Experimental report

15/02/2020

Proposal: **7-01-481**

Council: 10/2018

Title: Lattice dynamics of the low-dimensional Sr2CuO3

Research area: Physics

This proposal is a continuation of 4-04-481

Main proposer: **David MSIKA**

Experimental team: Dalila BOUNOUA
David MSIKA
Sylvain PETIT
Francoise DAMAY

Local contacts: Alexandre IVANOV
Andrea PIOVANO

Samples: Sr2CuO3

Instrument	Requested days	Allocated days	From	To
IN3	1	1	18/09/2019	19/09/2019
IN8	10	5	19/09/2019	24/09/2019

Abstract:

The Mott insulating spin chains system Sr2CuO3 was shown to exhibit a highly anisotropic magnetic heat conduction, which peaks at around 150 WK-1m-1 at 50K. Indeed, heat in Sr2CuO3 is conveyed by phonon and fractional S= 1/2 magnetic excitations, namely, spinons. Heat conduction by spinons occurs only along the spin chains direction and has been proven to be ballistic. The interplay between magnetic and lattice quasi-particles and chemical or structural defects during the heat transport process gives rise to various two-quasi-particles and quasi-particle-defects scattering mechanisms that govern the resulting heat conduction properties. A deepunderstanding of these different interaction paths is thus fundamental in order to have a clear insight of the heat transport process. However, such a microscopic description of the system is still lacking. We propose to measure the phonon spectra of phonon modes propagating along the spin chains and perpendicular to the spin chains in Sr2CuO3 on IN8 TAS .

Inelastic neutron scattering on Sr_2CuO_3 at IN8

David MSIKA* Dalila BOUNOUA† Romuald SAINT-MARTIN* Sylvain PETIT†
Francoise DAMAY† Loreynne PINSARD-GAUDART*

1 Motivations & measurement setup

Sr_2CuO_3 is a cuprate spin chain compound, with tremendous AF superexchange $J \approx$2000 K mediated by 180° Cu−O−Cu chemical bond. The inter-chain coupling being much smaller than J, the system is effectively a 1D spin chain while being a volumic material, suitable for neutron studies. In 1D spin chains, specific magnetic excitations exist: the spinons. We believe that spinon carry ballistically most of the thermal energy, until a scattering mechanism is thermally activated, such as spinon-phonon scattering. To show the existence of such coupling, we fist study the phonon dispersion in the material, and then look for a temperature effect on the phonon spectrum. A phonon softening with temperature could indicate the presence of such spin-lattice coupling, which would explain the decay of those exceptional thermodynamic properties. This report is a short summary of the experiment.

The sample was mounted in an Orange cryostat, to go down to 4 K, and the sample stage was wrapped in aluminum foil. The monochromator/analyser couple used was Si(111) on PG(002) (mosaic resp. of ≈ 0.4° and 0.30°), with a pyrolitic graphite filter to remove parasitic harmonics. We worked at constant $k_F = 2.662\,\text{Å}^{-1}$ during the experiment. No extra collimation was used. This setup allows a minimal energy resolution of 0.8 meV (on Vanadium), and allows a large energy-momentum region to be explored. We worked in phonon creation mode, and thus measured the dispersion at 10 K.

Γ_{020} ω (meV)	σ	Γ_{002} ω (meV)	σ
15.8	2.2	19.8	3.4
21.9	2.9	25.4	1.2
37.0	1.2	37.9	2.2
41.9	2.2	44.2	1.0

(a) Fitted modes around two reciprocal space nodes.

ω (cm^{-1})	ω (meV)	polar	Raman	IR
reset 168	20.8	E//c	○	●
181	22.4	E//b	○	●
205	25.4	z(xx)z	●	○
212	26.3	E//c	○	●
308	38.2	E//c	○	●
343	42.5	E//b	○	●

(b) Bibliography comparison with Lee (IR) and Misochko (Raman).

Y. S. Lee et al., Phys. Rev. B, vol. 62 (2000)
O. V. Misochko et al., Phys. Rev. B, vol. 53 (1996)

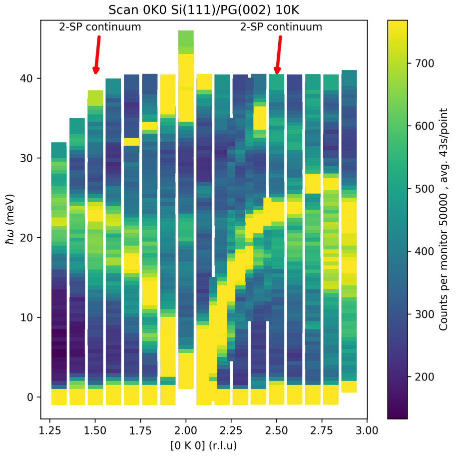
We begin by comparing the phonon mode energy at Γ on Table 1a to the literature on Table 1b, and infer their polarization.

2 Longitudinal phonon dispersion

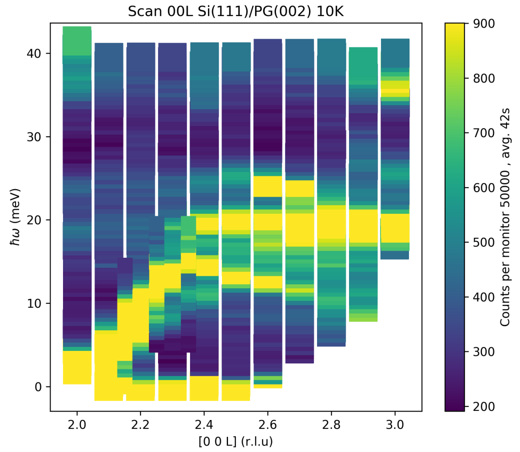
0K0 On Figure 1a is displayed the longitudinal phonon dispersion along the spin chain axis b^*. The result is similar in spirit with the compound $SrCuO_2Zn_{1\%}$ we measured before. A stiff acoustic branch meets an optical mode O1 at 0, 2.35, 0 (r.l.u.) and then disappears; it's intensity seems transferred to the optical mode. The two modes are closest at 0, 2.35, 0 (r.l.u.) with energies 17.7(13) meV and 21.7(15) meV. On this figure the 2-spinon continuum can be guessed at 0, 1.5, 0 and barely visible at 0, 2.5, 0. Its expected position is signaled by red arrows on Figure 1a.

00L The longitudinal phonon modes dispersion, propagating ⊥ to the spin chain, is reproduced on Figure 1b. The acoustic branch is softer than along the spin-chain axis. The acoustic branch peaks at (15.9(8) meV, $00Q_L$=2.4) and then softens to ≈ 10 meV at the zone border. At the point where the longitudinal acoustic branch brends down, a flat optical mode at 21.0(10) meV brightens, and remain very intense up to the zone border. Lastly a descending optical branch can be guessed at (37.5(21) meV, $00Q_L = 3$ r.l.u.). This mode brightens at the position corresponding with the intersection between the flat optical branch, with the acoustic phonon slope at the origin.

*SP2M-ICMMO UMR 8182 ,ORSAY FR
†CNCE/LLB-LAB LEON BRILLOUIN, SACLAY ,GIF-SUR-YVETTE FR

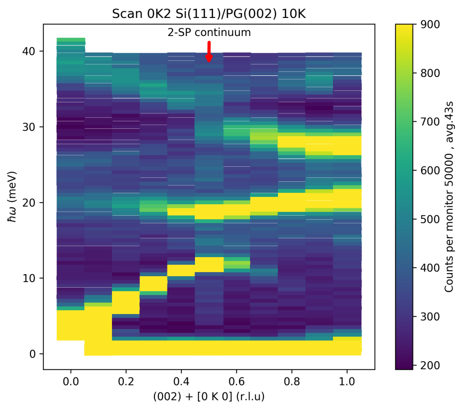


(a) Phonon dispersion relation along b^* at 10K.

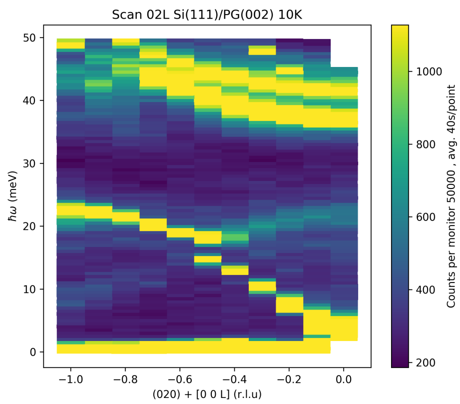


(b) Phonon dispersion relation along c^* at 10K.

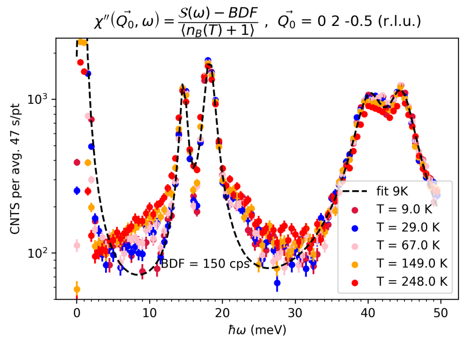
Figure 1: Longitudinal phonon modes in Sr_2CuO_3.



(a) Transverse Phonon dispersion relation along b^* at 10K.



(b) Transverse Phonon dispersion relation along c^* at 10K.



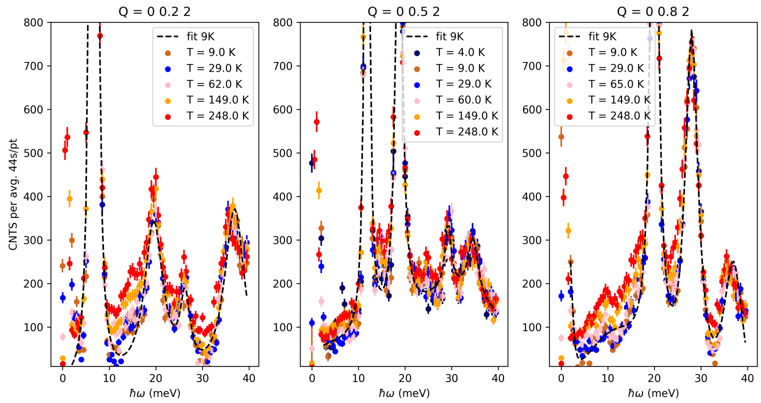
(c) Avoided crossing evolution with temperature of transverse phonons along c^* from 10 to 250 K.

Figure 2: Transverse phonon modes in Sr_2CuO_3.

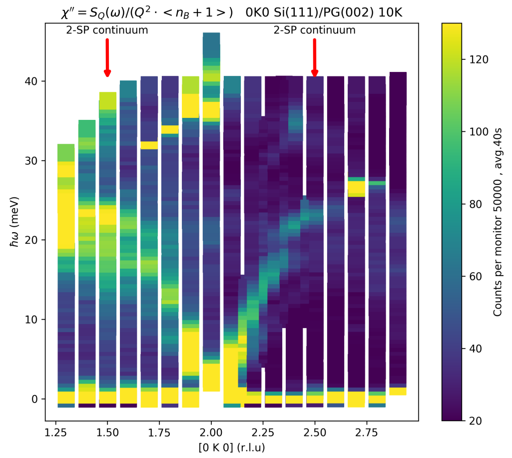
3 Transverse phonon dispersion

0K2 On Figure 2a is shown the transverse phonon dispersion, when $\vec{q}$ is along the spin chain axis [0K0]. The soft acoustic branch plateaus at (0, 0.5, 2, 11.8(4) meV) before fading into background. Stemming from Γ_{002} at 19.8 meV a flat optical branch, likely the IR-active E//c mode at 20.8 meV, picks-up the spectral weight of the acoustic branch starting at (0, 0.3, 2) and onward. At higher energy, one observes a flat optical branch starting at Γ_{002}, 25.4 meV, probably the Raman-active mode at 25.4 meV $E//c$, propagating along b^*. This branch is barely visible but jumps out of background at $(0, 0.5, 2, 30\,\mathrm{meV})$. A last optical mode is observed at high energy, observed at (Γ_{002}, 37.9 meV) with decreasing energy. This branch is probably the IR-active mode found at 38.2 meV. At $(0, 0.5, 2, 30\,\mathrm{meV})$ this branch seems avoiding crossing with the other TO and then curves upwards. At $(0, 0.5, 2)$ the phonon dispersion also meets the 2-spinon continuum which is remarkably bright. As so much is happening at this Q-point (possibly two avoided crossing and magnetic excitations), we looked for an effect of temperature on the inelastic spectrum at this position, and two positions aside at $(0, 0.2, 2)$ and $(0, 0.8, 2)$.

02L On Figure 2b is shown the transverse phonon dispersion propagating perpendicularly to the spin chain, $\vec{q}$ is along c^* (in the CuO_4 plaquette plane, $\perp$ to the spin chain). A soft acoustic phonon mode meets the optical mode found at 19.8 meV at Γ_{002}. This mode is probably the IR-active mode found at 20.8 meV at Γ when $\vec{E}//\vec{c}$. At higher energies two bright optical modes propagate with a smooth positive quadratic curvature; those modes most likely correspond to the two IR-active modes at 38.2 meV $\vec{E}//\vec{c}$ and 42.5 meV $\vec{E}//\vec{b}$. One can see a pronounced avoided crossing of the acoustic phonon branch and the optical one starting at 20 meV at Γ_{020}. The two modes are extracted at 14.7 and 18.2 meV at $Q_L = 2.5$.The low energy branch then softens to 8.4(35) meV at the Brillouin Zone boundary. This low energy mode might be called a soft mode, and its temperature dependence could be of interest in the future. The temperature dependence of the avoided crossing is shown on Figure 2c and discussed below.



(a) Avoided crossing evolution with temperature of transverse phonons along b^* from 10 to 250 K.



(b) Susceptibility along 0K0.

4 Avoided crossing with temperature

02L see Figure 2c. First the acoustic and optical modes, respectively at 14.7(4) meV and 18.2(7) meV seem unaffected by temperature, both in amplitude and linewidth. Same goes for the two high energy modes at 40.2(18) meV and 44.6(17) meV. Only the mode at 40.2 meV gets slightly asymmetric with increasing temperature. The simple model does not include a shoulder of the mode at 18.2 meV, probably due to another optical mode; this mode is however hardly seen on the map on Figure 2b.

0K2 On Figure 3a is shown three Q-cuts at different temperatures. The middle panel of the figure is taken at the 2-spinon continuum position. The phonon modes were fitted by a sum of 4 Lorentzians plus a constant background (the elastic line was not taken into account). Only the fit of the data at 9 K is shown with dashed lines as a guide to the eye. The four modes fitted do not shift with temperature, once the thermal balance factor is taken into account on the figure. However some intensity at 6.5(17) meV in the middle panel when $T < T_N$ is likely a transverse magnon mode. Finally the mode at $Q = 0\ 0.8\ 2$ with energy 37.0(27) meV at 9 K softens slightly to 36.4(22) meV at 250 K, but the difference is not significant. Both at $Q =$0 0.2 2 and 0 0.8 2 one can see a stronger intensity at energies 14 meV and 10 meV respectively, when $T >$100 K where the heat conductivity has decayed. At 250 K the fallout of the acoustic branch is more intense; the branch is almost flat at this position in the dispersion. As the acoustic branch is flat at this position, the density of states must peak at this energy; This branch is likely more occupied as temperature rises, and because it is not dispersive, this branch provides states not participating in the heat conduction. It may be a factor to explain the high temperature thermal conductivity behavior of the compound.

0K0 We observe that the phonon energies and linewidths are unaffected by temperature at scans recorded at 0 1.5 0.

5 A possible coupling with spin degree of freedom

On Figure 3b we observe an anomalous trend for the intensity; the scattered intensity seems too strong at low Q. The susceptibility should be constant with $\vec{Q}$ while it is not the case, as we observed with $SrCu_{99\%}O_2Zn_{1\%}$. At low Q the magnetic form factor is stronger, so we speculate that this scattered intensity may have a magnetic origin; a polarized neutron experiment would be necessary to test this hypothesis.

6 Conclusion

- Our measurement is comparable with Raman and IR previous measurements. A possible low energy branch was found at Gamma and might be a silent mode. The polarization of each phonon is however still lacking.
- Along the spin chain axis, low energy optical modes impede the propagation of the stiff acoustic branch, and results in many avoided crossings and spectral weight transfer. This indicates that the phonon-only part of the heat transfer is modified after 17 meV. In addition, the scattered intensity is stronger at low Q, in the same fashion as with $SrCuO_2$; it could indicate a coupling or hybridation between phonons and magnetism.
- Along the transverse directions, the acoustic branch avoids crossing with a flat optical mode, and end up with low energy ($\approx$ 8 meV) at the Brillouin Zone border. This mode could be a "sink" for thermal energy, and may explain high temperature properties of the compound. It could be of interest to follow these low energy modes with temperature in a future experiment.