

**La<sub>5</sub>(Sr,Ca)<sub>9</sub>Cu<sub>24</sub>O<sub>41</sub>: A 1D TOY MODEL FOR HOLE DOPED CUPRATES**

R. Klingeler<sup>1</sup>, A. Gukasov<sup>2</sup>, T. Kroll<sup>1</sup>, J. Geck<sup>1</sup>, M. Hücker<sup>3,4</sup>,  
U. Ammerahl<sup>4</sup>, A. Revcolevschi<sup>4</sup>, B. Büchner<sup>1</sup>

<sup>1</sup> Leibniz-Institute for Solid State and Materials Research IFW Dresden, 01171 Dresden, Germany

<sup>2</sup> Laboratoire Léon Brillouin, CEA-Saclay, 91191 Gif sur Yvette, France

<sup>3</sup> Physics Department, Brookhaven National Laboratory, Upton, New York 11973

<sup>4</sup> Laboratoire de Physico-Chimie des Solides, Université Paris-Sud, 91405 Orsay Cedex, France

The interplay between low-dimensional antiferromagnetism and the mobility of holes in layered cuprates is studied intensively nowadays. A different approach to investigate this interplay considers quasi-one-dimensional (1D) systems. For 1D systems, theory is much more predictive and experiments yield more pronounced results. In our 1D model system (Sr,Ca,La)<sub>14</sub>Cu<sub>24</sub>O<sub>41</sub> for doped cuprates, a small number of charge carriers significantly affects the magnetic properties. In particular, the mobility of holes can be tuned by a magnetic field and thus a long range AFM spin order, which is present if the holes are static, is destroyed by the motion of the depinned holes.

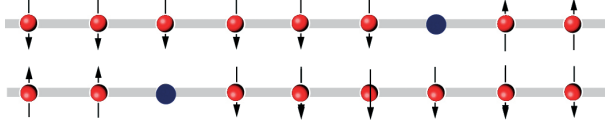


Figure 1. Sketch of adjacent doped spin chains.

In (Sr,Ca,La)<sub>14</sub>Cu<sub>24</sub>O<sub>41</sub>, two quasi-1D magnetic substructures are realized, i.e. Cu<sub>2</sub>O<sub>3</sub> spin ladders and CuO<sub>2</sub> spin chains. In the ladders, applying a hydrostatic pressure in Sr<sub>14-x</sub>Ca<sub>x</sub>Cu<sub>24</sub>O<sub>41</sub> ( $x \geq 11$ ) results in a superconducting state. However, the ladders do not contribute significantly to the low temperature magnetic response, since they exhibit a large spin gap of  $\Delta \sim 400$ K. The chains consist of edge-sharing CuO<sub>4</sub>-plaquettes containing Cu<sup>2+</sup>-ions with  $S = \frac{1}{2}$  in the undoped case. The Cu-O-Cu bonding angle amounts to  $\sim 93^\circ$ , which results in a ferromagnetic (FM) superexchange between adjacent (NN) spins and in an uniaxial anisotropy. Hole doping introduces non-magnetic Zhang-Rice singlets. The magnetic coupling of next nearest neighbor (NNN) Cu-spins via a hole is AFM, similar to the stripes in layered cuprates. For the low doped (Sr,Ca,La)<sub>14</sub>Cu<sub>24</sub>O<sub>41</sub> with  $x \sim 5$ , i.e.  $\sim 10\%$  holes, the Cu-spins in the chains form FM chain fragments which are coupled antiferromagnetically via the few non-magnetic holes (cf. Fig. 1). Due to a finite interchain coupling  $J_\perp \sim 10$ K a long range (LR) AFM spin order evolves below  $T_N \sim 10$ K [1].

### Experiments and results

We performed a detailed investigation of the low temperature spin correlations in the chains of

La<sub>x</sub>(Sr,Ca)<sub>14-x</sub>Cu<sub>24</sub>O<sub>41</sub> ( $x \geq 5$ ). The experiments were carried out on the 6T2 diffractometer with the 7.5T cryomagnet using unpolarized neutrons of wavelength  $\lambda_n = 0.90\text{\AA}$ . Polarized neutron flipping ratios were measured on the lifting-counter diffractometer 5C1 using neutrons with  $\lambda_n = 0.84\text{\AA}$  obtained with a Heusler alloy monochromator. We used single crystals of approximately  $0.2\text{cm}^3$  grown by the floating zone technique which were oriented with the  $b$ -axis either vertical or parallel to the magnetic field direction.

La<sub>5</sub>Sr<sub>9</sub>Cu<sub>24</sub>O<sub>41</sub> was characterized at 300K using the 6T2 diffractometer in the four-circle mode. A refinement on neutron scattering structure amplitudes has been done using the superspace formalism and the crystallographic program JANA2000. In contrast to recent findings for Sr<sub>14</sub>Cu<sub>24</sub>O<sub>41</sub> [2], our refinement for La<sub>5</sub>Sr<sub>9</sub>Cu<sub>24</sub>O<sub>41</sub> at room temperature yields the shift along the  $c$ -axis of adjacent chains in the  $ac$ -plane  $\delta = 0.01(1)$ , which corresponds to an *in line* arrangement of adjacent chains along the  $a$  axis.

LR spin order due to a finite  $J_\perp$  is a generic feature of quasi-1D magnets. Unusually for cuprates, for La<sub>x</sub>(Sr,Ca)<sub>14-x</sub>Cu<sub>24</sub>O<sub>41</sub> ( $x \geq 5$ ) there is a strong anisotropy of the magnetic phase diagram. As demonstrated in Fig. 2, the intensity of the AFM (1,1,0) reflection strongly decreases if a magnetic  $B \parallel b$  is applied. In particular, the intensity of the (1,1,0) reflection almost disappears for  $B \parallel b \geq 4$ T. The suppression of the LR spin order is confirmed by

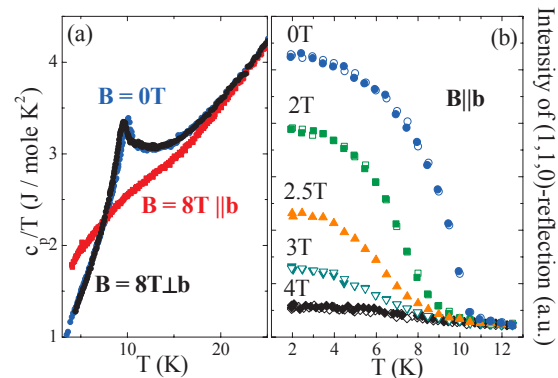


Figure 2: (a) Specific heat  $c_p/T$  of La<sub>5.2</sub>Ca<sub>8.8</sub>Cu<sub>24</sub>O<sub>41</sub> for  $B = 0$ ,  $B = 8T \parallel b$ , and  $B = 8T \perp b$ . (b) Maximum intensity of the AFM (1,1,0) reflection for different  $B$  at cooling.

the specific heat data. For  $B||b=8\text{T}$ , there is no signature of LR spin order at  $T \geq 3\text{K}$ . Whereas, the specific heat shows that the LR spin ordered phase is hardly influenced by  $B\perp b = 8\text{T}$ . The anisotropic phase diagram is also confirmed by neutron diffraction, magnetization and specific heat for  $\text{La}_x(\text{Sr,Ca})_{14-x}\text{Cu}_{24}\text{O}_{41}$  with  $x = 5, 5.2, 5.6$  [3].

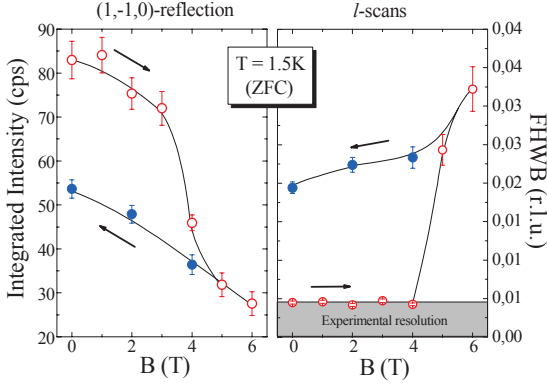


Figure 3. (a) Integrated intensity (from  $l$ -scans) and (b) FWHM of the AFM (1,-1,0)-reflection in  $\text{La}_5\text{Sr}_9\text{Cu}_{24}\text{O}_{41}$  at  $T=1.5\text{K}$ . Lines are guides to the eyes. Arrows denote the course of the measurements, starting from ZFC.

In Fig. 3, the magnetic field dependencies of the integrated intensity of the AFM (1,-1,0)-reflection of  $\text{La}_5\text{Sr}_9\text{Cu}_{24}\text{O}_{41}$  and of its width (FWHM) are displayed. The data show that small magnetic fields cause a moderate suppression of the AFM order parameter (OP), while any effect on the FWHM, respectively on the correlation length  $\xi$ , is beyond the resolution of the experiment. At  $B = B_C \sim 4\text{T}$ , a sharp decrease of the intensity of the (1,1,0)-reflection is observed, but the integrated intensity remains finite for  $B > B_C$ , where LR spin order is absent. In the mean time, the width of the (1,1,0)-reflection sharply increases at  $B_C$  but remains finite in higher fields. Both facts evidence short range (SR) AFM correlations for  $B||b > B_C$ . Note, that at  $B_C$  no spin reorientation occurs since the anisotropy field amounts to  $B \sim 7.5\text{T} \gg B_C$  [3]. The data in Fig. 3 exhibit a pronounced hysteresis, i.e. switching off the magnetic field does not restore the AFM order completely. This is most evident if the effect of switching off the field is considered in the field cooled (FC) state at  $T = 2\text{K}$ , i.e. after cooling in  $B||b = 4\text{T}$  (Fig. 4). The AFM OP in the FC state is considerably reduced compared to the zero field cooled (ZFC) value. Thus, additional thermal energy is needed to restore the ZFC properties. This also holds for cooling in  $B = 2.5\text{T} < B_C$ . The presence of the hysteresis implies a glassy

nature of the SR spin ordered phase at  $B||b > B_C$ .

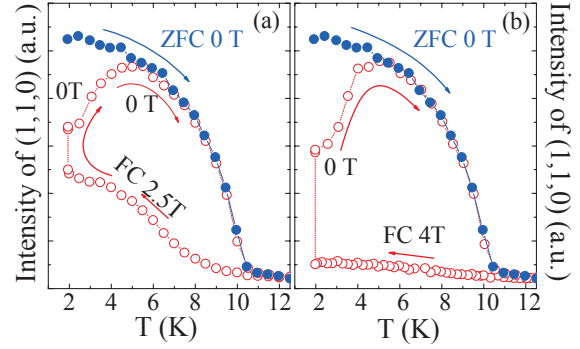


Figure 4. AFM peak intensity of the (1,1,0) reflection in  $\text{La}_5\text{Ca}_9\text{Cu}_{24}\text{O}_{41}$  for different  $B$  vs. temperature. ZFC (FC) data are shown by full (open) circles.

Our data rule out pure spin models to explain the anomalous melting of the LR spin order. In particular, the magnetic excitations induced by the field are no spin flips. This fact follows not only from the specific heat data but also from a very small value of the magnetization at  $B > B_C$ . Our polarized neutron study also yields  $\mu_{\text{FM}} \sim 0.07\mu_{\text{B}}/\text{Cu}$  at  $B = 6\text{T}$ , which is much smaller than the AFM sublattice magnetization ( $0.63\mu_{\text{B}}$  at  $2\text{K}$ ) of the Cu-ions. Hence, the data imply that  $B||b$  induces excitations, which are not purely magnetic but also include other degrees of freedom.

To explain our data, both the positions of the holes and the type of magnetic excitations must be considered. (1) If the holes are distributed randomly, the interchain coupling is frustrated and the LR spin order should not evolve (cf. Fig. 1). Thus, the observation of a LR AFM spin order implies a particular arrangement of the holes in adjacent chains. E.g., LR spin order is possible if the holes are arranged in stripes along the  $a$  axis. (2) Regarding the excitations, for an Ising-like spin chain a large magnetic field is necessary to induce a spin flop or a spin flip. However, even for small fields, a domain wall motion, i.e. a hole motion, along the chains results in a gain of Zeeman energy, since FM chain fragments parallel to  $B$  are enlarged. Therefore, we suggest that coupled spin-charge excitations, i.e. the motion of holes along the chains, occur when  $B||b$  is applied. This destroys the arrangement of holes, being present for  $B = 0$ , and yields a melting of the LR spin order. In contrast, applying  $B\perp b$  induces a conventional spin canting with no domain wall motion involved. Hysteresis effects are easily explained in this model, since hole motion is thermally activated.

## References

- [1] U. Ammerahl, B. Büchner, C. Kerpen, R. Gross, A. Revcolevschi, Phys. Rev. B **62**, 3592 (2000)
- [2] J. Etrillard, M. Braden, A. Gukasov, U. Ammerahl, A. Revcolevschi, Physica C **403**, 290 (2004)
- [3] T. Kroll, R. Klingeler, J. Geck, B. Büchner, W. Selke, M. Hücker, A. Gukasov, J. Magn. Magn. Mat., in print.