# MAGNETIC EXCITATIONS IN THE SPIN LADDER COMPOUNDS $Sr_{14-x}Ca_xCu_{24}O_{41+d}$

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At the boundary between dimensions one and two, spin-ladder systems are conceptually very interesting as they exhibit rather "exotic" properties. In particular, the spin-pairing expected to develop in a 2-leg ladder gives rise, upon doping, to a charge pairing and finally to a non conventional (i.e. non phonon mediated) superconductivity. In this report, we present recent neutron inelastic scattering results obtained on a single crystal of the undoped $Sr_{14}Cu_{24}O_{41+\delta}$ and doped $Sr_{14-x}Ca_xCu_{24}O_{41+\delta}$ 2-leg spin-ladder compounds.

After the discovery of the high-Tc superconductivity, a renewed interest in low-dimensional quantum magnetism has emerged, motivated by the possible role played by the magnetic interactions in the charge-pairing mechanism. One-dimensional antiferromagnets are particularly interesting to consider as they often exhibit unconventional phenomena. The first striking effect was discovered in the early 80's by Haldane[1], who suggested that Heisenberg antiferromagnetic chains with half-integer (S=1/2,3/2, ... ) and integer (S=1, 2, ... ) spin values behave quite differently at low temperatures. Whereas the former is expected to be gapless, the latter should have a non-magnetic singlet ground state and a quantum gap should open in the magnetic excitation spectrum. This non-intuitive prediction has been further very comprehensively verified from neutron-inelastic-scattering experiments performed on the spin-1 antiferromagnetic chain compound $Ni(C_2H_8N_2)_2NO_2ClO_4$.[2]. On the other side, the spin-1/2 square lattice with antiferromagnetic nearest-neighbour Heisenberg exchange couplings exhibits a quasi-ordered gapless ground state at T=0.

Spin-ladders can be viewed as an array of a finite number of coupled chains, allowing therefore to study the crossover between space dimensions 1 and 2. While spin ladders with an odd number of legs behave like the spin-1/2 antiferromagnetic chain (gapless excitation spectrum, power-law spin correlations, ... ), those with an even number of legs exhibit an exponential decay of the spin-correlations due to the opening of a spin-gap (with energy A) in the excitation spectrum[3]. Of particularly high interest is the 2-leg spin-ladder system, since the prediction that charge doping could induce a non-conventional superconductivity, in which a d-wave pairing could be achieved, driven by the magnetic fluctuations[3]. Indeed, superconductivity has been recently discovered in the Ca-doped spin-ladder family $Sr_{14-x}Ca_xCu_{24}O_{41+\delta}$ for x>11, under high pressure in the range 30-45 kbar[4] The understanding of the mechanism yielding to superconductivity in this material requires an accurate determination of both the temperature and doping dependencies of the magnetic excitation spectra. This can be achieved by neutron-inelastic- scattering investigations on undoped and doped single crystals.

As a first step, we have recently undertaken such a determination on the undoped $Sr_{14}Cu_{24}O_{41+\delta}$ and doped $Sr_{14-x}Ca_xCu_{24}O_{41+\delta}$ compounds. The structure of this material is a misfit stacking of layers of two distinct quantum spin systems : linear $CuO_2$ chains and 2-leg $Cu_2O_3$ ladders[5]. Figure 1 shows a "3D" view of the crystallographic structure, which emphasizes the chain and ladder subsystems. Quite interestingly, the pure material contains a large amount of holes mainly localized in the $CuO_2$-chains (0.6 hole/Cu), which play the role of a charge reservoir.

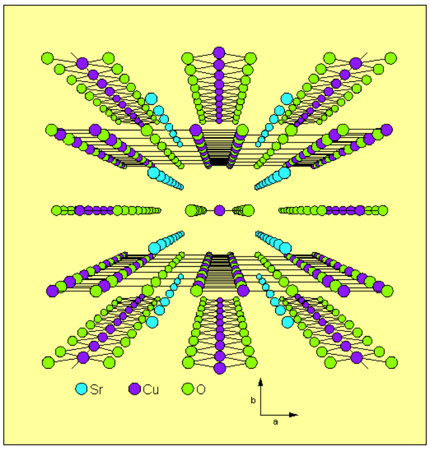


*Figure 1: Crystal structure of $Sr_{14}Cu_{24}O_{41+d}$ showing the chain and ladder subsystems (viewed along the c axis).*

Following the theoretical predictions, the ground state of a 2-leg spin-ladder system in the case $J_\perp < J_{//}$, where $J_{//}$ and $J_\perp$ represent respectively the exchange coupling constants along the legs and along the rungs, should be a non magnetic singlet ground state, well separated from the first excited states by an energy gap $\Delta \approx 0.4$

$J_\perp$. These two features have been unambiguously observed from neutron-inelastic-scattering experiments carried out on the 3-axes spectrometers (TAS) IN8/ILL, 1T/LLB and IN22/CRG-ILL. We show in Figure 2 two typical energy scans performed on TAS 1T/LLB at the scattering vectors **Q**=(4.5, 0, 0.5) (where one expects a strong signal originating from the ladders) and **Q**=(4.5, 0, 0.65) (where one expects a vanishing contribution of the ladders). The observed line shape is characteristic of a gapped magnetic response, with no signal detected at low energy and a gap value Δ=33 meV. The magnetic response can be understood from the existence of "two" contributions: one narrow contribution peaked at Δ and a second one, persisting at much higher energy and peaked around 40-50 meV. The temperature dependence of the magnetic response reveals several interesting and new features:

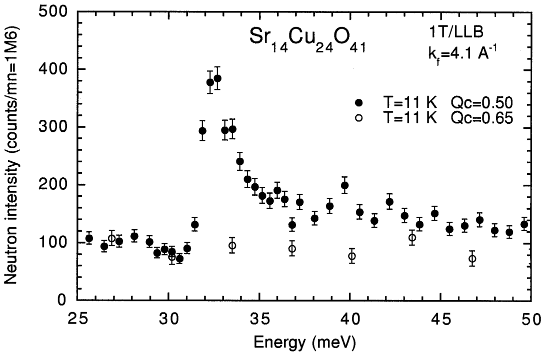


*Figure 2: Inelastic neutron scattering response of the ladder subsystem at 1.5 K, showing the presence of a quantum spin-gap with energy 33 meV.*

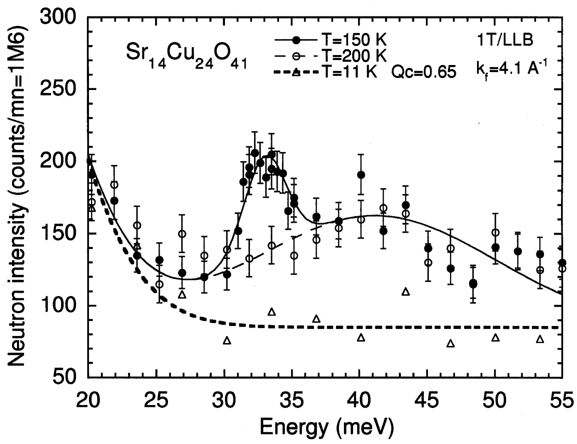


*Figure 3: Dynamical magnetic response at 150K and 200K showing the existence of two distinct contributions in $Sr_{14}Cu_{24}O_{41+d}$*

- the "33 meV-peak" disappears above roughly 200K, without strong renormalization nor damping below 150K. Figure 3 displays constant-Q scans at the scattering vector **Q**=(4.5,0,0.5) and two temperatures T=150K and 200K which demonstrate the presence of two distinct contributions.
- the second contribution is wealdy temperature dependent above 200K and extends at least up to 90 meV (data taken on IN1 and IN8/ILL).

We have presently no definitive explanation for these results, which however bear some ressemblance with those obtained on, e.g., $YBa_2Cu_3O_{6.5}$ in the normal state [6].

Upon Ca-doping, holes are progressively transferred from the $CuO_2$ chains to the $Cu_2O_3$ ladders which become metallic. Our neutron data show that the spin dynamics in the ladders is little affected by hole doping. The magnetic response, despite well visible broadening effects, remains peaked at 33 meV. At the reverse, the spin dynamics in the chain is much more affected, as a result of the partial destruction of the charge ordering in the chain subsystem.


**References:**

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