Experimental report

10/09/2022

Proposal: **4-01-1690** **Council:** 4/2020

Title: Spin-phonon coupling in spin chaincuprates

Research area: Physics

This proposal is a new proposal

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Samples: Ca2CuO3

Instrument	Requested days	Allocated days	From	To
IN8 Flatcone	5	5	02/10/2021	07/10/2021

Abstract:

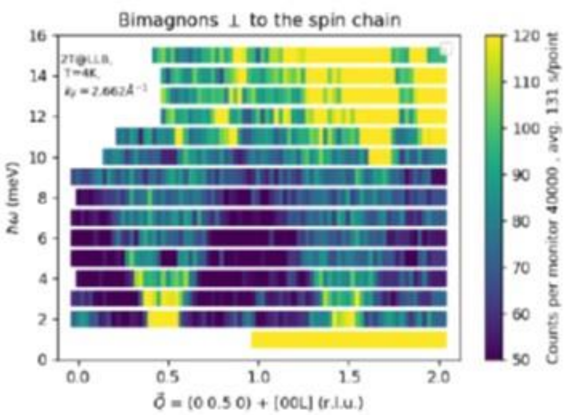
The magnetism of Heisenberg spin chains has aroused a lot of interest in the last decades. SrCuO2, Sr2CuO3 or Ca2CuO3 are emblematic realizations of this model. In those compounds, that align 180 degrees Cu-O-Cu bonds along one crystallographic axis, strong and anisotropic thermodynamic properties were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

In the present experiment, we propose to take advantage of the confinement of those spinons in conventional magnons in the low temperature 3D AF ordered phase, to look for a more conventional coupling between collective modes. We want to study the formation, dispersion and temperature evolution of the peculiar low energy magnons spectrum. Furthermore, we will focus on a point of reciprocal space where magnons cross the phonon spectrum, above and below TN, to show a possible anti-crossing.

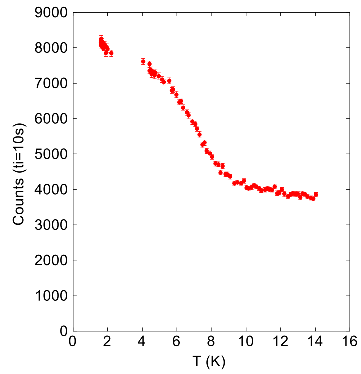
Exp report 4-01-1690 Spin phonon coupling in cuprate spin chain

Context: 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; for example the heat conductivity peaks at 500 W/m/Kat T = 22K in SrCuO2 a value comparable to that of 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. These S = 1/2 particles were made famous by the Des Cloizeaux-Pearson predictions and the observations of the 2-spinons continuum in KCuF3. At temperatures above this conduction peak, the conduction properties decay in a manner, which is not yet understood. We suppose that the spin excitations end up scattering with thermally activated phonons, and as their mean free path decays so does the heat conductivity. To shed light on this alleged spinon-phonon scattering, we carried out crystal growth by image furnace, and inelastic neutron scattering studies of Ca2CuO3, Sr2CuO3 and SrCuO2. We especially mapped out the Ca2CuO3 phonon dispersion and looked for a possible coupling with the spinon spectra. Yet, despite much effort, such a coupling has remained elusive so far. This study is made even more complicated by the fact that neutrons create pairs of spinons, preventing the observation of their individual dispersion. As a result, conventional signatures, like for instance, an anti-crossing cannot be unveiled.

In the present experiment, we proposed to take advantage of the confinement of those spinons in conventional magnons in the low temperature 3D AF ordered phase, to look for a more conventional coupling between collective modes. Indeed, these cuprates eventually order at low temperature, even if they are undoubtedly good 1D systems (running along b*), as inferred from the ratio of the Néel Temperature T_N and the superexchange J. In this low temperature regime, spinons should combine to form conventional magnons, while their dispersion would indicate the strength of the inter-chain superexchange J' (probed by measuring along a* or c*). Furthermore, in this classical regime, spin-phonon coupling may take the form of magnon-phonon interactions. In this experiment at IN8, we focused on the Ca2CuO3 case. Below 8 K, Ca2CuO3 enters a 3D AF long-range order. Actually, we found, during a previous experiment at 2T@LLB, two dispersion cones emerging from the magnetic Bragg peaks at (0, 0.5, 0.5) and (0, 0.5, 1.5) below T_N:

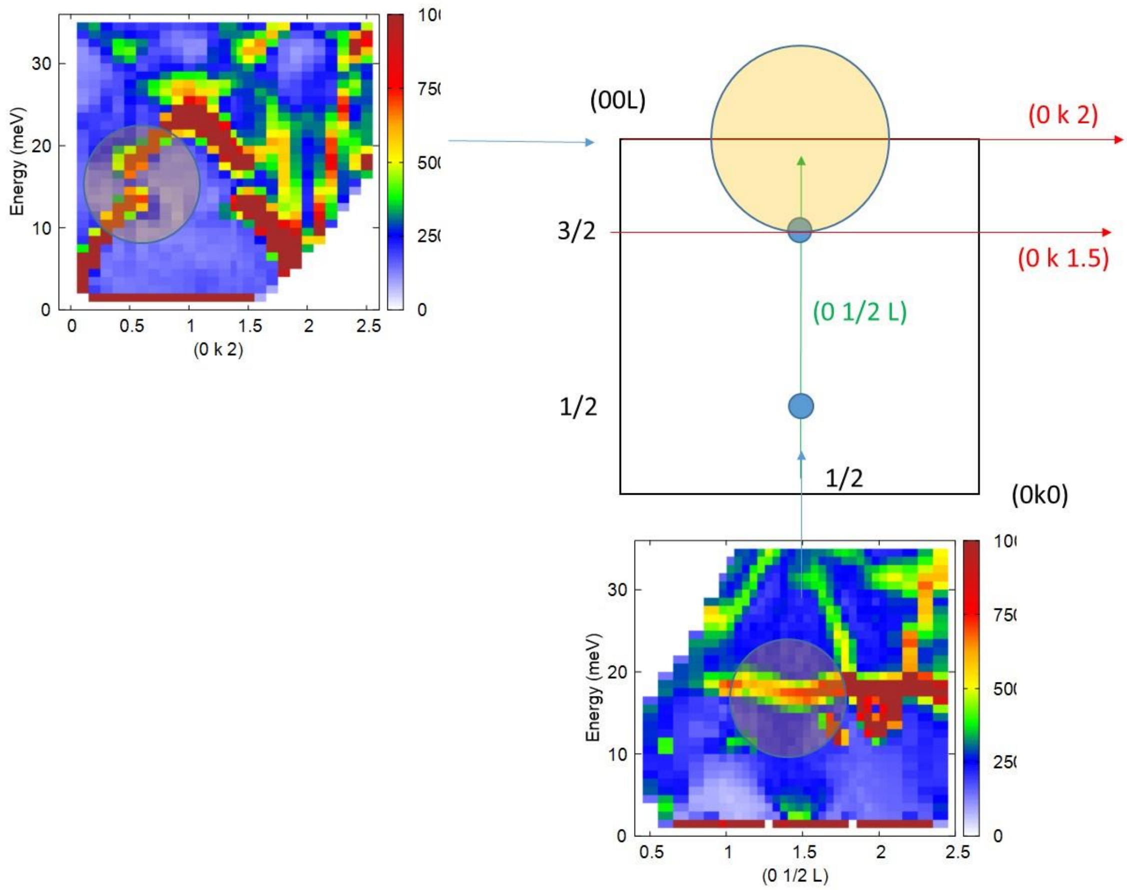


(a)

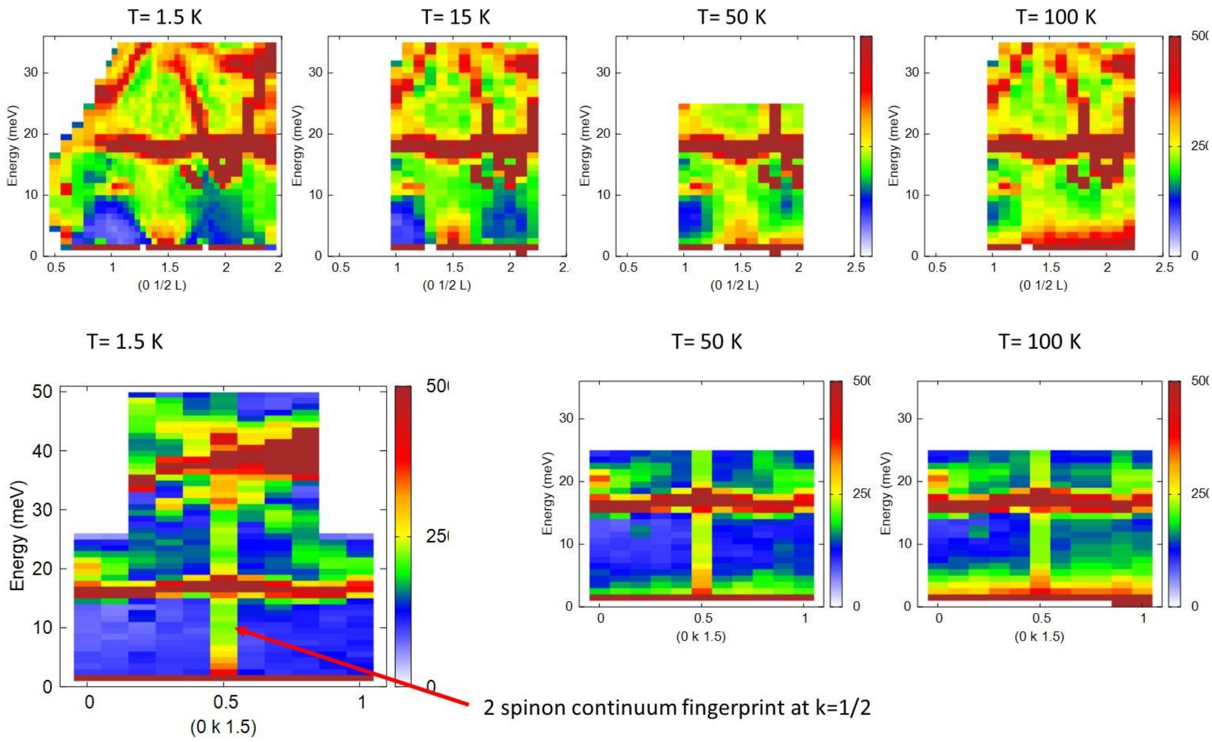


Intriguingly, the spectrum does not show well-defined excitations but is rather characterized by strongly enhanced two-magnons scattering (longitudinal fluctuations), with some spectral weight filling the region delimited by the spin wave dispersions. This result is consistent with strong quantum fluctuations and a significantly reduced ordered Cu magnetic moment. The rise of the Bragg peak was by the way quickly monitored as shown above. We also found that the magnon-like branch reaches about 12 meV at the magnetic zone boundaries as for instance Q=(0 0.5 2). This coincides with the energy range of two transverse phonons propagating along the chain axis b in Ca2CuO3 at 13 and 17 meV. As shown in the Figure below, anti-crossing between the two phonon modes appears at this point Q=(0 0.5 2).

To further investigate the spin-phonon coupling, we proposed to investigate this region of reciprocal space in more details as a function of temperature, from 50 down to 1.5 K, looking for instance for a "repulsion" or for a resonant coupling between the different modes. In the figure below, we present a sketch of this region highlighted by a yellow circle. Are represented the direction of (0 k 2) scan, unveiling phonons, the direction (0 0.5 L), probing the transverse dispersion of the magnons with the magnetic Bragg peaks (blue disks) at (0 0.5 0.5) and (0 0.5 1.5). Scans along (0 k 0.5) and (0 k 1.5) would probe dispersions along the chains. The figure shows maps taken along these directions. The spin wave dispersion is also visible on this map. IN8 was here operated Si(111) /PG and kF =2.662 A.



We then followed these data as a function of temperature, including (0 K 2), (0 ½ L) and also (0 K 3/2). The figure below shows (0 K 2) and (0 K 3/2). The spin wave dispersion softens and becomes quasi-elastic with increasing temperature, as expected, however, the phonon spectral weight apparently does not change. Analysis is ongoing; however, no clear signature of any spin lattice coupling could be evidenced in this experiment. The issue of the spinon contribution to heat thermal conductivity thus remains an issue.


T= 1.5 K
T= 15 K
T= 50 K
T= 100 K
Energy (meV)
(0 1/2 L)
(0 k 1.5)
2 spinon continuum fingerprint at k=1/2