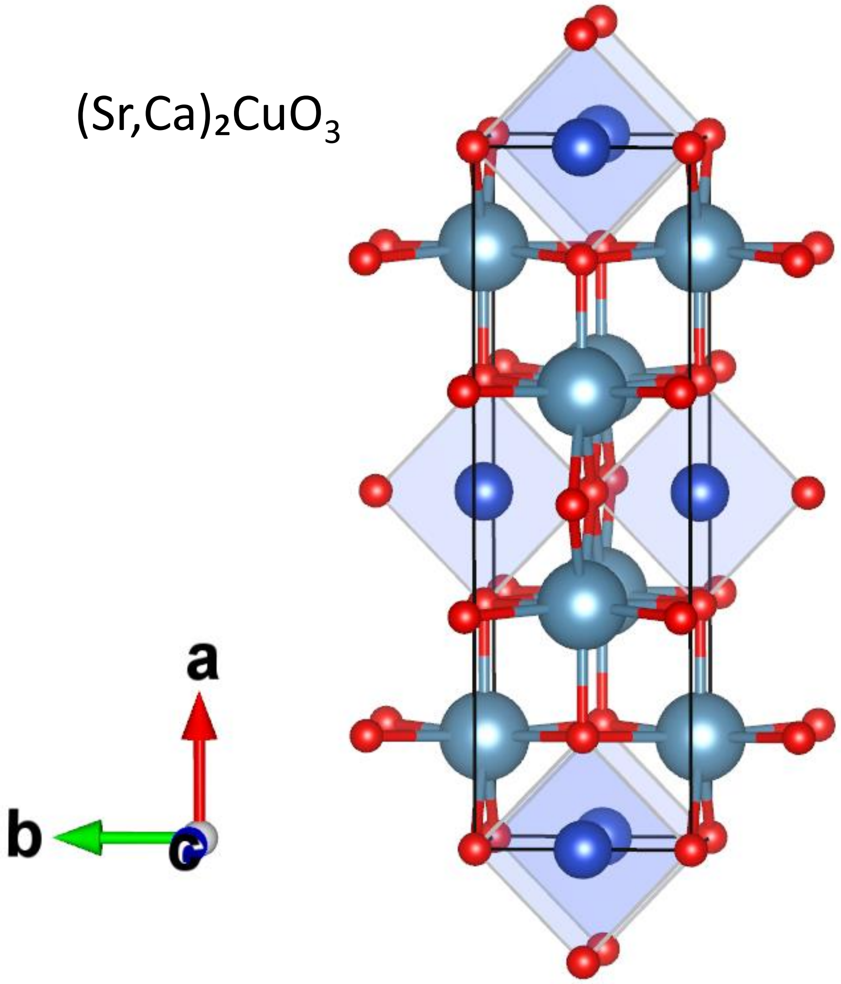
D. Bounoua, Phd thesis (2017)



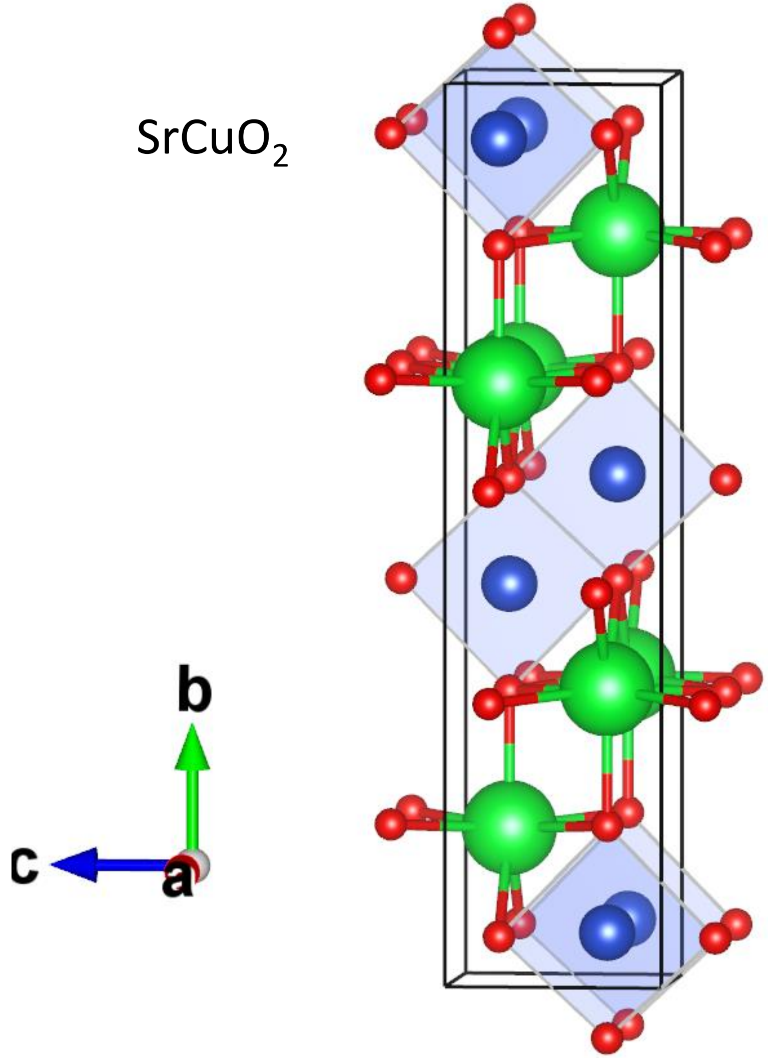
Heat is evacuated along 1D highways !!

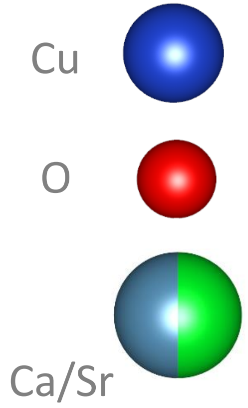
Spin chains cuprates $SrCuO_2$, Sr_2CuO_3, Ca_2CuO_3

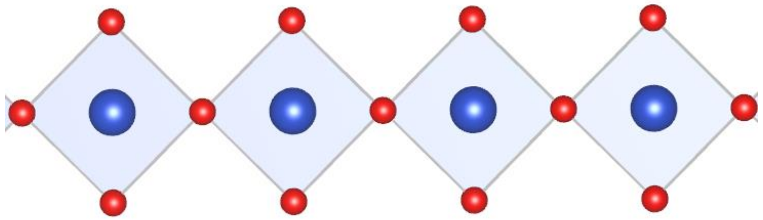
$(Sr,Ca)_2CuO_3$



$SrCuO_2$







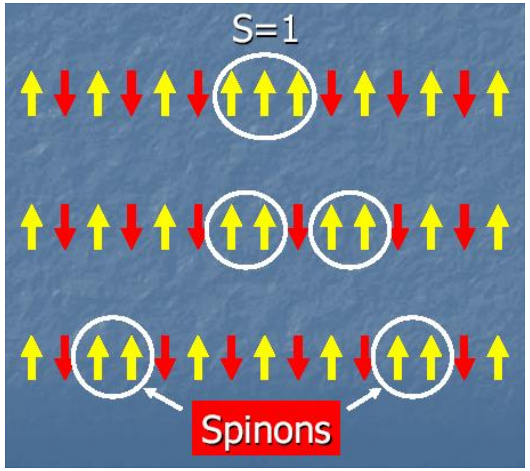
Regular or zig-zag Chains

Spinons in 1D chains

$$H_{\mathrm{XXZ}} = J \sum_i \epsilon(S_i^x S_{i+1}^x + S_i^y S_{i+1}^y) + S_i^z S_{i+1}^z$$

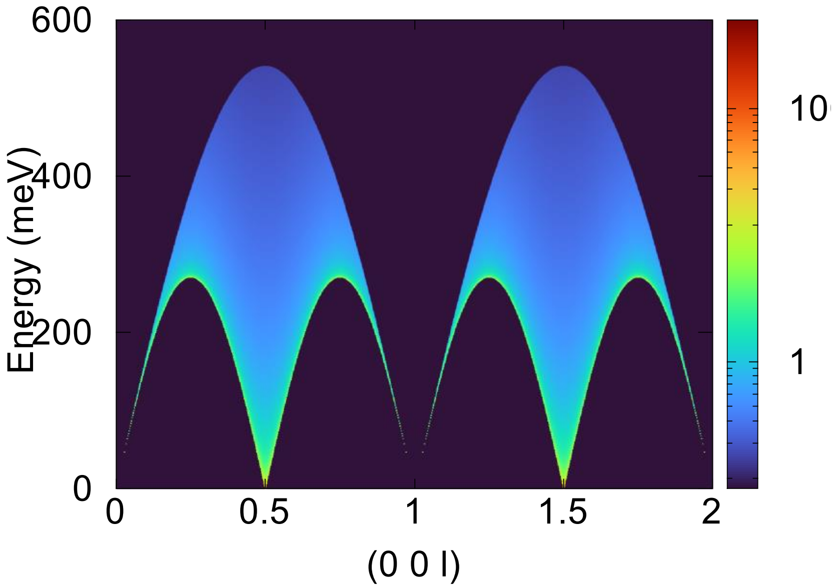
Spinons are very peculiar spin ½ excitations typical of 1D XXZ spin chains

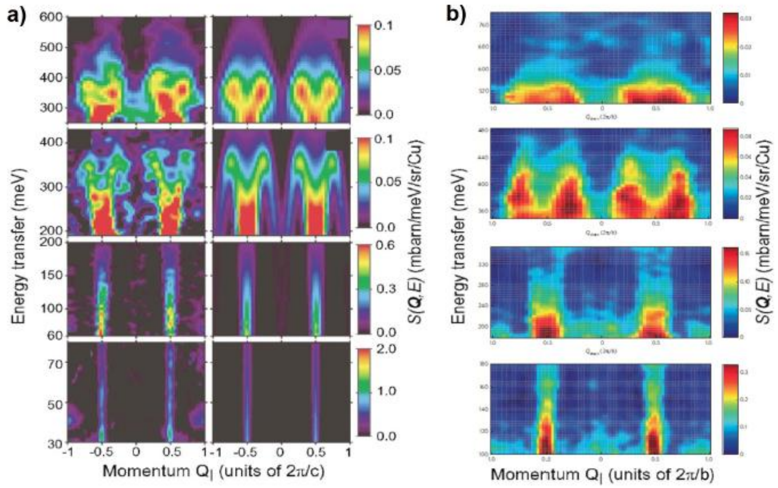
≈ domain walls



Courtesy F. Mila

Visible by means of neutron scattering (2-spinons continuum)





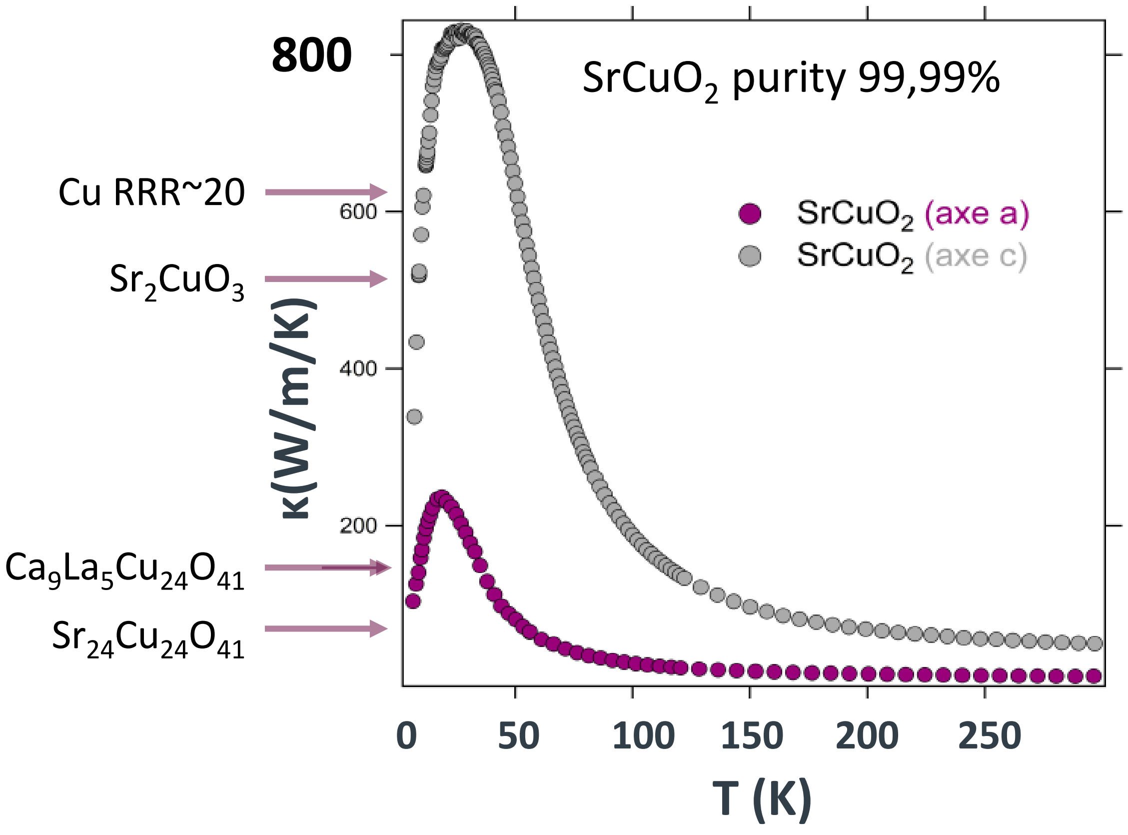
Zaliznyak et al.,Phys. Rev. Lett. 93, 087202 (2004)

Huge exchange coupling :
J ≈ 200 meV ≈ 2000 K

Spinons in 1D chains

Hlubeck et al., J. Stat. Mech. : Theory and Experiment (2012)

$\kappa_{\parallel}$ and $\kappa_{\perp}$



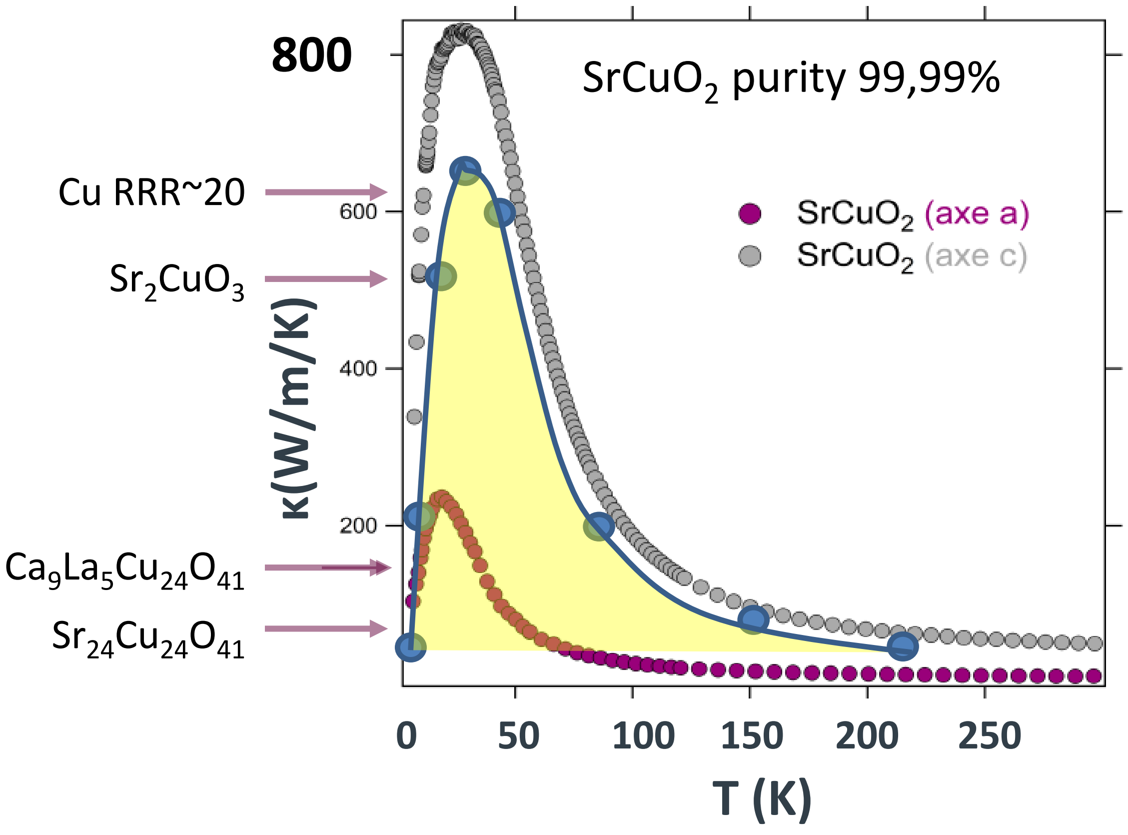
- Thermal conductivity is anisotropic
- Large maximum (HUGE J)
- Strongly decreases with increasing temperature

Hlubeck et al., J. Stat. Mech. : Theory and Experiment (2012)

$$\mathcal{H} = \sum_{\langle ij \rangle} J(\mathbf{r}_i - \mathbf{r}_j)\mathbf{S}_i \cdot \mathbf{S}_j,$$

Weak coupling — Strong coupling → g

$$\kappa_{mag} = \kappa_{\parallel} - \kappa_{\perp}$$



Two models, two different limits

Spinons in 1D chains

Hlubeck et al., J. Stat. Mech. : Theory and Experiment (2012)

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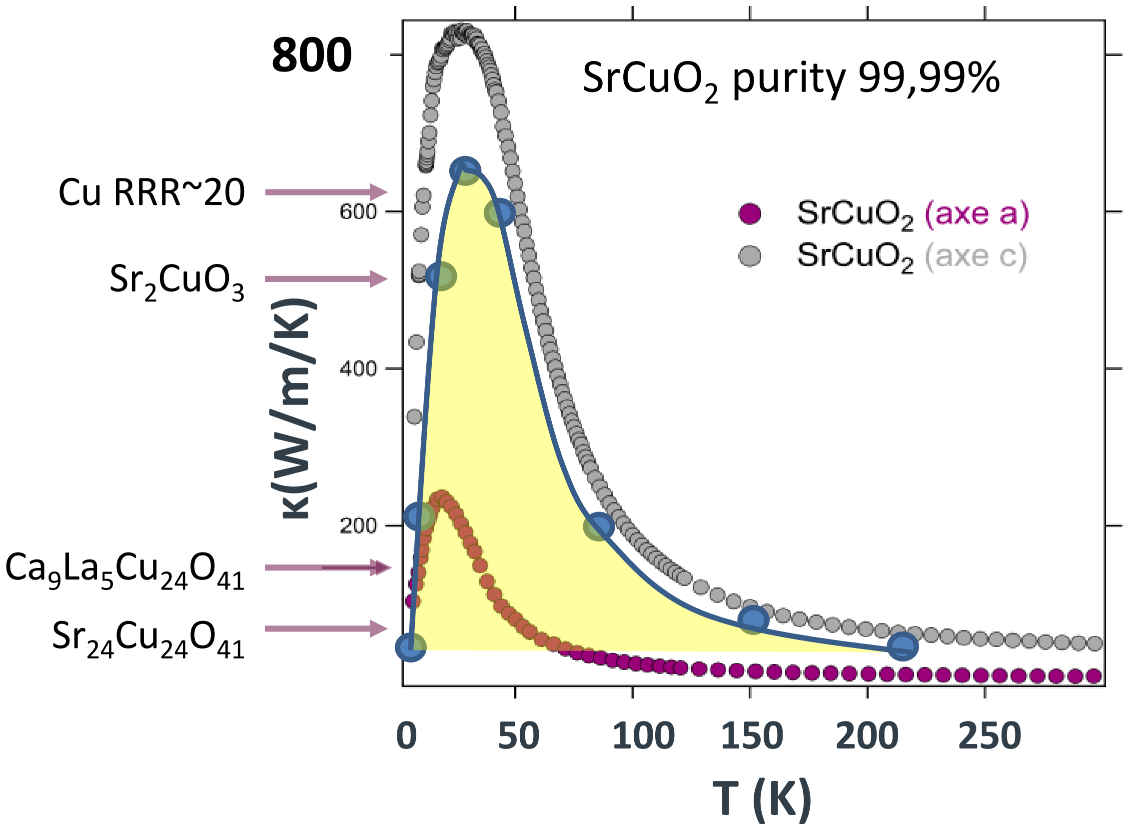
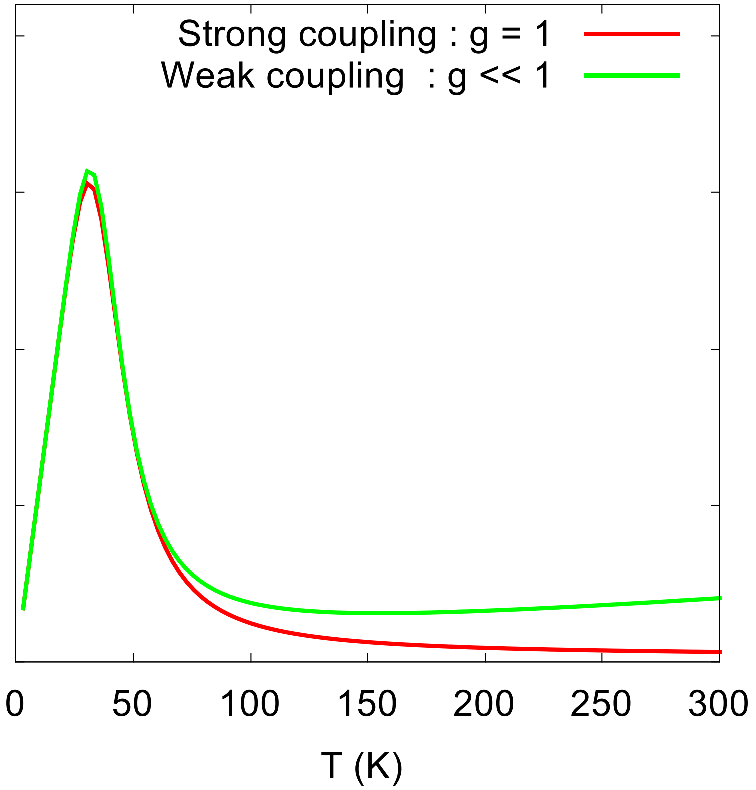




How to distinguish ?

Any effect on phonons modes ?

Chernyshev et al., PRL 116 017204 (2016)
Chen et al, PRL 122185901 (2019)

I. Sample synthesis & characterization (ICMMO)

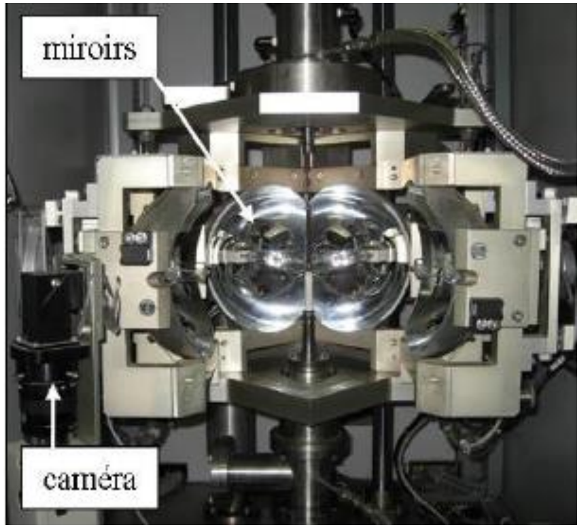
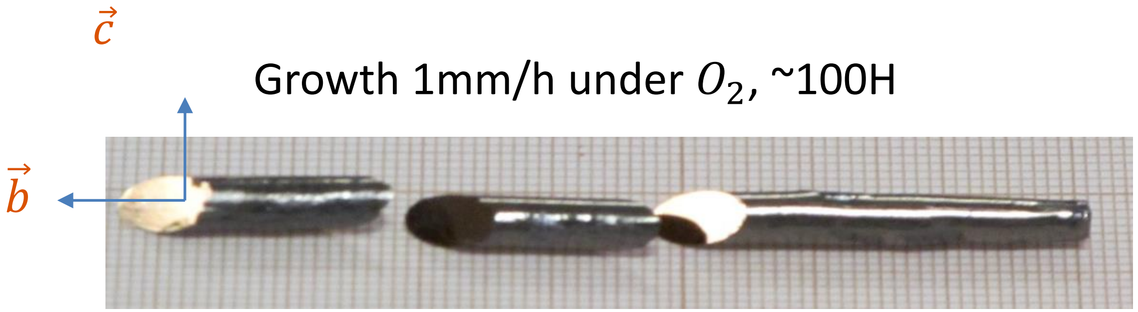


Image furnace @ ICMMO

Growth 1mm/h under O_2, ~100H

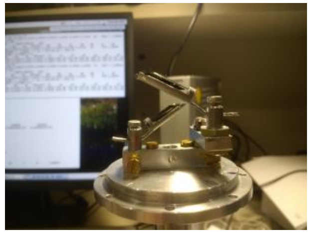


Sr_2CuO_3 Single crystals

Growth 0,7mm/h under O_2, ~140H

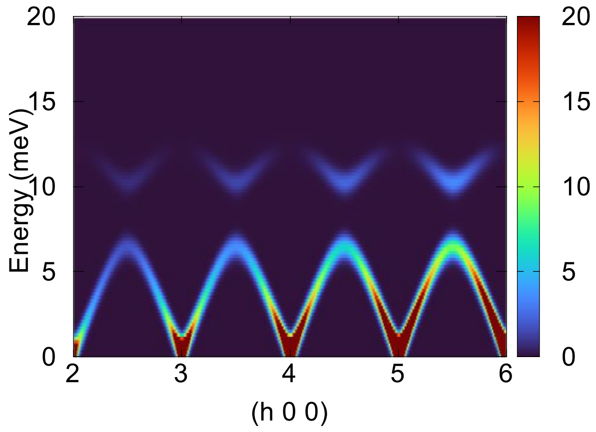
Ca_2CuO_3Single crystals

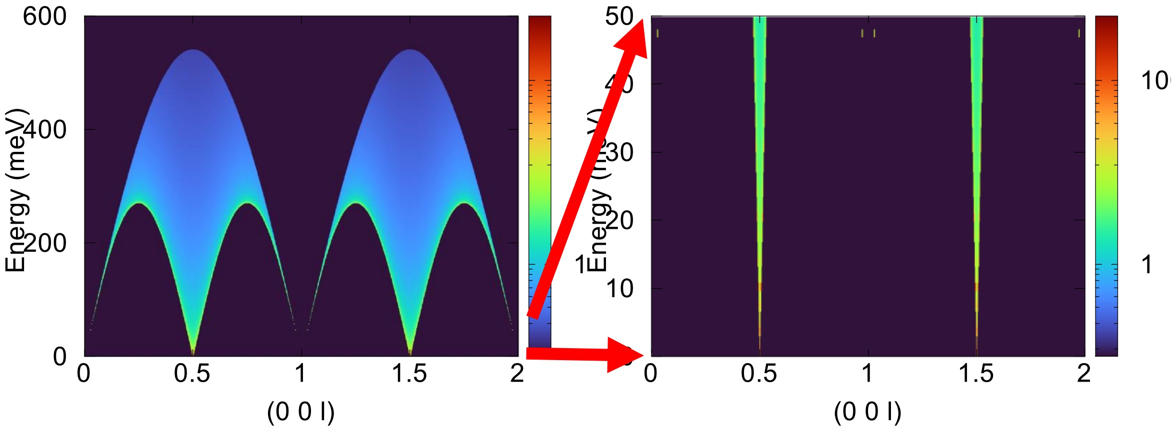
II. Inelastic neutron scattering (ILL & LLB-Orphée)

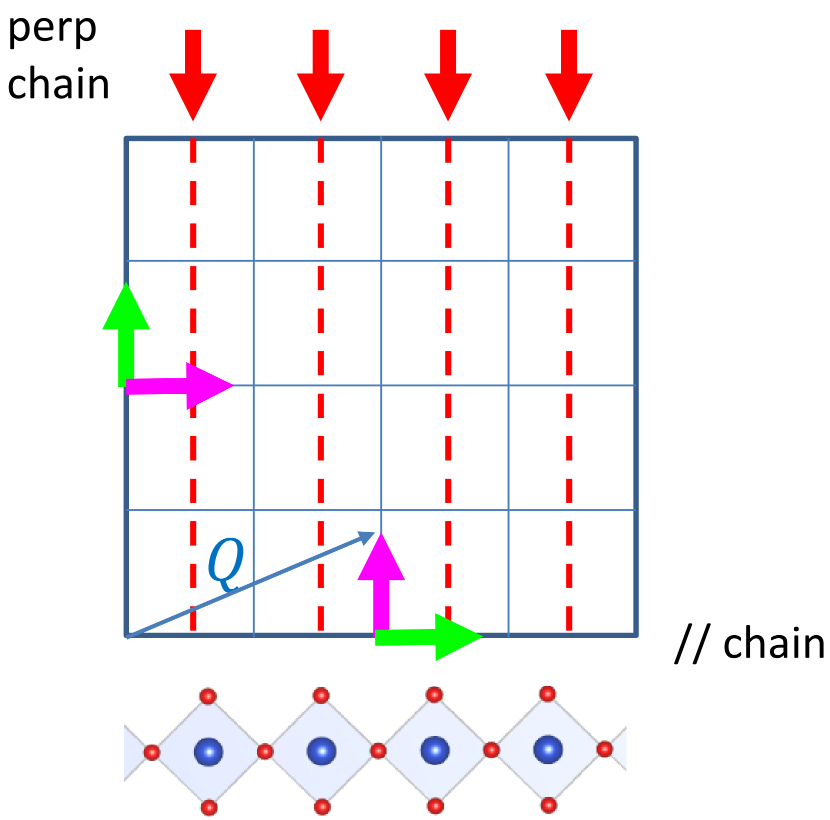


Map out (Q,ω) space to reveal phonons ans spinons. Keep in mind that spinons are :

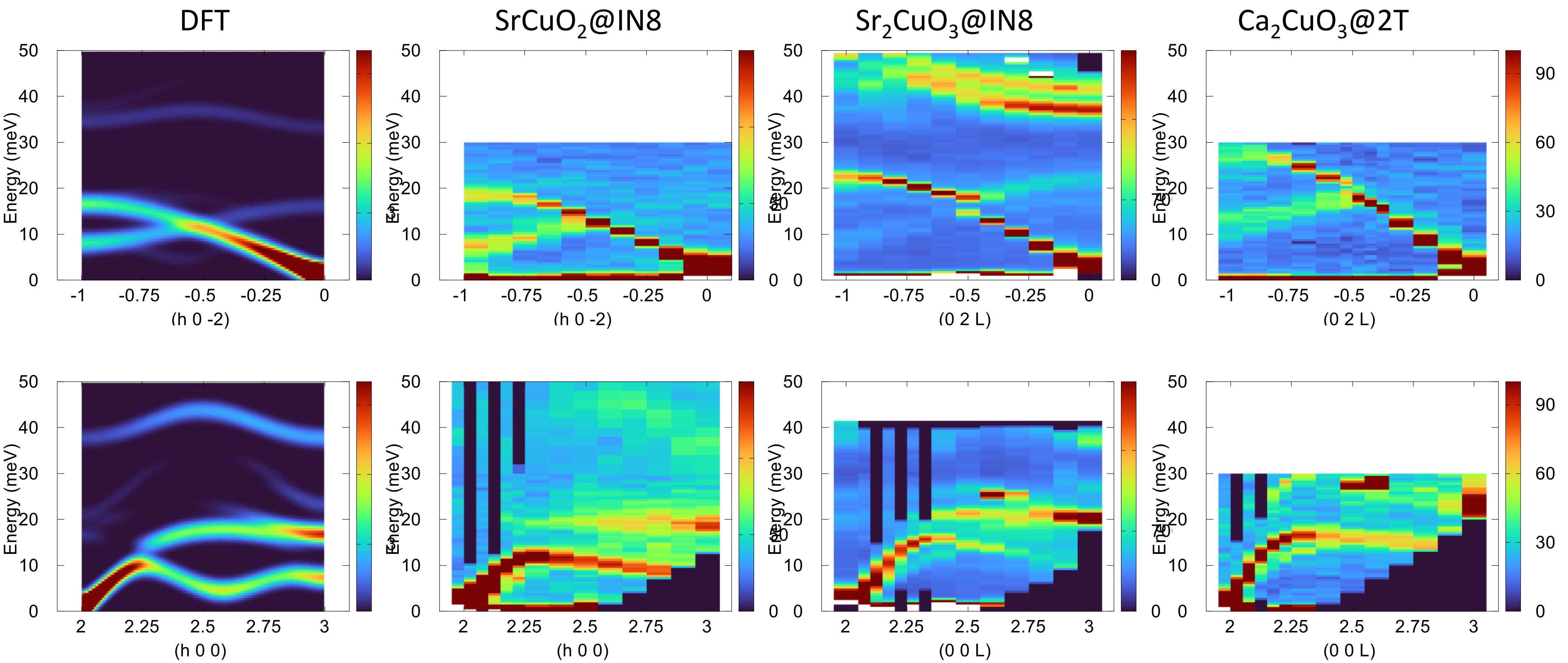
- 1D
- And much faster than phonons $J \gg \omega_{ph}$





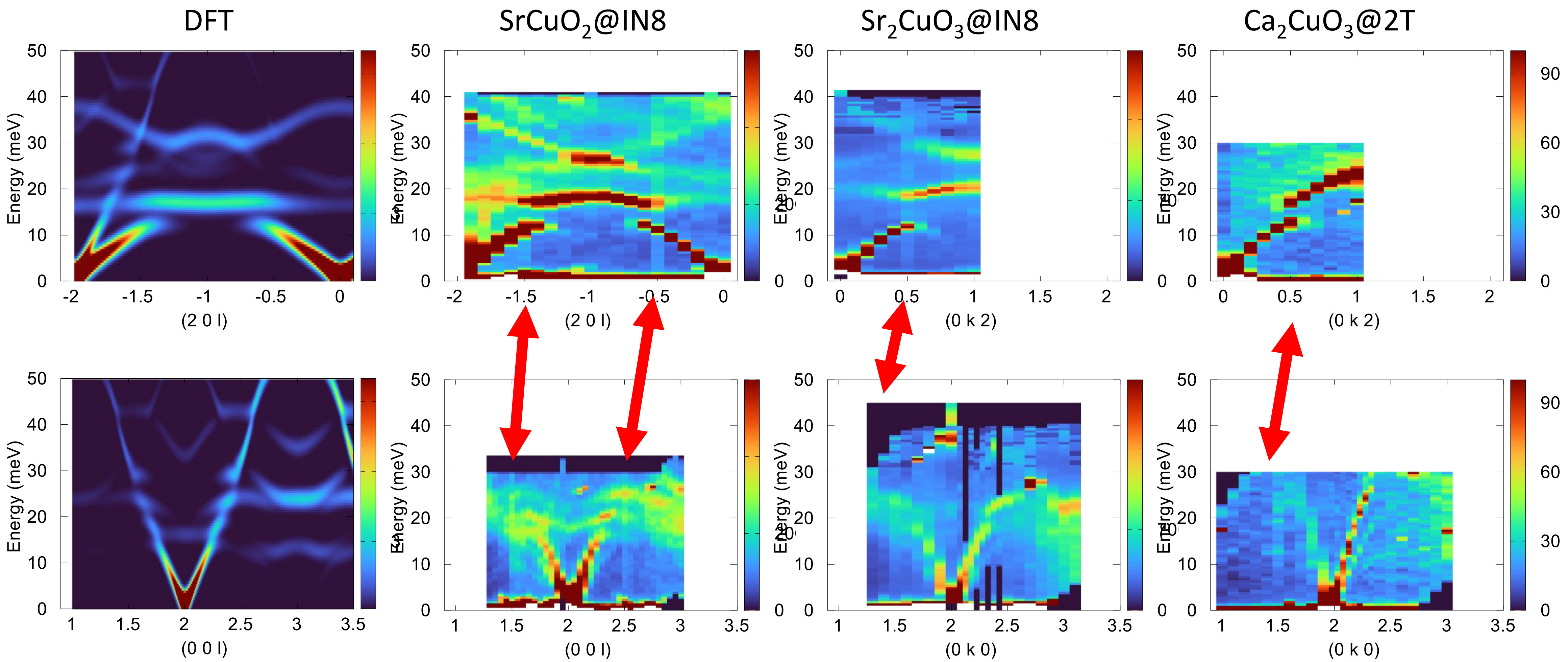


Survey of phonons propagating perp to the chains



Larger # of avoided crossings + « Soft » modes, all predicted by DFT

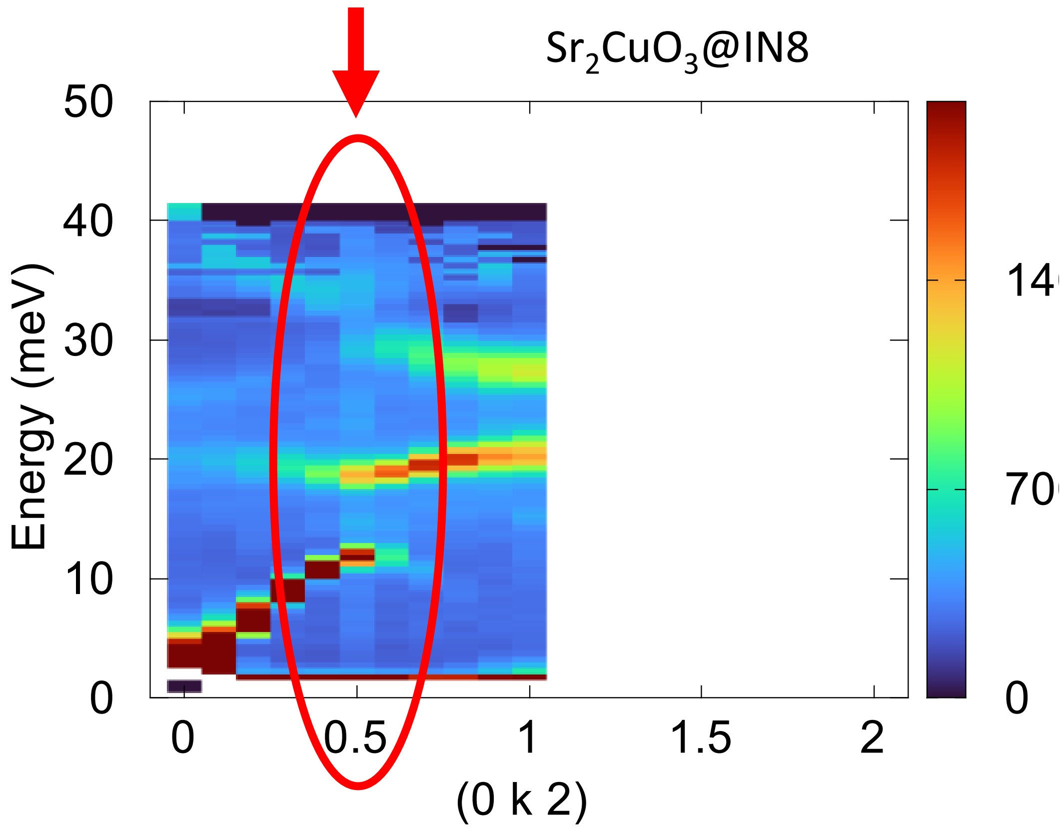
Survey of phonons propagating along the chains

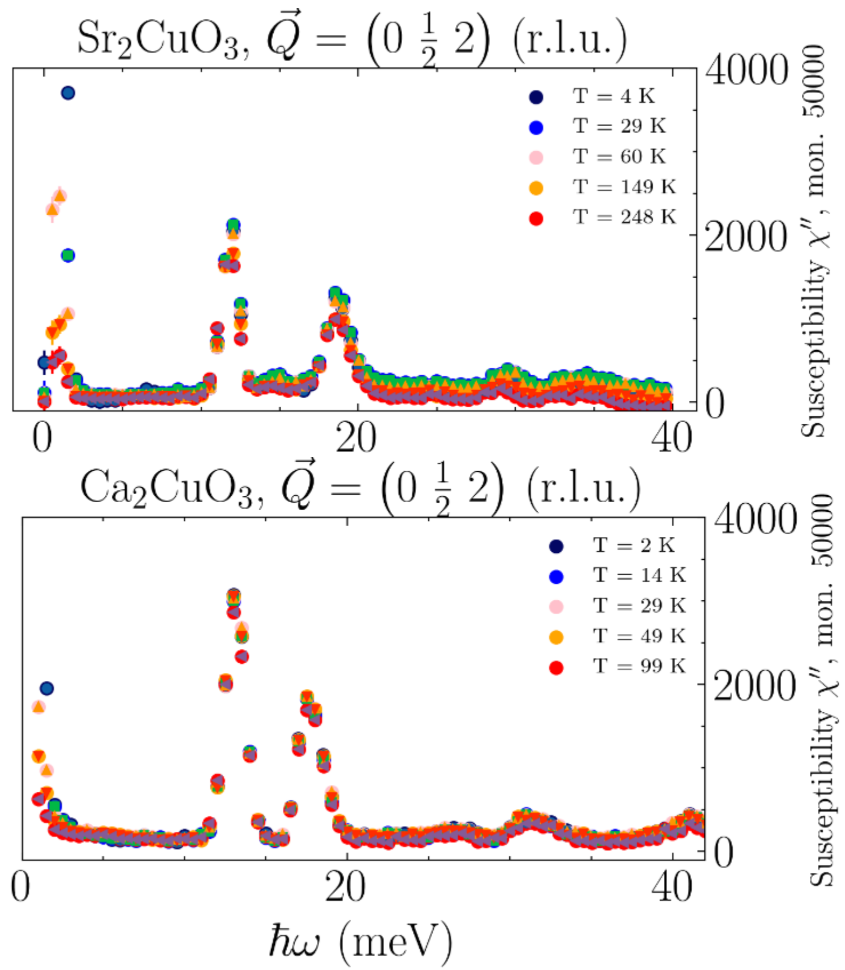


Larger # of avoided crossings + « Soft » modes, all predicted by DFT

When phonon cross the 2-spinons continuum :

Any significant broadening? NO …
Any significant evolution with increasing temperature ? NO …





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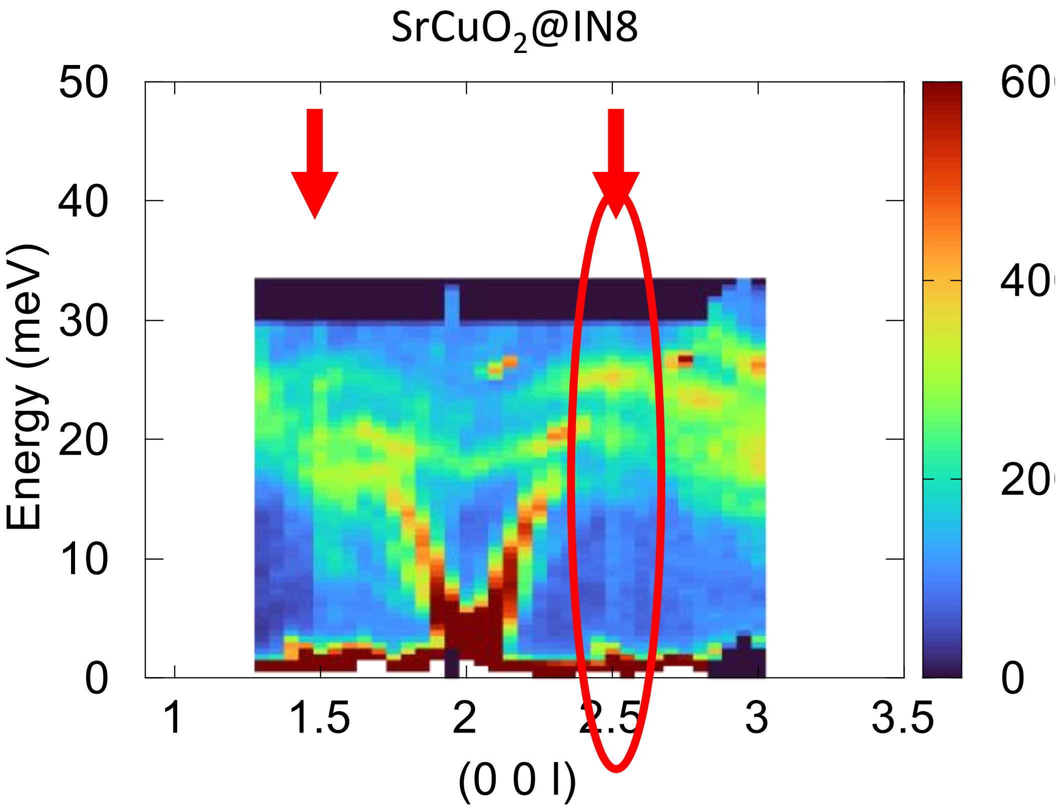
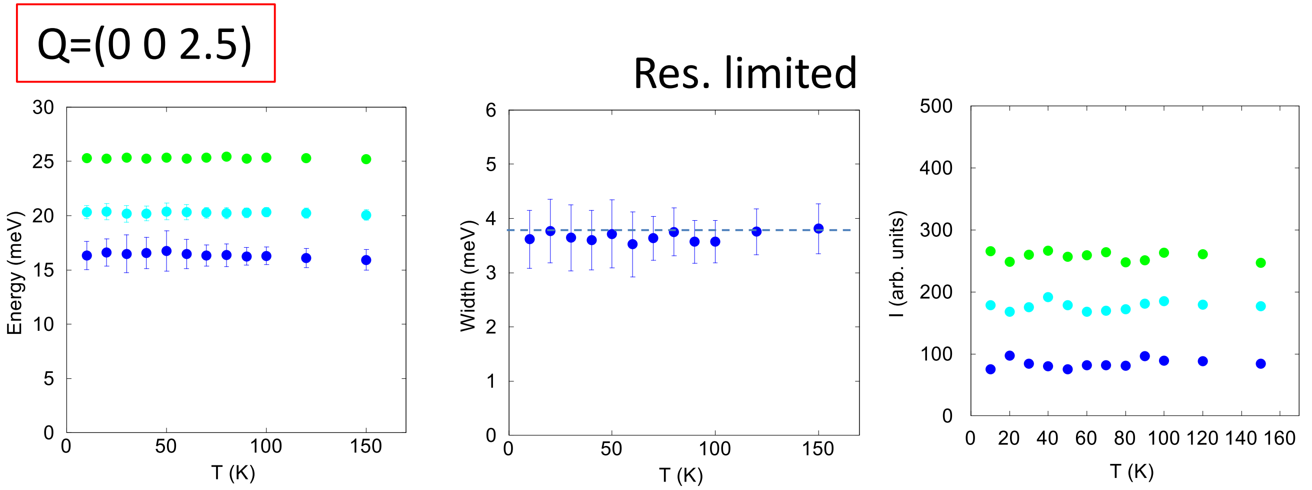
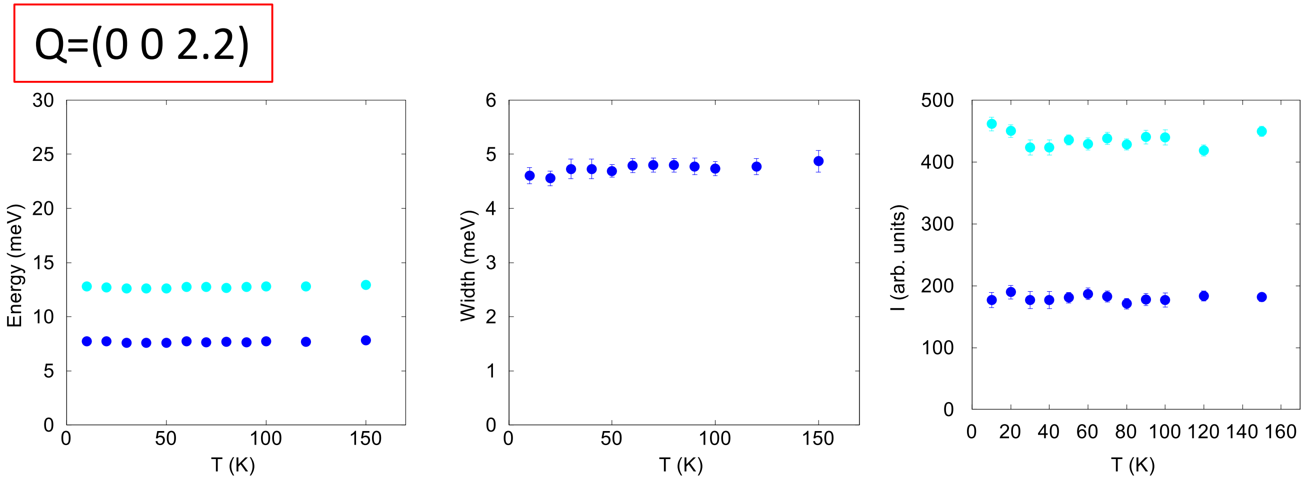






Conclusions

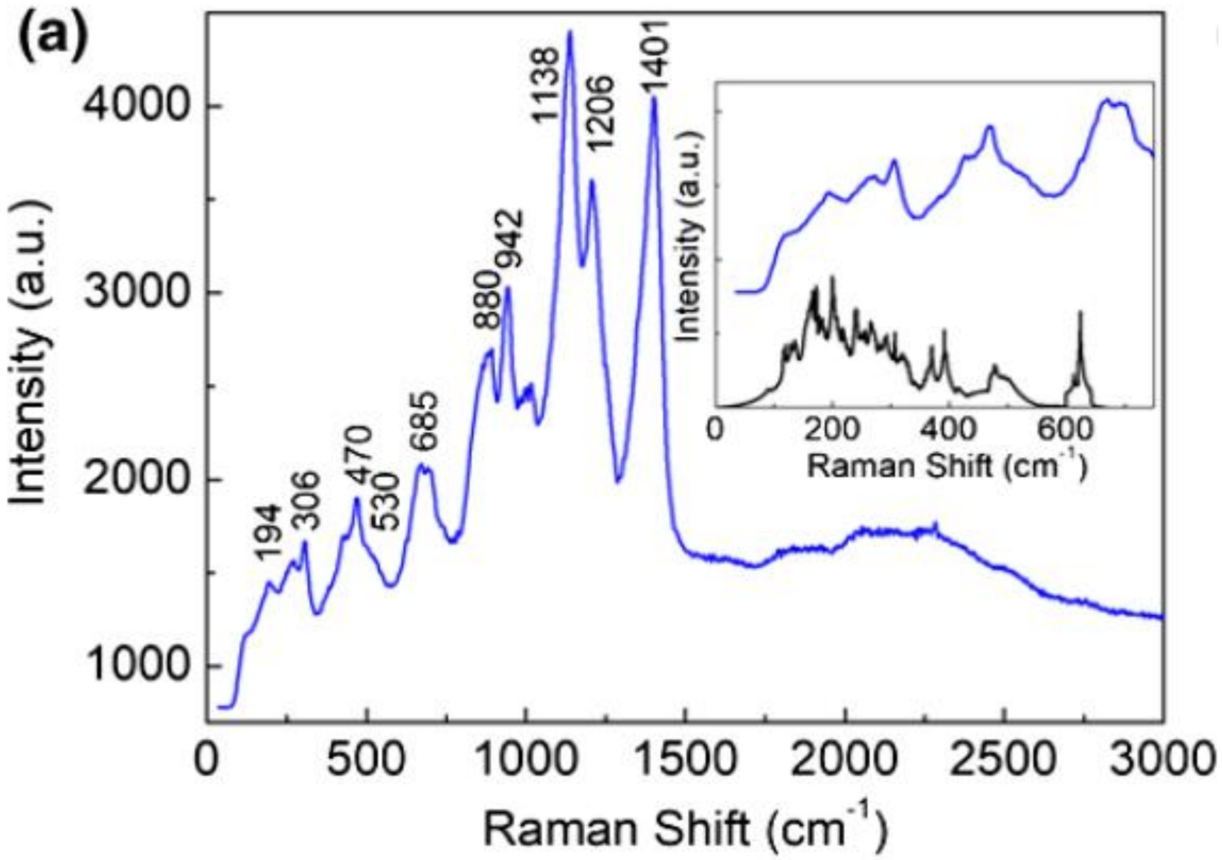
- Complicated lattice dynamics, wtih avoided crossings + low energy modes, indicating that the structure is soft in certain directions.

- These features are essentially predicted by DFT.

- NO remarkable anomalies as a fonction of temperature (within experimental resolution), suggesting that the spin-phonon coupling is WEAK.

Perspectives

- Improve energy resolution, theoretical background for phonon linewidth ?
- Going beyond the simple expression $\kappa_{mag} = \kappa_{\parallel} - \kappa_{\perp}$



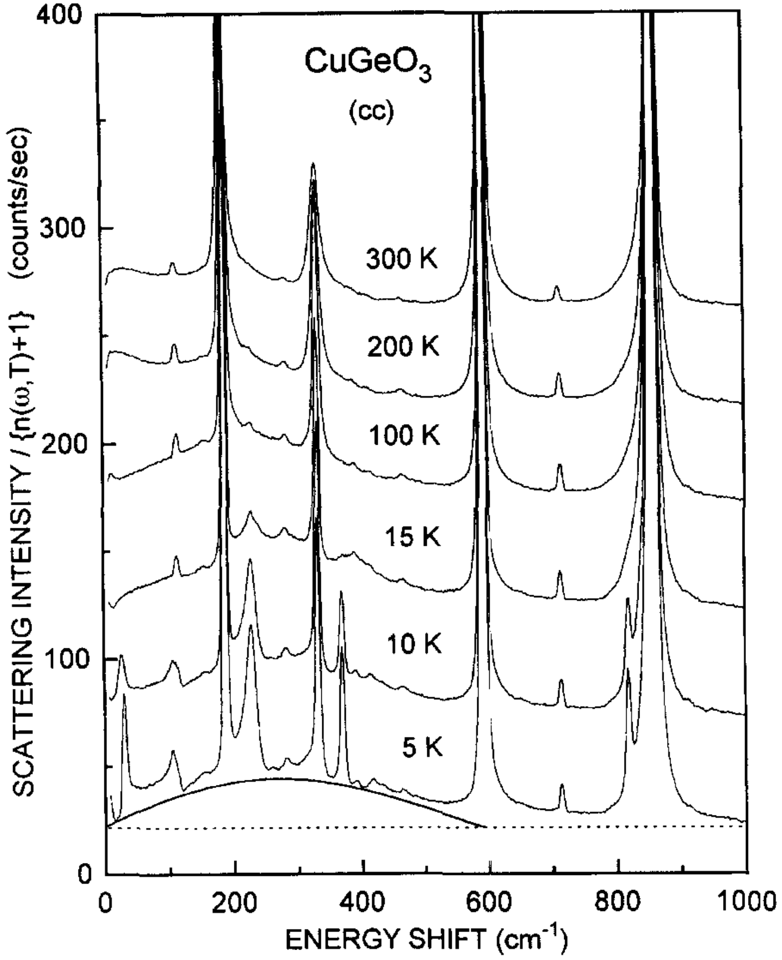
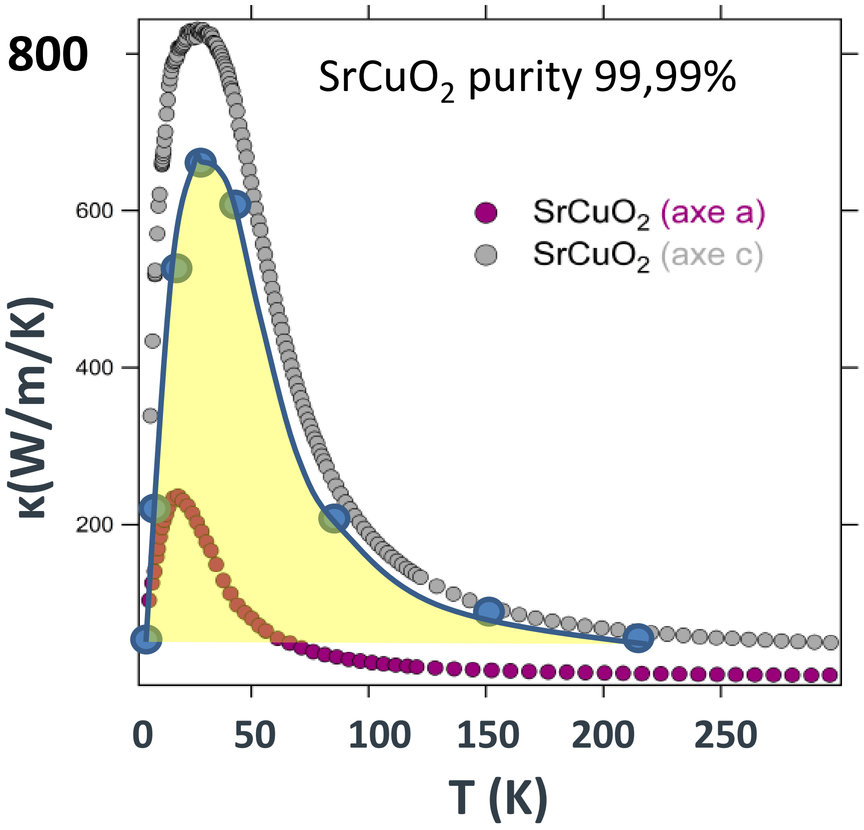


Fig. 3. Temperature dependence of low energy parts of (*cc*) Raman spectra of $CuGeO_3$.

Magnetic degrees of freedom in spin chains

$$\mathcal{H} = \sum_{\langle ij \rangle} J(\mathbf{r}_i - \mathbf{r}_j)\mathbf{S}_i \cdot \mathbf{S}_j,$$



Hlubeck et al., J. Stat. Mech. : Theory and Experiment (2012)

$$\kappa \approx \frac{\pi}{3}\, n_s\, k_B \frac{k_B T}{\hbar}\, \ell$$

Spinons scatter on optical phonons (essentially at rest)

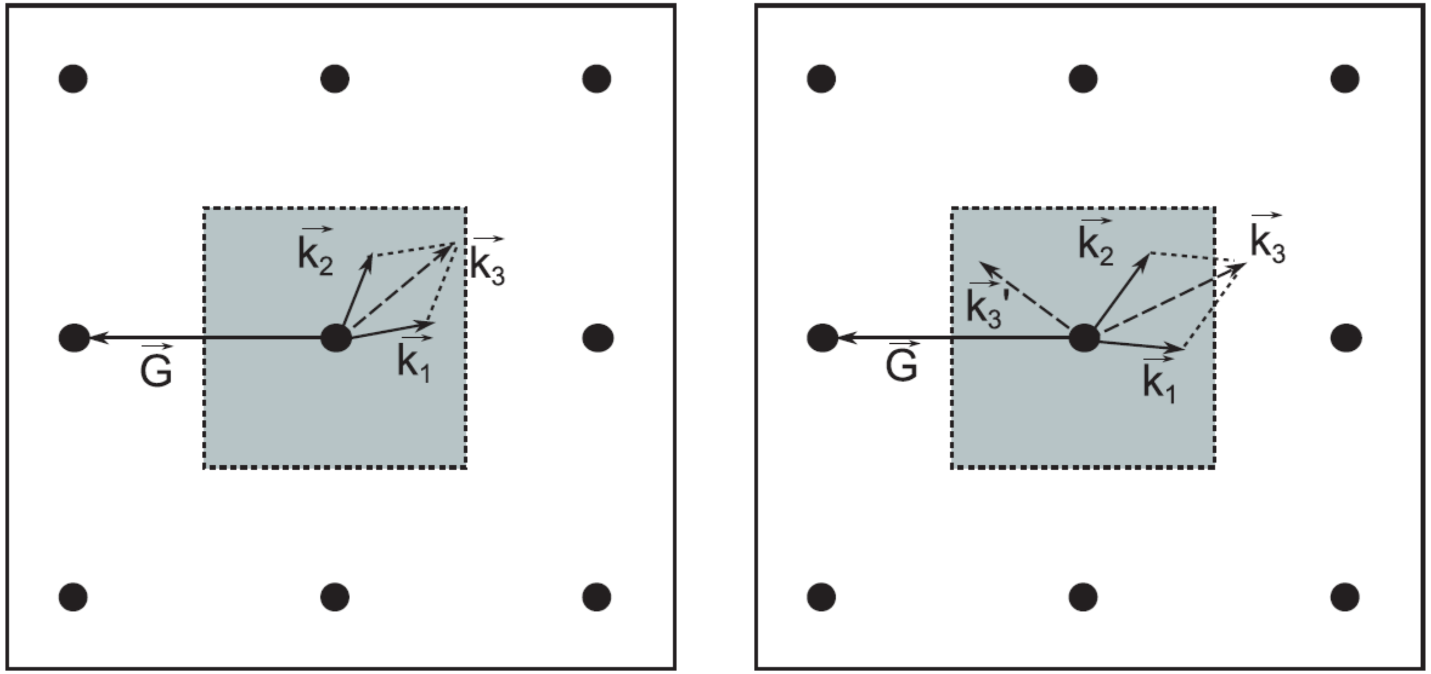
$$\frac{1}{\ell} \approx \frac{1}{\ell_d} + \frac{2g^2}{a}\frac{J}{T}\frac{1}{\sinh \omega_o/T}$$

Heuristic spinon mean free path
Spinons scatter on acoustic phonons

$$\frac{1}{\ell} \approx \frac{1}{\ell_d} + \frac{g^2}{a}\frac{T}{J}\, e^{-T^*/T}$$

Weak coupling → Strong coupling g

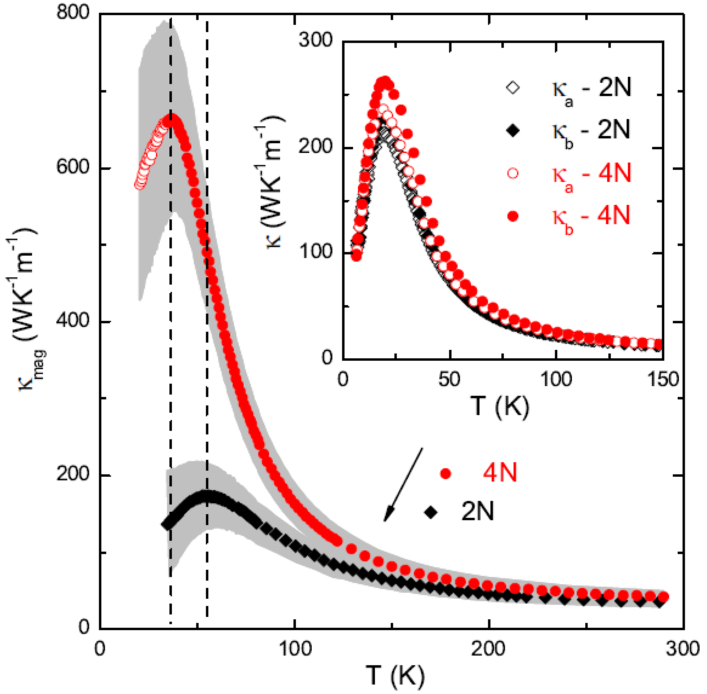
Spin Peierls (dimerization as in CuGeO3)



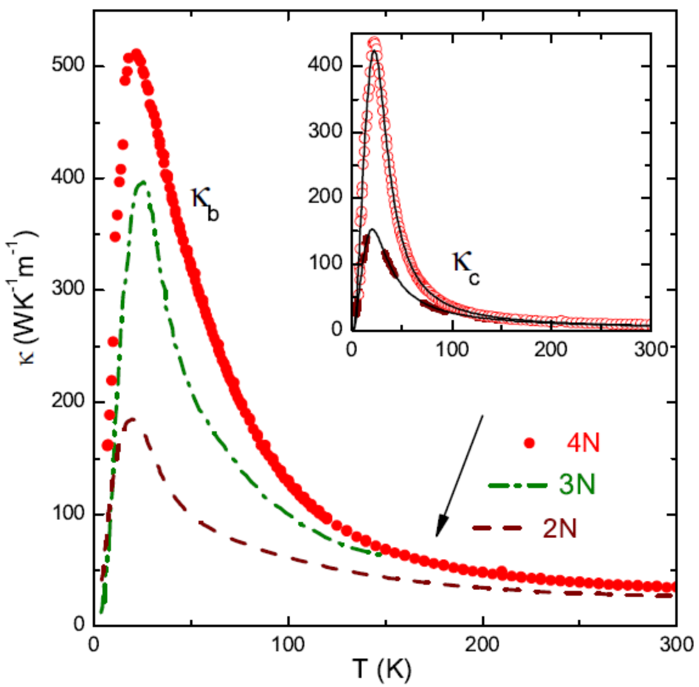
$$\hbar\omega_1 + \hbar\omega_2 = \hbar\omega_3$$

$$\vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{G}$$

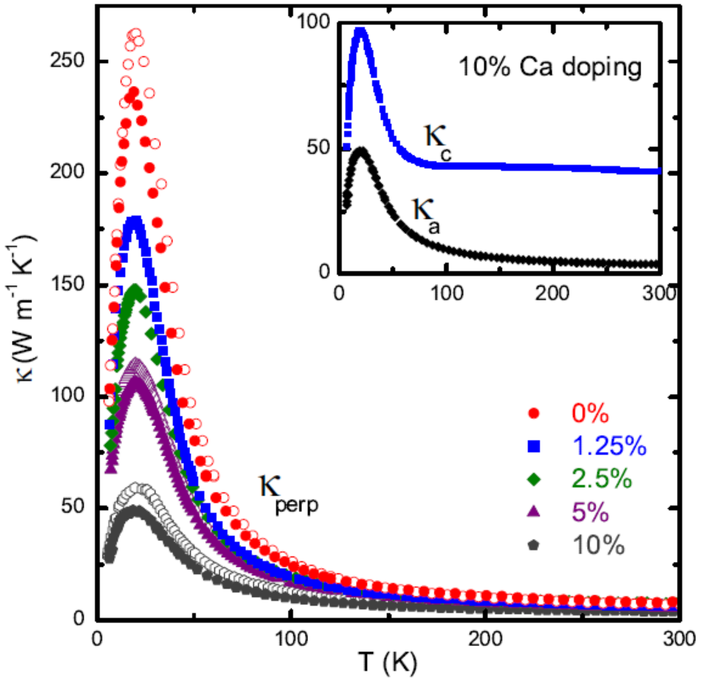
$SrCuO_2$

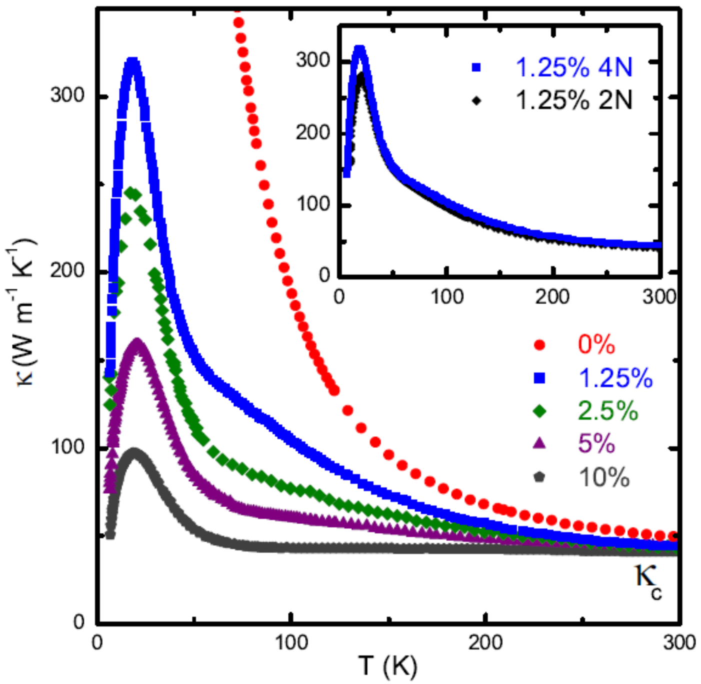


Sr_2CuO_3

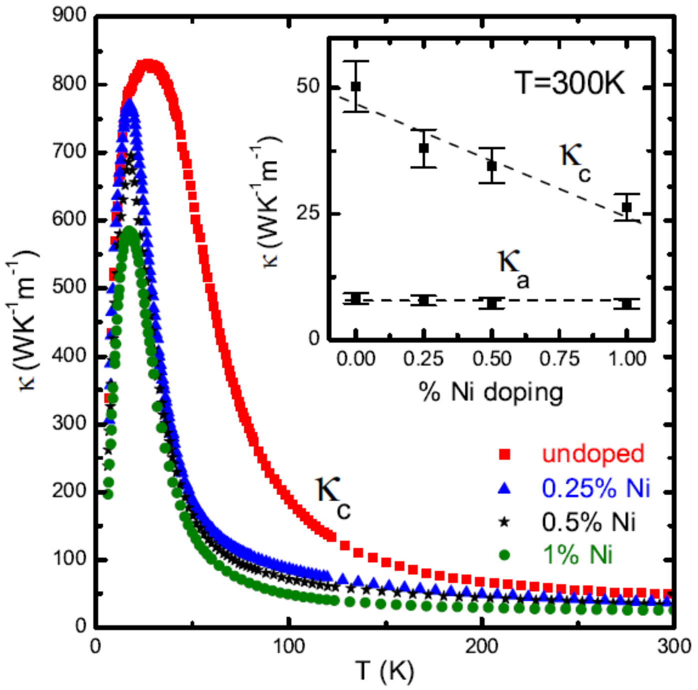


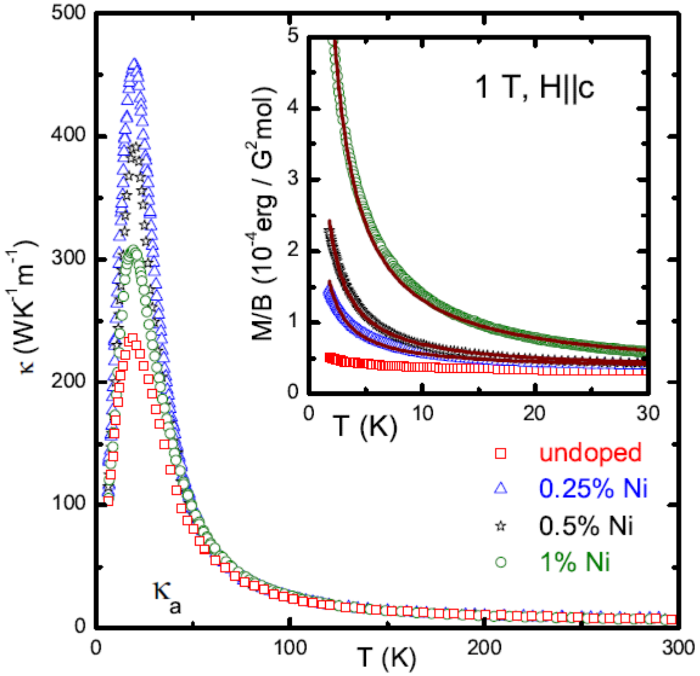
$Sr_{1-x}Ca_xCuO_2$



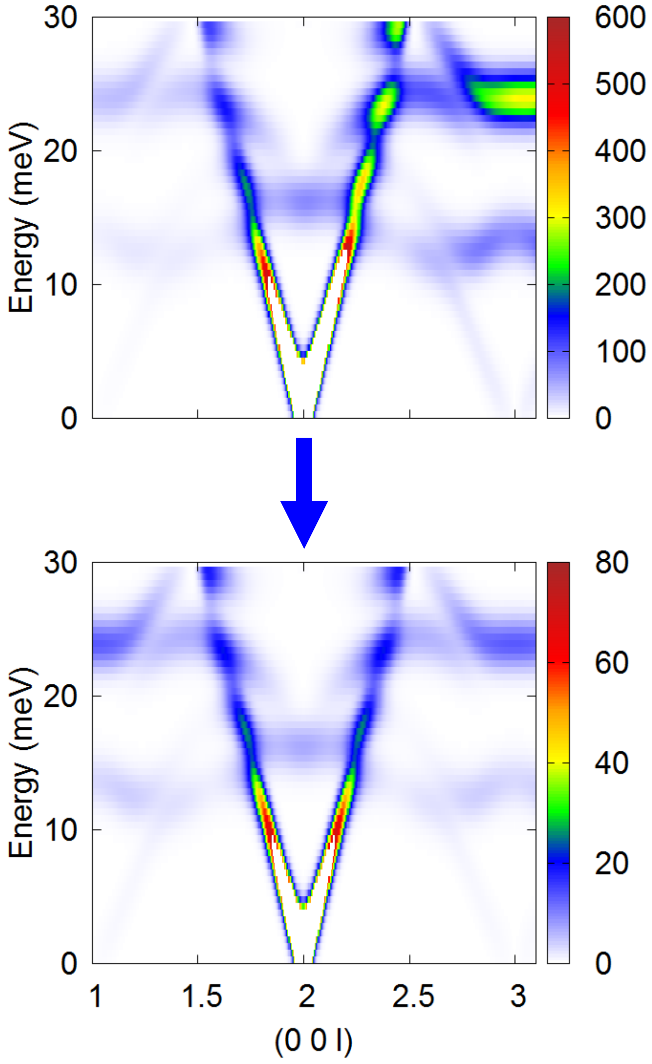


SrCuO$_2$ doped with Ni (S=1 impurity)





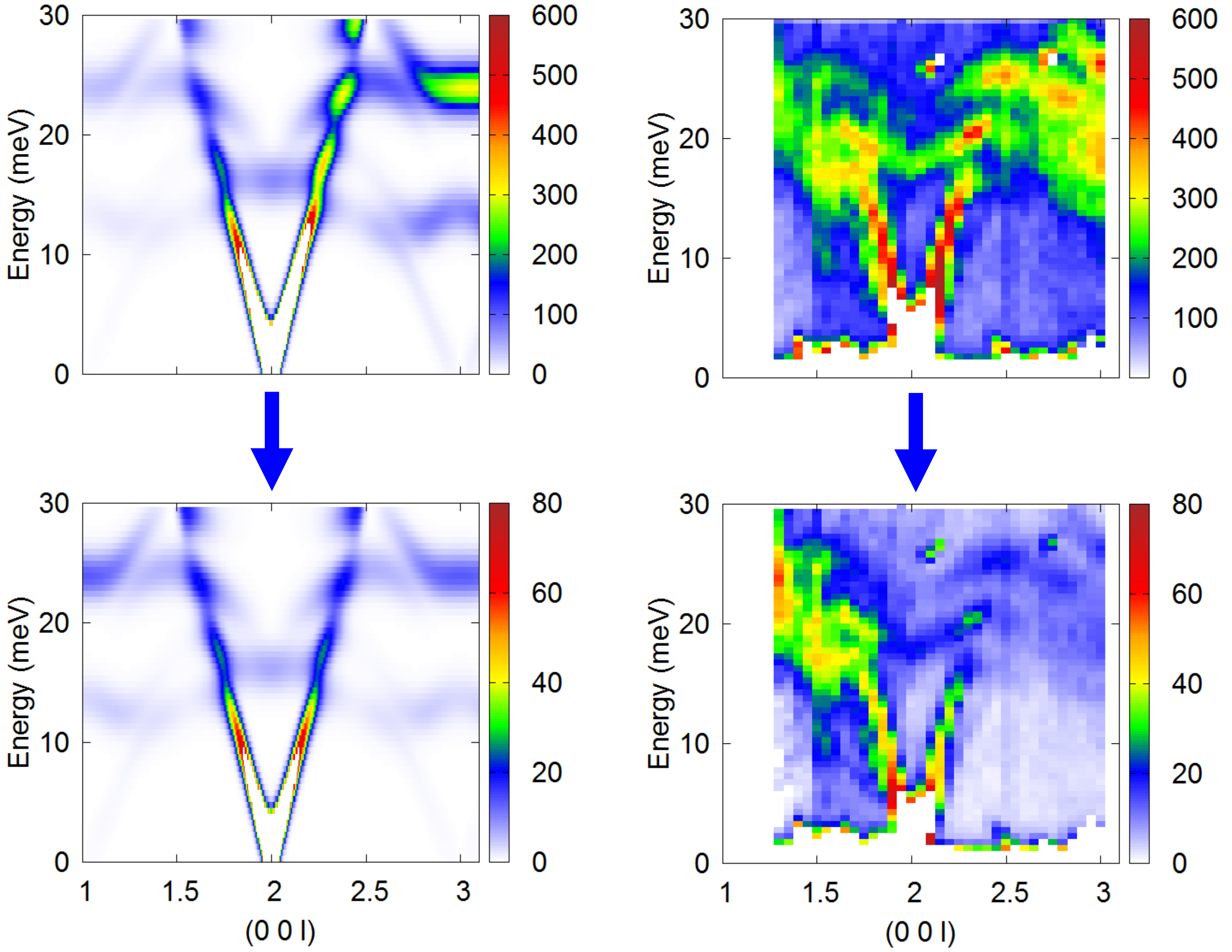
Any effect ?



Correct the data for classical Q^2 dependence and for the Debye-Waller :

$$\chi'' = \frac{I}{Q^2 \, e^{-W} [1 + n(\omega)]}$$

Any effect ?



The spectral weight is anomalously strong at low Q.

Is it related to the magnetic fom factor ?

No definitive conclusion