Experimental report

12/09/2022

Proposal: **7-01-532** **Council:** 4/2020

Title: High energy Spinon-phonon couplingin spin chain cuprates

Research area: Physics

This proposal is a new proposal

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Samples: Ca2CuO3

Instrument	Requested days	Allocated days	From	To
IN8	4	4	21/09/2020	25/09/2020

Abstract:

The magnetism of Heisenberg spin chains has aroused a lot of interest in the last decades. SrCuO2 , Sr2CuO3 or Ca2CuO3 are emblematic realizations of this model. In those compounds, that align 180 degrees Cu-O-Cu bonds along one crystallographic axis, strong and anisotropic thermodynamic properties were found. Those exceptional properties are due to the peculiar 1D S=1/2 magnetic excitations of the spin chains, the spinons. At temperatures above the conduction peak (22K), the conduction properties decay in a manner which is not yet understood. Spin excitations end up scattering with thermally activated phonons, and as their mean free path decays so does the heat conductivity.

Recently, Chen et al. measured the thermal conductivity of Ca2CuO3 and conclude from transport measurements that spinon scatter with phonons empirically. On the basis of a DFT calculation, they claim that two specific optical phonons at 25 and 75 meV, polarized along the spin chain, scatter with the spinons. As the argument of the chemical bond modulation is compelling, and could be the origin of the dissipation of spinon's energy, we want to show this spin-lattice coupling by a spectroscopic measurement.

Experimental report on Lattice dynamics in cuprate spin chains

For a comprehensive overview of the work that was carried out, this document gathers experimental reports from the following experiments:
7-01-473 Lattice dynamics in the low-dimensional $SrCuO_2$
7-01-532 High energy spinon-phonon coupling in spin chain cuprates
7-01-536 Soft mode temperature dependence in spin chain cuprates

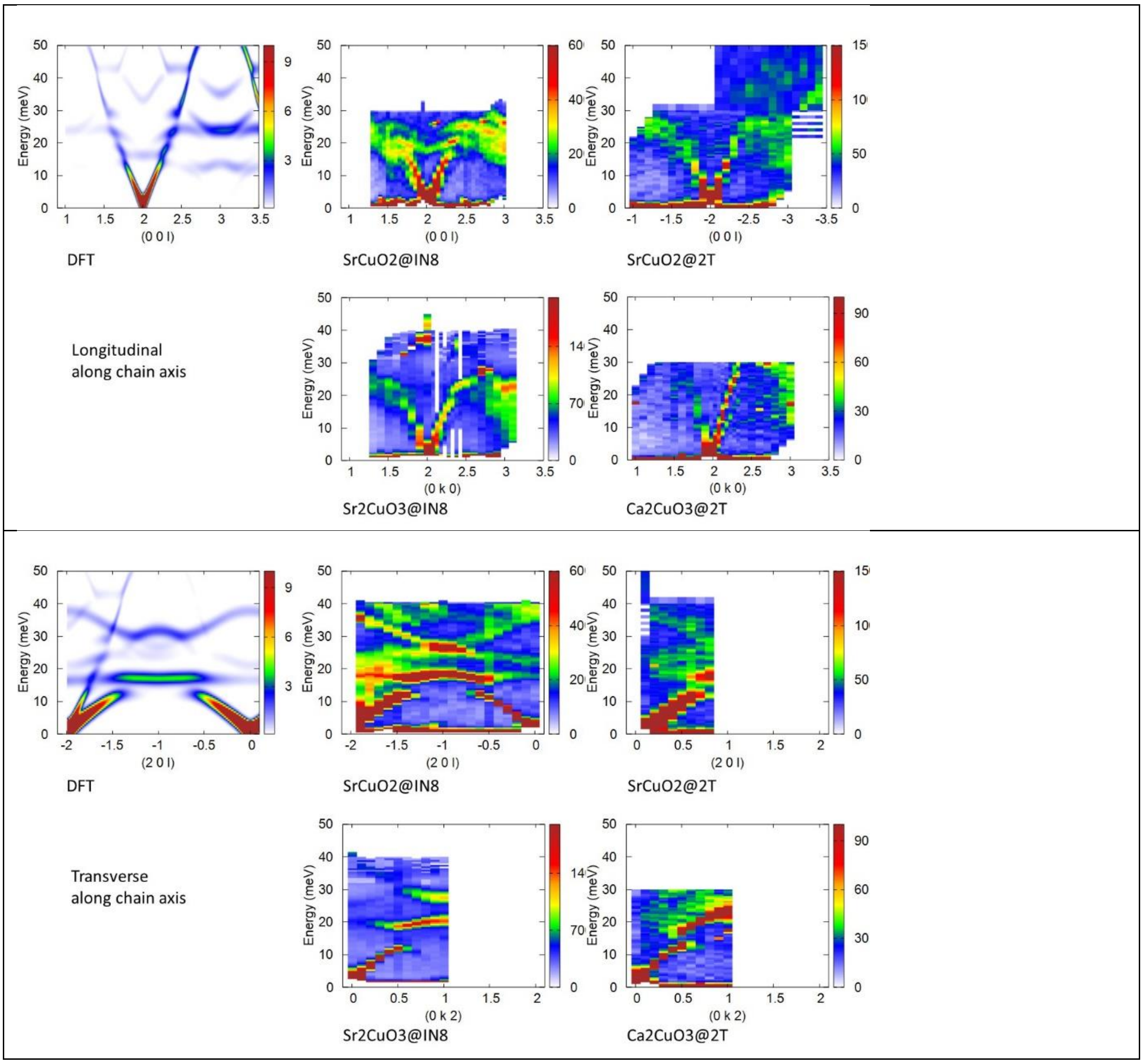
Introduction: The magnetism of Heisenberg spin chains has aroused a lot of interest in the last decades. $SrCuO_2$, Sr_2CuO_3 or Ca_2CuO_3 are emblematic realizations of this model. In those compounds, which are characterized by 180° Cu-O-Cu bonds along one crystallographic axis, unusual anisotropic thermodynamic properties have been found; for example, heat conductivity peaks at 800 watt/m/K at T=37 K in $SrCuO_2$, a value comparable to that observed in copper. Those exceptional properties are believed to be due to the peculiar 1D magnetic excitations of the spin 1/2 chains, the so-called spinons: in addition to phonons, which are the main heat carriers in insulators, spinons are also found, rather strikingly, to participate to these very peculiar thermal properties. In other words, two types of quasiparticles have to be considered to characterize heat transport; thermal conductivity thus writes $\kappa(T) = \kappa_{Phonons}(T) + \kappa_{Spinons}(T)$ and this naturally raises the question on how to single out the spinon contribution $\kappa_{Spinons}(T)$.

Context and methodology: in published studies, this contribution was estimated by measuring the heat conductivity along and perpendicular to the spin chains. As spinons only propagate along the chain direction, the subtraction between these two data was believed to give a good approximation of $\kappa_{Spinons}(T)$. Based on this estimation, it was found that at temperatures above the conduction peak, the spinon conduction properties decay in a way which is not yet understood. It is likely that the spin excitations end up scattering with thermally activated phonons, and as their mean free path decays so does the heat conductivity. The series of experiments performed (mostly) on $SrCuO_2$, along with Sr_2CuO_3 or Ca_2CuO_3 on IN8 and summarized in this report, were devoted to a better understanding of these issues. All experiments were carried out smoothly and efficiently, and we thank all the IN8 staff for their precious help and expertise.

I. we first investigated any manifestation of a coupling between phonon modes and the two spinon continuum.
II. we also mapped out the phonon dispersions in various directions, and compared them to theoretical calculations based on DFT force constants in $SrCuO_2$ (performed by Rolf Heid, KIT). As shown in the figures below, calculations were found to be in quite good agreement with experimental data. We thus decided to compute $\kappa_{Phonons}(T)$ from the phonon dispersion. Strikingly, it was found to be anisotropic, being larger in the $\vec{c}$ direction parallel to the spin chains. By subtracting the calculated $\kappa_{Phonons}(T)$ to the experimental $\kappa(T)$ along $\vec{c}$, we were then able to better estimate the contribution of spinons to the thermal conductivity along $\vec{c}$, $\kappa_{Spinons}(T)$. Our results suggest that the spinons mean free path is much shorter than the phonons' at low temperature, and drops sharply above 40 K, down to a few dozen ångström only.

Experimental details: we carried out inelastic neutron scattering measurements on Ca_2CuO_3, Sr_2CuO_3 and $SrCuO_2$ single-crystals (grown in an image furnace). We operated IN8 using SI/PG and k_f = 2.662 $Å^{-1}$. For completeness, some of the data shown below corresponds to additional experiments on 2T and

1T (LLB). We distinguish longitudinal and transverse phonons propagating along and perpendicular to the chains. Scans were also taken with the Cu/PG k_f = 4.1 Å^{-1} in the high energy range (not shown).



Interestingly, the spectra look similar for the three compounds (taking into account permutation of the crystallographic axes). Importantly, the two-spinon continuum, in these maps, takes the form of a (blue) vertical line, because the exchange is extremely strong, close to 2000 K, hence 200 meV. In all compounds, we looked for anomalies in the phonon spectra that would indicate a possible coupling with the spinon excitation. Yet, such a coupling has remained elusive so far. To be noticed is the excellent agreement of the data with DFT calculations in the case of $SrCuO_2$, except perhaps for precise values of the energies, along with some details of the dispersions : for instance the strong intensity of optical phonons along (02L) in Sr_2CuO_3, or the odd dispersion of acoustic phonons along (h00) in $SrCuO_2$, which are not captured properly. We also noticed a large number of avoided crossing leading to many low energy phonons. We anticipate that this feature might have a strong impact on the phonon contribution to the thermal conductivity of the three compounds.

It is also worth mentioning a « soft mode » at (-10-2) in $SrCuO_2$, (0 2 -1) in Sr_2CuO_3 and Ca_2CuO_3 in the transverse phonon modes propagating perpendicular to the Cu chains. This was the subject of a dedicated experiment. As determined by our analysis, this feature appears at different Q positions, but which are comparable considering the labeling of crystallographic axes in different space groups. More importantly, using DFT force constants, it was possible to determine that this soft mode actually arises because of intrinsic weakness in the crystal structure, related to the specific 5-fold coordination of the Sr or Ca ions. This feature could also be responsible for the low phonon contribution to the thermal conductivity.